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AUTHOR(S)

R.-D. Penzhorn, Kernforschungszentrum Karlsruhe, Germany

J. L. Anderson, Tritium Science & Technology Group
Materials Science & Technology Division

R. Haange, JET Joint Undertaking, England

B. Hircq, Atomic Energy Commission, France

A. Meikle, Canadian Fusion, Canada

Y. Naruse, Japan Atomic Energy Research Institute, Japan

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TECHNOLOGY AND COMPONENT DEVELOPMENT FOR A CLOSED TRITIUM CYCLE

R.-D. Penzhorn[1], J. Anderson[2], R. Haange[3], B. Hircq[4], A. Meikle[5], Y. Naruse[6],

[1] Kernforschungszentrum Karlsruhe, Institut für Radiochemie/Tritium Laboratorium Karlsruhe, Postfach 3640, 7500 Karlsruhe, Germany; [2] Tritium Systems Test Assembly, Los Alamos National Laboratory, Los Alamos, 87545 New Mexico, U.S.A.; [3] JET Joint Undertaking, Abingdon, Oxfordshire OX14 3EA, United Kingdom; [4] Atomic Energy Commission, Centre d'Etudes de Bruyères-le-Châtel, 91680 Bruyères-le-Châtel, France; [5] Canadian Fusion Fuels Technology Project, Mississauga, Ontario, L5J 1K3 Canada; [6] Japan Atomic Energy Research Institute, Tritium Engineering Laboratory, Tokai-mura, Ibaraki-ken 319-11, Japan

Abstract

A brief summary on recent advances in the field of tritium technology concerning the the most important subsystems of the fuel cycle of a fusion reactor, i.e. the plasma exhaust pumping system, the exhaust gas clean up system, the isotope separation, the tritium storage and the tritium extraction from a blanket is provided. Experimental results, single component developments, and technical tests including those with relevant amounts of tritium that constitute the basis of proposed integral process concepts are described.

1. Introduction

It is of crucial importance for the attainment of tritium self-sufficiency in a fusion reactor operated with D-T fuel that the tritium production in the blanket exceeds the rate of tritium burning in the plasma by at least the amount of tritium lost by radioactive decay and non-radioactive causes. In addition, the tritium needed as a hold-up inventory, which accounts for the time delay between production and use as well as reserve storage must be provided [1, 2]. Within this frame, it is necessary to define the selection criteria for design concepts and the range of acceptable performance parameters of all tritium processing systems, which comprise the plasma exhaust gas pumping system, the exhaust gas clean up system, the isotope separation, the tritium storage, and the tritium extraction from the blanket. For tritium self-sufficiency it is important that non-radioactive tritium losses - for instance by permeation through structural materials into coolants - be minimized, and that the mean residence time of tritium in tritium processing systems as well as the probability of failure and down-time to repair tritium processing units be kept as low as possible. Furthermore, it must be shown that tritium technology once developed and proven can be applied safely, reliably, and economically. Based on these judgments, strong fusion technology related research and development activities, increasingly using substantial amounts of tritium, are under way in Canada, the European Community, Japan, the U.S.A., and the USSR. In addition, much relevant experience will be gained at a large facility in Darlington, in which 2500 grams of tritium have been removed from heavy water in a period of continuous operation that started June 1990.

Because of the large increase in the number of relevant publications and the numerous new activities initiated in recent years, only some of the most recent developments in the field of tritium technology concerning a closed fuel cycle will be discussed in this paper. An attempt, however, will be made to cover all major and advanced experimental facilities designed for demonstration with tritium of components and processes that are commencing design, are under construction, or have already been used in a test program.

2. Plasma exhaust gas pumping

2.1. Process requirements

The plasma vacuum pumping system includes a primary vacuum system and a backing system. It evacuates the torus during all operational and maintenance phases and delivers the resulting gas stream to the fuel processing system at a pressure near atmospheric. The system must provide evacuation under conditions of burn and dwell, pump-down and conditioning. To satisfy all the multiple functional requirements the pumping system will have to include both mechanical and cryotransfer pumps [3]. To meet the burn and dwell cycle duty a helium pumping speed of the order of 400 m^3/s (such as required for the International Thermonuclear Experimental Reactor ITER) will have to be achieved. Options for this purpose are turbomolecular pumps and compound cryopumps together with regeneration pumps. For compound cryopumps two variants are under discussion, i.e. cryotrapping of helium by an argon spray technique or cryosorption of helium on activated charcoal. Advanced pumping systems, which offer a substantial potential saving in equipment are also being considered [4] but due to lack of sufficient test data, these systems will not be discussed in this paper. The pumps of the mechanical train must be available not only for the initial stages of pump-down from atmospheric pressure but also to back the turbomolecular pumps during torus conditioning and the later stages of pump-down. The installed capacity of the mechanical system is largely determined by the helium glow discharge duty.

2.2. Component development

2.2.1. Large roughing pumps

Large mechanical oil-free scroll pumps are proposed to pump air, helium, argon, hydrogen isotopes, etc., with and without impurities, in a number of subsystems such as plasma exhaust pumping, fuel clean-up, tritium extraction from the blanket, isotope separation, waste treatment, etc.

Since only little information was available on the performance of NORMETEX scroll pumps with fusion fuel relevant gasses, pumps with nominal pumping speeds for nitrogen of 15, 18, 60 and 150 m^3/h were tested either with the pure gasses N_2, air, Ne, Ar, CH_4, C_3H_8, He, D_2, and H_2 or their mixtures [5]. The data obtained revealed that whereas air and argon are pumped with the nominal efficiency, other gasses show pumping effciencies decreasing in the order He, deuterium, methane and hydrogen. To investigate the effect of suction pressure (0.1 - 1000 mbar) on throughput (1 - 10^4 [mbarl/s] for H_2, D_2, He. and mixtures of He/H_2, Ar/H_2, N_2/H_2, CO_2/H_2, CH_4/H_2 several pump combinations (including a metal bellows pump) were used. From the data obtained with all the tested scroll pumps it was concluded that for the gasses H_2, D_2, CH_4, and He an exhaust pressure of less than 400 mbar is needed to achieve the pumping speeds specified for nitrogen. The pumping speed curves specified for the 60 and the 150 m^3/h pumps for nitrogen could also be attained with H_2, when this gas contained about 10 % of either Ar, N_2, or CO_2 or about 70 % of He. Compression ratios for nitrogen, helium, methane, deuterium, and hydrogen were measured for all of the above mentioned pumps as a function of the exhaust pressure.

Tests with helium using NORMETEX scroll pumps of 600 and 1300 m^3/h connected in series have also been carried out at the Kernforschungszentrum Karlsruhe (KfK) [6]. Results on the throughput of this pump combination as a function of suction pressures down to 10^{-2} mbar were obtained.

A large oil-free, reciprocating, water cooled, single acting, four stage vertical piston pump has been developed for high level tritium service in Japan [7]. At suction pressures above 13 mbar pumping speeds of approx. 180 m^3/h were achieved with H_2, D_2, He and N_2. At lower suction pressures the pumping speed dropped drastically. The ultimate pressure for N_2 was slightly below 1,3 mbar and that for H_2 was 2.6 mbar. The loss of compression is believed to be caused by backflow of gas through the piston. A reciprocating, tritium compatible pump operated at 1000 mbar discharge pressure was tested in combination with an oil-free magnetic levitated turbomolecular pump at the Tritium Engineering Laboratory (TEL). With this combination it was observed that only N_2 is pumped down to $1.3 \cdot 10^{-4}$ mbar at pumping speeds of ca. 1080 m^3/h. The maximum pumping speed observed for He and D_2 was approx. 360 m^3/h at around $1.3 \cdot 10^{-2}$ mbar. The minimum hydrogen pressure achieved with this pump was approximately 1.3 mbar.

The function of the mechanical forevacuum system at JET is to provide the capability to pump down the torus, the neutral injector boxes or pellet injector boxes and thereafter back up the turbomolecular pumps [8]. Relevant information will be obtained from a corresponding mechanical forevacuum system, composed of one 600 m^3/h and two 150 m^3/h dry NORMETEX scroll pumps presently being installed in Culham. To cope with possible water inleaks into the torus or into the neutral injector box, a water condensor operated at 5 °C has been installed upstream and downstream of the mechanical pumps.

2.2.2. Cryosorption and cryotransfer pumps

From experiments at the Tritium Systems Test Assembly (TSTA) with helium/deuterium mixtures pumped with a single charcoal activated panel kept at 4 K it was observed that the capacity for helium is reduced from 1.8 $mbar \cdot l/cm^2$ to 0.9 $mbar \cdot l/cm^2$, when the amount of deuterium in the pumped mixture is increased from 0 to 33 % deuterium [9, 10]. A farther increase in the amount of deuterium up to 95 % caused only a small additional reduction in capacity to 0.5 $mbar \cdot l/cm^2$. In runs with preloaded deuterium in the range 0 to 13.3 $mbar \cdot l/cm^2$ of the charcoal surface frontal area, the helium capacity decreased steeply from 1.9 to 0.8 $mbar \cdot l/cm^2$ with a preloading of only 1.3 $mbar \cdot l/cm^2$, but leveled off with higher preloadings. At a preload of 13 mbar·l/cm^2 the helium pumping speed decreased to 75 % of that obtained in the reference case without deuterium. It was concluded, that charcoal has a high capacity and speed for helium and requires only minimal bakeout procedures.

For screening test purposes more than 300 combinations of various sorption materials and binders were subjected at KfK to temperature cycles in the range 323/573 K - $\mathrm{lN_2}$. Measurements on the pumping speed and sorption capacity of selected adsorption panels with Ø = 50 mm at 4.2 K yielded specific helium pumping speeds, S, in the range $5 \cdot 10^{-6} < S < 2.3 \cdot 10^{-1}$ l/s·cm^2 measured with a double baffle at feed rates of $Q > 10^{-4}$ mbar·l/s and $p \sim 10^{-4}$ mbar [11]. In general, activated charcoal showed better properties than zeolite 5A. In a facility with cryopanels of Ø = 400 mm the sorption characteristics of activated charcoal was examined with helium and helium containing several impurities at throughputs in the range 0.084 - 8.44 mbar·l/s.

Cryotransfer pumps will be used to collect gases like hydrogen and impurities from a helium carrier gas at JET [8]. By careful temperature control during warm-up pure hydrogen gas will be distilled off directly into the isotope separation system. Residual impurities will be transfered in a second process step into an impurity processing system. The cryotransfer pump at JET is operated at about 4 K. Impurities are released by cycling to room temperature and occasionally to approximately 470 K.

2.2.3. Turbomolecular pumps

A 25 000 l/s single turbomolecular pump designed for an ultimate vacuum of 10^{-7} Pa is under development in Japan and scheduled for tests in late 1991 [12].

3. Plasma exhaust gas clean-up

3.1. Process requirements

The fuel processing system removes the impurities from the "unburned" fuel during burn and dwell to concentrations lower than 1 ppm and recovers tritium from waste gases produced during bake out, glow discharge cleaning, and pump down from air or helium. For tritium economy and environment safety, all tritium bound to other elements in impurities must be recovered to concentrations low enough as to permit release of the produced waste gas via a tritium waste treatment system [3].

Total flow rates for NET II are estimated to 74 mol/h (69 mol/h hydrogen isotopes) during burn & dwell and 16 mol/h (1 % impurities) or 50 mol/h (0.1 % impurities during glow discharge cleaning with deuterium or helium, respectively. Flooding of the reactor with helium will be necessary about once a week. When accidental pressurization of the reactor occurs, the gas composition will be air with a concentration of water determined by the prevailing humidity, which will be contaminated with tritium desorbed from the reactor wall. The total quantity of gas to be pumped off in either case will be of the order of 100 kmol. The total tritium inventory of this subsystem (as of all other subsystems) shall be minimized (for ITER the max. limit is 200 g). The tritium content of all gaseous, liquid and solid impurity streams leaving the plasma clean up system shall also be reduced to a minimum.

3.2. Component development

3.2.1. Permeators

Palladium/silver alloy permeators separate specifically hydrogen isotopes from the fuel ash helium and from hydrogenated as well as non-hydrogenated impurities. Permeators are selected because they are widely used on industrial scale, deliver a hydrogen product stream of very high purity (99.9999%), operate with very low tritium inventory, and produce no process waste. Gases such as N_2, CO (partial pressure up to 1 bar), CO_2, CH_4, NH_3, and H_2O (partial pressure up to 200 mbar) do not influence the rate of hydrogen permeation at temperatures above 300 °C [13].

At the Tritium Engineering Laboratory a palladium/silver permeator of 48 permeation tubes has been tested succesfully at 573 K at a feed pressure of 1 bar and a permeate pressure of 13 mbar, indicating that more than 98 % of the hydrogen isotopes fed into the diffuser permeated to produce a pure product stream. The hydrogen feed rate was varied between 0.5 and 2 l/min, the bleed flow rate was 0.3 l/min. Several 3 - 5 day runs with 1 g of tritium were performed since early 1988 [14].

Experiments with a palladium/silver permeator having a total permeation area of approx. 0.14 m^2 have been carried out at 573 K with hydrogen/helium mixtures at Valduc [15]. The flow rate achievable with this permeator employing pure hydrogen at 2 bar was found to be 0.234 m^3/h. In a series of once-through runs using hydrogen/helium gas mixtures drawn with a pump from a reservoir and compressed into the permeator, the partial pressure of hydrogen served as the driving force. During the operation of the permeator a fraction of the gas from the feed side was continuously removed via a flow rate controller. The results provided information on the balance between the permeation rate (influenced by the blanketing effect) and the percentage of hydrogen carried over with the impurities. Following these tests with hydrogen/helium mixtures, a tritium compatible permeator of 1/5 NET scale has been designed and built.

A modified commercial palladium/silver permeator of 1/8 NET scale (0.12 m^2 effective permeation area), which has been constructed for work with tritium will be tested at the Tritium Laboratory Karlsruhe (TLK) in early 1992 for a period of at least one year, using gas mixtures that simulate the plasma exhaust [16]. Design improvement of the permeator is the result of extensive permeator cold tests in combination with a 150 m^3/h scroll pump and a Metal Bellows pump as well as detailed modeling calculations. The objectives of the experiment are a) to study the effect of decay helium in the membrane on the permeation rate of hydrogen isotopes through palladium/silver and its mechanical integrity and b) to evaluate the long-term resistance of the permeator towards possible poisoning by impurities in the gas mixture. The performance of in-line infrared spectrophotometers, designed for the continuous detection of CO, CO_2, and CH_4 or for the continuous long-term specific analysis of CO (detection limit < 10 ppm) will be examined in view of the development of process control in tritium conducing systems.

3.2.2. Electrolysis cells

Substantial progress has been achieved in recent years in the development of tritium compatible electrolysis cells. These cells may become crucial components of the plasma exhaust cleanup system or the tritium extraction system from a reactor blanket.

Several ceramic electrolysis cells that operate at elevated temperature with water vapour have been designed, manufactured and tested by JAERI scientists. The cell presently installed in a loop at TSTA contains ten sintered stabilized zirconia tubes with calcined platinum electrodes at the inside and outside surfaces [14]. Water vapour in the feed gas is decomposed at the inner surface of the electrode to form hydrogen/tritium, pure oxygen evolves from the outer electrode surface. From measurements on the conversion ratio as a function of the electrochemical potential across the cell a conversion efficiency lower than expected was observed. Special attention was focused on the electrolysis of impurities such as CO and CO_2. At the potential at which the cell is usually operated carbon dioxide electrolyzed to carbon and oxygen with a conversion of only 3 %. The electrolysis of water on the other hand is not influenced by the presence of this gas. Long-term tests at 913 K with $T_2O/CO_2/N_2$ at a carrier throughput of 0.4 - 1.0 l/min have been carried out succesfully.

An electrolysis cell with a hollow palladium/silver cathode coated with porous palladium black has been developed at Valduc and tested successfully with tritium with up to 1.2 x 10^{13} Bq/ml of water [17]. The cell uses NaOH as electrolyte and is operated at 353 K. Experiments with 3.7 x 10^{13} Bq tritium per ml water are forseen in 1991.

A low inventory capillary electrolyzer (ALICE) for the recovery of tritium from tritiated water with up to 3.7 x 10^{13} Bq/ml has been developed by SKC/CEN at Mol in Belgium [18]. Sulfuric acid has been chosen as electrolyte, thermoelectric heat pumps cool the cell to 281 K. With a prototype cell up to 3000 hours of continuous operation have been accumulated. Because of the high radiotoxicity of tritium and the need to work at constant tritium inventory a closed loop has been built that allows recombination of the electrolytically produced gases and their recirculation into the cell. To test and demonstrate the performance of the cell two experiments, each with a duration of 1000 h, will be carried out at Chalk River Laboratories in 1991.

3.2.3. Getter beds

Metal getters react with hydrogen isotopes containing impurities at elevated temperatures to give stable products and recoverable hydrogen.

Getter beds containing uranium metal powder or iron metal powder have been developed at JET to recover hydrogen isotopes from tritiated water by reacting to metal oxide and uranium hydride [19]. The beds are heated by two independent heating coils to operate between 290 and approximately 770 K (design temperature 770 K) and cooled by a separate gas cooling loop. The beds have sintered inconel tubular filters at the inlet and outlet to prevent dust migration, and are housed within an intermediate container to minimize heat and permeated tritium losses.

A getter bed containing 1.5 kg of uranium powder has been demonstrated for the recovery of tritium from HTO released during the thermal reactivation of molecular sieves [20]. For the investigation of the reaction of uranium with other tritiated impurities a uranium powder bed of 200 g designed for a max. temperature of 650 °C was employed. Series of tests with H_2/CH_4, H_2/NO, and $H_2/NO/CH_4$ at several temperatures up to 878 K give evidence that a high degree of purification can be achieved with uranium at temperatures higher than 973 K. Q_2/NO and Q_2/NH_3 (Q denoting H, D, T) as well as more complex mixtures containing in addition some carbon oxides were found to undergo radiolytical reactions within hours leading to changes of the original chemical composition. Experiments in another reactor at 963 K demonstrated that with inlet concentrations of methane in hydrogen in the range 2.3 - 2.7 vol. % outlet concentrations lower than 0.26 vol. % can be achieved in a once-through modus. The detailed design of a new high temperature getter bed is under way.

After successful tests with gas mixtures simulating TLK impurity streams employing three getters beds operated at 773 K (retention of oxygen and oxygenated compounds), at 973 K (retention of nitrogen and ammonia) and at 1173 K (retention of hydrocarbons) in series a Clean-up System for the TLK was developed [21]. For the second and third getter stage of the system now under constuction alumina ceramic vessels are used to contain the uranium.

The applicability of two commercial gas purifiers in series for the removal of carbon oxides, oxygen, and water at about 523 K and of nitrogen and hydrocarbons at temperatures above 673 K from a helium/hydrogen isotopes stream is under investigation in a technical facility to be operated with tritium [22].

3.2.4. Adsorber beds

In TSTA a bed with 1.6 kg of 5A molecular sieve at 77 K was used to remove impurities from a hydrogen gas stream containing 1 % impurities (90 % nitrogen and 10 % methane) [23]. The concentration of both impurities could be reduced to levels below the analytical detection limits. The average quantity of impurity adsorbed was 100.8 l/kg molecular sieve, corresponding to 84 % of the theoretical capacity.

Adsorption studies with deuterium/impurity mixtures are in progress at Bruyères-le-Châtel, employing a cryotrap that contains 128 g of type 5A zeolite activated at 498 - 573 K to a residual water content of 3 % by weight [20]. Impurities such as N_2, O_2, CD_4, CO_x and Ar are removed from deuterium carrier gas to levels below 10 vpm at temperatures below 113 K. Table I shows the effect of temperature on the cotrapping of deuterium in the molecular sieve. Evidently, at temperatures at which efficient removal of CD_4, N_2, O_2, and Ar from deuterium is achieved, i.e.: < 113 K, the cotrapped deuterium inventory becomes substantial. The residual water in the zeolite undergoes a slow exchange reaction with DT and T_2. The exchange rate constant was found to be directly proportional to the concentration of tritium.

Table I Cryotrapping of D_2 and impurities on zeolite 5A

Temperature	cryotrapping	
	deuterium + impurities	deuterium
K	ml(STP)/g	ml(STP)/g
153	32.8	9.2
133	35.4	11.8
113	43.1	19.9
103	58.5	34.9
93	82.5	59.5

At KfK the adsorption in zeolite 5A of hydrogen isotopes from mixtures with helium is under investigation at temperatures down to 53 K employing a cryostat integrated into a tritium compatible loop. The residual water in the zeolite can be reduced to levels below 0.5 % by weight without removal of the adsorption cell by heating the cell up to 723 K under vacuum [24].

3.2.5. Combined permeator-catalysis reactor

Catalytic reactions based on the establishment of thermodynamic equilibrium with formation of hydrogen isotopes as one of the products, i.e.

$$CH_4 = C + 2\,H_2 \quad \text{or}$$

$$CO + H_2O = CO_2 + H_2$$

could be influenced with a shift of the equilibrium towards the products (quantitative conversion) by combination of the catalytic reactor with a membrane selectively permeable for hydrogen. Experimental work for an evaluation of this option is being pursued at TSTA and in Karlsruhe.

3.3. Integral process concepts

In the following some of the integral process concepts under discussion or test will be briefly described.

3.3.1. Catalysis/permeator

For the processing of the primary exhaust gases of a fusion reactor during burn & dwell, glow discharge cleaning, bake out, and pump down (air or helium) a concept based on catalytic reactions combined with palladium/silver permeators has been proposed and an engineering design for NET II carried out [25].

The plasma exhaust gases from burn & dwell, and helium/deuterium glow discharge are first fed into a permeator of approx. 1 m^2 total permeation area where 99.85 % of the hydrogen isotopes are extracted by pumping the secondary side of the permeator with a 600 m^3/h scroll pump backed up by an oil-free metal bellows booster pump. Because the separated hydrogen isotopes are of high purity they can be fed directly into the isotope separation system. The effluent stream from the permeator, containing tritiated impurities (mainly hydrocarbons, residual hydrogen isotopes, and water) and non-tritiated impurities (noble gases, carbon oxides, nitrogen) are collected in a tank and processed in a semi batch way in two subsequent catalytic reaction steps: a) on a nickel catalyst at 773 K hydrocarbons and ammonia are decomposed into the elements up to thermodynamic equilibrium and b) on a copper chromite catalyst at about 473 K tritium containing water is converted by reaction with carbon monoxide into molecular hydrogen isotopes and carbon dioxide via the "water gas shift" reaction. The hydrogen isotopes liberated from these catalytic reactions are extracted with a second permeator integrated into the catalytic loop. To achieve a high degree of tritium recovery (and decontamination) the second permeator is backed up by a turbomolecular pump. Depending upon the partial pressure of water carbon dioxide may be added to the gas stream. This gas reacts with the carbon deposit on the nickel catalyst to produce the carbon monoxide needed for the water gas shift reaction, while at the same time increasing the life-time of the catalyst.

Pump down exhaust gas from torus flooding with helium or from air break-in is passed over a recombiner to oxidize molecular hydrogen isotopes and hydrocarbons into water and carbon dioxide (when the process gas is helium oxygen needs to be added). Water is first retained in a cryotrap and then volatilized with carbon monoxide as the carrier gas. The resulting gaseous mixture is passed through a copper chromite bed where the water is converted into hydrogen. The hydrogen is extracted by permeation and the bleed gas is sent once-through via the impurity processing loop into the tritium waste system for ultimate detritiation.

Every single step of this process has been studied in the laboratory. The kinetics and reaction mechanism of the catalytic decomposition of methane, ethylene, acetylene and other higher hydrocarbons has been studied in detail. Integral tests with relevant amounts of tritium have been carried out at TSTA [26]. The water gas shift reaction with CO as carrier gas has been demonstrated in a technical scale (attained conversion: 1.5 l of liquid water per day). A contract has been awarded to industry for the design, manufacture, assembly and commissioning of an experimental facility having 1/8 NET II scale to be installed in the TLK for the demonstration of the process.

3.3.2. Molecular sieves/palladium membrane reactor [23]

After the separation of the "unburned" DT fuel, the remaining impurity stream containing a small fraction of DT is first routed through a noble metal catalyst bed at 450 K in which oxygen is reduced into water. The hot gas leaving the recombiner is then cooled to room temperature before being passed through a molecular sieve bed kept at room temperature. In this bed water and ammonia are retained. Downstream the process gas passes through another molecular sieve bed cooled to 77 K, in which all other impurities, i.e. N_2, CH_4, and CO, but no He are trapped. To minimize DT cotrapping, the l N_2 cooled molecular sieve bed is preloaded with hydrogen (the impurities will displace the hydrogen and load completely the bed). To smooth the process three trains of molecular sieve beds will be used: one in operation, one in regeneration and one in stand by. The effluent gas from the molecular sieve beds is send to the cryogenic isotope separation system after passing through a last guard bed.

To process the impurities and some cosorbed DT, the room temperature and the l N_2 cooled beds are regenerated at 623 K and 400 K, respectively and the released gases directed to a palladium membrane reactor containing suitable catalysts. Hydrogen isotopes liberated in the reactor from a) the decomposition of hydrocarbons, b) the water gas shift reaction and c) the decomposition of ammonia (see 3.2.6.) are directly extracted by permeation and sent to the isotope separation system. Due to the continuous removal of hydrogen all chemical equilibria are shifted in the favourable direction. As a result very high conversions are achieved.

3.3.3. Hitex process

Recently a novel process concept for recovering tritium from the impurities in the fusion reactor exhaust stream has been proposed [27]. After the removal of the bulk of the "unburned" D,T in a first permeator, a batch of the impurity gases (tritiated and untritiated) with some D,T carry-over is collected in a previously evacuated vessel up to a preset pressure and then swamped with protium up to a pressure of 100 kPa. The resulting gas mixture is circulated trough a high temperature exchange (HITEX) reactor where isotopic equilibration between hydrogen and the impurity molecules takes place. The reactor consists of a horizontal tube with an axially coiled Pt hot wire operated at a temperature in the range 700-1100 K, depending on the amount of methane present. The hot wire is prevented from coming into contact with the wall of the reactor by suitably positioned spacers. To minimize tritium permeation, the walls are cooled to 350 K with a water jacket. The average temperature of the reactor is estimated to be 500 K. Tritiated hydrogen is extracted from the loop with the palladium/silver permeator S1. To compensate for the extracted hydrogen isotopes hydrogen is fed into the loop until the quantity added is equivalent to 20 volume changes of the buffer vessel. In this way the initial tritium content is reduced by a factor of the order of 10^{-9} and can be discharged into a conventional detritiation system.

Since the size of the isotope separation is determined by the waste water detritiation stream and the pellet injector propellant clean-up stream, the impact of the HITEX hydrogen isotope stream appears to be comparatively small.

3.3.4. Permeation/oxydation/electrolysis

Two concepts based on the combination of permeation, impurity oxidation and electrolysis as main process steps have developed.

At JAERI a fuel clean-up system was built that has been used with up to 1 g of tritium [28]. The first component is a precious metal diffuser to extract pure hydrogen isotopes from the process stream. The bleed gas from the diffuser is then routed through a catalytic reactor in which all impurities are oxidized to yield tritiated water. Oxygen required for the oxidation of the tritiated species is supplied by an electrolysis cell or from external sources. The exhaust gas from the catalytic reactor is passed through a cold trap held at 160 K. This trap, which retains only tritiated water, is periodically regenerated at 343 K. The vaporized water is decomposed in a ceramic electrolysis cell. Based on results obtained with this equipment a complete fuel clean up facility with a throughput of 1/5 ITER has been developed and designed by JAERI. The facility was installed in TSTA in early 1990 and is now being tested under cold conditions.

According to another concept [29] the gas from the torus is first submitted to a pretreatment, i.e. catalytic conversion of oxygen into water and removal of this species by condensation, to preclude the formation of nitrogen oxides. The "unburned" fuel is then extracted at a pressure of 2 bar with a permeator operated at 573 K. The bleed flow from the permeator is reduced to atmospheric pressure and passed through a nickel catalyst reactor for the cracking of ammonia. The hydrogen carried over with the impurities is oxidized catalytically mainly to protect the down stream catalyst (avoidance of hot spots) used for the oxidation of hydrocarbons. The oxidation of the hydrocarbons up to a conversion of 90 % occurs on an alumina supported palladium catalyst at 573 K. All oxidation reactions require the supply of an oxygen stream. The large number of single steps sofisticated control instrumentation mandatory. All water produced is collected by gravity and decomposed by electrolysis.

4. Isotope separation

The isotope separation system (ISS) separates molecular hydrogen isotopes into an enriched tritium stream for fuel preparation, a detritiated deuterium stream for recycling into the neutral beam injector and fuel preparation, and a detritiated hydrogen stream for pellet injector propellant or discharge into the environment.

4.1. Process requirements

To meet single event release constraints the ISS must be designed to have less than a given limited tritium inventory (for ITER the max. inventory shall be limited to 200 g). Normally the ISS will operate at pressures slightly above atmospheric and will be connected to an expansion tank to maintain the systems internal pressure at a reasonable level in case of loss of cooling [3].

4.2. Component development

4.2.1. Isotope separation by distillation

Cryogenic distillation using isotope hydrogens including up to 1.5 g of tritium have been carried out with two columns of different diameter and packing size [30]. HETP values (Height Equivalent to a Theoretical Plate) were correlated with the feed flow rates and vapour streams of the column as well as with the reflux ratio.

The ISS at TSTA has been operated for an extended period of time with a tritium inventory of approx. 100 g. Recently, the operation of two cryogenic columns with D-T mixtures and of four cryogenic columns with D-T mixtures containing impurities such as He and H_2 was investigated [31]. Observed and calculated mole fractions showed good agreement except for the concentration of HT and DT, which were found to be higher than the calculated values. HETP values of 4.5 - 6 cm were obtained independent of vapor velocities, when varied in the range 4 - 14 cm/s. The results lead to the conclusion that a helium concentration of 27 % is still acceptable for a normal operation of the columns.

Large cryogenic distillation columns have been operated succesfully at Ontario Hydro's Darlington Tritium Removal Facility. Integrated operation has been demonstrated and over 2500 g of > 99. % tritium gas has been extracted from D_2O containing (5.6 - 7.4) x 10^{11} Bq/kg.

4.2.2. Isotope separation by gas chromatography

For the separation of isotope hydrogens a displacement gas chromatograph [32] with a throughput of 20 mol/d, having four columns, each containing 5 l of a packing consisting of palladium on alumina has been installed at JET. The method is based on the strong isotope effect of the Pd-Q (Q = H, D, T) system in the β-phase region of the phase diagramm. Gas chromatography will be used in JET when the torus is operated with two neutral beam injectors at a maximum daily throughput of 5 moles T_2 and 15 moles D_2. Displacement gas chromatography has been succesfully demonstrated in extended use with large amounts of tritium [33].

4.3. Isotope separation systems [34]

A conceptual design description for an integrated ISS for ITER based on the requirements for a) plasma exhaust gas processing (71.4 mol/h with 1 % H, 49.5 %D and 49.5 % T; in case of HITEX clean up 457 mol/h with 99.564 % H, 0.218 % D and 0.218 % T), b) neutral beam injection deuterium clean up (378 mol/h with 0.03 % H, 98.47 % D and 1.5 % T), waste water detritiation (200 kg/h with 0.1 Ci/kg) and tritium recovery from a solid breeder blanket (105 mol/h with 99 % H and 1 % T) has been developed in a collaboration between CFFTP, Ontario Hydro (FLOWSHEET process simulator), the NET/ITER teams, and TSTA (dynamic cryogenic distillation code). The ISS products are detritiated water (with less than 1 lCi/kg of tritium), a protium stream (with a tritium atom fraction of $< 10^{-9}$), a deuterium stream (with less than 10^{-7} % T and 0.2 % H), and a tritium stream (with 80 % T, 20 % D and less than 10 ppm H).

The ISS comprises a waste water detritiation front-end and a cryogenic distillation. Two water detritiation options were considered, i.e. water distillation or CECE (combination of two liquid phase catalytic exchange columns with an electrolyser).

The components comprising the ISS are the most complex and expensive of all tritium processing systems.

5. Tritium storage

5.1. Process requirements

The general functional requirement of the storage system is to provide safe storage for all hydrogen isotopes during startup, rundown, and shutdown operations under normal and abnormal conditions [35]. Fuel storage has to satisfy the following functional requirements (based on ITER): a) storage of 2.4 kg of T_2 as purified DT after a normal shut down, b) storage of 10 600 moles of non-purified hydrogen isotopes after an unscheduled or abnormal shut-down, c) provide surge storage for the in-process tritiated gases and to receive and deliver at certain rates, and d) a make-up storage capacity for 1 kg of T_2 as DT, 3 kg of D_2 and 1.5 kg of H_2. As for other subsystems, the total amount of tritium releasable in a single initiating event must not exceed a certain max. value (for ITER 20.0 g).

Hydrogen isotopes may be stored in buffer tanks or as hydride (or deuteride or tritide) of metals or metal alloys.

5.1.1. Properties of uranium tritide and of alternative tritide forming intermetallics

Uranium hydride has been extensively studied in view of its use as hydrogen storage system and been amply demonstrated for the storage of tritium. In the JET Active Gas Handling System 16 uranium beds will be employed, 12 (for storage of isotope mixtures and pure D_2) contain 18 mols each of depleted activated U-238 (max. storage capacity 27 moles of hydrogen per bed) and 4 (for storage of pure T_2) contain 5 moles each of U-238, giving a max. storage capacity of 7.5 moles T2 per bed. Activated uranium is a fine powder, which reacts exothermically with oxygen to form uranium oxydes (ca. 850 kJ/mol O_2) and with nitrogen to form uranium nitrides (ca. 600 kJ/mol N_2). To assess safety hazards for the highly unlikely case of air ingress, only possible in case of multiple faults, a fully activated uranium bed was exposed to ambient air under controlled conditions [36]. The results indicate that not more than 30 bar·l of air are absorbed by the bed, the temperature increasing to only ca. 413 K. The reaction does not proceed to completion because of surface passivation and, to some extent, because of a blanketing effect caused by the continuous argon partial pressure increase in the vessel (the mol fraction % of argon in air is 0.93). Because less than 2 % of the helium-3 produced during an aging period of 280 days is released from stoichiometric uranium tritide (the release increases dramatically afterwards approaching the generation rate after about 1000 days) a pressureless tritium storage is possible over an extended period of time [37].

While uranium has been demonstrated as a tritium storage material there is continuing interest in the development of alternative getters having less chemical reactivity towards air and other gasses and no legal restrictions due to nuclear character.

ZrCo was shown to have adequate properties for the interim storage and transport of tritium [38, 39]. This intermetallic compound/hydrogen isotope system has been exensively characterized with respect to storage capacity, pressure concentration equilibria, recoverability of isotope hydrogens, effect of impurities, kinetics of hydrogen absorption, etc. [40]. An experimental facility for aging studies of ZrCo tritides is under construction in the TLK. Several ZrCo tritium storage beds have been built and tested succesfully with tritium by JAERI at the TPL and at TSTA [41].

In view of screening tests to select the most adequate material for tritium storage with several $LaNi_{5-x}Mn_xH_y$ ($0.5 < x < 2$) alloys, that indicated that the lowest equilibrium pressure is obtained for $x = 2$, the pressure-composition isotherms for the systems $LaNi_3Mn_2$-H_2, $LaNi_3Mn_2$-D_2, and $LaNi_3Mn_2$-T_2 were determined [42]. Another selection criterion is the loading capacity at a given pressure, which was shown to decrease with increasing x [33]. Work at Bruyères-le-Châtel therefore concentrated on $LaNi_{5-x}Mn_x$ for $0.5 < x < 1.5$. Experiments were carried out to investigate the kinetics with hydrogen isotopes as a function of grain size, the blanketing effect, and the thermal conductivity as a function of stoichiometry. Aging studies at room temperature covering a period of one year revealed that less than 1 % of the decay helium is released during this time.

5.1.2. Technical storage vessels and storage facilities

The concept design and construction of a storage unit comprising five storage modules in a double containment for storing up to 100 g of tritium (DT equimolar form) has been completed and installed in a glove box at Bruyères-le-Châtel [20, 33]. According to this concept the five storage modules are connected to a central thermal exchanger. The compound used for tritium storage is placed inside of the module in a porous containment of conical configuration. Testing of the facility is scheduled for 1991.

Based on the getter material preselected by the ITER Fuel Cycle design team, i.e. ZrCo, and the given process requirements a conceptual design description for a Fuel Storage System and the corresponding Fuel Management System was developed [43]. According to the concept, for the storage of DT 28 beds, each having a max. storage capacity of 60 mols of hydrogen isotopes, and for the storage of D_2/H_2, 18 beds each having a max. storage capacity of 2000 mols requiring a total of 4 t ZrCo will be needed.

6. Tritium extraction from a blanket

An ITER/NET size blanket, regardless of the selected concept i.e.: helium or water cooled solid ceramic blanket, aqueous liquid salt blanket, or lithium/lead blanket, is planned to breed 125 g/d of tritium. To maintain the desired tritium inventory constant a Tritium Extraction System (TES) must be designed capable of removing tritium at an average rate equivalent to the tritium breeding rate. For the case of a helium purged and helium cooled blanket with a lithium ceramic breeding material and a beryllium multiplier tritium is also bred within the beryllium zone, i.e. 1.52 g/d, of which a fraction of 14 % is released. Therefore an additional purging system must be included if the bryllium zone is physically separated from the lithium ceramic zone.

Some basic design requirements concerning the purge gases of the lithium ceramic zone and (eventually) the beryllium zone have been compiled in Table II [44].

The design requirements stipulate that the breeder material in the blanket is to be maintained at temperatures in the range 723 - 873 K and shall be purged with helium at 1.5-2.3 bar, containing 0.1 % hydrogen. The anticipated swamping ratio is $H/T = 100$. The blanket tritium recovery system shall produce a stream of dry hydrogen with a dew point of $T < 173$ K ($< 0.014\,\mu$bar H_2O) and contain less than 1 ppm total impurities, so that it can be fed directly into the Isotope Separation System.

In this paper only TES's proposed for solid ceramic blankets will be discussed.

2.5.1. Cryoadsorption/Permeation/Electrolysis Concept [45, 46]

One water cooled ceramic blanket option presently under discussion consists of water/beryllium/lithium zones paralell to the first wall, which are isolated from each other by stainless steel clads. According to this option, water in-leakage is only possible into the beryllium zone, because only this zone is immediately adjacent to the cooling water zone.

Under the assumption, that most of the tritium in the purge gas is in the molecular hydrogen form, a tritium extraction process concept based essentially on cryosorption, permeation, and water electrolysis has been developed. In a first step water (H_2O/HTO) is removed employing a cold trap at 173 K. The other tritium product, i.e.: H_2/HT, is separated from the helium purge gas together with all other tritiated and non-tritiated impurities and about 1 % of the helium by sorption on a down stream placed molecular sieve bed cooled to liquid nitrogen temperature (77 K).

The molecular sieve bed needs to be regenerated periodically. This is carried out sequentially employing an appropriate temperature program: a) to recover isotop hydrogens the beds are cycled between 77 and 173 K every two hours using warm H_2 gas to assist the desorption, b) to desorb impurities like O_2, N_2, CO, and noble gases the adsorber bed is heated about once a week to 298 K while purging with hydrogen, and c) to liberate water and carbon dioxide the molecular sieve is heated up to 523 K.

Table II Process requirements: Composition, flows, and other parameters of the purge gas through the lithium ceramic zone and the beryllium zone of a solid breeder blanket [45].

Ceramic blanket purge gas	inlet	outlet
Temperature K	723-873	723-873
Pressure [bar]	1.5-2.3	1.2
He flow [kmol/d]	2815	2815
Q_2 flow [kmol/d]*	2.815	2.815
Q_2O flow [mol/d]	2.818	28.01
Total impurities [mol/d]**	0.120	1.20
Total tritium flow [kCi/d]	166.2	1655

Beryllium zone purge gas	inlet	outlet
Temperature K	723	723
Pressure [bar]	1.5-2.3	1.2
He flow [kmol/d]	3.24	3.24
Q_2 flow [mol/d]	0.0532	0.532
Q_2O flow [mol/d]	0.00324	555.6
Total impurities [mol/d]*	0.000176	0.0499
Total tritium flow [Ci/d]	3.988	3247.2

* Q T, H

** Impurities: Ar, N_2, O_2, CO, CO_2, Kr, Xe

The isotop hydrogens recovered from the molecular sieve bed in high concentration are removed batchwise and with high efficiency with a palladium/silver permeator of comparatively small permeation area. The permeated tritiated hydrogen has a purity of 99.9999 %, it is fed directly into the isotope separation system.

To prevent that impurities arising from a possible leakage through one of the permeation tubes clog the cryogenic isotope separation system, a hot metal guard getter bed is installed between the permeator and the cryogenic columns.

Impurities, including those containing tritium, are sent to an interfacing impurity processing system. Water collected in the cooler is recovered periodically and decomposed into the elements hydrogen and oxygen using an electrolysis cell (options: wet electrolysis cell, solid polymer cell or high temperature solid oxide cell). While the electrolytical hydrogen is recycled into the TES, the oxygen is sent to the waste detritiation system.

Tritiated water produced from the oxidation of impurities such as ammonia and/or hydrocarbons in the waste disposal system is processed electrolytically together with the water collected in the cold trap.

Basically the same process steps as employed for the extraction of tritium from the blanket are used to extract tritium from the beryllium zones. The only important difference is that in this loop a larger electrolysis cell capacity is available to cope with possible water in-leakages.

2.5.2. Freezer/Adsorption Process Concept [47]

This concept assumes no physical separation between the breeder zone and the beryllium zone. The purge helium from the breeder ($2.4\ 10^5$ - $3.6\ 10^5$ mol/d) containing tritium as tritiated water and tritiated hydrogen is first cooled down by counter current to room temperature and then compressed to 10 bar. A process pressure of 10 bar is used to optimize the freezing operations and to reduce the size of the components.

After removal of the compression heat the gas is passed through a prefreezer, in which essentially all the water is frozen out. The water partial pressure is further reduced down to a dew point of 153 K employing a freezer panel cooled to 150 K with liquid nitrogen. The water freezer is regenerated every 24 hours. To recover the tritiated water (1 kCi/g) in the prefreezer panels, the panels are warmed up and the vaporized gases sent via a degassing column into an electrolyser. Hydrogen and oxygen produced from the electrolysis of water (about 21 mol/d throughput) are directly returned into the loop and compressed together with the helium purge gas coming from the breeder. In this way, drying of both gases is circumvented and oxygen is removed together with the other impurities by condensation in the cryoadsorption trap (see below).

Low boiling gases, such as N_2, O_2, CO, CO_2, CH_4, and residual H_2O, present as impurities in the purge gas (total impurities < 1.2 mol/d) are removed by adsorption at liquid nitrogen temperature (75 K). Regeneration of the adsorbers is carried out every two weeks at 423 - 473 K.

Hydrogen (22.8 mol/h) is separated from the purge gas helium in a freezer panel cooled to T < 10 K with liquid helium. The clean purge gas is returned through several counter current heat exchangers to the breeder. The hydrogen recovered from the freezer, which contains only helium as a minor impurity, can be fed directly into the isotope separation system.

2.5.3. Tritium extraction system for a NET blanket test plug [48]

According to the TES proposed for the NET helium cooled solid breeder blanket test plug the helium off-gas (6.5 mol/d with a total pressure of approx. 1 bar, containing 0.1 vol. % H_2) carries tritiated hydrogen and water. H_2O/HTO is removed in a large 31.6 kg 3A molecular sieve adsorber bed held at T < 313 K (max. sorption capacity at p_{HTO} = 0.133 Pa is 0.02 g_{HTO}/g_{MS}, which gives rise to a regeneration time = 1 a). After the removal of water the He is recirculated through a palladium/silver permeator of 6.3 m^2 total permeation area, which reduces the partial pressure of hydrogen from 100 Pa down to 1 Pa. The permeated hydrogen isotopes are collected as metal hydrides in a getter bed containing uranium (regeneration period of the getter bed = 4.2 d). The loop on the secondary side of the permeator contains helium recirculated with an oil-free pump.

Impurities in the helium purge gas, in particular nitrogen, are removed by adsorption on 33.1 kg charcoal cooled down to 96 K (loading capacity at p_{N2} = 0.1 Pa is 0.02 g_{N2}/g_C, regeneration period = 1 a).

7. Conclusions

Based on laboratory and technical scale experiments feasible design concepts have been developed for all fusion reactor fuel cycle subsystems. Further work is needed to optimize single process steps and to demonstrate the operation of integral facilities. Of crucial importance are steps towards further reduction of the tritium inventory in the various subsystems. Research activities in the future should concentrate on the demontration with relevant amounts of tritium of the safe and reliable operation of prototype components and prototype facilities.

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