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Informal Report

Production of Synthetic Gas from Nuclear Energy Sources

Prepared for the Texas Gas Transmission Corporation under contract No. EW-78-Y-04-4183 with the US Department of Energy.

University of California

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PRODUCTION OF SYNTHETIC GAS FROM NUCLEAR ENERGY SOURCES

by

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ABSTRACT

This report documents a survey of nuclear energy sources and their potential application to the production of synthetic gas. The state-of-the-art in commercial nuclear fission reactors and ongoing R&D in advanced reactors is described. The status of fusion energy research and estimated timing of commercial availability are reported. Detailed surveys of high-temperature electrolysis and thermochemical cycles as means for producing synthetic gas from process heat are given. Synthetic gas production from radiolysis is discussed. A description of the nuclear fuel cycle and uranium reserve and resource estimates are presented.

EXECUTIVE SUMMARY

INTRODUCTION

Because of its interest in identifying new sources of gas for transportation in its pipelines by the year 2000, the Texas Gas Transmission Corporation asked the Los Alamos Scientific Laboratory to survey various methods for producing synthetic gas using nuclear energy. The goal was identification of methods that might be commercially available and economically attractive by the year 2000. Under a contractual arrangement between Texas Gas and the US Department of Energy, these studies were undertaken from May 1978 through November 1978.

In the first phase of the study, the Los Alamos team surveyed the literature on the use of nuclear energy in processes for production of synthetic gas. At a review meeting with representatives from Texas Gas, an understanding was reached on the areas to receive further study, and on which areas not to pursue. As explained below, nuclear fusion as a source of nuclear energy, and radiolysis as a means of producing synthetic gas, were removed from further consideration.

Following the survey phase, emphasis was placed on nuclear fission as an energy source, and on coal gasification, thermochemical cycles, and high-temperature electrolysis as methods for synthetic gas production. The results of the initial survey and the later studies are summarized below and are presented in detail in the main body of this report.

NUCLEAR FISSION

Introduction

Uranium occurring in nature is composed of 0.7% of the isotope uranium-235 and 99.3% of the isotope uranium-238. Although uranium-235 decays spontaneously, its half-life is about a billion years and it can, for all intents, be regarded as stable. However, in 1938 it was discovered that the uranium-235 nucleus undergoes fission (i.e., splits, forming lighter elements and releasing energy) when bombarded with slow neutrons, which then offered the possibility of giving up its energy in a sustained chain reaction (the nuclear energy from 1 lb. of uranium-235 is equivalent to the energy in 29 million cubic feet of natural gas at standard conditions). For reactor applications in the US and worldwide, the naturally occurring uranium is first mined, then refined, and then fabricated into fuel elements. Before fabrication, though, the uranium is often enriched (increasing its uranium-235 content to 3% for light water reactor, LWR, or to 93% for high-temperature gas-cooled reactor, HTGR, applications) although several reactor systems can use the enriched fuel. The enriched fuel is then inserted into the reactor system where it fissions and produces energy and a further supply of neutrons. In thermal reactors (e.g., an HTGR or LWR) the neutrons are slowed down by the presence of a moderator (usually water or graphite), which allows them to be more easily captured by the heavy uranium-235 nuclei. The reactor is controlled by maintaining a neutron population in the reactor consistent with the energy demand on the reactor; typically reactor fuels are designed for burnup times on the order of 2-3 years.

Normally, the heat produced in the fuel is used to make steam, which then generates electricity in a conventional steam power plant cycle. In various designs, the nuclear reactor can produce heat at temperatures from 300°C to 1000°C. (In addition, nuclear fragments from the fission process can directly radiolyze various molecules, as discussed later). It is the potential for producing heat near 1000°C that makes fission a potential candidate for synthetic gas production, because many synthetic gas production processes either require high temperatures or operate more efficiently at high temperatures.

Virtually all commercial nuclear reactors in the US today are of the light-water type, meaning that ordinary water acts as the moderator and flows past the fuel elements to transfer the heat to the electric generation system. Because of engineering and thermodynamic limitations, these reactors can only produce temperatures up to about 320°C. In order to reach the temperatures desired for synthetic gas production, other types of reactors may be considered, such as the gas core and high-temperature gas-cooled reactors described below.

Gas Core Reactor

In this type of reactor, the core of nuclear fuel is in gaseous form, Two gas-core reactor concepts have been studied previously. The mixed-flow reactor uses a mixture of uranium hexafluoride (UF_6) and helium to reach an operating temperature of about 1000°C. Plasma core reactors, employing vaporized uranium as fuel, could reach temperatures above 4700°C.

Because of severe materials and other technical problems which must be solved in order to make these concepts commercially viable, and the fact that the technology program has been seriously curtailed, there is no potential commercial application for these concepts before the year 2000.

High-Temperature Gas-Cooled Reactors (HTGRs)

HTGR concepts use solid nuclear fuel and employ gas cooling to remove heat from the nuclear fuel. The nuclear fuel is bound inside strong graphite particles, which serve the dual purpose of containing the fuel and, as the fuel is utilized, containing the radioactive waste products. Solid graphite is used as the moderator. Because the inert gas helium is used for cooling, the reactor can operate at a temperature higher than water-cooled reactors and the issues of safety and materials requirements are more favorable.

In the prismatic-core concept, now under development at General Atomic Co. in the US, the graphite fuel particles are pressed into fuel rods, which are then placed inside a large block of graphite, and the gas-coolant helium flows through holes in the block. Approximately 5000 blocks make up the reactor core. Refueling of the prismatic core HTGR requires a reactor shutdown. The General Atomic HTGR, when used for electricity production through steam generation, has a coolant exit temperature of about 750°C. On the other hand, Japan is developing a VHTR (Very High Temperature Reactor) for nuclear steelmaking with a prismatic core and an exit helium coolant temperature of 1000°C.

The pebble-bed reactor concept, originally an American innovation, is under development in the Federal Republic of Germany. In this reactor, the nuclear fuel is sealed inside billiard-ball size graphite shells. A full-scale reactor would contain approximately 3 million balls. This concept has the advantage that the fuel balls may be fed into the reactor and removed continuously, as the fuel is consumed, thus avoiding planned shutdowns for fuel changes. In some concepts, nuclear fuel breeding (e.g., of nonfissionable thorium to the fissionable element uranium-233) can be designed, and the bred fuel would be recycled into the reactor with no intermediate fuel processing required, as is required with the Liquid Metal Fast Breeder Reactor (LMFBR).

In order to couple an HTGR to a gas production system, an intermediate heat exchanger would probably be required to keep the noxious reactor decay products out of the gasification process, as well as combustible products away from the reactor. The materials requirements for such a system are currently being studied.

The US commitment to the development of an HTGR for process heat applications is unclear at the present time. Although the General Atomic Co. steam cycle HTGR appears to be at an end in the US, it remains to be seen if that funding will be committed to a national effort for a process heat reactor development program. On the other hand, both the Federal Republic of Germany and Japan, because of limited resources and environmental concerns, have dedicated themselves to vigorous process-heat reactor development. It is possible that by the mid 1990s each of these countries could have a process-heat reactor (of differing designs) in operation characterized by an exit coolant temperature of between 900 and 1000°C.

Availability of Fuel for Fission Reactors

The long-term availability of fissionable fuel is critical in determining the viability of fission as an energy source for the production of synthetic gas. The major issues in the nuclear fuel cycle are resources of fissionable fuels, their efficient utilization, and the ultimate disposal of radioactive wastes.

Although there is general agreement on the proven reserves of uranium, there is heated disagreement on the magnitude of domestic (unproven) resources. The most pessimistic projections indicate that the US can sustain its light-water reactor industry for only 20 years--the most optimistic, roughly 40 years. These projections would be lengthened by 30 to 40% if reprocessing of spent fuel were undertaken.

If the US chooses to develop a breeder reactor industry, the fissionable fuel estimates jump 100-fold. The reason is that the breeder converts the nonfissionable uranium-238 (99.3% of the natural uranium) into the fissionable element plutonium-239. Thus, the potential resource jumps from 0.7% of the naturally available uranium to 100%. In a breeder industry, there would be associated with each breeder reactor three nonbreeders (e.g., HTGRs) whose role is to utilize the fuel produced by the breeders. Thus, the long-term availability of nuclear fission fuel is large if the breeder is used in the US, small if it is not.

There is a good deal of controversy about disposal of radioactive wastes. There is little disagreement among informed sources that adequate isolation in underground depositories is possible, but there is substantial disagreement about the particulars of such disposal.

FUSION

Nuclear fusion is the energy source of the stars. Light atomic nuclei (e.g., hydrogen, helium) collide, forming a heavier nucleus and releasing energy. The harnessing of nuclear fusion by mankind combines a great energy resource potential with immense technological difficulties: the deuterium (heavy hydrogen) present in one gallon of natural water could release, by fusion, the energy equivalent of 300 gallons of gasoline; however, in order to start the fusion process, the fuel gases must be heated to 50 million ^{0}C and held together long enough for the fusion reactions to proceed completely.

The National Fusion Energy Program had its beginnings 25 years ago, and has expanded to an operating budget of $400 million per year in the Department of Energy. One approach called inertial confinement fusion (which includes laser fusion) passed through a phase of unbridled optimism in the early 1970s, with respected figures predicting the commercialization of laser fusion as an energy source within ten to fifteen years. However, as the program matured and a base of experimental data was taken, it became clear that there was no straightforward scientific path leading to the commercial application of laser fusion. DOE has recently estimated that the earliest operation of a fusion demonstration plant would occur in the first decade of the 21st century, with the first commercial fusion electric power plant in operation by 2015 or 2020.

Based upon the present thrust of the DOE fusion programs, the design of synthetic fuel production as an integral part of a fusion reactor system would be a second-generation fusion technology, and therefore be available much later than fusion-based electric power plants. For these reasons, we recommended and Texas Gas agreed that synthetic gas production from fusion energy not be studied further in the context of the present contract. In looking to the future, however, a status report on the fusion program and its long-range plans was prepared, in order that Texas Gas might re-examine fusion in ten to fifteen years, in order to determine whether the National program has kept pace with its projections.

SYNTHETIC GAS PRODUCTION PROCESSES

Coal Gasification

The potential application of nuclear heat sources to coal gasification was considered by a detailed original design of a catalytic coal gasification process, analogous to Exxon's catalytic gasification process for the production of high-BTU gas. The Hygas process was considered, but because it uses char to provide the necessary process heat, there is no demand for heat inputs from a nuclear reactor.

A detailed chemical process analysis and a preliminary design were made, based upon Exxon's published catalytic data, in order to determine its heat input requirements. Economic comparisons were made between the cost of providing these heat requirements from a High-Temperature Gas-Cooled Reactor (HTGR) and from a conventional coal-fired plant.

Using the data from the preliminary design of a catalyzed coal gasification plant given later in this report, it would appear that one 800 MW(th) (65,500 x 10^6 BTU/d) HTGR could service about one standard-sized 250 x 10^6 SCFD coal gasification plant. The quantity of coal feedstock input for one standard plant is about 10 to 12 thousand tons per day, and because of the escalating costs and difficulties of coal transportation, the natural sites for these plants would be at the mine-mouth, where nuclear energy would find it most difficult to compete with coal burning. Other problems are the coupling of two highly capital intensive plants, a gasification plant and an HTGR, and the questions of reliability and availability of the latter.

In the preliminary design for catalytic coal gasification only about 15% of the HTGR output is required for the high-temperature phase of the steam gasification; about 45% goes to generating electricity for the power requirements of the system, and the remainder provides steam for the low-temperature parts of the process. Thus there appears to be a good match between the division of energy requirements of the gasification plant and the capability of the nuclear reactor.

Because of the strong economics of scale in nuclear plants, an 800-MWt nuclear plant would not be competitive with an 800-MWt coal-fired plant. However, if the thermal requirement were provided from a 1500-MWt (461,000 x 10^6 BTU/d) nuclear plant, the cost of the heat input to the coal gasification plant would be 20% less expensive than a conventional coal-fired plant, and 5% below the cost of an atmospheric fluidized-bed coal plant. Therefore, cogeneration of electricity and gas is a natural consideration, taking advantage of both the economies of scale of nuclear plants, as well as using economically the medium-temperature heat (of the helium after it has passed through the gasifiers) for electricity generation.

The suitability of a nuclear plant to provide process heat for a US coal gasification plant does not appear promising in the near term, largely because of economics. More severe environmental standards (CO_2 concerns) or restrictions on the burning of fossil fuels for boilers could put nuclear power back more strongly into the picture. In Germany and Japan, however, the environmental problems of coal burning and the economics of coal make nuclear process heat a more attractive alternative.

Thermochemical Cycles

Thermochemical cycles are chemical processes that take water as the input and then, in several chemical steps, produce hydrogen and oxygen. The energy required to decompose the water is provided in heat inputs to the chemical process. This area was surveyed because of the potential for fission energy to provide the heat requirements.

There are three thermochemical cycles receiving major attention, two in the US (General Atomic and Westinghouse) and one in Europe (ISPRA). These cycles are now operating at laboratory-scale production rates of 4 cubic feet of hydrogen per hour. These experiments are so small that reliable estimates of efficiency and cost are not yet available, although efficiencies of 35-45% and hydrogen costs of $7-10 per million BTU are projected. Commercialization of any of these cycles would require development of materials that are corrosion resistant at high temperatures.

The other major technology contender for future hydrogen production is electrolysis. At present, the differences in efficiency and cost between thermochemical cycles and electrolysis are small, particularly in view of the uncertainties. With an increased level of National effort in these areas, a decision could be made in 10-15 years on which of the two technologies to pursue.

Several schemes for removal of heat from a fusion reactor chamber were analyzed. It is estimated that process heat will be available at temperatures in the range 1200 to 1700°C. At these temperatures, it may be possible to design two-step thermochemical cycles (i.e., involving only two chemical reactions) at considerable savings in capital cost and increased efficiency as compared with the lower temperature thermochemical cycles applicable to nuclear fission.

High-Temperature Electrolysis

Electrolysis is a process in which electricity is passed through a cell containing water or steam; hydrogen is produced at one end of the cell and oxygen at the other end. The electrolysis of water at room temperature to produce hydrogen is inefficient: generation of electricity from thermal energy is only 33% efficient; the electrolysis cell is 60-80% efficient, yielding an overall efficiency (thermal to hydrogen) of 20-26%. Development of more efficient electrolysis cells could raise this figure to 31%.

Electrolysis of high-temperature steam in porous materials is attractive, because at elevated temperatures part of the energy required to produce the hydrogen comes from heat rather than from electricity, and so a larger part of the thermal-to-electrical energy inefficiency is circumvented. For example, at 930°C (achievable with HTGRs), 73% of the energy investment is electrical and 27% is heat; at 1830°C (expected to be achievable with fusion), 55% is electrical and 45% is from heat.

The advantages gained by using superheated steam from an HTGR as part of the energy input to an electrolyzer are marginal at best. The small thermodynamic gains at practical reactor outlet temperatures (around 1000°C) will likely be offset by the higher cost electrolyzers capable of operating in severe environments. Developmental problems for electrolyzers at these temperatures are significant.

Radiolysis

Radiolysis is a process in which the products of nuclear reactions - neutrons, gamma rays, nuclear fragments - directly break molecules apart. For example, water can be decomposed into hydrogen and oxygen; or carbon dioxide can be decomposed into oxygen and carbon monoxide. Because of the inherently low efficiency of radiolysis (only 5-10% of the fission energy release can be utilized in radiolysis of water), any viable system would have to find an economic use for 90% of the reactor output energy, probably for electricity generation. The plant would then be an electricity generator with hydrogen as a very minor by-product. Systems optimized for maximizing radiolytic hydrogen production such as an aqueous homogeneous reactor would require a substantial developmental program. In all cases, severe safety problems related to hydrogen explosions might be encountered.

Production of CO from radiolysis of CO_2 appears to be more efficient than radiolytic hydrogen production, but there are many developmental problems. Key issues in this development, which would require Federal sponsorship, would be how to provide intimate contact between CO_2 and the nuclear fuel particles to use the fission product energy, fission product removal, and the corrosion of steel components by the CO_2 which also serves as a coolant for the reactor. To produce pipeline gas, the CO would have to be put through a shift conversion process with steam to form hydrogen.

Fusion-driven radiolysis is somewhat more attractive than fission-driven radiolysis because of engineering considerations and because radioactive contamination of the radiolyzed product can be maintained at a lower level. However, systems studies of fusion reactors have shown that radiolysis is not competitive with electrolysis. Further, even if a 30% CO_2 radiolysis efficiency could be achieved, radiolysis is not even viable as a topping cycle in electric power generation.

SURVEY OF FISSION ENERGY SOURCES FOR THE PRODUCTION OF SYNTHETIC GASEOUS FUELS

INTRODUCTION

Synthetic gaseous fuels can be produced with the aid of energy from nuclear fission sources in a variety of ways. Hydrogen can be produced from direct radiolytic dissociation of water in the cores of certain reactors, from electrolysis of water using electricity generated by nuclear plants, and from thermochemical cycles and coal gasification using the heat from high-temperature reactors. Synthetic methane can be produced from coal with the assistance of high-temperature reactors. The types of reactors that can be used for such fuel production and the viabilities of such production methods in terms of economics and time scales are surveyed and discussed in this section.

A. Current Status of the High-Temperature Gas-Cooled Reactor (HTGR)

The application of nuclear reactor systems for the production of synthetic gaseous fuels is limited by the temperature of the coolant at its exit from the reactor core. Figure 1 illustrates the range of exit coolant temperatures that occur with reactor systems that are currently being operated throughout the world. Since the process temperatures for production of synthetic gaseous fuels (e.g., coal gasification) discussed later in this report are in excess of 800°C, it can be concluded from Fig. 1 that our attention must be focused on the HTGR or very high-temperature reactor (VHTR) systems.

Germany, Japan, and the U.S. currently have active programs for development of high-temperature (coolant temperature approximately 750°C) and very high-temperature (coolant temperature in excess of 900°C) reactor systems. Although the technology programs in the three countries have many similarities, they also have strong dissimilarities either because of the reactor design or because of the process temperature requirement.

The U.S. has the only HTGR in operation at the present time, a steam cycle prismatic core HTGR manufactured by General Atomic Company. The Fort St. Vrain Nuclear Generating Station operated by Public Service Company of Colorado for electricity production began a slow and protracted ascent to power at the beginning of 1976. Numerous technical difficulties have been encountered since then, most of which have involved the design of the helium circulators and the seals between the steam turbine and the circulator. To compound the difficulties, a core fluctuation phenomenon has been observed that was unexpected and is still not fully understood. The fuctuations involve changes in the neutron detector channels, refueling region outlet thermocouple readings, temperatures in the steam generator modules, and prestressed concrete reactor vessel (PCRV) motion. Because of the unpredictable nature of the fluctuation phenomena, the Fort St. Vrain plant has been limited to 70 percent of its rated power of 330 MWe.

In probably what is a related development, the U.S. Department of Energy in October, 1978, recommended to General Atomic Company and the Gas-Cooled Reactor Associates (a consortium of utilities representing thermal gas reactor development) that support of the HTGR steam cycle be discontinued and that efforts be concentrated on advanced gas cooled reactor concepts. For example, a direct cycle HTGR to be operational in 1992 was discussed. Also in the picture appear to be process heat nuclear reactors. In addition, cooperation with the German effort was encouraged (see below). The relationship of the events of the past year in the area of gas-cooled reactors is not clear, but it now appears certain that the steam cycle HTGR will not be developed in the U.S. in the forseeable future.[1]

In the Federal Republic of Germany during the past year there has also been an effort to concentrate the gas-cooled reactor work.[2] Two basic reactor concepts, a direct cycle gas turbine for electricity production and a process heat system are being studied. Both concepts employ the pebble-bed type of reactor with a prestressed concrete reactor vessel. A 600 MWe demonstration plant is being proposed for electricity production. The system has an outlet gas temperature of 950°C and an overall efficiency of 44.5 percent. The plant could be on-line in 1992. A 500-kWt process heat plant is being planned for somewhat later introduction. Much relevant German experience was accrued during the operation for ten years of their AVR - a 50 MWt VHTR with an exit coolant temperature of 950°C, which should allow them to confidently design the larger reactor systems.

At the moment, the top priority in the German gas reactor program is the completion of the 300 MWe thorium high-temperature reactor (THTR) plant located at Schmehausen. Good progress has been reported and it is now hoped that the plant will be completed in 1981.

Japan has mounted a vigorous program for development of a VHTR to be used in steelmaking using nuclear process heat. The lack of a sufficient quantity of fossil fuels and a severe environmental pollution problem caused by the present steelmaking process have combined to push the development of a multipurpose HTGR which will supply the required heat for a direct-reduction steelmaking process. The work in reactor development is directed by the Japan Atomic Energy Research Institute (JAERI) and is predicated on a reactor exit helium temperature of 1000°C. Some in-pile subassembly tests dealing with VHTR fuel irradiation,fission product transport, and component integrity are already in progress in the OGL-1, a helium gas loop located at the Oarai Research Establishment. In addition, material tests (on Hastalloy steels and Inconel 617 steel) are ongoing in a prototypical intermediate stage heat exchanger that has been designed for a hot side temperature of 1000°C. The large scale of the Japanese test loops and experiments indicate the degree of commitment that Japan is giving to the development of a VHTR (spending on the VHTR program is estimated at $100 million/year). Finally, an experimental prismatic core VHTR with a thermal output of 50 MWt and a coolant exit temperature of 1000°C has been designed with plans to have the reactor system built by 1986.[3] The estimated cost is $500 million.

In summary, it appears that the use of HTGRs in a steam cycle for electricity production has lost the potential that it once had. In the future HTGRs will be used in those applications that are unique to the HTGR, such as process heat.

B. Radiolytic Production of Hydrogen

The chemical bonds that hold the water molecule together can be broken by ionizing radiation. The principal ionizing radiations in a fission reactor power plant are fission fragments, beta particles produced by fission product decay, gamma rays produced directly by fission and also by neutron capture and fission product decay, and neutrons. The last two ionize indirectly, gamma rays by interacting with atoms to produce high-energy electrons and neutrons by producing recoil ions when they are scattered. Each of these radiations can give up its energy in water by producing various excited and ionized states of the water molecule and its constituent atoms. The resulting decomposition of water can be expressed as the number of molecules of a primary species produced per 100 eV of energy absorbed, G(X), where X is the product species. For a given product species the value of G(X) is a function of the rate at which an ionizing radiation transfers energy. For the hydrogen molecule $G(H_2)$ in water has a value of 0.3 to 0.7 for gamma rays, 1.8 for fission fragments, and 1.5 for recoil ions.[4]

The basic reaction for water in a radiation field is

$$H_2O \leftrightarrows H^+ + OH^- \quad . \tag{1}$$

For densely ionizing radiations characterized by a high $G(H_2)$, the ions are driven to recombine, to some degree, as[5]

$$2H^+ + 2OH^- \leftrightarrows H_2 + H_2O_2 \quad . \tag{2}$$

Although extremely simplified, this model gives physical meaning to the radiolytic phenomena accompanying the production of molecular hydrogen.

In conventional water moderated and cooled nuclear reactors, radiolytic reactions occur to such an extent that precautions have been taken to suppress the reactions and thus to eliminate the possibility that an explosive mixture of H_2 would form. An estimate of the hydrogen production rate (in kg/day) is given by[6]

$$18PfG(H_2) \quad , \tag{3}$$

where P is the reactor thermal power (MW), f is the fraction of thermal energy available for radiolysis, and $G(H_2)$ is the average molecular production rate per 100 eV averaged over all the ionizing radiations. The values of f and $G(H_2)$ vary with reactor type.

In an aqueous homogeneous reactor where the fissile material is in solution with the aqueous moderator-coolant, f is close to unity and $G(H_2)$ is approximately 1.6. Ninety-six percent of the H_2 is produced by fission fragments and the remaining four percent is produced by neutrons, gamma rays, and beta particles. For conventional light water reactors, fission fragments and beta particles are isolated from the water by the nuclear fuel cladding and radiolysis occurs only by fast neutrons and gamma rays. This limits the value of f to about 0.04 (the fraction contributed by neutrons and gamma rays) and limits the value of $G(H_2)$ to approximately 0.6 for boiling water reactors and 1.0 for pressurized water reactors.[6] The potential production of molecular hydrogen from these three reactor types is summarized in Table I.

We can calculate the energy efficiency for hydrogen produced by radiolysis. Using a heating value of hydrogen of 325 BTU/SCF, a straightforward calculation gives an efficiency of about 5 percent for the optimal case of an aqueous homogeneous reactor. This implies that a reactor system designed solely for the radiolytic production of hydrogen is not feasible, and that even in the optimal system, hydrogen generation by radiolysis must be ancillary to the production of electricity.

Molecular decomposition of carbon dioxide by ionizing radiations is another possible means of production of hydrogen. In this case the basic chemical reaction is

$$CO_2 \rightarrow CO + 1/2\ O_2 \ , \qquad (4)$$

whereupon the water-gas shift reaction

$$CO + H_2O \rightarrow H_2 + CO_2 \qquad (5)$$

can be used for production of the hydrogen. Since CO_2 has been used as the coolant in the British and French gas-cooled reactors (the MAGNOX and AGR reactors), it might prove economical to optimize these reactors for CO production.

There is a considerable amount of experimental data on the radiolysis of CO_2 including radiations with and without fission fragments, for which a

G(CO) equal to 10 and a resulting high conversion efficiency (~ 30 percent) is claimed. For radiolysis with radiations other than fission fragments, a value of G(CO) equal to about 4 is achievable, which corresponds to a conversion efficiency of about 12 percent. Furthermore improved efficiencies for radiolysis of CO_2 have been observed at elevated temperatures or by the use of additives and scavengers. Conversion efficiencies of 30 percent deserve further consideration as a means of hydrogen production although even here electricity production would be the major product.

In a fission nuclear system designed to radiolyze CO_2, the CO_2 would also be used as a coolant to remove the heat from the core of the reactor. Safety considerations would probably require that the fission product fragments be retained within a cladding of nuclear fuel. This, however, prevents attainment of the higher radiolysis efficiencies associated with fission fragments. Thus, the practicalities of developing and licensing a CO_2 gas reactor would preclude optimization toward radiolytic production of CO.

In conclusion, the low efficiency inherent in the production of hydrogen by radiolysis coupled with the technological problems associated with the handling of hydrogen and those of corrosion enhanced by the presence of hydrogen or carbon dioxide would seem to preclude radiolysis as a viable means of hydrogen production.

C. Gas Core Reactors for High-Temperature Process Heat

General - It has been suggested recently that gaseous core reactors have several attractive applications in meeting future energy needs.[7] Uranium fuel in gaseous or plasma form permits operation at much higher temperatures than possible with conventional solid fueled nuclear reactors. Higher working fluid temperatures in general imply higher thermodynamic cycle efficiencies for advanced closed-cycle gas turbine driven electricity generators and magneto-hydrodynamic (MHD) power conversion systems for electricity production. Of course, higher working fluid temperatures, also allow high process heat temperatures, which make many photochemical and thermochemical processes attractive, such as hydrogen production by dissociation of hydrogeneous materials.

The Los Alamos Scientific Laboratory has recently completed a study of gaseous core power plants designed for low proliferation potential.[8]

Characteristics of gaseous core power plants, excluding the low proliferation feature, are described in Sec. 1 below. Sec. 2 and Sec. 3 briefly describe the two proposed core concepts, the mixed flow and plasma core designs, respectively, and Sec. 4 summarizes the drawbacks of these reactors as far as production of synthetic gaseous fuels. Section 5 discusses the photochemical production of synthetic gaseous fuels using a plasma core reactor.

1. General Characteristics of Gaseous Core Power Plants. Because the fuel is a gas there is no need to manufacture fuel elements and, therefore, no need to process them. Nor is there a need to shut down the reactor to replace fuel elements because fuel can be introduced continuously while the reactor is operating. The fission products are generated in and carried by the gas, offering the possibility of removing them as the gases circulate in the loop.

If the reactor is designed as a sustainer, converting thorium to ^{233}U with a breeding ratio of one - and if a fluid breeding blanket such as the one developed by the Oak Ridge National Laboratory in the Molten Salt Breeder Reactor (MSBR) is used - the newly born ^{233}U can be continuously processed and fed back into the reactor to maintain continuous operation. This allows one-time movement of fissile material. Only the initial charge of uranium would have to be brought into the plant and no additional shipments would be required. In addition, this continuous feed of newly formed fuel makes it unnecessary to keep excess fissile material in the structure to sustain operations for a period of time.

Heat transfer rates are high. Heat conduction is not utilized in the reactor. Energy is transferred either by convecting the gaseous fuel itself or by thermal radiation from the fuel plasma.

2. Mixed Flow Reactor. In the mixed flow reactor, UF_6 and helium gas are intimately mixed and injected into a cylindrical cavity. By establishing a vortex flow in the cavity, a cooler outside gas flow and a hotter interior gas flow can be established, which accomplishes the goals of withdrawing a high-temperature process gas from the interior flow and protecting the cavity wall by the outer bypass flow.

Figure 2 illustrates a cross section of the mixed flow reactor showing seven cavities, each of which is surrounded by a beryllium moderator, the salt breeder blanket, a graphite reflector, and a pressure vessel capable of holding the 100 atmosphere internal pressure. The system shown produces 200 MWt, with a 1225 K mixture of UF_6 and helium, and has been neutronically designed for an inventory of about 100 kg of fissile material.[8] The UF_6-helium mixture is passed through a heat exchanger and the heat is dumped to a secondary helium loop (with a 50-100 K temperature loss) that can either be used to drive a gas turbine or can be used in process heat applications.

3. Plasma Core Power Plants. The impetus for developing plasma core reactors is their higher operating temperatures (above $4700^{\circ}C$),thus making attractive applications in advanced closed cycle, gas-turbine-driven electricity generators and in photochemical or thermochemical processes for the production of hydrogen. Because, in plasma reactors, the fuel gas (gaseous uranium rather than UF_6) reaches temperatures exceeding $4700^{\circ}C$, an argon buffer gas is circulated in a confining vortex to keep the gaseous uranium from the walls. Essentially the same cavity design is used in the plasma core design as is used in the mixed flow reactor except that the argon and uranium gases are not premixed. Breeder blanket, graphite reflector, and pressure vessel are much the same as in the mixed flow reactor.

Neutronic designs of two plasma core reactors have been presented in Ref. 5.

4. Problems with Gaseous Core Nuclear Plants. Gaseous core fission reactors possess the necessary high-temperature capability for process heat applications and also embody many desirable features in a nuclear heat source: self-sustaining in fuel, continuous operation, low potential for diversion of fissile material. However, numerous technological problems must be overcome in the development of such a heat source, particularly for the high-temperature plasma core reactor. Some of the technological areas that must be studied include

a. Fluid mechanics and thermodynamics (both reactors) together with buffer confinement (plasma core reactor).
b. High-temperature fluorine corrosion (mixed flow reactor) and high-temperature uranium chemistry (plasma core reactor).
c. Processes for removing fission products and fission product transport in the fuel loops for both reactor systems.

d. Optical properties of materials (plasma core reactor).

e. Reactor dynamics (both reactors).

A study program leading to the development of gaseous core reactors has recently been curtailed by its sponsor, the National Aeronautics and Space Administration. The reason given was lack of support by other government agencies, such as the Department of Energy. Until adequate funding is supported by the Federal government, the solution of the technological problems listed above will be postponed into the future. Once these problems have been solved, gas core reactors might then be engineered for various applications in the production of synthetic gaseous fuels. At best, it appears that 30 to 50 years would be required if a vigorous program were instituted now.

5. Photochemical Production of Synthetic Fuels

Dissociation of water into hydrogen and oxygen, or carbon dioxide into carbon monoxide and oxygen, can be accomplished by photochemical means in which photons of sufficient energy can break the chemical bonds. Since the energy from a plasma core reactor operating at 4700°C would be transferred largely by radiation, the feasibility of using this reactor concept as a photochemical synthetic fuel plant has been investigated.

To photochemically dissociate water or carbon dioxide requires photon energies of at least 5.0 eV and 7.5 eV, respectively. The energies of the photons emitted from the core plasma will not all be at the same energy, but will have an energy distribution given by the blackbody radiation law (see Appendix 2-A). At 5000 K, the average photon energy is only 0.43 eV and the proportion of the photons having energies of 5.0 eV or larger (the dissociation threshold for water) is only 0.6×10^{-3}. Thus, in a 3000 MWt ($246,000 \times 10^6$ BTU/d) plasma core reactor, which emits a total of 4.4×10^{28} photons/s, only 2.6×10^{25} would be above the water dissociation threshold. If all of these photons split water molecules, the daily hydrogen production would amount to 15000 lbs/d. In terms of BTU output, it would take over 250 such reactors to match one standard high-BTU coal gasification plant which produces 250×10^6 CFD of methane. The situation would be worse for CO_2 dissociation because of the higher threshold energy (7.5 eV compared with 5.0 eV for water).

We conclude from the above analysis that the production of synthetic fuels from plasma core reactors via photochemical splitting of water or carbon dioxide is not commercially viable.

APPENDIX 2-A

Assuming a blackbody radiation spectrum, the distribution of photons over the frequency spectrum is proportional to

$$\frac{\nu^2}{e^{\frac{h\nu}{kT}} - 1} \quad .$$

In the case of the reaction for production of hydrogen

$$\frac{E}{kT} = \frac{h\nu_o}{kT} = \frac{5.0}{.43} = 11.6 \ .$$

The proportion of photons above this energy is calculated from

$$\frac{N}{N_o} = \frac{\displaystyle\int_{(h\nu_o)/(kT)}^{\infty} \frac{\nu^2 \, d\nu}{e^{n\nu/kT} - 1}}{\displaystyle\int_0^{\infty} \frac{\nu^2 \, d\nu}{e^{h\nu/kT} - 1}} \quad .$$

At $h\nu_o/kT = 11.6$ this becomes

$$\frac{N}{N_o} = .60 \times 10^{-3} \ .$$

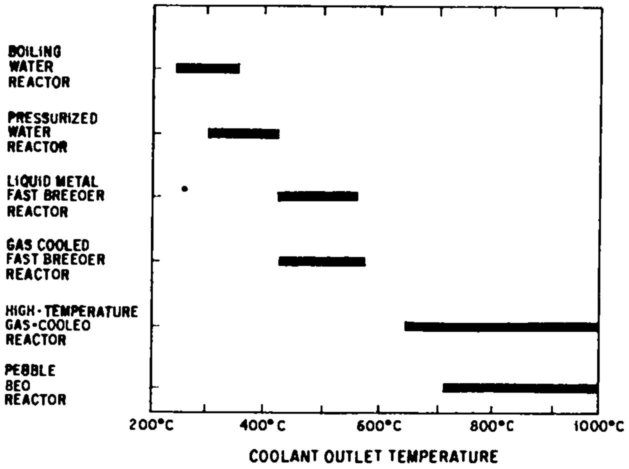


Fig. 1.

Coolant temperatures available from commercial and near-commercial power reactors.

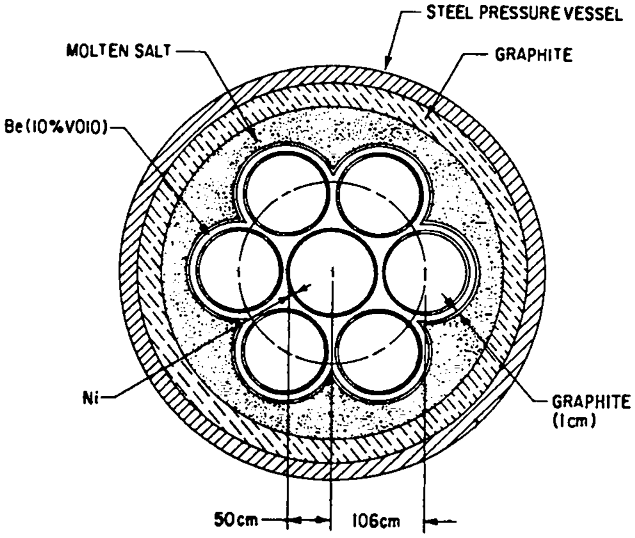
TABLE I

RADIOLYTIC PRODUCTION OF MOLECULAR HYDROGEN

Reactor Type	f	$G(H_2)$	H_2 Production in kg/MWt-day	H_2 Production for 1000 MWt reactor (10^6 SCF/day)
Pressurized water	0.04	0.6	0.4	0.16
Boiling water	0.04	1.0	0.7	0.28
Aqueous homogeneous	1.00	1.6	29	11.4

Fig. 2.

Mixed flow reactor cross section.



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DEVELOPMENT OF FUSION ENERGY - PRESENT AND FUTURE

INTRODUCTION

Thermonuclear fusion requires a process for supplying energy to fusion-fuel ions, such that the ions have sufficient kinetic energy to overcome electric repulsive forces, in order to fuse in exothermic reactions. Because the cross section for scattering is much greater than that for fusion reactions, this can only be done exoergically in a thermal plasma, thus fusion fuel must be heated to very high temperatures to initiate fusion reactions.

There are three principal fusion fuels based on the following reactions

$$\left.\begin{aligned} &D + D \rightarrow {}^{3}He + n + 3.27\ \text{MeV} \\ &D + D \rightarrow T + H + 4.03\ \text{MeV} \end{aligned}\right\} \quad \text{(50\% probability for either reaction)}$$

$$D + T \rightarrow {}^{4}He + n + 17.6\ \text{MeV}$$

$$T + T \rightarrow {}^{4}He + 2n + 11.33\ \text{MeV}$$

$$D + {}^{3}He \rightarrow {}^{4}He + H + 18.3\ \text{MeV}.$$

Deuterium is found in nature, the major source being heavy water (HDO) with a concentration of 0.015% in natural water, making this fuel source virtually inexhaustible. Tritium and helium-3 are produced by nuclear processes, including possible (D+D) fusion, but they do not occur naturally in significant quantities. From the aspect of resource availability, deuterium is the logical fuel choice. However, conditions required to achieve thermonuclear ignition are the primary consideration at this time, and these are related to the Maxwellian velocity-averaged reaction cross section $\overline{\sigma v}$. The (D+T) reaction is about 100 times more probable than the (D+D) reaction in the temperature range 10-100 keV (1 eV = 11600 K), and a given value of $\overline{\sigma v}$ can be achieved at a lower temperature for the (D+T) reaction than for other fusion fuels. Further the energy released from a (D+T) reaction is more than four times higher than that from its closest competitor for temperatures below ~20 keV. Thus (D+T) fuel is the leading contender for first generation fusion reactors.

Tritium is radioactive, with a relatively short half-life, and can be produced by reactions between neutrons and lithium. Fortunately, breeding of tritium is possible by placing lithium or lithium compounds in a blanket surrounding the fusion-reaction chamber.

Because the development of thermonuclear explosives in the early 1950's demonstrated the feasibility of producing energy from fusion, an international cooperative program was initiated in 1955 to discover and develop means of producing controlled thermonuclear energy releases, using magnetic forces to compress and heat thermonuclear material to ignition conditions, and to confine this material while it burns. Although much progress has been made in the understanding of the physics of fusion, the production of energy release in controlled thermonuclear reactions of sufficient magnitude for practical power production is yet to be demonstrated with magnetic confinement techniques.

Since about 1969 a new concept for achieving fusion has been under development--that of compressing, heating, and confining thermonuclear material by inertial forces generated by the phenomena subsequent to the interaction of an intense pulse of beam energy with a "pellet" containing the fusion fuel. As with magnetically confined methods, significant thermonuclear energy release for practical applications has yet to be demonstrated.

Below, we discuss the requirements that must be satisfied to make fusion energy commercially available when needed. We begin with a presentation of the current status of the two programs, including a discussion of research requirements, followed by a discussion of the technology development requirements, and conclude with a view of planning strategy requirements imposed on fusion by the dual nature of the program.

CURRENT PROGRAM STATUS

General

The largest fusion program in the world is supported by the US Department of Energy (DOE) at a level of $481 million in FY'79 for both magnetic and inertial confinement. This support has been supplemented by the Electric Power Research Institute (EPRI) at a level of 3 to 4 million dollars per year for system and applications studies. The second largest program is in the USSR at a level about two-thirds that of the US program. Information exchange between the US and USSR magnetic fusion programs is accomplished by a formal exchange agreement (no such agreement exists for inertial fusion information exchange). Smaller fusion programs are supported in Japan, the United Kingdom, and by the Organization for European Economic Development.

Because of the large cost of the US program, and because a first prototype reactor is not expected until after the end of this century, the DOE initiated a review process in early 1978. The six-month long review was directed by the Fusion Review Committee, comprising the DOE Assistant Secretary for Energy Technology, the Assistant Secretary for Defense Programs, and, as Chairman, the Director of Energy Research. The committee and an attendant working group scrutinized the program's objectives, strategies, and status; assessed the impacts of alternative budget scenarios, including some involving both small and large funding reductions; and examined the large experimental projects in cost and risk analysis. Also, an Ad Hoc Experts Group of distinguished scientists from outside the fusion community, which was chaired by John W. Foster of TRW, evaluated the status and prospects of the entire program.

From all of this examination and deliberation an extraordinarily high degree of consensus emerged. The most important conclusions were that the program is technically sound and that it holds excellent promise of ultimately achieving the goal of commercial fusion energy. Other important conclusions were: the United States now has world leadership in fusion energy research and development, and this lead should be maintained; the current level of funding was determined to be appropriate (decreases would, in fact, delay the date of commercial availability of fusion); and the general strategy for the solution of scientific and technological problems was upheld, with recommendations for some broadening and shifting of emphasis.

A paper[1] which explains in some detail the DOE policies governing current and future research and development in fusion energy was submitted to the Congress by John M. Deutch, Director of the Office of Energy Research, as part of testimony to the House Committee on Science and Technology. As stated in the Deutch paper, the objectives of the program are

"The goal of fusion research in the Department of Energy is to develop the highest potential for employment of fusion energy. The highest potential cannot be developed without an extensive technical base, both scientific and engineering. When choices are made, we must be confident that they are based on a firm understanding of all significant technical alternatives. This differs from a crash project, in which one strives to produce something that will work as soon as possible, subject only to a few minimal criteria of performance. The first device that would work at all might be far from fusion's highest potential, and the research that led to its construction would be narrowly focused."

The overall program strategy is stated as follows

"Thus, our philosophy in designing and managing this R&D program is to provide demonstration of that highest potential as soon as possible in an economically acceptable way that fully preserves and develops the technical base. This cannot be accomplished in a sudden plunge ahead, giving all or most resources to the leading prospect at some particular time. We must assure that fusion technology develops toward an economical and practical engineering system in a balanced progression and provides as wide a range of choices as possible throughout the progression."

With regard to the overall goal, he states

"Though most people in the program should look toward fusion power--that is, a pure fusion reactor--as its goal, we must consider other applications as well. The phrase fusion energy is meant to include fusion power, and also various hybrid fusion concepts: devices in which, for example, fusion reactions are used to breed fission fuels, or to produce other useful fuels, such as hydrogen."

Also, presented by Deutch, is an overall program plan shown in Fig. 1. This plan is based on the expectation of "scientific feasibility"* in the

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*Generally defined as greater thermonuclear energy output than driving energy invested in the process--driving energy is further defined as magnetic energy plus plasma heating for magnetic fusion (MFE) or beam energy for inertial confinement fusion (ICF).

early-80's for MFE and in the mid-80's for ICF. Upon achievement of scientific feasibility, a driver selection for MFE and ICF will be made for further R&D in Engineering Test Facilities (ETF). These facilities will be integrated systems producing net energy gain using fusion plasma techniques developed in the previous generation of experimental devices. The Engineering Test Facilities will also establish the technological requirements of each of the major components of a prototype reactor.

The next phase of the program, demonstration, will involve the operation of an Engineering Prototype Reactor (EPR), which first combines the elements tested in the superior Engineering Test Facility into a pilot plant in which the unknowns of reactor design can be tested and resolved. The EPR will approach, for the first time, complete energy gain, where the energy produced exceeds all energy consumed in keeping the entire plant running. Finally, demonstration will be completed with the construction of one or more commercial demonstration reactors, in which net power gain in excess of 100 MW per plant is produced with an economic efficiency that will make them attractive to industrial investors. Full commercialization of fusion energy will have been accomplished when about one tenth of a quad per year, which is about the equivalent of three 1,000 MW power plants, is produced.

Magnetic Fusion Program

The US magnetic fusion energy program is managed by the DOE Office of Fusion Energy and is organized into four interrelated subprograms: confinement systems, development and technology, applied plasma physics, and technical projects. The last of these is an office which supervises the construction of major facilities such as the Tokamak Test Fusion Reactor (TFTR).

The magnetic fusion program is carried out at five major sites: General Atomic Company, Lawrence Livermore Laboratory, Los Alamos Scientific Laboratory, Oak Ridge National Laboratory, and Princeton Plasma Physics Laboratory. Many smaller programs are also in progress at other laboratories, industries, and universities.

Confinement Systems

The confinement systems program is responsible for solving the experimental problems connected with the confinement of fusion plasma by magnetic fields; to demonstrate long-time confinement of high-temperature plasmas at power-producing reactor conditions and to optimize the plasma physics aspects of fusion reactor systems. The most important parameter for a fusion reactor is the plasma temperature and the so-called Lawson parameter, $n\tau$, where n is the number density of particles and τ is the confinement time. For a fusion reactor, the plasma temperature needs to be ~ 10 keV for both ions and electrons, and $n\tau$ must be of the order of 10^{15} s/cm^3.

The principal approach to the confinement of plasma is the tokamak which is a donut-shaped, long pulse time, moderate-density device. In addition, a strong effort is maintained in magnetic mirror systems, including both open and toroidally linked mirror systems. Smaller efforts are maintained in high-density short-pulsed systems, including the linear theta pinch, the toroidal Z-pinch, and imploding liner concepts.

Tokamaks - The major problem areas of the tokamak physics program are heating, transport and scaling, plasma shape optimization, impurity control and boundary effects, and fueling.

Heating refers to the process of producing the plasma temperatures necessary for a fusion reactor. Methods of heating include ohmic heating by inducing plasma currents with magnetic fields, induction heating by microwaves, and heating by injection of high-energy neutral particles.

Transport and scaling refers to the development of the physical laws which describe the measured transport of plasma energy in present experiments and the development of scaling laws to predict plasma behavior in larger, higher temperature devices. This area is, therefore, closely related to the heating program, and research on the two is conducted simultaneously.

This research has been carried simultaneously using the Alcator at the Massachusetts Institute of Technology, the ORMAK at Oak Ridge, T-10 in Moscow, and the PLT at Princeton. The experiments have been directed at understanding of the scaling of the $n\tau$ parameter, sometimes called the quality of confinement. This scaling in tokamaks depends fundamentally on four parameters: density, size, temperature, and magnetic field strength.

In early 1978, Alcator experiments indicated the unexpectedly favorable result that $n\tau$ increases as density squared over a range of several factors of ten and even beyond that required for a reactor.

With regard to size scaling, experiments without auxiliary plasma heating using the ORMAK, T-10, and PLT devices indicate $n\tau$ to scale as the square of the torus size, an expected result. Furthermore, these experiments have revealed no explicit dependence on magnetic field strength.

Recent experiments on PLT have indicated the dependence of $n\tau$ on temperature, the remaining parameter of concern. The most significant result from these experiments is that $n\tau$ does not decrease significantly at a temperature of ~ 6 keV, near that needed for a fusion reactor. This unexpected result is very favorable because it was previously believed that $n\tau$ would decrease markedly at this temperature due to plasma instabilities. This result has created confidence that the goal of "scientific feasibility" will be met with experiments on the TFTR.

Plasma shape optimization addresses the possibility, predicted by theory, that non-circular plasma shapes can be confined by lower strength magnetic fields and thus lead to lower fusion power plant costs.

Impurity control and boundary effects refers to problems resulting from the interaction of the plasma with its material boundaries. These interactions can result in an influx of non-hydrogenic (impurity) atoms into the plasma, which can cool the plasma core directly and/or can cool the plasma edge, causing the plasma to shrink and become unstable.

Mirrors - The mirror program consists of investigation of two configurations for ultimate steady-state operation: open systems, and toroidally linked mirrors. Experiments on open systems, currently considered the runner-up to tokamaks (conducted at Livermore), with the 2X-IIB "baseball" magnetic field configuration, have successfully demonstrated plasma scaling parameters, i.e., $n\tau \sim T^{3/2}$, at ion energies up to 13 keV and at an ion gyroradius, R/ $_i$, in the range of 2-3. Reactor conditions will require ion energies of $\geq$ 50 keV and $R/\rho_i > 40$. Experiments are planned for the MX device (operational in the early '80s) at ion energies of ~ 50 keV and R/ρ_i of ~ 13. Success with MX experiments will lead to tandem mirror experiments (TMX), in which a linear system of mirror configurations will be built using baseball coils to minimize end loss. This latter configuration would provide high enough energy gain for the open mirror concept to be economically viable.

The toroidally linked mirror concept, called the Elmo Bumpy Torus (EBT), is being studied at Oak Ridge. The major problem areas for EBT are plasma stability and microwave heating. Plasma stability has been demonstrated at densities up to $\sim 6 \times 10^{12}/\text{cm}^3$ at a microwave frequency of 28 GHz. However, microwave heating at ~ 100 GHz and at densities of $\sim 10^{14}/\text{cm}^3$ needs to be demonstrated for reactor conditions.

High-density systems - Experiments on these systems are conducted at Los Alamos. These systems include the linear theta pinch, z-pinch, and imploding liner concepts. Work has been abandoned on the toroidal theta pinch because of demonstrated plasma instabilities.

Linear theta pinch experiments are being conducted on the SCYLLA IV device. The major problems are end loss and high field operation. Without some form of end-stoppering, a fusion reactor based on this concept would be impractically long (many kilometers). A variety of methods are under investigation. High field operation is essential because reactor length varies as the inverse of the magnetic field strength squared, therefore the reactor length decreases dramatically at higher magnetic fields.

Z-pinch experiments on the ZT-1 device have indicated plasma stability and the potential for reaching ignition conditions by joule and shock heating. Experiments planned for the ZT-40 device will determine the validity of this potential.

The imploding liner concept offers the highest potential of achieving ignition without plasma instabilities among the various pulsed magnetically driven systems. Experiments are underway to establish the feasibility of this concept.

Development and Technology

The development and technology program provides both near-term engineering/subsystems support to existing and proposed experiments and longer term development of the necessary technology base to permit fusion energy to become a commercial reality. Development and technology program activities presently are organized in five related subprograms: magnetic systems, plasma engineering, reactor materials, systems engineering, and environment and safety.

Magnetic systems sponsors research and development of large superconducting magnet systems needed for fusion reactor engineering experiments within the next ten years. Plasma engineering is directed principally at the development of efficient plasma heating systems (neutral particle beams, radio-frequency waves, and electromagnetic plasma implosion systems) that are essential for the various confinement concepts.

The reactor materials activity is assigned the responsibility to develop (or invent) the materials required to permit the economical generation of energy from the fusion process. The principal focus is on materials that will be placed within the first ten centimeters or so of the plasma where the fusion radiation environment imposes the most difficult materials requirements.

Systems engineering focuses principally on the next generation and longer term fusion power reactor designs. Specifically, a major responsibility is to support the reactor designs necessary for Congressional approval (and funding) to build the first large fusion prototype experimental power reactor.

Environment and safety is charged with the responsibility of assuring that fusion power reactors will operate with the minimum possible hazard either to the environment or to plant personnel and nearby populations.

Applied Plasma Physics

The applied plasma physics program seeks the body of knowledge that predicts the behavior of fusion plasma confinement experiments and the operating characteristics of fusion power reactors. The program is composed of theoretical activities, including its computational components, and experimental plasma research, which supports a broad spectrum of experiments to attack problems related to the production and confinement properties of fusion plasma.

Tokamak Fusion Test Reactor

The Tokamak Fusion Test Reactor (TFTR) will be the first magnetic confinement fusion device to experimentally demonstrate the release of fusion energy from the deuterium-tritium reaction under conditions projected for future experimental power reactors. TFTR will represent an intermediate step between present, relatively small zero-power physics experiments and future experimental reactors. The TFTR will be located at the Princeton Plasma Physics Laboratory (PPPL). The project should be completed in mid-1981 at a total cost of $228 million.

The TFTR has major objectives in both physics and engineering. The principal objectives are

- To demonstrate fusion energy production from the burning of deuterium and tritium (DT) in a magnetically confined toroidal plasma system.
- To build a neutral beam heated tokamak in which hydrogen, deuterium, and DT plasma can be inserted in order to
  - study the physics of large tokamaks, and
  - verify advanced engineering concepts for DT tokamak systems.
- To experimentally demonstrate physics and engineering understanding of large fusion systems.

The unique features of the TFTR are its DT burning capability, its size which permits physics experiments in the reactor range of interest, and some of its engineering features, not heretofore tested. The experience to be gained in design, construction, and operation, and the information to be gathered in physics and engineering will provide a sound foundation for potential larger systems.

The specific objectives of the TFTR project are

- Attain reasonably pure plasma conditions at 5-10 keV temperature, approximately 10^{14} cm^{-3} density, and provide stable confinement with n equal to or greater than 10^{13} s/cm^3.
- Provide a neutral beam injection system capable of injecting into the plasma 20 MW of a 120-keV neutral deuterium beam, for at least 0.5 s.
- Provide a toroidal magnetic field of about 5 tesla (50 kG) (on vacuum chamber axis), for at least 3 s flattop time, with a 5 minute repetition rate.
- Develop plasma handling techniques and provide hardware capable of initiation, control (including feedback control and major radius compression), and dissipation of tokamak discharges up to 2.5 MA.
- Provide a vacuum chamber of adequate size (2.7-m major radius and 1.1-m minor radius), equipped for high-power discharge cleaning and capable of achieving base pressures below 5 x 10^{-8} torr.
- Provide capability for routine pulsed operation with H-H; D-D; D-He^3; or DT plasmas, with safe and reliable gas handling and support systems.

Inertial Confinement Fusion Program

The US inertial confinement fusion program is managed by the DOE Office of Laser Fusion and is conducted principally at the Lawrence Livermore Laboratory (LLL), the Los Alamos Scientific Laboratory (LASL), and the Sandia Laboratories at Albuquerque (SLA). Smaller research programs are conducted at KMS Fusion, Inc. and at the University of Rochester.

Consistent with the research nature of the current driver development and pellet-design phases of the program, the strategy is based on parallel investigations considering two ignition sources; these are lasers and electron beams. High-energy heavy-ion beams are a third possible ignition source, but they are not discussed here because of their present immaturity. Different ignition sources operate in different regimes of physical parameter space. Different energy-matter interactions are involved, each requiring somewhat different pellet designs.

The research requirements for the successful development of ICF may be grouped into the following three areas: (1) improved understanding of radiation-matter interactions at very high densities of energy and matter, (2) identification of a fusion ignition source (driver), (3) determination of a fuel pellet design with sufficient gain that, when coupled with driver efficiency, will result in competitive production of energy.

Lasers - The Nd:glass laser is the first-generation research photon-beam source used in most target irradiation facilities around the world. Since 1967 Nd:glass laser systems have been developed to high power levels. Experiments with the Livermore Argus system at powers up to 4.0 TW have resulted in neutron outputs greater than 10^9 from DT fusion targets. Neutron output (of $\sim 10^5$ neutrons) was first reported in 1973 by KMS Fusion, Inc., using a split-beam 0.3-TW laser system. Hundreds of experiments with this system and with Livermore's JANUS (up to ~ 1.0 TW) and ARGUS systems have verified results predicted by the simulation code LASNEX within this power range.

Early in 1978, the 10-kJ SHIVA Nd:glass system became operational at LLL. Experiments are underway at powers > 20 TW. Current plans are to construct a 100-kJ (100-200 TW) system called NOVA I. It is expected that breakeven gain, defined as thermonuclear output equal or greater than impinging beam energy, can be accomplished with this system.

Although the Nd:glass laser is capable of providing light pulses of specified high intensity for these initial studies, these laser systems require large investments in optical components and in large glass amplifiers. Glass lasers are inherently limited to a maximum efficiency of a few tenths of a percent and cannot be operated at high repetition rates. These features, along with uneconomical power-scaling constraints, make these systems unsuitable for commercial applications.

For ultimate commercial applications, a gas laser, in which the lasing medium can be circulated to remove waste heat, is considered essential for high-repetition rate operation.

The CO_2 gas laser is currently best developed in this class of lasers. Considered a second-generation laser for fusion research, its development has proceeded rapidly at LASL since 1969 with the invention of the electron-beam-controlled, electric-discharge pumping technique for high efficiency, short-pulse energy extraction. Development has produced an eight-beam, 10- to 20-TW target facility, called HELIOS, for fusion-pellet physics research at LASL. Target experiments are underway with this system. Also, construction is underway for a 100-kJ, 100-200-TW CO_2 laser system, called ANTARES, at LASL, with the goal of achieving breakeven gain.

The target physics program has emphasized the understanding of laser-beam target-interaction, and plasma physics with 10.6-µm radiation to address the uncertainties in radiation-coupling and compression efficiency as a function of wavelength. Results from experiments (1976-1977) with a single-beam 0.2-TW laser have indicated essentially no wavelength dependence, originally expected from classical theory based on critical-density/absorption considerations. For example, measurement of hot-electron temperatures at intensities of $5x10^{13}$ W/cm^2 for both 10.6-µm and 1.06-µm radiation-matter interactions imply a hot-electron temperature vs wavelength dependence proportional to the square root rather than the square of wavelength as predicted by classical theory. This phenomenon is theoretically predicted by inclusion of forces as a result of electric field gradients (ponderomotive force) present at the higher beam intensities. Similar results have been reported from experiments in ICF programs at CEA Limeil (France) and Osaka (Japan).

Results from experiments with a two-beam prototype module for LASL's HELIOS system at powers approaching 1.0 TW have further confirmed the absence of wavelength dependence with neutron yields within the same range of 1.06-μm results as a function of beam power. Although the range of power levels in these experiments is too limited for an accurate verification of power dependence, the results are encouraging and lend credence to the possibility that fusion targets might be designed without regard to wavelength. The results from experiments with the ANTARES system will definitively verify the suitability of the CO_2 laser.

Should the CO_2 laser prove to be uneconomical for energy production, a new advanced laser must be identified whose laser medium can be circulated to remove waste heat. This laser will necessarily be a gas laser and will probably be pumped electrically (electron-beam-controlled discharge or relativistic electron beam). Requirements for the so-called advanced laser include demonstration of saturated pulse output at the proper width, successful target experiments at 10% of the required intensity for breakeven gain, scalability to power levels of about 100 TW per beam, repetition rates of 1 Hz or faster, and an efficiency of at least a few percent. It is doubtful that such a laser can be developed within the time scale specified in Deutch's overall plan (Fig. 1).

Electron beams - Relativistic electron beams (REBs) are an alternative to lasers for initiating fusion-pellet microexplosions. Electron beam accelerators developed at Sandia are simple, efficient, and inexpensive compared to high-power laser systems. However, electron beams can be focused adequately for pellet initiation only if either the electrodes or the clouds of plasma or metal vapors are in contact with the pellet. Conceptual approaches to plasma production in electron-beam diodes have been suggested, but further research and design studies will be required to ensure that pellet microexplosions can be isolated to prevent damage to electron-beam pulse-forming lines and cathodes. Light and heavy ion beams are also potential candidates, and a modest effort is being expended on these backup possibilities.

TECHNOLOGICAL REQUIREMENTS

General

Although current programs are still directed toward demonstrating scientific feasibility, confidence in understanding the fundamentals involved has led to studies of conceptual commercial plant designs and to the initiation of technology development programs. On the basis of these conceptual studies, the thermal energy system is logically divided into four major subsystems: (1) the fusion reactor (e.g., plasma cavity, blanket); (2) the plasma-confinement driver system; (3) the lithium-tritium processing and fuel-cycle system; and (4) the heat transfer system, to extract heat from the reactor and to convert this energy to a usable form. Of these subsystems only the heat-transfer system and the tritium extraction systems have common features among the various plasma-confinement schemes. Major differences in the reactor and plasma-confinement driver systems exist between magnetic- and inertial-confinement concepts.

Heat Transfer System

Lithium has been proposed as the primary blanket coolant in many fusion reactor system studies and helium or boiling potassium has been proposed for cooling solid lithium-compound blankets such as LiAl, $LiAlO_2$, or Li_2O. Although little experience exists in lithium heat-exchanger design, the heat-transfer properties of lithium are similar to sodium for which experience in heat-exchanger design has been gained in the Liquid Metal Fast Breeder Reactor Program. In liquid-lithium blankets temperatures may be restricted to a maximum of ~ 750 K because of corrosion of stainless-steel-containing materials. However, refractory blankets should operate at higher temperatures, permitting temperatures as high as 1500 K.

In principle, direct conversion of fusion energy to a useful product in fusion reactors operating at extremely high reaction temperatures (in the keV range) and, particularly, in magnetically confined systems where the hot plasma does not contact any containing materials, should be possible at high efficiency. Unfortunately, both magnetic and inertial fusion reactor schemes are based on the (D+T) reaction, where the major fraction (0.8) of the total energy released (in a neutron) must be converted to thermal energy, and this conversion must be accomplished in a blanket containing lithium for tritium breeding. Here, the maximum temperature of the heat-transfer medium is limited by the physical and chemical properties of blanket materials. Therefore, the major fraction of the fusion energy is most conveniently converted to thermal energy, leaving only ~ 20% for direct conversion. For reactor concepts where this direct-conversion process is a natural consequence of the fusion process (i.e., no special costly equipment is required), such direct conversion has a significant, positive impact upon the overall energy balance.

Although direct conversion of only 20% of the (D+T) fusion energy generally may be economically marginal, a higher fraction of the fusion energy in the form of ionized plasma (such as that resulting from the (D+D) reaction or from the (D+^{3}He) reaction, in which a larger fraction of the energy appears as an ionized plasma) could make direct-conversion methods more attractive. However, in addition to technological difficulties associated with achieving the higher plasma temperatures required by these "advanced fuels", the radiation associated with these higher temperatures will limit the fraction of direct-conversion energy to 60% or less. Some form of direct conversion, however, will be used whenever it is either a direct result of the confinement scheme or where it is necessary to efficiently make up for large plasma energy losses.

Lithium-Tritium Processing and Fuel Cycle

Circulating liquid lithium is included in the blanket regions of most conceptual fusion reactors for the breeding of tritium and the removal of heat. The nuclear breeding of tritium in lithium-containing blankets that surround the burning (D+T) plasma can occur in a variety of chemical and physical configurations.[2] Two broad categories can be identified, depending on whether the lithium serves the active function of coolant or whether the lithium function is purely passive. Liquid-metal or molten-salt (FLiBe, nitrates) forms of lithium have been proposed for the former, whereas solid lithium compounds (ceramics, metal alloys, or combinations thereof) have been proposed for the latter.

Tritium recovery from the breeding medium depends on the active or passive mode described above. If passive (solid) breeders are used,[3] the tritium must be capable of diffusing to the flowing primary coolant (e.g., from which it could be separated by chemical oxidation, absorption, and reduction. Removal of the solid breeder material followed by external processing has also been proposed.[4] In the active mode, in which the bred tritium is contained in a lithium-bearing primary coolant, a variety of physical chemical separation techniques have been proposed.[5,6] The viability of a given separation method hinges primarily on the fraction of primary coolant flow that must be diverted to a side stream (processing system) and the separation efficiency (i.e., the tritium inventory entrained and actively circulating in the primary coolant).

Various methods have been proposed for separation of tritium from the liquid-lithium blanket, including (1) tritium diffusion through a semipermeable metal membrane, (2) cold trapping, (3) distillation or evaporation, (4) use of a solid sorbent, (5) gas sparging, and (6) liquid-lithium extraction.[7,8] Diffusion through metal membranes and liquid-liquid extraction seem the most promising possibilities. Multiple layers of permeable materials may be used in combination with chemical methods of removal (e.g., reaction in oxygen) to reduce the requirement for a very high vacuum otherwise needed in the first method.

The method by which (D+T) fuel is injected varies with the fusion concept. In inertial confinement reactors fuel is injected in the form of pellets, whereas in pulsed magnetically confined systems (theta- or Z-pinches)[9,10] gas-puff injection of appropriate mixtures of (D+T) is envisaged. The quasi-steady-state (tokamaks) and steady-state (mirrors) approaches will require more complex fuel injection schemes, involving either the injection of energetic neutral particles (~100 to 200 keV)[11] or of accelerated micropellets (few tens of microns in diameter at ~10^4 to 10^5 cm/s)[12] so that the fuel may penetrate to the core of the thermonuclear plasma.

For inertial confinement systems the fusion pellets may be fabricated either locally (cavity-coupled) or remotely, by batch or continuous processes. The processing method would be determined largely by the selection of pellet materials and design. While the number of pellets required for operation of a large central generating station (~2.5 million per day, in a

1000-MWe plant using 100-MJ yield pellets) suggest continuous operation, large-scale batch manufacture may be preferred for some fuel-pellet designs. Remote and/or batch fabrication of pellets would require larger storage capacity than local continuous production.

Accelerated, high-velocity injection may be required to meet pulse-rate requirements and to improve trajectory stability in inertial confinement systems. Mechanical, pneumatic, or electrostatic methods could be used to obtain high pellet velocities. The choice of method depends largely on pellet design and materials, and on injection requirements.

The recovery of tritium and deuterium from unburned fuel in the reactor cavity appears to present less severe problems than recovery from the breeding medium within the blanket. For inertial-confinement concepts, in which the pellets may contain other materials than tritium and deuterium, separate cleanup loops will probably be required to remove higher-Z materials. Watson[13] has suggested the use of parallel cryosorption pumps which allow recovery of the cavity gases without adding impurities. Commercial pumps that meet the requirements are available at reasonable costs (assuming no size scaling problems exist).

The remaining fuel-fabrication sequence of operations is the chemical purification of the tritium followed by liquefaction and cryogenic purification to produce liquid T_2 and DT. This mixture can then be either adjusted stoichiometrically by cryogenic distillation or with the addition of deuterium as required. The stoichiometric mixture of deuterium and tritium is then transported to the fuel preparation system.

Uncertainty exists with respect to the physical chemistry of tritium separation from certain breeding media, although these uncertainties may indicate higher tritium inventories rather than critical technology development areas. The separation and purification of the fuel gases should present no serious technological problems.

Fusion Reactors

The presently envisaged commercial applications of fusion are electric power generation, fissile fuel breeding, and neutron and/or heat generation for non-electrical applications (e.g., hydrogen production). For all applications, however, the reactor will be designed around the energy source; that is, the magnetically confined plasma or the beam-driven pellet microexplosion. Thus, it is necessary to consider the characteristic of the energy releases and their effect on the reactor vessel.

For magnetic systems, the fast (14 MeV) neutrons pass through the first wall of the vessel relatively unimpeded and interact with the blanket material, the major design consideration being the effect of neutron damage to the wall material. Because the plasma does not contact the wall, damage by energetic ions does not occur; however the brehmsstralung radiation from the plasma heats the wall material, requiring cooling of the first wall. The major engineering design constraints, however, are dictated by the geometry and plasma scaling parameters of the particular concept and by the presence of very high magnetic fields within the blanket. For example, the low aspect ratio toroidal geometry of the tokamak poses very difficult engineering design problems for this concept. Several conceptual reactor designs have been proposed and are discussed elsewhere.[14,17]

In inertial confinement reactors, the energy is released in the form of (a) fast (14-MeV) neutrons, (b) x rays, and (c) energetic ions comprising pellet debris. The neutrons pass through the first wall in the same manner as in magnetic systems; the x rays and pellet debris must be stopped by the first wall of the reactor vessel. Thus, the first technological requirement for commercialization of ICF is the development of materials for the construction of reactor vessels that will: (a) resist erosion by energetic ions (sputtering), (b) minimize evaporation caused by x-ray and debris impact heating, (c) tolerate cyclic thermal and mechanical stresses, (d) resist neutron damage and activation, and (e) be chemically compatible with lithium or its compounds at elevated temperatures.

In the case of electron-beam drivers, the much higher gas pressure in the vessel (~100 torr) is sufficient to prevent target debris and x rays from impinging on the vessel wall. The overpressure in the gas produced by absorbing this energy is not a severe problem in reactor-size vessels. The neutron damage to the vessel walls and blanket structures is not appreciably different between that of particle-beam and laser-driven systems.

The several approaches that have been proposed to mitigate the loading pulses and extend the lifetime of the first wall have been discussed in detail elsewhere[18-21] and will not be repeated here. Note, however, that the exact specification of the first wall design requirements will be determined to a large extent by the details of the fuel pellet structure, which will be determined by the outcome of investigations in the areas of driver development and driver-pellet energy coupling.

Driver Systems

Most approaches to controlled thermonuclear fusion power require the pulsed transfer of considerable quantities of energy for the purposes of heating and confining the (D+T) plasma, the actual transfer rate depending on the specific fusion scheme being considered. Generally, this transferred energy is sufficient to dominate, or at least to affect significantly, the fusion reactor energy balance.[18,22,23] Plasma confinement driver systems for fusion reactors are more generically characterized between magnetic- and inertial-confinement schemes; however, even among these generic categories, significant differences in power, pulse transfer times, and energy storage requirements exist, as shown in Table I.[24]

Among the representative magnetic-confinement schemes, the tokamak and mirror-reactor magnets are powered by steady-state dc supplies at voltages of tens of kilovolts. Because of excessive ohmic heating losses in conventional magnets, the magnets for these reactors will necessarily be superconducting. A fast-rising magnetic field (~1. T in ~1 μs) would require 200 MJ to implosion-heat a theta-pinch plasma to a temperature where adiabatic compression will be more effective.[9,16] Imploding-liner concepts[25] will require 1 to 10 GJ in 1 to 100 μs to electromagnetically drive metallic shells onto preheated plasma at velocities of 10^4 to 10^6 cm/s. Power supplies for these fast-pulsed needs will more than likely require a significant amount of capacitive energy storage. The adiabatic compression of an implosion-heated theta pinch[9,16] will require ~60 GJ of magnetic energy to be transferred in tens of milliseconds, whereas the ohmic heating supply used to preheat a tokamak reactor plasma[14] must deliver ~10 GJ in a few seconds. The toroidal field in a tokamak power reactor must be generated by superconducting coils and alone represents ~200 GJ of stored energy. The Z-pinch reactor[10] will require ~10 GJ for the ohmic heating current (and confining fields) to be transferred in 10 to 100 ms. Similar energy requirements are envisaged for long linear confinement systems[26-28] (theta-pinches, relativistic electron-beam-heated or laser-heated solenoids). For these requirements, nonconventional (i.e., high repetition rate, higher energy density, higher voltages) rotating machinery appears to be the most promising solution.[29,30]

In addition to the above mentioned needs, magnetically confined fusion reactors will require energy for supplemental heating and for feedback control of the plasma equilibrium and stability. For instance, neutron beam injectors at ~100 MW may be needed by tokamak[3] and (steady-state) mirror reactors.[15] The fast feedback power for the theta pinch[19,31] amounts to ~1 GW of reactive power during the pulsed burn, and would be supplied from a high-Q capacitive supply. Some tokamak reactor designs[14] propose magneto-acoustic heating of the ohmically preheated plasma, which would require ~50 MW at 60 Hz. The proposed steady-state operation of the Elmo Bumpy Torus Reactor[32] will require 200 to 400 MW of 100-GHz rf power, and ~100 GJ of magnetic energy would be stored in the steady-state superconducting magnets.

The fundamental requirements on the driver system for inertial confinement are established by the characteristic of fusion pellets. For laser fusion, the basic pellet-determined criteria are: (1) pulse intensity, (2) pulse duration, (3) wavelength, and (4) spatial and temporal pulse shape. For electron beam pellet fusion the electron beam energy is analogous to the wavelength criterion (and generally is not as important). A second set of criteria is determined by the energy balance and the economics in an energy production system: (1) net driver (laser or electron beam) efficiency, (2) pulse repetition rate, (3) costs (capital and operating), and (4) reliability and mean lifetime of components (especially power supplies and switches).

The most demanding requirement for lasers is the generation of high-energy pulses of a nanosecond or less duration, which requires the achievement of the inverted population state nearly simultaneously throughout the lasing medium. Several types of laser systems are being studied in laser fusion programs throughout the world.[33] These systems differ in their physical approach to produce population inversion in the respective lasing media. Laser development is advancing rapidly, and it is premature to predict the specific laser type that may ultimately be most advantageous for application in laser-fusion systems; however the lasing medium will necessarily be a gas for ease of waste-heat removal in high-repetition-rate operation and will probably be pumped electrically. Among prospective lasers, the electron beam-stabilized, electric-discharge-pumped CO_2 laser is the only one of sufficiently high efficiency and advanced stage of development to be presently considered for commercial applications. Pumping requirements for the CO_2 laser dictate a square-wave pulse of 300 to 500 kV and a duration of 2 μs. In current experimental systems, this requirement is fulfilled with a Marx-Guillemin pulse-forming network.

Electron beam machine technology is at present further advanced than laser technology. Beam energies of several tens of kilojoules at powers of tens of terawatts are attained, whereas the maximum operational CO_2 laser energy is ~10 kJ at a power of ~ 20 TW. Electron beam accelerators have the inherent advantage of higher electrical input to beam energy efficiency (up to 50%), but the electron-beam target (fuel pellet) is necessarily a part of the anode, creating more difficult engineering problems in designing the reactor and beam-transport system than in laser fusion concepts. Current electron-beam pulser designs include a pulse forming line for temporal pulse compression, and are driven by conventional Marx generators.[34,35]

The design of a suitable power supply for the driver of the ICF reactor offers a major challenge for the technology developments mainly because of the need to switch hundreds of kilovolts with an efficiency higher than 90% several times per second for many years.

In the current technology, the spark-gap switches have a lifetime limited to 10^5 to 10^6 pulses (less than one day's operation at 10 to 50 pulses per second), and the capacitors have a lifetime of 10^7 to 10^8 pulses (approximately one month at the required frequency of operation).

Therefore, the lifetimes of spark-gap switches and capacitors must be extended more than 100-fold. These goals may be achieved by either: (a) derating the presently available components to operation at lower voltages at the expense of size and cost, or (b) by investigating the potential advantages of semiconductors or ignition pulse transformers in place of spark-gap switches and identifying a new energy storage system in place of capacitors and inductors.

In summary, a wide range of uses, delivery times, and energy levels required to initiate, sustain, and confine the (D+T) reaction is envisioned. This energy must be carefully managed to ensure an economically acceptable energy balance. The size of present fusion experiments permits the exclusive use of capacitor banks and/or conversional rotating machinery to supply the energy. However, the efficiency of present power supply systems (for theta-pinch, laser, and electron beam systems) is less than 90% with capacitor lifetimes of the order of 10^7 pulses and switch lifetimes of the order of 10^5 pulses. The degree of reversibility required, the capital cost, and the ultimate technological feasibility will also play a major role in the selection of the energy storage and transfer system used for a particular function in establishing and maintaining thermonuclear energy for the production of economical energy forms.

CONCLUSIONS

The development of fusion into a technically and economically attractive energy source faces unique and challenging problems that have not been encountered in previous research and/or technology oriented programs. These problems are caused by the dual nature of the fusion program: It is simultaneously a research and a technology development program, and it is expected to succeed soon enough to meet national energy needs.

In general, research programs are established to acquire knowledge and understanding that may be useful in future applications, but economic and/or operational plans are not made to depend on unpredictable outcomes of scientific investigations. Plans and actions crucial to national well-being, therefore, prudently depend on the results of technology development programs for which scientific principles and feasibility are well established and whose success can be assured by the proper organization and implementation of the effort. In such programs a schedule of milestones can be established with reasonable certainty; consequently, a systematic and orderly integration of the results into the economy can be planned and carried out.

The successful commercialization of fusion, however, depends on the accomplishment of both the necessary scientific advances and the required technology developments within a fairly well specified time interval. The interval during which fusion must be made available extends from the time when depletion has increased the price of nonrenewable fossil fuels sufficiently to make fusion economically competitive, to the time when alternative energy sources (e.g., breeders) must be introduced to prevent economic collapse and political chaos. Results of many studies projecting future energy supplies and demands indicate that the "window" for the commercialization of fusion lies between the first and the second or third decade of the next century.

Because 20 to 30 years are needed to bring a major technology through research, development, and demonstration stages, and another 20 to 30 years are necessary to introduce such a technology into the economy, the requirements for commercial availability of fusion are being identified now so that the research and development efforts are properly emphasized and coordinated.

Because a new energy source will be urgently needed, and because of the large effort associated with its introduction and integration into the national economy, it is necessary to carefully plan the commercialization of fusion. However, the program is currently research-based, making the successful attainment of its goal depend more on the advances in understanding of yet unexplored areas of physics than on coordination of technology developments.

Therefore, to ensure success, i.e., to minimize the possibility of failure, the program strategy is based on the following principles:

- Investigate several approaches in parallel, with provisions to change emphasis without excessively perturbing the overall program when the results indicate the utility of such action;
- Establish milestones leading to an understanding of basic phenomena that opens the paths to further progress beyond special, specific, nongeneralizable results, and avoid committment to expensive experiments that may produce nonscalable (or unfavorably scalable) results and thus not lead to practical applications;
- Establish and maintain interactions and compatibility between the research and technology development phases of the program for their mutual benefit; and

- Establish intermediate objectives along the path to commercialization that will be realized and will have practical value before the ultimate goals are attained.

In summary, we conclude the following regarding the present and future status of fusion:

- Current status of the fusion program is research-based in the sense that its success depends more on results of basic investigations in unexplored areas of physics (e.g., radiation matter interaction at high energy and matter densities) than on coordination of technology developments;
- Generic and specific reactor designs have been and will continue to be investigated in sufficient detail to determine the technology requirements for their commercialization;
- Technology development requirements are challenging, some requiring long lead times; and
- For successful commercialization of fusion, the long lead time technology developments can and should be compatibly integrated into the program in parallel with the research efforts; however, the earliest date for significant penetration in the commercial market lies in 2030 to 2050 time frame.

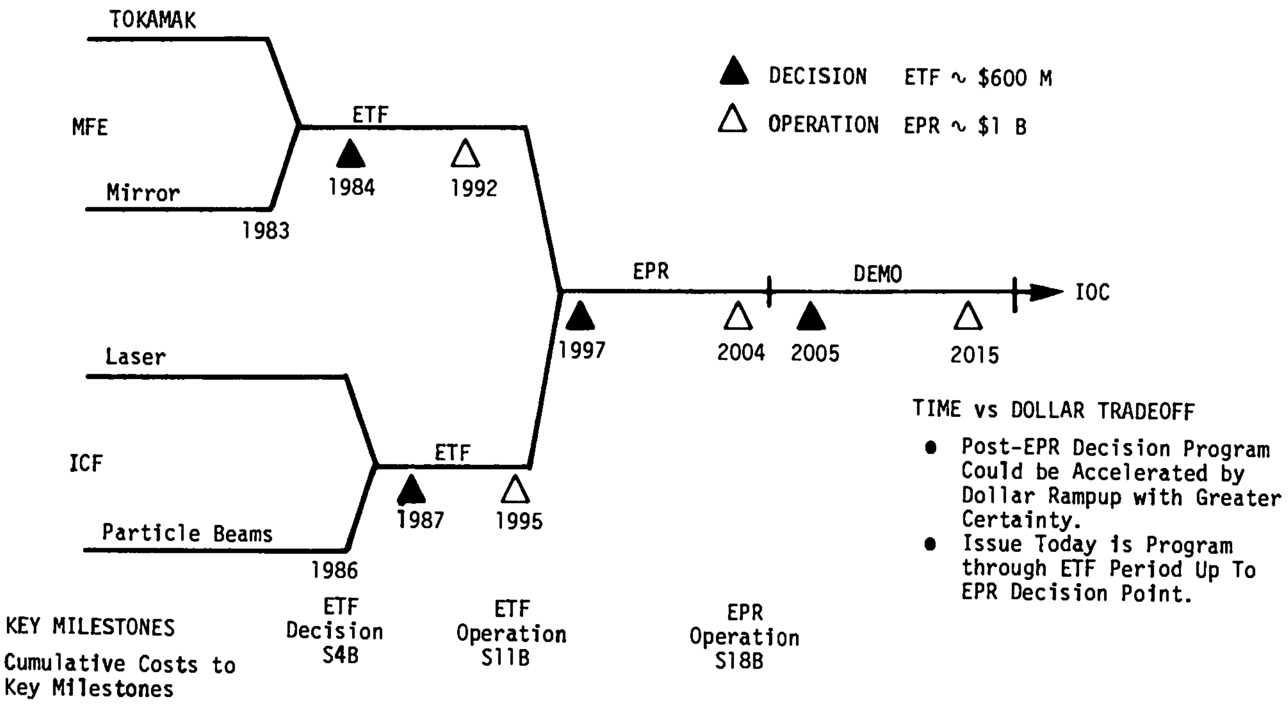

Figure 1. Fusion Development Program Presented to the Congress by John M. Deutch.[1]

TABLE I

DRIVER SYSTEM CHARACTERISTICS FOR VARIOUS FUSION REACTOR CONCEPTS

Reactor Type	Driver Pulse Width (μs)	Power Supply Voltage (kV)	Pulser Pulse Width (μs)	Maximum Switch Power (TW)	Stored Energy (GJ)
Tokamak (Plasma Heating)	10^6	10-100	10^6	10^{-2}	10
(Plasma Confinement)	(steady state)	10-20	---	---	200
Theta-Pinch (Implosion Heating)	1.0	100-200	1.0	200	0.2
(Adiabatic Compression)	$6(10)^4$	10-20	$6(10)^4$	1.0	60
Laser	10^{-3}	300-500	2-3	10	0.02
E-Beam	10	3000	0.1	100	0.01

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HIGH-TEMPERATURE GAS COOLED REACTORS

INTRODUCTION

Nuclear reactors may be classified in a number of ways depending on the type and arrangement of fuel, the moderator and coolant used and on the speed of the neutrons sustaining the fission chain reaction. Thermal reactors are ones in which the high speed neutrons born in fission are slowed down (moderated) to speeds where the neutrons are in kinetic equilibrium with the thermal motion of the moderator atoms. Moderator materials are primarily graphite, light water and heavy water. There are certain nuclear, engineering and economic advantages to the thermal reactors. Fast reactors are ones in which the neutron speeds are maintained as light as possible from birth through death, the principal advantage being a neutron economy in which the reactor can breed substantially more fissile material than it consumes. Fuel materials in reactors may be natural uranium, uranium enriched in the ^{235}U isotope, plutonium or ^{233}U. Coolants may be gases, liquid metals, light water, or heavy water, but since the last two are also good moderators they are excluded from fast reactor designs. Conventional light water reactors (LWRs) are slightly enriched uranium fueled, light water cooled and moderated reactors. The Canadian CANDU system employs heavy water as moderator and heavy water or light water as coolant. The Clinch River Breeder Reactor is a plutonium fueled, liquid metal cooled, fast breeder reactor. Fort St. Vrain is a high-temperature gas-cooled reactor using highly enriched uranium as fuel, graphite as moderator, and helium gas as coolant.

Gaseous coolants have been used in reactors for over twenty five years. The UK plutonium production piles at Windscale employed a once-through air cooling system. Later British designs, the Magnox and Advanced Gas Cooled Reactors, typified by Wylfa and Hartlepool, respectively, used carbon dioxide as coolants. However, because of the rapidly increasing reaction rate of CO_2 with graphite ($CO_2 + C \rightarrow 2\ CO$) with increasing temperature, CO_2-cooled graphite moderated reactors are limited to gas outlet temperatures below 700° C or so. In order to achieve the coolant outlet temperatures required for high temperature process heat (800 to 1000° C or more) helium is the best coolant.

Several high temperature gas cooled reactors (HTGRs) have been in operation over the last decade. In the mid-60s the Dragon reactor was built in England by a consortium of twelve European countries to demonstrate the technology on which the HTGR is based. This 20 MWe reactor was operated successfully from 1966 to 1977 and has now been decommissioned. Dragon operated at a power density of 20 MW/m^3, substantially greater than the 6-8 MW/m^3 of commercial HTGR designs, with 750^0 C helium outlet temperature. An important feature of the design was the use of coated fuel particles to minimize fission product release at high temperatures. The first US HTGR was Peachbottom, successfully operated by Philadelphia Electric between 1967 and 1976. The power output was 115 MWt (40 MWe) with a helium outlet temperature of 715^0 C. This was followed by the design, construction and operation of the Fort St. Vrain HTGR for the Public Service Company of Colorado. This demonstration plant is designed for 842 MWt (69,000 x 10^6 BTU/d) - 330 MWe with a 780^0 C helium outlet temperature.

The Dragon, Peachbottom and Fort St. Vrain reactors were all "prismatic" core reactors, so named because the holes for the coolant channels and the coated particle fuel rods were drilled into prismatic shaped blocks of graphite. A uniquely different design concept is the pebble bed fuel element in which the coated fuel particles are embedded in a spherical or spherical shell graphite matrix. Many thousands of such spherical fuel elements fill the cylindrical core cavity of a pebble bed reactor with burnt up fuel elements being discharged continuously from the bottom and fresh fuel elements being added to the top.

Although the pebble bed reactor concept was originally an American idea, the West Germans are the only ones who have built or operated a facility. Their AVR plant, a 46 MWt (3,770 x 10^6 BTU/d) - 15 MWe experimental reactor,[1] has been operating with great success. It has been operating since 1967 and since 1976 the average helium outlet temperature has been 950^0C. Over a million fuel pebbles have been circulated through the system and fission product release from the fuel has been a factor of a million lower than expected. For the first 10 years of operation the average plant factor was around 75% and in 1976 achieved about 92%, an important factor for process heat potential. The success of the AVR project has given the Germans the confidence to build a 750 MWt (61,400 x 10^6 BTU/d) - 300 MWe demonstration electricity generating pebble bed (THTR), to be operational in 1981, to design a 500 MWe process heat plant (PNP) for mid-80's operation and a later 1600 MWt (131,000 x 10^6 BTU/d) - 650 MWe plant (HHT).

The unique capabilities of HTGRs center around:

- The excellent neutronic, thermal, mechanical and chemical properties of graphite.
- The use of helium as a coolant (chemically inert and single phase).
- The ability to encapsulate fuel particles in nearly impervious graphite coatings.

HTGR COATED FUEL PARTICLE TECHNOLOGY

The two basic fuel particle concepts used in HTGR, the BISO and TRISO coatings, are shown in Fig. 1.[2] In the BISO concept a central ceramic fuel kernel (400 to 500 μm in dia.) is surrounded by two layers of pyrolytic graphite. The inner layer of porous low density pyrolytic graphite ($\sim$ 1.2 g/cm^3, thickness $\sim$ 95 μm) allows the kernel to swell and protects the outer layer from recoil damage. This outer layer is more dense (ρ = 1.80 to 1.85 g/cm^3, thickness = 75 to 110 μm) and serves to prevent fission product release. For high temperature operation (gas outlet temperatures above 950 ^{o}C) or for more conservative operation the TRISO concept is used. In this concept there are additional layers of a pyrolytic silicon carbide ($\sim$ 3.2 g/cm^3) and pyrolytic graphite. With both normally operating particles and particles with cracked coatings, the fission product release are orders of magnitude lower for the TRISO than for the BISO coatings.

HTGR FUEL ELEMENT DESIGNS

Prismatic Core

The fuel element for an HTGR of prismatic design is shown in Fig. 2.[3] The basis of the element is a hexagonal block of graphite (about .8 m long and .36 m across the plate) in which there are 72 holes for coolant passage, 132 holes for fuel rods and 6 holes for burnable poison. The fuel rods consist of vertical stacks of cylinders made up of coated fuel particles embedded in a graphite matrix. The elements are stacked on top of each other to form a column and many columns are stacked in a cylindrical array to form the reactor core. Column alignment is maintained by a dowel pin and socket arrangement. The number of elements forming a column and the number of columns would depend on the required power output from the core.

Not all of the columns in the core are identical. Some reactivity control is required for shut-down and operational controls. A certain number of the columns in the core are of the form shown in Fig. 3. The number of coolant, fuel and burnable poison holes are reduced to 43, 76, and 4, respectively, to make room for three large control channels.

PEBBLE BED FUEL ELEMENTS

The two forms of spherical fuel elements for pebble bed cores are shown in Fig. 4.(2) The conventional ball consists of a central graphite matrix containing the coated fuel particles. This 5 cm diam fuel region is surrounded by a 0.5 cm thick graphite shell which acts as cladding. In the shell ball the fuel matrix is in the form of a cylindrical shell (1.5 cm inner radius, 2.5 cm outer radius) with a graphite center and a 0.5 cm thick outer graphite shell. The advantage of the shell contribution can be seen by comparing the power profiles across the elements (show at the bottom of Fig. 4). For a 5MW/m^3 power density the maximum fuel temperature in the shell element is about 200^{o}C lower than that in the conventional ball.

HTGR DESIGNS

Prismatic Cores

As mentioned above the prismatic HTGR core is a packed assembly of fuel and control columns in a cylindrical core cavity, each of the columns being a vertical stack of prismatic elements. The core plan for a 3000 MWt (95,000 x 10^6 BTU/d) - 1160 MWe General Atomic design is shown in Fig. 5. There are 493 columns (including 73 control columns) each of which is a stack of eight elements. By varying the fissile fuel loading among each of the eight elements it is possible to shape the axial power distribution to give a more constant fuel temperature distribution, but in practice the number of distinct axial fuel zones is limited to about three, primarily because of fuel fabrication costs. The effective diameter of the GA core is 8.4 m and its height is 6.3 m. Figure 6 shows the general arrangement of the GA large HTGR nuclear steam systems. It will be noted that the circulators and steam generators are located within cavities in the prestressed concrete reactor vessel (PCRV).* The inside and outside diameters of the PCRV are 11.3 m and 30.6 m, respectively; the inside and outside heights are 14.4 m and 27.8 m, respectively. The design performance data of the GA core are:

Power	3000 MWt (246,000 x 10^6 BTU/d)
Power Density	8.4 MW/m^3
He Inlet Temperature	320 ^oC
He Outlet Temperature	775 ^oC
He Pressure	50 bar
Maximum Fuel Temperature	1350 ^oC
Fuel Lifetime (Th/ cycle)	4 yr at 80% LF
Fraction of Core Refueling each cycle	1/4
Average Burnup	98,000 MWd/Tonne
Net Plant Efficienty	38.6%

No GA 1160 MWe HTGRs have yet been built or sold. In the early 1970s ten orders were placed but these were later cancelled because of the general excess of electricity supply in the US.

In the US, one of the principal advantages of the prismatic designs is the already well established technology base. Also, in the event that part of the core has to be changed for some reason, the reactor can be shut down and that part changed fairly quickly. In the pebble bed design one would have to wait

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*In the Fort St. Vrain HTGR, noted previously, the steam generators and circulation are located under the platform that supports the core.

until the balls reach the outlet unless one is prepared to discharge most of the core.

Pebble Bed Reactors (PBR)

Figure 6 shows a basic and self-explantory longitudinal section of large process heat pebble bed reactor. The core dimensions would be about 6 m high and 11 m diam. and would contain about 2.7 million fuel balls. Figure 7 shows the integration of the same PBR with steam generators and steam reformers located within cavities in the PCRV.[4]

Typical performance data for such a design would be:

Power Output	3000 MWt (246,000 x 10^6 BTU/d)
Power Density	5 MW/m^3
He Inlet Temperature	280°C
He Outlet Temperature	980°C
He Pressure	40 bar
Mass Fuel Temperature	1170°C
Fuel Residence Time	4.5 yr
Fuel Feed Rate	3000 balls/d
Average Burnup	100,000 MWd/Tonne

About 1070 MW of high temperature process heat (700 to 950 °C) would be absorbed by steam reformers to produce around 49 MCF/h of hydrogen from methane. The other 1929 MW of heat would be absorbed in steam reformers to produce around 1.4 x 10^6 MWe. To date, the only PBR in operation is the 46 MWt (15 MWe) experimental AVR plant at Julich, West Germany. The West Germans are constructing a 300 MWe (electrical generation only) pebble bed (THTR) which is scheduled for operation in 1981.

RELATIVE ADVANTAGES AND DISADVANTAGES OF PBR OVER PRISMATIC CORES

The HTGR pebble bed concept offers a number of design, construction, and operational advantages:

- The fuel pebbles are easy to fabricate and, since a large number of identical units are required, mass production is appropriate.

- The fuel pebbles are rugged, having good dimensional stability under irradiation, good corrosion resistance and very low fission product diffusion loss rates.
- With the Once-Through-Then-Out (OTTO) fuel management scheme, there is a good match between the axial power distribution and the coolant gas temperature distribution (see Fig. 9). This results in the lower maximum fuel temperature for a given coolant outlet temperature.
- Because of the small size of the fuel element and the simple charge-discharge arrangement, on-line fuel changing is a major advantage of the PBR particular when employed as a process heat source.
- A smooth change over of fuel cycles is possible without shutting down or emptying the core vessel, again due to the small size of the fuel elements.
- The core geometry is defined by the core container walls. No individual fuel element positioning restraints or spacers are required.
- The natural packing of the fuel elements - the core gives a multiplicity of coolant passage interconnections which would tend to minimize the effects of blocked channels.
- The concept of the feed-breed fuel cycle with the initial fissile material being physically separate from the fertile material is more easy to apply in the PBR (separate feed and breed pebbles).

As mentioned above, most of the confidence in the performance of PBRs stems from the successful operation of the small AVR plant. There are, however, a few concerns about potential problems in large (3000 MWt) PBRs. In a small reactor, substantial reactivity effects can be achieved by moving control rods into the space between the top of the core and the upper reflector, and a small penetration of the core by the control rods will shut it down. With the large reactors, the reactivity worth at the core edge is much smaller and deep penetration of the bed by many control rods would be necessary to achieve a state of cold-shutdown. The West Germans are finding that injection of ammonia gas to reduce friction may be required to reduce the insertion force to reasonable levels. They are also working on a helical screw control rod which has a pitch of about a fuel ball diameter.

Another disadvantage of the pebble bed design is the difficulty of providing any interior core instrumentation, which however is a relatively simple arrangement in the prismatic design.

The reflector of a prismatic core is made up of prismatic graphite blocks which can be routinely replaced to avoid long-term damage from high fluences. In the case of the pebble bed, the reflector provides some of the structural containment for the core. Thus, the reflector has to be designed so that it will not break up in such a way as to affect core operation. One possibility might be an outer radial reflective zone of moving graphite balls which are small enough to not significantly degrade the gas outlet temperature but large enough to preclude penetrating the gap between the fuel balls.

HTGR FUEL CYCLES

Prismatic and pebble bed core HTGRs can operate on a number of fuel cycle concepts. Because of the superior netronic properties of U-233 in a thermal neutron spectrum and because of the desire to be able to exploit thorium ore reserves, the most appropriate HTGR fuel cycle is one in which U-233 and thorium are utilized in an nearbreeding system. Since U-233 does not exist in nature and very little man-made U-233 is available, HTGRs would have to start with U-235 and continue with some U-235 makeup (until U_{233}/Th breeders are developed with recycle U-233).

Prismatic HTGRs are usually designed to start off with a 93% enriched U-235 in thorium. A substantial fraction of the energy from the fuel element comes from in-situ fission of U-233 converted from Th. A prismatic HTGR operating on such a cycle and with a conversion ratio of 0.8 will have annual uranium requirements about half of that for an LWR operating on a plutonium recycle. The initial enrichment requirements are greater for the HTGR but over the life of the reactor it would be about the same as for the LWR.

Two basic fuel cycles are the focus of pebble bed reactor technology. One is the low enriched uranium (LEU) fuel and the other is the highly enriched U-235/Th fuel. With the LEU cycle the fuel balls operate on the (OTTO) concept and up to 90% of the plutonium bred in the core can be burnt up in-situ. The discharged elements have so little plutonium left that reprocessing may not be economical. With the highly enriched U_{235}/Th cycle all of the fissile material can be placed in one type of ball and all of the fertile material in another. The "feed-balls" will go through the core only once before reforming but the "breed" balls containing bred U-233 can be pushed through again. A conversion ratio close to unity can be achieved with such a fuel cycle with a minimum of reprocessing.

ENVIRONMENTAL AND SAFETY ASPECTS OF HTGRs

The inherent safety characteristics of HTGRs are probably superior to all other reactor types. The use of a large mass of graphite (with a high specific heat) as the moderator ensures that the effects of any sudden changes of energy input or energy extraction from the core will be slow and readily controllable. Graphite does not melt but sublimes at 3330 °C and its strength actually increases with temperature up to about 2500 °C. The helium coolant is a chemically inert and noncorrosive gas which is in one phase throughout all conditions of reactor operation, normal or otherwise. Thus there cannot be the type of loss of coolant accident that is postulated for light water reactors where a leak in the primary coolant vessel might result in the water coolant flashing out of the core vessel. In an HTGR a primary circuit leak could result in depressurization of the helium gas but some residual cooling capacity remains.. Because helium is transparent to neutrons there are no coolant reactivity effects, which lead to greater stability.

The environmental impact of an HTGR is relatively small. Liquid radioactive waste from the reactor plant is very low (10 Ci/yr for a 3000 MWt plant) and the gaseous radioactive waste is negligible. Systems for handling radioactivity wastes are provided so that releases to the environment are essentially zero.

COUPLING OF HTGRs WITH PROCESS HEAT PLANT

To obtain maximum thermal efficiency the most direct coupling of the HTGR heat output to the process plant would be required. Under present circumstances, however, it is unlikely that a nuclear process heat plant could be licensed without an intermediate heat exchanger (IHX) decoupling the nuclear from the process plant. One exception to this might be the use of a double walled steam·reformer but this would be more suspect under abnormal operation than would a conventinal IHX.

The purposes of the IHX are to include:

- Reduce or eliminate the transport of radioactivity into the product
- Reduce or eliminate the ingress of process materials (usually hydrogen or water) into the reactor.
- Reduce relability demanded of the PHX.
- Improve public acceptance and licensability.

The use of an IHX, however, introduces the following disadvantages:

- The cost of the proces heat increases by about 20%.
- Higher reactor outlet temperature required (50 to 100^0 C) for some process heat temperatures. This not only reduces thermal efficiency but also aggravates high temperature material problems.
- Need to develop high temperature values in the secondary helium system.
- More operating components, increasing probability of plant outages.

TECHNOLOGICAL PROBLEM AREAS OF PROCESS HEAT HTGRs

When HTGRs are designed solely for steam-electric generation there is generally no incentive for selecting a coolant outlet temperatue much above 750 ^{0}C since the region of rapidly diminishing returns has been reached. When the HTGR is required as a very high temperature process heat source a region of technological problems and uncertainties is entered. These problems are largely material problems outside of the reactor core and are associated with hot coolant ducts, insulation liners, process heat exchangers and various valves and seals.

The reactor's prestressed concrete pressure vessel (PCRV) must be protected from the hot coolant ducts and primary circuit components in PCRV cavities. Selection of materials for ducting and metal cover plates to hold the insulation to the PCRV liners represents a significant development problem.

Metals are currently not ASME code qualified for use much above 815 °C and very few alloys are approved at 815 °C. The basic problem is one of finding materials that will not lose their strength and ductility over many thousands of hours of operation at high temperatures and, in some components (PHXs), very corrosive environments.

Tables I-IV summarize the likely candidate materials for HTGR process heat systems, the problem areas and the probabilities of their successful application for four process temperatures: 760 °C, 815 °C, 870 °C and 1090 °C (without an IHX, 980 °C) respectively.(6) In all four cases the IHX/no IHX alternatives are evaluated. It can be seen that as the process temperature increases the uncertainty of finding satisfactory materials increases because materials data is incomplete or totally unknown.

The material in an HTGR core is virtually all graphite which has excellent very high temperature properties. In practice, the limiting temperature is governed by the ability of the fuel particles to retain fission products. The continuing development of HTGR fuel technology is being directed towards tighter fuel coatings, improved heat conduction between fuel kernel and coolant, and the development of extruded fuels for the prismatic design. However, there is great confidence in the capability of present and near state-of-the-art fuel technology.

Developmental Program for HTGR Process Heat Plants

The accumulated experience in US HTGR technology is totally with the prismatic core type. Although some paper studies and preliminary design work have been performed by General Electric,[7,8] there is no significant technology base for pebble bed reactors in this country. Nor does there seem to be any enthusiasm for them from DoE or the Gas Cooled Reactor Association recently formed by a group of US electric utilities. All information on PBR operating and fuel testing experience comes from the West German AVR plant.

As a result of the one-sided experience in HTGR technology in the US the developoment and construction of a lead PBR process heat plant could take up to ten years longer than its prismatic equivalent. Considering the special advantages that the PBR has in process heat applications, e.g. the potentials for higher temperature operations and very high plant factors, it would seem appropriate for the US to design and construct a small (~ 50 MWt) experimental PBR to develop the technology base (particularly fuel irradiation experience) which would be so important for the licensing of the lead plant. If pushed a small experimental PBR could be designed and constructed within three to five years because of the simplicity of the design concept and the information exchange which has already taken place through the FRG-US umbrella agreement on HTGR information sharing. Such an experimental program would involve a capital outlay of about $30 million. Whether or not such a step is essential depends on the acceptability of foreign information in the licensing process.

Many of the system components of an HTGR process heat plant are not very dependent on whether the core is a plebble bed or prismatic type. Much of the RλD requirements would therefore be common to both concepts. Development time-scales would depend to a large extent on the temperature requirements of the process and whether IHXs will be used. The effect of the temperature drop across the IHX would mean a totally new area of materials R&D. At this point in time it can probably be assumed that IHXs will be mandatory for the early plants.

An attempt to define the RλD program required for the construction of a lead HTGR process heat plant is given in Ref. 6, in which a five to eight year HTGR program definition phase is recommended, leading to the decision on whether to support development and construction of such a plant. The cost of these programs up to the decision point is estimated to be $110 M and $60 M for the reactor and process programs, respectively. A summary of the R&D elements and costs are shown in Table V. The proposed program does not involve the construction of a small experimental PBR, so for that type fuel, irradiation information would have to come from German sources and from small fuel testing programs on fuel elements in facilities such as FTR at Hanford. The timescale from the decision point to plant operation is difficult to estimate.

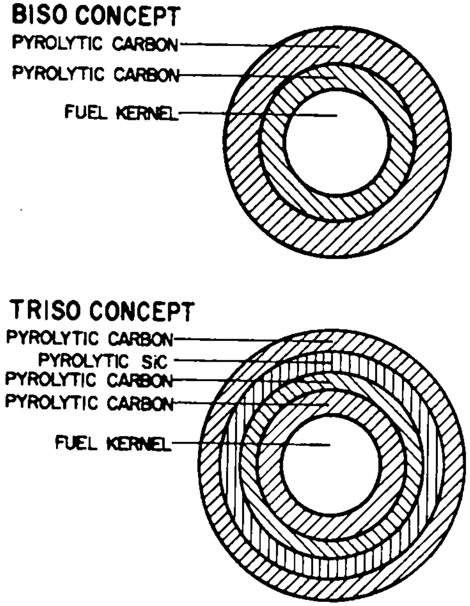

Figure 1. HTGR fuel particle concept.

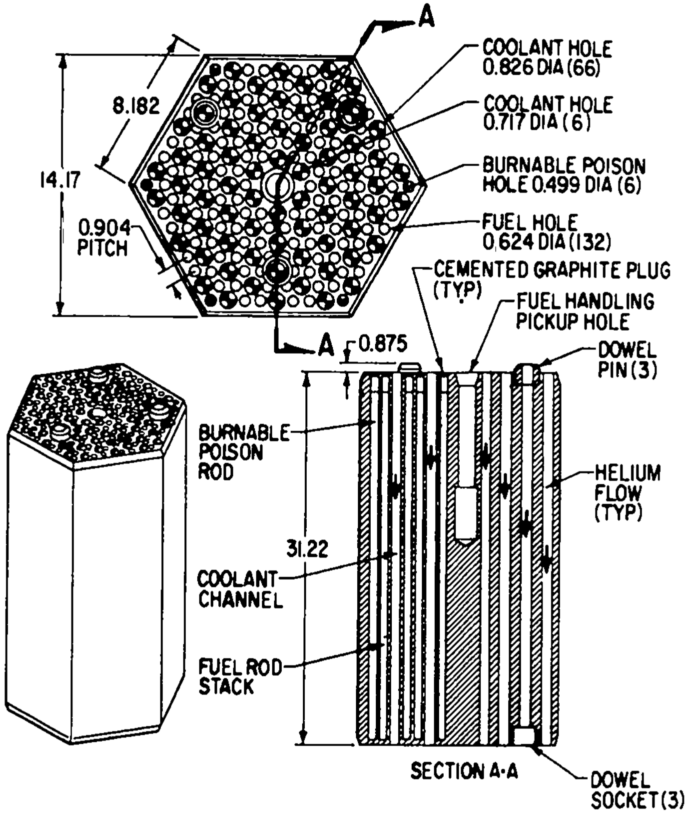

Figure 2. HTGR fuel element.

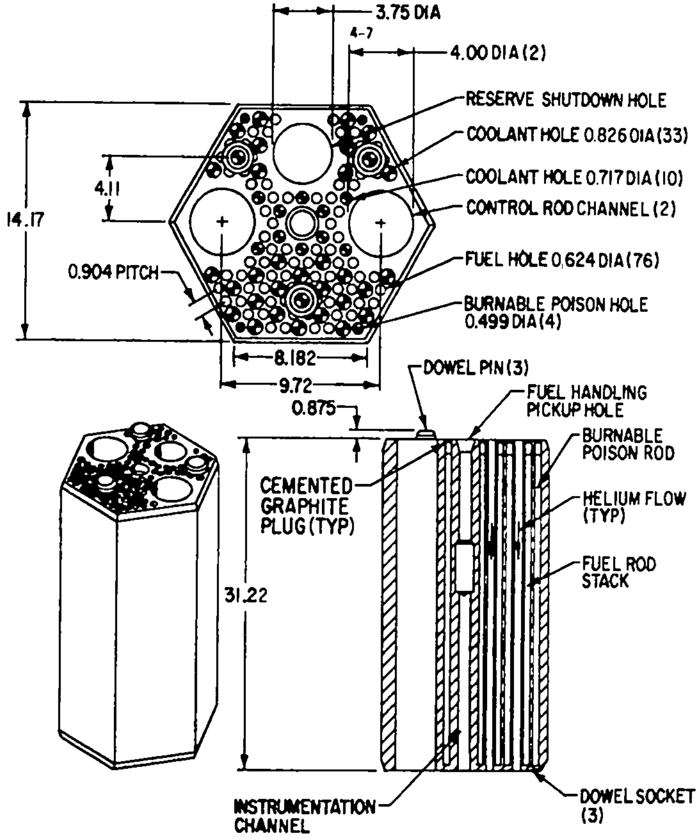

Figure 3. HTGR control fuel element.

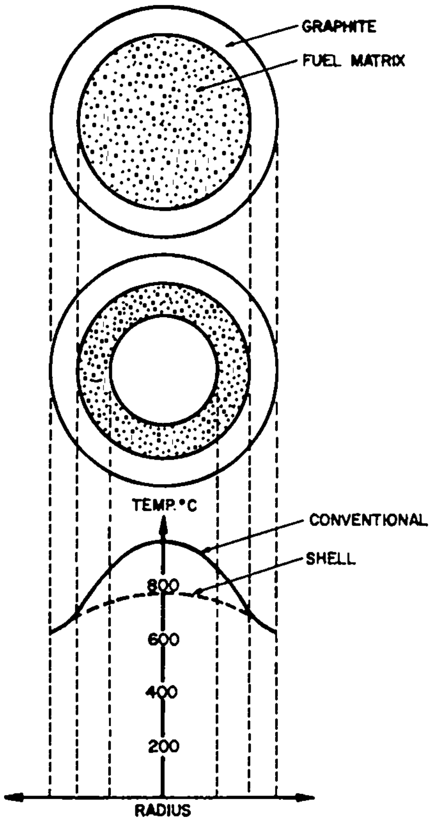

Figure 4. PBR fuel elements.

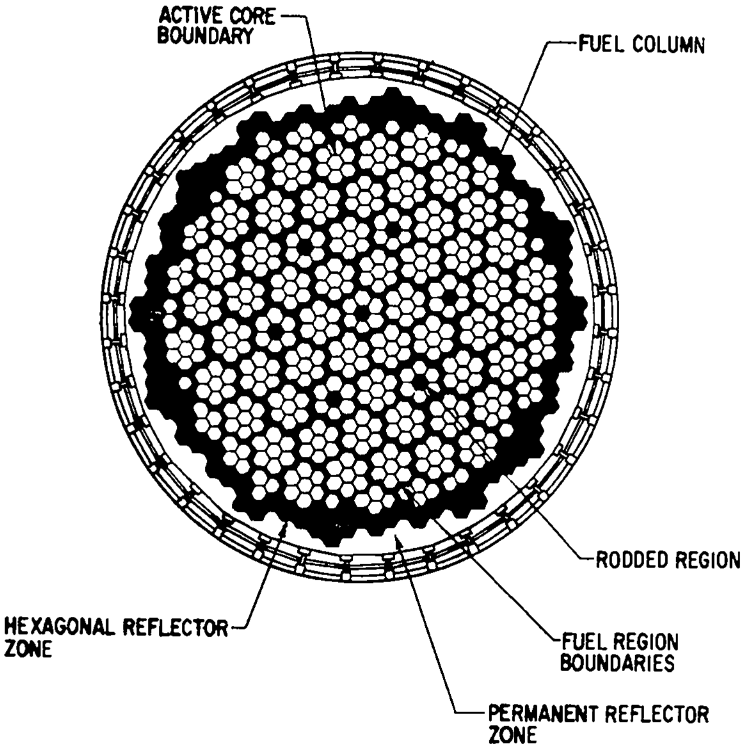

Figure 5. 3000-MWt core plan.

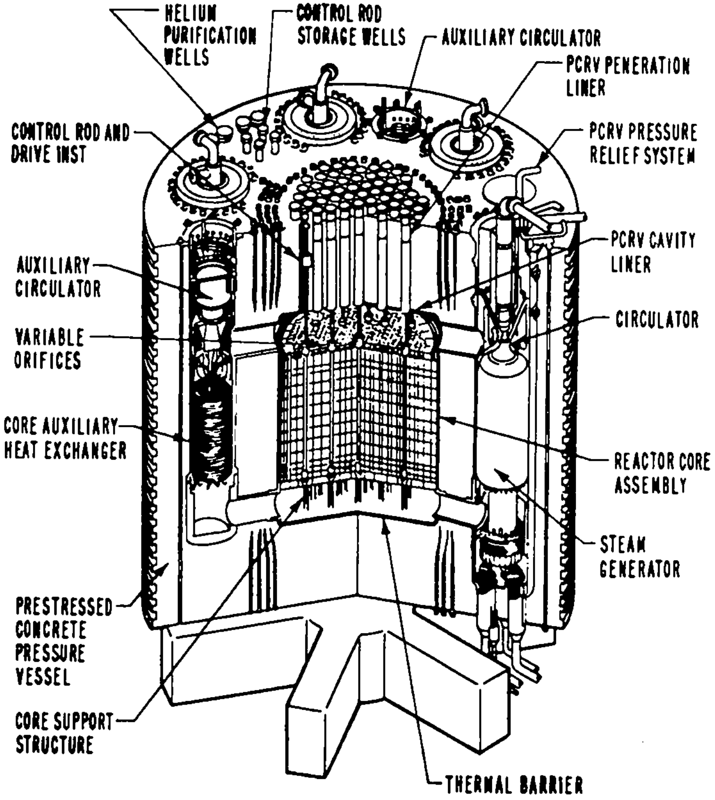

Figure 6. HTGR nuclear steam system.

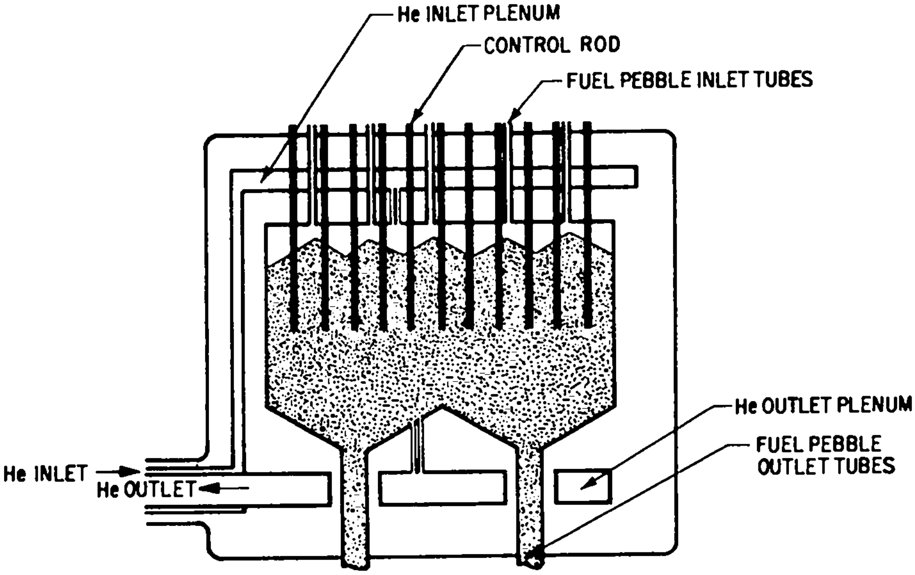

Figure 7. Longitudinal section of 3000-MWth process heat reactor.

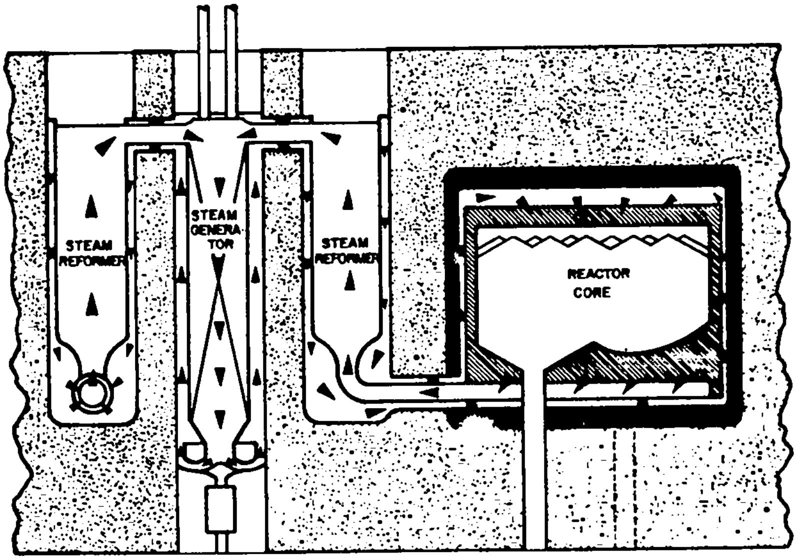

Figure 8. PBR process heat system.

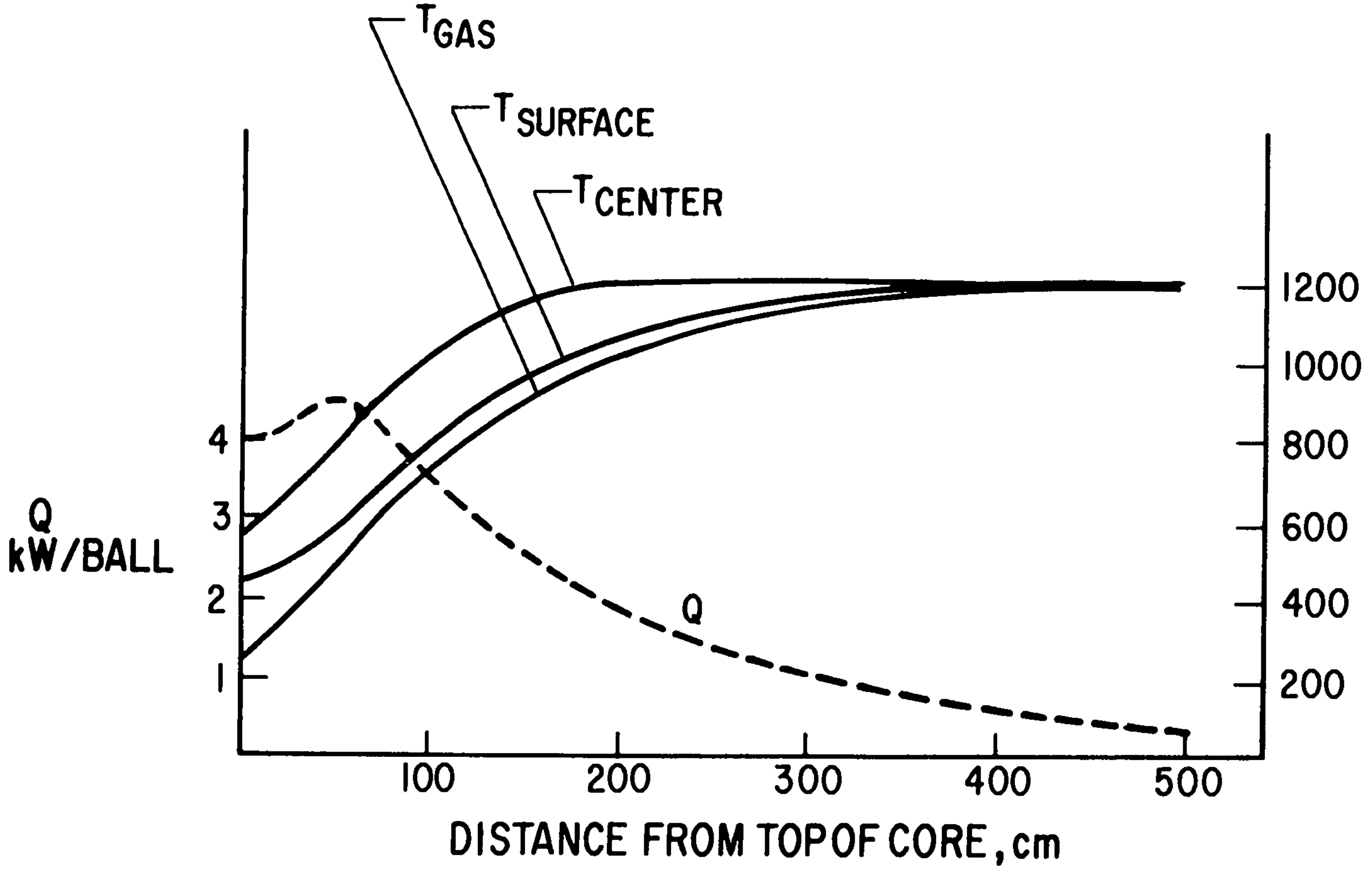
TGAS
TSURFACE
TCENTER
Q
kW/BALL
4
3
2
1
Q
1200
1000
800
600
400
200
T
°C
100
200
300
400
500
DISTANCE FROM TOP OF CORE, cm

Figure 9

TABLE I

MATERIAL CANDIDATES FOR CRITICAL COMPONENTS OF THE 760°C PROCESS TEMPERATURE VHTR

Component	Maximum Operating Temperature Range (C)	Typical Candidate Materials	Potential Problem Areas	Probability of Ultimate Applicability
			Without IHX	
Hot-duct insulation covers	730-900	Solid-solution-strengthened Ni-base alloys such as Hastelloy X, Hastelloy S, and Inconel 617	Thermal stability; effects of helium environment; not ASME Code approved; cobalt contamination (Inconel 617)	Good
PHX	732-900	Same as above	Same as above; effects of process environment	Potentially good, but effect of process environment largely unknown
Steam	565-675	Incoloy 800H, 304 SS stabilized high-alloy ferritics	Effects of helium and steam environments; Code approval of high-alloy ferritics	Excellent
Valves and seals	730-790	Ni-base alloys and austenitic steels such as Inconel 718 and 347SS	Effects of environment; thermal stability; Code approval; wear and galling problem	Good
			With IHX	
Hot-duct insulation covers	900-955	1. Same as w/o IHX 2. Mo-base alloy 3. Advanced superalloys 4. Ceramics	1. Same as without IHX, strength 2. Fabricability, environmental effects, Code approval 3. All aspects from alloy development through Code approval 4. Fabricability, shock resistance, environmental effects, Code approval	1. Fair 2. Potentially good, but unknown 3. Unknown 4. Unknown
IHX	900-955	Same as above	Same as above	Same as above
PHX	815-870	Same as PHX w/o IHX	Same as PHX without IHX	Potentially good but effect of process environment largely unknown
Steam generator	565-675	Same as steam generator without IHX	Same as steam generator without IHX	Excellent
Valves	815-870	Ni-base alloys, austenitic stainless steels	Effects of environment; thermal stability, Code approval material; wear and galling problems	Fair to good

TABLE II

MATERIAL CANDIDATES FOR CRITICAL COMPONENTS OF THE 815°C PROCESS TEMPERATURE VHTR

Component	Maximum Operating Temperature Range (C)	Typical Candidate Materials	Potential Problem Areas	Probability of Ultimate Applicability
			Without IHX	
Hot-duct insulation covers	900-955	1. Solid-solution strengthened Ni-base alloys	1. Thermal stability, effects of environment, ASME Code approval, strength	1. Fair
		2. Mo-base alloys	2. Fabricability, environmental effects, Code approval	2. Potentially good
		3. Advanced superalloys	3. All aspects from alloy development through Code approval	3. Unknown
		4. Ceramics	4. Fabricability, shock resistance, environmental effects. Code approval	4. Unknown
IHX	900-955	Same as above except omit Mo-base alloys	Same as above plus effects of process development	As above
Steam generator	610-720	Alloy 800H,304 SS, stabilized high-alloy ferr.	Effects of helium and steam environment; Code approval of high-alloy ferritics	Good to excellent
Valves and Seals	790-840	Ni-base alloys, austenitic stainless steels	Effects of environment, thermal stability, Code approval for 815°C applications (class 2,3), and all class 1 uses, wear, and galling	Good
			With IHX	
Hot duct insulation covers	955-1010	Same as hot duct w/o IHX except solid-solution Ni-base alloys	Same as hot duct without IHX	Same as hot duct without IHX
IHX	955-1010	Same as above	Same as above	Same as above
PHX	870-925	Same as PHX w/o IHX	Same as PHX without IHX	Fair to good but applicability of ceramics and advanced superalloys unknown
Steam generator	610-720	Same as steam generator without IHX	Same as steam generator w/o IHX	Good to excellent
Valves and Seals	870-925	1. Same as valves and seals w/o IHX but more restrictive	1. Effects of environments, thermal stability, code approval, wear and galling, strength	Fair to good
		2. Cast Ni-base alloys	2. Same as above but perhaps more serious stability problems and fabricability questions	Fair

TABLE III
MATERIAL CANDIDATES FOR CRITICAL COMPONENTS OF THE 870°C PROCESS TEMPERATURE VHTR

Component	Maximum Operating Temperature Range (C)	Typical Candidate Materials	Potential Problem Areas	Probability of Ultimate Applicability
			Without IHX	
Hot-duct insulation covers	955-1010	1. Advances superalloys 2. Mo-base alloys 3. Ceramics	1. All aspects from alloy development through ASME Code approval 2. Fabricability, environmental effects, Code approval 3. Same as (2) plus shock resistance	1. Unknown 2. Potentially good but unknown 3. Unknown
PHX	955-1010	Advanced superalloys and ceramics	Same as above plus process environment	Unknown
Steam generator	760-815	Austenitics	Effects of helium and steam environment, Code approval	Good
Valves and Seals	815-900	Ni-base and high-alloy austenitic steels	Effects of environment, thermal stability, strength, Code approval, wear and galling	Fair to good
			With IXH	
Hot-duct insulation covers	1010-1065	1. Mo-base alloys 2. Ceramics	1. Fabricability, environmental effects, Code approval 2. As above plus shock resistance	1. Potentially good but unkown 2. Unknown
IHX	1010-1065	Same as above	Same as above	Same as above
PHX	925-980	1. Advanced superalloys 2. Ceramics	1. Alloy development through Code approval 2. Fabricability, environmental effects, Code approval, shock resistance	1. Unknown 2. Unknown
Steam generator	760-815	Austenitics	Effects of helium and steam development, Code approval	Good
Valves and	925-980	1. Advanced superalloys 2. Cast superalloys 3. Ceramics	1. Alloy development through Code approval 2. Environment, stability, fabricability, wear and galling, Code approval 3. Same as (2) plus shock resistance	1. Unknown 2. Fair 3. Unknown

TABLE IV
MATERIAL CANDIDATES FOR CRITICAL COMPONENTS OF THE 1090°C PROCESS TEMPERATURE VHTR

Component	Maximum Operating Temperature Range (C)	Typical Candidate Materials	Potential Problem Areas	Probability of Ultimate Applicability
		Without IHX (980°C)		
Hot-duct insulation covers	1065-1120	1. Mo-base alloys 2. Ceramics	1. Fabricability, environmental effects, Code approval 2. Same as (1) plus shock resistance	1. Potentially good but unknown 2. Unknown
PHX	1065-1120	Ceramics	Same as (2) plus process environment compatibility	Unknown
Valves and	955-1010	1. Advanced superalloys 2. Cast superalloys 3. Ceramics 4. Mo-base alloys	1. Alloy development through Code approval 2. Environment, stability, fabricability, wear and galling, Code approval 3. Same as (2) plus shock resistance 4. Same as (2)	1. Unknown 2. Fair 3. Unknown 4. Fair to unknown
		With IHX (980°C)		
Hot-duct insulation covers	1120-1175	Same as hot duct w/o IHX IHX	Same as hot duct w/o IHX	Same as hot duct w/o IHX
IHX	1120-1175	Same as above	Same as above	Same as above
PHX	1040-1090	Ceramics	Same as PHX without IHX	Unknown
Valves and	1040-1090	1. Cast superalloys 2. Mo-base alloys 3. Ceramics	1. Same as valves and seals without IHX plus strength 2. As without IHX 3. As without IHX	1. Poor to fair 2. Poor to unknown 3. Unknown
		With and Without IHX (1090°C)		
Hot-duct insulation covers	1230-1290	Ceramics and Mo-base alloys	All problem areas apply, strength of Mo-base suspect at upper end	Unknown
IHX	1230-1290	Same as above	Same as above	Unknown
PHX	1150-1205	Ceramics	All problem areas apply	Unknown
Valves and seals	1150-1205	Same as valves and seals for 980°C process temperature with IHX		

TABLE 5

SUMMARY OF RλD PROGRAM ELEMENTS REQUIRED FOR LEAD BHTR PROCESS PLANT[a,b]

Element	Budget (millions of 1975 dollars)		
	Program definition (phase 5-8 years)	Construction phase	Total
VHTR process program			
VHTR development	110	110	220
Applications evaluation and process RλD	60	60	120
Lead plant design and detail development		120	120
Subtotal, VHTR process program	170	290	460
Supporting programs			
HTGR base program	60	70	130
Fuel reprocessing-recycle	50	100	150
Subtotal, supporing programs	110	170	280
Total	280	460	740

[a] Estimate does not include the construction cost of the lead VHTR process plant.

[b] Estimate assumes HTGR-steam cycle and HTGR-gas turbine plants are not developed, so that costs of the supporting programs are chargeable to VHTR.

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HYDROGEN FROM HIGH-TEMPERATURE ELECTROLYSIS OF STEAM

I. INTRODUCTION

Because natural gas has been so cheap and plentiful almost all the hydrogen used in the US is produced by steam-reforming of methane, the main constituent of natural gas. Virtually all of this hydrogen is used for chemical feedstock. The more costly electrolytic hydrogen has been used on a very small scale for specialized purposes. In some countries, in which either natural gas has been scarce or cheap hydroelectric power has been available, commercial electrolytic plants have been in operation for many years. Once natural gas becomes scarce in the US, advanced concepts may be developed to the point where electrolytic hydrogen becomes competitive with hydrogen produced by other methods. Almost a dozen companies in Europe and North America are presently engaged in the design and manufacture of commercial electrolyzers for the world market, and several new design approaches are being investigated. One interesting development is the use of high-temperature nuclear process heat for the high-temperature electrolysis of steam. As the operating temperature of the electrolytic process is raised, the proportion of electricity in the total energy requirement is reduced and cheaper, more efficiently produced heat energy can be partially substituted.

Present-day commercial electrolyzers are fairly efficient in converting electrical energy into hydrogen (60 to 80%), but, because up to 70% of the primary energy input to the electricity generating plants is rejected to the environment, the overall efficiency of electrolytic hydrogen production is only in the range of 20 to 33%. The prognosis for future developments is promising: cell efficiencies in the mid-90% range, with capital costs reduced 50% or more below the current value of 250$/kW electrical input.

As usual with energy supply technologies, the overall cost of electrolytic hydrogen production has two components - capital cost and operating cost. The operating cost of an electrolyzer is dominated by its efficiency and the cost of electricity. The capital cost can be reduced by using cheaper materials and construction techniques, but developments aimed at improving cell efficiency generally involve greater expense in those areas. Increasing the cell current density can usually more than counterbalance increased cost, but at the expense of cell efficiency. Thus, there are a number of design tradeoffs in electrolyzers, and the optimum design will depend on several factors, e.g., prevailing cost of electricity and particular application. An electrolyzer using relatively cheap off-peak electricity for only part of the day might be designed to have low capital costs. Electrolyzer manufacturers are reluctant to quote specific costs of hydrogen production because the electrolyzers can be fine-tuned to suit local needs and conditions. However, one fact is very clear: even if capital costs could be reduced to zero and efficiencies (hydrogen energy out vs electrical energy in) raised to 100%, the cost of hydrogen energy would be no cheaper than the busbar cost of electrical energy which, for the period 1985 to 2015, is estimated to range from 17 to 21$/10^{6}BTU for nuclear electricity and from 17 to 27$/10^{6}BTU for coal-fired electricity (both in current dollars), depending on geographic location.[1]

II. THEORETICAL BASIS FOR ELECTROLYSIS

Electrolysis is the process in which water is decomposed into hydrogen and oxygen by an electric current flowing through an electrolyte between an anode and a cathode. In the most commonly used electrolyte, aqueous potassium hydroxide, the electrode reactions are:

$$2\ H_2O + 2\ e^- \rightarrow H_2 + 2\ OH^-$$

at the cathode, and

$$2\ OH^- \rightarrow H_2O + 1/2\ O_2 + 2\ e^-$$

at the anode. With the General Electric solid polymer electrolyte[2] and the Westinghouse mixed-oxide electrolyte the charges are not carried by hydroxyl ions but by hydrated hydrogen ions and oxygen ions, respectively.

The basis for producing hydrogen from water by direct decomposition and thermochemical cycles is the same as for electrolysis, namely, the transformation

$$H_2O \rightarrow H_2 + 1/2\ O_2\ .$$

It can be shown from thermodynamic considerations that the input energy required to effect this transformation in a single-step process must be provided partly in the form of useful work (W) and partly in the form of heat (Q), as given by:

$$W = \Delta G_0$$

$$Q = T\ \Delta S_0\ ,$$

where T is the temperature at which the reaction takes place, ΔG_0 is the Gibbs free energy change across the process at temperature T, and ΔS_0 is the entropy change across the process at temperature T. The sum of W and Q equals the enthalpy change ΔH across the process. Figure 1 shows how ΔG and ΔH vary with temperature for the water decomposition process. Note that the magnitude of ΔG_0 decreases substantially with increasing temperature, whereas ΔH_0 increases only slightly. This means that the proportion of useful work (performed in the electrolytic process by electrical energy) decreases with increasing temperature until, at a temperature of ~ 3927°C, thermal energy alone would be capable of effecting the transformation. This decrease in the magnitude of ΔG_0 is the primary rationale for the development of high-temperature electrolysis and of thermochemical cycles, because heat can be produced at almost 100% efficiency whereas electricity generation is typically only 35 to 40% efficient, with a possible increase to 50% if advanced combined cycles are used.

Current process-heat sources, however, do not provide heat at 3927°C, in fact, we are limited to only ~ 1827°C for fusion reactors (if and when they are built) and to ~ 927°C for existing Very High Temperature Reactors (VHTRs), with a possible increase to 1077°C. Thus, in the near future, we might envisage a high-temperature electrolysis cell operating at 927°C with ~ 27% of the input energy being heat. As we shall see in the following sections, operation at even this temperature requires significant developmental work.

III. HIGH-TEMPERATURE ELECTROLYSIS

A. General

In addition to the decreasing need for electrical energy due to ΔG_0 reduction, several other advantages accrue from the operation of an electrolyzer at high temperature. The reaction kinetics at the electrodes increase with temperature, resulting in lower polarization losses; current densities can then be higher than at ambient-temperature operation. The total energy requirement for vapor-phase electrolysis is lower (by the latent heat of vaporization) than for liquid electrolysis. The latent heat can be provided by recuperated waste heat.

As might be expected, a number of problems are associated with the design of a high-temperature electrolytic cell, largely due to the hostile environment in which the components would have to operate for many thousands of hours. Among these problems are:[2]

- Loss of electrolytic ionic conductivity with time.
- Adequate stability of component materials in a hostile environment.
- Matching of thermal expansion coefficients of components for gas tightness.
- Interdiffusion of materials at boundaries.
- Prevention of tight sintering of porous electrodes.

Basically, an electrolyzer is made up of an electrolyte, electrodes, and interconnecting components linking a large number of electrolytic cells in series. The following desirable characteristics have been enumerated for high-temperature steam electrolysis:

Electrolyte	Low electronic conductivity.
	High and stable ionic conductivity.
	Nonporosity (to prevent H_2 and O_2 recombination).
	Mechanical stability.
Electrodes	High electronic conductivity.
	Good porosity.
	Stability to reducing environment (cathode).
	Stability to oxidizing environment (anode).

Interconnecting Material	High electronic conductivity. Stability to both reducing and oxidizing environments. Nonporosity. Good adhesion to electrolytes and electrodes.

In addition, the desirable characteristics of low cost and availability should be added to each of the above components.

The greatest interest and most developmental work on high-temperature steam electrolysis appear to exist at Dornier Systems GmbH in West Germany, and at Brookhaven National Laboratories and Westinghouse Corp. in the USA. The Germans are interested in the use of a pebble-bed VHTR to provide both electricity and high-temperature process heat ($900^{\circ}C$), whereas the US effort is focusing on the potential of fusion reactors as energy sources at a temperature in the range of 1377 to $1827^{\circ}C$. At $1827^{\circ}C$ the amount of electrical energy required would be only $\sim 55\%$ compared with $\sim 73\%$ at $900^{\circ}C$.

B. Electrolyte

The electrolyte that has been adopted by all developers of high temperature electrolyzers (HTE) is some form of metal oxide or metal-oxide mixture. Ytterbia-stabilized zirconia was one of the early prospective materials, but this has been superceded by the cheaper and more available yttria-stabilized zirconia.

The current through the oxide electrolyte is carried by oxygen ions and electrons. Because electronic conductivity shorts-circuits the cell potential, reducing the available voltage and efficiency, it is desirable that ionic conductivity carry over 99% of the current. Figure 2 shows the electrode reactions and the ionic movement in a basic high-temperature electrolyte cell. The flow of oxygen ions is controlled by the migration of vacancies in the lattice, the concentration of which depends on the concentration of the lower valency metal ions belonging to the dopant. Figure 3 shows the ranges of oxygen partial pressure and temperature over which calcium-oxide-doped zirconia would be suitable (proportion of ionic conduction, $\geq 99\%$). Because the partial pressure of oxygen depends on the ratio of steam to hydrogen partial pressures on the cathode-side of the electrolyzer (also shown in Fig. 3), the useful range of the electrolyte also depends on this parameter.

The long-term ionic stability of doped zirconia has been studied by Doenitz et al,[3] and their results are shown in Fig. 4. It would appear that 9 mol% yttria in zirconia gives good long-term stability.

To maximize cell efficienty it is important to reduce I^2R losses in the electrolyte. The electrolyte should be as thin as possible within the constraint imposed by mechanical strength requirements. Figure 5 plots the thickness required to hold the overvoltage due to I^2R losses below 50 mV as a function of temperature. In the potential fusion-reactor temperature range a thickness of 1 mm would be acceptable, but for a fission VHTR source the thickness could be lower by a factor of 10.

C. Anodes

The high operating temperature and the strongly oxidizing environment place severe constraints on the selection of material for HTE anodes. The noble metals are good electronic conductors and have good oxidation resistance, but they are generally ruled out on several other counts: They are expensive; gold and silver have melting points that are too low for HTE-VHTR coupling and, on the same grounds, platinum and palladium are undesirable for fusion reactor coupling; also, problems might arise in designing for gastightness due to poor matching of thermal conductivities. Doped oxides with a perovskite structure appears to be the most promising materials, the best choices being ones based on SnO_2 and In_2O_3.

D. Cathodes

For the hydrogen electrode in VHTR aplications nickel or cobalt cermets can meet the requirements of high electronic conductivity, high porosity, and the capability of matching thermal expansion coefficients; the skeletal structure of zirconia inhibits sintering. However, for fusion-reactor applications, conducting oxides would have to be used because sintering would destroy the porosity of the cermet.

D. Interconnections

Perovskite-structured oxides appear to be the most promising materials that satisfy the wide range of characteristics for the interconnecting material. The most promising materials are based on doped $LACrO_2$.

F. Higher Temperature Electrolyzer Programs

1. Dornier Systems GmbH

Dornier Systems GmbH of West Germany has been actively developing an HTE concept under the project name HOT ELLY (Hochtemperatur-Dampfphase - Elektrolyse) since 1975. A lot of experimental effort has gone into studying the characteristics of materials for electrolyzer components. The basic arrangement of components in this conceptual design is shown in Fig. 6. The electrolyte cylinders and short gas-tight cylinders of interconnecting material are joined by a special welding technique, and the porous electrodes are deposited with spraysinter techniques. Yttria-stabilized zirconia is the choice of electrolyte, with a nickel-cermet cathode and a doped perovskite-oxide (unspecified) cathode. Dornier's projected performance data for its HTE design are given in Table I. Overall efficiencies of an HTGR-HTE hydrogen production plant are projected to be in the range of 40 to 50%, depending on the type of electrical generating unit; the high-efficiency value corresponds to the use of a combined helium gas-turbine/steam-turbine cycle. From these overall efficiency projections, we infer that Dornier's electrolyzer is assumed to be 100% efficient.

A schematic of the VHTR-HTE hydrogen production plant is shown in Fig. 7.

2. Westinghouse Corporation

Westinghouse has been developing a HTE based on its fuel cell design; a schematic of the adaptation is shown in Fig. 8. Yttria-doped zirconia, doped indium oxide, and nickel cermet are the materials used for the electrolyte, the anodes, and the cathodes, respectively. The interconnecting material is based on a perovskite $LaCrO_3$.

3. Brookhaven National Laboratory(BNL)

BNL has performed conceptual design studies partially based on the Westinghouse high-temperature electrolyzer designs using steam and electricity generated from a fusion reactor.[4] Operating temperatures are to range from 1400 to 1800°C, which is, of course, much higher than can be achieved in fission reactors. Details of the proposed HTE vessel and cell-tube operations are shown in Figs. 9 and 10, respectively. The diameter of the vessel is 3.5 m and its length is 6.8 m. The two end sections of the HTE are at a low temperature and house electrical connections at the end of the electrolyzer tubes. To minimize thermal stresses, the outer region of the vessel is held at almost constant temperature at all times and the tubes are designed so that a free end can accommodate the large dimensional changes from shutdown to operating conditions.

The gas entering the first electrolyzer cell is steam, and in subsequent cells is a mixture of steam and hydrogen. The electrolyte (~ 10 μm thick) is supported by a porous zirconia tube similar to the one shown in Fig. 8.

The projected efficiencies of the design are 70% (fusion to hydrogen energy) for a 1800°C HTE unit and 50% for a 1400°C HTE unit. Modifications of the HTE design to a VHTR fission heat source (~ 1000°C) would likely reduce the overall efficiency to the range predicted by Dornier, i.e., ~ 40%.

G. Comparative Economics of High Temperature and Conventional Electrolysis

Let us assess the economics of generating hydrogen by comparing two plants:

Plant A is an advanced conventional electrolyzer using electricity generated by a 3000-MWt (1200-MWe) pebble-bed or prismatic-core HTGR, whereas

Plant B is a high-temperature steam electrolyzer using electricity and 900°C superheated steam from a 3000-MWt pebble bed or prismatic core.

At 900°C the split between electricity and heat-energy requirements of an electrolyzer is ~ 73 and 27%, respectively. Thus, if we assume electricity being generated at 40% efficiency and the 900°C steam at 95% efficiency, Plant B would be required to generate 380 MW of superheated steam and 1040 MW of electricity.

The following data are used in the cost calculations:

HTGR - Capital Cost	\$300/kWt
Steam Generator Capital Cost	\$ 40/kWt
Steam Superheater Capital Cost	\$ 40/kWt
Turbine-Generator	\$200/kWe
Fixed Charge Rate	18.6%
Fuel-Cycle Cost	\$0.50/10^6 BTU
Fuel-Cycle Cost Levelization Factor[+]	1.9

[+] Corresponds to annual fuel cost increase of 4%

Plant Capacity Factor	0.9
Hydrogen Output from Plant A*	2.91×10^{13} BTU/yr
Hydrogen Output from Plant B**	3.20×10^{13} BTU/yr

If C_L is the capital cost of the conventional electrolyzer in $\$/kW_{out}$ and C_H is the corresponding value for the high-temperature electrolyzer, then the hydrogen generation costs, H, for the two plants are given by:

$$H_A = 10.69 + 0.00767\, C_L \quad \$/10^6 \text{ BTU}$$
$$H_B = 9.53 + 0.00692\, C_H \quad \$/10^6 \text{ BTU} .$$

Projections for the capital costs of conventional electrolyzers (including subsystems) range from \$60 to \$255, depending on type and manufacturer.[5] We have performed parametric calculations of the above two equations with C_L and C_H varying from \$100 to $\$200/kW_{out}$. Table II shows the results.

The extreme environments in high-temperature electrolyzers will likely lead to higher capital costs than for conventional electrolyzers. The calculations are performed, therefore, for $C_L < C_H$. It can be seen from the table that the maximum gain in going to the high-temperature electrolyzer is of the order of 11%, which is marginal when one considers the relative difficulties in the technologies and in the required development program. The reactor in Plant B would be required to have a coolant outlet in the range 950^0 to 1000^0C, whereas the reactor in Plant A would not require an outlet temperature higher than ~ 750^0C. Thus, the assumption of equal capital costs for the two reactors is optimistic on the side of Plant B. These and similar considerations for other plant components make it difficult to project any cost effectiveness of high temperature electrolysis over conventional electrolysis using fission reactors with their temperature limitations around 1000 to 1100^0C.

H. Plasma Reactor Heat Source

As mentioned in Chapter 1 the plasma core reactor has the potential for operating at temperatures above 4727^0C. This is well above the temperature

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* Based on an electricity to hydrogen efficiency of 90%

**Based on Dornier Systems design[3]

(3927^{o}C) at which the chemical bonding between hydrogen and oxygen atoms in water can be broken by heat energy only. This very high temperature, however, is that of the UF_6 fuel gas which is confined in the vortex of a buffer gas to prevent fuel contact with the walls. Transfer of this thermal energy without large temperature drops will be a problem. Radiative heat transfer is the most obvious mechanism. This reactor concept does open up possibilities for process heat temperatures well above that of conventional HTGRs with their 1000^{o}C limits. At 2727^{o}C only about 30% of the input energy to an electrolyzer need be electricity. There would be extremely large developmental problems associated with electrolyzers above 1727^{o}C and any discussion of the viability of such a technology would be sheer speculation at this point in time. Also, this reactor concept is still in the conceptual stage and the funding and push for a developmental program to prove its feasibility has yet to materialize.

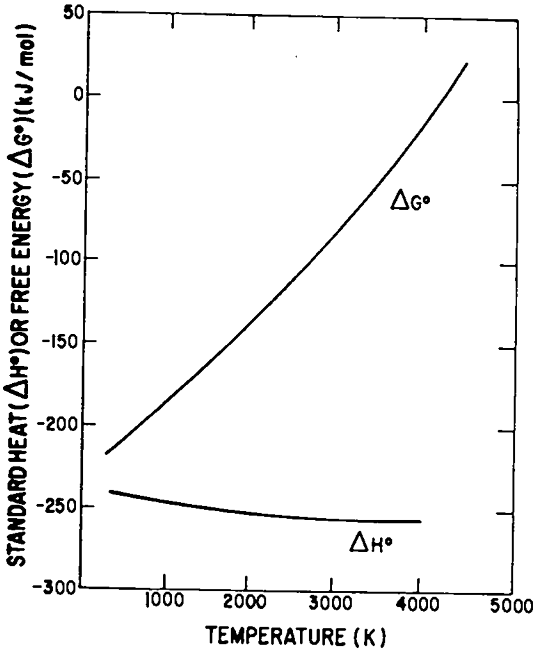

Figure 1.

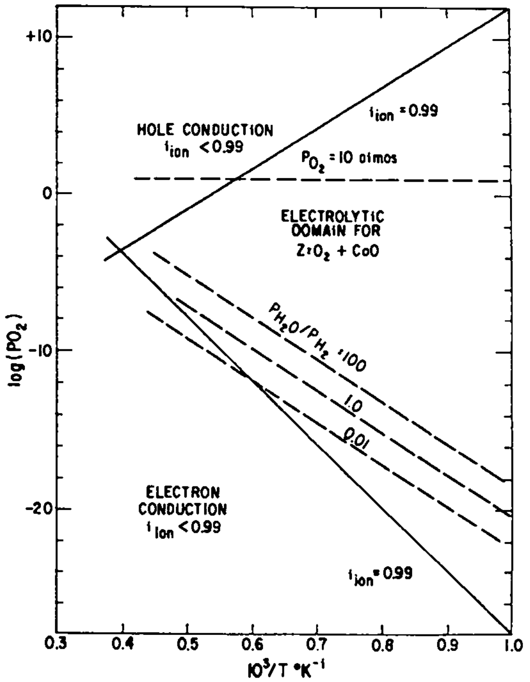

Figure 3.

Oxygen partial pressure as a function of temperature.

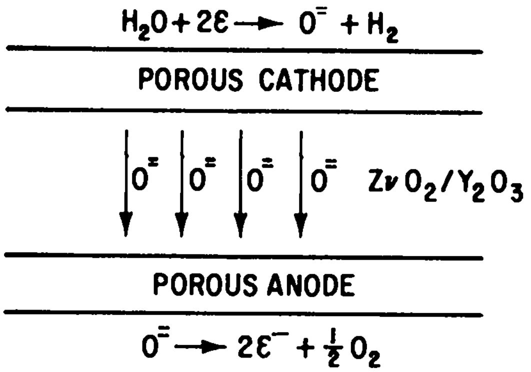

Figure 2.

High-temperature electrolytic cell.

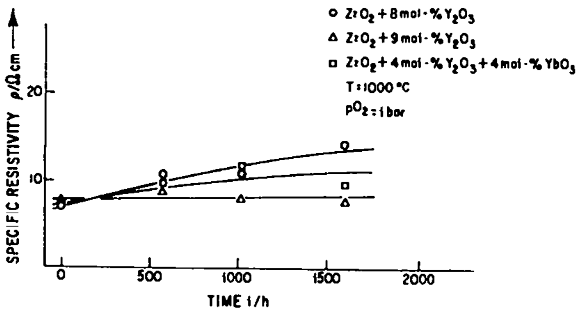

Figure 4.

Long-term stability of electrolyte ceramics.

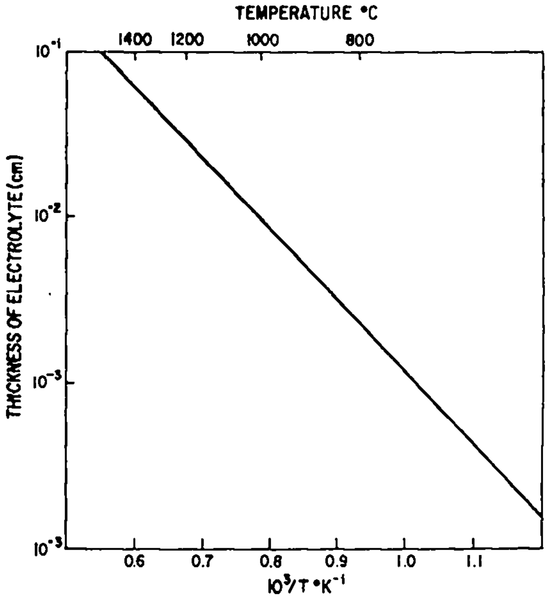

Figure 5.

Electrolyte thickness as a function of temperature.

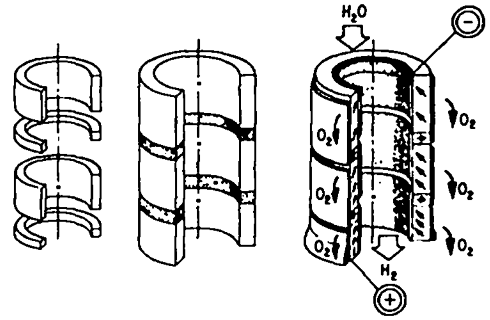

Figure 6.

Design and flow of current of interconnected cells in Dornier design.

TABLE I

PROJECTED PERFORMANCE OF DORNIER'S HTE

Average Cell Voltage, V	1.3
Current Density, A/cm^2	0.4
Operating Temperature, ^{o}C	900
Operating Pressure, bar (output pressure of H_2)	20
Gas Input	H_2O
Gas Output, Cathodic	0.75 H_2 + 0.25 H_2O
Gas Output, Anodic	O_2(+ H_2O)

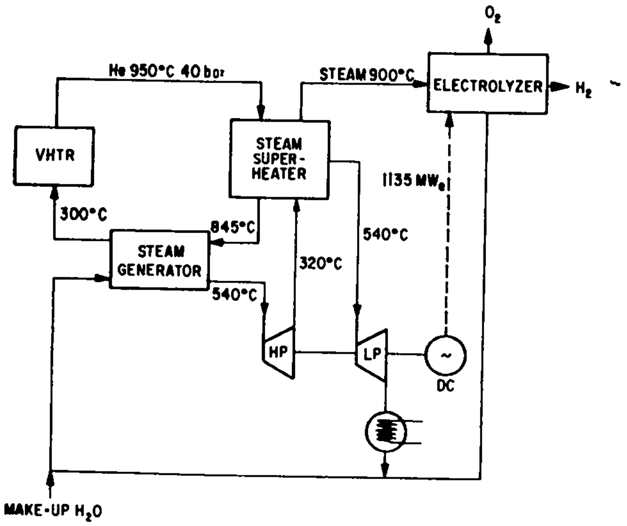

Figure 7.

High-temperature steam electrolyzer with VHTR supply.

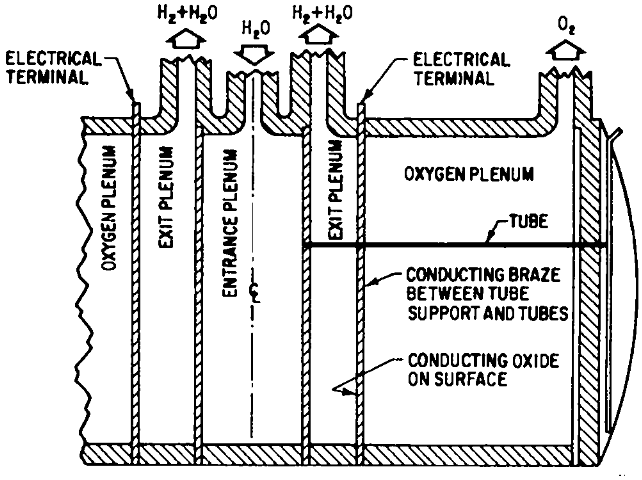

Figure 9.

HTE vessel.

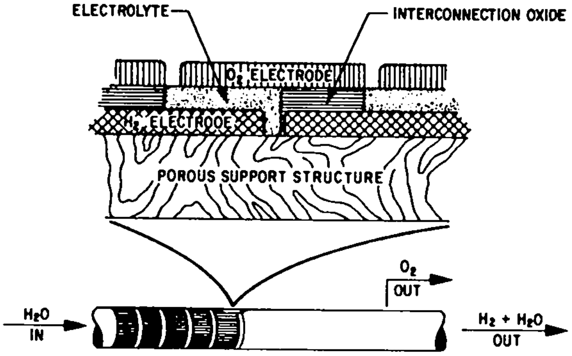

Figure 8.

HTE cell design (Westinghouse fuel cycle).

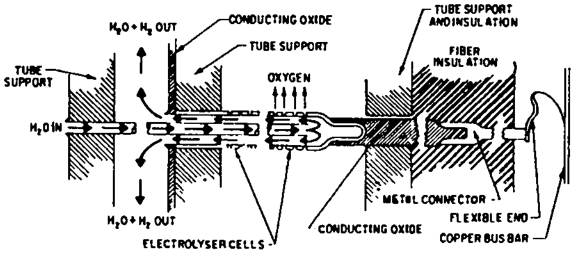

Figure 10.

Detailed cross section of HTE cell tube.

TABLE II
COMPARATIVE HYDROGEN PRODUCTION COSTS
($\$/10^6$ BTU)

C_L	C_H	H_A	H_B	H_B/H_A
100	100	11.46	10.22	0.892
200	200	12.22	10.91	0.893
100	200	11.46	10.91	0.951
300	300	12.99	11.61	0.894
200	300	12.22	11.61	0.950
100	300	11.46	11.61	1.013

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PRELIMINARY DESIGN OF A CATALYZED COAL GASIFICATION SYSTEM UTILIZING NUCLEAR HEAT

INTRODUCTION

The purpose of this evaluation is to determine whether or not nuclear heat can be economically and competitively wedded to a third-generation coal-gasification system. In most coal-gasification operations, endothermic energy must be provided to drive the steam carbon gasification or other similar endothermic reactions, such as the steam reforming of methane. However, recent information about catalyzed coal gasification indicates that the endothermic energy requirements for gasification may be substantially lower than thought. This study assesses the applicability of a nuclear reactor as a source of process heat to assist in catalyzed coal gasification. Recent developments at Exxon Corporation in catalyzed coal gasification are extensively used in this report.[1,2,3]

With catalyzed coal gasification, the overall system is somewhat simplified. No longer are separate parts of the system dedicated to the methanation step and the shift operation [see Eqs. (1) and (2)]. The complete conversion of the coal to produce methane occurs in the gasifier itself. The overall system is shown schematically in Fig. 1, and comprises four main steps. First, the coal is ground and impregnated with catalyst. Next, an extensive train of horizontal fluidized beds gasifies the coal and produces an almost carbon-free ash. Third, the off-gas from the gasifier system is purified in a purification and recycling section; the methane is removed while the carbon monoxide and the hydrogen are recycled to the gasifiers. Byproducts of hydrogen sulfide, carbon dioxide, and ammonia are also separated out in the gasification system. Fourth, the catalyst in the ash is leached out in a catalyst recycle system and returned to the coal-preparation operation.

The catalyzer gasification system offers the following advantages:

- The temperature of gasification can be reduced substantially from approximately 880-980°C down to approximately 700°C.
- The Exxon work[1,2,3] indicates that the potassium carbonate catalyst, when combined with coal char, performs a unique and useful catalytic action by equilibrating the shift and methanation reactions in the gas phase in each of the reactors. These equilibria permit more methane to be produced in each reactor than is now obtained in uncatalyzed systems.

- By recycling the carbon monoxide and hydrogen directly to the gasifier system, the equilibria can be shifted dramatically toward the production of methane. Thus, methane production is enhanced, and, by separating it out as the only product, the gasifier system performs all the operations of gasification shift and methanation that were previously performed in separate operations.
- The overall effect of the catalyzing-plus-recycling scheme is to gasify the coal with very little endothermic energy and with the release of only small quantities of exothermic heat. The overall energy efficiency is greatly increased.
- The potassium-carbonate impregnation produces a non-caking coal. Thus, no special preoxidation step is required, as it would be to process caking coals.

Disadvantages of a catalyzed coal gasification system are:

- More recycling and purification are required: The flow of gas around the loop from the gasifier through the purification system and back to the gasifiers is higher than in direct methanation systems. Therefore, the purification-equipment and operating costs may well be substantially higher. Also, the energy requirements for purification may be high because the system must be cryogenic to separate the carbon monoxide and hydrogen from methane.
- The catalyst must be reprocessed and recycled. Obviously, additional equipment and operational costs are incurred. Also, not all the catalyst can be recovered, and replacement cost will be substantial.

DESIGN CONSIDERATIONS

Obviously, the catalyzed coal gasification process offers many advantages, and may be much more cost-effective than conventional thermal gasification systems. However, the additional costs of recycling, purification, and catalyst preparation must be determined to assess the overall advantages of the system. Because the energy costs for conventional gasification are high due to the large amounts of endothermic energy required, the catalytic system may be vastly superior from an overall energy efficiency point of view. However, whether or not the endothermic energy reduction really is substantial in an actual reactor chain is the crucial question. Most of our effort is, therefore, directed toward the design of the gasification train and the overall assessment of energy requirements in that train.

The gasification train being considered is a series of ten horizontal fluidized beds through which the coal and ash flow countercurrently to the gasification products. A nearly pure ash with only 4% carbon is produced in Reactor 10. The compositions of gas out of each reactor, and the overall energy requirements for each reactor are assessed.

For a complete evaluation of the operation, the recycling system must be designed carefully. The elements of the system are therefore represented with calculated estimates of their energy requirements. Because the system is cryogenic, overall cost will be greatly affected by the choice of pressures and temperatures for the cryogenic operations. Pumping costs can be quite high because pressures must be lowered in the low-temperature separation. The cryogenic system with the refrigeration cascade is shown in Fig. 1.

For a full assessment, the catalyst regeneration system should also be designed. This was not done because equipment costs for this system are substantially lower than those of the other two important steps of the operation. About 20% makeup of the catalyst will be required.

COAL GASIFICATION CHAIN DESIGN

A. Gasification Chain Description

The coal is gasified in a series of ten horizontal fluidized bed gasifiers. The temperatures of the gasifiers are kept as low as possible to maximize methane production, but high enough to obtain reasonable gasification reaction rates. The gases flow countercurrently to the coal-ash flow from one gasifier to another. Gasifier 1 is at the coal inlet end and Gasifier 10 is at the ash outlet end. The system is sufficiently long to produce nearly carbon-free ash (~ 4 wt% carbon in the final ash).

The gas flow system is as simple as possible. The recycled gases -- hydrogen, carbon monoxide, and carbon dioxide -- are introduced into Reactor 10 along with water. The gases then flow countercurrently to the ash-carbon stream throughout the remaining nine reactors. More complex systems can be designed in which the water is introduced at appropriate points along the reactor chain to equalize the energy consumption or energy generation in the various reactors. However, in our initial design we are studying only the simple countercurrent system.

The calculational procedure comprises four steps.

1. The composition of the gas exiting each reactoris calculated first. The gases are in equilibrium in reference to the shift and methanation reactions. Thus, the mass balance calculation must account for the molecules entering and leaving the reactor along with the carbon being gasified. Also, the equilibrium constraints must be met.
2. The energy consumption or production by the reactor can be determined once the net reaction across the reactor is found from the mass balance. Thus, a stage-by-stage calculation first determines the exit gas compositions, from which the energy requirements for each reaction can be determined.
3. The reactor is then designed by determining the holdup volume required to gasify the carbon in the reactor. The reaction rates are a function of gas composition and can thus be determined once the mass balances have been completed. The length of the reactor is determined by the reaction rates in the reactor, whereas the actual bed configuration, the area of the gasification grid plate, and the height of the bed are determined by the fluidization characteristics of the gas-solid mixture.
4. Finally, the reactor costs are determined by sizing the wall of the reactor according to temperature, material, and pressure. Overall reactor costs can be estimated from the weight of material required.

B. Mass and Energy-Balance Calculations

The calculational procedure considers both the mass balance around the reactor and the equilibrium constraints. Exxon research[1-3] indicates that, with potassium carbonate as a catalyst impregnated in a char, the gas-phase shift and methanation reactions are brought to equilibrium at almost all reasonable flow rates. The shift and methanation reactions are, respectively,

$$CO + H_2O \rightarrow CO_2 + H_2 , \qquad (1)$$

$$3H_2 + CO \rightarrow CH_4 + H_2O \ . \qquad (2)$$

The mass-balance calculational procedure is very flexible because it allows for multiple inputs of all the molecules involved and gives the outputs of all these molecules in the proper amounts to account for the mass balance and for the equilibrium constraints. The equations as written can be considered as gasifying one mole of carbon. All other streams are normalized to this quantity. Therefore, the overall mass balance is based on one mole of char carbon flowing into Gasifier 1.

The calculational procedure has two variables, R_H (the ratio of water to hydrogen in the outlet stream) and K_1 (mole fraction of carbon monoxide in the outlet stream), which must be set to calculate all other streams. The calculation produces the number of moles, m, of water required to satisfy both the mass balance and the pressure, P_t, of the gasification reaction, as determined by the methanation equilibrium. Thus, because the values of m and P_t have already been determined by the design and by the flow of gases into the reactor being considered, a two-variable iteration must be performed to adjust R_H and K_1 to produce the correct values of m and P_t. The calculation was performed on an HP-25 calculator; in general, for each reactor, between six and thirteen calculations were required to achieve convergence.

Once the overall mass balance and equilibrium conditions are satisfied, the net reaction for the gasifier can be determined by subtracting the inlet flow rates from the outlet flow rates. The overall energy requirement or production for the gasifier is then easily determined by enthalpy calculations involving the reactants and products. In this case, the enthalpies are calculated at the reactor temperatures, with all the products in the gaseous phase.

As the overall result of the calculation, both the composition of all of the gases leaving the reactors and the energy requirement, or production, in each of the gasifiers are determined.

C. Reactor Design

The size of the gasifiers can only be determined after the reaction rates within the gasifiers are set. The reaction rates are a function of temperature and gas composition. The most important and recent work concerning rates of gasification for the catalyzed potassium carbonate char system has been recently published by Vadovic and Eakman of Exxon.[3] Their experimental data and the proposed rate equations have been reviewed carefully, and the equations, with some minor modifications, have been used in our discussion. Exxon's equilibrium and mass-balance calculations were made to simulate their experimental data, and then the experimentally observed rates of reaction were compared with the proposed rate equations. Our one major change in the rate equations was the elimination of the back reaction term associated with the CO/H_2 reaction to produce carbon. When the term was included, the results of the rate equation did not match the experimental data.

In addition, Ref. 3 does not include the possible carbon-hydrogen reaction and gasification through that step. Therefore, additional catalytic gasification work, by Gardiner, Samuels, and Wilkes,[4] and an additional rate equation were used to account for the carbon-hydrogen reaction. Again, the work of Vadovic and Eakman was simulated by utilizing the two equations, the modified Exxon equation and the equation of Ref. 4. The results were very satisfactory.

Therefore, once the composition of the gas leaving a given reactor was determined, the associated reaction rate equation was calculated with the two rate equations. By using a simple design equation, the length of the reactor (or the volume of the reactor) can be determined. In this case, the overall diameter of the reactor is 6 m, and the length was varied to give the proper volume. The design of the reactor is very similar to that proposed by Juntgen and Van Heek.[5,6] In their proposed gasification system design they utilized a pebble bed type nuclear reactor. The gasifier they proposed is a horizontal gasifier, 6 m in diameter, with the gases flowing vertically through a grid plate along the bottom of the tank. The advantage of this type of gasifier is a concentration gradient in the carbon ash-solid phase that can be maintained along the length of the reactor, so that a more carbon-free ash can be produced.

Once the length of the holdup volume in the reactor is determined by the appropriate reaction rates, then the final configuration of the fluid bed can be determined by the requirements of fluidization. A specific velocity must be maintained upward through the bed by the gas mixture to achieve the desired fluidization. Therefore, the configuration as far as height and width of the bed are concerned, must be modified to adjust the gas flow rate to achieve the required fluidization velocities. In our case, the density of the bed was maintained at 0.2 tonne/m^3, and the flow rates were in the 0.2- to 0.3-m/s range.

D. Reactor Costs

A rough estimate of the gasification reactor costs can be made by determining the weight of the reactors and ascribing a certain cost per unit of weight depending upon the material being used. Thus, the wall thickness is calculated for the pressure (in this case, 68 atm) and temperature of the system and the type of material being used. The overall costs according to weight were determined from Ref. 7.

E. Results of Calculations

The energy balance results from the mass and equilibrium calculationsare presented in Table I. The interesting part of the calculation, of course, is the energy requirement per gasifier on the ash end of the chain (Reactors 10, 9, and 8). A relatively small amount of endothermic energy is required. In the middle of the chain the gasifiers are almost thermally neutral. Then, in Sections 1, 2, 3, 4, and 5, a small amount of exothermic energy is released. The overall energy requirement per mole of carbon gasified in endothermic energy is -4.4 kcal/mole of char carbon. The amount of exothermic energy released at the carbon-rich end of the chain is +3.3 kcal/mole of char carbon. Recycling the carbon monoxide, hydrogen, and carbon dioxide did achieve the results expected. The net reactions for the gasifiers in most cases resulted in a direct conversion of water, hydrogen, carbon monoxide, and carbon into carbon dioxide and methane.

The overall energy requirements could possibly be further reduced in a more complicated flow system, with water being injected at appropriate points along the gasifier chain. However, the results for our simple countercurrent system are very encouraging and do indicate that the overall energy requirements for catalyzed gasification can be substantially lower than those for thermal gasification systems.

OVERALL ENERGY BALANCE, EQUIPMENT DESIGN, AND HEAT INTEGRATION

A. Energy Balance for the Gasifier System

Because only small quantities of energy, either exothermic or endothermic, were involved with each gasifier, the design of the gasifiers was somewhat simplified. Internal heat exchange was not required, and therefore, all energy to or from the gas streams for these gasifiers was transferred in external heat exchangers. As indicated in Fig. 1, each gasifier has an external heat exchanger following the gasifier in the exit gas stream. All the heat energies required by, or given off in, are shown in Fig. 1. The temperature gradient in the gasifier chain was such as to provide higher rates of reaction per unit of carbon in the low-carbon-concentration end of the chain. Thus, Gasifier 10 was at a temperature of 850°C, and Gasifier 1 was at 704°C. The lower temperature of Gasifier 1, at 704°C, was set so as to produce more methane; the equilibrium toward methane is enhanced at lower temperatures. The results of the overall enthalpy balances around the heat enchangers associated with the various gasifiers is shown in Fig. 1. Enough energy is either supplied to, or removed from, the heat exchanger so as to give the desired reaction temperature in the specified gasifier. Because the energy has to be removed from each exit gas stream to go to a lower temperature in the succeeding gasifier in moving from Units 10 to 9, 8, and lower, the actual energy required by the high-temperature end of the gasifier chain is minimal. Only Heat Exchangers 9 and 8 required energy. All other gasifier heat exchangers provided energy for the rest of the gasifier system.

B. Gasifier Output Cooldown Chain

The gases exiting from Gasifier 1 are at 704°C, and must be cooled to a temperature of -113°C to enter into the separation column. Thus, a series of heat exchangers are required for the cooldown and for the condensation of the water held in the stream from Gasifier 1. In Fig. 1, Units 21, 20, 19, 18, 17, and 15 are involved in this cooldown. The energy from this stream is carefully utilized throughout the rest of the gasification process. Interestingly enough, the condensation energy of the water, which occurs between 227°C and 163°C, cannot be effectively used anywhere else in the process. In this particular case, a steam work cycle is utilized to extract 2.06 kcal of work per mole of char carbon from the energy of condensation of 9.41 kcal per mole of char carbon. Otherwise, the integration of heat from this series of heat exchangers works well with the energy required for water evaporation and heatup of the stream leaving the cryogenic distillation system. For the cooldown below room temperature, the cooling energy is provided mainly by the evaporation of the cryogenic liquid methane coming from the distillation column reboiler. In addition, an isentropic expansion of the gases at -93°C and 68 atm is made to -113°C and 35 atm. The cryogenic distillation system is operated at 35 atm of pressure.

C. Cryogenic Distillation Separation of Methane from Carbon Monoxide and Hydrogen

The gasifier stream from Gasifier 1 has been reduced in volume by the condensation of the water at temperatures down to 25°C, and also by the removal of the carbon dioxide, hydrogen sulfide, and ammonia in the acid-gas scrubber system at about room temperature. Therefore, the recycle stream entering the cryogenic system contains only carbon monoxide, hydrogen, and methane. Before entering the cryogenic system, a series of zeolite adsorbers and molecular sieve adsorbers are required to remove the last small concentrations of the acid gases. Cooldown of these gases is achieved as indicated above, with the cooling energy available in the methanation evaporation and by the isentropic expansion. Incidentally, the work available in the isentropic expansion is used for the isentropic compression of the hydrogen and carbon monoxide after the separation. The isentropic expansion produces a small amount of liquid methane and, therefore, the liquid and vapors from the expander are charged into Separator Column 14, as shown in Fig. 1. A fairly large amount of energy is associated with this column. The largest amount of energy is that required to cool the condenser, 14 E. The amount of cooling is not large: 0.90 kcal per mole of char carbon. However, the refrigeration chain required to provide this cooling at -167°C does require ~ 7 kcal of work per mole of char carbon. The cascade of refrigeration is shown in Fig. 1. Incidentally, the energy available from heating the recycle gas up from -167°C to -23°C is utilized to assist in this cascade chain. The design of the cascade is certainly a very preliminary one, and a more efficient system can be conceived. This particular cascade appears to work and gives an estimate of the work required. The energy for evaporation of the methane in the reboiler for the distillation column is taken from Unit 17 to assist in the cooling of recycle gases from room temperature down to -113°C. The methane coming as a liquid from the reboiler is at 35 atm and is compressed as a liquid by pumping up to 68 atm. The evaporation to the gaseous phase occurs thereafter.

The overhead from the distillation column contains only carbon monoxide and hydrogen. It is heated in Unit 12 to -23°C, and then is compressed in Compressor 13 to 68 atm. The work of compression comes from the isentropic expansion of the gases being fed into the distillation column. The recycled gases are then heated to 882 C. Much of the energy comes from other portions of the process, as indicated in Fig. 1. Only 1.12 kcal per mole of char carbon is required from an external heat source such as the fast process helium stream from a nuclear reactor.

D. Evaporation and Heating of Water to 882°C and 68 atm of Pressure

One of the largest energy consumers in this process is the evaporation of water at 68 atm and 280°C, and the subsequent heating to 882°C. The units involved with this vapor production are Numbers 26, 24, 23, and 22. The high-temperature energy between 750 and 882°C is supplied by the external helium heating source. A portion of the evaporation energy is provided by heat from the gasifier system. However, a large quantity of energy, 14 kcal per mole of char carbon, is required for the evaporation of the water at 68 atm; 280°C cannot be provided by any convenient source within the gasifier and recycling system. This energy must come from an external source, such as the helium stream from the nuclear reactor. Many schemes were investigated to determine whether or not the energy available from the condenser in Unit 20 could be compressed and recycled to heat the evaporators at 280°C, or whether the pressure of the evaporation for Units 22, 24, and 26 could be lower and thus utilize the energy of condenser 20. None of these schemes led to any economy because of the large quantities of energy required in the compression of the vapor to 68 atm once it was produced in the evaporators.

E. Coal Preparation Operations

Units 28, 27, 26, and 25 are involved in the overall preparation of the coal for the gasification process. The coal must be pulverized to a particle size of ~ 0.3 mm in diameter. Pulverization occurs in Unit 28, and the work associated with this step is relatively low, ~ 0.3 kcal per mole of char carbon. In Unit 27 the coal is impregnated with the potassium-carbonate solution, which is recycled from the extractor system and from the makeup solution. The water from this impregnation is then driven off in Unit 26 at 68 atm and 280°C, and is injected into Unit 23 where it is superheated to 882°C. A pyrolysis is indicated in Unit 25. However, in this particular case, the pyrolysis operation will be combined with the gasifier fluid bed in Unit 1 and Heat Exchanger 1 into one unit. The overall energy requirement is about neutral for the combination of these three processes. The units are shown in the flow sheet in Fig. 1, as indicated to specify specifically the base for the calculation, which is one mole of char carbon entering Gasifier 1 after pyrolysis. Table II shows a composition of the gases leaving the pyrolysis at 704°C. This calculation was made by assuming equilibrium of the methanation and shift reactions to estimate the types of molecules that would be produced first during thpyrolysis at this temperature.

F. Other Miscellaneous Operations in the Cycle

The ash-leaching operation is shown as Unit 29. Very little effort went into the study of this unit, and the work required in agitation and pumping is estimated at 0.1 kcal per mole of char carbon.

The pump pushing the recycle gases through the total system is shown as a blower forcing gases into Gasifier 10. With a pressure drop of 2 atm throughout the total system, the amount of work is still small at ~ 0.2 kcal per mole of char carbon.

Also, the energy associated with cooling the ash from Gasifier 10 to room temperature is relatively small at ~ 0.4 kcal per mole of char carbon.

OVERALL RESULTS OF ENERGY CALCULATION FOR INTEGRATED CATALYTIC COAL GASIFICATION SYSTEM

A. Overall Energy Requirements

The overall energy required in the catalytic coal gasification system is summarized in Fig. 1. All energies required or given off by each of the various units are indicated. The energy units are either for thermal energy as heat required in heat exchangers or as work energy required in compressors or blowers. The basis for the calculations as shown in Fig. 1 is given in Table III. Again, one mole of char carbon entering Gasifier 1 is the overall basis for the calculation. To achieve the desired plant size of 3000 MW of higher-heating-value chemical energy in the methane being produced, the number of moles and the amount of coal required are: 5153 moles of char carbon per second. The overall quantity of dry coal from Kentucky Seams 9 and 11 is 377.5 tonnes/hr to provide this quantity of char carbon.

The overall energy balance for the catalytic coal gasification system is presented in Table IV. The energy required falls into three categories. First, high-temperature thermal energy is required by the gasification train in Units 9 and 8, and also in the heating of the water and recycle gases in Units 11 and 23; the total high-temperature energy required is 5.2 kcal per mole of char carbon. The second category is in the low-temperature range of 200 to 280^{o}C; the total energy required for evaporation in this low-temperature range is 14.0 kcal per mole of char carbon. In addition, a fairly substantial amount of work is required in the third category to operate compressors and blowers and other types of mechanical devices in the gasification system. Most of this energy is associated with the work of compression in the refrigeration cycles coupled to the condenser for the distillation column. Fortunately, some work is produced by the system in the expander and by the condensed steam from Unit 20. The net amount of work required by the system is 6.3 kcal per mole of char carbon. Of course, the production of this work requires a substantial quantity of thermal energy at a conversion efficiency of 40%. This requirement is 16.3 kcal of thermal energy per mole of char carbon.

B. Estimated Overall Efficiency of Catalytic Coal Gasification System

A summary of output and input energies for the coal gasification system is presented in Table IV. Most of the input, of course, is associated with the coal and its higher heating value of heat of combustion. For one mole of char carbon, 20.3 g of dry coal are required. The heating value as presented in data from Texas Gas Transmission is 6.80 kcal per gram of dry coal. In addition, the thermal input required, as detailed in the previous section, is 5.2 kcal per mole of char carbon for the high-temperature energy and 14.0 kcal per mole of char carbon for the low-temperature energy. Finally, a work energy of 6.30 kcal per mole of char carbon is necessary to run the mechanical equipment. The thermal energy associated with this quantity of work is 16.3 kcal per mole of char carbon. The sum of the total input is 173.4 kcal per mole of char carbon.

The output of the system is methane, ammonia, and hydrogen sulfide. Each of these compounds has heats of combustion, and the total energy associated with their production rates, as indicated in Table V, is 144.4 kcal per mole of char carbon. The overall efficiency of the plant, thus, is 83.3%. In other systems, gasification efficiencies associated with a temperature of 1000°C from a high-temperature gas-cooled reactor for synthetic natural-gas production are ~ 58%.[5]

UTILIZATION OF A NUCLEAR REACTOR TO DRIVE THE CATALYTIC COAL GASIFICATION SYSTEM

The fit of the thermal and power requirements of the catalytic coal gasification process if driven by a high-temperature helium stream from a high-temperature gas-cooled reactor is shown in Fig. 2. The fit is very good. High-temperature energy is required for part of the system. Enough low-temperature energy is required to permit a low-temperature recycle of 340°C back to the nuclear reactor. A middle portion of the helium heat is available for power production, which is directly required in the gasification system. As indicated in Fig. 2, the temperatures and quantities of energy fit well with a declining temperature in the helium stream from the nuclear reactor.

The total energy required is 35.4 kcal per mole of char carbon. The char carbon flow is 5153 mol/s to produce 3000 MW of higher heating value of chemical energy in the products. For this rate the nuclear reactor must have a capacity of 765 MWt. Unfortunately, such a nuclear reactor may be too small to be economical in this application. However, we do not itend to assess the viability of this kind of reactor for driving such a gasification system, we only wish to emphasize that the fit with the nuclear heat source from the nuclear reactor is excellent.

In summary, the system described herein is very efficient thermally. Almost all the energy sources and sinks are considered in this system. The overall efficiency of 83% is very high and warrants a continued study of the particular process. Optimization has not been tried, and even higher efficiencies may well be attainable.

To date, none of the equipment has been designed in detail. The overall gasification train has been approximately sized. The gasifiers are 6 m in diameter, and the total length of the gasification train is 185 m, if a 2%/min reaction rate of carbon existed throughout the chain. The next step in this analysis would be to design the gasifiers more carefully; the required rate equations are available. Neither has the rest of the system been designed in detail. In particular, the distillation column has not been designed because the phase equilibria were not determined in our study. An overall design of the distillation column can, however, be developed without great difficulty. The next step, therefore, would be to continue with the design and to determine whether or not the equipment costs of the overall system are competitive with or better than those of comparable second-generation coal gasification systems.

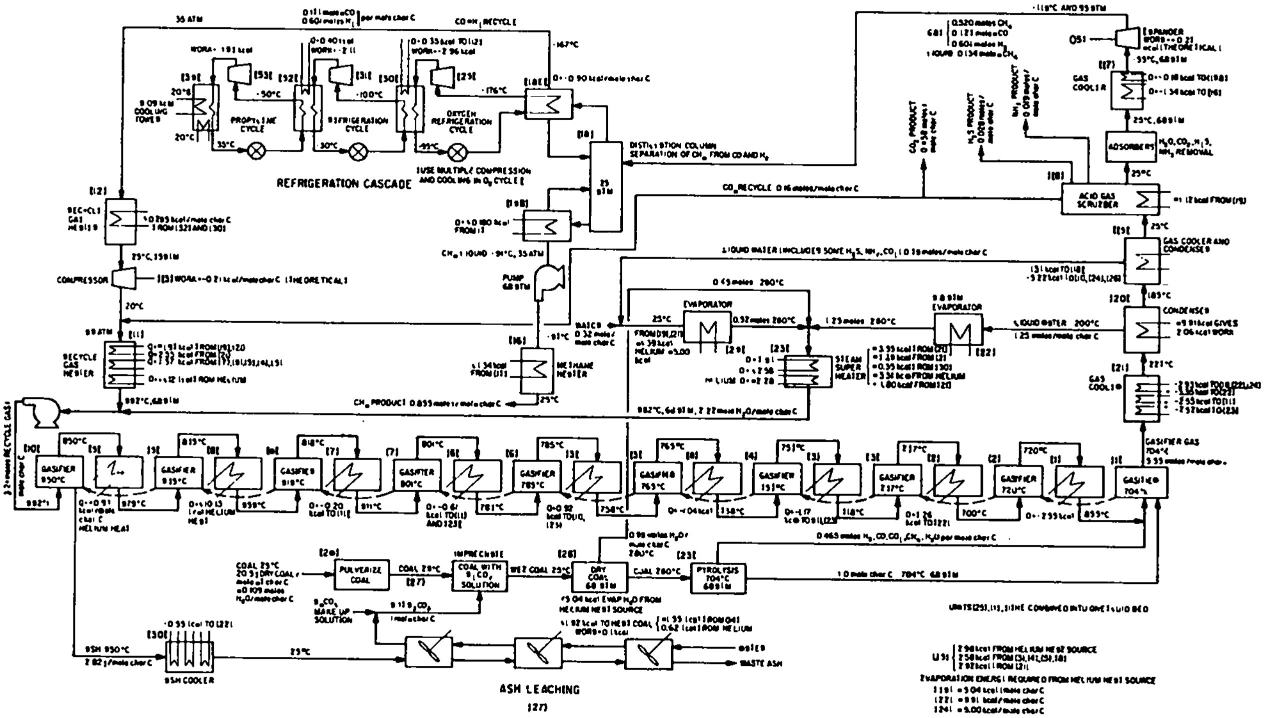


Figure 1. Catalytic coal gasification system.

TABLE I

ENERGY REQUIRED OR RELEASED FROM GASIFICATION REACTIONS IN GASIFICATION TRAINS

Gasifier	Energy Required, kcal/mol C (as char) gasified	
1 (Coal End)	- 1.774	
2	- 0.795	
3	- 0.684	Total energy released
4	- 0.573	=-4.397 kcal/mol C
5	- 0.443	
6	- 0.129	
7	0.292	
8	+ 0.654	Total energy required
9	+ 1.391	=+3.321 kcal/mol C
10	+ 0.984	

NOTE: 0.653 mole CH_4 produced/mol C (as char) gasified. Based on coal (day): 6182.2 kcal/kg coal (day) and 49.3 mol char C/kg coal (day).

TABLE II

COAL EQUIVALENT AFTER PYROLYSIS FOR KENTUCKY COAL (Nos. 9 and 11)
- TEXAS GAS TRANSMISSION -

(1 kg of dry coal after pyrolysis at 700°C - An estimate of products)

	moles	kg	moles/mole char C
C (char)	49.29	0.591	1.000
H_2	7.55	0.0 151	0.153
H_2O	2.60	0.0 461	0.0527
CO_2	0.55	0.0 242	0.0111
CO	1.28	0.0 357	0.0259
CH_4	5.30	0.0 848	0.1075
NH_3	0.94	0.0160	
HCl	0.03	0.001	2.819×10^{-3} kg/mol char C
H_2S	0.38	0.047	
Ash	---	0.139	
		1.000 kg	
H_2O with Coal	5.39	0.097	0.1093

TABLE III

BASIS AND PRODUCTION RATES

Calculational Basis: 1 mole of char C to Gasifier 1.

For 3000 MW in HHV chemical energy in CH_4 produced. The following rates are required.

5152.6 moles char C/s

3364.6 moles CH_4/s

104.6 kg dry coal (Kentucky Nos. 9 and 11 seam)

377.5 tonnes dry coal/h

9037.2 tonnes dry coal/day.

TABLE IV

ENERGY REQUIRED BY CATALYZED COAL GASIFICATION PROCESS

Heat Energy

Unit	kcal/mole char C	Temperature range
(11)	+1.12	767-882°C
(9)	+0.93	850-879°C
(8)	+0.13	855-839°C
(23)	+2.98	751-882°C
TOTAL:	5.16 High temperature energy.	
(22)	+3.31	200°C
(26)	+5.04	200°C
	+0.62	200-280°C
(24)	+5.00	280°C
TOTAL:	13.97 low temperature energy	

Work Energy

(29)	2.96
(31)	2.11
(33)	1.93
(29)	0.1 kcal est.
Recycle Gas Blower	0.24 kcal
Misc. Pumps	1.0 kcal est.
(28) Pulverizer	0.36 kcal
(13) Compresser	0.23
TOTAL:	-8.83 kcal/mole char carbon

Work Available from process:

(20) Condensed Steam	+2.06
(15) Expander	+0.27
TOTAL:	+2.33

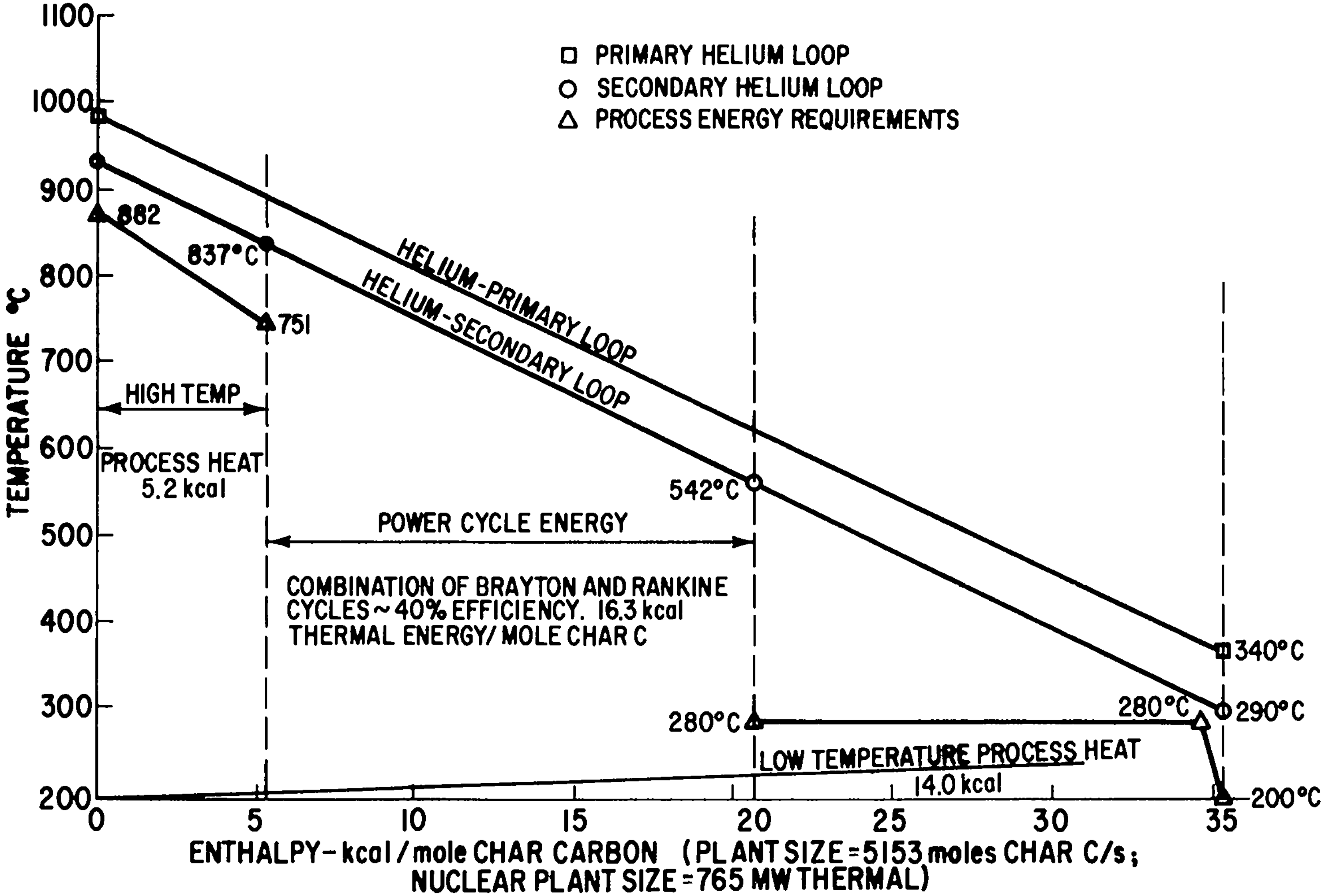
Net work required = -6.50 kcal/mole char C.

TABLE V

ESTIMATE OF OVERALL ENERGY EFFICIENCY

OUTPUT:		
Energy in methane: (HHV)		
0.653 mol/mol char C x 212.8 kcal/mol =		138.9 kcal/mol
Energy in NH_3 and H_2S		5.5 kcal char C
TOTAL:		144.4 mol char c
INPUT:		
Coal:		
20.3 g dry coal/mol char C x 6.80 kcal/g dry coal		137.9 kcal mol char C
Thermal Energy:	High Temperature	5.2
	Low Temperature	14.0
Work Energy:	6.30/(0.4 eff. of conv.)	16.3
TOTAL INPUT:		173.4
Overall Plant Efficiency		83.3%

Figure 2.



□ PRIMARY HELIUM LOOP
○ SECONDARY HELIUM LOOP
△ PROCESS ENERGY REQUIREMENTS
TEMPERATURE °C
1100
1000
900
800
700
600
500
400
300
200
882
837°C
751
HELIUM-PRIMARY LOOP
HELIUM-SECONDARY LOOP
HIGH TEMP
PROCESS HEAT
5.2 kcal
542°C
POWER CYCLE ENERGY
COMBINATION OF BRAYTON AND RANKINE CYCLES ~ 40% EFFICIENCY. 16.3 kcal THERMAL ENERGY/ MOLE CHAR C
340°C
280°C
290°C
280°C
LOW TEMPERATURE PROCESS HEAT
14.0 kcal
200°C
0
5
10
15
20
25
30
35
ENTHALPY- kcal / mole CHAR CARBON (PLANT SIZE = 5153 moles CHAR C/s; NUCLEAR PLANT SIZE = 765 MW THERMAL)

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THE NUCLEAR FUEL CYCLE

Because nuclear reactors utilize materials which were of limited value to our civilization before the beginning of the "atomic age" roughly four decades ago, and because the civilian nuclear power industry is young and still-developing, the industries and government agencies associated with the various portions of the nuclear fuel cycle have limited experience as a basis for planning and projecting the future. The nuclear fuel cycle, shown in a generalized schematic form in Fig. 1, consists of the conversion of fissionable nuclear isotopes into energy (primarily electricity) in nuclear reactor and ancillary activities. Those activities associated with preparation of fresh nuclear fuel (mining, milling, enrichment, etc.) are in the "front" of the cycle in the current terminology, and those associated with processing and disposal of wastes are in the "back" end of the cycle. Currently, because of the absence of recycle of recovered fissionable material from spent fuel, the nuclear fuel cycle is not actually a cycle, but is instead a once-through process.

THE POTENTIAL SUPPLY OF URANIUM

Because the estimates made by different groups and agencies of uranium potentially recoverable for use in the nuclear fuel cycle vary by at least a factor of two, and because this variance is significant in assessing the prospects of the light-water-reactor-based nuclear industry, an effort is made to describe the current projections and their bases.

The ultimate amount of uranium is large; it is widely dispersed in the earth's crust with a mean concentration of about 2-4 ppm.[1,2] It is found in more than 100 chemical forms in a large variety of geological formations. However, concentrations of uranium (400-2500 ppm) that can be mined economically are not common, and the amount that can be recovered economically is uncertain, but is clearly insufficient for building a stable, sustained, fission-based economy that does not include breeder reactors.

Almost all the mining activity in the US has been in sedimentary deposits of sandstone which contain 1000-2500 ppm of uranium. Most geologists believe that these deposits were formed by leaching of the uranium from surrounding volcanic or granitic formations with precipitation from ground water under reducing conditions. Such deposits in the Colorado Plateau occur in beds ranging from the late Paleozoic to Tertiary, but are mostly associated with sandstones of Jurrasic age in the Morrison formation. These deposits appear to be unique.

Extensive deposits of uranium-bearing quartz pebble conglomerates of pre-Cambrian age have been found in Canada and South Africa. These deposits apparently formed in the nearly oxygen-free reducing atmosphere of 2.5 billion years ago, and conatin other valuable minerals including gold and thorium. Uranium has been produced from these deposits as a by-product of gold. The concentrations in the Canadian deposits average 1200-1600 ppm of U_30_8. In the larger South African deposits, the concentrations are lower, generally 300-700 ppm U_30_8. Similar deposits of undetermined size have been identified in Brazil. There is speculation that similar deposits could lie at some depth under Montana, but no such formations outcrop on the surface in the US.

Large quantities of uranium are known to be present in western US lignites at concentrations between 100 and 2000 ppm. Higher concentrations, up to 5000 ppm, have been found in the ash after burning. (Western sub-bituminous coals contain much lower concentrations, in the range of 10 ppm).

Most phosphatic rocks contain uranium, with increased uranium content associated with increased phosphate content. Marine phosphorite deposits, commonly 5-10 feet thick, underly thousands of square miles of Idaho, Montana, Wyoming, and Utah. These beds have uranium contents in the range 10-750 ppm. In Florida, phosphate deposits cover several hundred square miles and contain uranium at the 120-240 ppm level. Similar deposits with concentrations near 100 ppm occur in countries along the Mediterranean Sea from Israel to Morocco. Somewhat richer uranium-bearing phosphate deposits have been found in central Africa and Brazil. Such deposits also occur in the USSR.

Black shales, resulting from marine deposits of organic-rich material, contain minor amounts of uranium. The upper portion of the Chattanooga Shale which covers several thousand square miles of Tennessee, Alabama and Kentucky to a thickness of 12-18 feet, contains uranium at average concentrations near

60 ppm. This formation extends, at greater depth and with lower uranium concentrations (~ 35 ppm), from eastern Tennessee to Texas and Montana with a thickness of 40 feet. Black shales in Sweden are much richer, if less extensive, averaging about 300 ppm.

Igneous rocks contain uranium, usually at concentrations near the average crustal abundance of 2-4 ppm, but some bodies of igneous origin are much richer. The Conway Granite formation of New Hampshire, which is exposed over 300 square miles and extends to a depth of about 1000 feet, contains 10-30 ppm U_3O_8.

Uranium also occurs in veins, filling fissures in many types of rocks and geological formations. Individual veins are a few inches to a few feet wide and extend to depths of from several hundred to several thousand feet. Veins have been mined with concentrations from 1000-10000 ppm of U_3O_8. In the US, such deposits have accounted for an insignificant fraction of the uranium recovered to date. High grade vein deposits in Zaire, formally the Belgian Congo, and Canada, have been depleted. Veins containing uranium at about 1000 ppm and thorium at about 200 ppm form fairly large deposits in Canada. Vein-deposited uranium is being mined at concentrations greater than 1000 ppm in Australia.

Sea water contains about 1.5 ppm. Although the concentration is very low, th total amount of uranium in sea water is very large. There has been a good deal of interest in the extraction of uranium from sea water, particularly in nations without other sources, such as Japan.

From the point of view of the nuclear fuel cycle, the key question concerns the amount of uranium which can be economically extracted as a function of time, rather than the total natural abundance. The answer to this question is central to the development, by both industry and government, of a strategy for dealing with decisions about the LWR fuel cycle longevity, uranium and plutonium recycle, and breeder reactors.

At the present time there is considerable uncertainty about the projected uranium reserves and resources. (Reserves are generally understood to be that uranium which is extractable at today's technology and near today's costs. Resources are more vaguely defined, and this is the hub of much of the disagreement, as that material which is postulated to be ultimately extractable).

Potential resources represent appraisals of the undiscovered uranium that is presumed to be present from geological evidence on hand. In recent years they have been broken into several subcategories; "probable" potential resources as extensions of known deposits or evidenced by exploration experience; "possible" potential resources as new deposits in geological formations known to be productive; and "speculative" potential resources in formations which have not been previously shown to be productive.

Mineral reserve and resource assessment, whether oil, copper, or uranium, is fraught with difficulty as a matter of course. In addition, uranium has a special set of circumstances. It has only been in production for 25 years or so. Until the late 1950's the US government purchased most of its uranium abroad, from the Belgian Congo, Canada, and South Africa.[3] In addition, originally there was a single buyer, the US government, for a non-commercial defense-related use. Commercial buyers did not emerge significantly until a decade ago. In this environment of government monopoly, the concept of "forward cost" was developed early in the history of the uranium mining industry to give a measure of resource supply. This cost is based on projected capital and operating costs. These forward costs, given as \$8/lb, \$15/lb, etc., are used to represent categories of uranium reserves which are intended to represent a supply function. Because costs for exploration, property purchase or lease, mine development, or other work costs or profit are not included, the projected costs do not reflect actual costs, and they are not related to actual selling price. Also, the forward costs used to categorize reserves refer to the upper cutoff costs for a category of material, and not an average cost for that category. In addition, because of inflationary pressures, the categorization of material at a given cutoff cost will bracket a different segment of material at the same cutoff cost in a different year.

For these reasons, the supply function represented by these forward cost categories has led to a good deal of confusion in recent attempts to use such data in decision making about preferred strategies for the nuclear fuel cycle. For many years \$8/lb of U_3O_8 was the reference resource cost used in the statistical data base. Currently, most data is referenced to \$15 or \$30 per pound material.

There is little or no disagreement about the amount of uranium in the proven reserve category. The resource estimates, however, vary by a factor of two in recent and on-going studies. The Ford-Mitre report[4] which appeared in 1977, argued that sufficient reserves and resources at $40/lb (1976 dollars) will be available to meet requirements for a LWR economy through the year 2000 and, at costs to $70/lb (1976 dollars), well into the next century. This report is of interest because it apparently reflects, to some degree, current policy.

The Ford-Mitre report argued that because early estimates were based on material recoverable at $8/lb by the government, industry had little incentive to explore for more expensive uranium and for this reason, uranium resource data are based on the low side. The increase in uranium prices in the past few years, they argue, will lead to additional supplies which will be further expanded by continuing price increases. They give, as their judgment, that uranium resources should be capable of development to support LWRs up to and beyond the end of the century at costs of about $40/lb (1976 dollars).

Ford-Mitre based its analysis on resource assumptions of 2.5 million tons of uranium at $30/lb and 5.5 million tons at $100/lb. These estimates in turn, rested on resource data indicating that US resources at $30/lb are projected at 3.7 million tons[5] (Table I). Of this total, 17% are classified as reserves, another 4% as reserves as byproducts of copper and phosphate production, 29% as probable resources, 34% as possible resources and 16% as speculative resources.

The Ford-Mitre report points out that 95% of the US reserves and 80% of the probable resources are in sandstone, whereas uranium is found in the rest of the world in other types of formations (Table II). Canada's uranium is found in veins and conglomerates, Africa's in a variety of rocks, but mostly veins and conglomerates, Sweden's is in shale (richer by a factor of 5 than the Chattanooga Shale), Brazil's in granite and conglomerate, and in veins or other non-sandstone formations in the rest of the world. It seems odd that such a difference in the nature of the deposits between the US reserves and those of the rest of the world should exist. Either major deposits of uranium in formations other than sandstone do not exist in the US, or the exploration in other formations has not been adequate to uncover major deposits.

Geologists have suggested that, in the US, alternatives to the sandstone deposits could be found in vein deposits, "porphyry" deposits similar to South Africa, and as a byproduct in lignite, phosphate rocks and copper ores.[6] Conglomerates in the Great Lakes region are possible sources. Abundant quantities are present in granites and shales, but at recovery costs estimated to be substantially above $100/lb. To mine the Chattanooga Shale would require an equivalent tonnage of ore to be mined to support a 1000-MWe nuclear plant as is required to support a coal plant of equal capacity, with the associated environmental impacts of large strip mines.

Several recent studies are more pessimistic about the ability of the uranium industry to meet projected demands.[7,8] Preliminary reports from the Committee on Nuclear and Alternate Energy Sources (CONAES) working under the National Academy of Sciences, project only 1.76 million tons of US uranium reserves and resources. This figure represents 640,000 tons of $30/lb reserves, the same as the previous estimates, but only 1.06 million tons of resources at $30/lb. Further the CONAES study predicts less than 3 million tons of resources at $100/lb.

DOE's preliminary 1978 reserve estimates are in bold contrast to the CONAES study results. They estimate $30/lb reserves up to 690,000 tons and potential resources at 3.5 million tons.

It is certainly beyond the scope of this study to resolve the issue. The divergence of expert opinion is an indication of the incomplete data upon which these estimates are made.

Factors that indicate that further exploration will confirm the higher estimates include:

- to date, there has been a good correlation between drilling footage and discovery rate;
- higher prices will encourage a search for lower grade ores (the US has depended on uniquely rich uranium depoists in the past);
- there are many areas known to be mineralized but not fully explored;
- due to the limited state of knowledge about uranium deposits, it is likely that new deposits will be found; and

- the historically uncertain uranium market has hindered exploration, and with a stable market, the exploration effort could be greatly increased.

On the other hand:

- the most favorable areas in some basins have been explored already, so the correlation between drilling footage and discovery rate will continue to decline, having fallen from 14.7 pounds of U_3O_8 per drilling foot in 1955 to 4.7 pounds per foot in 1971;[2]
- while known mineralization is widespread, most of the reserves are found in a few deposits (95% of reserves in 150 of 4500 properties listed);
- there is a deficit of information about lower grade (500-1000 ppm) deposits in sandstone; and
- drilling in areas previously estimated to contain potential resources has not always been successful and exploration experience has contradicted projections of potential ore in the past.

URANIUM PRODUCTION - MINING

Beyond the question about the amount of uranium potentially available, there is the near-term question about the adequacy of the rate of extraction to meet projected demand.

Projections of uranium demand have been nearly as volatile as projections of ultimate supply during the past several years. Every year since 1974 has resulted in a lowering of sights on the part of forecasts of nuclear demand to the year 2000. Often the low-range projection of one year has become the high range of the next.

The National Energy Plan (NEP) released by the President in April 1977 envisaged energy requirements of about 93 quads in 1985, and assuming continuation of policies consistent with the President's original energy policy, nearly 124 quads in 2000.[9] Without NEP, i.e., without the policies in the original energy bill, projections in late 1977 were for energy demand of more than 137 quads in 2000. This is quite similar to the mid-range projection of 1976.

The NEP foresees nuclear growth at 16% per year until 1985, and at a slower 7.3% rate between 1985 and 2000. This projected growth in nuclear power is faster than that of coal (6.4% to 1985, and 2% annually from 1985 to 2000). This rate of nuclear growth is projected despite the delay of the breeder and the limitation on reprocessing of spent fuel. The 1977 forecast for installed nuclear capacity was for 127 GWe to be installed in 1985 and between 380 and 440 GWe in 2000. The lower value assumes NEP policies are followed, and the higher value rates on the assumption that they are not. The demand for uranium, assuming NEP in force, between 1977 and 2000 is on the order of a million tons of U_3O_8 (Table III).

If the trend of an annual reduction in forecasted demand continues, as it seems it will, with some indications of less than 300 GWe by 2000 in the low range of the latest estimates, the uranium demand values should be scaled down accordingly. The latest mid-range estimates are for 111 GWe in 1985 and 325 GWe in 2000. The corresponding low and high estimates for 2000 are 256 and 396 GWe, respectively.

Note that these requirements are quite significant when compared to the reserves and resources.

These cases do not allow for reprocessing spent fuel and using the recovered uranium and plutonium to fuel LWRs. The spent fuel inventory, if processed for uranium to 2000, could reduce the U_3O_8 demand by 20%. If the spent fuel could be reprocessed for uranium as well as plutonium, the savings in U_3O_8 would grow to 30%.

Another 30% could be saved by more efficient enrichment of the fissile uranium-235 isotope. Operation of the gaseous diffusion plants is optimized, at current economics, with relatively inefficient recovery of the uranium-235, characterized by a "tails" or waste stream content of uranium-235 at an assay near 0.25%. Development of more efficient laser-isotope-separation processes could allow almost complete recovery of uranium-235 with an attendant reduction of U_3O_8 requirements.

There seems to be little chance that breeder reactors could significantly enter the picture before 2000. The Ford-Mitre report suggests 2020 as the earliest time for breeder entry, and then on a small scale, initially.

With current production less than 13,000 tons per year (11500 in 1975, see Table IV), expansion to the annual requirements indicated in Table III will require a significant increase, more than a doubling by 1985, in mining and milling capacity. Added to the capacity requirements shown in Table III is the effect of reduced capacity and recovery efficiency in the milling process with declining ore grade.

There is serious concern that uranium production could be limited by the rate of expansion of mines and mills.[10] Industrial milling groups worry that overcapacity in milling could result from a lag in the expansion of mining capacity. Conversely, mine operators are concerned about the falling projections of demand, which could lead to a limited market, on one hand, and a potential lag in milling capacity expansion on the other. Over the next ten years, the rate of expansion of production capability is a key issue. Planned uranium capacity additions are shown in Table IV.

Uranium mining and milling operations are very capital intensive. The domestic uranium mining industry uses variations of established mining techniques to exploit the sandstone deposits. The larger deposits are mined by open-pit methods where the depth is less than 350-400 feet. At greater depths, underground techniques are used. About 70% of the US deposits are being worked as underground mines, 20% by open-pit, and the remainder are by-products or in low-grade stockpiles.

In most open pit mines the relatively unconsolidated sands can be stripped by wheeled scrapers, dozers and power shovels. (Exceptions are the Jackpile and Paguate pits near Grants, NM.).

The underground mines employ the same mining techniques as used for similar deposits of other minerals. Vertical shafts are used to get to large deep deposits. Most mines drive drifts under the ore body. Nearly all mining is a variation of room and pillar or long wall retreat. The long wall allows recovery of up to close to 100% of the ore while the room and pillar method requires 20-30% of the total reserve to be left in place for roof support.[11]

Almost all the uranium mined currently is extracted by conventional techniques. However, increasing attention is being given to solution mining (the extraction of the uranium without extraction of the rock). About two-thirds of the yellowcake production costs are incurred in stripping and extracting the ore. Thus, a lower cost alternative has the potential for producing substantial savings. There are deposits which are not economical to exploit with conventional techniques, which could be potentially extracted with solution mining techniques. The solution mining technique, in a general sense, involves the injection of a leaching solution into the mineralized zone through a series of drill holes. The leaching solution migrates to a production well where it is recovered and processed through a conventional mill. It has been estimated that, by 1985, 10% of the domestic uranium production could come from solution mining if the current R&D effort results in a successful technology base.

The milling process involves the treatment of a uranium ore to produce a high-grade concentrate. The method of treatment is determined largely by the composition of the uranium mineral and by other minerals present in the ore. A schematic of the milling process is shown in Fig. 2. The ore is crushed, wet or dry, and then ground, usually wet in rod or ball mills. Leaching is performed with either an acid or an alkaline carbonate. The most common acid is sulfuric, the most common alkaline carbonate is sodium carbonate.

The quality of the product varies with a large number of parameters. The purity, given as the content of the oxide, U_3O_8, usually ranges from 75% to nearly 100%. The quality of the uranium ore has been dropping over the past decade or more. In 1966 the average concentration of U_3O_8 in the ore mill feed was 2300 ppm. In 1976, it had dropped to 1500 ppm. Concurrently with this drop in ore feed quality, the efficiency, defined as recovery of contained U_3O_8, has also declined from about 95% in 1966 to about 93% in 1976. Because of the declining ore quality, the ore processing rates or throughput capacities have nearly doubled from about 12500 tons of ore per day in 1966 to about 24000 tons of ore per day in 1976, while the uranium concentrate production has only slightly increased ($\sim$ 10500 tons of U_3O_8 in 1966 to 12600 tons of U_3O_8 in 1976). In 1976, the 18 domestic mills operated close to capacity. Table V shows the capacities of the mills in operation in 1973.[12]

UF_6 CONVERSION

The UF_6 industry has been relatively trouble-free to date. The industry is concentrated in five firms world-wide, two of which are located in the US. The US Government also has UF_6 conversion facilities at Paducah, Kentucky.

The first commercial UF_6 plant in the US was built by Allied Chemical Corp. at Metropolis, Illinois. This plant has a capacity of 12700 metric tons of uranium (MTU)/yr, accounting for 35% of the existing world-wide UF_6 production capacity. It uses the dry-fluoride volatility process shown schematically in Fig. 3. The volatility process utilizes fluid bed reduction, fluid bed hydrofluorination, and fluid bed fluorination to produce impure UF_6. The impure UF_6 is then double distilled to produce the 99.5% pure UF_6.

The other domestic manufacturer of UF_6 is Kerr-McGee which has a nearly 5000 MTU/yr facility at Gore, Oklahoma. This plant is presently in the process of being effectively doubled to about 9000 MTU/yr. This plant uses a solvent extraction-fluorination process shown schematically in Fig. 4. In this process, the uranium solution is first purified by solvent extraction with tributyl phosphate, then heated to form UO_3 which is reduced to UO_2 prior to fluorination. The UO_2 is converted to UF_4 by anhydrous HF and the UF_4 is reacted with elemental fluorine to produce UF_6. This process yields a product with a purity of 99.97% and with an overall yield of 99.5%.

The main problem facing UF_6 producers is associated with impurity levels in the U_3O_8 concentrates entering their plants. There are two reasons for this. The first has to do with the changing nature of the uranium mining industry. As new deposits are developed, changes in the levels and nature of impurities in the concentrates may force modifications to the processes. The other problem with impurities has to do with receiving concentrates from different sources controlled by a utility rather than the UF_6 manufacturer.

In perspective, UF_6 conversion is effectively a trouble-free portion of the nuclear fuel cycle. There are indications of a shortage of UF_6 conversion capacity as soon as 1981. The UF_6 conversion industry faces the same uncertainty as the rest of the nuclear fuel cycle, however, and this is affecting expansion plans. The potential cancellation of nuclear power plant orders could significantly decrease the demand for conversion services.

URANIUM ENRICHMENT

In natural uranium, the uranium-235 is present as a trace impurity in uranium-238 at approximately 0.7%.

Light water reactors require fuel with a fissile uranium-235 content of 2.8% or higher. The current technology for enriching the assay of the uranium-235 isotope is based on three government-owned gaseous diffusion plants at Oak Ridge, Tennessee, Portsmouth, Ohio, and Paducah, Kentucky. The next increment of enrichment will utilize the gaseous centrifuge process. A very active program in advanced isotope separation (AIS) technology holds the promise of a much more efficient process which could make significant contributions in the 1990s.

A gaseous diffusion plant depends on the very slight differences in weight between $^{235}UF_6$ and $^{238}UF_6$ which results in the molecules of $^{235}UF_6$ moving at a slightly higher velocity than those of $^{238}UF_6$. A high-pressure stream of hot UF_6 vapor flows past a porous membrane, or barrier, and because the $^{235}UF_6$ molecules move faster, they pass through the barrier at a slightly higher rate than the heavier $^{238}UF_6$. The separation coefficient is not large, and so the process must be repeated in many stages in series to achieve the desired enrichment. For example, to enrich the uranium to 4% uranium-235 requires about 1500 stages. The detailed specifications of the gaseous diffusion process are tightly held by the government which owns all of the domestic enrichment capacity.

The three government plants work together to produce materials of varying assay for military and civilian needs. The gaseous diffusion plants were constructed between 1943 and 1955 to a capacity of about 17 million separative work units (SWUS). Separative work is a measure of the increase effected in the value of the effluent streams with respect to the feed stream. It requires about four units of separative work to produce 1 kg of 3% uranium-235 from natural uranium with a tails assay of 0.2%.[13] Two programs are currently underway to modernize these facilities and to increase their capacity to 27 million SWUS.

The gaseous diffusion process is characterized by high capital costs (the facilities are very large) and high electrical demands (over 7000 MWe will be required for the expanded facilities). Even so, the energy requirement of the enrichment step represents only 4.4% of the final electrical power produced in the nuclear fuel cycle. For many years, the high cost and large minimum size of this process served as a major barrier against proliferation of nuclear weapons.

The ultra-centrifuge process also depends on the mass difference between the vapor phase $^{235}UF_6$ and $^{238}UF_6$ molecules. A more efficient separation can be affected in an ultra-centrifuge than in a single-stage of gaseous diffusion. Hence, to produce a significant enrichment requires only a few stages of centrifuges in series. Because the throughput of centrifuges is limited, however, a large number of units are required to work in parallel with each other to achieve the required separative work requirements. A centrifuge plant is, therefore, large and capital intensive, but advances have been made in centrifuge technology which allow operation with only a fraction, about 4%, of the electrical power requirements of an equivalent diffusion plant. A 2.2 million swu per year increment of separative capacity is being added to the Portsmouth, Ohio diffusion facility. This add-on facility is expected to come on line in 1989, which illustrates the long lead times required for both centrifuge and diffusion enrichment technology. The efficiency and inherent scalability of gaseous centrifuge plants has generated a great amount of concern about the proliferation potential of this technology.

Laser enrichment technology is based on the difference in vibrational frequencies of the two isotopes rather than on mass differences. Very high separation factors can be achieved in these processes. Two approaches are being followed. One utilizes uranium in the molecular UF_6 form and the other uses atomic uranium vapor. The details of the technology are controlled by the government. Projected plant designs which could become available in the late 1980s and 1990s are much less capital intensive (~ $700 million for a 9 million swu per year facility compared to more than $4 billion for an equivalent centrifuge facility) than either gaseous diffusion or centrifuge technology and, the energy requirements are low, on the same order or less than the centrifuge requirements (Fig. 5).

REACTOR FUEL FABRICATION

The enriched uranium must be converted from the volatile hexafluoride form to the stable oxide UO_2, and then compressed and canned, or clad, to be usable as LWR fuel. The method currently dominant in the US is termed the ADU process, for ammonium diuranate, one of the intermediate chemical forms involved. Ammonium diuranate is produced by adding water and then ammonium hydroxide to heated UF_6. Heating the ADU results in U_3O_8 which is reduced at high temperature to form UO_2. The UO_2 is then ground, pressed, and pelletized, after which it is sintered in a furnace. The sintered pellets are then ground to size, cleaned and loaded into cladding tubes to form the fuel assemblies for LWRs. These fuel assemblies, which differ for different types of reactors, are subjected to a vigorous inspection and quality control procedure. A schematic of the individual steps in the ADU fuel fabrication process is indicated in Fig. 6.

The ADU process is only one of four which have been developed to industrial scale. Two, the ADU and the ammonium uranyl-carbonate (AUC), are based on precipitation from aqueous solutions. The other two are dry processes involving fluidized beds or rotating kilns to achieve the decomposition of UF_6.[4] The AUC process, which has been developed in Germany, requires fewer steps than the ADU process.[15] The dry processes, developed at Argonne National Laboratory and United Nuclear Corp., have lower operating costs.[16] The difference in overall cost performance between the processes is small--too small in today's environment to counterbalance the risk involved in going to the new processes. However, it is likely that future expansion of nuclear fuel cycle will involve other methods for fuel fabrication than the ADU-based procedure. Whether such a shift in the process occurs or not, the fuel fabrication step in the nuclear fuel cycle is not a potential trouble area or bottleneck. It is relatively efficient and cost effective.

NUCLEAR REACTORS

There are many potential and actual variations in nuclear reactor design which lead to different reactor concepts. In the US, the dominant reactor concept is labeled the light water reactor (LWR). It exists in two variations, the pressurized water reactor (PWR) and the boiling water reactor (BWR). LWRs require slightly enriched uranium (~ 3% uranium-235) and utilize ordinary, or light, water as coolant and neutron moderator.

While most of the energy produced in the reactor comes from fissioning of the uranium-235, a significant fraction comes from plutonium which is bred from the uranium-238 (which is 97% of the uranium in the fuel) and then fissioned in place. For each gram of uranium-235 in a LWR core, more than one-half gram of plutonium-239 is produced, about half of which is burned or fissioned in place. Reactors in which the ratio of plutonium-239 produced to the initial uranium-235 (or other fissile fuel) loading approaches one are called "converters" and those in which the amount of plutonium-239 produced exceeds the original loading of uranium-235 are called breeders. Isotopes such as uranium-235, plutonium-239, plutonium-241, and uranium-233 are easily fissioned and are equivalently good reactor fuels to a good approximation. Isotopes such as uranium-238 (which constitutes 99.3% of the uranium in nature) and thorium-232 are termed fertile because they can be readily converted, in the right conditions, to one of the easily fissioned species.

While there are not-insignificant differences between PWRs and BWRs and between PWRs of different configurations and manufacture, from the point of view of the overall nuclear fuel cycle, such differences are not important. Hence, the performance of BWRs manufactured by General Electric and of the PWRs produced by Westinghouse, Babcock and Wilcox, and Combustion Engineering will be all considered to be the same for this overview. The major differences concern safety systems and environmental effects. Efficiencies and cost performance for BWRs and PWRs are quite similar.

Other reactor concepts which show promise for possible introduction are the high-temperature gas-cooled reactor (HTGR) in its many variations, the gas-cooled fast-breeder reactor (CGFR), the heavy-water reactor (LMFBR), the light-water breeder reactor (LWBR) and the molten-salt breeder reactor (MSBR). There are several significant variations in design and approach for HTGRs and LMFBRs in particular. In addition, variations and hybrid reactor types such as spectral shift and tandem-cycle (LWR-HWR) concepts have been put forth. Each concept has its advantages.

The LWRs are the basis for the US nuclear fuel cycle and industry, and will retain that role until the end of the century. Currently, there is no recovery, with subsequent recycle, of the uranium and plutonium in the spent fuel and overall fuel efficiencies are low. Even with recovery and recycle of uranium and plutonium, the overall efficiency of the conventional LWR fuel cycle is not high (Fig. 7). The main reasons for the low efficiency are: (a) the loss of fissile uranium-235 to enrichment plant tails, (b) the low thermodynamic efficiency of the LWR caused by relatively low operating temperatures, and (c) the low breeding ratio in the reactor core. In the absence of recovery and recycle of the residual fissile material in the core, the net fuel efficiency of the LWR cycle is very low indeed. Because of the concern about nuclear proliferation and the identification of the fuel reprocessing step in the fuel cycle as most dangerous from a proliferation viewpoint, the very inefficient once-through (no uranium or plutonium recovery) LWR cycle in Fig. 7 is the basis for current policy in the US. Nearly all projections as to the adequacy of the resource and industrial base are made with reference to this LWR reference fuel cycle without spent fuel recovery. Figure 8 shows the effect of enrichment efficiency (as indicated by the assay of the enrichment waste) and of uranium and plutonium recycle. Both laser isotope separation and recycle show significant increases in the efficiency of the light water reactor fuel cycle.

The heavy water reactor (HWR) can utilize natural uranium and effectively burn most of the uranium-235 present. For this reason, losses associated with the enrichment step in the fuel cycle are avoided and a lower penalty is associated with disposal instead of recovery of the fissile fuel value in the spent fuel elements. This type of reactor has been successfully developed as the CANDU series in Canada. While the HWR fuel cycle is more efficient in terms of U_3O_8 to energy conversion than a once through LWR fuel cycle, it is not more so than the LWR fuel cycle with laser isotope separation and recycle of uranium and plutonium. The economics of HWR's do not match those of LWR's and tritium can cause an added environmental problem. These factors have showed the expansion of HWR technology. Figure 9 shows schematically the HWR fuel cycle with efficiencies with and without spent fuel reprocessing.

More efficient and with lower environmental impact than HWRs or LWRs, the high-temperature gas-cooled reactor (HTGR) provides a flexible and versatile concept. Modified HTGRs have been designed for use as process heat sources as well as for electricity generation. They can be operated either with uranium-235 or uranium-233 bred from thorium. Because of the opportunity for expansion of the resource base to include thorium as well as uranium and of the good thermodynamic efficiency as well as the good conversion ratio of fertile fuel to fissile forms, this reactor concept is a very attractive follow-on to the LWR. An extension of HTGR technology is manifest in the GCFR breeder concept. Considerable design flexibility exists to accomodate proliferation concerns (with denatured fuel) or to maximize overall fuel efficiency. The HTGR (Fig. 10) is classified as an efficient converter because the reactor core can be designed so that the breeding ratio approaches unity. Reactors of this type can achieve a significant role during the first half of the next century as advanced converters merging into a symbiotic relationship with fast breeders. Advanced converters such as the HTGR are projected to coexist with fast breeder reactors in a mature breeder environment with an equilibrium ratio of three converters for each breeder.

The light-water seed-blanket breeder reactor utilizes near-term technology to achieve a breeding ratio near or slightly above unity. With the HTGR, the LWBR could fill the gap between the LWR-based industry and a fast breeder (FBR) based industry. These reactors, although developed as near-term breeders, could be effective advanced converters operating on a thorium cycle with uranium-233 recycle. As seen by comparing Fig. 11 with Fig. 10, the LWBR with full uranium and plutonium recycle promises to be more efficient than the HTGR.

As indicated by the efficiencies shown in the schematics illustrating recycle options for the burner and advanced converter nuclear fuel cycles, only a small part of the potential energy in the U_3O_8 or ThO_2 mined is used by LWRs, HWRs and HTGRs. If all of the fertile atoms could be fissioned in a reactor, the fuel efficiency would be very large. However, even in the most efficient breeder reactors, only about one-half to two-thirds of the fertile and fissile atoms will be fissioned. The rest are lost by nonfission capture of in fuel reprocessing. In addition, the thermodynamic efficiencies associated with steam power cycles allow only about 40% of the energy to be converted to electricity. With cogeneration, of course, a large fraction of the remaining energy could be used. The maximum energy yield to be expected in a breeder economy is in the range 3500-4700 million kWhe/short ton U_3O_8 or roughly two orders of magnitude more than the energy yield in the LWR once-through or no recycle option.

This enormous advantage of breeders with respect to fuel efficiency (compare Fig. 12 with 7) is currently being weighed against the potential for nuclear weapons proliferation inherent in such systems. In order to realize the potential of a breeder-based nuclear fuel cycle, the spent fuel must be reprocessed to recover and concentrate the fissile material bred during the residence of the fertile material in the reactor core. The reprocessing involves the most sensitive part of a nuclear fuel cycle with regard to diversion of nuclear weapons materials.

The primary issue in the once-through conventional LWR fuel cycle involves the availability of sufficient high-grade uranium resources to support the industry. The uncertainty is in the range up to 20-40 years. With the lowering of sights in projections of the size of the nuclear-based generating capacity by the turn of the century, sufficiency of resources to 2000 appears probable. However, if the lower projections of uranium resources prove correct, breeder introduction between 2000 and 2010 is necessary to maintain a nuclear option, the ten year uncertainty depending on the reserve and resource uncertainty and on the level of tails assay in the enrichment process.[17] If the higher estimates of resources are correct, the resources may be adequate for several decades beyond the turn of the century. After the high-grade deposits are depleted, extraction of lower quality ore will sustain a nuclear industry but an inefficient fuel cycle will lead to higher costs and substantial environmental disruption at the front end of the fuel cycle.

From the view point of efficiency in the utilization of the uranium and thorium resources available, breeders which utilize a fast neutron spectrum convert the uranium-238, which constitutes 99.3% of the available uranium, into a very high value fuel. Currently the value of uranium-238 is low enough to allow the substitution of uranium for lead in bullets. Similarly, breeders can convert thorium, which has very little value to our society at present, into fissile uranium, uranium-233, which also has a very high energy value.

REPROCESSING SPENT FUEL

The current US policy and planning is based on a once-through fuel cycle after identification of fuel reprocessing as the key to nuclear proliferation. The uranium utilized in the LWR cycle (and in the low assay HTGR cycles) is not suitable for diversion to nuclear weapon production without further enrichment. Because enrichment facilities are either inefficient and thereby very costly, or are technically sophisticated and thereby access-limited, the uranium enrichment step has formed a barrier to nuclear weapon proliferation. This barrier is being eroded somewhat by centrifuge and centrifuge-related aerodynamic processes, but the conversion of reactor-grade uranium to weapons-grade uranium is quite difficult compared to the procurement of weapons-grade plutonium in a spent fuel reprocessing facility. Plutonium is produced from the non-fissile uranium-238 in all reactors, LWRs as well as fast breeders. While much of the plutonium is converted to energy or burned in place, there is a buildup of the fissile plutonium-239 and plutonium-241 isotopes in the fuel.

Breeder reactors are designed so as to optimize the production of fissile plutonium from the fertile uranium-238, but because some plutonium is produced in all reactors fueled with a uranium-235/uranium-238 mix, it is potentially available for separation in a fuel reprocessing facility. Fuel reprocessing to recover and recycle plutonium is an essential part of a nuclear breeder fuel cycle. It is not essential to a LWR-based fuel cycle, although it can extend the resource base by up to 40%.[18]

Although more than thirty[19] different processing techniques have been proposed, all existing reprocessing plants are based on the Purex process (Fig. 13). A plant designed by General Electric at Morris, Illinois based on the Aquafluor process was a complete failure and after a $64 million investment was effectively abandoned. In the US, Nuclear Fuel Services has operated a reprocessing plant at West Valley, New York, for six years, before shutting down for planned expansion. However, due to increasing projected costs to meet environmental costs, this plant was terminated in 1976. A new facility built by Allied-General Nuclear Fuel Services (AGNS) at Barnwell, South Carolina, is complete, but unable to process fuel without government license.

Because the most sensitive portion of the fuel cycle to diversion and proliferation is the step involving the separated plutonium, several modifications of the Purex process, termed coprocessing, have been examined which produce an end product in which the uranium and plutonium are not separated.[20] In the Civex process, in particular, plutonium is never physically separated and is fresh fuel is fabricated in a shielded facility at the spent fuel processing plant. The Civex process[21,22] is designed to be a proliferation-resistant portion of a fast breeder fuel cycle. Coprocessing can be adapted to the reprocessing step in the LWR fuel cycle as well. However, extra enriched uranium would have to be added to the coprocessed product to bring the fuel up to approximately 3% fissile content as required for LWRs. If shipment is required between the reprocessing facility and a fuel fabrication facility, spiking of the plutonium bearing material with a radioactive substance so as to make it very unattractive for weapons purposes has been suggested.

Because of the problem with the GE plant at Morris, Illinois, the termination of the NFS plant in New York, and the delay in authorization of operation in the AGNS plant at Barnwell, a deficit in nuclear fuel reprocessing will exist if a go-ahead is given to LWR fuel recycling. Before such a decision can be made, it will be necessary to satisfactorily resolve the technical concerns about proliferation and establish the international controls that will inhibit the spread of nuclear weapons. Such a decision is several years away, and so the reprocessing deficit will grow. Given the experience of the industry and the lead-time required to bring additional capacity on-line, it appears unlikely that sufficient reprocessing capacity could be added to meet projected demand in this century. Hence, projections about the adequacy of uranium resources over the next 20 years will of necessity have to be based, at least in part, on a once-through LWR cycle. As indicated above, this is the most inefficient of the nuclear fuel cycles addressed in this study.

INTERIM WASTE STORAGE

A short-term problem has emerged as a result of the indecision about the back end of the fuel cycle. Because of the lack of reprocessing of long-term interim storage facilities such storage as exists at reactor cooling pools is projected to be inadequate from 1979 on. This has been of immediate concern because of the potential of reactor shut-downs caused by a backup of spent fuel. It is, however, a solvable problem in the longer view--being a result of a combination of management errors and delays in developing a reprocessing capability.

ULTIMATE WASTE DISPOSAL

The objective of waste management planning in the long term is to provide reasonable assurance that nuclear waste from both military and civilian reactors can be adequately isolated from the biosphere. High-level fission products and transuranic wastes pose the biggest problem. Isolation in underground formations of demonstrated geological stability can be expected to meet the technical requirements for long term disposal. However, the selection of a particular formation and a specific site has become a public issue and a political problem. As such, the "nuclear waste disposal" issue is less likely to be quickly resolved than it could be as a primarily technical issue, although it is solvable in the long term.

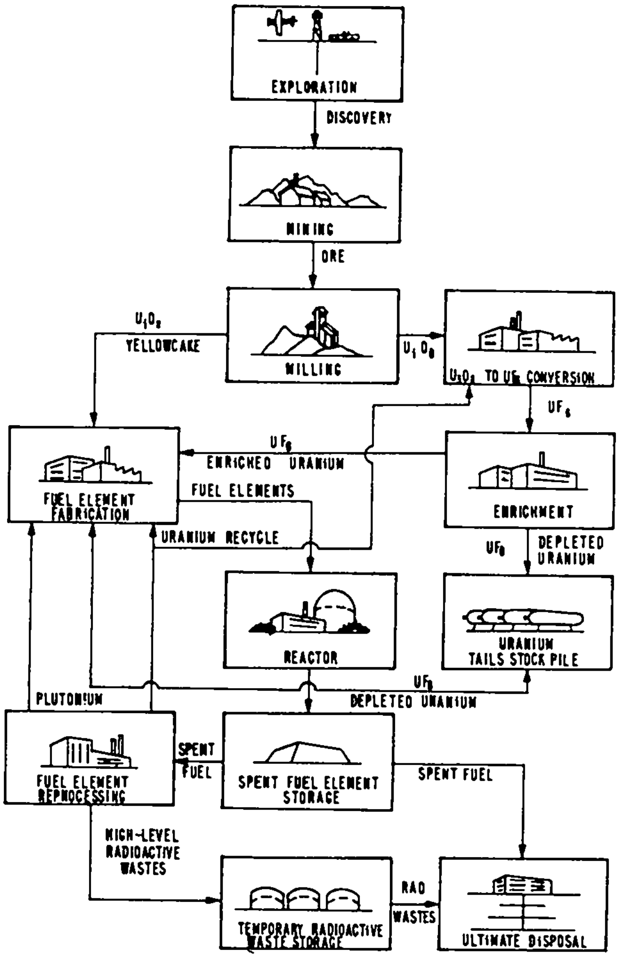

Figure 1. Schematic of generalized uranium-based nuclear fuel cycles.

TABLE I

United States Uranium Resources as of January 1, 1976

Tons U_3O_8

		Potential		
	Reserves	Probable	Possible	Speculative
\$10	270,000	440,000	420,000	145,000
10-15 increment	160,000	215,000	255,000	145,000
\$15	430,000	655,000	675,000	290,000
\$15-30 increment	210,000	405,000	595,000	300,000
\$30	640,000	1,060,000	1,270,000	590,000
By-product 1976-2000 1/	140,000	--	--	--
	780,000	1,060,000	1,270,000	590,000

1/ Estimated by-product of phosphate and copper production.

TABLE II

U.S. Estimated Distribution of $30/lb. U_3O_8 Potential Resources by Host Rock (Thousand Tons U_3O_8)

Host Rock	Probable	(%)	Possible	(%)	Speculative	(%)
Sandstone	847	(80)	820	(64)	358	(61)
Conglomerate	56	(5)	76	(6)	53	(9)
Veins	100	(9)	202	(16)	162	(27)
Limestone	16	(2)	5	(1)	13	(2)
Lignite	15	(1)	2	(1)	4	(1)
Phosphate Rocks	26	(3)	165	(13)	0	
Total	1,060	100	1,270	100	590	100

Energy Research and Development Administration, Uranium Industry Seminar. 1975 (Grand Junction, Colorado. 1975), p. 113.

TABLE III

Uranium Requirements with and without NEP
(thousands of short tons of U_3O_8), 1977 Forecast.
No allowance for existing contracts; spent fuel not reprocessed.

With NEP

	0.15% Tails		0.25% Tails	
	Annual	Cumulative	Annual	Cumulative
1977	10	12	12	12
1980	16	51	18	60
1985	27	166	32	194
1990	40	340	47	399
2000	68	902	80	1060

Without NEP

	0.15% Tails		0.25% Tails	
	Annual	Cumulative	Annual	Cumulative
1977	10	10	12	12
1980	16	51	18	60
1985	28	168	33	196
1990	45	359	52	420
2000	81	1010	95	1180

TABLE IV

Uranium Production Capacity Additions
(Does not include by-product or solution mining type facilities)

Likelihood λ Year of Operation	Owner	Capacity: Tons Ore/day	Capacity: Tons U 308/yr*
Firm 1978	Rocky Mountain Energy	800	3.200
	UNC	3,000	
	Anaconda	4,000	
		7,800	
Firm 1979	Western Nuclear	2,000	800
Probable 1979 to 1983	Kerr-McGee (Powder River)	1,500	4,500 to 4.800
	Exxon (expansion)	1,000	
	Western Nuclear (expans.)	500-1.000	
	Homestake	600	
	Cotter	1.000-1,200	
	Chevron	2.500	
	Conoco-Pioneer (expansion)	1,050	
	Union Oil (Red Desert)	3,000	
		11.150-11,850	
Possible 1980 to 1985	Getty (renovation)	1,750	5.700 to 6,700
	TVA	500	
	Gulf	3,000-4.000	
	UNC (Morton Ranch)	1.000-2.000	
	Conoco (Bernabe)	2,000	
	Kerr-McGee (Church Rock)	2.000	
	Uranerz	500	
	Pioneer (Uravan)	400-750	
	Minerals Exploration	3,000	
		14,150-16.500	
Possible 1983 to 1988	Cleveland Cliffs-Getty	1,000	4,300 to 5,000
	Conoco (Crown Point)	2.000	
	Phillips	2,000	
	Union Carbide	750	
	UNC (Dalton Pass)	1,000-2.500	
	Kerr McGee-Ariz PS	2.000	
	Mobil	2,000	
		10.750-12.250	

*Conversion from tons ore/day to tons U_3O_8/year is based on an average grade of 0.145% and a capacity factor of 76.5%. No loss factor is incorporated in the U_3O_8 production figures.

From Nuclear Fuel - May 29, 1978, p 16.

TABLE V

US URANIUM ORE MILLS OPERATING OR ON STANDBY (DECEMBER 1973)

Company	Location	Nominal Capacity (tons ore per day)
Anaconda Company	Grants, New Mexico	3.000
Atlas Corporation	Moab. Utah[a]	1,500
Conoco and Pioneer Nuclear. Inc.	Falls City. Texas	1,750
Cotter Corporation	Canon City. Colorado	450
Dawn Mining Company	Ford. Washington	400
Federal-American Partners	Gas Hills. Wyoming	950
Exxon Company	Powder River Basin. Wyoming	2.000
Kerr-McGee Nuclear Coporation	Grants. New Mexico	7.000
Petrotomics Company	Shirley Basin, Wyoming	1,500
Rio Algom Corporation	La Sal. Utah	500
Union Carbide Corporation	Uravan. Colorado	1.300
Union Carbide Corporation	Natrona County. Wyoming	1.000
United Nuclear-Homestake Partners	Grants. New Mexico	3.500
Utah International. Incorporated	Gas Hills, Wyoming	1.200
Utah International. Incorporated	Shirley Basin, Wyoming	1,200
Western Nuclear, Incorporated	Jeffrey City, Wyoming	1,200
TOTAL		28.450

Source: AEC. 1974b: 62.

[a] Uranium production facility on standby at end of 1973.

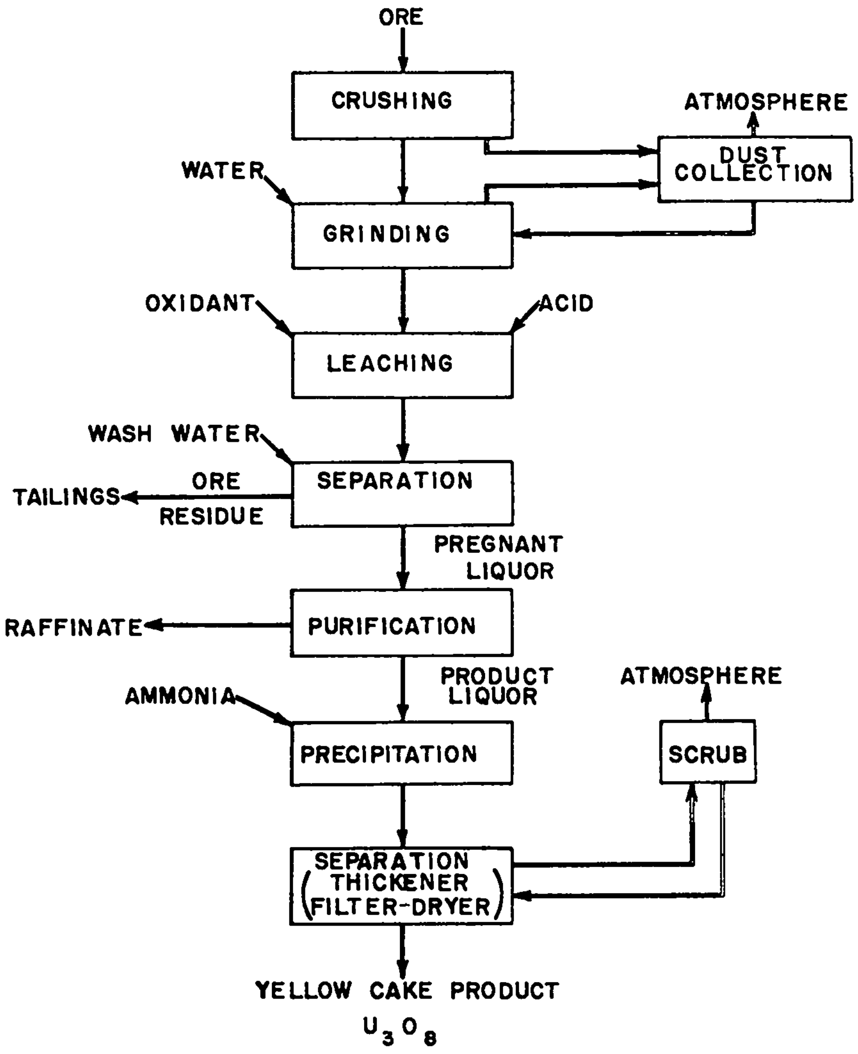

Figure 2. U_3O_8 milling process flow schematic. (Source: AEC, 1974c:B-8)

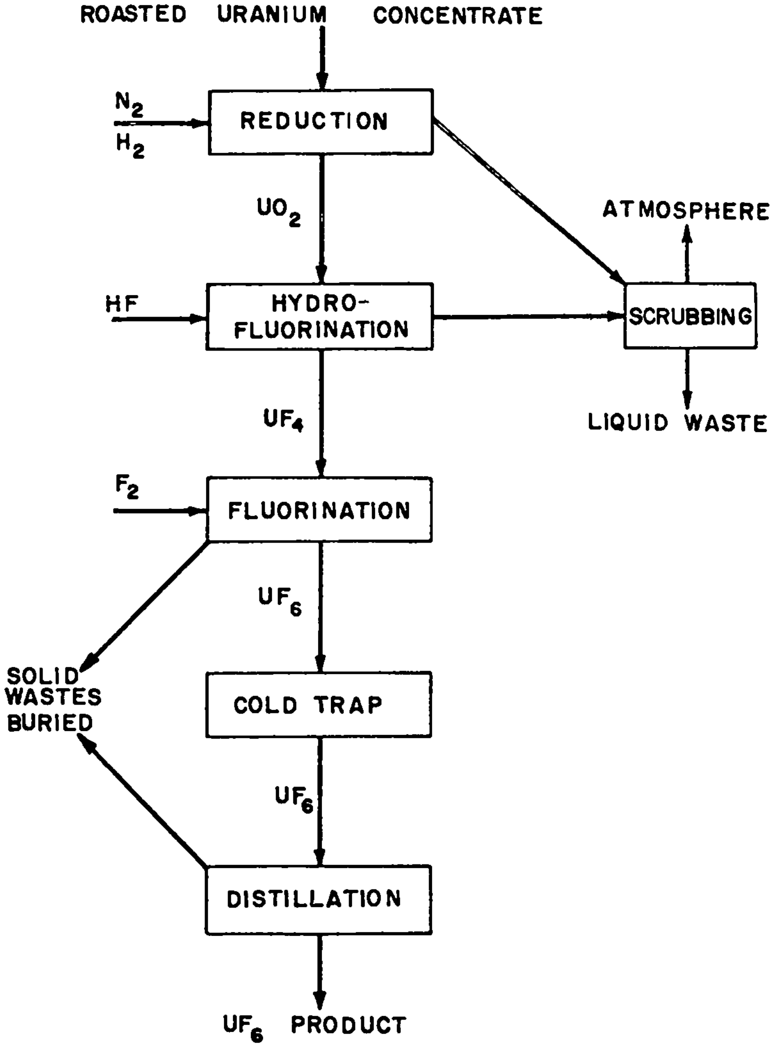

Figure 3. UF_6 production -- wet solvent extraction-fluorination. (Source: AEC, 1974c:C-9)

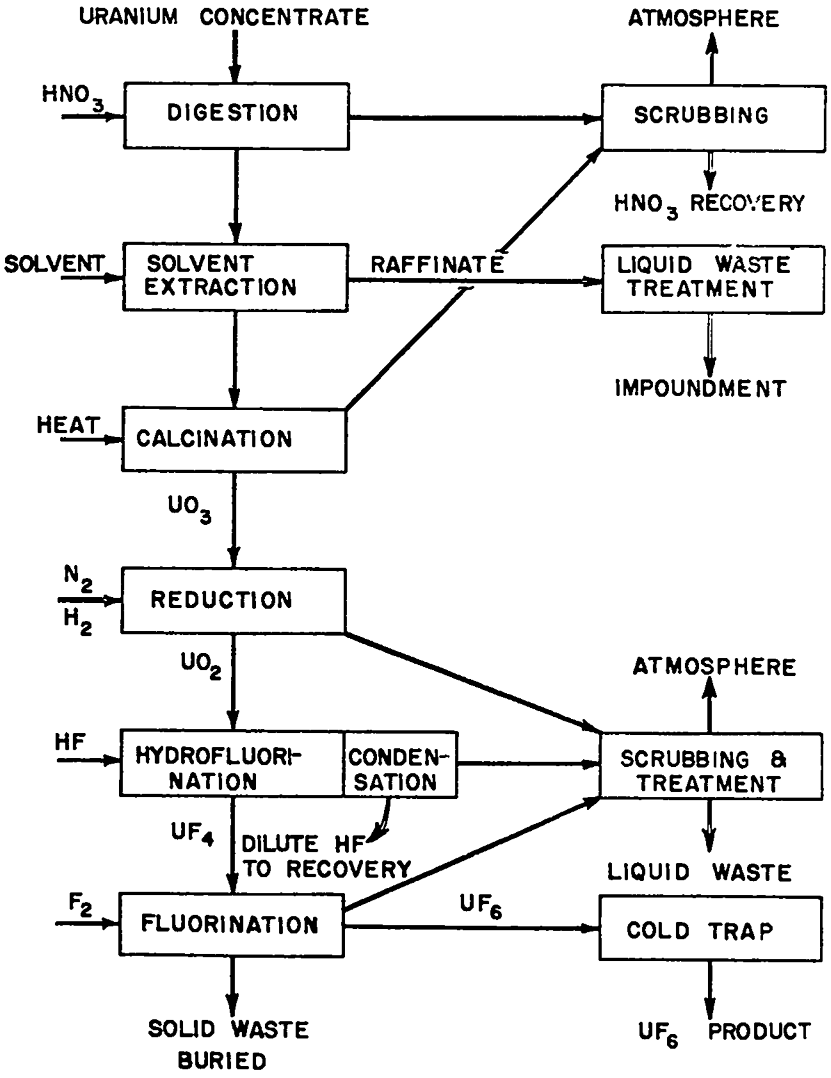

Figure 4. UF_6 production -- wet solvent extraction-fluorination. (Source: AEC, 1974c:C-9)

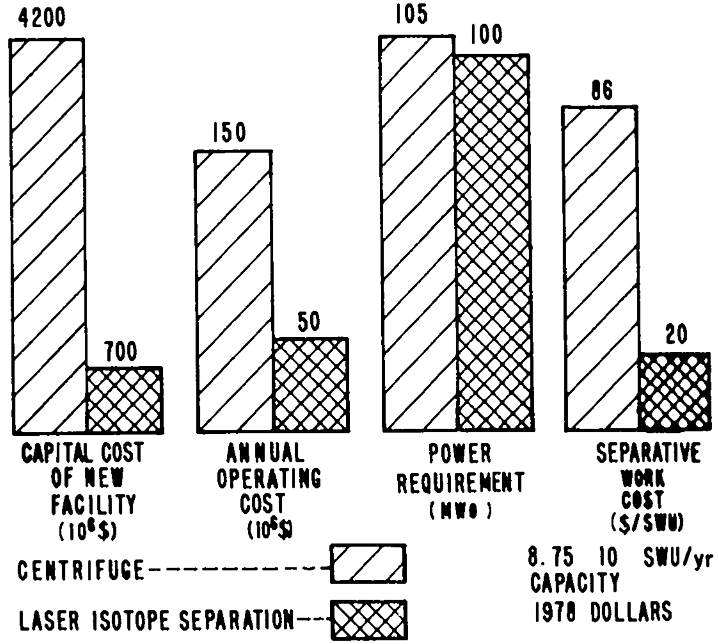

Figure 5. Comparison of performance of the molecular laser isotope separation and gaseous centrifuge processes.

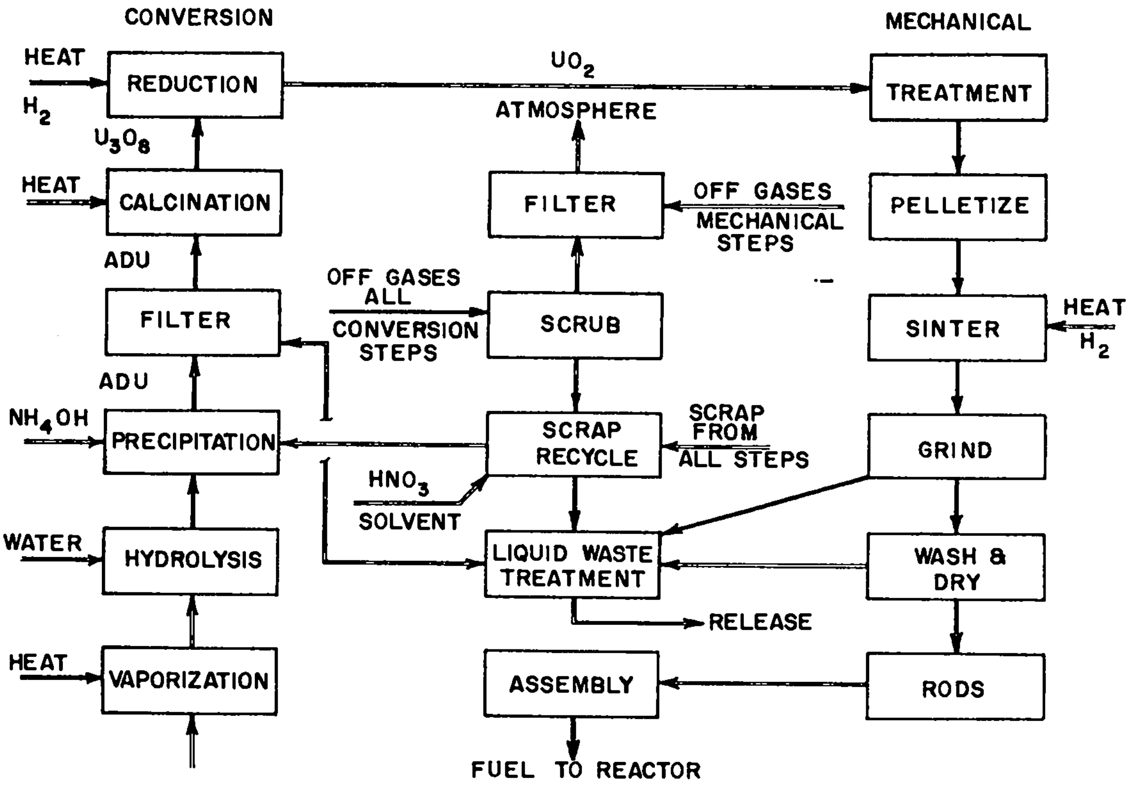

Figure 6. Fuel fabrication -- ADU process.
(Source: AEC, 1974c:E-10)

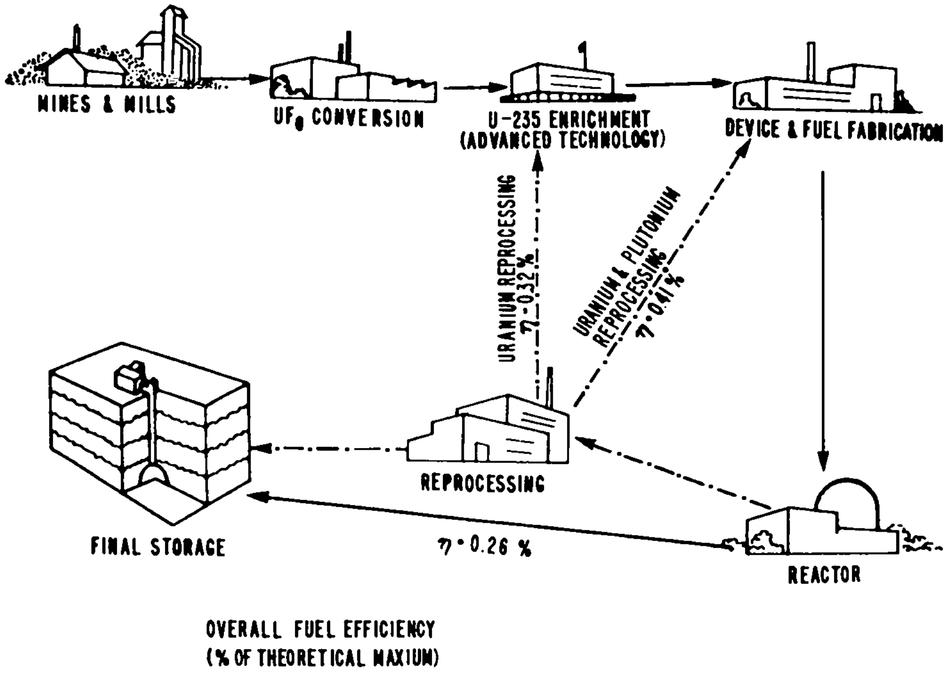

Figure 7. Light-water reactor fuel cycle (advanced technology).

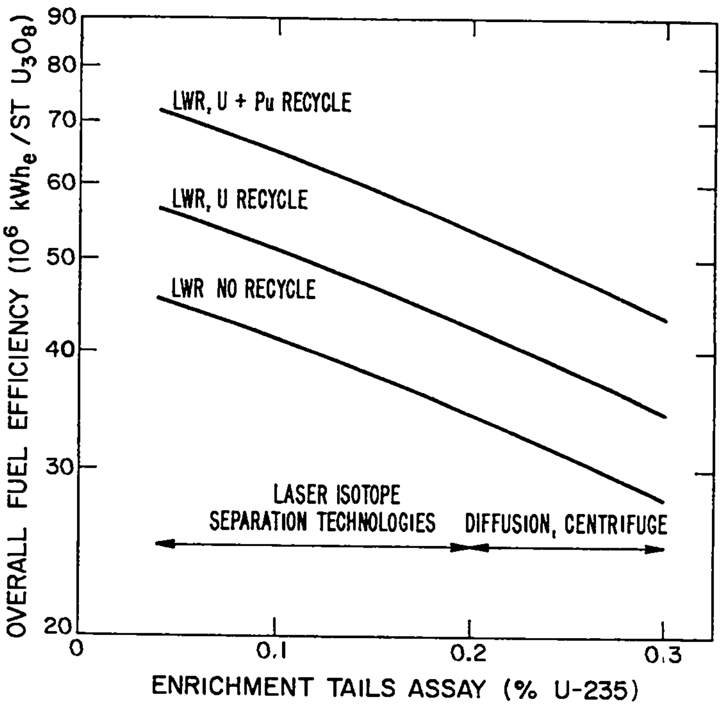

Figure 8.

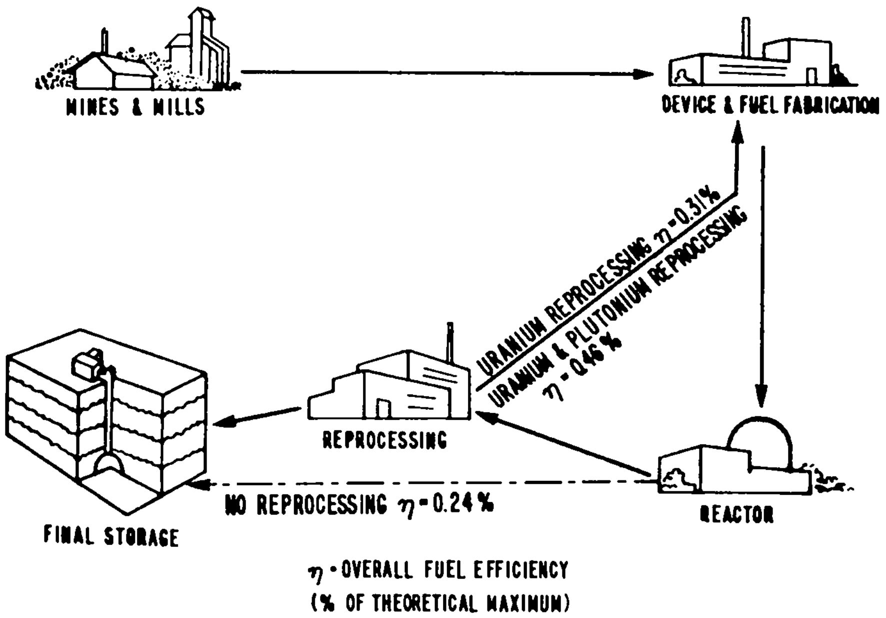

Figure 9. Heavy-water reactor fuel cycle.

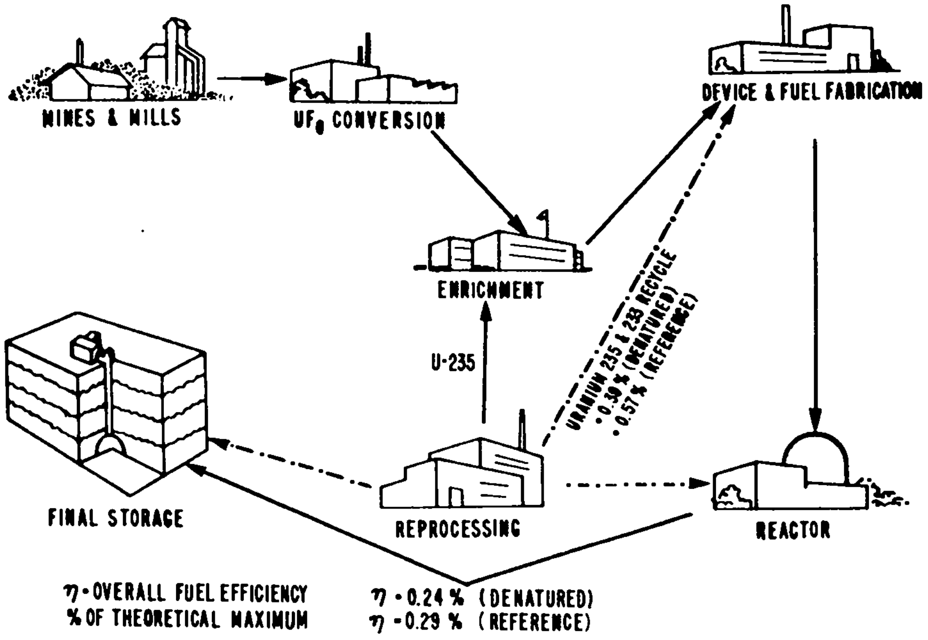

Figure 10. High-efficiency converter (HTGR) fuel cycle, conventional enrichment.

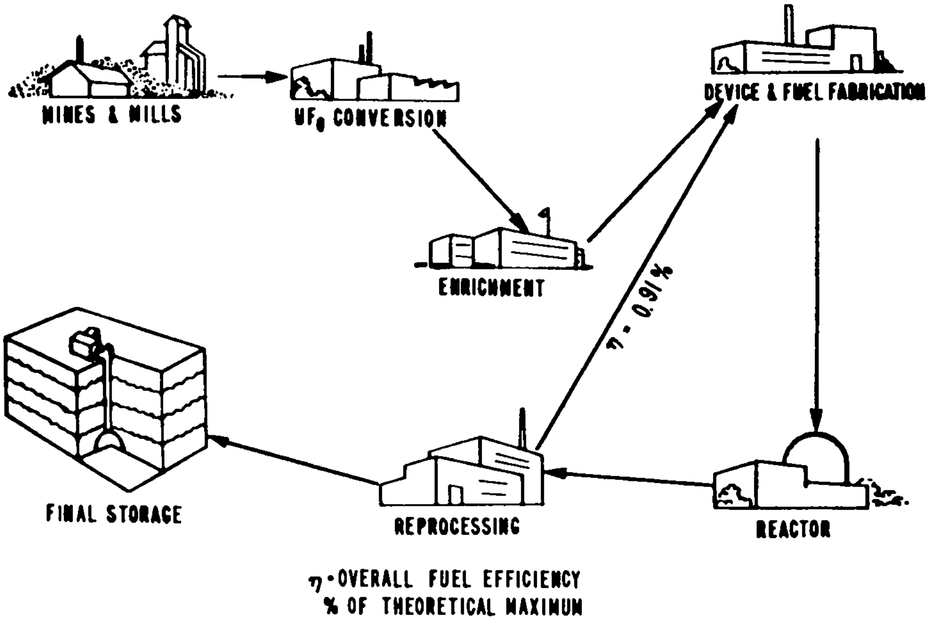

Figure 11. Low-efficiency breeder (LWBR) fuel cycle (conventional enrichment).

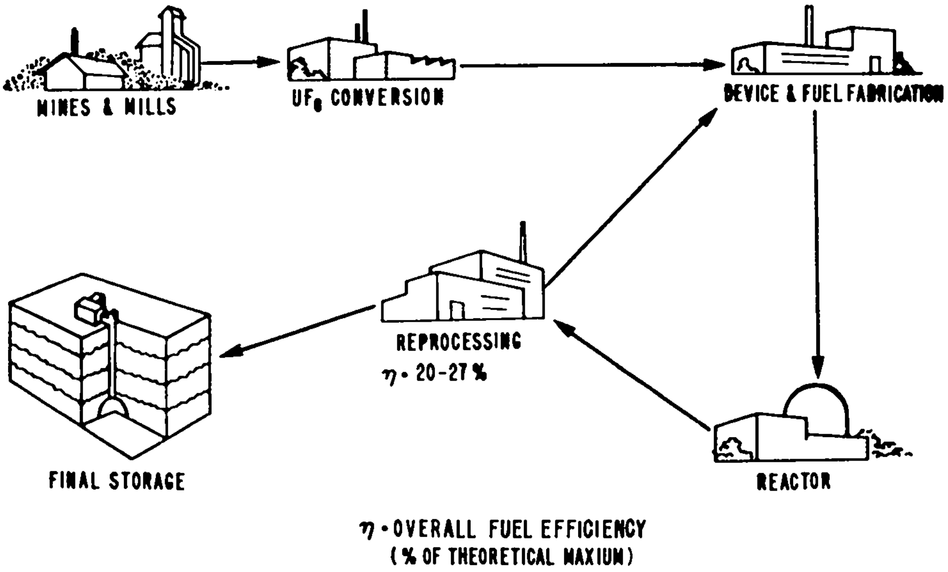

Figure 12. High-efficiency breeder (FBR) fuel cycle.

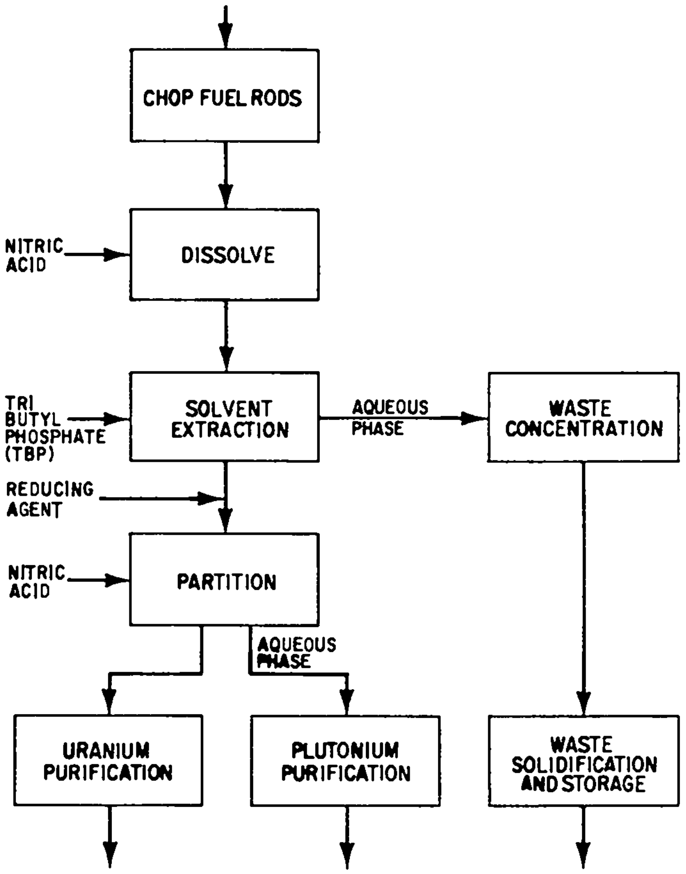

Figure 13. Basic steps in the Purex nuclear fuel reprocessing process.

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PRELIMINARY ECONOMIC ANALYSIS OF COAL GASIFICATION USING FISSION REACTORS TO PROVIDE THERMAL ENERGY

SUMMARY

This report assesses the economic viability of using thermal energy from nuclear fission for coal gasification. The report is divided into three sections: (1) a brief description of fission power plant characteristics, (2) a description of coal gasification requirements, and (3) an economic analysis of nuclear fission for coal gasification.

There are only two fission power plants that have high enough temperatures for coal gasification: High Temperature Gas Cooled Reactors (HTGR), and Pebble Bed Reactors (PBR). Because of its inherent reliability advantage the PBR was selected for this analysis. Only catalytic coal gasification appears amenable to using energy from nuclear reactors at present. Two processes were examined, one developed by Exxon, the other by J. Biery. It was originally intended that the full cost of high Btu gas be derived for all applicable processes. Thus a gas production cost model was developed (see Appendix A). However, cost data on the Biery process are incomplete while those for the Exxon process are not consistent with PBR cost estimates. Therefore an alternative approach was adopted. The costs of providing thermal energy from a mature technology PBR were estimated and compared with provision of the same energy by conventional coal systems. The Pebble Bed Reactor is not economic for pure process heat supply under these conditions. Under the most optimistic assumptions it has higher average annual costs for the Biery method than a coal system, and requires an increased investment of $80 million (in 1977 dollars). The PBR under the same optimistic assumptions is only slightly (4 to 11%) less expensive in the average year for the Exxon process and results in an increased investment in excess of $85 million (in 1977 dollars). This does not mean that the PBR is without prospects. Because PBR's have substantial economies of scale, cogeneration where the plant is owned by electric utilities, does show some promise.

NUCLEAR FISSION POWER PLANTS FOR COAL GASIFICATION: A DESCRIPTION

Only two of the over half a dozen different fission power reactor systems have outlet temperatures which are high enough for coal gasification (700-900°C). These are the high-temperature gas-cooled reactor and the pebble bed reactor. Table I lists operating characteristics of both plant types. As can be seen, both use highly enriched ^{235}U as their basic fissile material. There are also substantial amounts of ^{238}U or ^{232}Th in the fuel. These are converted to fissile ^{239}Pu or ^{233}U, though not at a sufficiently high ratio to be breeder reactors. Coolant temperatures of at least 950°C are feasible if needed. Better heat transfer materials would result in higher temperatures. There is very little experience with either system. Indeed, there are no pebble bed reactors in the US. Combined licensing requirements and construction times plus this lack of experience indicates that new HTGR or pebble bed reactors in the US cannot be expected before 1990. As it takes at least several units for a technology to mature, neither technology can be expected to mature before the year 2000 unless an "Apollo-type" crash effort were to be mounted. The HTGR must shut down to refuel, while the pebble bed can stay on line. This gives the pebble bed an important edge in reliability. Thus the best expectation for HTGR capacity factor is slightly above 80% while the pebble bed is cited at as high as 90%, although at the temperature required for gasification, pebble bed availability is estimated to drop to less than 85%. The question of reliability and its impact on costs is addressed in Appendix B.

Several estimates of capital costs for mature prismatic and pebble bed reactors are listed in Table II. There are substantial discrepancies among these estimates. This appears to result from (1) differences in the cost estimating process; (2) economies of scale; (3) the different years when the analysis was performed; and (4) the uncertainty attached to an undeveloped technology. In two cases, Sources 1 and 2, it seems that not all indirect costs were included. For the prismatic estimate, inclusion of interest expenses during construction raises the cost estimate per KWt above those of the equivalent size pebble bed reactor from Source 4. Source 2's methodology was very sketchy. It may well not include any indirect expenses. If so, then costs rise to nearly the same level as the Source 3 estimate for a 500 MWt reactor. It is widely recognized that substantial economies of scale exist for nuclear power plants. This seems to be particularly true for small, less than 1000 MWt, plants, when compared to large plants (Appendix C treats this topic in more detail). Differences in estimating these scale economies account for some of the divergencies in cost estimates. The cost estimates in Table II were generated in several different years and normalized to January, 1977, dollars using the Handy-Whitman Index. Depending on the time of year the estimates were generated and accuracy of this approach, this normalization could lead to as much as a 10% distortion. Finally, with undeveloped technologies there always exists significant uncertainty on costs. Thus, while the cost estimates in Table II cover a wide range, they are not necessarily inconsistent. Rather, the estimates reflect changes in the state of knowledge over the time these estimates were developed. This was confirmed by communication with General Electric personnel who had generated most of these estimates. Based upon current knowledge, their feeling is that for privately owned PBRs the inflation-adjusted Source 3 estimates are the best available.

COAL GASIFICATION TECHNOLOGY

Although there are a number of high-Btu coal gasification technologies under development, only the catalytic processes appear suitable for combining with nuclear power. There are two such processes considered: the Exxon process, which is now at the pilot plant stage, and the Biery process, which was developed for this project to take advantage of the high-temperature heat available from a nuclear reactor. Other promising coal gasification processes with currently foreseen technology create waste fuels which are used to satisfy most if not all thermal requirements.

The Exxon process uses potassium carbonate as the catalyst. Gasification takes place in one gasifier which operates at $700^{\circ}C$, 35 atm. A 250×10^{6} cubic foot per day gas plant requires four such gasifiers.* The methane is

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*See L. C. Furlong and N. C. Nahas, Exxon Research and Engineering Co., "Catalytic Coal Gasification Process Research and Development," presented at the 10th Synthetic Pipeline Gas Symp., Chicago, ILL (October 1978), and J. E. Gallagher, Jr., and H. A. Marshall, Exxon Research and Engineering Co., "Production of SNG from Illinois Coal via Catalytic Gasification," presented at AIChE Symp. on Reaction Engineering in Processing Solid Fossil Fuels, Miami Beach, FL (November 1978).

then separated from the carbon monoxide and hydrogen by cryogenic cooling. There are three principle energy requirements to the system: gasifier operation and preheat, heat for recycled gas streams, and energy for the cryogenic distillation. Overall converson efficiency is 63% including ammonia and other salable products. This is for a coal burning plant which imports electricity.

The capital cost for a 257 x 10^9 Btu per day pioneer (i.e., first of a kind) plant has been estimated at $1,640 million (in 1978 dollars). This is quite high relative to estimates for other types of gasification plants. Exxon feels there are three reasons for this:

1. Their approach aims at most likely final cost for a pioneer plant. Thus, investment is based on a process basis supported by the current data base -- potential future improvements not being considered. The design philosophy incorporates features to achieve a high service factor. The utility capacities include contingency allowances based upon past experience. Final equipment specifications were developed in detail to avoid the omissions common to overly simplified approaches.
2. The inclusion of substantial investment contingencies totaling to $470 of $1640 M.
3. The inclusion of added costs to cover the effect of "diseconomies of scale" on field labor construction costs for large projects.

Based upon Exxon's cost estimates, amended where possible to Texas Gas's circumstances, the cost of methane would be $5.80 per 10^6 Btu in the first year while the average cost is $5.35 per 10^6 Btu. These costs are broken down in Table III. Exxon estimates that these costs will drop $0.75 to $1.00 as the technology matures.

The Biery process also uses potassium carbonate as the catalyst. However, it aims at maximizing thermal efficiency by taking full advantage of the exothermic reactions in the gasifiers. The high-temperature bed requires an input temperature of about 900°C. The principle energy inputs are the same as in the Exxon process: the gasifier, waste stream reheat, and cryogenic cooling. However, the energy balancing in the gasifier string substantially increases conversion efficiency. Biery estimates the overall plant efficiency at 83.3%. (If thermal energy is provided by efficient combustion and electricity is purchased, this drops to about 80%). About half the energy input reflects the thermal requirement for the electric (or shaft) power used in cooling. Unfortunately capital cost estimates for this process are not available.

ECONOMIC ANALYSIS

There is only limited data available on the catalytic processes. Therefore a null hypothesis approach was adopted. This hypothesis is, that using the most favorable reasonable assumptions for pebble bed reactors, they would still be more expensive than equivalent fossil fueled plants. Thus, instead of determining the cost of gas produced and the total investment, this analysis only concerns itself with the economic aspects of energy provision. For the Biery process this approach is necessitated by the lack of data. For the Exxon process this approach was adopted because of inability to normalize the Exxon process cost estimates and those of the pebble bed reactor.

In performing the economic analysis the following assumptions were made:

1. The nuclear reactor and gasifiers (as well as alternative fuel source) have 90% capacity factors, and there is a one-to-one correspondence among outages. This is quite optimistic; Appendix B notes that lower reliability favors fossil fired versus nuclear plants.
2. Capital costs for the nuclear reactor are the lowest estimate for the mature technology in the reasonable cost range. See Appendix C for these estimates.
3. Costs for all power plants are based upon the following formula, direct and indirect costs multiplied by 1.15 for A and E work, that multiplied by 1.2 for contingencies and the total by 1.185 for interest expenses during construction. (This normalizes power plant cost estimates to Source 3, Table II).

4. The cost of nuclear fuel, in 1977 dollars, is $.54 per million Btus.* Coal cost is $1.00 per million Btus in 1977 dollars (cost provided by Texas Gas).
5. The capital charge rate in the first year is 22.11%. The average capital charge is 18.48% for a twenty-year project lifetime.
6. The fossil-fired alternative is a conventional boiler with flue gas desulfurization. See Appendix D for costs.
7. The construction time is equivalent for either a nuclear or fossil fuel-fired plant and there would be little discernable difference in the permit process time requirement.
8. The PBR delivers 90% of the heat generated in the core and the coal-fired units are 87% efficient.**
9. Operating and maintenance costs are equal. See Appendix E.
10. Because of differing scale economies the nuclear plant will have one unit, the coal-fired, two.
11. Methane production is 257 billion Btu per day.

The Exxon process requires thermal input of about 1500 MW. As can be seen in Table IV, the PBR results in slightly lower costs, less than 1% the first year and only about 4% in the average year. However, there is a significant increase in the initial cost of plant of about $100 million. The situation is somewhat more favorable if the size is increased and about 160 MWe of electric generating capacity is added to allow the provision of all energy requirements. As can be seen from Table V, the PBR now has almost a 7% cost advantage in Year 1 and about 11% in the average year. Moreover, the increased investment appears to drop slightly, to about $90 million. This scheme is likely, however, to reduce PBR reliability slightly as the entire facility

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*Based on SRI International "Fuel and Energy Price Forecasts: Quantities and Long Term Marginal Prices," EPRI EA-433 (September 1977). Estimate escalated from mid-1975 dollars. If their coal estimate, $.80 per 10^6 Btu (1977 dollars) for high sulfur interim coal, is indicative, this estimate is on the low side.

**See General Electric, "The VHTR for Process Heat," op. cit. p. 36; and Dale Brown, cit. Appendix D, p. 9.

could be forced out due to turbine related problems. This annual cost saving must be balanced against the increased investment. Thus even under the most favorable circumstances, the nuclear unit is at best marginally attractive. Indeed, if the alternative is the atmospheric fluidized bed, then even on an annual cost basis coal is the preferred fuel.

For the biery process, nuclear power fails the null hypothesis. It is not economic. Because of its greater efficiency this process only requires about 800 MWt of input for all purposes.* As can be seen in Table VI, the PBR is both more expensive in the average year and requires a significantly higher investment, $80 million.

These results indicate that even when the technology matures, fission reactors are not economically viable for coal gasification at today's relative prices of input factors. There is, however, a means to possibly overcome this situation: cogeneration. By building a 3000 MWt nuclear plant and generating electricity for export with the thermal energy not used for gasification, capital costs per KWt are considerably reduced. There are three institutional mechanisms to achieve this: gas utility ownership, electric utility ownership, and joint ownership.

Gas utility ownership is not promising. This route considerably increases the initial investment. On top of that, the price that TVA is willing to pay for electricity is insufficient to cover the increased reactor costs, let alone the turbine generator. It is possible a private utility might be willing to pay more, but not likely.

Electric utility ownership is promising, at least from the gas utility's standpoint. There are four important economic/financial advantages to this proposal: (1) reduced investment by Texas Gas, who would not have to build any power plant facilities; (2) assuming TVA is the utility, a reduced cost of gas due to TVA's lower cost of capital, (3) reduced energy costs for gasification from a sharing of the savings that accrue from cogeneration, and (4) likely elimination of the reliability problem as the utility would probably build a

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*This assumes 40% electric conversion efficiency once the steam is generated. This is readily attainable with state-of-the-art technology.

multi-unit plant.* (See Appendix F for economic analysis of cogeneration.) There are, of course both considerable contractual and scheduling problems.** The benefits to the gas utility, however, appear attractive enough to surmount these if the electric utility is similarly inclined.

Joint ownership would involve the gas utility owning some part of the thermal plant. It is somewhat less attractive than full electric utility ownership.

This analysis indicates that gas utility ownership of fission reactor(s) for providing energy to a coal gasification plant is not economically viable. It may, however, be viable if the reactor(s) were owned by an electric utility with operated it (them) as a cogenerator(s) to take full advantage of economies of scale.

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*With the possible exception of (4), electric utility ownership of coal-fired units offers the same advantages, though they are of comparatively less value.

**That these problems are by no means insignificant is apparent from even a cursory glance at the Dow-Consumers Power, Midland Plant fiasco. Originally plans called for Dow to receive process steam from a Cosumers Power nuclear plant in 1976. Our latest information is that this will not occur until 1983. At the same time, projected costs have skyrocketed.

APPENDIX 8-A

GAS PRODUCTION COST MODEL

In this appendix we describe the model developed to compute the cost of gas production from coal. Certain variables were included with an eye to further development, and so are defined here but were not used in our analysis. The variable names, definitions and initial values are given in Table A-I. In Table A-II we list the equations used in our analysis. A flow chart for this computation is included as Figure A-1. Our actual computations were performed using a simple FORTRAN program, COAL GAS, based upon the analysis described in Table A-II. A source listing of this program is given in Table A-III. Finally, in Table A-IV, we present sample input data and output for COAL GAS.

TABLE A-I

VARIABLES USED IN GAS PRODUCTION COST MODEL

Variable:	Definition:	Initial Value:
CC	Capital Construction Cost	$\$1 \times 10^{10}$
PE	Percent Equity	25%
EC	Equity Cost	15%
IR	Interest Rate	10%
YRS	Number of Years in Model Run	30
T	Project Year	1 to 30
MTB	Marginal Tax Rate	48%
FCT	Factor for Conversion of Taxes (= State Income Tax/Federal Income Tax)	.02
PTI	Property Tax and Insurance	2%
OE	Operating Expense (as a percent of CC)	2%
MFC	Hourly Fuel Consumption (10^6 Btu per MWt)	4
NCM	Nuclear Fuel Cost (per 10^6 Btu)	$.75
AMT	Annual MWH Thermal	7×10^7
TCG	Tons of Coal per 10^9 Btu Gas Output	50
GAS	Gas Output (10^9 Btu per Year)	6.5×10^5
CCT	Cost of Coal (per Ton)	$20.00
DCM	Nuclear Disposal Cost (per 10^6 Btu Fuel)	$.15
DCT	Coal Disposal Cost (per Ton)	$1.00
MEG	Electricity Generated via Turbines (MWH)	0
OMM	Nuclear Operating and Maintenance Cost (per MWH thermal)	$.01
OMT	Gasifier Operating and Maintenance Cost (per 10^6 Btu Gas)	$.04

TABLE A-I (Continued)

OME	Turbine Operating and Maintenance Cost (per MWH)	.001
MP	MWH of Electricity Purchased	0
PM	Price per MWH	0
PD	Peak Demand for Purchased Electricity (MW)	0
PMD	Demand Charge (per MW)	0
PER	Interval to Print our Costs (not used)	10
YRST	Year Analysis Starts (not used)	1990

TABLE A-II

DEVELOPMENT OF GAS PRODUCTION COST MODEL

In our computation we have divided production costs into costs of service (capital and associated costs) and operating costs. The latter are further subdivided into fuel cycles, operating and maintenance costs, and costs of purchased electricity. After computing each cost term, we develop average and yearly costs per 10^6 Btu of gas produced.

1. Cost of Service:

Return on Equity	$RE = PE \times EC$
Depreciation	$DR = 1/yrs$
Federal Income Tax	$FIT = RE \times MTB/(1 - MTB)$
State Income Tax	$SIT = FIT \times FCT$
Interest Expense	$IB(T) = (1 - PE) \times IR \times (YRS - T + 1)/YRS$
Capital Recovery Factor	$CRF(T)F = RE + DR + FIT + SIT + PTI + IE(T) + OE$
Cost of Service	$CS(T) = CRF(T) \times CC$

2. Fuel Cycle Costs:

Nuclear Fuel Cost	$NFC = HFC \times NCM \times AMT$
Coal Cost	$CFC = TCG \times GAS \times CCT$
Total Fuel Cost	$FC = NFC + CFC$
Nuclear Disposal Cost	$NDC = HFC \times AMT \times DCM$
Coal Disposal Cost	$CDC = TCG \times GAS \times DCT$
Waste Disposal Cost	$WDC = NDC + CDC$

3. Operations and Maintenance Costs:

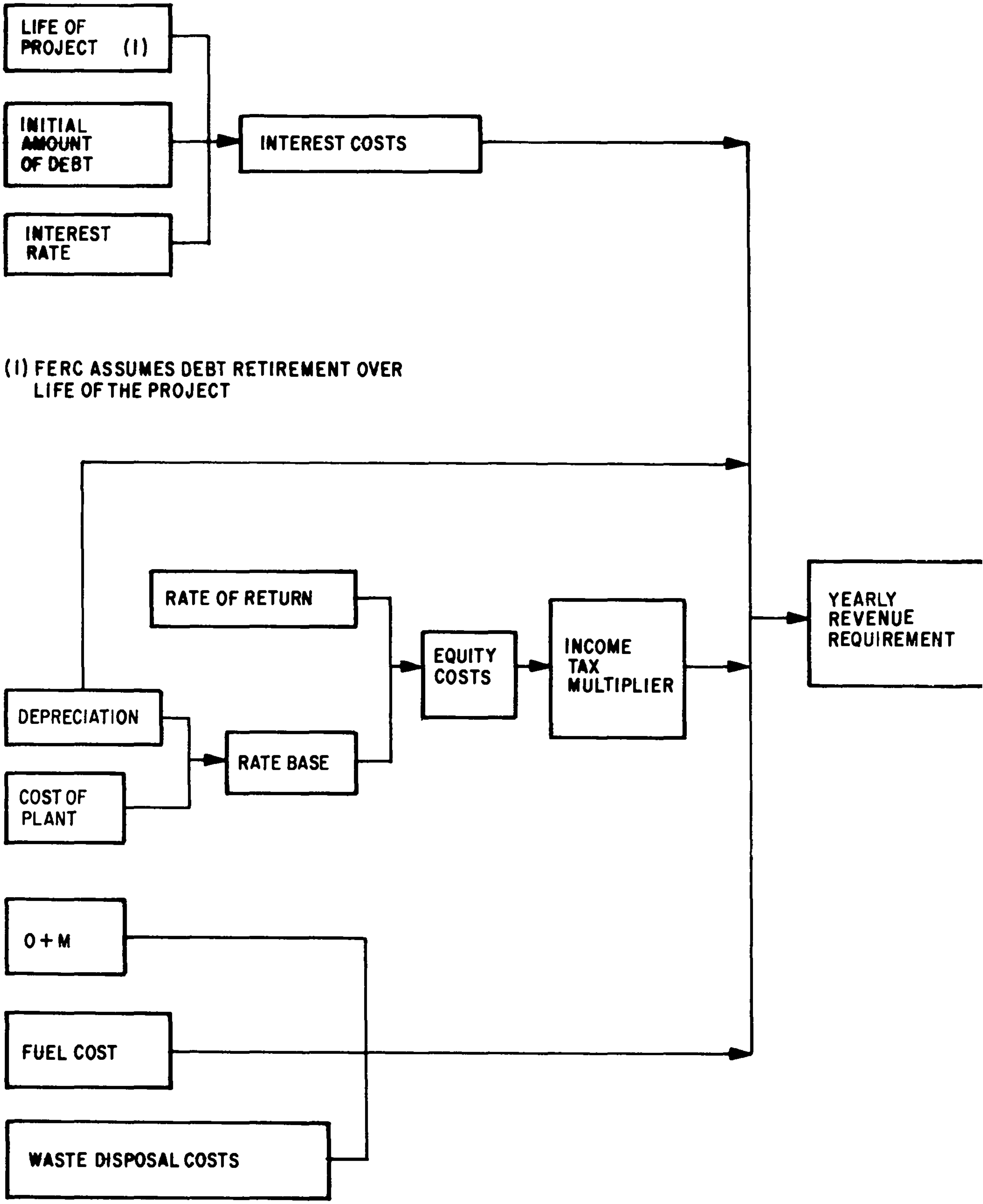
Nuclear O and M Costs	$NO = AMT \times OMM$
Gasifier O and M Costs	$GO = TCG \times GAS \times OMT$
Turbine O and M Costs	$TOE = MEG \times OME$
Total O and M Costs	$OM = NO + GO + TOE$

4. Purchased Electricity Costs:

Energy Cost plus Demand Charges	$COP = MP \times PM + PD \times PMP$

5. Total and Average Costs:

Total Operating Costs	$OC = OM + FC + WDC$
Yearly Cost per 10^9 Btu Gas	$CMG(T) = (CS(T) + COP + OC) \times 1000/GAS$
Average Cost per 10^9 Btu Gas	$AVRGE = \underset{T=1}{\overset{30}{}} CMG(T)/30$



(1) FERC ASSUMES DEBT RETIREMENT OVER LIFE OF THE PROJECT

ANNUAL COST MODEL

TABLE A-IV

SOURCE LISTING FOR COAL GAS

```
      IMPLICIT REAL (I-N)                                               COA00010
      INTEGER PER,YRST,YRS,T,TYR                                        COA00020
      DIMENSION CS(50),CMG(50)                                          COA00030
      NAMELIST /OUTPUT/ CS1,CRF,FC,WDC,OM,COP,OC,CMG1,AVRGE             COA00040
      EQUIVALENCE (CS(1),CS1),(CMG(1),CMG1)                             COA00050
      READ (11,*) CC,PE,EC,YRS,MTB,FCT                                  COA00060
      READ (11,*) PTI,IR,OE                                             COA00070
      READ (11,*) HFC,NCM,AMT,TCG,GAS,CCT,DCM,DCT                       COA00080
      READ (11,*) OMM,OMT,OME,MEG,MP,PM,PD,PMP                          CA000090
      READ (11,*) PER,YRST                                              COA00100
C COMPUTE COST OF SERVICE                                               COA00110
      RE=PE*EC                                                          COA00120
      DR=1./YRS                                                         COA00130
      FIT=RE*MTB/(1.-MTB)                                               COA00140
      SIT=FIT*FCT                                                       COA00150
      ID=(1.-PE)*IR                                                     COA00160
      DO 1 T=1,YRS                                                      COA00170
      IE=ID*(YRS-T+1)/YRS                                               COA00180
      CRF=RE+DR+FIT+SIT+PTI+IE+OE                                       COA00190
1     CS(T)=CC*CRF                                                      COA00200
C     COMPUTE OPERATING COSTS                                           COA00210
C     COMPUTE FUEL CYCLE COSTS                                          COA00220
      NFC=HFC*NCM*AMT                                                   COA00230
      CFC+TCG*GAS*CCT                                                   COA00240
      FC+NFC+CFC                                                        COA00250
      NDC=HFC*AMT*DCM                                                   COA00260
      CDC=TCG*GAS*DCT                                                   COA00270
      WDC=NDC+CDC                                                       COA00280
C COMPUTE O AND M COSTS                                                 COA00290
      NO=AMT*OMM                                                        COA00300
      GO=TCG*GAS*OMT                                                    COA00310
      TOE=MEG*OME                                                       COA00320
      OM=NO+GO+TOE                                                      COA00330
      OC=OM+FC+WDC                                                      COA00340
C COMPUTE PURCHASED ELECTRICITY COST                                    COA00350
      COP=MP*PM+PD*PMP                                                  COA00360
      DATA ACMG/0./                                                     COA00370
      DO 2 T=1,YRS                                                      COA00380
      CMG(T)=(CS(T)+COP+OC)/GAS/1000.                                   COA00390
2     ACMG=ACMG+CMG(T)                                                  COA00400
      AVRGE=ACMG/YRS                                                    COA00410
      CRF=CS1/CC                                                        COA00420
      WRITE (6,OUTPUT)                                                  CA000430
      STOP                                                              CA000440
      END                                                               COA00450
```

TABLE A-IV

DATA AND OUTPUT FOR COAL GAS

COAL GAS DATA FILE

```
R; T=0.03/0.36 14:34:09

.TYPE FILE FT11F001

1E10 .25 .15 30 .48 .02
.02 .1 .02
4 .75 7E7 50 6.5E5 20 .15 1
.01 .04 .001 5*0
10 1990

R; T=0.01/0.05 14:34:25
```

COAL GAS OUTPUT

```
.LOAD COALGAS
R; T=0.09/0.23 10:45:19

.START
EXECUTION BEGINS...
 OUTPUT
CS1=  .221140762E+10,CRF=  .221140742    ,FC=  859999744.     ,WDC=  74499984.0
  ,OM=  1999999.00    ,COP=  .0           ,OC=
  936499456.    ,CMG1=  4.84293270    AVRGE=  4.28523350
 END
R; T=0.03/0.08 10:45:35
```

APPENDIX 8-B

IMPACT OF POWER PLANT RELIABILITY ON COST OF GAS PRODUCTION

Coal gasification plants are expected to produce gas at 90% of their rated capacity to minimize the capital component of gas costs. Therefore, thermal energy must be available both 90% of the time, and concurrent with the gasifiers to minimize cost. Otherwise, one of the following solutions must be adopted:

1. Addition of thermal energy back-up,
2. Acceptance of less than 90% capacity factors and increased sizing of all systems to obtain the same net gas output, or
3. Thermal storage.

The last alternative is feasible only if the thermal source has about a 90% capacity factor and the durations of thermal requirement to availability mismatch are short. It is therefore unlikely. The second alternative is also not likely. The cost of other components to the gasification plant are high. Building additional capacity beyond that needed, therefore, is to be avoided. This option would be adopted only if it cost less than back-up power. Therefore, we shall endeavor to obtain some insight into the cost of backup.

An important question is how reliable is the thermal supply. This will set the back-up requirements. Coal burners tend to be quite reliable. A 90% capacity factor for a two unit plant does not seem unrealistic, though sulfur removal could reduce this by several percent. However, to demonstrate the impact of reliability, we shall assume coal plants have the same capacity factors as nuclear plants.

The experience of nuclear power reactors in the US to date has been disappointing. For the period May 1974 to June 1977 they were unavailable 27% of the time.* Of this, 40% of the unavailable time, or nearly 11% of the total period, was due to refueling. The Canadian Candu reactors have done much better. They hve been unavailable slightly more than 20% of the time.* This

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*Science Applications, Inc., "Refueling Outage Trends in Light Water Reactors," Electric Power Research Institute, EPRI NP-842 Project 705-1 Interim Report (August 1978).

improved availability results from their on-line refueling capability. The pebble bed reactor also has on-line refueling, while the HTGR does not. Therefore, it should have a greater availability. Ten years' experience with the only pebble bed reactor operating, the AVR in Julich, Federal Republic of Germany, has resulted in 78% availability, although there recently was a year of slightly over 90% availability.** The target for process heat pebble bed reactors has been set at 90% availability.*** Should this be attained, then reliability is not a problem. However, for the relatively high temperature, about 950°C, required, expected availability falls to about 80%.[+] This corresponds to German experience. At 80% availability, the need for backup is obvious. For a coal thermal plant with two units this requires a third unit, which would operate 20% of the time (needed plant availability (.9) - actual (.8) plant percent required (.5)). At this low load factor, an oil-fired backup may be economical. The combined thermal plantsd now have a 100% of requirements availability of 89.6% and a 50% of requirement availability of 9.6%. There are two options for the nuclear plant: (1), full backup with a single plant which would have a 10-16% load factor, or (2) build two reactors and backup of the same size as that in the coal case. The least expensive option must be determined. However, compared to coal, either result has higher relative costs than when availability is 90%. For example, going from a single 800-MWt plant to two 400-MWt plants is estimated, in Table C-II to cost at least $85 per kWt. Therefore the cost of the backup to the single reactor system must be less than $170 per kWt or the two reactor system will be chosen.

*Personal correspondence, D. G. Martin, Reliability Evaluation Section, Ontario Hydro (September 1978).

**G. C. Leath and C. C. Bussey, "The Pebble Bed Reactor with Chemical Heat Pipe and Steam Turbine Electric Output--A Multiplex Energy System," General Electric (March 1978).

***General Electric, "The VHTR for Process Heat," GEAP 14018, UC 77 (September 1974) and General Electric, "Small Nuclear Process Heat Plants Using Pebble Bed Reactor," GEEST 75-001, ORNL-Sub-4352 (September 1975).

[+] General Electric, "The VHTR for Process Heat," GE AP 14018, UC77 (September 1974).

What this simplistic approach demonstrates is that much less than 90% nuclear availability is likely to result in a significant cost penalty (quite possibly over $100 per kWt) for the nuclear as compared to the coal thermal energy supply option.

APPENDIX 8-C

ESTIMATES OF CAPITAL COSTS OF PEBBLE BED REACTORS

Two reports estimate the cost of pebble bed reactors over a range of sizes. These estimates are presented in Tables C-I and -II. There is a wide variation in the two cost estimates. Communication with General Electric personnel involved in both studies indicates that the discrepancy is basically due to improvements in the state of pebble bed reactor knowledge. They felt that the cost range presented in Table C-II is realistic based upon present (November 1978) knowledge, though they feel the low-side estimates are somewhat more probable. These cost estimates were extended from 1000 MWt to 3000 MWt and are presented in Table C-III. This cost extension is based upon the relationship between cost estimates in Tables C-I and -II. These estimates are also in accordance with the sizing exponential factor, .7, commonly used in estimating costs for different size nuclear plants.

TABLE C-I

ESTIMATED EFFECT OF PLANT SIZE ON PEBBLE BED REACTORS (IN 1977 DOLLARS)

	Cost per kWt			
Plant Size, MWt	Reactor Plant	Undistributed Costs	Other	Total
500	260	245	295	800
1000	185	160	150	495
2000	145	80	115	340
3000	135	55	95	285

Source: General Electric, "The VHTR for Process Heat, Vol. II," GEAP-14018 UC-77 (September 1974), p. 38.

TABLE C-II

ESTIMATED RANGE OF CAPITAL COSTS FOR SMALL PEBBLE BED REACTORS (IN 1977 DOLLARS)

Plant Size, MWt	High Cost Estimate per kWt	Low Cost Estimate per kWt
200	635	505
400	530	405
600	460	360
800	390	320
1000	360	285

Source: General Electric, Energy Systems and Technology Division, "Small Nuclear Process Heat Plants Using Pebble Bed Reactor," GEEST 75-001, ORNL-Sub-4352-1 (September 1975), pp. 10-19.

TABLE C-III

EXTENSION OF CAPITAL COSTS FOR SMALL PEBBLE BED REACTORS (TABLE C-II) TO LARGER REACTORS (IN 1977 DOLLARS)

Plant Size in MWt	High Cost Estimate per kWt	Low Cost Estimate per kWt
1000	360	285
1500	330	260
2000	290	235
3000	265	215

APPENDIX 8-D

COST OF COAL-FIRED THERMAL POWER PLANTS

These cost estimates are based upon coal-fired boilers with flue gas desulfurization. As some of the thermal demands will be met by burners this approach slightly overestimates costs. Moreover, boiler costs may vary by as much as 40% depending upon location, efficiency, and reliability. The estimates in Table D-I are based upon efficient, reliable boilers and represent the middle to high part of the cost range. The atmospheric fluidized bed technology is included because it is expected to be commercially available about 1990, considerably sooner than the pebble bed reactors (at 2000 or later).

TABLE D-I

COST OF COAL-FIRED BOILERS BY SIZE (IN 1977 DOLLARS)*

Size, MWt	Conventional (Flue Gas Desulfurization) Estimate per kWt	Atmospheric (Fluidized Bed) Estimate per kWt
400	220	175
750	195	150
1500	190	150

*Based upon the following:

O. H. Klepper and W. R. Smith, "Studies of a Small PWR for On-Site Industrial Power," Proc. American Power Conf., V. 39, Illinois Institute of Technology (1977), pp. 744-754.

Dale H. Brown, "Conceptual Design and Implementation Assessment of a Utility Steam Plant with Conventional Furnace and Wet Lime Stack Gas Scrubbers," Energy Conversion Alternatives Study, National Aeronautics and Space Administration NASA CR-134950, 5RD-76-064-4 (December 1976).

National Aeronautics and Space Administration, Lewis Research Center, "Evaluation of Phase 2 Conceptual Designs and Implementation Assessment," Resulting from the Energy Conversion Alternatives Study, NASA TM Y-73515 (April 1977).

and personal communications with A. Jonke, Argonne National Laboratory; H. Marshall, Exxon; and D. Klepper, Oak Ridge National Laboratory.

APPENDIX 8-E

ESTIMATED OPERATING AND MAINTENANCE COSTS

The maintenance costs for pebble bed reactors are shown in Table E-I. The estimates for 500- and 1000-MWt plants were from the 1975 GE study cited earlier. These cost estimates were inflated to 1977 dollars and then extended to larger size plants.

There is some divergence in the operating and maintenance cost estimates for coal-fired plants as can be seen in Table E-II. Note how these estimates bracket the pebble bed estimates.

TABLE E-I

OPERATING AND MAINTENANCE COSTS FOR PEBBLE BED REACTORS BY SIZE
(IN 1977 DOLLARS)

Size in MWt	Operating and Maintenance Cost, 10^6 1977 Dollars
500	4.25
1000	7.8
2000	14.2
3000	21.3

TABLE E-II

OPERATING AND MAINTENANCE COSTS FOR COAL-FIRED THERMAL POWER PLANTS*

Size in MWt	Operating and Maintenance Cost Range, 10^6 1977 Dollars
500	2.8 - 4.4
2000	11.0 - 15.5
3000	16.6 - 22.9

*Based upon Dale Brown, op. cit., and Klepper and Smith, op. cit.

APPENDIX 8-F

IMPACT OF COGENERATION UPON COMPARATIVE COSTS OF PEBBLE BED REACTORS AND COAL-FIRED UNITS

It is not possible to estimate the cost of energy with utility ownership because the utility capital charge rate is lacking and there is no mechanism for allocating energy savings between thermal and electric use. Therefore, for analytical purposes it is assumed that the plant is owned by an electric utility having the same capital charge as Texas Gas, that Texas Gas is charged for its proportional share of the plant (i.e., if 800 MWt were required for a coal gasification plant, they would be charged for 800 MWt at the average cost of construction per kWt) and that the electric utility keeps all energy cost savings that may result from cogeneration. The results of this comparison are found in Table F-I. They indicate that the mature technology PBR could be about 5% less expensive than an atmospheric fluidized bed coal plant, and offers a considerable savings, about 20%, over conventional technology.

TABLE F-I

COST COMPARISON OF PEBBLE BED REACTOR AND COAL-FIRED UNITS TO PROVIDE THERMAL POWER WHEN COGENERATING (IN 1977 DOLLARS)

	Unit Type		
Cost Category	Conventional Coal	Atmospheric Fluidized Bed Coal	Pebble Bed Reactor
Unit Size	1500 MWt	1500 MWt	1500 MWt
Size Assignable to Gasification	800 MWt	800 MWt	800 MWt
Cost per kWt	\$190	\$150	\$215
Capital Paid Cost per kWt	\$190	\$150	\$215
Total Investment	$\$152 \times 10^6$	$\$120 \times 10^6$	$\$172 \times 10^6$
Annual Fuel Cost	$\$24.7 \times 10^6$	$\$24.7 \times 10^6$	$\$12.9 \times 10^6$
First Year Costs:			
Capital Charge	$\$33.6 \times 10^6$	$\$26.5 \times 10^6$	$\$38.0 \times 10^6$
Total Cost	$\$58.3 \times 10^6$	$\$51.2 \times 10^6$	$\$50.9 \times 10^6$
Average Year Costs:			
Capital Charge	$\$28.1 \times 10^6$	$\$22.2 \times 10^6$	$\$31.8 \times 10^6$
Total Cost	$\$52.8 \times 10^6$	$\$56.9 \times 10^6$	$\$44.7 \times 10^6$

TABLE I

OPERATING CHARACTERISTICS OF FISSION POWER REACTOR SYSTEMS

	HTGR	Pebble Bed
Fuel type:	93% enriched ^{235}U in UC_2 microspheres	93% enriched ^{235}U coated with pyrolytic C or SiC and embedded in a graphite matrix ball, 6 cm in diameter
Moderator:	graphite	graphite
Coolant:	helium	helium
Coolant Temperature: (for process heat)	980°C	950°C
Experience:	one commercial unit which has had substantial start-up problems	four years with AVR (a German reactor) with coolant temperatures = 950°C
Refueling:	shut down to refule	on-line
Reliability	80%+	80 to 90%

TABLE II

CAPITAL COST ESTIMATES OF SINGLE REACTORS OF "MATURED" TECHNOLOGY IN 1977 DOLLARS

Reactor Type	Source	Size in MWt	Cost per KWt
Prismatic (HTGR)	1	3000	$270*
Pebble Bed	2	500	$290**
Pebble Bed	3	500	$445
	3	1000	$285-360
Pebble Bed (outlet temperature of coolant 925°C)	4	500	$790
	4	1000	$495
	4	2000	$340
	4	3000	$285

* does not include interest costs during construction; these run 15.2 to 18.8% of total costs in 3 and 4; including interest costs raise estimate to $320-$330 per KWt.

**methodology for deriving cost data is very cursory; it seems that all indirect costs, about one third of total costs, were excluded.

Sources

1. General Atomic Company, "Nuclear Process Heat (VHTR) Commercialization Study: Volume 1," 6A-A14668-Vol. 1, UC-77 (December 1977).
2. G. G. Leeth and C. C. Hussey, "The Pebble Bed Reactor with Chemical Heat Pipe and Stea Turbine-Electric Output--A Multiplex Energy System," General Electric Company (March 1978).
3. General Electric Company, Energy Systems and Technology Division, "Small Nuclear Process Heat Plants (SNPH) Using Pebble Bed Reactors," GEEST 75001, ORNL-Sub-4532-1 (September 1975).
4. General Electric Company, "The VHTR for Process Heat: Draft Volume 1," GEAP-14018 UC-77 (September 1974).

TABLE III

COST OF SYNTHETIC NATURAL GAS USING THE EXXON PROCESS, IN \$ PER 10^6 BTU

Cost Category	First Year	Average Cost
Coal Feed	1.50	1.50
Gasification Catalyst*	0.25	0.25
Other Operating Costs*	1.60	1.60
By-Products Credits*	(0.20)	(0.20)
Capital Charges	2.65	2.20
Total Cost	\$5.80	\$5.35

*See Gallagher and Marshall, op cit.

TABLE IV

COST COMPARISON OF PEBBLE BED REACTOR AND CONVENTIONAL COAL-FIRED UNIT TO PROVIDE THERMAL ENERGY TO THE EXXON PROCESS (IN 1977 DOLLARS)

Cost Category	Coal:	Nuclear:
Unit Size	750 MWt	1500 MWt
Number of Units	2	1
Capital Cost per KWt	\$195	\$260
Total Investment	\$292 x 10^6	\$390 x 10^6
Annual Fuel Cost	\$46.4 x 10^6	\$24.2 x 10^6
First Year Costs:		
Capital Charge	\$64.6 x 10^6	\$86.2 x 10^6
Total Cost	\$111.0 x 10^6	\$110.4 x 10^6
Average Year Costs:		
Capital Charge	\$54.0 x 10^6	\$72.1 x 10^6
Total Cost	\$100.4 x 10^6	\$96.3 x 10^6

TABLE V

COST COMPARISON OF PEBBLE BED REACTOR AND CONVENTIONAL COAL-FIRED UNIT TO PROVIDE ALL EXXON PROCESS REQUIREMENTS

Cost Category	Coal:	Nuclear:
Unit Size: Thermal Plant	950 MWt	1900 MWt
Number of Units	2	1
Unit Size: Turbine	160 MWe	160 MWe
Capital Cost per KWt	\$194	\$240
Total Investment	\$368 x 10^6	\$456 x 10^6
Annual Fuel Cost	\$58.8 x 10^6	\$30.7 x 10^6
First Year Costs:		
Capital Charge	\$81.4 x 10^6	\$100.8 x 10^6
Total Costs	\$140.2 x 10^6	\$151.5 x 10^6
Average Year Costs:		
Capital Costs	\$68 x 10^6	\$84.3 x 10^6
Total Costs	\$127.6 x 10^6	\$115.0 x 10^6

TABLE VI

COST COMPARISON OF PEBBLE BED REACTOR AND CONVENTIONAL COAL-FIRED UNIT TO PROVIDE THERMAL ENERGY TO THE EXXON PROCESS (IN 1977 DOLLARS)

Cost Category	Coal:	Nuclear:
Unit Size	400 MWt	800 MWt
Number of Units	2	1
Capital Cost per KWt	\$220	\$320
Total Investment	\$176 x 10^6	\$256 x 10^6
Annual Fuel Cost	\$24.7 x 10^6	\$12.9 x 10^6
First Year Costs:		
Capital Charge	\$38.9 x 10^6	\$56.6 x 10^6
Total Cost	\$63.6 x 10^6	\$69.5 x 10^6
Average Year Costs:		
Capital Charge	\$32.5 x 10^6	\$47.3 x 10^6
Total Cost	\$57.2 x 10^6	\$60.2 x 10^6

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THERMOCHEMICAL PRODUCTION OF HYDROGEN FROM WATER, A CRITICAL REVIEW

EXECUTIVE SUMMARY

This report has assessed the current status of thermochemical hydrogen technology as regards process chemistry, preliminary chemical engineering design and techno-economics for a number of cycles undergoing active research and development efforts throughout the world at this time.

Three cycles are receiving the bulk of the total effort and most of the funding:

- In the USA, the cycles are:
    1. The Hybrid Sulfuric Acid cycle - Westinghouse.
    2. The Sulfuric Acid-Hydrogen Iodide cycle - General Atomic.
- In Europe:
    3. The Hybrid Sulfuric Acid-Hydrogen Bromide Cycle - Euratom (Mark 13).

All three cycles are at the stage where a laboratory scale continuous plant can be or is in operation. The only plant (100 liters of hydrogen per hour) in operation is one at Ispra, Italy on the Mark 13 cycle. Both Westinghouse and General Atomic have been funded and expect operation of their closed-circuit laboratory units by the end of 1978 or early 1979. These plants will develop data to assess:

- reference design conversions and concentrations,
- control problems for operating equipment,
- materials problems, especially corrosion in sulfuric acid service, and
- possible by-product formation and their elimination.

At the design level of these plants, no accurate evaluation of plant capital cost or overall thermal efficiency of the cycle is possible, however, data for the design of the next (larger) scale-up should be obtained that will aid in determining these quantities.

Two other cycles were noted:

4. The Hybrid Bismuth Sulfate cycle - Los Alamos Scientific Laboratory, and
5. The Magnesium-Iodine cycle - NCLI, Japan.

The LASL cycle, in principle, offers an improvement over the earlier-mentioned cycles in two areas. By avoiding the handling of sulfuric acid other than at reasonable temperature, pressure and concentration (5-50%), the corrosion and heat penalty problems are minimized; in addition, the electrochemical voltage for the hydrogen generation reaction may be lowered as a result of operating at low acid strengths. The Japanese cycle is included to illustrate efforts in other countries. Serious difficulties exist in this cycle as a result of low conversion, mutual solubility of intermediate compounds and large quantities of water that require evaporation.

Materials problems are endemic to all cycles. In most cases reference materials for the sulfuric acid vaporization stages and the sulfuric acid or sulfur trioxide decomposition vessels have not yet been defined. A prime difficulty is the need for the vessel walls to transmit heat to interior fluids as well as withstand their corrosive effects. Serious efforts must be undertaken in the materials area prior to demonstration of any of the sulfuric acid-based cycles on a pilot plant scale under realistic pressure (30 atm) and temperature conditions.

In the area of techno-economics, several studies have been done mainly under assumed conditions. The most studied cycle has been the hybrid sulfuric acid cycle (Westinghouse and Ispra Mark 11). Values of efficiency and cost were developed in early reports by Westinghouse on the basis of "overly optimistic" operating conditions. These gave efficiencies in the 50%+ range at costs for product hydrogen at around $\$5/10^6$ BTU. Since that time, Euratom (Ispra) and further Westinghouse studies have shown values in the 35 to 45% range for the efficiency, and costs from \$7 to $\$10/10^{16}$ BTU for the hydrogen produced.

Heat penalty analysis has been applied by Funk and Knoche to determine the irreversibilities in the different steps of a thermochemical cycle. These heat penalties can be directly related to the capital cost and the hydrogen production cost for a thermochemical cycle. The method has been applied with success to the hybrid sulfuric acid cycle (however, under assumed operating conditions) to obtain cost and efficiency similar to the latter ones quoted above. Part of the problem of using this method is the lack of reliable thermodynamic data. These are gradually being accumulated for key substances such as sulfuric acid, etc.

In the comparison of electrolysis of water with thermochemical cycles for producing hydrogen, exponents of both technologies have emerged. Techno-economic assessments of these competing processes to produce hydrogen have been performed at both Euratom and at Westinghouse recently. These have shown small differences in efficiency and cost between thermochemical cycles and electrolysis. The values obtained are as follows:

Process	Efficiency (%)	Cost ($/10^6/BTU)
Thermochemical		
Ispra Mark 11	41.2	8.02
Ispra Mark 13	37.2	8.88
Westinghouse HSA	47.0	7.30
Electrolysis		
(Ispra)	32.7	8.54
Westinghouse	40.9	7.80

It appears reasonable to state that at this point in thermochemical cycle development, the differences shown above are not truly significant in view of the uncertainty in the estimation procedure. Both the thermochemical and the water electrolysis systems require further development to substantiate the assumptions used in flowsheet definition, performance capability, component design, and process economics. In view of this point, continued efforts in both technologies should be strongly supported by vigorous funding designed to obtain factual information to make a clear-cut case favoring either one or the other options for hydrogen production from water. This will probably take a 10 to 15 year developmental time period and, in view of the elasticity afforded by the price of synthetic hydrogen, it will allow adequate time to fully explore options before choosing a single thermochemical cycle or water electrolysis process for commercialization.

INTRODUCTION

Currently there is widespread interest in the development of a "hydrogen economy" as an eventual solution to many of the problems associated with the energy crisis. Hydrogen deserves serious consideration in ensuring a continuing gaseous fuel supply as it can be manufactured from a variety of thermal energy sources, and water - a relatively inexhaustible resource. Many studies have been published that discuss the advantages and disadvantages associated with the use of hydrogen as an energy carrier or "medium" for energy storage, energy transmission, and indeed for large-scale use as a non-polluting fuel. Technologies that produce hydrogen at high energy efficiencies are being developed and improved to provide a usable technology base for the future.

In addition to the potential for a "hydrogen economy", it is important to emphasize that hydrogen is a very valuable chemical commodity that is used in large volume for the production of ammonia, methanol, and in chemical processing. Requirements for these applications are increasing rapidly and it is clear that expanded production of hydrogen is necessary. It is equally clear that fossil fuel supplies are becoming inadequate to satisfy the demand for hydrogen, and that coal, a major fossil resource, not only is finite, but its use involves placing severe burdens on the environment such as the increasing level of carbon dioxide in the earth's atmosphere. Large-scale hydrogen production must, therefore, utilize "renewable" primary energy sources such as nuclear fission, fusion, and/or solar energy for the decomposition of water by thermochemical cycles, electrolysis, or perhaps, by hybrid combinations of these methods.

Hydrogen is attractive as an alternative fuel for several reasons, some of which are listed: (a) It provides a high energy density storable chemical form of energy; (b) It can be synthesized from "renewable" energy supplies and water; (c) On combustion, water is essentially the only product, thus completely compatible with the environment. After substitute natural gas (SNG), hydrogen has the best prospects for supplementing natural gas supplies (to 45 million US customers) without major changes to existing equipment for delivery and use of fuel gas.[1]

In regard to producing hydrogen by water-splitting, the potential higher efficiency and lower cost for thermochemical cycles, versus the overall electrolysis path (involving large losses due to mechanical irreversibilities in power generation) has been rather widely recognized. As a consequence, several laboratories throughout the world are conducting programs to develop thermochemical processes for water decomposition. A large number of thermochemical cycles have been conceived. Unfortunately, many of these have been published without experimental verification of the reactions in the cycle. As a result of this, most evaluations and/or comparisons of thermochemical processes for process efficiency and cost have been on assumed data or on reaction conditions that have not been actually achieved. Nevertheless, several cycles have now been published where all of the reactions in the cycle have been proven experimentally. As a consequence, the development of methodology for the engineering and cost analysis for this new technology can now be based with some firmness on the actual chemistry involved in the demonstrated cycles.

There are three important and inter-related parameters which characterize a thermochemical hydrogen production process:

- Thermal efficiency,
- Capital cost, and
- Operating cost.

The meaning of capital cost and operating cost is clear, however, it is necessary to carefully identify all of the assumptions that enter into deriving these values. The thermal efficiency of a thermochemical cycle is defined as the ratio of the higher heating value of hydrogen (325 BTU/SCF, 12,100 kJ/m^3, 286 kJ/mol) to the thermal equivalent of the total energy entering the hydrogen production process.

Figure 1 illustrates an electrolysis process for hydrogen production and a thermochemical process. The efficiency of the conventional electrolysis process--from the primary energy source to hydrogen output--is approximately 28%. Increases in the efficiencies of the power generation step as well as in the electrolytic cell might increase the overall efficiency to about 40%. Indeed, a number of electrolyzer manufacturers are projecting efficiencies of the order of 90-100%. The single most attractive feature of the thermochemical process is that it offers the potential of a high thermal efficiency by elimination of the power generation step. There are thermodynamic restrictions and irreversibilities in the thermochemical process arising from incomplete reactions and separations that have to be overcome. These are somewhat more obscure and have not been formalized to nearly the same extent as those in power generation. While it is possible to derive theoretical efficiency limitations from thermodynamic constraints, the practicalities and the resulting cost of the product hydrogen can only be determined fron engineering design work. It is probable that such engineering assessment will reveal serious flaws in most cycles, but in many cases changes in process flow sheets will be possible that will minimize the flaws. It is anticipated that this process of iteration will not only yield improvements in existing cycles, but also lead to the development of criteria to guide the search for and evaluation of newer and possibly better (in terms of efficiency) thermochemical cycles.

THERMOCHEMICAL WATER DECOMPOSITION

In its most general sense, thermal water decomposition implies the splitting of water into its elements, hydrogen and oxygen, by the use of heat. Water (liquid state) has an extremely high enthalpy and free energy of formation (-286 and -237 kJ/mol) that decrease slowly as the temperature increases. For this reason, direct or one-step processes to decompose water are impractical. Temperatures in excess of 3000 K are required to obtain a reasonable yield of hydrogen and one is faced additionally with separating this hydrogen from oxygen and the unreacted water before the products recombine. The reaction is also favored by low pressure which is detrimental if the final product is hydrogen at pipeline pressure.[2]

To improve on direct water-splitting, researchers have tried methods that deompose water in a number of steps. These processes, by which water is decomposed by a set of chemical reactions at various temperatures with complete recycling of the intermediate reactants, are known as thermochemical cycles.

Thermochemical Efficiency

The definition of efficiency, η, adopted by the International Energy Agency,[3] is the ratio of the theoretical energy required, ΔH^o, (286 kJ) to the total heat input required, Q_T, for the decomposition process, based on one mol of water. Thus,

$$\eta = \frac{\Delta H^o}{Q_T} = \frac{286}{Q_T} \quad . \tag{1}$$

The efficiency is sometimes defined on the basis of the free energy of formation of liquid water rather than on the enthalpy:[4]

$$\eta' = \frac{\Delta G^o}{Q_T} = \frac{237}{Q_T} \quad . \tag{2}$$

This definition takes into account the pressures at which the gases are produced. Under standard conditions, the ratio of the two efficiencies is 1.2:

$$\eta/\eta' = \Delta H^o/\Delta G^o = 286/237 = 1.2 \ . \tag{3}$$

The upper limit on thermochemical cycle thermal efficiency, , was first defined by Funk and Reinstrom[5] as:

$$\eta = \frac{\Delta H^o}{\Delta G^o} \quad \frac{T_h - T_c}{T_h} \quad , \tag{4}$$

where T_h and T_c represent the maximum and minimum temperatures in the cycle.

The cycle efficiency has an upper limit of 1.2 multiplied by the efficiency of a Carnot engine operating between the same temperatures in the cycle. For temperatures of 1000 K and 400 K, a 72% cycle efficiency is theoretically attainable.

The Step-Wise Decomposition of Water

The basic thermochemistry involved in the step-wise decomposition of water was published in 1966.[5] A large ΔS value is required so that the $T\Delta S$ term equals the H term for the high temperature reaction of a two-step cycle. It was concluded that simple two-step cycles would not be feasible for the 1150 K maximum temperature available from a nuclear high-temperature reactor at that time. Recently, other workers have considered the thermochemistry of water decomposition cycles and essentially confirmed the conclusions of Funk and Reinstrom. Bowman[6] has repeated the analysis in order to point out that specific values for the sum of the ΔS^o and the ΔH^o terms are required for the endothermic reactions if maximum heat efficiencies are to be realized. These values depend on the maximum temperature at which heat is available and the ΔG_f^o of H_2O at the low temperature. Thus, for a general two-step decomposition cycle:

1. R + AB → RA + B at T_1,
2. RA → R + A at T_2.

The "ideal" ΔS^o and ΔH^o values are given by,

$$\text{ideal } S^o = \frac{-G^o(AB)}{(T_2 - T_1)} , \tag{5}$$

$$\text{ideal } \Delta H^o = \Delta S^o \times T_2 . \tag{6}$$

For decomposition of water with T_1 = 400 K and T_2 = 1100 K, i.e.,

3. R + H_2O → RO + H_2 at 400 K,
4. RO → R + 1/2 O_2 at 1100 K.

For reaction 4, $\Delta S^o \sim$ 320 J/K, and $\Delta H^o \sim$ 350 kJ.

The striking feature of the above analysis is the large ΔS^o value required for the decomposition reactions. Typically, reactions such as 4 above exhibit ΔS^o changes of about 100 J/K. Thus, it is quite clear that simple two-step cycles for H_2O decomposition will not be found unless temperatures very much higher than 1100 K are used. Temperatures of 1223 K (950 C) available from HTGRs such as the pebble-bed reactor developed in Germany will not permit simple two-step thermochemical cycles.

Examination of the ideal ΔS^o values emphasizes the value of reactions with large entropy changes in water splitting cycles in order to minimize the number of reactions required. This, of course, suggests gaseous reactants and/or products to provide for large entropy change.

Practical considerations that have to be met before conceptual cycles become reality include the following criteria:

Process:
- Availability of accurate thermodynamic and equilibrium data
- Kinetic data
- Effect of losses of intermediate compounds
- Effect of competing reactions and side products

Engineering:
- Development of separation methods to allow for reactant recycle and product separation
- Minimization of heat exchange area
- Materials to withstand high temperature and hostile environments

These are the primary reasons why cycles have not yet been developed commercially although three 'laboratory-scale' models have been set up at this time. Mention has already been made of the large amount of scientific activity in this field; much of it is devoted to laboratory testing of the key reactions and to engineering evaluation of the cycles undergoing examination.

THERMOCHEMICAL CYCLES UNDER ACTIVE RESEARCH AND DEVELOPMENT

Research programs in the United States and abroad (Europe, Japan) have identified large numbers of prospective thermochemical cycles since the inception of this technology in 1972. These cycles have been screened through a series of laboratory and preliminary engineering/economic tests to determine their potential for further effort.

Support for cycle development has been granted by both government agencies (DOE, in particular) and by private industry. At present, cycles must show economic competitiveness with other cycles under development as well as with conventional and future electrolysis schemes in order to obtain funding. The economic analysis that ultimately determines product cost starts with a detailed engineering flow sheet based on (reliable) laboratory data. Workable separation schemes for process and product streams are required for product recycle and recovery. Kinetics and reaction yield obtained in the laboratory define the sizes and configuration of the needed chemical reactors as well as the amount of chemical inventory on hand. Heat exchange, an important factor in determining cycle efficiency, must be optimized both for heat recovery and minimization of heat exchange surface area. The cycle process efficiency is an important parameter that may be used, with due care, to monitor the effect of changes in process conditions and of other variables, such as alternate separation processes.

Three cycles, not necessarily the "best" ones, have survived the screening process and are presently being tested in continuous-circuit bench-scale units. Typically, these units are designed to produce hydrogen at a rate of 2 liters/min (4.25 SCF/hr) and use recycle chemicals. The basic purpose of the bench-scale tests is to demonstrate "operability" rather than to obtain serious information on cycle efficiency and cost.

The cycles are:

- Hybrid sulfuric acid cycle - (Westinghouse/Euratom Mark 11)
- Sulfuric acid - hydrogen iodide cycle - (General Atomic/Euratom Mark 16)
- Hybrid sulfuric acid - hydrogen bromide cycle - (Euratom Mark 13)

The Hybrid Sulfuric-Acid (HSA) Cycle

Hybrid cycles are those in which some of the reactions are thermally driven and others are effected by electrolysis at a lower voltage than that employed in the electrolysis of water (1.6-2.0 V). This is one of the hybrid cycles studied at an early stage in the LASL Thermochemical Hydrogen Program.[7] The two step cycle is written as:

$$1.\quad SO_2(g) + 2H_2O(l) \xrightarrow{350\ K} H_2(g) + H_2SO_4(sol)\ \text{elec.}$$

$$2.\quad H_2SO_4(g) \xrightarrow{1100\ K} H_2O(g) + SO_2(g) + 1/2\ O_2(g)\ .$$

In the United States, active development of this cycle has been pursued by the Westinghouse Electric Corporation.[8] The cycle has also received attention in Europe, primarily at the Euratom J.R.C. laboratory at Ispra, Italy. The latter have termed the HSA cycle - Mark 11. Fundamental work on the electrochemical step 5 above has recently been disclosed at the 2nd World Hydrogen Energy Conference by Appleby[9] and by Struck.[10] Due to the basic simplicity of the reaction sequence, this cycle has had detailed design studies done on it by Westinghouse,[7] Funk under contract to the Electric Power Research Institute,[8] and by Euratom.[9]

The cycle has many features which make it attractive for hydrogen production, including:

- The cycle consists of only two chemical reactions,
- The hydrogen is produced in a pure state, and
- All reactants and products are either in the liquid or in the gas phase.

A simplified flowsheet of the process is given in Fig. 2. The cycle can be divided into four major sections: the electrolyzer, the acid concentrator, the acid decomposer, and the separation system.

In the electrolyzer system, sulfur dioxide is mixed with makeup and recycle water. This solution is transferred to the anode side of the electrolyzer in which approximately 50% of the sulfur dioxide is oxidized to sulfuric acid, while hydrogen (99.9%) is evolved at the cathode. Sulfur will be produced at the cathode if sulfurous acid migrates from the anode to the cathode compartment. This will result in a loss of faradaic efficiency for hydrogen generation for the cell. To prevent migration of sulfurous acid, a membrane is placed between the two electrode compartments and a slight electrolyte overpressure is maintained in the cathode compartment. The membrane and overpressure cause an increase in the cell internal resistance and a net flow of sulfuric acid from the cathode to the anode compartment. Subsequently, the sulfuric acid stream is purged of sulfur dioxide and sent to the acid concentrator.

In the earlier Westinghouse designs,[11] an acid concentrator section was not included as the electrolyzer effluent was at 75 or 80% acid (by weight). Serious doubts as to whether electrolysis can be performed at these high concentrations have been raised by both Appleby[9] and Struck.[10] In their opinion, 55% acid is the highest concentration practical in this application. In all likelihood, therefore, an acid concentrator section, possibly a multiple-effect evaporator, will be required to concentrate the acid from 55% to the 75-80% design basis or further to the 98% azeotrope.

The decomposition system consists of the equipment required to decompose the acid into sulfur trioxide and water, and thermally reduce the sulfur trioxide to sulfur dioxide and oxygen. The acid from the concentrator is preheated and further concentrated to greater than 98% (azeotropic composition) by contact with the hot effluent of the acid decomposer and vaporized in the acid vaporizer section. The gas mixture enters a convectively heated catalytic reactor where the decomposition products, water, undecomposed sulfur trioxide, sulfur dioxide and oxygen result. On removal of the undecomposed sulfur trioxide and part of the water, this gas mixture is sent to the sulfur dioxide-oxygen separation system.

Water and the remaining sulfur dioxide can be removed by several conventional separation schemes. Compression and cooling is the scheme adopted in the Westinghouse design. Ispra is testing a gas adsorption scheme using activated charcoal as the adsorbent species. The purified oxygen is vented to the atmosphere and sulfur dioxide is returned to the electrolyzer portion of the process.

A complete description of the Westinghouse design may be found in recent reports[8,36] with efficiency and economics. In their Case 3, utilizing a Very High Temperature Nuclear Reactor (VHTR) rated at 2790 MW(t) producing 380 x 10^6 SCFD of hydrogen, an efficiency of 54.1% overall was obtained. An earlier study, published as NASA CR-134976,[11] arrived at an efficiency of 45.2%. An EPRI report authored by Funk quotes an efficiency of 44% using the same design bases (compare to 45.2%).[12] The 44% efficiency was obtained by the Lummus Company. The Euratom Laboratory using a different design basis than Westinghouse, i.e., 100,000 m^3/hr of hydrogen (roughly 1/4 the Westinghouse size) arrived at an overall thermal efficiency of 41.4%. In this author's opinion, the design bases were somewhat "over-optimistic" as regards the performance of key plant facilities, especially the electrolyzer unit. In reality, with the use of actual laboratory data for design, the plant efficiency would be below 40%. The efficiency/cost values will be reviewed in a later section of this report.

Funding for the development of this cycle is being obtained from DOE primarily, DOE-Solar is supporting some work in the sulfur trioxide decomposition area. Corporate funding is also being applied to the laboratory-scale facility that will illustrate the "proof-of-concept" for this cycle.

Construction of a continuous closed-cycle bench-scale unit is ongoing with a scheduled start of operation by November 1978 at the Westinghouse Advanced Energy Systems Division's laboratory in Pittsburgh, Pennsylvania.

Key Problem Areas - Hybrid Sulfuric Acid Cycle

Electrolyzer
- Operating voltage, current density and effluent acid concentration. These parameters will bear heavily on the cycle efficiency/cost.
- Electrode materials, should be inexpensive and long-lasting. Presently, carbon electrodes with platinum loading.
- Cell construction, to withstand 30 atm operating pressure and temperature in the 50-80°C range.

Acid concentrator and vaporizer - No reference materials for these units have been identified. Duriron and Durichlor have been suggested, but both are difficult to form and expensive.

Capital cost estimate - This is deemed inaccurate for two reasons. One, the design basis was predicated on a highly idealized conceptual design of the process (using overly optimistic assumptions as to operating data), and two, materials are as yet unidentified and thus costs are difficult to determine. Using typical chemical engineering estimating methods, based on the cost-factor approach, it is approximated that the total installed cost may be 1.5 to 3 times that presented.

In a positive light, one might add that this cycle is one of the "best" of those under present-day development and that there is sufficient evidence to state with considerable confidence that this process can be made workable technically. The process design is acceptable from the point of view of feasibility. The various steps in the cycle have been demonstrated in the laboratory or are well known in the chemical industry. However, if the proposed plant were to be built with current technology, the system could be made operable but the cost of hydrogen would be higher than that derived from the optimistic economics given by Westinghouse.

The Sulfuric Acid Hydrogen Iodide Cycle

This cycle differs from the others under development in that it is a "pure" thermochemical cycle. No electrolytic steps are included. A cycle with the same chemical steps is undergoing evaluation at the Euratom Laboratory, Ispra, Italy, and has been named the Mark 16 thermochemical cycle there. In the United States, its development is being conducted by the General Atomic Company (GA).[13]

This water-splitting process consists of the three following chemical reactions:

1. $2\ H_2O(l) + SO_2(g) + xI_2 \overset{300\ K}{=} H_2SO_4(sol) + 2\ HI_x(sol)$

2. $2HI_x(g) \overset{600\ K}{=} H_2(g) + xI_2(g)$

3. $H_2SO_4(g) \overset{1100\ K}{=} H_2O(g) + SO_2(g) + 1/2\ O_2(g)$

The first reaction is conducted at around room temperature in aqueous solution. Use of excess iodine by GA, or of excess sulfur dioxide by Euratom, causes the formation of two distinct liquid phases which can be separated by decantation. The lighter phase is predominantly sulfuric acid and the heavier (lower) phase contains the HI_x.

By itself, the second reaction is relatively straightforward, however, conversions are low (roughly 20%) at the temperatures indicated, 600-700 K, for rapid reaction kinetics. The separation of HI_x from the aqueous heavy phase of the first reaction is complicated by the formation of a HI-water azeotrope. Phosphoric acid, H_3PO_4, is used as an azeotrope breaker forming a preferential aqueous solution that must be evaporated.

The final reaction in this cycle is the decomposition of sulfuric acid that is common to this and the previous cycle under discussion. Concentration of the acid is done by multiple-effect flash evaporation in order to improve the thermal efficiency and matchup with the heat source.

A schematic diagram of the process is given in Fig. 3. The water, iodine, and sulfur dioxide enter the low-temperature (368 K) reactor where the two liquid phases are formed. The lower phase of this reaction, containing the HI_x aqueous solution, is sent to a vacuum still and desulfurizer to remove trace sulfur dioxide and sulfur. After concentration and recovery from aqueous solution, the hydrogen iodide is thermally cracked to give hydrogen and iodine vapors. The iodine is condensed from this stream and recycled to the first reactor leaving a pure hydrogen product. The sulfuric acid is concentrated and then vaporized at about 98% acid. The sulfur trioxide in this stream is thermally decomposed in a catalytic reactor to produce sulfur dioxide and oxygen. General Atomic has discovered that this gas mixture can be passed directly into the first reactor to effect the separation. The sulfur dioxide takes place in the reaction and oxygen passes through without effect and is vented to the atmosphere. This obviates the need for a separate sulfur dioxide-oxygen separation step.

The overall process efficiency, as determined from an early flowsheet prepared by the Lummus Company, was 36.2%.[12] A later flowsheet, prepared and analyzed by GA, and quoted in the EPRI report, indicates an efficiency of 41.4%. In late 1978, with 60% of the latest flowsheet optimized, GA expects an efficiency in the neighborhood of 45% or higher for their cycle.[13] This in turn contrasts with the Euratom estimate for the Mark 16 flowsheet of approximately 40%. An independent estimate of the Mark 16 flowsheet made by the Chemical Engineering section of the Belgian SCK/CEN Laboratory gives a value of 31% for the efficiency.[14] Energy consumption due to the product separation steps, for example HI decomposition, leads to this value.

As with Westinghouse, GA hopes to have in operation a continuous, closed-circuit laboratory-scale unit of their process in operation by the end of 1978. GA is receiving funds from the Gas Research Institute (GRI), DOE, and corporate sources to develop this cycle.

Key Problem Areas - Sulfuric-Acid Hydrogen Iodide Cycle

Main solution reaction - Degassing of the sulfur dioxide from the solutions. Elimination of the oxygen effluent without loss of intermediate species, sulfur dioxide or iodine. Handling and recovery of large quantities of iodine are required.

Acid concentration - Tradeoff between amount of heat recovery and capital cost. In the GA flowsheet, six flash evaporation stages are called for to concentrate sulfuric acid from 55% to 98%. These are highly capital-intensive. Materials are a major consideration here as boiling sulfuric acid is being handled.

Hydrogen iodide recovery and decomposition - Phosphoric acid is used to separate the HI-H_2O azeotrope. The resulting phosphoric acid solution must be distilled resulting in a heat penalty for the process. HI decomposition may be improved by use of a catalyst that allows iodine to be recovered in the liquid state.

GA concludes that this cycle appears to be a promising approach to producing hydrogen from non-fossil sources matching the thermal output capability of the High Temperature Gas-Cooled Reactor (HTGR) rather well.[13] The all-liquid and gas phase characteristics of the cycle are claimed to give this cycle a considerable advantage over cycles requiring solids handling. This last point has not been verified, however.

The Sulfuric-Acid Hydrogen Bromide Cycle

This cycle is another example in the class of "hybrid" cycles, one of the chemical reactions being carried out in an electrochemical cell, obviously at a lower voltage than that of water electrolysis. The Mark 13 cycle under development by the Euratom Laboratory, Ispra, Italy, consists of the following three reactions:[15]

1. $SO_2(g) + Br_2(l) + 2\ H_2O(l) \overset{350\ K}{=} 2\ HBr(g) + H_2SO_4(sol)$

2. $2\ HBr(sol) \overset{350\ K}{=} H_2(g) + Br_2(l)$ elec.

3. $H_2SO_4(g) \overset{1100\ K}{=} H_2O(g) + SO_2(g) + 1/2\ O_2(g)$.

A flowsheet of this cycle is shown in Fig. 4. Reaction 1 is performed with an excess of bromine to produce a sulfur dioxide-free gas stream containing only hydrogen bromide and bromine as well as a concentrated sulfuric acid solution at 70-80% concentration. The HBr/Br_2 gas stream is fed together with the recycle fluid from the HBr electrolysis cell, reaction 2, to a Br_2 distillation-HBr absorption tower to separate bromine for recycle to the first reaction step. The concentrated sulfuric acid is decomposed, reaction 3, in a manner similar to that already described in the previous cycles. The gaseous product containing water, sulfur trioxide, which are recycled, and sulfur dioxide and oxygen is then separated to recover sulfur dioxide. The oxygen, as before, is vented to the atmosphere. In the electrolytic cell, the entering fluid consists mainly of HBr solution practically free of bromine. The effluent contains approximately 4% bromine. The inlet HBr concentration to the cell is 45% and at the outlet decreases to 41%.

Designs on this cycle have been performed at Euratom to obtain efficiency and hydrogen cost. The cell voltage for the electrolytic step, reaction 2, is a key parameter in the process. Laboratory work has indicated a cell voltage in the range of 0.8-1.0 V at a current density of 2000 Am^{-2} at the HBr concentrations indicated above.[16] Using a value of 0.8 V for the electrolyzer voltage, an efficiency of 37.2% was obtained for this cycle.[17]

A complete, continuous, closed-cycle laboratory-scale unit operating at 100 liters/hour (4 mol H_2/hr) is now in operation at the Ispra facility of Euratom. This unit is the first working model of a hybrid thermochemical process in the world and, as such, represents a new frontier in hydrogen energy technology. The aims of this plant are to study the following:

- Whether the reference design conversions and reactant concentrations can be achieved and maintained.
- Testing of control and analytical equipment under actual, corrosive conditions.
- Detemination of possible by-product formation and developing suitable remedies.
- Obtaining data for plant scale-up.

Observation of the plant in operation during a visit to Ispra in August 1978 indicated that most of the above objectives were being met. Hydrogen was being produced in the unit and the HBr/Br_2 streams were being recycled and reused without significant loss or by-product formation. A novel feature of the plant was the use of a membraneless electrolyzer to cut down on losses due to internal cell resistance. At the time of the visit, this electrolyzer was operating at a voltage higher than the 0.8 V design specification, however this was to be remedied by the use of an electrolyzer of newer and better design (as regards electrode materials, configuration, flow passages, etc.).

Key Problem Areas - Sulfuric-Acid Hydrogen Bromide Cycle (Mark 13)

Electrolyzer - Operability at design conditions of 0.8 V or less. The electrical requirements of this cycle are rather severe as 0.8 V is approximately half the voltage requirement for water electrolyzers using advanced technology, such as the GE SPE electrolyzer. This problem may be very difficult to overcome; it also has a large effect on the efficiency.

Materials - for containment of the HBr and Br_2 species.

ALTERNATIVE CYCLES UNDERGOING ACTIVE RESEARCH

A selection of two of the cycles under experimental research was made. The cycles chosen were the Los Alamos Scientific Laboratory (LASL) Bismuth Sulfate Cycle and a Japanese cycle known as the Magnesium-Iodine Cycle. There are many other cycles being actively pursued at other laboratories such as the Institute of Gas Technology,[18,33] Argonne National Laboratory,[19] Oak Ridge National Laboratory,[20] and the Lawrence Livermore Laboratory[21] in the USA alone. The cycles are shown in Tables IA and B. However, it was felt that preliminary data on these cycles indicated either a low efficiency or a low promise of commercialization.

The LASL Bismuth Sulfate Cycle[24,25]

We have seen in previous sections that sulfuric acid is a common denominator to the hybrid cycles as well as to the GA cycle. Sulfuric acid poses serious problems, both in its handling which requires materials resistant to corrosion as well as in its concentration to the azeotropic composition, 98%. The latter step involves evaporation of sulfuric acid solutions. This operation incurs a large heat penalty unless the latent heat in the vapor is recovered. Multiple effect evaporation may accomplish this recovery but, in doing so, adds largely to the capital cost of the plant.

Avoidance of these problems can be achieved by the use of an insoluble metal sulfate that is precipitated from the sulfuric acid solution. To be most efficient, the metal sulfate should additionally not possess water of hydration. Bismuth sulfate was found to have the desired properties and the cycle thus formed is shown below:

1. $SO_2 + 2\ H_2O(l) \overset{350\ K}{=} H_2SO_4(sol) + H_2(g)$ elec.

2. $H_2SO_4(sol) + 1/3\ Bi_2O_3(s) \overset{350\ K}{=} 1/3Bi_2O_3{\bullet}3SO_3 + H_2O(l)$

3. $1/3Bi_2O_3{\bullet}3SO_3(s) \overset{900\text{-}1100\ K}{=} 1/3Bi_2O_3(s) + SO_2(g) + 1/2\ O_2(g)$.

A flowsheet of the cycle is seen in Fig. 5. The bismuth sulfate precipitates on adding bismuth oxide to the sulfuric acid produced in the electrochemical step, reaction 1. In this manner, sulfur trioxide is recovered from the sulfuric acid without the need for a costly and corrosive acid drying step. In addition, use of the bismuth sulfate or a bismuth oxysulfate may allow the electrochemical reaction to proceed at a lower acid concentration than 55% or higher required in the hybrid sulfuric acid cycle, possibly lowering the voltage requirements and therefore improving the efficiency of this cycle.

Preliminary calculations based on these effects shows a potential 10-12% efficiency improvement when solid sulfate processing is substituted for sulfuric acid in these cycles. The major challenge is to devise the means of handling large quantities of solids and process them at high temperatures (1000-1500 K).

Bismuth sulfate, $Bi_2O_3\ 3SO_3$ or $Bi_2(SO_4)_3$, decomposes with increasing temperature to a series of bismuth oxysulfates and sulfur trioxide. The latter in turn decomposes to sulfur dioxide and oxygen as shown:

4. $Bi_2O_3{\bullet}3SO_3(s) = Bi_2O_3{\bullet}2SO_3(s) + SO_3(g)$

5. $Bi_2O_3{\bullet}2SO_3(s) = Bi_2O_3{\bullet}SO_3(s) + SO_3(g)$

6. $Bi_2O_3{\bullet}SO_3(s) = Bi_2O_3(s) + SO_3(g)$

7. $SO_3(g) = SO_2(g) + 1/2\ O_2(g)$.

As a result, the options for generating SO_3 over a temperature range that includes intermediate temperatures, in addition to higher temperatures for SO_3 decomposition exist, and should be useful in achieving efficient extraction of heat from a HTGR, a fusion reactor or a solar heat source.

A laboratory-scale unit to test the decomposition of bismuth sulfate is being designed at LASL with operation contemplated for later in 1979. Initially electrical heating will be used with plans to implement the heating with a solar source later. To avoid the circulation of large quantities of bismuth sulfate, it is proposed to heat and decompose sulfur trioxide. The product sulfur dioxide and oxygen will recombine in another section of the chemical reactor giving up its thermal energy in the form of exothermic heat of reaction. We hope to try this new concept of a "chemical heat pipe" as an efficient method for heat transfer in the unit to be built later this year. The unit will be sized at approximately 100 liters/hr to be comparable in size and output with the other bench-scale units under construction, presently.

Key Problem Areas - LASL Bismuth Sulfate Cycle

Decomposer - In general, the major unknown in this cycle is solids handling. The bismuth sulfate must be decomposed to yield bismuth oxide and sulfur trioxide. Reaction rate and heat transfer to the solid phase must be rapid. The mechanism of heat transfer by use of a "chemical heat pipe" to avoid the circulation of large amounts of gases must be proven. The configuration of the solids decomposer, i.e., fixed bed, moving bed, or fluidized bed must also be selected and verified.

Electrolyzer - Electrolysis at significantly lower voltages (in dilute acid solution) must be demonstrated; the electrochemical reaction must also be tested to determine the effect of the low concentration of bismuth ion present in the solution.

The Magnesium-Iodine Cycle (Japan)

This cycle comprising four thermal steps may be carried out at temperatures around 900 K or below. It is being investigated by the National Chemical Laboratory for Industry in Tokyo, Japan. The chemical reactions included in the present cycle are represented as follows:[26]

1. $6/5\ MgO(s) + 6/5\ I_2(s) \xrightarrow{450\ K} 1/5\ Mg(IO_3)_2(s) + MgI_2(sol)$

2. $1/5\ Mg(IO_3)_2(s) \xrightarrow{900\ K} 1/5\ MgO(s) + 1/5\ I_2(g) + 1/2\ O_2(g)$

3. $MgI_2 \bullet 6\ H_2O(s) \xrightarrow{700\ K} MgO(s) + 2\ HI(g) + 5\ H_2O(g)$

4. $2\ HI(g) \xrightarrow{600\text{-}700\ K} H_2(g) + I_2(g)$.

The cycle is similar in many respects to a calcium-iodine cycle that was reported earlier by the authors.[27] The cycle flowsheet is shown on Fig. 6.

In this cycle, redox reaction 1 of iodine with magnesium oxide in aqueous solution forms magnesium iodide in the aqueous phase and the iodate as a precipitate. Thermal decomposition of the magnesium iodate results in magnesium oxide, iodine and oxygen in reaction 2. Hydrolytic decomposition of magnesium iodide into more magnesium iodide and hydrogen iodide follows with the final reaction 4 being the thermal dissociation of hydrogen iodide into product hydrogen and recycle iodine. Experiments have proven the validity of this cycle in good agreement with thermodynamic estimates. Disadvantages in the cycle with calcium showed an excess of water used to obtain a high degree of reaction 3 as well as melts of calcium oxide forming in reaction 2. These difficulties were partly remedied by the substitution of magnesium for calcium in the cycle. In addition, the temperature for reaction 2 is lowered from 1100 K to 900 K with the rates of reactions 2 and 3 being speeded up.

Cycle efficiencies for this type of cycle are not expected to be high largely as a result of having to handle chemical species in solution which involves the use of large amounts of low-temperature thermal energy for drying. In addition, the final reaction 4, decomposition of HI is energy-inefficient due to the separation problems involved and low conversion. An estimate of the efficiency for this cycle is in the 20-30% range, based on results for the similar calcium-iodine cycle.[27]

Key Problems in the Magnesium-Iodine Cycle

Reaction 1 - Both magnesium iodide and magnesium iodate are formed in this reaction with the latter precipitating. Practical problems are the degree of completion of the reaction, the solubility of the iodate, and evaporation of the solution to recover the hydrated magnesium iodide species.

Reaction 2 - Although not shown, the magnesium iodate is actually a magnesium iodate tetrahydrate, this involves driving off the water of hydration and more importantly, recovering their latent heat of vaporization in an energy-efficient cycle.

Reaction 3 - The problem is similar, six waters of hydration have to be removed in addition to supplying the endothermic heat for decomposition of magnesium iodide.

Reaction 4 - Separation of hydrogen from iodine and undecomposed hydrogen iodide must be effected.

With these difficulties and the need to evaporate large amounts of water in this cycle, it is doubtful if this cycle will be competitive with the previously mentioned cycles. The chemical studies being done will, no doubt, contribute to a greater understanding of cycles involving solution chemistry.

The Japanese have also proposed a cycle in which a mixture of barium and magnesium oxides are reacted with iodine in the first reaction. The barium/magnesium-iodine cycle may offer some interesting possibilities that have not yet been determined.

HEAT PENALTY ANALYSIS OF THERMOCHEMICAL CYCLE

A key parameter associated with any thermochemical process is the efficiency, η, that is redefined below.

Process Thermal Efficiency, $\eta = \frac{\Delta H}{Q_T}$,

where ΔH = total enthalpy associated with hydrogen produced, and Q_T = total thermal energy required from primary energy source to produce the hydrogen.

The value of H is 68.3 kcal/mol (286 kJ/mol) for all cycles and Q_T includes all the heat and work energy required by the process. Since ΔH is constant, the value of the efficiency clearly depends on the method used to compute Q_T.

It appears obvious that the most accurate method of obtaining the cycle efficiency is that of conventional chemical engineering. A complete flowsheet is drawn up based on the chemical reactions in the cycle and the separations required. Heat and mass balances are then used to calculate the process efficiency. This method is not amenable to a quick determination and thus early efforts in thermochemical hydrogen were spent in obtaining a rough estimate of this parameter for the purpose of cycle selection.

A computer program, HYDRGN, was written at the University of Kentucky under the direction of Funk to estimate the efficiency in a quick and rather simple manner.[28] The HYDRGN program used thermodynamic data for the pure components involved in each chemical reaction to calculate enthalpy, entropy, and free energy changes for each reaction step as well as for the heating and cooling steps in the cycle. A heat exchange routine determines the net heat by balancing the endothermic requirements against the exothermic heat liberated in the process. Finally, the program calculates the efficiency estimates which are used in evaluating the process.

The estimate used is shown in the equation below, and is obtained after heat matching. The heat needed by the cycle, Q', is the sum of the endothermic heats remaining after the matching process. The work of separation, calculated from the ideal work of separation, is reduced by the work that can be generated by unmatched exothermic heat and it is further assumed that the separation processes are only 50% efficient.

$$2. \quad \eta = \frac{\Delta H}{Q' + \dfrac{W_{sep}/0.5 - W_{gen}}{0.3}} \quad .$$

The 0.3 figure in the denominator is the value assumed for the conversion from heat to work (30%).

Since then, Funk and Knoche have devised a more sophisticated and accurate means of determining the process efficiency and relating this parameter to capital and operating costs, as well as to the final production cost of hydrogen.[12] The method, known as heat penalty analysis, is based on the result that the hydrogen production cost is linear with the product of the heat penalty and direct capital cost of each component or collection of components (battery) of the thermochemical plant. The heat penalties are the result of irreversibilities, or entropy production, associated with the process equipment and they are an indication of the quality, from a basic thermodynamic viewpoint, of a particular flowsheet or process design. The sum of all the heat penalties, plus an ideal heat requirement, is the total amount of energy, Q_T, required of the primary heat source. The formalism of this methodology is indicated in the equations below (process thermal efficiency expressed in terms of second law effects on "heat penalties"):

$$3. \quad \frac{1}{\eta} = \frac{1}{\eta_{id}} + \frac{1}{\Delta H} \sum_j Q_{pj} ,$$

where η_{id} = ideal efficiency - a function of the temperature of primary heat source and properties of material input and output, and Q_{pj} = heat penalty associated with the jth battery.

The heat penalty, Q_p, can be related to the entropy production by 4 (heat penalty Q_p, and entropy production, s):

$$4. \quad Q_p = \frac{T_m}{T_m - T_o} \bullet T_o \bullet s$$

where T_o = sink temperature; where T_m is a characteristic of the primary energy source,

$$5. \quad T_m = \frac{T_{out} - T_{in}}{\ln \frac{T_{out}}{T_{in}} + \frac{R}{C_p} \ln \frac{P_{in}}{P_{out}}} .$$

Combining these expressions, we get,

$$6. \quad \frac{1}{\eta} = \frac{T_m}{T_m - T_o} \left(\frac{\Delta G}{\Delta H}\right) + \frac{T_m}{T_m - T_o} \frac{T_o}{\Delta H} \sum_j s_j r_j ,$$

where ΔH and ΔG are kcal/kg H_2, s is kcal/kgX-^oK, and r is kgX/kg H_2.

Table II shows a heat penalty analysis as performed by Funk for the hybrid sulfuric acid cycle using highly optimistic values for the electrochemical reaction parameters (80% H_2SO_4, 0.45 V).

A battery in the chemical plant is usually the collection of process equipment required to accomplish a necessary step in the process. The total plant is the collection of batteries required to accomplish the overall process. There are two important characteristics associated with each battery - the capital cost and the heat penalty - and trade-offs may be made among these to minimize the production cost.

For a fixed primary energy source output:

$$7. \quad P_c = \frac{1}{\eta}\left[K_1 + K_2 \sum_j (DCC)_j\right] ,$$

where P_c = production cost, $\$/10^6$ BTU or \$/GJ; η = process thermal efficiency, $(DCC)_j$ = direct capital cost of the j^{th} battery; K_1 = constant related to the cost of primary energy; and K_2 = constant related to capital recovery and operating costs for chemical plant.

Combining the expressions for the production cost and heat penalties:

$$8. \quad P_c = \left[\frac{1}{\eta_{id}} + \frac{1}{\Delta H} \sum_j Q_{pj}\right] \cdot \left[K_1 + K_2 \sum_j (DCC)_j\right] .$$

Note the important characteristics of each battery, (1) heat penalty, Q_p, and (2) direct capital cost, DCC. The tradeoff is between capital cost and heat penalties.

The procedure is applied to the hybrid sulfuric acid process and the results are shown in Table III and plotted on a production cost, capital cost, thermal efficiency diagram, Fig. 7.

Under conditions approaching reality (point X, 0.8 V), the efficiency of the hybrid sulfuric acid process is 40% rather than 51%, with the hydrogen production cost reaching $\$10.55/10^6$ BTU (\$10.00/GJ).

MATERIALS

Key questions arise concerning the materials requirements of any thermochemical process involving sulfuric acid. The first concerns technical feasibility: i.e., whether materials can be found to effect the vaporization and decomposition of sulfuric acid at the relatively high temperatures needed for the endothermic stage of these cycles. The second question is unique to hybrid cycles, and is one of economics: can materials be found that are inexpensive enough to offset by the energy savings due to the hybrid electrochemical step relative to that of water electrolysis?

The unique materials requirements for sulfuric acid processing occur in the acid vaporizer and SO_3 reduction reactor. Candidate materials for the vaporizer that have been selected are:[11] previous metals, ceramics, superalloys, and cast high-silicon irons (duriron). Conventional experience with these materials do not match process requirements in three important areas: temperature, pressure, and acid concentration. In industrial practice with sulfuric acid, the flow is normally from the acid to the containment wall; thus, the vessel walls can be kept cooler than the acid itself. In sulfuric acid decomposition, heat must be transferred to the acid, so that wall temperatures in the heat exchanger necessarily must be higher than in the bulk acid. Current data on sulfuric acid corrosion are limited to approximately 150 C and most have been obtained only at atmospheric pressure.

In addition, the changing composition of the acid solution, the conversion to a two-phase vapor-liquid mixture, and finally the superheating to a vapor in the vaporizer section represent a diversity of chemical environments that may require more than one containment material. The problem is further exacerbated by the requirement that the containment material conform to a geometry which provides efficient heat flow to the acid. It is difficult to accomodate materials such as duriron and silicon nitride in conventional heat exchanger designs.

In summary, the feasibility and economic aspects of materials for thermochemical cycles are open questions at the present state of development. The feasibility question hinges strongly on the degree to which process operating parameters (particularly temperature and pressure) can be adjusted to accomodate materials capabilities. Experimental data on the corrosion and mechanical behavior of materials must also be provided to answer this problem.

HYDROGEN PRODUCTION - THERMOCHEMICAL CYCLES OR WATER ELECTROLYSIS

Two competing methods are available for the production of massive quantities of hydrogen for the future. These methods, thermochemical cycles and water electrolysis, both employ water as the starting raw material and are coupled to a high-temperature heat source. Water electrolysis is a well-known technology with present, relative poor characteristics which may offer improvement through some amount of research. Most hydrogen produced in the industrial countries of the world today is derived from hydrocarbons, indeed, the abundance of cheap hydrocarbons until recently, slowed efforts in the development of efficient electrolyzers. Less than 1% of the world's hydrogen supply derives from electrolysis which is used only where electricity generation is favorable or where product purity is needed for some specific application. With estimated increases in both the efficiency of power generation and of the electrolysis process forecast, thermochemical or hybrid cycles must achieve higher levels of performance and cost to be competitive.

Attempts to estimate costs have been made recently at Euratom, Ispra[31] and those at Westinghouse.[32] The Ispra efforts have attempted to estimate the costs of advanced electrolytic systems on par with hybrid thermochemical cycles. As an example, using the OPTIMO code, they have calculated the hydrogen production cost and efficiency of Mark 11, Mark 13, and Advanced water electrolysis. Their data are shown in Table IV.

Interestingly enough on the "bottom line," the hydrogen production cost is remarkably similar despite differences in the three processes. Mark 11 has a cost of $\$8.02/10^6$ BTU, Mark 13: \$8.88 and advanced electrolysis: \$8.54. Westinghouse has recently concluded a study for the Electric Power Research Institute (EPRI) that indicates much the same thing.[32] In their work, a comparison was made on the economics and efficiency for the hybrid sulfuric acid cycle and a water electrolysis process using sulfuric acid (rather than potassium hydroxide) solution as the electrolyte. The two processes were based on the same assumptions as much as possible, i.e., the same VHTR supplying thermal energy, etc. Their results show for the thermochemical cycle a 47% efficiency (at a voltage of 0.6 V in the electrochemical cell and 80% sulfuric acid) resulting in a hydrogen cost of $\$7.30/10^6$ BTU. (Note: It may be optimistic to expect hydrogen generation at 80% sulfuric acid at 0.6 V). The water electrolysis process indicated that at a cell voltage of 1.68 V, an efficiency of 41% and a cost of $7.80 for the product hydrogen would be obtained.

As all values are based on future technology, which has not yet been developed, it is "safe" to say that thermochemical hydrogen costs are in the same range as those for electrolytic hydrogen and hence continued RλD efforts in both the thermochemical (pure or hybrid) and the water electrolysis areas should proceed in parallel until one technology clearly demonstrates superiority over the other on factual, rather than on assumed, grounds.

ECONOMICS AND EFFICIENCY

Clearly the best method to determine the economics of a particular process for hydrogen manufacture is to base the determination on a realistic flowsheet of the process that in turn is based on a design supported by laboratory evidence. In the case of thermochemical hydrogen, the technology has not yet progressed to this stage of refinement, thus cost estimates are often made based on flowsheets put together on the basis of assumed design information. These assumptions are made on the hope that continued research and development will yield the desired results. This approach has the effect of yielding results that are somewhat over-optimistic when viewed in the light of actuality.

Better estimates of the efficiency and production cost of hydrogen have been made with the use of the Funk-Knoche heat penalty analysis and the OPTIMO computer code developed at the Euratom Laboratory, Ispra, Italy.[29] The heat penalty method has been described in the previous section and the Euratom methods are shown here.

The OPTIMO code uses a modular cost estimating technique based on the process flowsheet. The flowsheet must include all the unit operations necessary for the technical feasibility of the cycle and show the principal recycle streams. The operating conditions should be fixed as a result of experimental data or estimated carefully from thermodynamic considerations. A detailed plant cost estimation then can be performed after definition of the process units from the flowsheet knowing the mass flows through each unit. Knowing the mass flow and the necessary outlet conditions, the dimensions of the individual units can be calculated. As an example of this procedure, the mass flow indicates the diameter of a separation tower while the outlet conditions determine the number of plates. Once these dimensions are found, the process unit costs may be evaluated after materials considerations.

The key to obtaining the thermal efficiency of a thermochemical cycle is the heat exchange network. Heat recovery largely affects this parameter. However, there is always a compromise between the amount of heat recovered and the cost of the heat exchange surface required. It should also be evident that the heat exchanger capital cost plays a large role in determining the total plant investment. In OPTIMO, a heat recovery routine was developed to calculate the cost of each chemical process heat exchanger, in order to optimize the total heat exchange network. The calculation employs the following equation:[30]

$$\begin{array}{c} \text{Installed} \\ \text{Cost} \end{array} = \begin{array}{c} \text{Basic} \\ \text{Cost} \\ \$/\text{m}^2 \end{array} \times \begin{array}{c} \text{Size} \\ \text{Factor} \end{array} \times \begin{array}{c} \text{Pressure} \\ \text{Factor} \end{array} \times \begin{array}{c} \text{Heat} \\ \text{Transfer} \\ \text{Area, m}^2 \end{array} \times \begin{array}{c} 4.57+1.42\times \\ \text{Materials} \\ \text{Factor} \end{array}$$

All costs are based on the cost per unit area of carbon steel corrected by size, pressure, and materials factors to give the final installed cost. 4.57 is a constant relating basic to installed cost for equipment. The materials factor used is shown in Table V.

Application to the Hybrid Sulfuric Acid Cycle

The OPTIMO code was applied to a flowsheet, Fig. 8, derived at Euratom, Ispra for the hybrid sulfuric acid cycle. It must be emphasized that the results shown in the tables only refer to the flowsheet presented. A different flowsheet as developed in the US for this cycle would have different process conditions, etc., giving a completely different picture of the estimates in these tables. One distinct advantage of computer estimation such as OPTIMO is that it allows one to perform parametric analyses on key variables in the process. Parametric analysis will show the influence of these variables and indicate where more work is necessary to better define values. As an example, in the electrochemical step of this cycle, the cell voltage is closely linked to the investment cost through the current density. Parametric analysis shows that the cell voltage has a strong influence on the hydrogen production cost thus necessitating a major research effort to reduce it.

Table VI indicates the main characteristics assumed in the Euratom design of the Mark 11-V6 (hybrid sulfuric acid) process. The cell voltage was chosen at 0.62 V at an acid concentration in the cell of 75% H_2SO_4. (Author's note: These conditions may be somewhat optimistic in practice). The end result of the estimation is to obtain the thermal efficiency of the process, the total capital investment and the hydrogen production cost. The Euratom design is based on a hydrogen production rate of 29.03 x 10^3 (10^6 BTU/d) (100,000 m^3/hr) of hydrogen. The values obtained are shown in Table IV. The thermal efficiency is 41.4%, the investment cost for the chemical portion of the plant is $119.8 million resulting in a product hydrogen cost of \$8.02/10^6 BTU (7.62/GJ). The cost of process heat from the nuclear reactor (HTGR assumed) was taken as \$1.61/10^6 BTU ($1.70/GJ).

The parametric analysis for this process flowsheet is shown in Fig. 9. As the cell voltage rises to a value of 0.8 V, the thermal efficiency decreases to under 37% and the hydrogen cost increases to \$10/10^6 BTU. Other important variables influencing these values are the primary energy cost (cost of thermal energy from the primary heat source), and the minimum T that is used in the design of the heat exchangers. Increases in T lead to a reduced area, hence a decreased overall capital cost, however, the process S is increased leading to a lowered efficiency and greater energy requirements (for the same net amount of hydrogen produced). These competing effects lead to a trade-off situation; optimization of this position gives a minimum production cost, the position of which depends on the relative importance of the cost of capital and energy.

Other investigators have made estimates of the energy efficiency and hydrogen production cost for thermochemical hydrogen. Funk detailed costs for the hybrid sulfuric acid cycle and a methanol cycle (Author's comment: technically unworkable as one of the reactions that of methanol with sulfur dioxide and water to form methane and sulfuric acid, though thermodynamically feasible, does not "go" as written), in an earlier EPRI report.[12] Another appraisal of these variables was done in a similar study by Westinghouse for the same cycle.[8] In addition the Euratom Laboratory has performed an analysis using the OPTIMO code on their Mark 13 cycle.[30] These data have been collected in Table VII and are plotted on Fig. 10. The data from the table and plot show a minimum cost of

$4.90/10^6 BTU at 45% efficiency from an early Westinghouse report,[11] a later report gives the cost as $5.56 at an efficiency of 54%.[8] These costs and efficiencies were obtained for certain assumed conditions in the electrochemical reaction that have not been, and are not likely to be achieved in the near future (as a result of fundamental chemical mechanisms in the reaction). A recent Westinghouse report[32] comparing the techno-economics of the hybrid sulfuric acid cycle with that of sulfuric acid (water) electrolysis using a more realistic voltage of 0.6 V arrives with an efficiency of 47% at a hydrogen cost of $7.30/10^{6}BTU. In comparison, their assessment of the water electrolysis, of which more will be added in the next section, comes to a 41% efficiency at a cost of $7.80.

The maximum cost is for the Lawrence Livermore Laboratory Zinc-Selenium cycle indicating a $13.50 cost at 42% efficiency.[21] Part of the reason for the high cost for this cycle is a result of a large amount of material circulation, mainly water, hence capital intensive. The design was also based on laboratory data rather than on assumed conditions, which affects the flowsheet design and ultimately the hydrogen cost.

From the data presented, it appears likely that a successful thermochemical hydrogen process will have a thermal efficiency in the 40-45% range with the cost of product hydrogen being in the $8.00 to $10.00/10^6 BTU bracket. Costs lower than this will result if there is a drop in the cost of the primary heat source and/or capital equipment, both of which are very unlikely to happen.

CONCLUSIONS

- Three thermochemical processes are under active development today with efforts being made to produce hydrogen in laboratory-scale units in a continuous mode at 100 liters per hour. Design data for a larger unit will result, but data on efficiency and cost estimates will not be much furthered.
- Lesser efforts in researching other cycles are being done at laboratories both in the USA and abroad. Many seek to avoid materials and heat penalty problems caused by the use of sulfuric acid in the three leading contenders. The LASL option uses an insoluble metal sulfate to transfer sulfur values in the cycle.

- Materials problems are endemic to all cycles. A healthy effort is needed to develop high-temperature, corrosion-resistant materials for thermochemical cycle usage.
- Improvements are being made in estimating the cost and efficiency of hydrogen produced from water and a thermal energy source either by thermochemical cycle technology or by water electrolysis. These include the heat penalty analysis (Funk) and the OPTIMO computer code (Euratom). Costs of thermochemical hydrogen have been found to fall in the \$7 to \$10/10^6 BTU range with efficiencies in the 35 to 45% bracket.
- A 10 to 15 year developmental effort with increased funding of both options (thermochemical and water electrolysis) should find a clear-cut solution and resolve the situation of the "best" option to use for producing synthetic hydrogen from water.

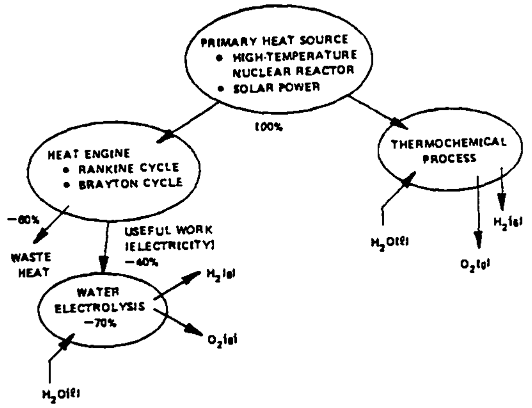

Figure 1.

Hydrogen from a thermal energy source.

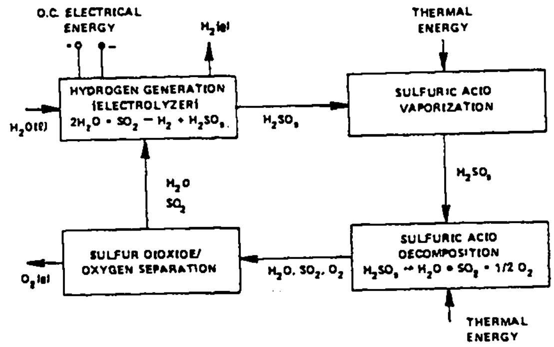

Figure 2.

Hybrid sulfuric acid cycle, schematic diagram.

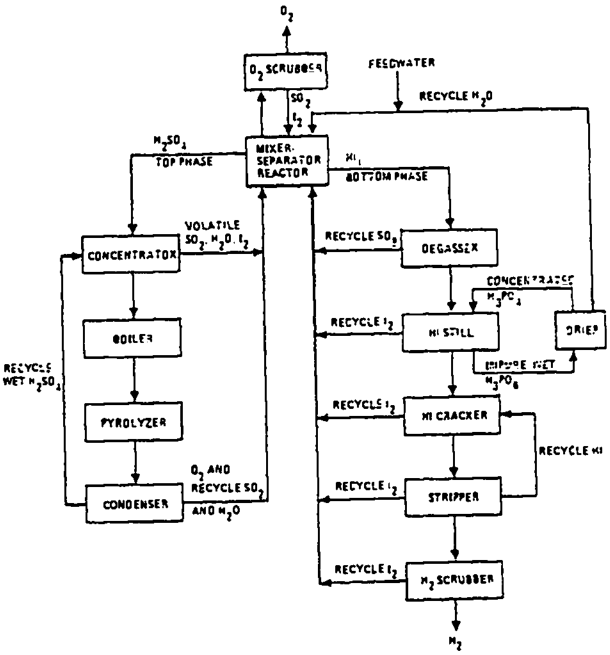

Figure 3.

Sulfuric acid-hydrogen iodide cycle, schematic diagram.

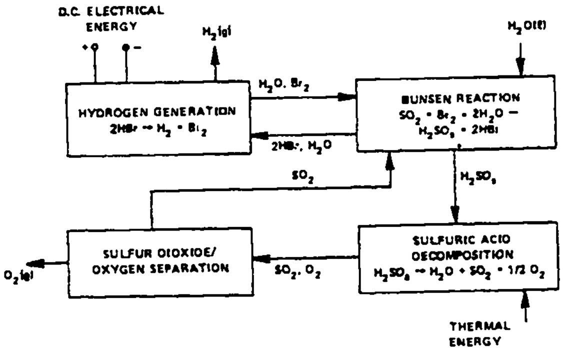

Figure 4.

Hybrid sulfuric acid-hydrogen bromide cycle, schematic diagram.

TABLE IA

COMPILATION OF OTHER THERMOCHEMICAL CYCLES UNDER DEVELOPMENT

1. Institute of Gas Technology (USA)[18]

   Cycle H-5

   1. $CuO + 6H_2O + SO_2 \rightarrow CuSO_4\ 5H_2O + H_2$
   2. $CuSO_4 \cdot 5H_2O \rightarrow CuSO_4 + 5H_2O$
   3. $CuSO_4 \rightarrow CuO + SO_3$
   4. $SO_3 \rightarrow SO_2 + 1/2\ O_2$

2. Argonne National Laboratory (USA)[19]

   Cycle ANL-4

   1. $2NH_3 + 2KI + 2CO_2 + 2H_2O \rightarrow 2NH_4I + 2KHCO_3$
   2. $2KHCO_3 \rightarrow K_2CO_3 + CO_2 + H_2O$
   3. $Hg + 2NH_4I \rightarrow 2NH_3 + HgI_2 + H_2$
   4. $HgI_2 + K_2CO_3 \rightarrow 2KI + Hg + CO_2 + 1/2O_2$

3. Hitachi (Japan)[23]

   Na_2CO_3-I_2 Cycle

   1. $2NaI + 2NH_3 + 2CO_2 + 2H_2O \rightarrow 2NaHCO_3 + 2NH_4I$
   2. $2NaHCO_3 \rightarrow Na_2CO_3 + CO_2 + H_2O$
   3. $2NH_4I + Ni \rightarrow NiI_2 + 2NH_3 + H_2$
   4. $NiI_2 \rightarrow Ni + I_2$

4. Lawrence Livermore Laboratory (USA)[21]

   L^3 Zinc-Selenium Cycle

   1. $2\ ZnO(s) + Se(l) + SO_2(g) \rightarrow ZnSe(s) + ZnSO_4(s)$
   2. $ZnSe(s) + 2\ HCl(aq) \rightarrow ZnCl_2(aq) + H_2Se(g)$
   3. $ZnCl_2(l) + H_2O(g) \rightarrow ZnO(s) + 2HCl(g)$
   4. $ZnSO_4(s) \rightarrow ZnO(s) + SO_2(g) + 1/2\ O_2(g)$
   5. $H_2Se(g) \rightarrow Se(l) + H_2(g)$

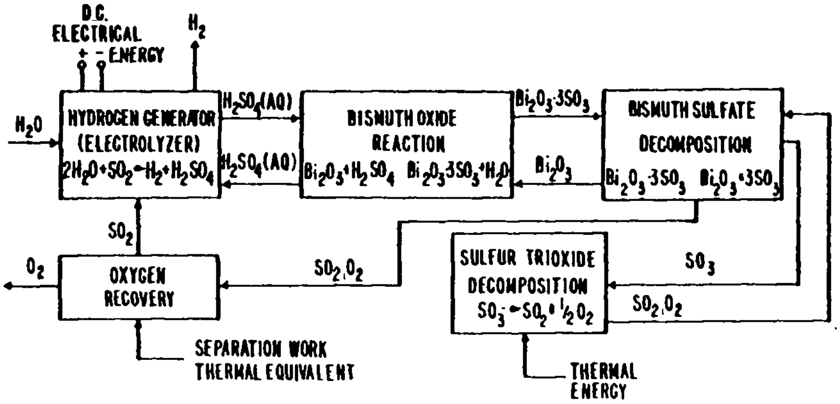

Figure 5.

Hybrid bismuth sulfate cycle, LASL, schematic diagram.

TABLE IB

CYCLES WITH DEMONSTRATED REACTIONS

1. Institute of Gas Technology (USA)[33]
   Cycle B-1
   1. $3FeCl_2 + 4H_2O \rightarrow Fe_3O_4 + 6HCl + H_2$
   2. $Fe_3O_4 + 8HCl \rightarrow 2FeCl_3 + FeCl_2 + 4H_2O$
   3. $2FeCl_3 \rightarrow 2FeCl_2 + Cl_2$
   4. $Cl_2 + H_2O \rightarrow 2HCl + 1/2O_2$

2. Oak Ridge National Laboratory (USA)[20]
   Cu/Cu (Ba, F) Cycle
   1. $2Cu^0 + Ba(OH)_2 \rightarrow BaCu_2O_2 + H_2$
   2. $BaCu_2O_2 + H_2O \rightarrow Ba(OH)_2 + CU_2O$
   3. $2Cu_2O + 4HF \rightarrow 2CuF_2 + 2Cu^0 + 2H_2O$
   4. $2CuF_2 + 2H_2O \rightarrow 2CuO + 4HF$
   5. $2CuO \rightarrow Cu_2O + 1/2\ O_2$

3. Los Alamos Scientific Laboratory (USA)[22]
   LASL Cerium-Chlorine Cycle
   1. $2CeO_2 + 8HCl = 2CeCl_3 + 4H_2O + Cl_2$
   2. $2\ CeCl_3 + 2H_2O = 2CeOCl + 4\ HCl$
   3. $2CeOCl + 2H_2O = 2CeO_2 + 2HCl + H_2$
   4. $Cl_2 + H_2O = 2HCl + 1/2O_2$

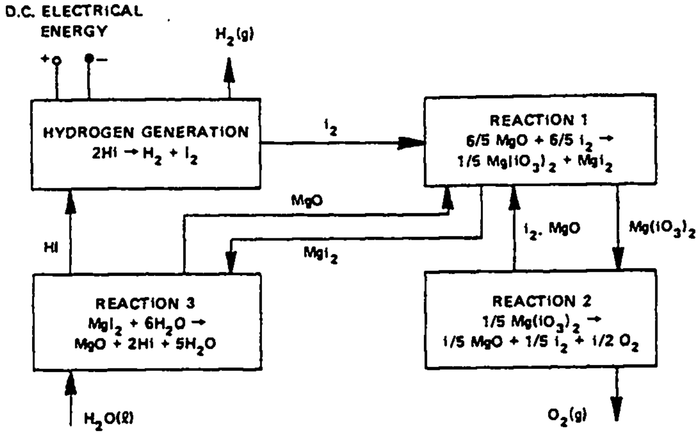

Figure 6.

Magnesium-iodine cycle, NCLI, Japan, schematic diagram.

TABLE II (Ref. 12)

HEAT PENALTY ANALYSIS
HYBRID SULFURIC ACID CYCLE

T_m = 962° K

$\frac{\Delta H}{\Delta G} = 1.2$ ΔH = 33,900 kcal/kgH_2

T_o = 300° K

		X	$\dot{s}, \frac{\text{kcal}}{\text{kgX - °K}}$	r	$\dot{s}r$	$\frac{T_m}{T_m - T_o} \frac{T_o}{\Delta h} \dot{s}r$
IHX		H_2	8.2	1	8.2	0.015
Battery						
F + G	Elec.	H_2	7.7	1	7.7	0.099
H	H_2SO_4 Decomp.	H_2SO_4	0.35	49	17.1	0.220
I	SO_2 Sep.	SO_2	0.058	32	1.8	0.023
J	Power Gen.	H_2	21	1	21	0.270
	Misc.	H_2	3	1	3	0.038
						0.755

$$\frac{T_m}{T_m - T_o}\left(\frac{\Delta g}{\Delta h}\right) = 1.206$$

$$\eta^{-1} = 1.961$$

$$\eta = 51\%$$

TABLE III (Ref. 12)

HEAT PENALTY AND CAPITAL COST ANALYSIS
HYBRID SULFURIC ACID CYCLE

1510 MW H_2
T_m = 962°C
T_o = 300°C

0.48V* at 2000 A/m2
80% H_2SO_4 *
No O_2 credit

			MW	DCC, 10^6\$
A.	Theoretical (ideal) Heat Requirement		1820	
B.	Heat Penalties			
		IHX	155	83.4
	Battery			
	F + G	Electrolyzer and Power Cond.	145	200.6
	H	H_2SO_4 Decomposition	323	167.6
	I	SO_2 Separation	35	23.6
	J	Power Generation	396	57.1
	Miscellaneous		88	
			2962	532.3
			η = 51%	

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*Overly-Optimistic assumed conditions, actual conditions are 0.6V at 50% H_2SO_4, at 2000 A/m^2.

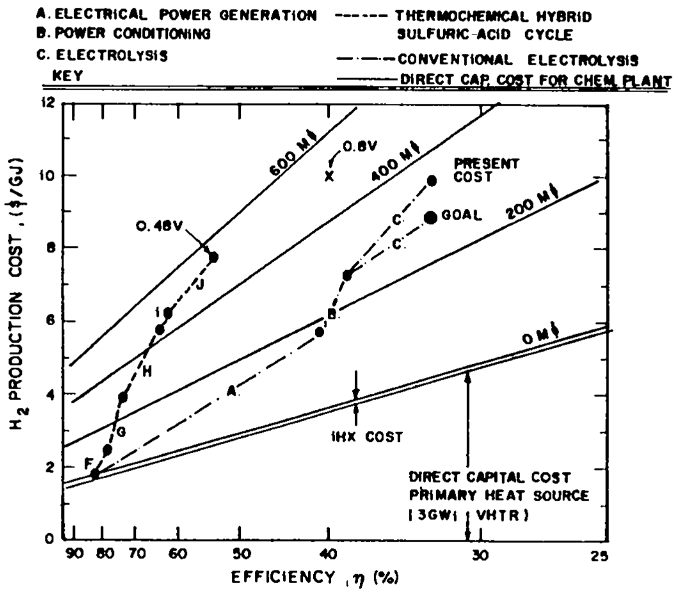

Figure 7. Production cost, capital cost, and efficiency; hybrid sulfuric acid cycle.

TABLE IV

COSTS FOR THERMOCHEMICAL AND ELECTROLYTIC HYDROGEN - 10-15 YEAR FUTURE TECHNOLOGY (Ref. 35)

	Mark 11 - V6					Mark 13 - V2					Electrolysis				
	Specifi-cations	Unit. Cost ($/kW)	Grass Roots (M$)	Fix. Cost (%)	Ann. Char (M$)	Specifi-cations	Unit. Cost ($/kW)	Grass Roots (M$)	Fix. Cost (%)	Ann. Char (M$)	Specifi-cations	Unit. Cost ($/kW)	Grass Roots (M$)	Fix. Cost (%)	Ann. Char (M$)
Annual Production (GJ/yr)x10^6	8.92					8.92					8.92				
(10^6 BTU/yr)x10^6	8.46					8.46					8.46				
(10^6 BTU/d)x10^3	29.0					29.0					29.0				
Overall Thermal Efficiency (%)	41.4					37.2					32.7				
Nuclear Heat Cost ($/GJ)	1.75					1.75									
($/$10^6$ BTU)	1.66					1.66									
Process Heat Required, (GJ/yr)x10^6	14.2				24.9	13.8				24.1					
(10^6 BTU/yr)x10^6	13.5					13.1									
Electricity Cost (Mills/kWh)	20.0					20.0					20.0				
Electrical Generating Efficiency (%)	38.0					38.0					38.0				
Electricity Required, (kWh/yr)x10^6	771.8				15.4	1076.				21.5	2889.				57.7
Non-energy Utilization (% of energy)	10.0				4.0	10.0				4.5	5.0				2.8
Hydrogen Plant Investment			119.8		23.6			126.3		25.0			73.9		11.7
Total Annual Charges					67.9					75.1					72.2
Hydrogen Production Cost (u/m^3)	9.69					10.72					10.30				
Hydrogen Production Cost ($/GJ)	7.61					8.42					8.09				
Hydrogen Production Cost ($/$10^6$ BTU)	8.03					8.88					8.53				

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*In this table, the prefix k = 10^3, M = 10^6.

TABLE IV

COSTS FOR THERMOCHEMICAL AND ELECTROLYTIC HYDROGEN - 10-15 YEAR FUTURE TECHNOLOGY (Ref. 35)

	Mark 11 - V6					Mark 13 - V2					Electrolysis				
	Specifications	Unit. Cost ($/kW)	Grass Roots (M$)	Fix. Cost (%)	Ann. Char (M$)	Specifications	Unit. Cost ($/kW)	Grass Roots (M$)	Fix. Cost (%)	Ann. Char (M$)	Specifications	Unit. Cost ($/kW)	Grass Roots (M$)	Fix. Cost (%)	Ann. Char (M$)
Chemical Installations			10.5	25	4.6			23.1	25	5.7					
Process Heat Exchangers			24.7	25	6.1			23.3	25	5.8					
Interest During Construction	13.0		(5.6)	11	0.6	13.0		6.0	11	0.6					
Electrolyzers, Output (In MW-thermal) (In 10^6 BTU/d)x10^3	 354.4 29.03	118.8	42.1	15	6.3	 354.4 29.03	118.8	42.1	15	6.3	 354.4 29.03	118.8	42.1	15	6.3
Electrolytic Cell Voltage (V)	0.62					0.80					1.64				
Required Electricity, DC (MWe)	148.3					191.3					400.0				
Compressors Power (MWe)	5.5		0.2	15	0.0	4.8		0.2	15	0.0					
Transformers and Rectifiers (MWe)	152.8	77.1	11.1	15	1.6	197.2	77.1	14.4	15	2.1	412.3	77.1	31.7	15	4.7
Required Electricity, AC (MWe)	158.4					202.1					412.3				
Recovered Electricity, (MWe)	48.3	476.7	23.0	15	3.4	48.5	476.7	23.1	15	3.4					
Interest During Construction (%)	8.0		(6.1)	11	(0.6)	8.0		(6.3)	11	(0.7)	8.0		(5.9)	11	0.6
Hydrogen Plant Investment[a]			119.8		23.6			126.3		25.0			73.9		11.7
H_2 Capacity (m^3/h)x10^3	100.0					100.0					100.0				
(10^6 BTU/d)x10^3	29.0					29.0					29.0				
Capacity Factor (%)	80.0					80.0					80.0				

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[a] Discrepancies in summation totals due to computer truncation.

TABLE V

MATERIALS FACTOR (Ref. 25)

Carbon Steel	1	Incoloy 6F	10
Stainless Steel	2	Hastelloy C4	11
Incoloy 800	6.5	Titanium	12
Graphite	8		

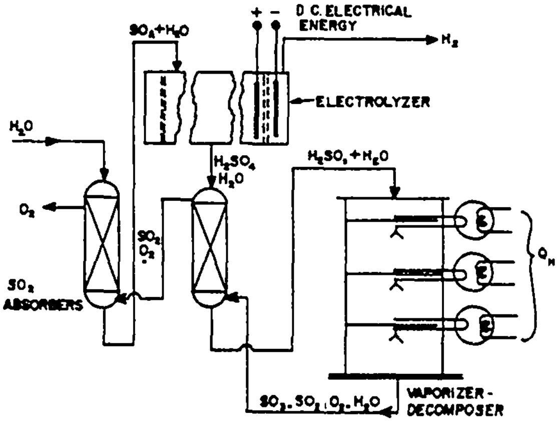

Figure 8.

Hybrid sulfuric acid cycle, ISPRA Mark 11-V6 flowsheet (Ref. 30).

TABLE VI

MAIN CHARACTERISTICS OF THE HYBRID SULFURIC ACID. MARK 11-V6 CYCLE (Ref. 30)

Plant Capacity	100.000 Nm^3H_2/h
Electrolytic Cell Voltage	0.62 V
H_2SO_4 Concentration in Electrolytic Cell	75 wt%
Electrolytic Cell Temperature	363 K
Electrolytic Cell Pressure	30 bar
H_2SO_4 Train Pressure	10 bar
SO_3 Decomposition Temperature	1083 K
SO_3 Conversion	52%
Hydrogen Delivery Pressure	30 bar
Electricity Generation Efficiency	38%
Overall Thermal Efficiency	41.4%

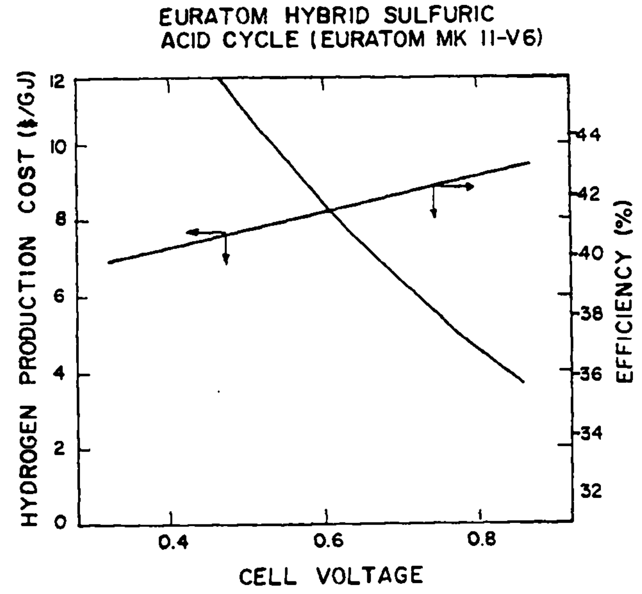

Figure 9.

Parametric analysis of hydrogen production cost and efficiency vs. electrochemical cell voltage (Ref. 30).

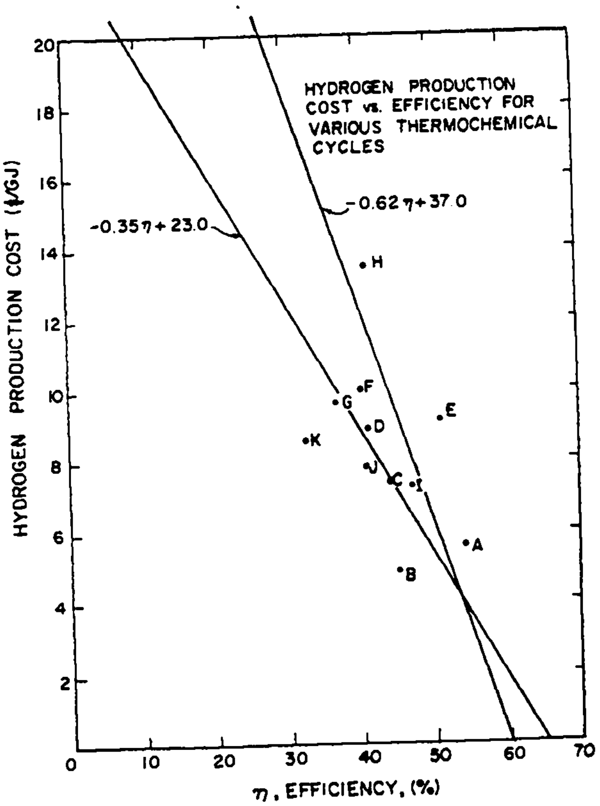

Figure 10.

Hydrogen production cost vs efficiency for thermochemical cycles.

TABLE VII

EFFICIENCY AND HYDROGEN PRODUCTION COST*
THERMOCHEMICAL CYCLES AND ELECTROLYSIS

Cycle	Efficiency (%)	Cost $\$/10^6$BTU	Done By	Reference	Point, Fig. 10
1. Hybrid Sulfuric Acid					
	54.1	5.56	Westinghouse	8	A
	45.18	4.90	Westinghouse	8	B
	47.0	7.80	Westinghouse	32	I
	44.0	7.40	Funk-Lummus	12	C
	41.3	8.85	Euratom	25	D
	51 (0.48 V)	8.20	Funk-DOE Panel	26	E
	40 (0.8 V)	10.00	Funk-DOE Panel	26	F
2. Hybrid Sulfuric Acid-Hydrogen Bromide (Ispra Mark 13)					
	36.9	9.70	Euratom	25	G
3. Zinc-Selenium					
	42.0	13.50	LLL	27	H
4. Sulfuric Acid-Hydrogen Iodide (G.A Ispra Mark 16)					
	45+	NA	General Atomic	13	
	31	NA	SCK/CEN, Belgium	14	
	41.4	NA	Funk-Lummus	12	
	36.2	NA	Funk-Lummus	12	
5. Hybrid Bismuth Sulfate					
	41	NA	LASL	24,25	
6. Hybrid Copper Sulfate					
	37.1	NA	IGT	18	
7. Sulfuric Acid (Water) Electrolysis					
	41	7.80	Westinghouse	32	J
8. Water Electrolysis (Advanced)					
	32.7	8.54	Euratom	31	K

*Nuclear heat cost is taken as $\$1.50-2.00/10^6$ BTU.

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APPENDIX A

FUSION REACTORS AS PROCESS HEAT SOURCES

INTRODUCTION

The primary goal of most United States fusion research and development programs is commercial production of electric power, and will probably so remain unless pressure for change is brought to bear on organizations sponsoring fusion research and development.

The present pattern of energy end-use in the United State [1] is indicated in Table I. According to these figures about three-quarters of the total energy consumption in this country is for space heating, industrial process heat, transportation, etc. The energy required for these purposes is provided almost exclusively by fossil fuels. Electric power either cannot or will not be used to meet many of these nonelectrical energy requirements for reasons of cost, efficiency, or insurmountable technological barriers. Therefore, if fusion energy is to become a primary source of energy when fossil fuels are no longer available, fusion reactors must be adapted to meet these nonelectrical energy requirements, especially process heat and synthetic transportable fuel requirements. Otherwise, either mismatches between production capacities and requirements for nonelectrical energy will develop or large-scale changes in energy consumption patterns must occur.

We suggest that fusion reactor concepts for process heat generation which would tend to not improve or to exacerbate the potential mismatch between capacity and demand for nonelectrical energy are not very attractive. In particular, concepts which require export of large fractions of total thermonuclear energy release as electric power for commercial success will be dropped from further consideration here, although moderate fractional electric power export will be treated as allowable. We feel that, in general, fusion plants optimized for either process heat or electric power generation will be more economical in producing these energy forms.

The results of a recent study[2] of present large-scale industrial process heat requirements and the potential of inertial confinement fusion for meeting these requirements conducted at Battelle Columbus Laboratories in the United States under the combined sponsorship of the Department of Energy and several industrial concerns are summarized in Table II. The applications which are listed are those for which the survey indicated the present existence of at least fifty such installations worldwide.

There are several points to be made concerning this list. First we note that unless facilities with the smaller listed process heat requirements can be clustered together, present projections of fusion energy production cost scaling with plant size suggest that the smaller listed requirements may not be met economically with fusion energy sources. Second, we remark that one potentially very important application was not included in the list, namely synthetic fuel production, which might be driven by thermal energy from fusion reactors. In general, the required plant thermal powers would be large, i.e., 3000 MWt (2.5×10^5 million BTU/day) or greater.

Our final point is that the lower temperature process heat requirements can be satisfied by several different technologies, including fission reactors, adaptations of electric power producing fusion reactor concepts, etc. If economically-competitive fusion electric power production technology can be developed for use when fossil fuels are no longer available, it seems reasonable to project that low temperature process heat can be provided by adaptations of such technology at competitive costs. It is only in the area of high-temperature process heat production that special high-temperature process heat fusion reactor concepts offer significant advantages relative to fission heat sources and more conventional, if this is the right word to use in describing a technology currently in a very early stage of development, fusion reactor concepts.

Therefore, we will concentrate our attention on current concepts for and thinking regarding high-temperature process heat production by fusion reactors, which incidentally corresponds to the large capacity applications considered more likely to be economically viable applications of fusion-generated thermal energy. In particular, we will consider only fusion reactor process heat blankets which can deliver thermal energy at temperatures over 1500 K (1227C or 2240F). Because heat is heat regardless of source or application, particular applications will be mentioned only for illustrative purposes.

However, we note that most attention has been focused on use of high-temperature thermal energy from fusion reactors to drive processes for hydrogen production for direct use, or as a feedstock for methane production, e.g., by direct hydrogasification of coal, to be used as substitutes for present fossil fuels such as gasoline, oil, and natural gas used as transportable fuels, chemical feedstocks, etc.

Another recent report[3] treated briefly a wide range of potential applications of fusion energy using all forms of fusion energy, including electrical, plasma, ultraviolet, x-ray, gamma-ray, and neutron energies, and thermal energy at temperatures up to about 2800 K (4580°F). Estimates of energy efficiencies, production costs, and energy requirements for a number of the processes for the year 2000 are presented. Many of these estimates, as does the projection of large scale commercial application of fusion energy by the end of the century, appear to be overly optimistic. However, the survey of industrial energy requirements is interesting.

The fundamentals of fusion were discussed in a preceeding section and the reader is referred to that section. However, several topics discussed in that section will be reviewed in greater detail here, because of their particular relevance to the present discussion. We predicate this discussion on successful development of fusion as an energy source, at least for electric power production.

The energy and power units used by the electric power industry are quite different from those used in fossil fuel industries, but each set of units is useful in the proper context. In what follows, we will provide both the units of the electric power industry which are widely used in the fusion community, and units familiar to fossil fuel supplies and consumers. For preliminary orientation purposes, however, we offer the following equivalences:

$$\begin{aligned} 1000\text{ MW} &= 8.1936 \times 10^{4}\text{ million BTU/day} \\ &= 8.1936 \times 10^{7}\text{ ft}^{3}\text{/day of 1000-BTU/ft}^{3}\text{ natural gas,} \end{aligned}$$

for consideration.

Much of what follows is necessarily rather general and somewhat vague or preliminary. Controlled thermonuclear fusion is a technology which is in a rather early stage of development, with proof of principle still not yet accomplished. Design studies of commercial applications of fusion are still primarily conceptual in nature, with many practical details having been either ignored or treated only superficially. Fusion electric power generation has received far more attention than process heat applications, and the economics of fusion reactors as process heat sources, the ultimate criterion of commercial success assuming technological feasibility, have only been addressed in preliminary fashion.

One example of such a first-order economic study is discussed in the following section on fusion-driven radiolytic production of synthetic fuels. Another such study[2] presently underway is the Battelle Columbus Laboratories study mentioned previously. We are planning a similar study at Los Alamos Scientific Laboratory in the near future. However, we believe that the discussion of issues, general principles, design options, etc., that follows will provide useful guidelines for informed assessment of progress of, and proposals for, research and development, technological validity of concepts, and potential for commercial applications in this area.

We will not attempt to characterize fusion reactor energy forms in detail here, but instead will defer such discussion to the following section on direct radiolysis. However, it is important to note that about 80% of total fusion reactor energy release is associated with highly-penetrating neutrons whose energy can be utilized outside fusion reactor cavities and hence with considerable freedom from cavity phenomenology. This is in direct contrast to fission reactors in which about 80% of the energy is carried by fission fragments of very low penetrating power. The remainder of the fusion energy release, in the form of x-rays and energetic ions, is effectively trapped inside reactor cavities. Recovery of this energy as high-temperature heat may be possible, but plant circulating power requirements may also consume all electric power which could be obtained by conversion of this energy to electric power. Therefore, design of fusion reactors to permit recovery of the x-ray and ion energy at high temperatures must be justified primarily on the basis of higher efficiency for conversion to electric power.

Although the three types of inertial confinement fusion and many types of magnetic confinement fusion can apparently be adapted relatively easily for high-temperature process heat generation, a few magnetic confinement fusion reactor types present special problems. For example, the toroidal geometry of tokamak-type reactors makes efficient high-temperature process-heat blanket design for such reactors difficult. This topic is discussed in more detail later.

In general, adaptation of fusion reactors for supply of high-temperature process heat is expected to involve only moderate changes in scale, and little or no changes in plant technology for components not concerned directly with conversion of the primary fusion energy forms into thermal energy. For example, although electric power generation capacity may be reduced to only that required for recirculating power to drive lasers, electron beam machines, or heavy ion accelerators for inertial confinement fusion or neutral beam injectors, magnets, etc., for magnetic confinement fusion, and to operate other plant systems for those process heat fusion reactor concepts in which electric power export can be avoided, the thermal energy necessary for generating recirculating power typically represents a large fraction of total plant-thermal capacity and relatively conventional electric power cycles, of the same types that would be used if the plant produced only electric power, would normally be used to supply recirculating power. Conventional ultimate heat rejection systems, e.g., once-through cooling, wet or dry cooling towers, district heating networks, etc., would be used for either type of plant. The characteristics of lasers, electron beam machines, heavy ion accelerators, neutral beam injectors, superconducting magnets, power supplies, control systems, beam transport systems, etc., required to induce fusion would be similar for both applications. Maintenance equipment and facilities buildings, radioactive waste handling, treatment, and disposal systems, fuel processing systems, etc., would also be similar.

Therefore, in devising fusion reactor concepts to supply high-temperature process heat, the reactor systems which require most attention are those which convert primary fusion energy forms into high-temperature heat and deliver it to process materials and those systems whose characteristics are significantly affected by the presence, characteristics, design requirements and restrictions, etc., of the high-temperature process heat generation and delivery systems. A list of major plant systems on which heat generation and delivery system design requirements can have significant impacts would include tritium breeding blankets, bred tritium recovery systems, first-wall protection systems, and engineered barriers to tritium escape into the biosphere.

Operation of fusion reactor blankets at higher temperatures to permit delivery of thermal energy at higher temperatures is attractive for several reasons:

- higher efficiencies for conversion of thermal energy to electric power, with associated reductions in thermal pollution by reject heat and potential reductions in electric power generation costs, become possible;
- electric power generation methods, e.g., thermionics, gas turbines, and magnetohydrodynamics, which cannot be effectively driven by lower temperature heat sources, can be efficiently used, especially as topping cycles;
- greater efficiencies in conversion of thermal energy to stored chemical energy of synthetic fuels, e.g., methane, hydrogen, or carbon monoxide, by means of thermochemical or electrothermochemical (hybrid) cycles, by high-temperature electrolysis, etc., can be achieved, provided processes such as two-step thermochemical cycles, direct thermal decomposition, etc., which can take advantage of higher temperatures can be developed;
- fusion reactors can supply thermal energy for industrial processes, e.g., coal gasification, steel making, and ammonia production, which presently consume fossil fuels for lack of suitable high-temperature nuclear heat sources; and
- delivery of thermal energy at higher temperatures permits greater flexibility in matching fusion energy delivery characteristics to application requirements.

Increases in the efficiency of thermochemical and hybrid cycles with increase in temperature at which thermal energy delivery to the process occurs are possible because the efficiencies of cycles for conversion of thermal energy to stored chemical energy are subject to the same thermodynamic limitations that the efficiencies of cycles for conversion of thermal energy to electric power are subject to.[4] Overall efficiencies of high-temperature electrolytic processes for synthetic fuel production can be improved both through decreases in free energy required to drive the dissociation reaction, and hence substitution of thermal energy for part of the electrical energy requirement, at elevated temperature and through conversion of higher-temperature thermal energy to electric power with greater efficiency.[5]

Delivery of thermal energy by fusion reactors at higher temperatures allows cascading of reject heat from higher-temperature process steps to supply heat to lower-temperature process steps to be accomplished in cases in which lower-temperature heat sources would not permit such utilization of reject heat from higher-temperature process steps by lower-temperature steps. Even in cases in which such utilization of reject heat is possible with thermal energy delivery at more modest temperatures, higher temperatures permit the use of greater temperature differences for heat transfer which can mean significant savings in process equipment costs because of decreased heat transfer area requirements. Also, chemical reactions and other unit operations can be carried out at temperatures which are optimum with respect to kinetics, separation of products and reactants, mechanical design of process equipment, etc., without significantly affecting the characteristics of other process steps, resulting in increases in overall plant efficiency.

The following is an example of a two-step thermochemical process for synthetic fuel production, which might be made practical by development of a fusion reactor concept capable of delivering thermal energy at high enough temperatures.[6] Unfortunately, this process also presents commercialization design problems that may be insurmountable. There may be other, more suitable two-step cycles, for the effort expended in the search for such cycles has been very modest when compared with that expended on lower-temperature cycles to be driven by fission reactors. Reduction of the number of steps in thermochemical cycles offers the possibility of reduction in the capital cost for process equipment and higher efficiencies through reductions in separation steps, elimination of some highly irreversible chemical reaction steps and heat transfers, etc.

The two-step cycle that we will discuss is one member of a larger family of water splitting cycles characterized by the generalized reaction steps:

$$Me_nO_m + H_2O \rightleftharpoons Me_nO_{(m+1)} + H_2$$

$$Me_nO_{(m+1)} \rightleftharpoons Me_nO_m + 1/2\ O_2\ .$$

The first is a low-temperature exothermic hydrolysis reaction of a lower valance metal oxide which liberates hydrogen and yields a higher oxide, and the second is a higher-temperature , endothermic decomposition reaction which liberates oxygen and regenerates the lower valance metal oxide. Because oxygen and hydrogen are liberated in separate reactions, they are automatically separated.

We will consider the particular member of this family defined by:

$$3FeO(c) + H_2O(g) \rightleftharpoons Fe_3O_4(c) + H_2(g)$$

$$Fe_3O_4(l) \rightleftharpoons 3FeO(\ell) + 1/2\ O_2\ .$$

The first reaction can be conducted at temperatures below about 700 K. The second can be carried out with a 1-atm oxygen partial pressure at temperatures above about 2200 K (3500 F) and at lower temperatures at lower oxygen partial pressures, e.g., at about 2000 K (3140 F) for 0.1-atm oxygen partial pressure.

One proposal[6] calls for direct neutron energy deposition in the Fe_3O_4 to accomplish the decomposition to FeO and O_2, with the suggestion that radiolysis will enhance the reaction rate and yields. Alternatively, pure thermal decomposition can be considered. The addition of fluxing agents such as CaO to the decomposing Fe_3O_4-FeO mixture to further lower the decomposition temperature, or the discovery of alternative metal oxide systems are also suggested as possibilities.

One process scheme[6] which has been suggested is to alternate the reactions in two separate blanket regions. Stationary iron oxide beds would be alternatively exposed to steam to produce Fe_3O_4 and liberate H_2, and then heated to high temperature to decompose the Fe_3O_4 and release O_2. The development of an energy-efficient design for such a process appears difficult. The Fe_3O_4-FeO system is molten above about 1775 K (2735 F) and the corrosivity of molten iron oxides is considered to represent a very serious obstacle to development of successful commercialization.

Activation of the process streams is also problematical. Another suggested process design would remove the cycle from the neutron field by using an inert gas and/or steam as heat transport fluids which are circulated through fusion reactor blankets and then through process reactors, which could involve a regenerative arrangement of static beds or circulation of solid and molten iron oxides around a circuit. Fused iron oxide mixtures are sometimes referred to as the aqua regia of high-temperature chemistry because almost all solids are soluble in them. Only one way has been seriously suggested for containment of such molten iron mixtures. One might hope to contain them by surrounding the melt with cooled solid shells of the oxides as liners to protect containment structure from attack. However, mere containment is not the only problem. Successful pumping of such mixtures is extremely difficult to envision. Also, emergency shutdown could result in fusion reactor blankets, if direct heating of the oxide mixtures is contemplated, and/or thermochemical process units plugged by solid oxides with little hope of successful startup following the end of the conditions which led to the shutdown. Maintaining a suitable solid oxide liner thickness over the entire range of conditions from normal startup to full power will also be difficult. Alternative melting and solidification of the oxides as they move around a cycle are viewed with some trepidation even for normal operations.

Japanese workers in the field of thermochemical cycles for hydrogen production by water splitting also apparently regard commercially viable operation of cycles involving molten FeO/Fe_3O_4 mixtures as a nearly impossible problem. On the other hand, as we shall discuss later, Russian workers seem quite interested in this cycle and some of its variants. It must be admitted that if the potentially fatal obstacles to commercialization of this cycle which have been described could be overcome, the iron oxide cycle would be very attractive because of potential very high cycle efficiencies.

Hydrogen can be produced by direct thermal decomposition of water to hydrogen and oxygen or carbon dioxide to carbon monoxide and oxygen followed by the water gas shift reaction.[7] At sufficiently high temperatures and with effective thermal energy recuperation or efficient conversion to work, thermal efficiencies of direct thermal decomposition processes for hydrogen production can in theory be very high, i.e., greater than 90%. However, the temperatures required are very high, much higher than we have been considering up to this point, as indicated by the entries in Table III. In computing the product mole fractions, dissociation of H_2 and O_2 to the atomic species H and O, important only at very high temperatures, were neglected. Lower pressures, of course, mean greater dissociation of the water or carbon dioxide, but at the cost of very large energy and cost penalties for compression to pipeline pressures. The magnitudes of these penalties are given approximately in Figure 1.[8]

The materials problems associated with direct decomposition of water or carbon dioxide are perhaps as severe as for any of the other high-temperature processes which have been mentioned, because of the extreme temperatures involved, even though carbon dioxide and water are relatively benign substances. There are not many materials of construction which have adequate strengths and creep resistances at temperatures of 3000 K and above and all are ceramics or graphite. Materials which will be contacted by the process streams under consideration will almost assuredly have to be oxides, e.g., thorium oxide which has a melting point of 3573 K (5971°F). Other potential materials of construction which, however, must be protected from attack by these process streams include HfC (M.P. 4611 K or 7840°F), TaC (M.P. 4148 K or 7006°F), NbC (M.P. 3773 K or 6331°F), ZrC (M. P. 3533 K or 5899°F), and TiC (M. P. 3523 K or 5881°F). All of these potential materials of construction will be difficult to fabricate and costly. Cooled walls thermally isolated from the process streams by insulation made of e.g., thorium oxide represents another approach to engineering of a fusion-driven direct thermal decompositon process for synthetic fuel production.

In general, direct neutron energy depositon in a bed of refractory oxide particles contacted directly by the decomposition reaction mixture will probably be necessary for achievement of the required temperatures, although other energy delivery systems have been discussed. Tritium breeding, if mandated, will almost assumedly result in significant, i.e., to 50% or less, reduction in the fraction of fusion energy recovered at the high temperatures under consideration. Therefore, fusion reactors used to drive such processes will probably be either non-breeders, or sub-breeders, or be faced with the necessity for development of coupled applications of lower-temperature process heat.

Another very important problem for direct thermal decomposition processes for synthetic fuel production is separation of dissociated products from non-decomposed feed and from each other to prevent recombination of the dissociation reaction products. Some recombination can, of course, be tolerated if thermal energy recuperation or conversion to work is sufficiently efficient, but excessive recombination will seriously reduce process efficiency. There are at least three approaches to prevention of recombination:

- rapid quenching of the reaction mixture to temperatures at which recombination rates are sufficiently small by mixing with a cold gas or a liquid spray;
- rapid removal of energy from the reaction mixture and its conversion to work, e.g., by expansion in a turbine or a so-called energy exchanger;[9] and
- separation of decomposition reaction products at temperature.

Achievement of the cooling rates necessary for success of the first approach to recombination prevention, estimated to be as high as 10^5 K/sec or greater, may not be possible in economically viable, industrial scale plants. In addition, although thermal energy at a few hundreds of degrees centigrade can be used for electric power generation, overall plant thermal efficiencies will be significantly affected by decrease in reaction mixture energy availability resulting from the quench operation.

The second approach can result in a very efficient combined synthetic fuel - electric power plant. Gas turbines may not be suitable for this purpose because of materials limitations, slow rates of cooling of gases, and large expansion ratios required to achieve the necessary temperature decreases. So-called energy exchangers have been suggested for this application and studies of this device are currently funded at a modest level.

The third method for limitation of decomposition reaction product recombination is very appealing. Absorption of oxygen in regenerable molten metal or oxide baths or removal by diffusion through oxide membranes has been suggested as possible approaches.[7]

Combinations of high-temperature fusion reactor process heat blankets with some form of one type of new high-temperature chemical reactor, the so-called Thagard reactor [10], whose development was announced recently, appears to offer considerable promise. A number of potential applications have already been identified for this high-temperature chemical reactor and pilot plant scale demonstrations, part of an extensive research and development program sponsored by private industry, have been made, with commercialization projected for the near future, if not already accomplished, for an electrically heated version.

Process Radioactivity Hazards

In our studies of the potential utilization of fusion reactors as process heat sources, we have adopted as a ground rule the requirement that unless it can be demonstrated for a particular application that induced radioactivity will not pose significant problems, we will not consider exposing process streams to high-flux neutron irradiation. We, of course, recognize that each potential application is different and that each case must be examined on its own merits. The reasons for this ground rule are the following.

The neutron fluxes from fusion reactors are exceptionally large. Induced radioactivity in process streams resulting from neutron irradiation can cause several types of problems on the applications side of a plant in which fusion reactors are the process heat source. If the radiation emitted by the activated species consists, for example, of high energy gamma photons, which are highly penetrating, then part of or all of the process equipment may have to be surrounded by thick shielding for protection from exposure to radiation of the general public and plant operations and maintenance personnel. Considerable redesign of process equipment and large additional capital expenditures, relative to the same processes coupled with more conventional heat sources, would be required and would have a major impact on economic viability of the processes. Operations and maintenance would also be greatly complicated and more expensive, with much more extensive instrumentation and monitoring, decontamination, and perhaps even remote maintenance being required.

Even if the radiation from activated process streams is relatively nonpenetrating, e.g., consisting of alpha and beta particles which are stopped by very small thicknesses of solids and liquids, but which are often far more dangerous than gamma emitters if ingested or inhaled because of localized energy deposition, leakage of process materials during norm of operation would have to be reduced to rates far below the levels tolerated for more conventional processes and special measures, i.e., additional safety and backup systems, more stringent specifications and quality control, etc., would have to be taken to prevent exposure of the public and operating and maintenance personnel to radiation hazards in the event of natural disaster, equipment malfunction, operator error, etc.

Long-lived contamination of process equipment could result in elaborate and expensive decommissioning procedures. Industrial process equipment lifetimes are often relatively short compared to nuclear power plant design lives and the additional expense of increased process equipment lifetimes would have to be weighed against costs associated with more frequent replacement of contaminated equipment. Similar considerations apply to scheduled maintenance.

Product contamination by radioactive substances must be maintained at acceptable levels, and maximum permissible concentrations of radioactive species are being continually revised downward by regulatory agencies. Certain elements which are contained in molecules of main constituents of process streams which would be exposed to neutron irradiation for many potential applications of fusion generated thermal energy are readily transmuted through interaction with neutrons to yield long-lived and dangerous isotopes. The maximum permissible concentrations of such radioactive isotopes will be much smaller than the maximum permissible concentrations for most ordinary chemical contaminants of industrial products. In addition, the radioactive isotopes will in many, if not most, cases be of the same chemical species as the species from which they were produced, so that rather than standard separations techniques based on gross chemical or physical differences being applicable, much more difficult and expensive isotope separation techniques must be developed if the dangerous primary process stream constituents atoms are also contained in primary process product molecules. Even if main process stream constituent atomic species subject to transmutation into dangerous isotopes are not desirable species in process products, the separations required to achieve maximum permissible or lower concentrations of the activated species in process products may be very difficult and expensive.

Even if activation of atoms contained in primary process stream constituent molecules does not pose problems, process stream contaminants entering in process feed streams at concentrations considered acceptable for more conventional processing, may pose problems similar to those resulting from activation of main constituent atoms. Although, the concentrations of such contaminants can be reduced to acceptable levels by appropriate feed stream purification steps, the additional cost may threaten process economic viability.

Corrosion of activated reactor or process equipment materials of construction by process stream constituents can also introduce radioactive species into process streams. Direct contact between primary blanket coolants and process streams which might contribute significantly to heat transfer efficiency is generally ruled out because activated primary coolant impurity, or corrosion product atoms can be transferred from coolants to process streams and activatable species from process streams to coolants and because of potential contamination by tritium bred in or leaking from breeding blankets into the coolant. It is important to note that purity standards applied to and affordable for closed systems, e.g., coolant loops for fission power plants, are generally much higher than those usually applied to and affordable for conventional industrial processes, especially large-scale processes with low unit product values.

Materials Limitations

In most cases, properties of materials of construction determine limitations on maximum temperatures at which fusion reactor high-temperature process heat blankets can deliver thermal energy. For the temperature range which we are considering, i.e., over 1500 K, metallic materials of construction either do not possess adequate strengths or creep at intolerable rates under even moderate stresses. Therefore, if such materials are to be used for load-bearing structure, then clever designs which permit location of load-bearing structure outside high-temperature regions must be developed. Otherwise, the use of relatively difficult to fabricate and brittle materials such as graphites,[10-21] ceramics,[22-31] e.g., silicon nitride, silicon carbide, alumina, or sialon (alumina and silicon nitride mixture), or glass-ceramics,[32] with glass-ceramics offering advantages relative to ceramics in fabricability at penalty of somewhat lower service temperatures, will be necessary. There appears to be no way to avoid the use of such materials for main heat exchangers. However, materials of this type would probably be necessary in any event for heat exchanger surfaces in contact with many process streams, especially for oxygen-rich process streams or process streams containing halides. Fortunately, the resistances to thermal shock and neutron irradiation damage of many such materials are greater at elevated temperatures.

In the temperature range of interest, over 1500 K, successful development of pumps for circulation of coolant fluids through high-temperature fusion reactors becomes problematical, because only graphites, ceramics, glass-ceramics, etc., possess adequate strengths and resistances to creep at such temperatures. Of course, if only sensible heat transfer to and from coolants is involved or subcooled condensates are recirculated to the blanket, then conventional pumps located in the cold legs of coolant loops can be used to provide the head necessary for coolant circulation. If liquid-metal coolants are involved, then electromagnetic pumps, with pumping chambers constructed of ceramics, ceramic-glasses, or graphite for strength, with protective coatings for corrosion resistance and with pumping coils maintained at modest temperatures by insulation and/or active cooling, can be used up to very high temperatures. Such pumps have no moving parts, are available for high flow rates and high pressures, and have performance characteristics resembling centrifugal pumps. However, for magnetic confinement fusion reactor blanket cooling with single-phase liquid metals, mechanical pumping losses resulting from flow of the electrically conductive fluids across magnetic field lines of the powerful magnetic fields required for plasma confinement can be intolerable.[33,34] If phase change, i.e., boiling, provides most of the heat transport capacity of the coolant as latent heat, then pumping losses become less serious because coolant circulation rate to meet heat removal requirements are greatly reduced.[34]

However, if return of fluids to high-temperature blankets cannot be accomplished at moderate temperatures and liquid metals or other highly-conducting fluids are not involved, then some method other than mechanical pumping for circulating primary coolants must be devised or methods of thermal energy transport other than by circulation of coolant fluids must be devised. An important example which falls into the first category is the use of liquid metal-filled heat pipes or, more generally, coolant boiling in the blanket and vapor transport to, and condensation on, process heat exchanger surfaces, with gravity return of condensate to the blanket. An important example of the second type is removal of heat from solid blankets by direct thermal radiation to process streams or to process heat exchanger surfaces.

If primary heat transport is to be accomplished by circulation of fluids, then obvious leading contenders for the role of primary coolant are helium as a gaseous coolant and lithium for either single-phase or two-phase applications. Apparently few other pure fluids have been seriously considered as high-temperature fusion reactor blanket coolants. Helium possesses the well-known advantages of a relatively high specific heat capacity for a unimolecular gaseous substance and a low-viscosity characteristic of low density gases, which tend to lower pumping work requirements, and chemical and nuclear inertness. On the other hand, helium-loop pressures required for efficient loop heat transport are relatively high, i.e., at least tens of atmospheres, with obvious design implications.

Liquid lithium has a very low density (~ 0.5 g/cm^3 at room temperature, declining to less than 0.4 g/cm^3 at temperatures approaching 2000 K), a relatively high heat capacity (~ 1.0 cal/g) and latent heat of vaporization (~ 4680 cal/g at its normal boiling point), a relatively low vapor pressure at elevated temperatures (~ 0.4 atm at 1500 K, rising to ~ 15 atm at 2000 K), a high thermal conductivity (~ 0.4 to 0.7 W/cm^2-s, K/cm, in in the temperature range 300 to 2000 K) typical of liquid metals, and a low viscosity (~ 1 cp over the entire liquid range). These physical property values[35] make lithium a good single-phase, in the absence of strong magnetic fields, or two-phase coolant and liquid lithium is a superior tritium breeding material.

Thermal expansion and differential thermal expansion of dissimilar materials in reactor structure will always be a critical design topic. Care and cleverness will be necessary if credible designs are to be developed. However, there are many potential solutions to such design problems and we have not yet encountered any insurmountable problems in our preliminary design studies of high-temperature process heat fusion reactor concepts.

TRITIUM FUEL CYCLE CHARACTERISTICS AND IMPACTS ON FUSION REACTOR DESIGN FOR HIGH-TEMPERATURE PROCESS HEAT

Present indications are that at least the first generation of fusion reactors will be fueled with a mixture of deuterium (D) and tritium (T). The conditions required to induce thermonuclear reaction to proceed to the extent that more than the amount of energy required for conversion to electricity to power the devices which induce reaction, i.e., lasers, electron beam machines, heavy ion accelerators, etc., for inertial confinement fusion or superconducting magnets, neutral beam injectors, etc., for magnetic confinement fusion, a combination of temperature, density, and reaction time, is released, are less extreme for the DT fusion reaction than for other fusion reactions. The DT fusion reaction,

$$D + T \rightarrow {}^{4}He + n + \text{energy},$$

liberates one neutron per tritium nucleus undergoing fusion. These neutrons, born with approximately 14-MeV of kinetic energy, have considerable penetrating power and can pass through thin solid walls into blankets containing pure liquid lithium, molten lithium alloys, solid lithium compounds, etc., in order to breed new tritium to close the tritium fuel cycle. Fusion-born neutrons can induce the reaction ${}^{7}Li(n,n'\alpha)T$ at high energies and the reaction ${}^{6}Li(n,\alpha)T$ at low energies to yield a breeding ratio, tritium atoms produced per fusion-born neutron, in excess of unity. Tritium breeding ratios can be further improved somewhat by including materials, of which there are a number, including beryllium (for which there is a potential resource problem), lead, zirconium, niobium, molybdenum, in tritium breeding blankets to multiply the number of neutrons available for tritium breeding through (n,2n) neutron interactions.

However, tritium is a moderately dangerous radioactive isotope. Although it is only a weak beta emitter (0.01861 MeV), its physicochemical similarity to ordinary hydrogen means that it is relatively mobile and exchanges readily with protium in living organisms. Reviews of the radiological hazards of tritium have been recently published.[36,37]

Efficient recovery of bred and unburnt tritium from fusion reactor cavities, breeding blankets, and other plant subsystems and control of tritium migration within fusion electric power and process heat plants and from them into the biosphere is essential for achievement of self-sufficient tritium fuel cycles. Tritium is too rare in nature for natural sources to constitute the basis for a viable fusion economy and it can only be manufactured at unacceptably high cost, e.g., by neutron irradiation of lithium-containing materials in fission reactors, until it can be manufactured in fusion reactor breeding blankets. Some thermonuclear reactor concepts do not permit achievement of breeding ratios much greater than one and hence would consume virtually all of their bred tritium. Nonbreeding reactors and reactors with breeding ratios less than unity may have desirable characteristics for particular applications, but must be supported by reactors whose breeding ratios significantly exceed one.

Protection of the public, plant personnel, and the environment from and minimization of the hazards associated with the presence of tritium in thermonuclear reactors becomes a primary concern once the possibility of breeding and recovery of tritium adequate for sustained reactor operation is established. Tritium hazards minimization requires minimization of plant inventories, especially in those subsystems in which tritium is most difficult to control and where tritium is in its most hazardous form, e.g., due to extremes of temperature and/or pressure and/or particular physicochemical characteristics. Tritium hazards minimization also requires minimization of tritium escape into the biosphere under both normal operating conditions and abnormal conditions reslting from plant malfunction, natural disaster, human intrusion, etc.

Efficient recovery of bred and unburnt tritium is essential to maintenance of low tritium inventories. Escape of tritium to the biosphere can be minimized by maintenance of low tritium inventories and by provision of engineered barriers to tritium escape under both normal operating conditions and abnormal conditions. The fusion reactor subsystems usually considered to represent the greatest hazard under emergency conditions are the breeding blanket and main coolant loops. Thin, hot reactor components, e.g., steam generator or process heat transfer surfaces, lying across direct paths to the biosphere are considered to be critical to tritium escape into steam cycle or process streams, and thence into the biosphere. This is generally regarded to be the most important radioactivity hazard associated with normal fusion reactor operation, but one that is controllable within acceptable limits by tritium recovery systems and escape-prevention barriers.

Early estimates of rates of escape, which will probably have to be limited to no more than a few curies per day, of tritium into steam cycles from primary or secondary coolant loops through steam generator heat transfer surfaces suggested that tritium concentrations in fusion reactor breeding blankets and/or coolant loops might have to be maintained at 1 ppm or less. Because the difficulties attendant on recovery of tritium at such concentrations have become apparent to the fusion community, more effective concepts for prevention of tritium escape have been developed, and the importance of the potential hazards associated with large tritium inventories relative to other potential fusion hazards under normal operating conditions and under abnormal conditions resulting from natural disaster, plant malfunction, or sabotage have been more accurately assessed, operation with tritium concentrations in breeding blankets and/or coolant loops of one to two orders of magnitudes greater has been accorded serious consideration. To permit this, better barriers to tritium migration during normal operations and safety systems for preventing tritium escape during abnormal conditions will be necessary.

In general, tritium migration within and escape from fusion reactors is more rapid at elevated temperature and thus minimization of tritium escape from high-temperature process heat fusion reactors will be more difficult than for, e.g., fusion electric power reactors which operate at lower temperatures.

Thermodynamic and transport rate data for tritium in many advanced materials of construction, especially graphites, ceramics, glass-ceramics, etc., are scarce. Frequently, one must extrapolate over large temperature and/or concentration ranges, use data for protium and deuterium in the same materials to estimate such data for tritium by attempting to adjust them for isotopic differences, estimate on the basis of hydrogen isotope behavior in similar systems, or guess. The same situation obtains to an even greater degree with regard to our knowledge of the characteristics of solid tritium containing materials which have been proposed for use in fusion reactor tritium breeding blankets.

One widely observed, although, as we shall see, by no means universally adhered to, ground rule followed by conceptual design engineers in attempting to develop high-temperature fusion reactor process heat blankets is that such blankets must breed tritium at a sufficiently high ratio that a self-sustaining tritium fuel cycle is possible. There are several reasons for this.

One fact that it is important to take note of is that statements, made by some who have not thought about the question enough, to the effect that in a mature fusion economy, excess tritium breeding capacity will exist and therefore tritium will be available at essentially no cost from tritium breeding reactor cavities for support of nonbreeders or subbreeders are simply not true. Tritium breeding blankets and the plant subsystems necessary for containment, recovery, and processing of bred tritium are expected to contribute significantly to fusion plant capital, operating, and maintenance costs, and to impact on the economics of production of other reactor products such as electric power and process heat. Therefore, excess tritium breeding capacity will be relatively costly and hence excess capacity will only be provided for in fusion reactor design if market prices for excess tritium exceed marginal production costs.

If a process heat fusion reactor breeds tritium at a ratio less than one, or does not breed tritium at all, then other fusion reactors, which may be either electric power or process heat reactors, that breed tritium at breeding ratios greater than one, must supply tritium to the subbreeding and nonbreeding reactors. There are fusion reactor concepts that involve the use of efficient neutron multipliers such as beryllium, for which a resource availability question exists, and large lithium-containing material thicknesses, which theoretically can achieve tritium breeding ratios approaching two. Such reactors could conceivably support nearby nonbreeding process heat reactors of the same thermal power and even more subbreeders. We say approximately, because different reactor concepts involve different materials of construction, coolants, etc., which result in differences in heat generation resulting from exoergic neutron interactions with these materials, but, more importantly, because many neutronics calculations are presently done for rather simple conceptual models using cross-section data and simple one-dimensional mockups which are of limited accuracy. Thus, although in theory one need only achieve a tritium breeding ratio of almost exactly one, in view of the small tritium loss rates which must be maintained if fusion is to be a socially acceptable energy source for the future, rapid turnover of bred tritium, and moderately long half-life of tritium, to permit a self-sustained tritium fuel cycle for a reactor, conceptual fusion reactor design engineers usually attempt to attain calculated breeding ratios of at least 1.1 to 1.2. They know that more detailed design will usually result in incorporation of additional neutron-absorbing structure, loss of neutrons through necessary reactor cavity ports, etc., and they want to be realistic about the potential tritium breeding capabilities of their concept.

On the other hand, most fusion reactor concepts have calculated breeding ratios much less than two, and some are marginal breeders which consume essentially all their bred tritium. In some of these cases, the reasons for the low breeding ratios are inherent in the characteristics of the materials of construction, reactor coolants, and/or lithium-containing breeding blanket materials and/or other reactor design characteristics. In other cases, systems studies of the prospects for successful commercialization of reactor concepts indicate that economically optimum characteristics will be obtained with tritium breeding blankets which breed at ratios near one. Such a conclusion might be reached if, for example, the unit production cost of process heat or electric power were a strong function of breeding blanket thickness, total reactor size, bred tritium containment system requirements, bred tritium recovery system capacity, etc., and there were no market for excess tritium and disposal costs were high or market prices for byproduct tritium were low.

In any event, more than one megawatt of fusion reactor thermal power capacity in a reactor with a tritium breeding blanket which achieves a breeding ratio greater than one will be required to support each megawatt of nonbreeding process heat fusion reactor capacity even if breeding fusion reactor cavities are optimized for tritium breeding with probable detrimental effects on electric power or process heat production. More likely, several electric power producing reactors would be required to support a single nonbreeding, high-temperature, process-heat reactor of about the same thermal power, but perhaps only one would be required to support a subbreeder. In any event, the potential for aggravating imbalances between electric power capacity and demand is obvious.

In addition, transportation of large quantities of tritium from site to site may be deemed excessively hazardous by regulatory authorities in the future and banned. Breeding reactors could be paired with nonbreeders/subbreeders at a single site, but this may not always be feasible. Another reason for attempting to devise high-temperature process heat blankets which breed most or all of the tritium required for a self-sufficient tritium fuel cycle is that not all tritium which is introduced into fusion reactors as fuel is consumed by the thermonuclear reactions. In fact, for most fusion reactor concepts fractional burnup of injected fuel is expected to be rather modest. Therefore, fusion reactor designers will be faced with most of the tritium handling, recovery, contamination, etc., problems associated with self-sufficient tritium fuel cycles for fusion reactors whether or not they breed tritium and none of the benefits of a self-sufficient or nearly self-sufficient cycle.

A fairly substantial amount of effort has been expended in devising engineered barriers to tritium escape through steam generator heat exchanger surfaces.[38] Among the methods which appear promising are:

- selection of materials to give naturally-formed corrosion layers, e.g., oxide films, which present large resistances to tritium permeation because of low solubility of tritium in them, low diffusivities for tritium in them, slow surface process, etc.;
- engineered ceramic, e.g., oxide, barriers to tritium permeation;
- special alloys with low permeation rates; and

- double-walled tubes with a vacuum space from which tritium is pumped or a space through which helium is pumped to sweep tritium out, after the tritium has been converted to tritiated water through reaction with oxygen maintained at low partial pressures.

Tritium permeation rates through many materials, including most common metallic materials of construction, at low temperatures are acceptably small, so that many such materials can be used as tritium barriers if they can be located out of high-temperature regions, e.g., as tritium containment plena, which are kept cool by interposing insulation between the plena and the piping, around coolant loop hot leg piping.

As noted previously, the main heat exchangers represent the critical path for escape of tritium. Calculations based on the conservative assumption that the only resistance to permeation is bulk material resistance to diffusion indicate that rates of escape of bred tritium from tritium breeding blankets into the biosphere by permeation through primary heat exchangers to process streams can be controlled at acceptable levels by interposing vacuum spaces or spaces filled with a circulating fluid, such as helium, between double-walled heat exchanger transfer areas, at least one of whose walls is a ceramic. Only materials such as ceramics, glass-ceramics, graphites, etc., have the necessary strengths and creep resistance at temperatures appraoching 2000K and are therefore apparently obligatory materials of construction for primary heat exchangers at such temperatures in any event. Many of these materials display very low permeabilities for hydrogen isotopes and contribute greatly to reducing tritium leakage. Maintenance of low tritium partial pressures in a vacuum space between the walls by pumping and/or by introduction of modest amounts of oxygen to convert permeating tritium to tritiated water vapor, which does not readily permeate solids, also contributes heavily to controlling tritium permeation.

Other resistances to permeation of tritium can contribute significantly to limiting permeation rates. A list of such resistances includes slow surface processes, such as adsorption, dissociation, and dissolution on the blanket sides of the two walls and, coming out of solution, slow recombination and desorption on the process sides of the two walls, kinetic limitations on transfer in fluid phases, thermal diffusion opposing ordinary diffusion, corrosion layers, etc. Thus, permeation rate calculations which assume that only bulk materials resistances to diffusion are significant are always conservative and can be very conservative.

Heat transfer across the vacuum space is by thermal radiation. The heat transfer rate can be increased by putting helium in the space between the two walls of the heat exchanger surfaces to provide a second, parallel, mechanism for heat transfer conduction. Helium is essentially transparent to thermal radiation. Tritium entering the space is converted to tritiated water and is swept out of the space by flowing the helium through the space. The helium pressure can be adjusted to divide the total pressure loading due to difference in pressure between the blanket and process sides of the heat exchanger between the two walls. At temperatures greater than 1500K with gaseous process streams, the most likely case, the largest heat transfer rate limiting resistance will usually be the gas side film resistance. With liquid process streams, the largest resistance could be, but need not be, the resistance to conduction in the solid walls, the resistance on the blanket side, or the resistance to transfer across the space between the walls.

Fractional burnups of tritium fuel fed to fusion reactors are projected to be relatively modest and therefore unburnt tritium fuel can constitute the majority of tritium which must be recovered and recycled in fusion reactors. Depending on reactor concept, the same recovery method for recovery of unburnt tritium from reactor cavities as that used for recovery of bred tritium from tritium breeding blankets or different methods may be used. For example, if sacrificial liners or magnetic fields for deflection of ions onto engineered energy sinks, or first-wall cooling are used to prevent excessive rates of material loss and other damage to first walls, then some sort of processing of cavity exhaust gases will be necessary. There is considerable experimental and developmental activity in this important area for magnetic confinement fusion and much less for inertial confinement fusion. On the other hand, if liquid lithium thin films or thick curtains are used to protect reactor cavity first walls, and liquid lithium is used as the tritium breeding material, then the same or similar methods for recovery of bred and unburnt tritium seem appropriate.

A number of bred tritium recovery methods have been proposed. Most of these can be used only with specific classes of fusion reactor concepts, some are applicable to recovery of tritium at relatively low temperatures, and some are useful primarily at relatively high temperatures and thus can take advantage of the temperatures provided by high-temperature process heat fusion reactors. Among the methods for bred tritium recovery which have received serious consideration are:

- the use of tritium windows;
- high-temperature distillation of liquid lithium;
- cold-trapping of liquid metal, e.g., sodium or sodium-potassium, intermediate coolant loops;[39]
- gettering by solid metal alloys which have a higher affinity for tritium;[40]
- extraction of tritium from liquid lithium into molten salts[41] or liquid metal alloys;[42] and
- trapping of tritiated water or T_2 from helium coolant streams by adsorption, cryogenic condensation, absorption, etc.

A tritium window consists of a large area of a material, e.g., a vanadium or niobium alloy, through which tritium permeates at exceptionally high rates into, e.g., a vacuum or helium-filled space. Compact heat exchanger designs for tritium windows are appropriate and pressure differentials across the tritium window membranes are kept low so that membrane thicknesses can be minimized. Tritium is recovered from vacuum spaces as T_2, or T_2O if small concentrations of oxygen are maintained for conversion of T_2 to tritiated water, by adsorption, cryogenic condensation, absorption, etc. Circulation of helium results in sweeping of tritium or tritiated water from the tritium window apparatus for collection by the same methods as for vacuum tritium windows.

Tritium windows have fallen into some disfavor for lower temperature fusion reactor concepts, because of the following results of extensive research into tritium permeation through a wide variety of membranes:

- rates of permeation, an activated process whose temperature dependence is usually accurately represented by an Arrhenius-type relation, are unacceptably small at lower temperatures, and
- the discovery that in many instances slow surface dissociation or recombination processes, dissolution steps, etc., also activated processes, which have unfavorable concentration driving force dependences, are rate limiting.

Unacceptably large tritium window areas, and hence costs, are projected in many cases.

However, there is some experimental evidence that alloys or intermetallic compounds with significantly higher tritium solubility and significantly lower bulk diffusive resistances to diffusion can be found and that catalytic surface coatings capable of greatly increasing surface process rates can be developed. Furthermore, the higher permissible concentrations of tritium in fusion reactor blankets which are being considered at present and the higher temperatures which are under consideration here suggest that tritium windows should still receive serious consideration as a method for bred-tritium recovery from high-temperature process heat fusion reactors. This method is thus one which would take advantage of the high temperatures characteristic of such reactor concepts.

We will not discuss high-temperature distillation of lithium as a bred-tritium recovery method here, but will defer discussion of this method until we treat our most promising high-temperature process heat fusion reactor blanket concept, the lithium-boiler concept, for which this bred-tritium recovery method appears to be among the most promising. We will also not discuss the other bred tritium recovery methods in greater detail, because with the exception of the last one, which is clearly promising, but also largely self-explanatory, they are primarily lower-temperature processes and we are attempting to utilize methods which take advantage of the temperature provided by our high-temperature process heat fusion reactor blanket concepts.

As indicated previously, the question of simultaneous conversion of most fusion energy release to high-temperature thermal energy and tritium breeding at a ratio of one or greater is of great importance and hence we will discuss it in more detail and illustrate our discussion with some examples.

"Conventional" fusion reactor concepts, including recent concepts investigated here and elsewhere for high-temperature thermal-energy generation for electrolytic and thermochemical hydrogen production by water splitting, employ either:

- relatively low-temperature ($\lesssim$ 1000K) blankets containing pure liquid lithium, or liquid lithium-containing alloys (e.g., lithium-lead), or solid (e.g., lithium oxide), or liquid lithium-bearing compounds (e.g., "flibe", F_3LiBe) for both tritium breeding and conversion of neutron kinetic and interaction energy, which constitute ~ 80% of total fusion reactor energy release, to heat, or
- relatively low-temperature lithium-containing blanket regions or modules (possibly also containing neutron multiplying materials such as lead or beryllium for tritium breeding) and solid, high-temperature blanket regions for conversion of neutron energy to heat (plus perhaps additional tritium breeding at a ratio less than one if high-temperature lithium-bearing substances, e.g., lithium meta-aluminate are included).

A blanket concept, which involves conversion of neutron thermal energy in a refractory blanket, e.g., graphite, with no breeding capability and with energy transport by thermal radiation across a vacuum to process heat tubes, has also been proposed.[43,44] Because of extreme corrosivity, the use of molten salts as tritium-breeding blanket materials at temperatures of 1500 K or higher appears very unlikely to be feasible.

Examples of laser fusion reactor concepts which fall into the first category include the Los Alamos Scientific Laboratory wetted-wall[45] and magnetically protected wall[46] concepts, the Lawrence Livermore Laboratory lithium-curtain HYLIFE concept,[47] and the University of Wisconsin SOLASE concept.[48] Most magnetic-confinement fusion reactor concepts also fall into this category. Included in the second category are confinement-independent and magnetic-confinement-fusion process-heat blanket concepts developed recently at the Los Alamos Scientific Laboratory[49] and at Brookhaven National Laboratory.[5]

To initiate our ongoing study of the potential for application of high-temperature thermal energy from fusion reactors to drive thermochemical and electrothermochemical cycles for hydrogen production by water splitting, we examined the tradeoffs between achievement of high tritium breeding ratios and recovery of most of the fusion neutron energy as high-temperature process heat for a relatively conventional fusion reactor blanket concept. The concept, which is described in more detail in a recent paper,[49] involved a lower-temperature (800K or 980F maximum coolant temperature) tritium-breeding zone containing various combinations of lead and lithium or pure lithium and cooled by circulation of the breeder materials, close to the fusion reactor cavity and an outer nonbreeding graphite blanket region cooled by circulating helium, heated to a maximum temperature of 1500K (2240F), and insulated to minimize leakage of heat from the high-temperature region into the lower-temperature tritium breeding zone. Some results of our neutronics studies, which give us tritium breeding ratios and neutron energy deposition in the high-temperature blanket region, of a simple mockup of the concept, illustrated schematically in Fig. 2, are given in Figs. 3, 4, and 5. The implications of these results of the neutronics calculations are clear. If a tritium breeding ratio of one or greater is mandated, then only about half or less of the neutron energy, and even less of the total fusion energy release, can be converted to high-temperature, as we have defined it, thermal energy in fusion reactor blankets of this type.

A companion study[5] concerned with high-temperature water electrolysis conducted recently at Brookhaven National Laboratory considered an alternative high-temperature fusion reactor blanket configuration that, however, involves essentially the same limitation on simultaneous achievement of high tritium breeding ratio and conversion of most fusion neutron energy to high-temperature heat. This concept, which was intended for use with a tokamak-type reactor, involves blanket modules which breed tritium and blanket modules which do not, and which are operated in parallel, rather than in series as the above-described concept is operated.

THE LITHIUM BOILER HIGH-TEMPERATURE PROCESS HEAT FUSION REACTOR BLANKET CONCEPT

Two alternative configurations for our most promising high-temperature process heat fusion reactor blanket concept, the lithium boiler, are displayed schematically in Figs. 6 and 7. Many variations on these basic themes are possible, and we have not attempted here to illustrate design details. We will, however, discuss this concept in some detail because many of the design problems faced and solutions developed for high-temperature process heat fusion reactor blanets are illustrated by this concept. This concept also shows considerable promise for electric power reactor applications.

We are presently in the midst of detailed neutronics and thermal-mechanical-hydraulics design studies aimed at developing particular designs for process heat and more efficient electric power laser fusion reactors derived from the Los Alamos Scientific Laboratory wetted-wall and magnetically-protected wall electric power reactor concepts. We have also initiated similar studies to develop a lithium boiler blanket design for one magnetic confinement fusion reactor concept, the tandem mirror machine,[50] as part of our ongoing study of the potential of fusion reactors as process heat sources for thermochemical and hybrid cycles for hydrogen production by water splitting. The tandem mirror machine consists essentially of a long, horizontal cylindrical cavity, where most of the fusion reaction occurs, in which plasma confinement is accomplished with solenoidal magnet coils and magnetic mirrors at each end. This choice of a magnetic confinement concept for mating with a lithium boiler blanket was made because this type of reactor is beginning to show some promise after a relatively slow start, and because the geometry appears to be especially favorable for wrapping a lithium boiler blanket around. On the other hand, geometrical constraints on lithium boiler design are few.

Preliminary investigations[44] of potential applications of the use of thermionic devices to generate DC electric power for high-temperature water electrolysis or electrolysis steps of hybrid thermochemical cycles for hydrogen production by water splitting, with rejection of heat at temperatures high enough to meet the temperature requirements for thermal energy consumed in such processes, have indicated some promise. In particular, we have considered the coupling of thermionic devices with solid, thermally radiating blankets, to be discussed later, and with lithium boiler blankets. In both cases only modest adaptations of existing high-temperature blanket concepts and thermionic device designs are required.

The reliability of thermionic diodes for use for topping of fossil-fuel fired steam cycles for electric power generation and in fission reactors has been demonstrated in tests of several tens of thousands of hours. They are simple, rugged, and have no moving parts. They are, however, expensive and only moderately efficient and the economics of mating them with lithium boiler blankets has not yet been addressed adequately.

The former alternative, a pressure vessel concept, is perhaps simpler and is expected to involve less neutron absorption and energy deposition in structure, while the latter, a pressure tube concept, would require less massive components to be constructed of advanced materials and permit isolation of leaks by tube plugging, but would require that all structure be at temperature or result in more neutron energy deposition in cooled structure and perhaps problems in achieving tritium breeding ratios greater than one.

In both basic alternatives, 14-MeV fusion neutrons, on the average, deposit only a few percent of their energy, and an even smaller percentage of the neutrons is absorbed, in passing through first wall structure. Most of the fusion neutron kinetic and capture energy is deposited in a blanket region containing boiling lithium. Most of the neutrons passing through first wall structure can be absorbed in a liquid lithium thickness of less than two meters and tritium breeding ratios substantially greater than one can be achieved with such liquid lithium thicknesses. However, actual blanket thicknesses will be greater because of significant fractional vapor volumes. In general, the thinner the blanket, the less total blanket capital cost will be. Because of the unusual nature of the boiling and the complex geometry, vapor volume fractions are very difficult to estimate accurately and depend on various design parameters, but are projected to be in the range 0.1 to 0.3. This is a matter which may require experimental work for resolution.

However, with liquid-lithium thickness of this order of magnitude, significant escape of gamma energy, produced by (n,γ) reactions of neutrons with reactor structure and lithium, through exterior blanket walls can occur, with the definition of significant depending on context. Similarly, neutron leakage can be excessive. While we desire to deposit as much of the fusion energy as possible in our high-temperature blanket, capturing the last few percent of the neutron energy is more difficult than capturing all the rest. On the other hand, greater gamma and neutron leakage means that thicker biological shielding and more cooling of irradiated structure will be required. In the case of magnetically-confined fusion, the penalty for leakage of neutron and gamma energy is especially severe, for the leakage energy will be deposited in part in the superconducting magnets necessary for plasma confinement. These magnets are operated at very low temperatures, with current designs calling for temperatures below 20 K, and deposited energy can be removed only at expense of considerable refrigeration work. The problem of neutron and gamma energy leakage is common to all high-temperature process heat blanket concepts and not just a problem with the lithium boiler concept.

The use of lead-lithium mixtures, to be discussed in more detail later, can significantly reduce the problem of gamma leakage. Efficient absorbers of thermalized neutrons, e.g., boron-10 carbide or $^{10}B_4C$, canned in a lithium-resistant refractory alloy, can be placed around the outside of the blanket in the hot zone so that neutron interaction energy absorbed in these materials will be converted to high-temperature thermal energy. The neutron absorbers need not bear structural loads other than those resulting from the requirement that they be self-supporting. Gamma energy can also be absorbed in this manner by canned high-atomic-number materials.

Virtually all first wall protection schemes, e.g., wetted walls, magnetic fields, replaceable sacrificial liners, residual cavity atmospheres, that do not call for thick molten lithium layers, as in the Lawrence Livermore Laboratory laser fusion lithium curtain concept, or thick solid layers inside the cavity etc., can be utilized. Thermally radiating sacrificial liners can be used to transfer x-ray and ion energy into the lithium boiler blanket at high temperatures so that almost all fusion energy can be converted to high-temperature heat, if desired. In this case, the inner structural wall must run hot and hence must be made of graphite, ceramic, or glass-ceramic. Cooled structural walls, e.g., of low-alloy ferritic steel, refractory metal alloys and cooled by lithium as in the Los Alamos Scientific Laboratory wetted-wall laser fusion reactor concept or by water, can also be used. For this option, thin (to minimize neutron absorption and neutron energy loss), very efficient insulation between the high-temperature blanket and the cooled structural wall is necessary.

Identification of economical materials of construction capable of performing satisfactorily under the extreme conditions of temperature under consideration clearly represents a formidable problem. The first problem we addressed was selection of a material resistant to attack by liquid lithium at temperatures up to 2000 K (3140 F). There are apparently no materials with adequate strengths and creep resistances at temperatures substantially over 1500 K (2240 F) to serve as load-bearing structure which are also resistant to attack by lithium at such temperatures. Only such materials as graphite, ceramics, and glass-ceramics possess the necessary structural properties at such high temperatures and only certain refractory metal alloys possess the necessary resistance to attack by lithium at such temperatures.

This apparently leaves us with only two options:

- ceramics, glass-ceramics, or graphites coated with a thin (a few mils), adherent refractory metal layer or supporting a thicker (a millimeter or so) independent refractory metal alloy layer, or
- thicker (a millimeter or so) independent refractory-metal layers supported by actively cooled structural walls at substantially lower temperature, e.g., low alloy steels at a few hundreds of degrees centigrade, with an insulation layer capable of transmitting internal blanket pressure loads to the cooled structural walls, i.e., possessing substantial compressive strength, between the refractory metal liner and the structural wall.

Heat pipes using lithium as a working fluid and constructed according to the first option have been operated for long periods at 2000 K (3140 F).[51] Sealing problems in such systems have been successfully addressed.

However, we have opted for the second alternative wherever possible in the belief, as yet not thoroughly tested, that reliability would be higher and cost lower. Our refractory metal liner material is TZM (0.5 wt% Ti, 0.08 wt% Zr, balance Mo), a gettered molybdenum alloy, which was not detectably attacked in 1000-hr reflux experiments at 1923 K by high-purity lithium.[52] Low, i.e., less than 100 ppm, oxygen concentrations are essential for such performance, but are necessary for resistance to corrosion by lithium at most temperatures for most metallic materials.

We have tentatively selected for our insulation a carbon foam, made by sintering together hollow carbon spheres manufactured from petroleum coke, one form of which has a density of only 0.39 g/cm^3, a very low thermal conductivity of about 1.2 x 10^{-3} W/cm^2 (K/cm) at room temperature, and a substantial compressive strength of 1500 psi, also at room temperature.[53] The temperature dependence, especially over 1500 K, of its thermal conductivity and its compressive strength have not been measured and are difficult to estimate for such complex materials. If graphite thermal conductivity temperature dependence can be used as a guide, then decrease in thermal conductivity with temperature is to be expected. On the other hand, the thermal conductivity of amorphous carbon increases somewhat with temperature. The compressive strength is not expected to change significantly up to 2000 K (3140^{0}F). However, the behavior of the carbon foam under neutron irradiation must be investigated before a final choice can be made.

Using the room temperature thermal conductivity, we see that the heat leak through only a 1-cm thickness of this foam for a 1000 K (1800^{0}F) temperature drop through the foam is only 1.2 W/cm^2, which may be compared with typical fusion reactor neutron energy wall loadings of hundreds or even thousands of W/cm^2. First wall insulation thicknesses will apparently have to be limited to such values to hold temperature maxima in the carbon foam to acceptable levels. The temperature maxima result from energy deposition in the carbon foam and reduce heat leak through the insulation to zero. The insulation for the outer walls are not subject to significant energy deposition and can be made as thick as necessary, within limits determined primarily by economic considerations, to reduce heat leakage to specified levels.

The vapor pressure of lithium varies from about 0.4 atm at 1500 K (2240^0 F)to about 15 atm at 2000 K (3140^{0}F), well below the reported compressive strength of the foam, and sufficiently low that cooled steel structural walls need only be a centimeter or so thick. There are also other insulating materials which show promise of acceptable performance, but this carbon foam is our first choice at present. A molybdenum carbide interface a few mils thick will probably be required on the carbon-foam side of the TZM layer for compatibility, but there is established technology, some developed at Los Alamos Scientific Laboratory, in this area.

Thus the lithium boiler blanket concept involves, with the exception of heat exchangers and the thin TZM liner, relatively economical materials of construction which are relatively easily fabricated, for which a technology base already exists, and for which there appear to be no significant resource limitations.

Flow of lithium vapor to, and its condensation on, power cycle or process heat exchanger surfaces constitutes the primary blanket heat transport mechanism. Boiling, vapor flow, and condensation are very efficient heat transfer mechanisms.

Although our schematic illustrates a primary heat exchanger that is closely coupled with the reactor cavity and blanket, this is not necessary. Lithium vapor can be conducted through ducts to separate heat exchanger modules, which can be large enough to accept lithium vapor from more than one reactor cavity in the case of inertial confinement fusion plants which contain many reactor cavities or from more than one magnetic confinement fusion reactor blanket segment. In this way isolation of process fluids from the neutron environment can be assured, access for maintenance to heat exchanger modules and module isolation from remaining plant components is made easier, and module capacity can be adjusted for different applications or to meet design or manufacturing limitations with some degree of independence of reactor capacities. The penalty to be paid for these benefits is that additional expensive containment, shielding, piping, etc., will be required.

Condensate can be returned by gravity, eliminating all lithium pumping requirements, if the heat exchangers are located above the blankets. Subcooling of lithium can be performed to recover additional heat from the condensate and permit mechanical pumping of lithium back to the blanket if desired. The penalty associated with pumping of conducting fluids through magnetic fields, which can be a source of significant inefficiency for magnetic confinement reactors, has already been mentioned. There is no such penalty for the vapor.

Strong magnetic fields, such as those characteristic of magnetic confinement fusion, can inhibit boiling and gravity return of condensate, but only to a degree, and not completely.[33-34,54-56] The superheating required for rapid boiling and the gravity head required for condensate return are increased, but because the system is self-pumping, there is no loss of mechanical work which would have to be provided, e.g., by conversion of thermal energy to electricity with modest efficiency.

An important feature of the thermal energy delivery characteristics of the lithium boiler is that all thermal energy can be delivered at temperature, except for temperature drops through the heat exchanger surfaces, as latent heat of condensation. Even with substantial subcooling of condensate, the large latent heat of vaporization, about 4680 cal/g, ensures that almost all of the thermal energy will be delivered as latent heat because the specific heat of the liquid is only about 1 cal/g. Matching of reactor thermal energy delivery characteristics to application process stream heat acceptance characteristics is made much easier by this characteristic. The large latent heat of vaporization of lithium further means that vapor boilup rates are relatively low.

If a close-coupled main heat exchanger is selected, then enough depth of lithium or neutron-absorbing baffles, not shown, will be necessary to provide adequate limitation of neutron irradiation of process streams. No design problems, but a small additional cost, are foreseen.

The boiling within a lithium boiler blanket is very interesting. Neutron energy deposition within the boiling lithium is highly nonuniform, with energy deposition densities being much greater near the inside wall and declining roughly exponentially with radial distance from the neutron source. However, high liquid thermal conductivity, vigorous boiling, and large effective heat capacity promote temperature uniformity. The large effective heat capacity results from absorption of deposited energy as latent heat of vaporization and means that local temperatures in the lithium are largely independent of neutron energy deposition density. The boiling ensures rapid mixing. The nature of the boiling is unusual in the sense that energy responsible for boiling is deposited volumetrically, rather than being transferred through a solid surface. Thus, the relevance of much of our knowledge of nucleate and film boiling at surfaces is limited and experiments will probably be necessary to firm up our understanding of the phenomenon.

Nucleation is not expected to present any problem whatsoever. Control of vapor generation distribution within the blanket for optimum performance may be more important and difficult to achieve. The free surface and bubbles in the lithium boiler blanket are expected to eliminate stress waves resulting from pulsed nonuniform neutron energy deposition characteristic of inertial confinement fusion as a significant design consideration. As little as one percent vapor volume is projected to reduce these stresses, important, e.g., in the conventional wetted-wall laser fusion concept, to negligibility.[57] The expected temperature uniformity within the blanket should aid in minimizing thermal stresses in structure and material transport by the lithium.

We are examining high-temperature distillation of the blanket lithium for bred tritium recovery. This method appears to be simple and efficient, involves the same materials of construction and design problems of the lithium boiler blanket itself, is self-pumping if located above the reactor, consumes no fusion energy, either thermal or electrical, because heat removed in the overhead condenser can also be delivered to process heat streams at temperature, takes advantage of the high temperatures which are available, and a modest diversion of only a few percent of the lithium vapor from the main heat exchangers is required. This method of tritium recovery is an example of one which cannot be used at much lower temperatures, for below about 1230 K (1754^0 F) the vapor-liquid equilibria becomes unfavorable in the deuterium-lithium system and presumably the characteristics of the lithium-tritium system are similar. The ratio of deuterium atom fraction in the vapor phase to that in the liquid phase rises from 1 at about 1230 K (1754^{0}F) to 10 at about 1850 K (2870^{0}F). If this method were to be used for bred tritium recovery from reactors with lower operating temperatures, an auxiliary source of high-temperature heat would be necessary.

The use of high-temperature distillation for bred tritium recovery may provide other benefits as well. The tritium breeding reactions and some neutron interactions with reactor structure release alpha particles, which are nothing more than helium nuclei. Helium in the lithium boiler blanket, being noncondensible, will tend to be swept toward and trapped against the heat exchanger surfaces by the condensing lithium vapor. At sufficiently high concentrations, condensation rates can be reduced so much by the requirement that condensing lithium atoms diffuse through the trapped inert gas to the heat exchanger surfaces that the condensation step could become the heat transfer rate limiting step. We suggest that the best place to extract tritium vapor for introduction into the distillation column may be near the main heat exchanger transfer surfaces in hopes that the inert gases can be removed with the tritium-rich distillate from column top. Otherwise, an independent aspiration system for noncondensible gas removal would apparently have to be provided.

As noted previously, the second structural wall design option is not available for heat exchanger heat transfer surfaces. Thus, in some ways heat exchanger design represents the most critical aspect of lithium boiler development. At present we are concentrating on design of a heat exchanger for coupling the lithium boiler to a thermochemical cycle in which high-temperature heat is transferred to sulfur trioxide which is decomposing to sulfur dioxide and oxygen at temperatures up to 1500 K (2240^{0}F) and pressures of a few tens of atmospheres.

Our present approach involves shell-and-tube heat exchangers, with lithium condensation on the tube side, which would be located in separate modules. The tubes are double-walled, consistent with our earlier discussion, with inner walls of ceramic, glass-ceramic, or graphite protected by either a thin (a few mils), adherent molybdenum coating or a more massive (one or two millimeters), freely floating (to allow for differential thermal expansion) TZM layer on the lithium side. If an oxide ceramic is used for the inner wall, then attack by process gas which is rich in oxygen will be minimal in the event of a leak. Hopefully, sufficient thermal mechanical shock resistance can be provided that sudden failure of an outer tube can be contained. The outer tube must be constructed of an oxygen-resistant material. If its inner surface is protected, e.g., by a few-mill thick, adherent layer of molybdenum, then a lithium leak will not result in catastrophic failure if a design with adequate resistance to thermal-mechanical shock upon sudden failure of the inner tube can be developed. Leakage of either lithium or process gas into the space between the tubes would be detected by monitoring the effluent from the vacuum pumps or the helium stream used to remove and recover tritium permeating the inner tube wall from the lithium into the space, as was discussed previously. Because the violent reaction of oxygen in the process stream with hot lithium resulting from massive leakage of either lithium or process gas is potentially the worst credible accident, this double containment is especially important for this application. Of course, it is important for tritium containment as well.

Placing the lithium on the tube side requires higher strength for the shell because of the higher, but still moderate, pressures on the process side. However, shell-side design can be similar to that for the blanket outer shell. Also, this puts the somewhat brittle materials of the heat exchanger tubes in compression and the greatest resistance to heat transfer, the gas film resistance, is thereby associated with the largest transfer area.

Molten metal mixtures containing sufficient lithium to assure adequate breeding ratios may prove superior to pure lithium for use in the lithium boiler concept for the following reasons:

- such solutions may have significantly lower vapor pressures than pure lithium in temperature ranges of interest, thereby easing blanket containment requirements and perhaps even permitting blanket pressure at design operating temperature to be exactly matched to external pressure through modest adjustment in mixture composition;
- such mixtures can have significantly higher cross sections for neutron capture and gamma photon energy deposition, and permit thinner blankets to be used for deposition of a specified fraction of fusion neutron and gamma radiation energy, reduction of escaping neutron and gamma photon fluxes to specified levels, reduction of required shielding thicknesses, and/or reduction in external structure cooling requirements;

- the volumetric rate of vapor generation necessary for a specified blanket thermal power and operating temperature and pressure can perhaps be reduced somewhat, permitting reductions in blanket thickness, vapor conduit sizes, etc.;
- lessening of corrosion problems may be possible;
- significant adjustments in bred tritium solubility, which could lower steady-state bred tritium inventories and promote easier tritium recovery, may be possible; and
- the freezing points of such mixtures are generally lower than the freezing points of the pure constituents and lower melting points ease startup and shutdown liquid metal handling requirements.

Lead-lithium represents a system which offers at least the first, second, third, and last potential advantages. The possibility of lead-lithium mixtures offering the other two advantages listed above cannot be accurately assessed at present because the necessary experimental data is not available. However, we do know that introduction of lead into a fusion reactor blanket in the form of a solution in lithium should enhance tritium breeding ratios somewhat, owing to the significant (n,2n) reaction cross section of lead. Because of the potential for significant improvement in fusion process heat blanket characteristics through the use of such mixtures, their impact on lithium boiler blanket characteristics are scheduled to receive at least modest attention at Los Alamos Scientific Laboratory in the relatively near future.

Thermal Radiator Blankets

Solid blankets which slow and capture fusion neutrons and operate at temperatures sufficiently high that deposited energy can be efficiently removed by direct thermal radiation to process streams or by thermal radiation to process heat exchanger surfaces offer many potential advantages.[41,42] Such blankets do not involve circulation of coolants and cannot develop coolant leaks, need not fulfil any structural role, have no moving parts, are simple, and can be inexpensive and easily repaired or replaced, can be subjected to any hydrostatic pressure level that is convenient, and a combination of a vacuum space surrounding the blanket and ceramic heat exchangers constitutes an excellent tritium barrier.

Resistance to radiation damage and minimization of thermal stresses are important in determining service lines of solid radiator blankets. Essentially no radiation damage information has been published for solid lithium containing compounds, but considerable information is available for such non-breeding materials as graphites, amorphous carbon, and some ceramics, although not for 14-MeV neutron irradiation. The voluminous literature [13-20,24,26] on swelling and changes in such properties as thermal conductivity, tensile strength, etc., in graphite, amorphous carbon, and selected ceramics subjected to neutron irradiation suggest that satisfactory service lives may be possible if ordinary radiation damage is the limiting phenomenon, especially when the fact that such blankets need not be completely constrained geometrically and must only support themselves is realized.

Thermal stress considerations could be especially important for inertial confinement radiating blanket concepts because of their short pulse operation, whereas magnetic confinement reactors are more nearly steady-state, although duty cycles may be relatively short, e.g., only a few tens to a few hundreds of seconds, for some concepts, and thermal stress can still poses problems for this class of fusion reactors. Careful design and limitations on power density can greatly aid in eliminating thermal stress problems. Segmenting can be used to reduce thermal stresses due to time-averaged temperature gradients, while holding time-averaged thermal power densities to moderate levels, e.g., of the order of a few to a few tens of W/cm^3, can keep maximum pulsed temperature rises to a few to a few tens of degrees centigrade. We do not know if deleterious synergistic combinations of thermal shock and radiation damage will cause problems. Only extensive experimental work can provide answers to some of these important questions.

Radiator blankets with sacrificial liner first walls of the same material would be particularly simple. The maximum temperature would be located at the inner surfaces and would have to be maintained low enough that rates of material loss by evaporation and sputtering were acceptable. If a cooled first wall were used, then the maximum temperature within the blanket would be somewhere in its interior and could perhaps be higher than the maximum temperature in the first case, provided that structural integrity were not affected and surface temperatures were not so high that evaporation rates became unacceptably large.

It is also possible that some process streams might be compatible with the radiator blanket material or with a suitable protective layer on the blanket outer surface. In such cases, heat exchangers could perhaps be eliminated, and convection and thermal radiation directly to process streams used.

The inclusion of a replaceable outer layer which consists of an efficient absorber of slowed-down neutrons, e.g., boron-10 carbide, would, in general, be necessary for minimization of neutron leakage from the blanket which would irradiate process streams. Gamma leakage would not cause significant problems, because it would simply represent a mechanism parallel to thermal radiation for removal of fusion energy from the blanket.

We have briefly investigated[44] the use of a nonbreeding graphite radiator blanket with a $^{10}B_4C$ outer neutron absorber and a sacrificial graphite liner for direct process heat supply and for coupling with thermionic devices for DC electric power generation for electrochemical processes with heat rejection to process streams. Calculations which assumed thermal radiation to a process stream at 2000K (3140F) and large heat fluxes indicated that inner surface temperatures would approach the one-atmosphere sublimation temperature of carbon.

However, surface temperatures of only 1750 to 2000K would give thermal radiation fluxes adequate for many purposes, albeit with blankets of greater inner and outer radii, and lower power densities and greater blanket size would result in smaller temperature gradients within the blanket. Also, cooled insulated inner walls could be used to prevent evaporation from the inner surface of such a blanket. Thus, there does appear to be a window in parameter space for a workable design for such a blanket for supply of high-temperature process heat.

Because the thermal radiator concept is so attractive, a discussion of the potential for discovery of a high-temperature lithium-containing compound which would permit tritium breeding ratios near or greater than one to be achieved seems warranted. Table IV contains a list of lithium compounds or alloys that have received some consideration for use in fusion reactor blankets and some relevant characteristics of these substances. The possibility of using these materials as low-tritium inventory breeding blanket materials in electric power fusion reactors which operate at temperatures substantially lower than those presently under consideration is presently being studied.[48,60-64] The information presented in this table, and in subsequent discussion, indicates that probably none of these substances have sufficiently high service temperatures for use in what we have designated as the high-temperature regime. Worse yet, some of the information in the table which was altered and added to only slightly from the original [58]in presenting it here, is no longer completely valid because of new, albeit still very incomplete data. These discrepancies illustrate the scarcity of data concerning lithium-containing solids, much of which is summarized in a recent review of binary lithium-containing systems.[65] For example, the listed compound Li_4Sn is now recognized to be $Li_{22}Sn_5$ on the basis of structural analysis and, as we shall see, the melting point for $LiAlO_2$ listed in handbooks appears to be in error. However, because such materials are being seriously considered for a number of fusion reactor conceptual designs, this situation will, of necessity, no doubt be remedied in the near future.

One of the columns of Table IV indicates whether or not there are potential sintering problems with the various lithium-containing solids. Sintering can be a problem if the solid lithium-containing substance is to be used in powder, pellet, or granular form, but might be beneficial for repair of thermal stress- or radiation-induced cracking of more massive breeding blanket elements such as would be desirable for a thermally-radiating blanket. The air-reaction information in another column is important in giving an indication as to whether or not special techniques, say, involving inert cover gases, will be required for fabrication of blanket elements from such materials.

The tritium breeding capacity of a given quantity of lithium-containing material which contains a specified total amount of lithium depends primarily on four factors:

- lithium atom density in the material relative to atom densities of other elements;

- neutron interaction characteristics of other elements contained in the material which competitively absorb neutrons or release additional neutrons through (n,2n) reactions;
- neutron energy spectra changes resulting from passage of neutrons through such materials, for, as may be recalled, one of the two tritium breeding reactions occurs with high probability only at low neutron energies, and the other only at high energies, while neutron absorption and (n,2n) reactions for other elements usually have larger cross sections at low and high neutron energies respectively, and
- differences in first wall, blanket containment, shielding, coolant loop, etc., structural requirements resulting from different physicochemical characteristics of the breeding material which impacts on the number and energies of fusion neutrons which interact with it.

The first factor also determines in part how thick the breeding zone of a solid thermally-radiating tritium breeding fusion reactor blanket has to be to achieve a specified tritium breeding ratio. It is not essential that such a blanket need consist entirely of lithium-containing material. To the contrary, as indicated in prior discussion of radiator blankets, this would in general not be the case. However, the thickness of the tritium-breeding zone could have an impact on total blanket cost. Sample number densities, a function of the number of lithium atoms per molecule of the material, the mass density of the material, and the total molecular weight of the material, include approximately 0.03248 x 10^{24} atoms/cm^3 for liquid lithium, 0.05708 x 10^{24} atoms/cm^3 for Li_2O, 0.03670 x 10^{24} atoms/cm^3 for Li_2C_2, and 0.04648 x 10^{24} atoms/cm^3 for Li_3N at room temperature.[62] These are all materials which permit tritium breeding ratios greater than one to be achieved without the use of neutron multipliers.

There are a number of important considerations, other than those mentioned previously, which include attainable tritium breeding ratio, resistance to thermal shock and ordinary radiation damage, etc., which bear on selection of a solid fusion reactor tritium breeding blanket material. For example, the consumption of lithium by the tritium breeding reactions can result in significant changes in physical and chemical properties of the lithium-containing solid, including lower melting points, production of volatile species, decrease in breeding ratio, etc.

The following examples illustrate some of these considerations.[62,63] Among the higher-melting-point solid lithium-containing materials which have been suggested as having potential for fusion reactor tritium-breeding blanket applications, lithium compounds of aluminum oxide and silicon dioxide seem to be practical to prepare reasonbly pure on a large scale. The orthosilicate Li_4SiO_4 has a high lithium atom density, but melts by a peritectic reaction at about 1528K (2290F), with rapid vaporization of the Li_2O at this temperature, unless the orthosilicate is canned. Also, as the lithium in the orthosilicate is consumed in producing tritium, a liquid of composition between the orthosilicate and the metasilicate, Li_2SiO_2, will form. The eutectic temperature is only about 1301K (1882F), so that the true service temperature limit of the orthosilicate is probably less than about 1300K.

The Li_2O-Al_2O_3 system apparently can form only two compounds: $LiAlO_2$ and $LiAl_5O_8$. The melting point of $LiAlO_2$ has been reported to lie between 1983 and 1973K (3109 and 3091F), substantially below the 2173 to 2293K listed in standard handbooks. A eutectic liquid is reported at approximately 1943K (or 3037F) between the compounds $LiAlO_2$ and $LiAl_5O_8$ and as lithium is consumed in breeding tritium the system melting point will decline. It has been suggested that this phenomenon plus excessive rates of evaporation of Li_2O at elevated temperature when th solid lithium-containing system is exposed directly to a helium coolant stream or a vacuum limits the maximum temperature of usefulness to less than 1673K (3551F). If this material is to be used in granular fixed, fluidized, or moving beds falling under the influence of gravity, then the possibility of sintering is projected to reduce the maximum service temperature by another 100K (180F).

Tritium can be bred in and recovered from lithium-containing solid breeding blanket materials by diffusion into a helium coolant, converted to tritiated water as it emerges from, e.g., an oxide solid and trapped, e.g., by adsorption on a number of possible adsorbents or trapped as T_2 by, e.g., metal hydrides. This approach to recovering tritium is a feature of a number of fusion reactor concepts of both confinement types.

However, there is some controversy and conflicting experimental data concerning the forms in which bred tritium might escape from solid lithium-containing compounds into helium coolant streams. For example, in some studies escape of tritium from $LiAlO_2$ mainly as tritiated water was claimed, while in others, recovery of the majority of escaping tritium as molecular tritium rather than T_2O was reported. On the basis of the nuclear reactions involved in breeding tritium and the stoichiometry and thermodynamics of tritiated water formation, it would appear that tritiated water should be the primary product. The conflicting experimental evidence suggests that bred tritium recovery systems for such materials either may have to possess the capability for removing tritium from helium coolant streams in both forms or reduce the tritiated water or oxidize the tritium. A similar situation may arise with any solid tritium-breeding material which contains elements capable of chemically reacting with tritium.

Much of the interest in solid lithium-containing substances as potential fusion reactor tritium-breeding blanket constituents results from pressure on developers of commercial electric power reactor concepts to reduce blanket tritium inventories in order to minimize inventory-related tritium inventories. Low tritium inventories, perhaps as much as two or more orders of magnitude lower than with liquid lithium, are apparently possible and bred tritium recovery is simple and efficient with a substantial technology base already extant. Long-lived neutron activation can be low or high, depending on the particular lithium containing material. Although little is known about the solubility and transport of tritium in the solid lithium-containing materials under consideration, consideration of the limited available experimental data and reasoning by analogy with the behavior of tritium is non-lithium containing ceramic materials, such as Al_2O_3 and SiO_2, indicates that if small tritium inventories are desired, then the lithium-containing solids must be relatively finely divided, whether used in static, moving, or fluidized beds.

This conclusion implies that tritium-breeding radiator blankets would contain relatively large tritium inventories, for a finely divided static bed from which thermal energy is removed solely from its boundaries, e.g., by thermal radiation, would not be efficient in a thermal sense. The effective conductivity in such a bed would be so low that neutron energy deposition would have to be very low to prevent excessively high maximum temperatures within the blanket. A few large tritium escape channels and/or metallic blanket materials which typically display larger tritium permeabilities than ceramics could ease the tritium inventory problem in thermally radiating, tritium-breeding blankets constructed of massive elements.

One possibility for high-temperature, tritium-breeding blanket materials that remain solid at temperatures approaching 2000K (3140°F) that has received very little consideration is lithium-containing intermetallic compounds. Published experimental data, compiled in a recent review,[65] on even binary, much less higher-order, lithium-containing intermetallic compounds is very sparse. However, some binary compounds of this type which are relatively rich in lithium, so that tritium breeding ratios approaching, if not exceeding, one can be achieved, and which have melting points much higher than the melting points of either constituent are known. For example, Li_3Bi has a reported melting point of ~1418K, while the melting points of pure Li and Bi are respectively 453.7K and 544.5K. The Li(M.P.453.7K) - Sn(M.P.505K) system encompasses several intermetallic compounds, of which $Li_{22}Sn_3$ (M.P.1038K) and Li_7Sn (M.P.1056K) are noteworthy. Some of these intermetallic compounds were mentioned earlier and, of course, do not possess sufficiently high service temperatures for use in high-temperature fusion reactor blankets as we have defined them.

As an example, lithium and bismuth have (n,2n) and thermal neutron capture cross sections of similar magnitude and lithium also has the relatively large tritium-producing reaction cross sections, suggesting that three-fourths or more of incident neutrons would interact with lithium in Li_3Bi rather than with bismuth. The possible existence of binary, or higher order, lithium-containing intermetallic compounds with melting points greater than 2000K and sufficiently rich in lithium that interesting tritium breeding ratios can be achieved with them is also suggested. Lithium-refractory metal intermetallic compounds would appear to represent possible candidates. Because such materials are potentially very attractive for fusion reactor blankets in electric power producing, and fusion-fission hybrid plants if suitable fertile material-lithium compounds can be discovered, as well as for process heat fusion reactors, further investigation of the possibility seems warranted.

OTHER FUSION REACTOR BLANKET CONCEPTS WHICH SHOW PROMISE FOR HIGH-TEMPERATURE PROCESS HEAT APPLICATIONS

A tritium breeding blanket concept,[48] which has potential for development for high-temperature process heat applications if lack of tritium breeding can be accepted, has been developed for both magnetic and inertial confinement fusion electric power reactors. This concept could also be a breeder if a solid compound rich in lithium with a higher melting point can be found. The concept involves, in its electric power reactor version, gravity flow of 100 to 200 micron Li_2O particles through the blanket with noncirculating neon gas at low pressure, i.e., about 10 torr, filling the spaces between the particles. The flow of Li_2O particles is divided into several layers by spacers and the flow in each layer is adjusted to match heat transport rate capacities to neutron energy deposition rates in that blanket zone so that exit temperatures from the various blanket zones can be made roughly equal.

The gravity flow Li_2O blanket represents an attempt to provide a high heat capacity per unit mass of cooling medium circulated together with considerable decoupling of wall and blanket medium operating conditions. Because the blanket containment wall areas are relatively modest and the wall heat transfer coefficients in such systems are also relatively small, the blanket medium and the walls can operate at significantly different temperatures without large heat transfer rates between them, at least under the moderate (1123K or 1561 F maximum blanket temperature) operating conditions projected for, e.g., the SOLASE laser fusion reactor concept.[46]

The 100 to 200 micron Li_2O_3 particle size range was selected on the basis of compromise between tritium diffusion, structural erosion, particle attrition, particle circulation, etc., considerations. A low void fraction is maintained in the blanket to minimize required blanket thicknesses and capital cost. The neon gas pressure is a compromise between blanket-wall heat transfer rate limitation requirements in the blanket and requirements for adequate heat transfer rates in primary heat exchangers. The claim is made that under SOLASE operating conditions an intermediate heat transfer loop is not required as a tritium barrier because the rates of tritium escape into the steam cycle can be maintained at acceptably low levels without one. Steam leaks into the Li_2O coolant are said to be not hazardous. Multistage Archimedes lift pumps in the cool leg of the coolant loop have been suggested as the best choice among a number of particle circulation schemes based on equipment maintenance, particle attrition, and circulation power considerations. However, it is important to note that the above materials, parameter choices, equipment selections, etc., may not be optimum for higher-temperature applications, either with nonbreeding or breeding solids. Also, some of the design ideas discussed in the section on lithium boiler blankets may be useful in devising a viable high-temperature, solid-particle, gravity-circulation bed concept.

The Battelle Columbus Laboratories, Inc., study of process heat applications of laser fusion energy mentioned previously, currently includes consideration of a number of process heat reactor concepts, but only one that falls into the high-temperature category as we have defined it.[66] Their high-temperature concept, to be described in a forthcoming report, is a non-breeder in which neutron energy is deposited in a stationary bed of alumina beads which is cooled by circulating helium. The blanket containment is also lined with alumina. First-wall protection is provided by graphite sacrificial liners, although most other methods which have been proposed could be used. Steel structural walls are water cooled. Maximum helium temperature is set at 3000F or 1922K, with steel-making as the intended application.

The simplicity of this concept is appealing, but the lack of tritium breeding capability is a serious drawback, and the concept is still in a relatively primitive state. Nevertheless, there seems to be little doubt that a blanket concept of this type can be made to work very much as intended, although the materials of construction now advocated may not be optimum. If a solid lithium-containing material which has a sufficiently high melting point, and which permits a tritium breeding ratio greater than or equal to one to be achieved can be discovered, this concept could be quite attractive because of its simplicity, even though all heat is transferred as sensible heat, and operating pressures are relatively high. Tritium recovery would be by adsorptive trapping of tritiated water either emerging from the solid breeding material or formed as a result of the presence of trace quantities of oxygen in the helium coolant stream from a side stream of the coolant stream.

Another recent study of applications of fusion energy, including thermal energy, which was mentioned previously, suggested that regenerative pebble-bed heater pairs with thermal energy transport by 30 atm helium could be used to achieve temperatures approaching 2800K (4580F). The beds would be alternatively heated by switching the helium primary reactor coolant stream from one bed to the other and alternatively cooled by switching a gas process stream from one bed to the other. However, design details, the tritium breeding issue, etc., were not discussed and operation in a regenerative mode will not be compatible with all potential applications of fusion-generated high-temperature process heat. Limitation of helium loss rates to the process stream and contamination of the helium coolant by process gases to acceptable levels appear to be serious obstacles to commercialization.

SURVEY OF RECENT RUSSIAN ACTIVITIES IN DEVELOPMENT OF HIGH-TEMPERATURE PROCESS HEAT APPLICATIONS OF FUSION ENERGY

The following is a brief summary of some activity in the USSR aimed at development of high-temperature process heat applications of fusion energy as described at a joint USA/USSR technical information exchange and workshop on alternative uses of fusion energy held in Moscow and Leningrad on November 20-27, 1978.[67] We will begin our summary with general impressions and then proceed to more detailed descriptions of high-temperature processes and fusion reactor blanket concepts which the Russians are investigating.

First we note that the percentage of energy end use in the form of electricity in the USSR is even lower than in the US by a modest amount. However, the percentage end use for transportation is also somewhat lower. Therefore, the Russians have a larger percentage requirement for stationary application thermal energy than we do in the United States. Thus, the Russians deem unwise the use of nuclear energy, fission or fusion, solely for electric power generation, and are therefore emphasizing end uses of nuclear energy as thermal energy in long-range planning. District heating using fission nuclear sources, not our concern here, is already being practiced in the USSR. The vast distances within the USSR, highly-nonuniform distribution of population, and equally highly-nonuniform distribution of energy resources which does not coincide with population distribution nonuniformity, which we have in the US to a lesser degree, compounds synthetic fuel production strategy assessment there.

The Russian workshop participants indicated that they believe that direct neutron irradiation of process streams will not be feasible for many industrial processes, in accordance with our earlier discussion, but agreed that elimination of intermediate heat exchangers, if possible, offered significant potential capital cost and operating efficiency advantages. They also suggested that fusion/fission hybrids for fissile fuel breeding in support of nonbreeding fission reactors represented the most significant near term application of fusion energy. Furthermore, because of their emphasis on thermal energy end uses of fusion energy, they are apparently quite interested in fusion/fission/process heat hybrids. Our studies here indicate that such fusion-driven fissile fuel breeders will probably have to be relatively power-rich to be commercially viable and hence could export significant quantities of either electrical or thermal energy, but the Russians suggested that the demand for fissile fuel might be so great as to force optimization of fusion/fission hybrids for fissile fuel production with severe limitations on export energy in the near future. They also felt that for direct thermal decomposition, and radiolysis as well, in which quenching of back reactions is important, pulsed fusion, i.e., inertial-confinement, fusion energy sources might have some advantages over both fission and magnetic-confinement fusion reactors. They also recognize that the question of simultaneous achievement of tritium breeding ratios of one or at least near one, and conversion of at least most fusion neutron energy into high-temperature process heat is of major importance for many process heat application of fusion energy. They appear to be somewhat pessimistic in this regard, but then they are not yet aware of our lithium boiler concept. Finally, the Russians agree that tokamak-type fusion reactor geometry can involve serious drawbacks for process heat applications.

The Russians are apparently keenly interested in magnetohydrodynamic topping of fusion reactor process heat and/or electric power cycles. Some of their MHD concepts exhaust gases at over 2000K, too high for conventional steam cycles for electric power generation, and therefore they are interested in intermediate temperature processes, e.g., for synthetic fuel production to fill the temperature gap between MHD exhaust and steam cycle conditions. One Russian high-temperature blanket concept for combined MHD-steam cycle power generation involves a completely-closed cycle using cesium injection into the primary helium coolant at 2000K for the MHD power generation, no coolant intermediate loop, and a complex regenerative heat exchange scheme with at least three stages in the main steam generator to provide a 50% conversion efficiency. Efficient removal of the cesium seeding from the helium prior to recirculation through the fusion reactor blanket is difficult, but also is considered essential if neutron activation of the primary coolant stream is to be maintained at levels which did not require exceptionally expensive shielding. This would also be a significant design consideration for process heat applications and many other blanket design considerations are common to be both process heat and electric power generation applications.

Their blanket concept involves, proceeding radially outward, a cooled, but relatively high-temperature (1100K or 1520F) coolant exit refractory metal first wall, high-temperature insulation, a solid $LiAlO_2$ tritium-breeding zone, a ZrC neutron-multiplier region with the zirconium being the active material, a second $LiAlO_2$ tritium-breeding zone, and finally a very-high-temperature graphite zone from which the helium, which passes in turn through these several blanket regions, exits at about 2000K (3140F). This elaborate blanket design achieves a calculated tritium breeding ratio of only about 0.93.

Coupling of another tokamak-type reactor concept with a three-step variant of the iron oxide cycle for thermochemical hydrogen production by water splitting which was discussed earlier, was also described at this meeting. Their iron oxide thermochemical cycle is defined by:

$$H_2O(g) + C(s) \xrightleftharpoons{1200K} H_2(g) + CO(g)$$

$$CO(g) + 2Fe_3O_4(s) \xrightleftharpoons{600K} C(s) + 3Fe_2O_3(s)$$

$$3Fe_2O_3(l) \xrightleftharpoons{1700\text{-}1900K} Fe_3O_4(l) + 1/2\ O_2(g)$$

and was selected by the Russians for investigation because of its simplicity and a claim of extensive knowledge of reaction kinetics resulting from steel-industry experience. The method proposed for achievement of the temperatures necessary to drive the last reaction step is unusual and makes the energetics of the cycle similar to that of a relatively inefficient electrothermochemical cycle. The significant fraction of the thermonuclear energy release which is deposited in lower-temperature structure and blanket regions is converted to electric power and part of it used to power a plasma torch to raise the approximately 1500K (2240F) helium exiting the fusion reactor blanket to about 1800K (2780F).

A Russian blanket concept for a tokamak-type fusion reactor, which displays certain similarities to our lithium boiler concept, to provide process heat to drive the two-step iron oxide thermochemical cycle for hydrogen production by water-splitting described previously was also discussed. In order to improve yields and kinetics, the high-temperature reaction to decompose Fe_3O_4 to FeO and oxygen is to be conducted at a very high 2500K (4040F). The primary blanket coolant is supposed to be a suspension of solid Li_2O particles in molten silicon (MP 1700K or 2600F). The primary means of heat transport is to be vaporization of silicon and its transport to and condensation in a heat exchanger which thermally radiates to the FeO/Fe_3O_4 process stream. Because the melting point of Li_2O is substantially less than the maximum thermochemical cycle temperature, some disproportionation of oxygen between silicon and lithium oxide with consequent intersolubility is expected, and the dissociation pressure of Li_2O is apparently greater than the vapor pressure of silicon in this temperature range, we believe that the concept as described in unworkable.

The FeO/Fe_3O_4 melt would then flow to an argon-cooled prilling tower into which the melt would be sprayed to form spherical pellets to be circulated through another reactor to accomplish the lower temperature water-splitting reaction. The role of the argon is heat recuperation to improve cycle efficiency.

The study which led to the reactor blanket concept described in the two preceding paragraphs, was judged by some participants to exhibit more overall balance in addressing key design issues than other Russian studies, although it was noted that many of these key design problems have not been successfully addressed. Overall thermochemical cycle efficiencies were computed to vary from 0.3 to 0.7 as the fractional completeness of the Fe_3O_4 reduction to give FeO varies from 0.4 to 0.9 and fractional neutron energy recovery as high-temperature heat varies from 0.7 to 0.85. Tritium breeding ratios greater than one were claimed for these large high-temperature thermal energy recovery fractions for combinations and thicknesses of blanket neutron multiplying regions, tritium breeding regions, and high-temperature zones which appear to be quite inadequate.

A novel Russian approach to fusion-driven combined direct-radiolysis/direct-thermal decomposition for synthetic fuel production was described at this workshop. This process was designed to take advantage of the special characteristics of inertial confinement fusion. Specifically, it is proposed that fuel pellets of 1000 MJ (~ 0.949 million BTU) yield be surrounded by an 0.35-m (~ 1.15 ft) thick, 1.1-m outer-radius (~ 3.61 ft), 8.7-tonne (~ 9.59 ton) spherical shell of solid carbon dioxide which would be radiolytically decomposed and raised to very high temperatures with further thermal decomposition to yield carbon monoxide and oxygen. Operation at 0.1 hertz (one microexplosion every ten seconds) was projected.

A similar concept for a laser fusion electric power reactor developed at Los Alamos Scientific Laboratory to a very primitive stage which involves fewer major design problems has not received much favorable attention. One obvious difficulty with this scheme is the problem of accomodating the relatively high repetition rates, i.e., opening up reactor cavities and inserting large premanufactured CO_2 ice-fuel pellet energy "capsules" every ten seconds with a single cavity or every hundred seconds with ten cavities, etc. The modest value of the energy release from such a large "pellet" per pellet is also discouraging. The problems would be eased considerably if higher, e.g., by an order or two in magnitude, per-pellet thermonuclear energy releases, as suggested for the Los Alamos Scientific Laboratory concept, could be obtained economically.

Combined direct radiolytic and thermal decomposition of CO_2, to be used in conjunction with the water gas shift reaction to produce hydrogen, was selected to minimize contamination of hydrogeneous product with bred and unburnt fuel tritium. Seeding of the CO_2 ice spheres with lithium carbonate to permit tritium breeding was proposed, but even with as much as 20 wt% $LiCO_3$ tritium breeding ratios of one could not be achieved. Then an inner 6-cm-thick shell of lead oxide to provide neutron multiplication sufficient for achievement of a tritium breeding ratio of one was added to the energy capsule concept. What happens to this lead oxide and the economics of its manufacture were not addressed. The separation of tritium from the reaction mixture at low concentrations is a major process design problem.

A peak process mixture temperature of about 4100K (6920F) and a peak process mixture pressure of approximately 30 atm was calculated for this latter energy capsule concept, assuming thermochemical equilibration. At these conditions the composition of the reaction mixture was calculated to be 21.8 mol% CO_2, 50.8 mol% CO, 15.9 mol% O_2, and 0.11 mol% O_2. The reaction mixture would be transported rapidly to a liquid CO_2 injection quench chamber to limit back reactions. The quenched gas mixture, now at 800K, would then be passed successively through cyclones for separation of condensed phases, a tritium separator whose characteristics were not specified, and a $CO/CO_2/O_2$ separator, followed by hydrogen production via the water gas shift reaction of CO wth H_2O and eventual recycle of CO_2. A large amount of power is required for CO_2 cooling and solidification and liquifaction and much of it is not recoverable in useful form. All in all, the lack of detailed examination of important engineering and economic questions and the extremely complex nature of the proposed synthetic fuel production scheme makes it difficult to accept it as a serious candidate for commercialization. In general, the Russian workers in the field of high-temperature process heat applications do not appear to have been able to devote much effort to their investigation and, as a result, their fusion reactor blanket concepts do not appear to be serious candidates for commercialization.

TABLE I

PRESENT PATTERN OF END-USE ENERGY CONSUMPTION IN THE UNITED STATES

Energy Form	End Use	% of Total
Electric Power	Industrial	14
	Residential and Commercial	8
	Other	2
Total		24
Fossil Fuels	Transportation	24
	Residential and Commercial Heating	20
	Low Temperature Process Heat	10
	High Temperature Process Heat	9
	Conversion to Other Fuels	7
	Non-Energy Uses	5
	Other	1
Total		76

TABLE II

POTENTIAL APPLICATIONS FOR FUSION GENERATED PROCESS HEAT

Application	Typical Application Size MW(th)	Typical Application Size 10^6 BTU/day	Typical Temperature Range K	Typical Temperature Range F
Steelmaking	3000	2.5×10^5	1700-1750	2600-2700
Inorganic Chemicals (including pulp and paper)	300	2.5×10^4	1100-1150	1500-1600
Petroleum Refining	500	4.0×10^4	800- 850	1000-1100
Petroleum Refining	500	4.0×10^4	700- 750	800- 900
Many	500	4.0×10^4	450- 500	350- 650

TABLE III

EQUILIBRIUM THERMAL DISSOCIATION OF WATER AND CARBON DIOXIDE

$H_2O(g) \rightarrow H_2(g) + 1/2\ O_2(g);\ CO_2(g) \rightarrow CO(g) + 1/2\ O_2(g)$

Temperature K	Temperature F	Pressure atm	Mole Fraction* Hydrogen	Mole Fraction* Carbon Monoxide	Fractional Dissociation** Water	Fractional Dissociation** Carbon Dioxide
2000	3140	0.01	0.025	0.062	0.025	0.064
		0.10	0.012	0.030	0.012	0.030
		1.00	0.006	0.014	0.006	0.014
		10.00	0.003	0.007	0.003	0.007
2500	4040	0.01	0.150	0.369	0.162	0.452
		0.10	0.080	0.223	0.083	0.251
		1.00	0.039	0.119	0.040	0.127
		10.00	0.019	0.060	0.019	0.062
3000	4940	0.01	0.395	0.606	0.492	0.869
		0.10	0.245	0.507	0.279	0.679
		1.00	0.134	0.362	0.144	0.442
		10.00	0.069	0.218	0.071	0.245
3200	5300	0.01	0.476	0.632	0.625	0.354
		0.10	0.325	0.571	0.388	0.587
		1.00	0.188	0.453	0.208	0.799
		10.00	0.098	0.301	0.103	0.942

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*Maximum value is 2/3.

**Maximum value is 1.

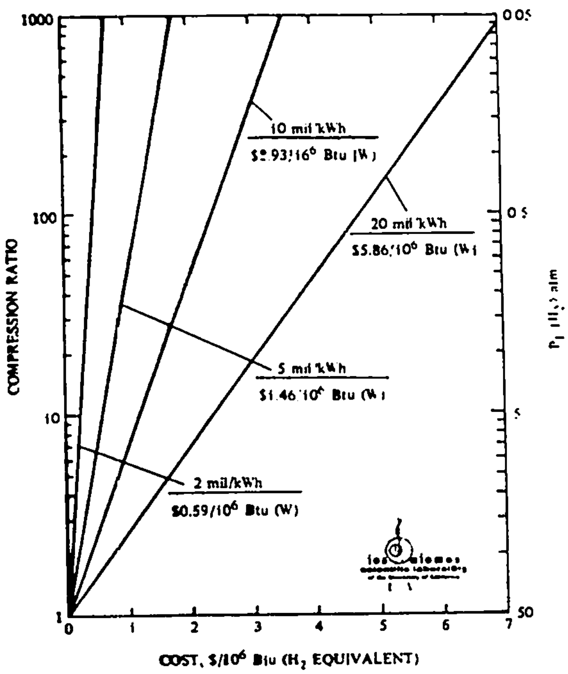

Figure 1.
Cost of compression of hydrogen as a function of compression ratio or initial pressure resulting in final pressure of 50 atm with cost of electric power or other energy source to drive compressors as a parameter.

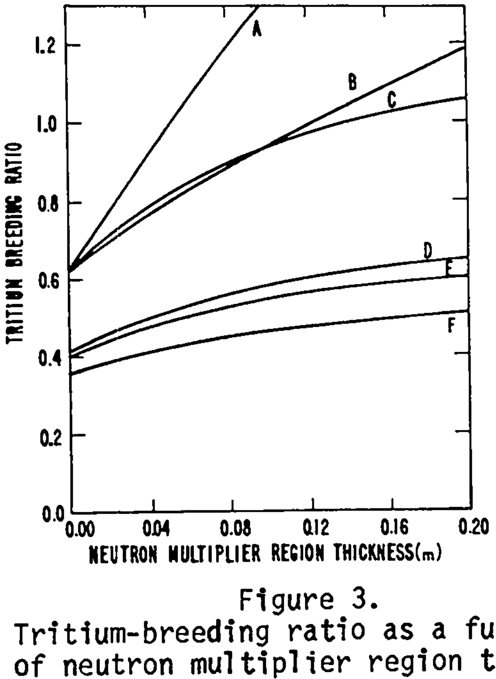

Figure 3.
Tritium-breeding ratio as a function of neutron multiplier region thickness (v/o = % by volume before mixing). A=Be, 20v/o void, 0.1 m Li; B=Li, 0.1 m Li; C=Pb, 0.1 m Li; D=Pb, 0.1 m 25 v/o Li, 75 v/o LiPb; E=Pb, 0.1 m 50 v/o Li, 50 v/o LiPb; F=Pb, 0.1 m 75 v/o Li, 25 v/o LiPb.

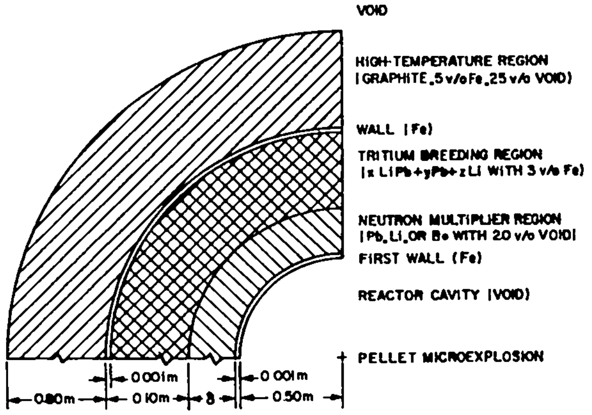

Figure 2.
Schematic of blanket mockup used for neutronics studies of tradeoff between tritium breeding and high-temperature process heat generation.

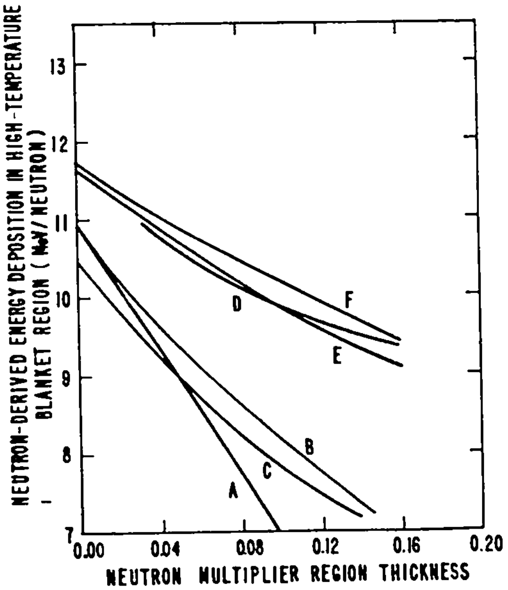

Figure 4.
Fraction of neutron-derived energy deposition converted to high-temperature process heat as a function of neutron multiplier region thickness (v/o = % by volume before mixing). A=Be, 20 v/o void, 0.1 m Li; B=Li, 0.1 m Li; C=Pb, 0.1 m Li; D=Pb, 0.1 m 25 v/o Li, 75 v/o LiPb; E=Pb, 0.1 m 50 v/o Li, 50 v/o LiPb; F=Pb, 0.1 m 75 v/o Li, 25 v/o LiPb.

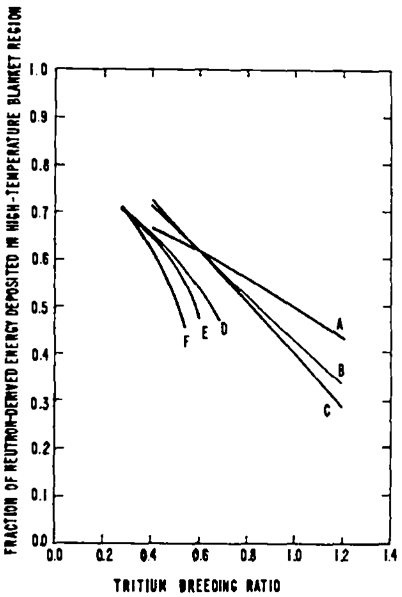

Figure 5.

Fraction of neutron-derived fusion energy release converted to high-temperature heat as a function of tritium breeding ratio (v/o = % by volume before mixing). A=Be, 20 v/o void, 0.1 m Li; B=Li, 0.1 m Li; C=Pb, 0.1 m Li; D=Pb, 0.1 m 25 v/o Li, 75 v/o LiPb; E=Pb, 0.1 m 50 v/o Li, 50 v/o LiPb; F=Pb, 0.1 m 75 v/o Li, 25 v/o LiPb.

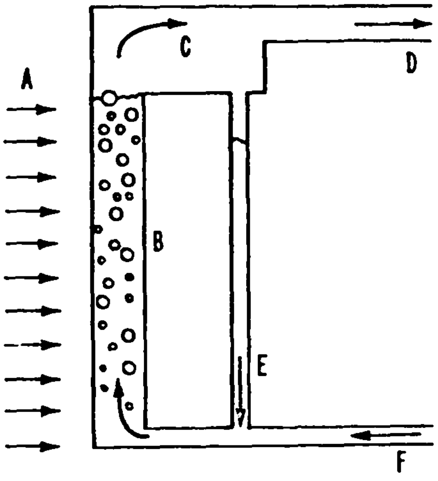

Figure 7.

Lithium-boiler pressure-vessel concept, A, fusion energy source; B, lithium two-phase mixture in pressure tubes; C, vapor-liquid disengagement region; D, Saturated lithium vapor to process- or power-cycle heat-exchanger tubes; E, disengaged liquid lithium recycle; F, saturated or subcooled condensed liquid lithium.

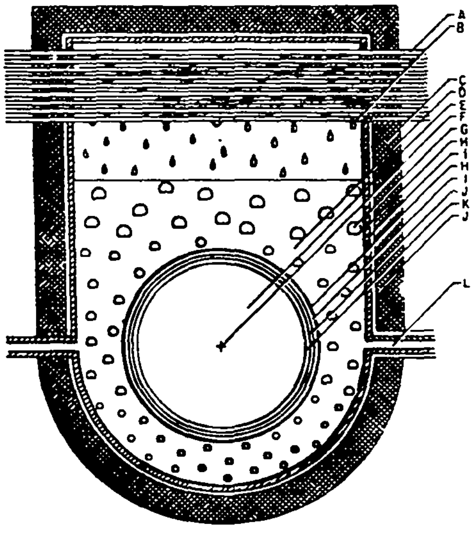

Figure 6.

Lithium-boiler pressure-vessel concept. A, process or power-cycle heat-exchanger tubes with tritium escape barrier and/or tritium recovery system; B, refluxing liquid lithium; C, shielding, other reactor structure, and/or reactor subsystems; D, liquid lithium blanket at 1500 to 2000K, 0.4 to 15 atm; E, reactor cavity; F, fusion energy source; G, lithium vapor bubble; H, liquid-lithium-resistant refractory-metal (e.g., a gettered molybdenum alloy such as TZM); I, structural wall (e.g., a ceramic at temperature, graphite at temperature, or a cooled-metal wall); J, thermal insulation (e.g., evacuated multiple metal foils, graphite cloth, or conventional high-temperature insulation); K, cavity first wall; L, duct to lithium purification and tritium recovery (if required) subsystem.

TABLE IV

CHARACTERISTICS OF SOME SOLID LITHIUM-CONTAINING MATERIALS WHICH HAVE BEEN CONSIDERED AS POTENTIAL FUSION REACTOR TRITIUM-BREEDING BLANKET MATERIALS UNDER PROJECTED ELECTRIC POWER REACTOR OPERATING CONDITIONS

Lithium Compound or Alloy	Melting Point		Tritium Solubility under Projected Operating Conditions?	Neutron Multiplier Needed for Tritium Breeding Ratio of One?	Low Residual Radioactivity?	Reacts with Air During Fabrication?	Sintering Problems Under Projected Operating Conditions?	Chemically Stable Under Projected Operating Conditions?	Resource Limitations?
	K	F							
LiAl	991	1324	very low	yes	yes	slowly	maybe	yes	no
$LiAlO_2$	2173-2273	3451-3631	very low	yes	yes	no	probably not	yes	no
Li_2SiO_3	1477	2199	very low	yes	yes	no	probably not	yes	no
$Li_2Be_2O_3$	1423	2101	unknown	no	yes	no	unknown	yes	Be
Li_7Pb_2	999	1338	very low	no	yes	slowly	unknown	yes	no
Li_3Bi	1418	2092	unknown	no	some Po^{210}	probably slowly	unknown	yes	Bi
Li_4Si	908	1174	unknown	no	yes	probably slowly	unknown	probably	no
Li_3N	1073	1471	unknown	no	yes	probably slowly	unknown	probably requires N_2 pressurization	no
Li_4Sn	1038	1408	unknown	no	no	probably slowly	unknown	probably	no
$Li_2B_4O_7$	1703	2605	unknown	yes	yes	probably not	unknown	yes	requires separated ^{11}B
Li_2O	1973	3091	very low	no	yes	with CO_2 in air	probably not	yes	no

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APPENDIX B

FUSION-DRIVEN PRODUCTION OF SYNTHETIC FUELS BY DIRECT RADIOLYSIS

INTRODUCTION

Investigations of the potential for utilization of fission reactor radiations, e.g., gamma photons and neutrons, but with emphasis on fission fragments which carry off the bulk of the energy released by fission reactions, for direct radiolytic production of large volume chemicals, e.g., synthetic fuels by decomposition of water to hydrogen and oxygen and carbon dioxide to carbon monoxide and oxygen, fixed nitrogen from air, ozone, etc., have been performed over the past two decades.[1-6] More recently, there has been interest in the use of fusion reactor radiations for direct radiolysis, especially for synthetic fuel production.[7-10]

As we shall discuss in more detail below, interest in the use of fission reactor radiations for direct radiolysis has largely vanished because of apparently insurmountable obstacles which prevent simultaneous achievement of high conversions of radiant energy to stored chemical energy of desired products of radiolysis and acceptable levels of product contamination at affordable production costs. The characteristics of fusion reactor radiations appear to be much more suitable for direct radiolytic production of, e.g., synthetic fuels from water and/or carbon dioxide, but many of the same obstacles to commercialization of such processes with fission energy sources must also be faced for commercialization of direct radiolytic chemical production using fusion energy sources, albeit to a lesser degree. We will attempt here to present a frank discussion of these obstacles to commercialization, but will also provide information on all grounds for optimism of which we are aware.

As of now, the direct radiolysis process which appears to have the greatest chance for commercialization assuming that there is sufficient demand for the product, is ozone (O_3) production from ordinary oxygen (O_2) because of inherently low efficiencies in energy utilization of present primary production methods, i.e., electric discharge.[1-3, 10-20] The primary application of ozone is drinking water and waste water treatment.[21-23] However, we will concentrate here on the potential use of fusion reactor radiations for commercial processes involving direct radiolysis as the primary production step or as part of more complex processes.

Much of the discussion in the section on high-temperature process heat applications of fusion reactors concerned with mechanical-thermal-hydraulic design of high-temperature process heat fusion reactor blankets is applicable to design of fusion reactor blankets for direct radiolysis applications, especially when radiolysis is intended to top other energy conversion cycles, be a high-temperature step in a combined radiolysis-thermochemical cycle, etc. We will, however, mention a number of additional considerations where appropriate.

Essentially all the discussion on the problems inherent in direct neutron irradiation of process streams which appears in the section on high-temperature process heat applications of fusion reactors is relevant to direct radiolysis applications of fusion reactors. These problems are, of course, some of the most serious obstacles to commercialization of direct radiolysis applications of fusion reactor radiations.

The tritium breeding issues, as discussed in the section on high-temperature process heat applications of fusion energy, are essentially the same for proposed direct radiolysis applications of fusion neutron energy as they are for high-temperature process heat applications, with one exception. It is very difficult to see how fusion reactors used for direct radiolysis which apply more than 50% of the fusion neutron energy to radiolysis can ever be more than substantial subbreeders at best, and it is not at all clear that nonbreeders supported by breeder cavities would not be more nearly cost effective. This opinion is based on the reasonably thoroughly tested assumption that combinations of:

- neutron multiplying materials,
- only modest reductions in neutron energy used for radiolysis,
- the use of efficient low neutron energy tritium breeding materials such as liquid lithium enriched in ^{6}Li, and
- radiolysis process streams with low neutron absorption cross sections and/or high (n,2n) reaction cross sections,

cannot be used to obtain simultaneously and economically tritium breeding ratios greater than or equal to one and deposition of a majority of fusion neutron energy in a radiolysis process stream.

CHARACTERIZATION OF FUSION REACTORS AS SOURCES OF RADIATION FOR DIRECT RADIOLYSIS

The mixture of radiant energy forms characteristic of fusion reactors which burn deuterium and tritium is unusual, consisting of:

- photons (x- and gamma-radiation);
- energetic ions (plasma consisting of fusion reaction products, i.e., alpha particles, unburnt fuel, i.e., deuterium and tritium, and, in the case of inertial confinement fusion, pellet debris); and
- high energy neutrons.

The fraction of the total thermonuclear energy release and the energy spectrum for each type of radiation can be controlled to some extent through seeding of magnetically-confined plasmas with suitable materials or pellet design for inertial confinement fusion.

The photon energy spectra resulting from deuterium-tritium burn in fusion reactors may involve both essentially blackbody or equilibrium thermal components and highly nonequilibrium components. Nonequilibrium photon energies in the several MeV range and equivalent blackbody temperatures from hundreds of keV down to a few hundred eV are possible, depending on magnetic-confinement fusion plasma or inertial-confinement pellet characteristics, with large masses of pellet structure composed of heavy elements favoring lower photon energies and less total energy release in the form of photons. Photon energy typically represents only a few percent of total fusion reactor energy release.

Fusion plasma ions can have kinetic energies ranging up to a few MeV, but are usually slowed to the few-tens-of-keV range by inertial confinement fusion pellet structure. Inertial-confinement fusion pellet debris ions typically have similar kinetic energies, with more structure leading to more ions of lower energy. In general, particles possessing such kinetic energies cannot penetrate solids or liquids to an extent which would permit their utilization outside fusion cavities. The primary fusion energy carried by such ions is typically about 20 to 30%.

Fusion neutrons:

- are born with 14.1-MeV kinetic energies, typically represent about 80% of total fusion reactor energy release when exoergic interactions with reactor materials are included;
- lose very little of their kinetic energy in escaping from magnetic-confinement fusion reactor plasmas, but can deposit substantial fractions of their kinetic energy in, be absorbed by, or undergo other nuclear interactions such as (n,2n) reactions with structural materials of inertial confinement pellets with massive cases, with the neutron energy being converted to photon and ion energy and moderately reducing the percentage of total fusion energy release carried by energetic neutrons;
- suffer only moderate fractional absorption, kinetic energy loss, and other nuclear interactions in penetrating moderate thicknesses of reactor structure to enter blankets surrounding fusion reactor cavities; and
- may release significant additional energy (a few MeV/neutron) through exoergic nuclear interactions with blanket materials.

The neutron energy deposited in fusion reactor blankets can generally be used under conditions which are largely independent of reactor cavity phenomenological constraints. The availability of this major fraction of total energy release outside the reactor cavity is an important feature of fusion reactors.

Radiolytic decomposition of reactants could be accomplished directly with any of the primary energy forms released by the fusion-reaction. In addition, substantial increases in x- or γ-radiation, or production of ultraviolet radiation, could be accomplished by including suitable materials in magnetic-confinement fusion plasmas or inertial confinement fusion pellets. Radiolysis using ultraviolet radiation produced in fusion reactors is considered impractical because:

- efficiencies of conversion of primary fusion energy forms to ultraviolet radiation are low (estimated to be less than 10% for Tokamak reactors);
- engineering concepts for efficient extraction of ultraviolet energy from fusion reactor cavities, where it is, of necessity, produced, for utilization outside reactor cavities are lacking, owing to the low penetrating power of ultraviolet radiation in typical reactor materials of construction; and

- utilization of ultraviolet radiation within fusion reactor cavities is subject to the severe constraints imposed by the conditions required for thermonuclear reaction, e.g., the very low plasma densities required in magnetic confinement fusion reactors, very low vapor densities required in laser fusion reactors for laser beam transmission, and moderate densities reqired in electron-beam or ion beam fusion reactor cavities for beam propagation.

With regard to the last point, typical magnetic confinement plasma densities are of the order of 10^{14} ions/cm^3 or about six orders of magnitude less than sea level atmospheric density. Introduction of significant quantities of synthetic fuel process reagents into such dilute plasmas would interfere with the processes which are responsible for thermonuclear burn, i.e., the plasma would be quenched. Residual gas pressures remaining in laser fusion reactor cavities following clearing of the cavities of pellet microexplosion debris which will permit laser beam transmission to pellets without unacceptable energy loss, beam defocusing, etc., are estimated to be 0.1 torr or less Electron beam and ion beam fusion relies on cavity gas pressures of the order of one atmosphere for beam propagation. In addition, because fractional burnups of tritium in both magnetic and inertial confinement fusion reactors are projected to be modest, unacceptable contamination with tritium of synthetic fuel process reactants and products introduced into fusion reactor cavities in order to make direct use of fusion energy forms which are of low penetrating power apparently cannot be avoided.

These same considerations preclude efficient use of primary x-ray and plasma and/or pellet debris ion energy for radiolysis, owing to their lack of penetrating power. Thus the only practical means of utilizing these forms of energy which are trapped in reactor cavities are conversion to more penetrating radiation, e.g., gamma radiation, conversion to thermal energy, with or without subsequent conversion to electric power, or direct conversion to electric power.

More penetrating gamma radiation produced by inelastic scattering reactions in fusion plasmas, with pellet constituents or reactor structure, or by

(n,γ) reactions with structural or blanket materials streams throughout reactor and blanket and is available for utilization free of cavity constraints. Therefore, we will consider further only utilization of gamma and neutron energy from fusion reactors for radiolytic production of chemicals.

Neutron energy can be transferred to reactants in fusion reactor blankets through scattering interactions, i.e., collisions with atoms or molecules, which typically ionize or otherwise excite, including chemical bond disruption which is the essence of radiolytic decomposition, and impart kinetic energy to the atoms or molecules struck by the neutrons. The atoms or molecules undergoing neutron bombardment can in turn strike other atoms or molecules, resulting in further ionization, distribution of kinetic energy, disruption of chemical bonds, etc. Neutron interactions, $(n,2n)$ reactions, (n,γ) reactions, etc., also result in transfers of energy from incident fusion neutrons to the constituents of fusion reactor blankets. These processes proceed with continual degradation of energy quality until all incident neutron energy is completely thermalized or converted into stored chemical energy of the products of radiolysis.

Gamma-ray energy is initially absorbed in fusion reactor blanket materials, primarily through such electron production or excitation processes as pair production, Compton scattering, and the photoelectric effect. Energetic electrons and ionzied atoms or molecules resulting from these processes can ionize and otherwise excite other atoms and molecules and induce chemical reactions, with continual degradation of energy quality unless storage in the form of chemical bond energy of radiolytic products occurs.

Neutron interaction cross sections are large enough that the majority of incident neutron energy can be absorbed in fusion reactor blankets containing materials at normal liquid and solid densities of only a few meters thickness. Densities substantially lower, i.e., corresponding to dilute gases, require blanket thicknesses of many meters, leading to prohibitive blanket dimensions for such radiolysis mixtures. Gamma ray interaction cross sections, on the other hand, for the low atomic weight substances which would be used in most radiolytic process for synthetic fuel production are so small that even with condensed phases in fusion reactor blankets, large blanket thicknesses may be required for capture of significant fractions of the gamma energy in the materials to be radiolytically processed. Therefore, we will henceforth accord gamma radiation only minor consideration as a form of fusion energy release important for radiolytic production of synthetic fuels.

FUNDAMENTALS OF RADIOLYSIS

The essence of a chemical reaction is the breaking and forming of chemical bonds by energy supplied in certain forms to molecules in amounts equivalent to binding energies. For quasistatic thermally-driven processes, which are governed by the principles of equilibrium thermodynamics, there are certain thermodynamic restrictions on yield, products and product concentrations, reaction rates, etc. However, chemical reactions can also be induced by ionizing radiation with photon or particle energies of the order of molecular electronic binding energies (a few electron volts) and above, through disruption of chemical bonds with few of the restrictions associated with thermal equilibrium, i.e., independently of equilibrium energy level distributions or temperature. Thus the difference between radiation-driven and thermally-induced processes is similar to the difference between processes in which free energy is supplied in the form of work, e.g., mechanically or by electricity, rather than thermally. In particular, systems subjected to irradiation can be driven away from thermochemical equilibria. Furthermore, radiant energy deposition typically occurs in the form of energy "spikes" along primary and secondary particle and/or photon tracks in which initial energy densities are so high that corresponding temperatures, if thermal equilibrium could be instantaneously attained, would be very high. Of course, conditions within such a spike are far from equilibrium, but the effect is to induce processes which would not spontaneously occur under conditions of bulk material thermal equilibrium to proceed at rates which far exceed those observed under typical thermally-driven reaction conditions.

It is important to note, however, that under such conditions, the rates of reverse reactions are also greatly increased within the spikes and can proceed at ordinary thermochemical rates in the bulk of the irradiated materials which are outside such spikes unless the radiation field intensity is so great that substantial overlap of particle and/or photon tracks occurs. Furthermore, rapid transfer of energy and diffusion of reactants and products from the spikes also affect which species are formed and in what amounts, and results in rapid degradation of quality of the energy originally deposited in the spikes. The rapid reactions, including back reactions, which are induced by the extreme conditions in the spikes are rapidly quenched by energy loss to bulk material whose temperature may be maintained at relatively modest levels. The courses of competing reactions can be affected differently by the complex concentration, pressure, and temperature histories of irradiated material in and near such spikes. Track overlapping can introduce further complexity.

Thus, the physics and chemistry of radiolysis reactions are complex and the simple models of radiolytic processes whose behaviors have been analyzed have achieved only limited success in predicting radiolytic yields and steady-state concentrations.[24-41] Therefore, accurate assessment of the potential of radiolytic processes for synthetic fuel production must be based on experimental data. Unfortunately, while radiolysis has been studied extensively for several decades, these studies have mostly involved irradiation with heavy ions (alpha particles, fission fragments, etc.), photons (x- and gamma-radiation), and electrons (from accelerators and beta particles). There is very little experimental data concerned with radiolysis by neutrons, and apparently none for radiolysis by 14.1-MeV fusion neutrons. However, because, as previously noted, the primary mechanism of neutron kinetic energy deposition in fusion reactors involves production of energetic heavy ions, G-factors, to be discussed below, for neutron-driven radiolytic processes are expected to be similar to those for heavy ion radiolysis of the same systems.

We hasten to note that there is some experimental evidence and the opinions of some investigators[9] which tend to contradict this opinion, which, however, is shared by other workers in this field. Certainly that portion of fusion reactor energy release resulting from (n,γ) reactions will have a significant gamma radiation component and LET effects, to be discussed shortly, may be quite different for such radiation, depending on particular radiolysis conditions and systems.

RADIOLYTIC YIELDS AND ENERGY EFFICIENCY OF RADIOLYTIC PROCESSES

Radiolytic yields are usually expressed in terms of G-values for each substance produced by the various radiolytic reactions induced by irradiation. The G-value for a particular product is defined as the number of atoms or molecules of that product formed per 100 eV of radiation absorbed by the reaction mixture. Efficiency of conversion of absorbed radiation energy to stored chemical energy of a particular radiolysis product destined to be used as a fuel which is burned in air or oxygen, and which is produced by radiolytic decomposition of the combustion products for that substance, is defined as the ratio of the standard heat of combustion for a unit of that product to the total radiant energy which must be absorbed in the reaction mixture to produce a unit of that product. More generally, the efficiency of radiolytic conversion is defined as the standard heat of reaction for the reaction which produces that product from the reactants, divided by the amount of radiant energy which must be absorbed in the reaction mixture to effect the reaction. Perhaps a better way to express radiolytic efficiency would be to use the actual temperature - and reaction mixture composition - dependent heat of reaction corresponding to actual radiolysis conditions, provided composition dependence of heats of reaction, which are usually not available, were available. Temperature dependences of heats of reaction are frequently relatively modest, e.g., for water vapor, the heat of formation varies by only about 4% between room temperature and 2000K.

Conversion of radiolytic G-values to a fractional radiolytic efficiency using the standard definition involving the standard heat of reaction ΔH_R^o is simple: $\eta = 4.2882 \times 10^{-4}\ G\Delta H_R^o$. Efficiencies for some radiolysis reactions of interest as functions of G-values are displayed in Fig. 1 for orientation purposes. Clearly, the smaller the heat of reaction, the more of the product of interest which must be formed per unit of radiant energy deposited in the reaction mixture to achieve a specified radiolysis efficiency. Here we are of course concerned with endothermic reactions, for which heats of reaction are more positive than free energies of reactions, and are basically interested in converting radiation energy into stored chemical energy of synthetic fuels, the magnitude of which is represented at least approximately by the heat of reaction used in computing the radiolysis efficiency. There are, however, many reactions of commercial interest which are exothermic and are merely initiated or induced to occur more rapidly by means of irradiation and a few for which free energies of reaction are positive while heats of reaction are negative and radiation is used to supply free energy of reaction rather than heat of reaction. For these cases, the foregoing definition of radiolytic efficiency is, of course, meaningless.

Radiolytic G-values can also be defined for the destruction of specific reactants as well, with the definition being strictly analogous to that for products of radiolysis. A G-value for a particular product P of radiolysis is often denoted by G(P) and for a particular reactant R by G(-R), where P and R can be molecules, free radicals, atoms, ions, etc.

LIMITATIONS OF G-VALUES AND PRODUCT CONCENTRATIONS BY BACK REACTIONS INDUCED BY RADIATION

An important property of many, but not all, radiolytic processes is that as radiolysis product concentrations build up, back reactions to form the initial reactants become important and hence only limited product concentrations can be achieved. Because the types of radiolytic processes under consideration involve driving the systems being irradiated far from thermodynamic equilibrium, the back reactions are not those characteristic of a system approaching thermodynamic equilibrium and which, together with the forward reaction, determine the equilibrium state. Instead, the limiting product concentrations correspond to a steady state in which the rate of the forward reaction, which would not occur spontaneously because the free energy change for the reaction is positive, driven by the irradiation of high concentrations of reactants, equals the rate for the backward reaction. The backward reaction can occur spontaneously because the free energy change is negative, but only very slowly in the absence of irradiation because product concentrations are small and/or intrinsic nonradiolytic kinetics are slow, but which is induced to occur at higher rates by the irradiation responsible for the radiolytic reaction of interest. The steady state product concentrations will, in general, be functions of the type and rate of irradiation, temperature, total pressure, phases present, phase compositions, etc.

These back reactions result in effective G-values, which may be much lower than intrinsic G-factors which correspond to negligible back reaction and can often be estimated from initial rates of production of desired product, i.e., before significant product concentrations build up. In general, if low product concentrations must be tolerated to ensure acceptable G-values and the radiolysis system is single-phase, separation of product and reactant for recycle of reactant may be difficult to accomplish at acceptable cost.

There are exceptions to the general rule. For example, the radiolytic reaction for fixation of nitrogen from air as NO_2 can apparently be driven essentially to completion under the proper circumstances. More importantly, for radiolytic processes in which separation of radiolysis products from reactants can be readily and promptly accomplished, no such restriction on G-values due to back reactions need apply. An example of such a process would be one in which a solid or liquid is being radiolytically decomposed and the products of radiolysis are gaseous, not highly soluble in the reactants, and can readily escape from the reactant phase and be removed from the reactor. Therefore, we feel that successful radiolysis processes for synthetic fuel production are more likely to be heterogeneous in nature than to involve homogeneous reactions.

An example of reduction of effective single-phase radiolysis G-values by back reactions as product concentrations increase is the experimental values of Table I for radiolysis of O_2 with ^{60}Co gamma radiation to produce O_3 at 195K and 460 torr total pressure.[11] Experimental results for fission fragment and alpha particle radiolysis of aqueous 0.4N H_2SO_4 solutions with various additives, which indicated that the total yield of H_2 gas is proportional to total irradiation time with steady irradiation, illustrate a case for which back reactions induced by the irradiation are apparently not significant because of separation of radiolytic products from reactants due to the heterogeneous nature of the radiolysis system or because of the additives. We will also present considerable experimental data somewhat later which indicates a significant effect of pulsed dose rate on G-values which can probably be attributed to reduction in recombination of radiolysis products under pulsed irradiation conditions.

There exists another method for limiting back reactions in radiolysis processes involving decomposition reactions. This method is the use of additives which either result in more or less selective deexcitation of radiolysis products so that concentrations driving back reactions involving excited radiolysis products are reduced or changes in chemical characteristics, e.g., by chemical binding of radiolysis products, which limit their ability to participate in back reactions whether excited or not. The use of such additives, often referred to as scavengers, is widespread in experimental studies of radiolysis fundamentals and may be very significant in development of commercial radiolysis processes for synthetic fuel production.

Clearly, if such additives are to be useful for commercial synthetic fuel production by radiolytic means, they must be stable or easily regenerated and inexpensive, not introduce unacceptable corrosion or neutron activation problems, not tie up principal reactants or the desired radiolysis products in a highly irreversible manner, etc. In a combined radiolytic-thermochemical cycle for synthetic fuel production, modest concentrations of unseparated primary cycle chemicals, present deliberately or because of separations inefficiencies, might play this role. For example, simple inorganic compounds could be useful scavengers for commercial processes if their principal radiolytic decomposition reactions are highly endoergic and do not involve highly irreversible combinations with main reactants or desired radiolysis products and the back reactions to reconstitute the original compound are subject to little competition from other reactions, especially reactions with main reactants or radiolysis products of interest. Most organic compounds, however simple, do not, in general, satisfy these criteria and, therefore, although widely used in experimental work, are not likely to be satisfactory commercial scavengers and often tend to make interpretation of experimental studies difficult, e.g., because of contributions to $G(H_2)$ when used as additives in water radiolysis experiments. Because we will be discussing numerous examples of such additives and their effects as we proceed, we will forego presentation of examples here.

LET EFFECTS ON RADIOLYTIC YIELDS

One of the characteristic properties of energy deposition by ionizing radiation which was recognized early as having potentially significant influence in determining radiolytic G-values, and steady-state radiolysis product concentrations under constant irradiation, is the so-called linear energy transfer (LET) of a particular form of radiation in a specific reaction mixture. The mean linear energy transfer is the mean rate of deposition of energy by photons or particles along their tracks through the medium, calculated by dividing initial energy by average total path length.

In general, particle and photon path lengths are random variables with considerable variances, initial particle and photon energies are also random variables, and their interaction cross sections vary significantly with energy. Thus the mean linear energy transfer, although widely used, is a grossly lumped parameter with which to characterize density of energy deposition along photon and particle tracks. Nonetheless, because radiolytic yield and product concentrations are expected to depend on energy deposition density along particle and photon tracks, the concept is important. Local linear energy transfers for well-characterized locations, e.g., initial values, or points on representative average photon or particle tracks are also occasionally measured and reported. However, unless specifically qualified, LET will be used to denote mean LET's consistent with common practice. Mean LET's and initial LET's for important types of radiation of representative particle of photon energies in air and water are listed in Table II. The concept of LETs for neutron irradiation is probably only meaningful when related to energy deposition by "knockon" or secondary ions produced by scattering of neutrons by atomic nuclei, for in between such collisions, almost no neutron energy deposition occurs because neutrons bear no electric charge. Charged particle energy deposition occurs in what is for many purposes a continuous fashion along the particle tracks. Similar considerations apply to x-ray and gamma-ray photon energy deposition.

Because radiolytic processes are so complex, theoretical predictions of LET dependences of G-values and product concentrations based on present understanding are unlikely to be very accurate. Experimental evidence indicates increases in G-values and product concentrations with increases in LET in some cases, and decreases in others. We will discuss in more detail the results of some experimental studies of LET effect on G-values for water radiolysis to illustrate the complexity of the problem.

Measurements[42] of the LET dependences of $G(H_2)$, $G(H)$, $G(-H_2O)$, and $G(Fe^{+3})$ in aqueous 0.8 N sulfuric acid solutions containing iron sulfate revealed that for LET values in the range 0.02-1.0 eV/A (2 to 100 MeV/cm), which includes the LET values for the gamma rays, x rays, fast electrons, accelerated deuterons, and neutrons used in the extensive experimental work, the following approximate relationships were valid:

$$G(H_2) = 0.44 + 0.5\ |LET|$$

$$G(Fe^{+3}) = 16.0 - 7.0\ \sqrt{|LET|}$$

$$G(H) = 3.80 - 1.75\ \sqrt{|LET|} - 0.25\ |LET| \sim 3.80 - \sqrt{|LET|}$$

$$G(-H_2O) = 4.68 + 0.75\ |LET| - 1.75\ \sqrt{|LET|} \sim 4.6 - 1.22\ \sqrt{|LET|}$$

For LET values in the range 3 to 25 eV/A (300 to 2500 MeV/cm), corresponding to alpha particles of various energies, but much less than the mean LET value of about 385 eV/A (3.85×10^4 MeV/cm) of fission fragments, the corresponding results were:

$$G(H_2) = 1.1 + 0.02\ |LET|$$

$$G(Fe^{+3}) = 8.0 - 0.78\ \sqrt{|LET|}$$

$$G(H) = 1.45 - 0.195\ \sqrt{|LET|} - 0.01\ |LET| \simeq 1.55 - 0.25\ \sqrt{|LET|}$$

$$G(-H_2O) = 3.65 - 0.195\ \sqrt{|LET|} + 0.03\ |LET|\ .$$

One reviewer[43] ventured the following opinions concerning the LET effect on G-values for pure water vapor radiolysis compared to that for radiolysis of the pure liquid. He suggested that because of the low density of the vapor, LET effects would be expected to be much less significant for radiolysis of the vapor than for radiolysis of the liquid. All but a few studies of water vapor radiolysis have been conducted using gamma rays, x rays, and electron beams which have low LET's. However, he did note that some studies had been conducted using fission fragments for irradiation of water vapor with and without scavenging additives and remarked that observed $G(H_2)$ values were similar in magnitude to those obtained using gamma rays. On the other hand, he also noted that marked LET effects on G-values had been observed with water vapor to which substances such as cyclohexane had been added in concentrations sufficiently great that radical concentrations large enough to cause radical-radical reactions to become important. Finally, he remarked that because of obstacles which prevent changing the value of only one experimental variable at a time in such experiments, definitive interpretation of experimental results is difficult.

The difficulty of theoretically predicting only the LET dependence of G-values is illustrated by the following. A theoretical study, based on consideration of available experimental data and a simple qualitative model of the radiolysis process performed at William H. Johnston Laboratories, Inc.,[11] led to the conclusion for radiolysis of oxygen to give ozone that $G(O_3)$ should increase in passing down the LET scale from fission fragments through alpha particles and electrons to gamma rays, with limits of 6 and 12 respectively for high and low LET values were predicted by this first-order thoery. But, other investigators contend that their research indicates that with fission-fragment irradiation of O_2 at high pressures (68 atm) and temperatures below ambient, $G(O_3)$ values of 10-15 can be achieved.

We mentioned previously that G-values for fast neutron radiolysis were expected to be similar to those for heavy ion radiolysis. This is because the primary energy transfer mechanism is through collision with atomic nuclei, to which significant kinetic energy is imparted, and it is these knock-on atoms which are believed to be responsible for most of the radiolytic effects. Thus, LET-dependences of G-values for heavy ions are believed to be applicable for fusion neutron radiolysis. The LETs for fast neutrons, although little information is available, are expected to be similar in magnitude to those for gamma radiation and are probably misleading if used in attempts to infer LET effects on G-values for fast neutron radiolysis.

REPRESENTATIVE G-VALUES

We will not attempt to survey all reported radiolysis results, but will present only representative G-values for carbon-dioxide, water, and hydrogen halide decomposition here. Other experimental radiolysis data will be presented where appropriate. Comparison of G-values reported by different workers is difficult because there is no universally accepted basis for absolute dosimetry for different types of radiation and radiolysis systems. Therefore, such comparisons will often be only qualitative rather than quantitative. The data that we discuss here will reveal strong dependences of G-values on dose rate, the presence of scavenging additives, temperature, and in the case of gas-phase radiolysis, pressure.

There is a vast body of experimental data concerned with carbon dioxide radiolysis. Some of this data is listed in Tables III and IV and Fig. 2. Steady state concentrations of CO during radiolysis of pure gaseous CO_2 appear to increase with dose rate, e.g., see Table III, especially for pulsed irradiation, G(CO) apparently increases with increase in pressure at low dose rates and high densities, e.g., see Fig. 2, contrary to many other radiolytic decomposition reaction G-value pressure dependences, but also appears to be subject to little variation with increase in pressure for pulsed, high-dose-rate, low-pressure radiolysis, and G(CO) seems to decrease with increasing temperature, as opposed to increase in $G(H_2)$ with temperature for water radiolysis. Values for G(CO) for radiolysis of liquid CO_2 have been reported to decrease somewhat with increase in dose rate and, unexpectedly, to lie in the range from about 3.5 to about 5.0 independent of the nature of radiation characteristics from gamma radiation to fission fragments.[46] Solid carbon dioxide G(CO)-values which have been published do not seem to show much agreement, ranging from about 0.3 to about 10.[46] Similarly, there is considerable variability of reported values for G(CO) for scavenged gas-phase carbon dioxide radiolysis, as indicated by the data of Table IV. We also see that the largest reported CO_2 radiolysis values, about 10, still correspond to rather low radiolysis efficiencies, i.e., approximagely only 30%.

The totality of water radiolysis data is even greater than that for carbon dioxide radiolysis. As liquid-phase water radiolysis $G(H_2)$-values are typically less than those for vapor-phase radiolysis, we will concentrate on vapor-phase water radiolysis. Figures 3 and 4 indicate the effects of various scavenging additives on $G(H_2)$-values for, respectively, steady low dose rate x-radiolysis and pulsed, high-dose-rate electron radiolysis of water vapor. Figures 5 and 6 respectively show measured temperature and pressure dependences of $G(H_2)$-values for steady low-dose-rate γ-radiolysis at low pressures. These data appear to represent the highest $G(H_2)$ values that have been reported for water vapor radiolysis and hence are most favorable insofar as radiolytic hydrogen production is concerned. A $G(H_2)$-value of 15, corresponding to very low pressures, represents a radiolysis efficiency of only about 38%. The economic penalties associated with synthetic fuel production at low pressures, discussed in the preceding section on fusion process heat, are severe and, as we have already indicated, low-pressure gas-phase radiolysis is generally not compatible with fusion reactor blanket design requirements, although inertial confinement fusion is less constrained in this regard.

We present data in Table V and Fig. 7 for hydrogen halide, HCl and HBr specifically, because of potential applications of radiolysis in combined radiolytic-thermochemical cycles for synthetic fuel production by water splitting which we will discuss subsequently. The pulsed, high-dose rate data of Fig. 7, which agrees very well at the higher pressures with the low-dose rate data of Table V, indicates an unfavorable pressure dependence for $G(H_2)$-values. For steady, low-dose rates there is apparently very little dependence of $G(H_2)$ values on pressure to rather low pressures.[48]

One very important feature of all the data which has been reviewed is that the highest G-values are often observed with high-dose-rate, pulsed irradiation. The high dose rates, usually obtained with very short pulse (as short as a few tens of nanoseconds), electron accelerators, and the pulse durations are similar to those expected in inertial confinement fusion reactor blankets. Another very important feature which was not emphasized by the selected data, and which depends on so many factors that accurate predictions are almost impossible, but to which we have alluded previously, is that the concentrations of radiolysis products which can be achieved are often very low.

POTENTIAL ADVANTAGES OF FUSION REACTORS RELATIVE TO FISSION REACTORS AS SOURCES OF RADIATION FOR DRIVING RADIOLYTIC PROCESSES

As noted previously, G-values for neutron-induced radiolytic reactions are expected to be similar in magnitude to those for heavy ion-induced reactions under the same conditions. Therefore, no particular advantage, beyond that for heavy ions relative to radiolysis with electron and gamma radiation, can be claimed for radiolysis with fusion neutrons under steady irradiation conditions, and these advantages would probably only be available for condensed-phase radiolysis. However, although there seems to be no published fusion neutron radiolysis experimental data upon which to base definitive judgment, the considerable pulsed electron beam radiolysis experimental data which was discussed above suggests that high-dose-rate, pulsed irradiation may result in higher effective G-values, if not intrinsic G-values as well, for at least some radiolytic reactions than does steady irradiation. One reason which has been suggested for these results is that the interpulse periods permit deactivation of excited radiolysis products before recombination can occurt. Therefore, inertial confinement reactors may possess a significant advantage in this regard relative to both fission and magnetic confinement fusion reactors.

Studies aimed at development of commercial radiolytic processes for production of synthetic fuels and other chemicals, such as ozone and nitrogen fixed from air, driven by fission reactors have generally concentrated on the use of fission fragment energy which represents about 80% of the total fission energy release, with alpha particles, neutrons, and gamma photons carrying off the rest. Fission fragments are often very efficient radiation energy transfer agents, as indicated by the discussion on LET dependences of G-values.

The primary problem, which has not been definitively overcome, with the direct use of fission fragment energy for radiolytic chemical and synthetic fuel production is, of course, contamination of radiolysis products with radioactivity. The use of fission fragments for direct radiolytic chemical and synthetic fuel production could be contemplated for fusion-fission hybrid reactors. However, such installations would involve the same product contamination problems as fission reactors used for direct fission-fragment radiolytic synthetic fuel and chemical production. Furthermore, one would have an incredible conglomeration of technologies, all of which presumably would be important in determining commercial success.

The major advantage of being able to utilize a large part of fusion energy release outside the reactor cavity with relative independence of cavity phenomenology, not the case with fission reactors, has already been mentioned. The limited ranges of fission fragments, useful in that there is no problem in absorbing the radiation, severely limits the dimensions of fuel elements in fission chemonuclear reactors, making design of reliable fuels for such reactors difficult. The use of very thin plates or foils of bare fuels and fuels coated with very thin, i.e., a few microns thick, coatings and of powdered fuels to be intimately mixed with radiolysis process fluids have been suggested. Neutron activation of main reactants and products and impurities and corrosion of activated structure and fuel elements also present severe problems. Pure fusion reactors do not, of course, produce fission fragments, but neutron activation of main reactants and impurities and corrosion of activated structure, as discussed in the section on high-temperature process heat from fusion reactors, must be considered. Tritium contamination of radiolysis products must also be minimized in fusion chemonuclear reactors. Finally, the neutron economy of fission reactors can be severely compromised by excessive neutron absorption in process materials and therefore, the range of systems which can be processed radiolytically in a fission reactor is somewhat limited.

That tritium contamination of radiolysis products can be limited to acceptable levels seems likely. Various aspects of fusion reactor tritium fuel cycles were discussed in the section on fusion process heat sources. Whether or not neutron activation and activated structure corrosion will lead to unacceptable contamination, which cannot be economically eliminated, of radiolysis product streams from fusion chemonuclear reactors is more problematical. Certainly such contamination represents a major obstacle to commercialization of fusion-driven radiolytic processes for synthetic fuel production. The ability to utilize a majority of fusion energy, that associated with the neutrons, free of cavity phenomenology, not a feature of fission reactors, has already been mentioned.

MECHANICAL DESIGN PROBLEMS FOR FUSION REACTOR BLANKETS FOR GAS-PHASE RADIOLYSIS

The following remarks concerning fusion reactor radiolysis blanket design considerations when gases are to be radiolyzed further illustrate the difficulties which must be overcome in developing commercial synthetic fuel production processes involving radiolysis of gaseous species. If a majority of fusion neutron energy is to be absorbed by acceptable thicknesses, i.e., no more than a meter or two, of gaseous radiolysis systems, then operation of fusion reactor blankets at hundreds of atmospheres pressure will be required. At such pressures, the only practical means of containment of the gases to be radiolyzed appears to be the use of relatively small diameter, i.e., no more than a few to a few tens of centimeters, tubes. The stresses imposed on the walls of larger ducts, vessels, etc., become unacceptably large.

Vessels for containing high-pressure gases are almost never built with flat sides, but are almost always spherical or cylindrical with hemispherical or ellipsoidal heads because thinner walls are required for the same internal pressure. Many inertial-confinement fusion reactor concepts are compatible with these geometries. The blankets for many magnetic-confinement reactor concepts, however, must be constructed of large numbers of modules of unusual shapes, at least some of which must have flat or nearly flat sides if efficient utilization of space inside the large cryogenic magnets is to be accomplished. This topic is of considerable importance for magnetic-confinement fusion because if the large cryogenic magnets which provide the magnetic fields necessary for confinement must be made larger to accomodate inefficient blanket packing schemes, the required magnetic field strengths and the magnet sizes, power consumption, and costs increase rapidly. The geometries of present tokamak-type and simple mirror reactor concepts are particularly unsuitable in this regard.

The amounts of structure required to contain gases in large blanket modules with flat sides at pressures over 100 atm would be enormous, with consequent great weights to be supported, high costs, and large inventories of activated structure. Replacement of these modules of unusual geometry with cylindrical or spherical elements is possible, but at cost of high void fractions or filling of the interstices with cooled irregular solid shapes or channels. The latter approach would involve enormous increases in complexity of blanket "plumbing". It has been claimed that any reasonable estimate of mean time to failure for welded joints in such designs would show that this approach would lead to unacceptable short times between failure somewhere in the coolant system, i.e., unacceptably high incidences of forced outage.[52]

A detailed study[53] of radiolytic decomposition of CO_2 to give CO and O_2 in magnetic confinement fusion reactor blankets considered the use of titanium, steel, and a high-strength aluminum-alumina fiber composite for radiolysis mixture containment. One of the conclusions of this study was that even if relatively high, i.e., up to 50% of ultimate tensile strengths, operating stress levels in the tube walls could be tolerated, a majority of the fusion neutron energy would be deposited in blanket structure and tritium breeding blanket regions rather than in the process gas. This judgment was based on an estimate of deposition of at least one-third of the neutron energy in structure, and about one-third of the neutron energy in tritium breeding regions. Recent studies[54] concerned with the use of fusion energy to drive thermochemical cycles for hydrogen production by water splitting suggest that even higher fractions of the neutron energy will be deposited in tritium breeding zones. The probable necessity for tritium self-sufficiency of individual fusion reactors was discussed in the section on process heat applications of fusion energy.

Based on an assumed maximum G-value of about 10 for radiolysis of CO_2 to produce CO and O_2, corresponding to a potential radiolysis efficiency of about 30%, the radiolysis efficiency is reduced to only about 10% if the fraction of neutron energy absorbed by blanket structure and tritium breeding regions together is taken as two-thirds. Furthermore, if tube wall operating stress levels must be reduced by a factor of two, the actual radiolysis efficiency would also be roughly halved to only about 5%, which is generally considered to be far too low to be marginally interesting even for topping cycle applications because:

- operating stress levels of 25% of ultimate tensile strengths are considered to be still relatively high by fusion reactor blanket design engineers, partly because of creep at elevated temperature, and special measures would have to be taken to ensure acceptable blanket lifetimes under such conditions;
- at such low efficiencies, the grafting of complex radiolysis blanket structure and process equipment onto relatively simple power production blankets and cycles is expected to result in unacceptable capital cost;
- the increased probability of blanket structural failure because of greatly increased containment structure wall area, more welds, joints, seals, etc., and additional corrosion problems could have a major impact on blanket reliability; and
- a large imbalance between electric power supply and demand would result from the use of such radiolytic-topping cycles for production of enough synthetic fuel to satisfy a significant portion of the nation's requirement for such fuels.

Thus we contend that simple radiolytic decomposition of gaseous species to yield a synthetic fuel directly probably will never be the basis for a commercially viable fusion-driven synthetic fuel production process. Unfortunately, if this judgment is valid, the simplest radiolytic processes for synthetic fuel production must be elimiated from further serious consideration.

RADIOLYTIC PROCESSES AS TOPPING CYCLES FOR THERMOCHEMICAL, ELECTROTHERMICAL (HYBRID), OR ELECTROLYTIC PROCESSES FOR SYNTHETIC FUEL PRODUCTION

Assume that a fusion reactor radiolytic blanket concept which shows promise of being relatively economical to construct, and compatible with other fusion reactor blanket requirements can be developed for a radiolytic process for synthetic fuel production in a direct fashion. Topping of a pure thermochemical, hybrid, or electrolytic process for synthetic fuel production by such radiolytic processes does not result in the same imbalance in electric power production versus synthetic fuel production that topping of electric power generation cycles with low-efficiency radiolytic processes leads to, provided that the radiolytic process rejects heat to the bottoming cycle at a temperature compatible with bottoming cycle requirements.

In general, if thermal energy can be delivered to, and utilized by, thermochemical, hybrid, or electrolysis processes at higher temperatures, cycle efficiencies can be increased. However, many thermochemical cycles involve relatively corrosive substances whose rate of attack on materials of construction increase as temperature increases so that the inability of affordable materials to withstand attack for adequate times at sufficiently high temperatures often places limits on maximum cycle temperatures. If a direct radiolysis process involves less corrosive process substances, then operation of the radiolytic topping cycle at temperatures above the maximum temperature of the bottoming cycle may be possible, which would permit efficient utilization of topping cycle reject heat. The desired product of the radiolytic process need not be the same as that produced by the bottoming synthetic fuel production process, thereby allowing for greater flexibility in satisfying the nation's synthetic fuel demands.

COMBINED RADIOLYTIC-THERMOCHEMICAL OR RADIOLYTIC-ELECTROTHERMOCHEMICAL (HYBRID) CYCLES FOR SYNTHETIC FUEL PRODUCTION

The efficiencies of many thermochemical and hybrid cycles for synthetic fuel production are limited by the rates at which one or more spontaneous chemical reaction steps can be performed and by reaction yields at temperatures compatible with process equipment materials of construction limitations or energy source characteristics. Catalysts, which may, however, be unacceptably costly, can be used to improve reaction kinetics, but cannot overcome thermodynamic limitations on equilibrium yields. Electric power can be used to provide free energy to drive reactions which would proceed spontaneously only in the wrong direction under desired cycle conditions.

Radiation can also be used to improve reaction rates or to drive reactions which otherwise would not proceed in the desired direction. In the former instance, the radiation may be viewed as performing the function of a catalyst and probably not required to provide reaction energy, while in the latter the radiation provides free energy and heat of reaction to drive the reaction.

The latter case represents the type of radiolysis process which is of interest here, for the amounts of radiation energy required merely to improve the kinetics of spontaneous reactions are, in general, expected to be small and probably would not be supplied by a fusion neutron source. However, the use of ionizing radiation must offer some advantage over alternative methods, e.g., electricity, photochemistry, etc., for supplying free energy and enthalpy to drive such reactions.

Potential advantages relative to electrolysis offered by radiolysis include:

- higher reaction rates per unit volume of reactor, unit energy transfer area, etc., resulting from circumvention of usual restrictions on electrolysis cell current densities, heat fluxes in heat exchangers, etc.;
- greater overall fuel production cycle efficiencies;
- lower process equipment, fusion reactor, and electric power system capital, operating, and maintenance costs;
- greater plant availability;
- greater process flexibility; and
- replacement of reaction sequences by smaller numbers of reactors.

The suggestion that one or more steps in a thermochemical or hybrid cycle for synthetic fuel production be driven by radiation has been advanced by many persons. The role of the radiation is similar to that of electricity in the electrolytic step of a hybrid cycle and one could imagine replacement of an electrolysis step of a hybrid cycle by a radiolytic step. However, because radiolytic processes tend to be, at best, only moderately efficient, in contrast to many electrolytic processes which can exhibit relatively high efficiencies, it appears unlikely that such radiolysis steps will be useful unless reject heat can be used efficiently elsewhere in the cycle. If this judgment is correct, then such radiolysis steps must, in general, be conducted at or above the maximum temperature of other, purely thermochemical, endothermic steps in the cycle, while there is no such restriction on electrolytic steps of high efficiency. On the other hand, many electrolysis reactions can be conducted efficiently only within narrow ranges of temperature, composition, and pressure, while many radiolytic processes are subject to fewer such restrictions.

Because radiolysis product concentrations may be, but, as noted previously, need not be, relatively low, some possible radiolytic steps may involve excessive separation and recycle systems process equipment and operating and maintenance costs and energy requirements. Only radiolytic steps in which radiolysis products are automatically separated from reactants, e.g., steps involving radiolysis of heterogeneous systems, or for which efficient, low-cost means of effecting sharp separations are available, seem likely candidates for commercialization.

The fact that the role of radiolysis in combined radiolytic-thermochemical cycles is analogous to that of electrolysis in present hybrid cycles means that radiolysis can be used to reduce the number of reaction steps in a cycle just as electrolysis is used to reduce the number of reactions by replacing a sequence of two or more reactions required to effect a given chemical change by a single reation which will not otherwise occur spontaneously and for which electrical energy is used to provide the free energy to drive the reaction. The potential for savings in process equipment capital, operating and maintenance costs, through reduction in the number of process steps is obvious, but may or may not be realizable. An example of a family of cycles in which reductions in the total number of reaction steps by introduction of a radiolytic step is the following:

$$H_2SO_4(g) \xrightarrow{\text{high temperature}} H_2O(g) + SO_2(g) + 1/2\ O_2(g)$$

$$SO_2(aq) + 2H_2O(l) + Br_2(aq) \xrightarrow{\text{low temperature}} H_2SO_4(aq) + 2HBR(aq)$$

$$2HBr(aq) + M\ Br_x(aq) \xrightarrow{\text{low temperature}} M\ Br_{x+2}(s) + H_2(g)$$

$$M\ Br_{x+2}(s) \xrightarrow{\text{high temperature}} M\ Br_x(s) + Br_2(g)$$

where M represents a metal atom, in which the last two reaction steps would be replaced by a single radiolytic step for decomposition of HBr following separation of HBr from the aqueous solution.

This family of thermochemical cycles was once considered likely to produce a strong candidate for commercialization because of inherent potential for high efficiency provided the kinetics of all the listed reactions were favorable.

The first two reactions with chromium as the metallic element are known to give satisfactory yields at satisfactory rates under the listed conditions. However, extensive efforts to achieve acceptable reaction rates for the third reaction with a variety of choices for the unspecified metallic element M have been unsuccessful. The third and fourth reactions whose function is to accomplish the decomposition of HBr for recycle to the first reaction can be replaced by a single electrolytic step to close the cycle, but apparently this has not been considered sufficiently promising to attract much interest. The substitution of a radiolytic step has not been extensively investigated, but as indicated by the remarkable consistency of the data in Table V and Fig. 7, some good data on radiolysis of HBr have been published. Unfortunately these data suggest very low efficiencies for such a radiolysis step, i.e., only about 3.7% for the higher-pressure $G(H_2)$-values. Some consequences of low radiolysis efficiencies are discussed in the context of our next example.

A West German patent, Ger. Offen. 2,454,561 with H. J. Gomberg listed as the discoverer, has been obtained by KMS Fusion, Inc. for combined radiolytic-thermochemical cycles for synthetic fuel production which are claimed to be especially suitable for mating with fusion energy sources and in which the effectiveness of radiation energy is claimed to be considerably enhanced. There is a corresponding United States patent application, US Appl. 416,998, on file, but information concerning patent applications in the United States is not released prior to final granting of a patent. Therefore, the following discussion of these claims is based on a translation of the German patent document.

We remark first that, as in the field of hydrogen production with presently conceived pure or hybrid thermochemical cycles, few other investigators would claim that the cycles described in the patent documents display obvious and exceptional promise for successful commercialization or represent the final word in this area, even assuming successful development of fusion-driven radiolysis technology. They are presented merely as examples of possible combined radiolytic-thermochemical cycles for synthetic fuel production.

As outlined sketchily in the patent application, one of the combined radiolytic-thermochemical cycles for hydrogen production by water splitting considered by KMS Fusion, Inc., involves the reactions:

$$H_2O(g) + Cl_2(g) \xrightarrow{800C} 2HCl(g) + 1/2\ O_2(g)$$

$$2HCl(g) \rightsquigarrow H_2(g) + Cl_2(g)\ .$$

We will not comment on the first, pure thermochemical, reaction except to note that it will proceed spontaneously at the indicated temperature and that the reaction rate would probably be acceptable. Instead we will concentrate on the radiolytic step.

The $G(H_2)$-value, 8, which is claimed for the radiolytic reaction in the patent application, is confirmed by experiment, with remarkable agreement of vapor-phase G-values for HCl radiolysis, listed in Table V, reported by independent investigators, and little apparent effect of LET. The latter characteristic of the G-value data means that G-values for fusion neutron radiolysis should be similar. However, this G-value corresponds to a radiolysis efficiency of only about 7.7%. Although it is realized that the proposed cycle must be viewed in its entirety for a proper assessment of its potential, it is difficult to see how this cycle can be developed into a commercially-viable process for synthetic fuel production. We claim that this cycle can be part of an energy efficient energy production plant only if it is used to top another synthetic fuel production process, or electric power cycle. This follows from the fact that even if a radiolysis efficiency of 10% can be achieved, the reject heat from the radiolysis step will be about 200 kcal/gmol HCl (ΔH_F^o = 22.03 kcal/gmol for HCL at 25°C), while the thermal energy which can be absorbed by the first, endothermic reaction, is only about 7 kcal/gmol HCl (ΔH_R^0 = 14.158 kcal/gmol HCl at 800°C), assuming that the radiolytic step can be carried out at temperatures above 800°C so as to reject heat to the first step.

In addition, both reactions are gas-phase reactions and some of the fusion reactor radiolysis blanket design problems resulting from the high pressure required to obtain reasonable stopping powers for fast neutrons in gases have already been discussed. The necessary separations of reactants and products for both steps in the cycle are also likely to be problematical because conversions in the radiolytic step are expected to be quite low and gas-phase separations are notoriously difficult. Recirculation rates will be large if only low conversions can be achieved. Finally, there is some cause for concern regarding activation of chlorine and bromine, as indicated by the thermal neutron absorption cross section data of Table VI for chlorine and bromine.

Another of the combined radiolytic-thermochemical cycles described in the KMS Fusion, Inc., patent involves the reactions:

$$CaBr_2(s) + 2H_2O(g) \xrightarrow{730C} Ca(OH)_2(s) + 2HBr(g)$$

$$2HBr(g) \longrightarrow H_2(g) + Br_2(g)$$

$$Ca(OH)_2(s) + Br_2(g) \xrightarrow{100C} CaBr_2(s) + H_2O_2(l)$$

$$H_2O_2(l) \longrightarrow H_2O(l) + 1/2\ O_2(g)\ .$$

Once again, the radiolysis step is to be conducted in the gas phase and the radiolysis efficiency, as noted previously, is even lower, only about 3.7 based on the data of Table V. The additional reaction steps in this cycle apparently do not permit achievement of greater cycle efficiencies, compared to the first example, but will require more process equipment.

Thus, while there appears to be some potential for commercialization of fusion-driven combined radiolytic-thermochemical cycles for synthetic fuel production, the examples drawn from the KMS Fusion, Inc. patent document do not possess this potential and involve direct contradictions of obvious principles for selection of suitable cycles of this type. Higher G-values and condensed-phase, or two-phase, radiolysis would seem to be essential features for a commercially-viable fusion-driven combined radiolytic-thermochemical cycle. Whether or not such combined cycles exist is a question which can only be settled by extensive experimental programs.

ECONOMICS OF SYNTHETIC FUEL PRODUCTION BY DIRECT RADIOLYSIS USING FUSION REACTOR RADIATIONS

The viability of any technologically successful process for producing a new product or a conventional product in a new way depends ultimately on economic considerations. Although we have already indicated that there are significant technological barriers to successful commercialization of processes for synthetic fuel production by direct radiolysis of water or carbon dioxide to yield hydrogen or carbon monoxide, we will proceed here as if these technological barriers did not exist. The following simplified analysis of the economics of synthetic fuel production by such methods is based on laser fusion technology, but conclusions based on the results of similar analysis for other types of fusion reactors are expected to be similar.

The uncertainties involved in projecting production costs of synthetic fuels produced using combinations of undeveloped technologies such as laser fusion and radiolysis are large and conclusions based on comparisons of such estimates with hard-to-predict future costs of increasingly scarce conventional fuels are difficult to justify. However, we have attempted to provide just such estimates of production costs of methane by one particular process which involves fusion-driven radiolysis and to compare the estimates to methane produced by conventional coal gasification. Our analysis[56] is mostly based on optimistic assumptions, which we will discuss below, that tend to favor the use of fusion energy sources.

We first concentrate on estimates of costs of neutron energy produced in fusion reactors adapted for generalized radiolytic processes. As we have previously indicated, only fusion neutron kinetic energy can be effectively utilized for direct radiolysis. Therefore, we stipulate that other fusion reactor radiations, i.e., pellet debris ion kinetic and ionization energy, x-ray and gamma-ray photons emitted from the pellets, gamma-ray photons released by (n,γ) neutron interactions with structure and blanket materials, etc., will not be used for radiolysis, but that the energies could be converted first to thermal energy and subsequently to electric power to meet recirculating power requirements and/or for export. We further assume that inclusion of capability for tritium breeding, even at ratios less than one, in fusion reactors adapted for radiolytic production of synthetic fuels cannot be economically justified. Therefore, tritium to fuel such reactors would have to be purchased from outside sources.

A somewhat simplistic adaptation of a laser fusion electric power plant model has been analyzed to provide the desired fusion neutron energy cost estimates. The plant model adaptation includes four magnetically-protected-wall laser fusion reactors[57] with a common CO_2 laser driver and sequential switching of laser beams to successive reactors, and heat exchanger and electric power generation capacities adequate for supply of plant recirculating power requirements by conversion of reactor thermal energy. Reactor radiolysis blanket structure consists of the blanket structure normally required for containment of the liquid-lithium tritium-breeding-blanket and primary coolant fluid of the electric power reactor model which was adapted. Without lithium, the blanket, of course, has no tritium-breeding capability. Except for first-wall structure of niobium, reactor structure is made of stainless steel. No special apparatus was included for separation of radiolysis products from unconsumed reactants, for recirculation of unreacted reactants, etc., resulting in a conservative model because low steady-state product concentrations are projected.

The fusion reactor energy release per fusion pellet microexplosion is 100 MJ, of which 76.6 MJ is neutron kinetic energy and the remainder appears in the form of pellet debris ion energy and x rays which can be converted to electric power with a thermal efficiency of about 35 to 40%. Exoergic neutron interaction energy is ignored for simplicity, which tends to balance out the neglect of neutron kinetic energy losses to reactor structure, i.e., the stipulation that all neutron kinetic energy is deposited in the radiolysis process stream. The repetition rate for each reactor is ten for a total plant primary thermonuclear power of 4000 MJ (or 3.28×10^5 million BTU/day). The net rate of neutron kinetic energy production available for radiolysis after satisfaction of circulating power requirements is 2056 MW (or 1.68×10^5 million BTU/day).

Tritium to fuel the plant is purchased from fusion electric power plants with excess tritium capacity. The cost of tritium was based on estimates of marginal production cost of tritium in such installations.

Neutron energy production cost in 1973 dollars is given in Fig. 8 as a function of the cost of supplementary electric power which must be purchased to supply recirculating power, assuming that no such capacity for conversion of thermal energy were provided for the plant. The cost of neutron energy production if such capacity were provided for conversion of neutron energy to provide recirculating power is also indicated. For the range of electric power costs considered, neutron energy production costs lie in the approximate range $2.00 to $2.50 per million BTU of neutron energy. A cost breakdown for the point plant design under discussion is given in Table VII. The production cost of neutron energy clearly places a floor on synthetic fuel production costs using fusion energy sources.

Estimates made in 1973, and hence presumably comparable on a relative, if not absolute, basis, of production costs of gaseous synthetic fuels, e.g., H_2 or CH_4, from coal produced by standard coal gasification processes range from about $1.00 per million BTU to $1.50 per million BTU for commercial operation in the 1980s.[58] Even if neutron energy can be utilized at 100% efficiency for radiolytic decomposition of water or carbon dioxide, with hydrogen or carbon monoxide as the final product, and complete separations at no cost are possible, this process for synthetic fuel production using laser fusion energy sources could not compete on a production cost basis with standard coal gasification processes in the indicated economic environment. Actual radiolysis efficiencies are not expected to exceed 20 to 30% even under favorable conditions and gas separations tend to be relatively expensive.

The synthetic fuels which might reasonably be produced through radiolytic processes using fusion reactors as radiation sources include hydrogen, carbon monoxide, and methane. The raw materials for synthetic fuel production must be relatively inexpensive, not subject to resource limitations, and must contain hydrogen or carbon, or, in the case of methane production, both. Candidate materials include water, carbon dioxide, coal, and possibly carbonates, such as limestone.

Synthetic fuels must not only be competitive on the basis of unit cost, of production, or as delivered, but must also gain market acceptance. Currently neither hydrogen nor carbon monoxide are complete suitable as natural gas substitutes. Carbon monoxide can be used directly as a low BTU industrial or power fuel, but not as a natural gas substitute for commercial and residential use. The use of hydrogen, although it has many attractive characteristics, is receiving widespread attention, and is an indispensable chemical feedstock, has not been, and probably will not be in the near future, considered to be superior to methane as a fuel for a variety of reasons.

If carbon monoxide were to be produced on a large scale by direct radiolysis in fusion reactor blankets, much of it would probably be used to produce hydrogen via the water gas shift reaction:

$$CO(g) + H_2O(g) \longrightarrow H_2(g) + CO_2(g) .$$

The hydrogen could then be used as a fuel, reacted further with carbon monoxide to make methane:

$$CO(g) + 3H_2(g) \longrightarrow CH_4(g) ;$$

used for direct hydrogasification of coal:

$$C(s) + 2H_2(g) \longrightarrow CH_4(g) ;$$

or used for direct reduction of iron ore, as a chemical feedstock, etc. The reactions involved in the methane production process are either:

$$2H_2O(g) \rightsquigarrow 2H_2(g) + O_2(g)$$

$$\underline{C(s) + 2H_2(g) \rightarrow CH_4(g) \qquad\qquad}$$

$$C(s) + 2H_2O(g) \rightarrow CH_4(g) + O_2(g)$$

or:

$$2CO_2(g) \rightsquigarrow 2CO(g) + O_2(g)$$

$$2CO(g) + 2H_2O(g) \rightarrow 2CO_2(g) + 2H_2(g)$$

$$\underline{C(s) + 2H_2(g) \longrightarrow CH_4(g) \qquad\qquad}$$

$$C(s) + 2H_2O(g) \longrightarrow CH_4(g) + O_2(g) \; .$$

To further illustrate the economics of fusion reactor driven radiolytic processes for synthetic fuel production, we will now treat methane production by direct hydrogasification of coal using hydrogen produced by either direct radiolysis of water or radiolysis of carbon dioxide, followed by the water-gas shift reaction. The energetics of the water-gas shift reaction are ignored. We adopt the following optimistic scenario.

In addition to the previous favorable approximations concerned with fusion reactor and radiolytic process performance, we add the following assumptions which are also favorable for fusion driven radiolytic synthetic fuel production. The minimum net overall chemical reaction stoichiometry is assumed to be closely approached in practice. The result is an approximate 50% savings in coal feed compared to standard coal gasification processes for which the minimum net overall chemical reaction stoichiometry is given by:

$$2C(s) + 2H_2O(g) \rightarrow CH_4(g) + CO_2(g) \; .$$

We further assume that because plant coal feed is halved, coal-related chemical plant, coal handling, etc., capital and nonfeedstock operating and maintenance costs are also halved. These costs determine coal-related synthetic fuel production costs, which, if added to the fusion reactor radiolytic processing costs, give total methane production costs.

Based on these assumptions, the cost of thermonuclear neutrons which would result in the same methane production costs as the standard coal gasification processes can be calculated as functions of efficiency of radiolytic conversion of neutron kinetic energy to stored chemical energy of carbon dioxide or hydrogen and coal feed cost. The results of such computations are shown in Fig. 9.

These results suggest that fusion neutron energy costs must be much lower than the fusion neutron energy production cost estimates displayed in Fig. 8 if the savings in coal feed and coal-related processing costs are not to be exceeded by the neutron energy production costs. Thus, it seems clear that synthetic fuel production involving fusion-driven radiolytic processes would not be competitive in the context of 1973 market values and probably not for some time to come unless there are dramatic changes in market values for coal.

A Ford, Bacon and Davis Utah, Inc., study[59] led to similar conclusions regarding commercial viability of hydrogen production by laser fusion reactor radiolysis of water. This study involved the following ground rules for radiolytic fusion reactor design:

- alpha irradiation of the process fluid was to be maximized; and
- water vapor, rather than the liquid, was to be radiolyzed because of higher G-values for vapor radiolysis.

The radiolytic fusion reactor concept which evolved from these design constraints was a variant of the Los Alamos Scientific Laboratory wetted-wall laser fusion reactor concept for electric power production that had the following characteristics:

- 14-MeV fusion neutrons pass through a lithium film used to protect the first wall and the first wall structure and enter a pure steam blanket at 1000 to 2000 psia where radiolysis and heating of the water vapor occurs;
- the epithermal neutrons leaving the first blanket region pass through a structural wall into a second steam blanket, into which an unspecified boron-containing material is injected to enhance radiolytic hydrogen production by means of the $^{10}B(n,\alpha)^{7}Li$ reaction which is supposed to cause significant alpha particle irradiation of the steam in this blanket to further radiolyze and heat the steam; and
- the superheated steam from the second blanket region is passed through a turbine for electric power production and then boron, lithium, and hydrogen are separated before recirculation of water to the reactor blanket.

The estimated range of costs of hydrogen production in such fusion-driven plants, apparently in 1969 dollars, is listed in Table VIII, along with similar estimates for hydrogen production costs for alternative means of hydrogen production. We see that all the alternatives which were considered are projected to produce hydrogen at lower costs.

Great accuracy is not claimed for either the simple economic analyses of fusion-driven synthetic fuel production which were described above, or the projected costs of production for the alternative synthetic fuel production processes, even in the context of the 1969 and 1973 time frames. The important point is that simple direct radiolysis has not emerged from such comparative studies showing promise of being commercially viable approaches to synthetic fuel production.

POTENTIAL METHODS FOR IMPROVEMENTS IN RADIOLYSIS YIELDS UNDER INDUSTRIAL CONDITIONS

In this section, we speculate briefly on some possible approaches to improving radiolysis yields so as to enhance the attractiveness of radiolytic topping cycles and combined radiolytic-thermochemical cycles for synthetic fuel production, In general, however, accurate assessment of the potential of these approaches using energetic neutrons for irradiation will require substantial experimental programs, because present theoretical models are unable to predict effective G-values as a function of radiolysis product concentrations with acceptable accuracy and there is very little relevant experimental data.

Independent investigators in the modern era of radiolysis research report experimental values for radiolytic G-values obtained under similar conditions for single-phase systems near ambient temperature and pressure, with which most such studies were concerned, that are in substantial agreement for the systems and types of radiation studied. This fact suggests that if conditions under which significant improvements in G-values and radiolysis product concentrations can be effected are to be found, then unusual conditions of temperature and pressure, probably for two-phase systems, must be explored. For single-phase high dose-rate, pulsed radiolytic gas decompositions, the effect of pressure increase often seems to be decrease in G-values, as discussed previously for water vapor and hydrogen halide radiolysis. Carbon dioxide may be an exception as noted before. Therefore, increases in pressure, which might be beneficial for gas phase radiolysis insofar as radiation energy absorption in the radiolysis mixture is concerned, do not appear to be helpful in improving radiolysis efficiencies for gas-phase radiolysis, but may have little effect on condensed-phase or two-phase radiolysis. Higher pressures are expected to promote back reactions of both excited and nonexcited gas-phase product molecules in general. If the opinion that radiolytic processes for synthetic fuel production, nitrogen fixation, etc., can be economically viable only when used in conjunction with other applications of laser fusion energy, e.g., as a topping cycle for an electrical power generating reactor, in a combined radiolytic-thermochemical cycle, etc., is valid, then clearly radiolysis at high temperature must be examined experimentally in depth. The radiant energy not used in radiolytic processes can, of course, be more efficiently used in a conventional thermal power cycle or thermochemical cycle when available at a higher temperature.

An example[60] of such an unusual condition, for which higher G-values have been reported is the following. $G(H_2)$-values of about 40, i.e., corresponding to roughly 100% radiolysis efficiency, are claimed for radiolysis of water absorbed on "impure" silica gel at less than one monolayer. However, because most of the radiant energy in such systems is absorbed by the silica gel substrate, determination of the amount of radiant energy actually delivered to adsorbed water (which is in an unusual state whose characteristics are only partly understood), the basis for computation of $G(H_2)$-values, is difficult and development of a commercially-viable direct-radiolysis process for hydrogen production based on this type of system does not appear to be possible. The following are a selection of conditions for which there is at least some experimental and/or theoretical bases for suspecting that significant increases in G-values may be possible.

Effect of Temperature Increases on G-Factors

Significant increases in radiolytic G-values with increase in temperature have been reported for a number of substances whose decomposition is endothermic in nature. For example, studies of the effects of changes in temperature on G-values for various decomposition products of NH_3, CH_3OH, C_2H_5OH, n-C_3H_7OH, and C_2H_5OH in the presence of CCl_4 vapors under gamma irradiation in the temperature range 50-300°C resulted in observations of significant increases with increase in temperature, often with relatively abrupt jumps over narrow temperature ranges which are apparently critical in some sense, with "plateaus" in between. Increases over the entire temperature range 50 to 300°C of the order of a few percent for some products to several hundred percent for others were observed with these substances. A similar investigation was performed with water vapor containing 5 mol% NH_3, and water vapor containing 5 mol% C_2H_5OH over the temperature range 100 to 350°C with similar results. Observed increases in $G(H_2)$ were of the order of 15% to over 100%, with the highest values observed at the lower pressures. Experiments involving accelerated-electron radiolysis of pure water vapor as a function of temperatures revealed that the $G(H_2)$ value at 400°C was roughly 100% higher than the $G(H_2)$ value at 164°C. A sample of experimental results was given previously in Fig. 5. The authors of these reports presented theoretical explanations for the observed behavior. However, it does not appear that continued increase in G-values for water decomposition with further increase in temperature can be expected on the basis of their arguments. However, this theory has been criticized by one reviewer[43] as perhaps qualitatively correct, but not quantitatively correct.

Further experimental support for this theory, plus contradictory results, are also discussed by this reviewer. The conclusion of the reviewer was that further investigations over wider temperature ranges with different additives would be necessary for resolution of this problem.

Experimental evidence for increase in G(CO) for CO_2 radiolysis with temperatures reported in the literature include:

- a report[61] that G(CO) values for vapor-phase radiolysis of CO_2 with approximately 1 mol% additions of Xe, C_3H_8, or propylene increased on going from room temperature to 400°C from 0.6 to 2.0, 5.3 to 7.8, and 6.0 to 6.8 respectively; and
- a review[46] of experimental work suggesting that with certain scavengers, e.g., H_2 and CH_4, necessary for achievement of respectable values for G(CO), the general effect of increase in temperature is increase in G(CO), while with others, e.g., NO_2, the effect is decrease in G(CO).

For example, with CH_4 as a scavenger, and using gamma irradiation, an increase in G(CO) from 6.3 to 10 ± 0.8 was noted in one experiment on raising the temperature from 350 to 500°C, but with NO_2 as a scavenger and gamma irradiation, G(CO) declined from 4.5 ± 0.4 to 1.7 over the temperature range 20 to 270°C in another experiment, and results with fission fragments indicated little effect of G(CO) in the range 70 to 416°C with NO_2 as a scavenger.

The implication of the limited available data concerning the effect of increases in temperature on G(CO) for radiolysis of CO_2 is that significant increases in G(CO) may be possible, but that no firm conclusions can be drawn at present.

There appears to be little published information on temperature dependences of G values for decomposition of other compounds, e.g., hydrogen halides, which might, for example, be involved in one or more steps of a combined radiolytic-thermochemical cycle for synthetic fuel production. It is also important to note that if dissociation energy requirements for radiolytic decomposition reactors decrease with increasing temperature as is often the case, then even if G-values increase with increase in temperature, radiolysis efficiencies can decline at high temperatures.

Improved Scavengers

Investigations of high-dose-rate, pulsed radiolysis with accelerated electrons of H_2O, H_2O-HCl mixtures and H_2O-HBR mixtures in the vapor phase at 160°C and 1000 torr resulted in observations of increases in $G(H_2)$ with increase in both HCl and HBr content over that observed with pure H_2O vapor under the same conditions. From 0 mol% HCl to about 3 mol% HCl, $G(H_2)$ increased sharply by about 250%, but further increases in HCl content up to around 8 mol% had only minor effects. Similar increases were observed with HBr, but only about 1 mol% sufficed to accomplish the bulk of the increase in G-value for H_2 production. The experimental data, summarized for HCl in Fig. 7, was discussed previously. However, it should be noted that the asymptotic G-values are not in excess of what has already been claimed for water with various other additives, e.g., alkanes, or resulting from temperature increases. On the other hand, it is important to note that HBr or HCl would be much more stable additives than, say, alkanes, although corrosion problems might be more severe.

Radiolysis of Two-Phase Systems

A recent paper[62] discussed radiolysis of a two-phase mixture of liquid water and water vapor. The emphasis was on examination of explosion and corrosion hazards arising from radiolytic decomposition of the two-phase light-water coolant of the Italian CIRENE fission reactor concept (heavy-water moderated, natural uranium fueled). The experimental study involved $G(H_2)$ values for and steady-state hydrogen concentrations resulting from radiolysis of the steam-liquid water mixture as functions of steam quality (weight fraction or percentage vapor) under typical CIRENE reactor conditions (260-285°C, 62.9-67.7 atm, 0-100% steam quality).

Observations that the steady-state concentrations and rates of production of hycrogen and oxygen pass through maxima and that $G(H_2)$ values increased steadily as steam quality increased from 0 to 100%, summarized in Figs. 10, 11, and 12 in an irradiation facility of fixed geometry with a radiation source of fixed radiosity were explained by the authors of the paper as follows. The initial rise in H_2 and O_2 steady-state concentration and rates of production and the rise in $G(H_2)$ with increase in steam quality were attributed to great differences in solubility of O_2 and H_2 in liquid water, i.e., by a factor of about 300, with effective separation of the products of radiolysis and consequent reduction in reverse reaction rates, being claimed. This is consistent with our earlier discussion of reduction in steady state radiolysis product concentrations and effective G-values due to back reactions. The decreases in steady state radiolysis product concentrations and radiolysis rates at higher steam qualities were attributed to reductions in total water, and hence reduced absorption of radiation, in the radiolysis chamber as vapor fraction increased. The claimed radiolysis products interphase separation effect would presumably be greater for phase ratios, gas to liquid, near unity and to decrease to zero as steam quality goes to 0% or 100%.

If the suggested interphase disproportionation of radiolysis products is indeed effective in increasing $G(H_2)$ values and radiolysis product formation rates and concentrations, then a significant impact on efficiency and economics of direct radiolytic processes for hydrogen production by water splitting is possible. Also interesting is the fact that these relative high $G(H_2)$-value data were obtained at relatively high pressures and hence would appear to contradict other experimental data discussed previously which suggested that $G(H_2)$-values for water vapor radiolysis decline significantly with increase in pressure at pressures near atmospheric.

However, we hasten to point out that not all details of the experimental procedures used were reported, so that accurate assessment of the reliability of the reported G-values is not possible. Some critics of this work have suggested that unreported, and perhaps unrecognized, factors, e.g., trace impurities in the radiolysis mixture which acted as scavengers may have contributed to anomalously high G-values. In addition, we remark that the reported radiolysis product concentrations are still relatively low and may have to be substantially greater if separation work and process fluids circulation rates are not to be excessive.

However, what first attracted attention about this work was the fact that the values for $G(H_2)$ for 0 and 100% water quality were respectively roughly twice the values reported for room-temperature radiolysis of single-phase liquid water and single-phase water vapor. However, as discussed under the heading of effect of temperature on G-values, the higher values reported for high steam qualities appear to be consistent with other reports of increase in $G(H_2)$ for water vapor radiolysis with increase in temperature. Therefore, it seems reasonable to speculate that no special significance should be attributed to the reported high values of $G(H_2)$ insofar as the two-phase nature of the radiolysis experiments is concerned. These results appear merely to indicate a smooth transition between $G(H_2)$-values for single-phase liquid water and steam of 100% quality as steam quality is increased.

Another report[63] described increases in $G(H_2)$ upon bubbling N_2 gas through liquid water and attributed the effect to escape of gaseous species from sites of formation so rapidly that recombination was prevented. An increase from 5×10^{-3} to 0.26 was reported. However, it should be noted that these G-values are relatively small to begin with, and the observed behavior may not be representative, although certainly suggestive. There appears to be no more evidence bearing on this problem. Radiolysis of two-phase systems does not appear to have attracted that much attention.

Radiolysis in the Critical Region

The kinetics of diffusion processes in the vicinity of the critical points, i.e., the temperature and pressure at which liquid and vapor properties become identical, of binary solutions have been investigated for a number of homogeneous and heterogeneous reactions. The interest lies in the fact that at the critical point derivatives of chemical potentials of the components with respect to composition vanish. Because diffusion fluxes are, at least to the first order, proportional to chemical potential gradients, the implication is that diffusion will be very slow in the region of the critical point. Therefore, in the critical region, chemical reaction rates should be very slow if diffusionally influenced.

Marked retardation, i.e., by several orders of magnitude, of homogeneous bimolecular reactions in the critical region have been established experimentally for the recombination of atoms of iodine formed by photolytic decomposition of I_2 molecules in carbon dioxide and of clorine atoms formed by photolytic decomposition of Cl_2 molecules, with Cl_2 the solvent, according to a recent review.[64] The suggestion has also been made that advantage be taken of the special properties of the critical region for isotope separation.[65]

One principal reason for low G-factors in many endothermic radiolytic processes appears to be recombination of dissociated species, i.e., ions, free radicals, etc., to form the original molecules before they can be separated. The retardation of diffusion of dissociated species near the critical point may be effective in preventing recombination before excitation energy can be dissipated to surrounding molecules. The fact that this appears to be the case for photolytic dissociation, a closely related process, encourages this belief.

Unfortunately, the critical region appears to have been largely neglected insofar as radiolysis studies are concerned. There seems to have been only one experimental investigation concerned with radiolysis near the critical point. A study[66] of gamma radiolysis of ethane near its critical point led to observations of an average increase of about 55% in the total yield of radiolysis products at the critical point compared to yields obtained at the critical pressure, but a few degrees above and below the critical temperature. Therefore, an accurate assessment of the potential for increasing G-factors by radiolysis in the critical region is not possible at this time. However, the concept appears to merit further investigation.

Table IX is a short list of critical properties of some hydrogen-containing substances which we have previously mentioned in connection with synthetic fuel production by radiolysis. An important point concerning temperature levels for typical radiolysis systems which are illustrated by the values of Table VIII is that, unfortunately, the critical temperatures are all relatively low. Therefore, efficient utilization of reject heat will generally not be possible with such systems and G-value enhancement resulting from radiolysis in the critical region must be substantial and/or other candidates for radiolysis with higher critical temperatures must be considered.

Laser-Enhanced Radiolytic Processes

One of the principal reasons that G-values for many endothermic radiolytic processes are so low is that much of the energy deposited within the substance to be decomposed is transformed into molecular excitation energy short of that necessary to cause dissociation. The proposal[67] has been advanced to introduce small amounts of additional energy into the excited, but non-dissociated, molecules resulting from interaction of radiation with the feed substances in the form of laser light of selected wavelengths in order to cause significant additional dissociation to form desired products, e.g., hydrogen and carbon monoxide for fuel, fixed nitrogen, ozone, etc. Because of expected relatively low efficiencies of conversion of thermonuclear energy into laser light it is foreseen that for such combined laser-light-radiolytic processes to be economically competitive relatively large increases in effective G-values with only modest additions of laser light energy will be necessary. The use of carefully selected frequencies should lead to efficient utilization of the laser light energy. The emphasis should probably be on states of excitation that are relatively near dissociation in energetic terms and are densely populated.

Unfortunately, this suggested symbiotic process does not appear to have been investigated to any significant degree. Therefore, without considerable further research, it does not appear to be possible to make a critical judgment concerning the viability of this concept.

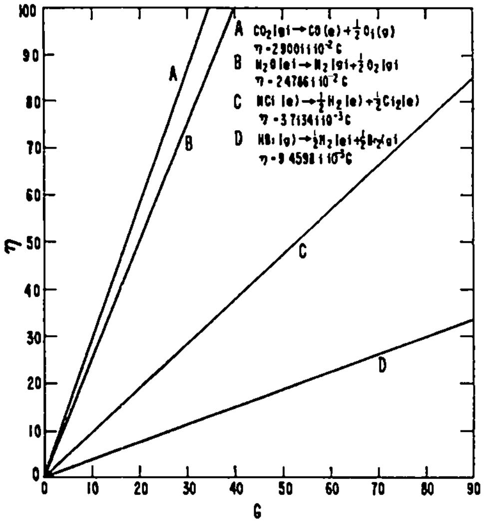

Figure 1.
Radiolysis efficiency as a function of G-value for selected gas-phase radiolysis reactions.

TABLE I

EFFECTIVE G-VALUES FOR RADIOLYSIS OF OXYGEN TO GIVE OZONE AS A FUNCTION OF PRODUCT CONCENTRATION AT 195K and 460 TORR

Ozone Concentration, ppm	Effective G (O_3)
43.6	5.33
46	5.6
304	6.0
5100	4.1
7000	3.65

TABLE II

TYPICAL RANGES AND LET VALUES FOR VARIOUS TYPES OF RADIATION IN AIR AND WATER

Radiation Type	Particle Mass, amu	Particle Charge, Electronic Changes	Particle Energy, MeV	Energy to Mass Ratio, MeV/amu	Average Range* in Air at STP, cm
^{60}CO gamma photons	0	---	1.25		1.48×10^4
electrons	5.4×10^{-4}	1	2.0	3.69×10^3	7.9×10^2
protons	1.0	1	5.0	5.0	3.4×10^1
^{210}Po alpha particle	4.0	2	5.3	1.33	3.84
fission fragments	95-139	20-22	65-97	0.47-1.02	2.2
fast neutrons	1.0	0	5.0	5.0	---

Radiation Type	Average Range* in in Water, cm	Initial LET in Water MeV/cm	Mean LET in Water MeV/cm	Relative Mean LET in Water MeV/cm	Mean LET in Air, MeV/cm	Relative Mean LET in Air, MeV/cm
^{60}Co gamma photons	1.59×10^1	---	---	---	---	---
electrons	9.5×10^{-1}	1.9	2.1	1.00	2.55×10^{-3}	1.00
protons	3.6×10^{-2}	8.16×10^1	1.39×10^2	6.62×10^1	1.47×10^{-1}	5.77×10^1
^{210}Po alpha particles	4.0×10^{-3}	9.38×10^2	1.32×10^3	6.30×10^2	1.38	5.42×10^2
fission fragments	2.1×10^{-3}	---	3.85×10^4	1.84×10^4	3.68×10^1	1.44×10^4
fast neutrons	1.0×10^1					

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*correspond to one relaxation length.

TABLE III

SOME G(CO)-VALUES FOR RADIOLYSIS OF PURE CARBON DIOXIDE[45,46]

Radiation Type	Pressure, torr	Dose Rate, eV/g s	G(CO)	Steady-State CO Concentrations, ppm
α	40	4.1×10^{19}	7	25%
α	150	1.4×10^{18}	0.09	70
α	60	1.5×10^{18}	0.015	< 1000
α	56	0.5×10^{16}	0.09	< 600
n,γ	56	2×10^{16}	0.005	200
n,γ	128	1.0×10^{16}	< 0.001	5
n,γ	530	1.3×10^{16}	< 0.001	5
γ	29 atm	1.2×10^{16}	< 0.05	100
γ	40-50	4.9×10^{14}	< 0.07	< 100
γ	30	2.3×10^{15}	< 0.0016	20
e^-	6	2×10^{25}	7.4 ± 0.4	---
e^-	400-1500	2×10^{27}	7.8 ± 0.3	---

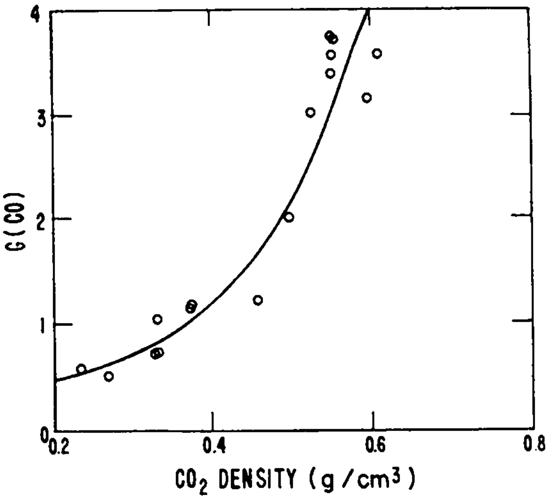

Figure 2.
G(CO) for steady pure CO_2 radiolysis as a function of vapor density at 50 C.[44]

TABLE IV

SOME G(CO)-VALUES FOR RADIOLYSIS OF CARBON DIOXIDE IN THE PRESENCE OF SCAVENGERS[46]

Scavenger	Radiation Type	Temperature, °C	G(CO)-Values
NO_2	α	100	10
NO_2	n,γ	60	3.1±0.2
NO_2	n,γ	140	9
NO_2	fission fragments	70	8.5±0.5
NO_2	fission fragments	240-416	8.2
NO_2	γ	26	4.5±0.4
NO_2	γ	270	1.7
NO_2	γ	20	3.51±0.23
SO_2	protons	25	3.92±0.08
CH_4	γ	17	4.3±0.4
CH_4	γ	200	6.3
CH_4	γ	350	10±0.8
CH_4	γ	500	10±2
CH_4	γ	350	6.5
CH_4	pulsed e^-	25	5.3-B.5
CH_4	fission fragments	220-615	6.6-7.6
H_2	α	20	4.7
H_2	γ	400	4.0
H_2	γ	100	1.7
H_2	e^-	20	0.3

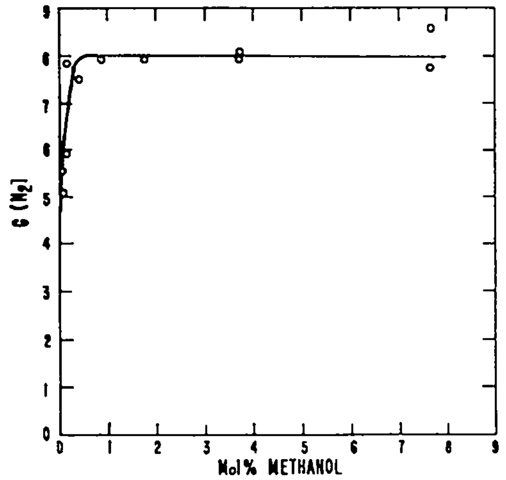

Figure 3.
$G(H_2)$-values for steady, low-dose-rate, x-ray water vapor radiolysis at 25 C and 1 atm as a function of methanol concentration.[26]

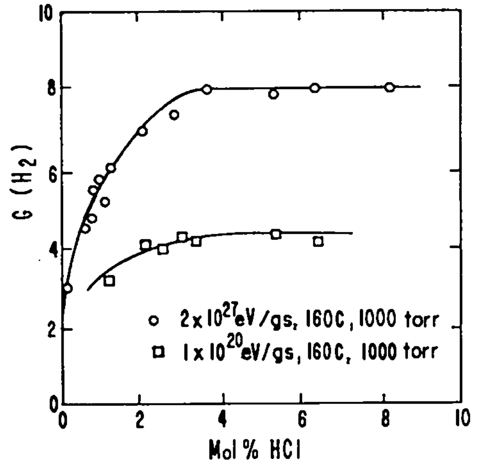

Figure 4.
$G(H_2)$-values as a function of HCl concentration for high-dose-rate, pulsed, accelerated electron radiolysis of water vapor (the data for H_2O-Br radiolysis under the same conditions is very similar to the HCl data).[47]

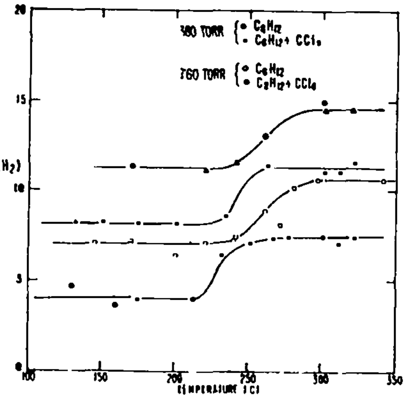

Figure 5.
Yield of hydrogen for steady gamma radiolysis of water vapor with 5 mol% C_6H_{12} and of water vapor with 5 mol% C_6H_{12} and 0.2 mol% CCl_4 as a function of temperature for pressures of 380 torr and 760 torr.[41]

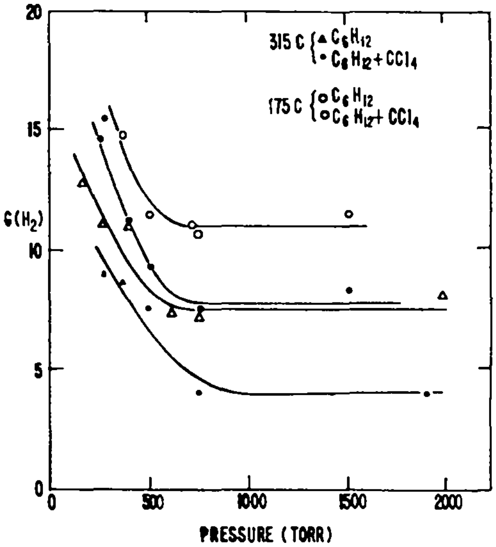

Figure 6.
Effect of pressure on yields of hydrogen at 175 C and 315 C for steady gamma radiolysis of water vapor with 5 mol% C_6H_{12} and for water vapor with 5 mol% C_6H_{12} and 0.2 mol% CCl_4.[41]

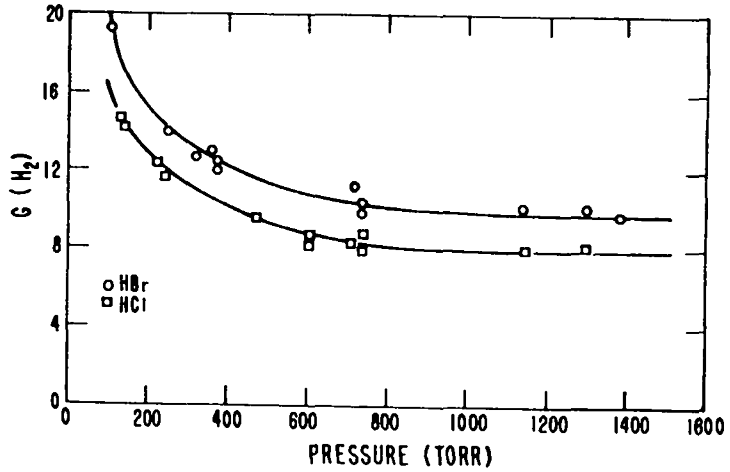

Figure 7.
Effect of pressure on pure hydrogen halide vapor radiolysis at pulsed high dose rate (10^{27} eV/gs).[49]

TABLE V

G-VALUES FOR VAPOR-PHASE RADIOLYSIS OF HCl AND HBr AT 298 K[48-50]

Compound	$G(H_2)$	Radiolysis Efficiency %	Radiation Type
HBr	9.8 ± 2	3.67	gamma and x radiation
	9.9 ± 0.3	3.71	field emission
HCl	8.0 ± 0.3	7.64	gamma and x radiation
	8.0 ± 2	7.64	gamma radiation
	8.1 ± 0.2	7.74	field emission

TABLE VI

NEUTRON ACTIVATION CHARACTERISTICS OF CHLORINE AND BROMINE ISOTOPES

Product or Reactant Species (and Natural Abundance	Potentially Dangerous Radioactive Isotope Produced	Half-Lives of Radioactive Isotope, Years	Type of Radiation (Energy, MeV, Intensity, %)
(1) $_{17}C^{35}$ (75.53%)	$_{17}Cl^{36}$	3.1×10^5	(1) β^- (0.714 MeV, 98.1%) β^+, EC (1.14 MeV, 1.9%)
(2) $_{35}Br^{79}$ (50.54%)	$_{36}Kr^{81}$	2.1×10^5	(2) EC (0.29 MeV, 100%)

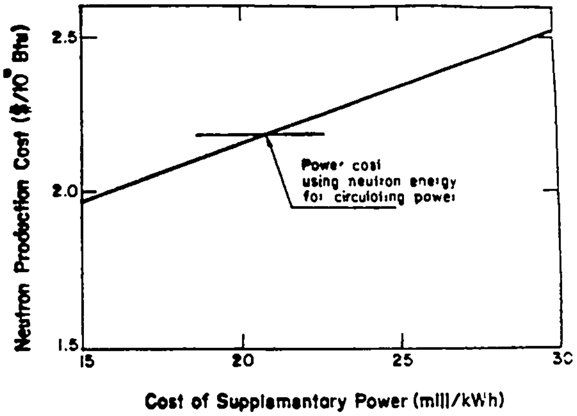

Figure 8.
Neutron energy production cost as a function of cost of supplementary electric power to provide recirculating power.

TABLE VII

COST BREAKDOWN OF LASER FUSION NEUTRON PRODUCTION PLANT

Plant System Parameters	
Number of reactor cavities	4
Energy release/cavity/pulse	100 MJ
Pulse rate/cavity	10 pulses/s
Total thermal power output	4000 MW
Plant power requirement (electric)	742.6 MW
Generating plant conversion efficiency	38.2%
Neutron power output (thermal)	2056 MW
Plant Cost Summary (10^6 $)	
Direct Costs	
Land and land rights	1.0
Structure and site facilities	35.0
Reactor and fuel system	95.8
Laser and beam transport system	88.3
Electric generating plant	67.8
Special laser cooling equipment	33.0
Miscellaneous plant equipment	26.4
Total Direct	347.3
Indirect Costs	
Construction facilities and equipment	17.6
Engineering and management	46.0
Interest during construction (7.5% for 90 mos.)	92.1
Total indirect	155.7
Total Capital Cost	503.0
Production Cost of Neutrons	
Plant duty factor	.84
Annual fixed charge rate (investor owned)	15%
Production cost summary ($/$10^6$ BTU)	
Capital	1.46
Fuel (@ 5 mil/pellet)	.10
Operating and Maintenance	.68
Total Production Cost	2.24

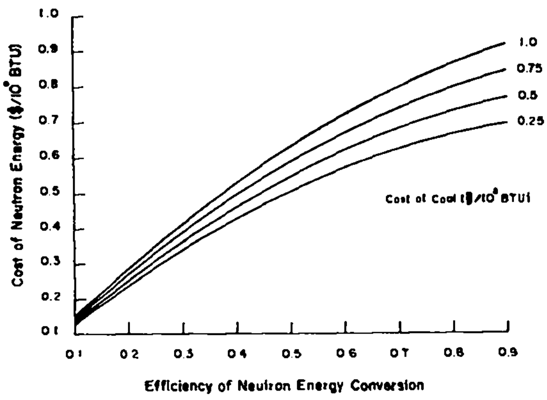

Figure 9.
Cost of 14-MeV neutrons for which the synthetic fuel production cost by direct radiolysis is equal to the synthetic fuel production cost by standard coal-gasification processes.

TABLE VIII

FORD, BACON AND DAVIS, INC., ESTIMATED COSTS OF HYDROGEN PRODUCTION BY FUSION-DRIVEN WATER RADIOLYSIS AND BY VARIOUS ALTERNATIVE PROCESSES

	Production Cost Range	
Method	$/10^9 J	$/10^6 BTU
Electrolysis with electric power from hydroelectric or fossil fuel-fired plants	2.80-11.00	2.95-11.60
Electrolysis with electric power from fission reactors	4.40-5.00	4.64-5.27
Conventional coal gasification	1.20-5.20	1.27-5.49
Natural gas reforming	1.00	1.05
Solar-driven thermochemical cycle	5.00	5.27
Fission-driven thermochemical cycle	1.80-2.00	1.90-2.11
Laser fusion radiolysis	5.00-10.00	5.27-10.55

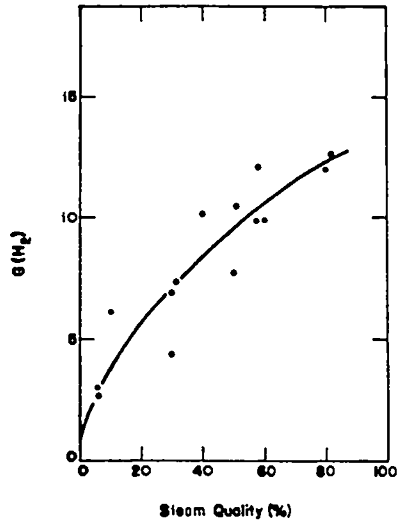

Figure 10.
$G(H_2)$ as a function of steam quality.[62]

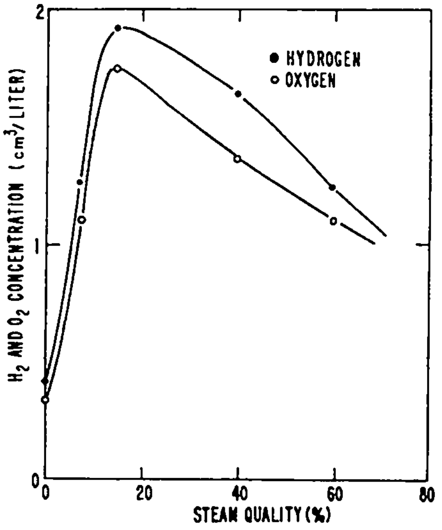

Figure 11.
Steady-state concentrations of hydrogen and oxygen in feed to radiolytic chamber with closed-loop operation.[62]

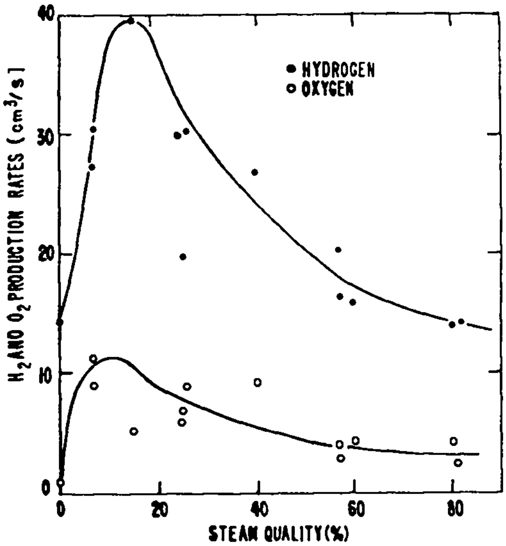

Figure 12.
Hydrogen and oxygen production rates as a function of steam quality.[62]

TABLE IX

CRITICAL PROPERTIES OF SOME HYDROGEN-CONTAINING COMPOUNDS OF INTEREST IN SYNTHETIC FUEL PRODUCTION

Substance	Critical Temperature, C	Critical Pressure, atm
H_2O	374.1	218.3
CO_2	31	72.9
HCl	51.4	82.1
HBr	90	84.5

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