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HYDROGEN ISOTOPE DISTILLATION FOR THE TRITIUM SYSTEMS TEST ASSEMBLY

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A system of four, interlinked, cryogenic fractional distillation columns has been designed as a prototype for fuel processing for fusion power reactors. The distillation system will continuously separate a feedstream of 360 g moles/day of roughly 50-50 deuterium-tritium containing ∿1% H into four product streams: 1) a tritium-free stream of HD for waste disposal; 2) a stream of high-purity D_2 for simulated neutral beam injection; 3) a stream of DT for simulated reactor refueling; and 4) a stream of high purity T_2 for refueling and studies on properties of tritium and effects of tritium on materials.

INTRODUCTION

The purpose, functions, and major subsystems of the Tritium Systems Test Assembly (TSTA) are described elsewhere in these proceedings$^{(1)}$. A key subsystem of TSTA is the Isotope Separation System (ISS), which the present paper will discuss in depth as to purpose and functions; design concepts and considerations; design method, including laboratory measurements; and the detailed design of the final process.

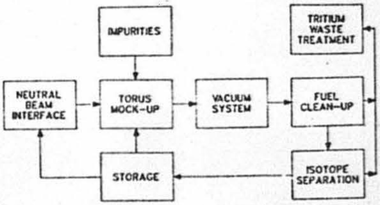

Figure 1. Functions of ISS in TSTA Process.

PURPOSE AND FUNCTIONS OF THE ISS

The role of the ISS in the overall TSTA process is summarized in Figure 1. Details are available$^{(1)}$ and need not be repeated here. The principal separation duties required of the ISS are shown in Figure 2.

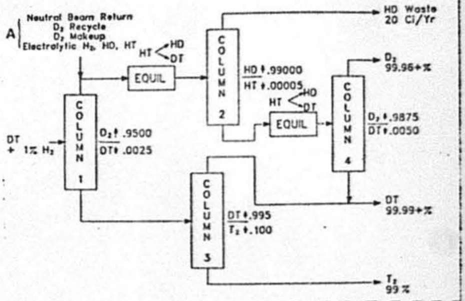

Figure 2. Principal Separation Duties of ISS.

Product Streams

The four product streams were selected to give a full verification and demonstration of the techniques of isotope separation needed for fusion power reactors and to provide maximum flexibility in the operation of TSTA. Product streams needed by eventual commercial reactors may be fewer in number or have different purity requirements from these; however, the cost to TSTA for the four-product system chosen would not have been significantly decreased by deleting one stream or by relaxing any purity requirements, while flexibility to meet changing requirements would have been substantially impaired.

The purpose of the HD waste stream, the topmost stream shown in Figure 2, is to eliminate from the fuel process loop unwanted hydrogen (H) which appears as a result of outgassing, minor leaks, diffusion, and isotope exchange reactions. The ISS provides a hydrogen-rich stream which can be discharged directly to the environment free of tritium down to a level of 20 Ci/yr. The second stream in Figure 2, the D_2 stream, has a design purity of 99.96+% D_2 with most of the remainder (>80%) being HD rather than DT or HT. The purity specification of this stream, to simulate a stream for neutral beam injection, was set in coordination with TNS design teams.

The third stream shown is the primary stream for simulated reactor refueling, consisting of a nearly equimolar mixture of D-T. The bottommost stream shown in Figure 2 is a high-purity stream of T_2 to be used at TSTA for laboratory studies on properties of tritium and on effects of tritium on materials. A commercial reactor perhaps would not require such a stream unless a tritium neutral beam is used for additional heating of the plasma.

Feed Streams

The feed rate to the ISS is that required for a full-scale (several hundred MW) commercial fusion reactor. Two feed streams are shown in Figure 2 - namely, the feed to Column 1, which is the primary TSTA process flow coming directly from the Fuel Cleanup System (see Fig. 1) of 360 g moles/day of D-T plus 1 mol% H_2, containing less than 1 ppm total noncondensible impurities; and, secondly, the stream (A) shown joining with the distillate from the top of Column 1. To understand fully the reasons behind this secondary feed, it is necessary to understand both the internal ISS process, which will be discussed in the succeeding section, as well as the interrelationships between the ISS and the Neutral Beam Interface, the Fuel Cleanup System, and the Tritium Waste Treatment. These interrelationships are shown schematically in Figure 3.

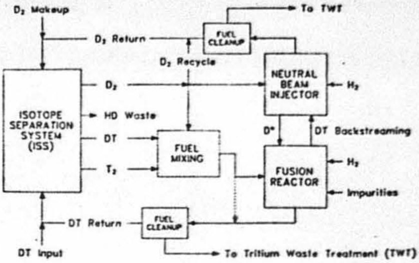

Figure 3. Interrelationships of ISS with other Subsystems.

The four components of feedstream A are listed as follows: 1) neutral beam return; 2) D_2 recycle; 3) D_2 makeup; and 4) electrolytic H_2, HD, HT. The first of these has a flowrate of about 35 moles/day consisting primarily of D_2 (98%) contaminated with hydrogen and with tritium from backstreaming up the neutral beamline. The "D_2 recycle" component is a relatively large flow (248 moles/day) of almost pure D_2 whose purpose is to help split HT into HD and DT in the two equilibrators, as will be explained more fully later. The "D_2 makeup" component is simply to replace D which unavoidably leaves the system with HD waste and is designed to be less than 4 moles/day of D_2, but will, of course, depend on the rate of H ingrowth. Finally, the electrolytic hydrogens processed in the ISS arise from electrolytic decomposition of all waters formed and/or trapped out in the Fuel Cleanup System and the Emergency Tritium Cleanup System. This feature permits the recovery and reuse of virtually all tritium that combines with carbon, oxygen, or nitrogen impurities in the fueling process or accidentally spills into the process room.

DESIGN CONCEPTS AND CONSIDERATIONS

Cryogenic fractional distillation was selected as the process to effect the separations just discussed after an appraisal was made of previous work in the separation of hydrogen isotopes. In the late 1940's and early '50's, studies by the U. S. Atomic Energy Commission on separating deuterium from hydrogen for heavy water production(2,3) concluded that cryogenic distillation was an attractive choice because of its advantages of relatively large separation factors, low power consumption, high throughputs, relatively short start-up times, and flexibility of design. A plant was designed(4) but never built in the U. S., but similar plants were built in both France(5,6) and Russia(7). In the 1960's and '70's, interest arose in separating tritium from isotopic mixtures for recovering tritium or reducing contamination. Again cryogenic distillation proved attractive. A batch still for tritium recovery was built in 1966 at Savannah River Plant and a multicolumn, continuous still was built in 1971 at L'Institut Max Von Laue-Paul Langevin in Grenoble, France(8,9). Thus, in addition to the advantages found previously, the advantages of building on existing commercial experience were considered in the selection of cryogenic distillation.

Equilibrators

Besides the distillation columns, the two catalytic equilibrators located before and after Column 2 (Fig. 2) play an integral part in the separation scheme. Table I lists the six isotopic species present in order of increasing molecular weight (or decreasing volatility).

TABLE I

1.	H_2	discard to environment
2.	HD	
3.	HT	
4.	D_2	
5.	DT	retain in process
6.	T_2	

The functions of the ISS as described are to provide usable products of #4, #5, and #6, and to discard #1, and #2 to the environment. This leaves the problem of separating #3 (HT) so as to discard the H and retain the T, which obviously cannot be accomplished by distillation alone, but can be done with the room-temperature, catalytic equilibrators. In the equilibrators, the HT molecule is split in the presence of D_2 into HD and DT according to the reversible reaction $HT + D_2 \rightleftharpoons HD + DT$. However, this will not proceed far to the right, as desired, expect in the presence of a large excess of D_2, which is most conveniently provided by the "D_2 recycle" stream shown in Figures 2 and 3. The second equilibrator, following Column 2, carries the splitting of HT still further after the removal of the reaction product HD by Column 2. Thereby, both the HD waste stream and the D_2 neutral beam stream are produced with adequately low levels of tritium (0.1 and 30 ppm respectively).

Tritium Inventory

Because cryogenic distillation involves tritium in a condensed state, a disadvantage of the process is its relatively large tritium inventory. In TSTA, a majority of all process tritium (~100 out of 170 g) is contained in the ISS; though this would not be true for a commercial reactor. Minimization of tritium inventory is not a goal of TSTA; rather TSTA is to demonstrate that tritium can be handled routinely and safely in quantities exceeding 100 g. Nevertheless tritium inventory is a design consideration because an important goal of the project is to understand the relationships between design parameters and tritium inventory.

The tritium inventory in each column in the final design of the ISS is shown in Table III. Most of the total of ~100 g is seen to be in Column 1 (34 g) and Column 3 (58 g). The inventory in Column 1 could be reduced in a commercial reactor by leaving more D_2 in the bottom stream from this column, which would reduce the size of the column and would leave more D_2 in the final DT product stream. The key to being able to extract less D_2 at the top of Column 1 and still have sufficient D_2 to drive the two equilibrators is the previously discussed "D_2 recycle" stream. This was not done extensively in TSTA because the cost of the ISS would have been substantially unchanged and it would have reduced the flexibility to meet changing specifications on product or feed streams. Also inventory could be reduced in a commercial reactor, if no high-purity tritium stream is needed for neutral beams, by greatly reducing or perhaps eliminating Column 3 with its 58 g inventory.

Column Pressure

The nominal operating pressure of the distillation columns was set at 101.35 kPa (760 torr) following consideration of the several factors involved. The separation factors (relative volatilities) of all components increase as pressure decreases below the critical. Thus the necessary column heights are less the lower the operating pressure. However, it was thought undesirable for safety reasons to operate below the local atmosphere (585 torr). Because the ISS is a prototype for nationwide application, pressures no lower than a standard sea level atmosphere were judged appropriate. Higher operating pressures would decrease column diameters (while increasing the heights), but because the required diameters were already small there was no advantage to increasing the pressure above 760 torr.

Refrigeration Cycle

Refrigeration for the ISS will be provided by a standard commercial refrigerator with a capacity of 450 W at 20 K. Alternatively, energy input requirements could be reduced by about one-third by integrating the refrigeration cycle with the distillation process by utilizing the column reboilers as precoolers in the refrigeration cycle. This more efficient cycle was not chosen for TSTA because operating ease and flexibility would be lost and savings would be nominal in this size system.

Computer Control

Like all major subsystems of the TSTA, the ISS is designed to be integrated with a large central computer, which will monitor and control the process. A special feature of this integration will be the inclusion of four or five quadrupole mass spectrometers, which will provide on-line analyses of stream compositions to the computer for use in its control functions.

DESIGN METHOD

The major process design calculations in their final form were made by means of a computer code for standard multicomponent distillation, modified for use with tritium systems. The original code was generously furnished to LASL by Professor Donald Hanson of the Chemical Engineering Department at the University of California, Berkeley, and was modified by one of us (R. H. S.) to include the following:

i) The actual molal heats of vaporization were used instead of the usual assumption of equal molal heats and equal molal overflows within columns.

ii) Provision was made to allow both a constant and a composition-dependent addition (or subtraction) of heat on any theoretical separation plate. This permitted proper accounting for the radioactive heat of decay of tritium.

iii) Experimentally determined values (see the following section) were used for relative volatilities of the three principal isotopic species - D_2, DT, and T_2.

iv) Experimentally determined values (see the following section) were used for the separating efficiency or Height Equivalent to a Theoretical Plate (HETP) of specific column packings for the isotopic mixtures.

v) An important design consideration, tritium inventory, was computed from compositions on each plate, summed, and printed out.

The design code can treat columns having multiple feed streams, but does not treat multiple, interlinked columns together as a system. Interlinking the columns was done by hand for TSTA.

Experimental Determination of Design Parameters

Because only limited data were available for tritium systems, laboratory measurements were made to determine two of the key design parameters on which the least information was available - namely, the relative volatilities for $D_2/DT/T_2$ and the separating efficiency (measured as HETP) of commercial distillation column packings. A third parameter, the allowable vapor velocity, was also explored in the laboratory for comparison with other sources.

The relative volatility of two components is, of course, related to their vapor pressures and is essentially the separation factor for a single stage distillation of those components. Values were measured for D_2/DT and for D_2/T_2 and were found to lie about 5% below the ideal values computed as the ratio of vapor pressures of the pure components.(10)

HETP's were measured in a small, prototype distillation column of 9.5-mm i.d. x 457-mm in packed height. Series of tests were made using two different commercial column packings, as described in Table II. Both packings indicated a void fraction of 0.88 as packed. The first packing was tested only on mixtures of hydrogen (protium) and deuterium and the second was tested both with H-D and D-T mixtures. The results indicated values of HETP of 50 ± 5 mm. A value of 50 mm was used in the process design calculations

Superficial vapor velocities were also measured in the prototype still. Velocities up to 66 mm/sec were obtained with no sign of deterioration in separating efficiency or column performance. Other workers have operated successfully using vapor velocities above 125 mm/sec in mixtures of hydrogen isotopes and similar packings. A vapor velocity of 89 mm/sec was used in the process design calculations.

FINAL PROCESS DESIGN

Within the constraints mentioned, a system was designed by use of the computer code to perform the required separations shown in Figure 2. The resulting system specifications are summarized in Table III.

The four columns - together with redundant instrumentation, necessary interconnecting and refrigeration piping, and a thermal shield at 77 K - are enclosed in a single, stainless steel, vacuum

TABLE II. Specifications of Packing Materials Tested in Prototype

mfg.	name	model no.	mat'l	descript.	dimen.
Eglin	col. packing	1664	st. st.	wire coils	1.5 x 1.5 mm
Podbielniak	Heli-Pak	3012	st. st.	flat'n'd helices	0.03 x 0.07 x 0.07 in.

TABLE III. Final Design Specifications of the ISS

	col. packed ht., m	col. inside diam., mm	Refrig. @ 20 K, W	tritium inventory, g
Col. 1	4.11	29.0	38.6	34.3
Col. 2	4.06	19.0	18.2	0.1
Col. 3	3.20	23.2	24.1	57.5
Col. 4	4.11	38.0	64.6	10.3
Over-all Total	5.5[a]	711.[a]	145.5[b]	102.2[c]

[a] outer vacuum shell

[b] excluding heat leak

[c] including reboilers

shell, which also serves as secondary containment for the process tritium. This unit will be supplied and tested commercially at a cost of about $1.2 million, with installation at TSTA scheduled for 1981.

SUMMARY

A description is presented of the functions, design concepts, and final design of a system of four, interlinked, cryogenic distillation columns for separating a stream of mixed hydrogen isotopes into four high-purity product streams needed in a prototype D-T fueling system for fusion power reactors.

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