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FUEL CLEANUP SYSTEM FOR THE TRITIUM SYSTEMS TEST ASSEMBLY: DESIGN AND EXPERIMENTS

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ABSTRACT

A major subsystem of the Tritium Systems Test Assembly is the Fuel Cleanup System (FCU) whose functions are to: (1) remove impurities in the form of argon and tritiated methane, water, and ammonia from the reactor exhaust stream and (2) recover tritium for reuse from the tritiated impurities. To do this, a hybrid cleanup system has been designed which utilizes and will test concurrently two differing technologies - one based on disposable, hot metal (U and Ti) getter beds and a second based on regenerable cryogenic adsorption beds followed by catalytic oxidation of impurities to DTO and stackable gases and freezeout of the resultant DTO to recover essentially all tritium for reuse.

INTRODUCTION

The Tritium Systems Test Assembly (TSTA) is under construction at Los Alamos Scientific Laboratory (LASL) to develop and demonstrate the D-T fuel handling technology needed for commercial fusion power reactors. A major subsystem is the Fuel Cleanup System (FCU). The main functions of the FCU are twofold:

1. to remove accumulated impurities from the reactor exhaust gases and the neutral beam line gases, and
2. to recover tritium from these impurities for reuse in the fuel cycle and to keep tritium out of the environment.

Although the species and concentrations of impurities in reactor exhaust gases are not known with certainty, the best current estimates have led to the design quantities shown in Table I.

No helium is shown in the feed to the FCU because helium is separated from the impure reactor exhaust gases by operation of the compound cryopump at the reactor. This pump collects helium on one adsorbent surface at 4 K and collects all other species on another surface. Experiments have shown this separation of helium is easily, almost unavoidably, maintained during regeneration of the cryopump surface (1).

The purity requirements for the effluent fuel stream from the FCU are determined not by the requirements of fusion fuels, but rather by the needs of the cryogenic fractional distillation system (Isotope Separation System) which processes the cleaned fuel stream from FCU. Experience has shown that impurity levels must be maintained below 1 ppm to ensure safe, long-term operation of such equipment without blockages or hazardous accumulation of impurities (2).

PROCESS SELECTION

A wide variety of process alternatives are available for meeting the needs outlined above. However, none has been proven in a continuous operating mode with tritium. The chief alternatives given consideration included the following process elements: freezeout, cryogenic adsorption, catalytic oxidation, electrolysis, activated metal getters, and palladium diffusers.

The use of palladium diffusers was eliminated fairly early from further consideration. Their disadvantages were the need for elevated pressures, reported brittle failures during temperature cycling, reported poisoning by ammonia and methane, and the fact that, although diffusers can produce a hydrogen isotope stream free of impurities, they cannot produce an impurity stream free of hydrogen isotopes.

Two basic process schemes were given intensive evaluation. The first was a scheme based on adsorption of impurities on molecular sieve beds at 75 K, followed by catalytic oxidation of recovered impurities to form tritiated water and tritium-free compounds, freezeout of the water, and electrolysis to recover tritium. The second was based on hot uranium getter beds operated at 1170 K to remove impurities as uranium oxides, carbides, and nitrides. Both schemes have advantages and disadvantages and, in the end, both will be used at TSTA. Table II summarizes the relative strengths and weaknesses of the two schemes.

The cryogenic process at this time appears to be the one ultimately preferred. However, the lack of a proven HTO electrolyzer forces the interim use of alternative processes.

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*Work performed under the auspices of the U. S. Department of Energy

TABLE I

FCU INLET QUANTITIES (MAXIMUM)

Component	Ratio to $(D_2 + DT + T_2)$	G-Mols/Day
Q_2^* $(=D_2 + DT + T_2)$	1.0	356.4
HQ (= HD + HT)	0.02	7.20
C $(= \underline{CQ_4} + CO)$	0.001	0.36
O $(=\underline{Q_2O} + CO = NO + 1/2O_2)$	0.005	1.80
N $(=\underline{NQ_3} + NO + 1/2\ N_2)$	0.001	0.36
Ar	0.05	18.00
TOTAL FLOW		384.12

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*Q = any mix of D and T atoms

Principal species underlined

PROCESS DESCRIPTION

From the above considerations, a hybrid system has been developed, utilizing both hot metal getters and cryogenic adsorption with catalytic oxidation, as shown in Figure 1.

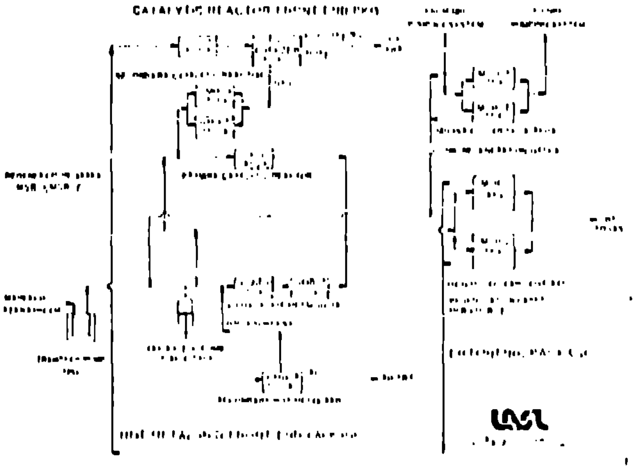

Figure 1 -
Schematic Diagram of FCU Process

The Catalytic Reactor Package and the Hot Metal Bed Package are parallel process trains, each containing elements of separate impurity removal processes which, when followed by the common Cryogenic Package, are capable of achieving the specified level of purification. Each front end package also contains separate processes for impurity treatment and tritium recovery. Process flow will be switched alternately between the two front end packages to achieve continuous cleanup and full operating experience with both technologies.

Passing stepwise through the FCU proceeds as follows. Regardless of path, the first element is the variable volume surge tank, which is used to remove flow fluctuations and provide a constant feed pressure of 1000 torr. If the Catalytic Reactor Package is on-stream, flow proceeds through the catalytic reactor, CR-1, in which any free oxygen is combined with hydrogen over a 'DEOXO' type catalyst at 450 K. The removal of all impurities takes place in the following molecular sieve bed, MSB-1 or -2, in which a small freezer section and 5A molecular sieves (60-80 mesh) at 75 K utilizes condensation and adsorption to produce a fuel stream directly suitable for cryogenic distillation. When the capacity of MSB-1, for example, is exhausted, flow is switched to the regenerated bed, MSB-2. MSB-1 is heated to 400 K to drive the contained impurities to the catalytic reactor, CR-2, where catalytic oxidation at 800 K produces tritiated water, DTO, and tritium-free compounds, CO_2, N_2, and Ar. DTO is subsequently frozen out at 160 K and periodically vaporized and routed to a hot uranium bed, HMB-4 or -5, in which DT is freed for reuse by the formation of stable oxides of uranium at 750 K. HMB-1 and -2 eventually will be replaced by an electrolysis cell when such becomes available. The waste gases, CO_2, N_2, O_2, and Ar, receive final detritiation in the TSTA Tritium Waste Treatment System -TWT- (catalytic recombiner and drying beds) before being stacked to the atmosphere.

When the alternate front end package, the Hot Metal Bed Package, is on-stream, the main feedstream leaves the variable volume surge tank and enters the primary uranium beds, HMB-1 and -2, at 1170 K. In these beds, impurities are removed by chemical reactions that form uranium oxides, carbides, and nitrides. The inert argon, with traces of the other impurities, passes through to be picked up in MSB-1 or -2, as before. The regenerated argon, with a small

TABLE II

Advantages and Disadvantages of Alternative U Processes

	Process	Advantages	Disadvantages
1.	cryogenic	no solid wastes; simple, non toxic waste gases go to atmosphere low operating cost	a proven safe, reliable electrolyzer with low inventory for DTO has not been developed
2.	hot U getters	available minimizes handling tritium in the more hazardous oxide form	operating temperature of 1170 K causes permeation and materials problems operating costs of $100,000 per year for replacement of disposable getter beds.

amount of tritium, from this bed is sent to a titanium bed, HMB-3, at 500 K, which collects DT and passes on an argon stream containing only tenths of a ppm of DT to go to TWT. The DT collected in HMB-3 is periodically recovered for reuse by heating the bed to 1170 K.

The only remaining elements of FCU are the molecular sieve beds MSB-3 and -4 shown in Figure 1. These beds collect small amounts of impurities from the neutral beam line and on a schedule of approximately once per week regenerate their accumulated impurities into the inlet to FCU.

DESCRIPTION OF COMPONENTS

General overall descriptions of the major components are given in Table III. Because of the permeability of metal walls to DT at the working temperatures of the hot metal beds (117" K), these beds require a special triple-wall design to prevent the diffusion of unacceptable amounts of tritium into the glove box containing the FCU.

ANALYTICAL CONTROLS

Gas chromatography will be used as the principal analytical method in the TSTA fuel cleanup system. Samples taken with probes inserted within the molecular sieve beds will allow a sufficient margin of safety to switch to the alternate path without total impurity breakthrough. Typical analyses can be performed using a helium carrier gas and columns packed with carbon granules. Such analyses should require only a few minutes for data collection, reduction, and transmission for automated computer control.

A microprocessor-controlled infrared spectrometer with a 20-meter-path gas cell is available for overall monitoring of the exit gas stream with detection limits of less than 1 ppm for impurities such as CH_4 and NH_3.

TABLE III

Description of Major Equipment Components

Component	Diameter, cm i.d	Length, m	Materials
Cat. React. Pack.			
CR-1	working vol. ≈ few hundred cm³		Cu; precious metal catalyst
CR-2	working vol. ≈ few hundred cm³		Cu; precious metal catalyst
DTO Freezer	working vol. ≈ few hundred cm³		Cu; plate or tube-fin
HMB-4 & -5	1.3	1.2	Cu and st. st.; 4 kg U
Cryo. Pack.			
MSB-1,-2,-3,-4	5	1.1	Cu; 1.6 kg mole. sieves
Hot Metal Pack.			
HMB-1 &2	3.8	1.2	Cu and st.st.; 4 kg U
HMB-3	2.5	0.6	Cu and st.st.; [illegible] Ti;
surge vol.	vol. = 5.5ℓ (max) - 1.7ℓ (min)		st.st. bellows

SUPPORTING EXPERIMENTAL RESEARCH

An experimental flow system has been constructed in order to test the feasibility of some of the various operational methods and to obtain quantitative performance data for the more promising unit operations. This system contains four parts, a) a gas mixing and storage system in which appropriate concentrations of impurities can be mixed with the carrier gas - hydrogen isotopes and helium - and stored for subsequent experimental tests, b) a flow metering and temperature control system, c) a modularly interchangeable unit operations system in which various adsorption tubes, catalytic converters, or metal getter units can be installed for testing, and d) an analytical system using a microprocessor-controlled infrared spectrometer and a quadrupole mass spectrometer for determining concentrations of inlet and outlet gas. The overall flow system was usually operated at flows approximately one tenth that of the full scale TSTA system.

Using inlet concentrations similar to those shown in Table I, experiments have been performed on the adsorption of ammonia and carbon dioxide on molecular sieves of various mesh sizes at ambient temperatures, and on the adsorption of methane on charcoal and molecular sieve materials at 75 K. In all cases, sharp "break-through" curves were obtained which gave the amount of adsorbate per unit adsorbent. These data together with the dimensions of the experimental adsorption tubes permit scaling to the required dimensions for the full scale TSTA system adsorption units. A typical experimental "break-through" curve is shown in Figure 2.

Tests have been made on the simulated regeneration of adsorption beds with the subsequent catalytic burning of the contained methane on a Hopcalite catalyst at 450 C. It was shown that by recycling a relatively small amount of helium gas as a carrier gas, the concentration of the desorbed methane could be reduced from about 20,000 ppm to less than 3 ppm in about five hours, provided that additional oxygen was added to the circulating helium. This latter step is necessary because the circulating gas lacks sufficient residence time on the Hopcalite to utilize all the contained oxygen originally in the Hopcalite.

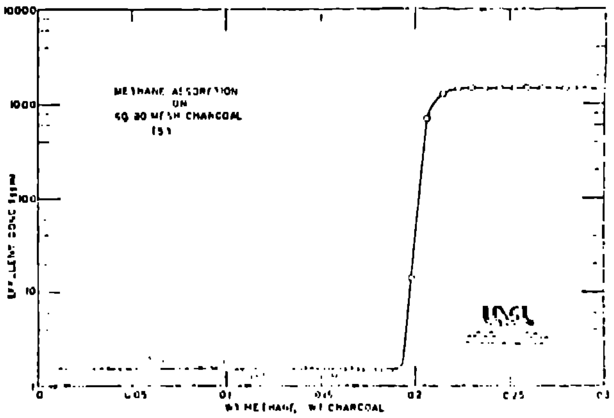

Figure 2-
Methane Adsorption on Charcoal

A number of experiments were done to measure the amount of coadsorbed hydrogen isotopes which would remain on molecular sieves after a preliminary controlled pumpout period before warmup of the molecular sieve bed during the regeneration process. Under typical proposed operating conditions for the TSTA system, this amount of hydrogen isotope would be a maximum of about 35 standard cm^3 per gram of molecular sieve. This gas would be burned to water along with the methane type compounds during regeneration and oxidation of the effluent from the molecular sieve bed.

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