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FUSION TECHNOLOGY INSTITUTE
UNIVERSITY OF WISCONSIN
MADISON WISCONSIN

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Fusion Technology Institute
University of Wisconsin
1500 Engineering Drive
Madison, WI 53706

<http://fti.neep.wisc.edu>

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L. J. Wittenberg¹, W. F. Vogelsang², and A. B. Johnson, Jr.³

¹Monsanto Research Corp., Mound Lab., Visitor to Nuclear Engineering
Dept., University of Wisconsin - Madison

²Nuclear Engineering Department, University of Wisconsin - Madison

³Pacific Northwest Lab., Battelle Memorial Institute

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Abstract

A separation technique based upon the reversible formation of yttrium tri-tide for the extraction of tritium from a liquid lithium blanket was applied to a 5000 MW(th) Fusion Reactor Design. Based upon extrapolated thermodynamic parameters, the technique reduced the tritium concentration to the low level required, 5 ppm (wt), but because of the large lithium inventory nearly 8.5 kg remained in the reactor system. Experimental information of the kinetics and thermodynamics in the temperature range of interest are needed to complete the study. An isotopic dilution technique with deuterium was shown to be useful to reduce the tritium inventory by a factor of 1000. Preliminary considerations of an isotopic separation technique for D₂ and DT indicated that a cryogenic distillation system gave good separation at the required flow with a small tritium inventory.

I. A. Introduction

Liquid lithium has been proposed as both the heat transfer fluid and the fertile tritium breeding material in several fusion reactor design studies. The amount of tritium contained in these lithium blankets is an important consideration in the normal operation of the reactor and in the potential hazard associated with the accidental release of tritium from the plant to the environment. These considerations favor a low tritium inventory. On the other hand, the solutions of tritium in liquid lithium are expected to be very stable, with low tritium partial pressures; consequently, the extraction of tritium from the lithium to the low concentrations desired will be difficult.

In order to assess this problem, this study was made of a potentially attractive tritium separation technique and applied to the UWMAK-I, a 5000 MW_{th} fusion reactor design of the University of Wisconsin.⁽¹⁾ The tritium inventory in the lithium blanket and its diffusion into the steam system were calculated. Additionally, the technique of isotopic dilution with deuterium was explored briefly in order to reduce the tritium inventory and diffusion. The concomitant requirements of a deuterium-tritium isotopic separation technique prior to fabrication of plasma fuel were defined by the use of a cryogenic distillation system.

In the fusion reactor design selected,^a a 316 stainless steel structure is cooled by a primary lithium blanket, containing nearly 1.7×10^6 kg Li, which transfers heat to an intermediate sodium circuit ahead of a steam generator. Because of possible corrosive effects, the maximum lithium temperature was limited initially to 483°C while the heat and hydrodynamic flow considerations

^a See Figure 1

required a 200° temperature differential for the working fluid so that the minimum lithium temperature was 283°C. In order to confine the tritiated fluids as closely as possible to the reactor, the recovery of tritium directly from the lithium was explored rather than from the sodium.

I. B. Tritium Removal Technique

The various techniques proposed for the removal of tritium from liquid lithium were considered for this study in order to select a potentially useful method. Only tentative results were obtained in most cases because known thermodynamic data had to be extrapolated over wide temperature ranges or for some techniques no experimental data existed. Also, no attempt was made at this time to combine two attractive techniques to form a system. The techniques^(2,3,4) which have been proposed are: diffusion through a metal membrane, distillation, helium gas sparging, cold-trapping of LiT, freezing of Li, zone refining, absorption on a metal absorber, electromagnetic separation, thermal diffusion, reaction with a non-metal, extraction into a second immiscible phase, and electrolytic transport through an ionic conducting electrolyte. For the particular reactor concept under consideration, the tritium extraction system based upon the absorption on a metal absorber appeared to be a simple technique which was selected for further study. In addition, the stabilities of several metal hydride systems have been examined so that some thermodynamic data were available for this study.

II. A. Tritium Absorption from Lithium Blankets

The optimum metal for the extraction bed would combine a high affinity for tritium compared to lithium and a low affinity for impurities such as oxygen, nitrogen and carbon, which may reduce the surface activity of the metal bed. Several metals form hydrides more stable than LiH, including Ca, Sc, Y, Zr and certain rare earths.⁽⁵⁾ Yttrium hydride has the highest stability. Unfortunately, the oxygen affinities of the metals listed above are similar to or greater than the oxygen affinity of lithium.^(6,7) The literature appears to be essentially devoid of information regarding the kinetics of hydrogen isotope uptake into metals in contact with liquid lithium and no information of the effects of impurities; consequently, the selection of the metal for the extraction bed is based principally on thermodynamic considerations. On this basis, yttrium was selected for the tritium extractant in UWMAC-I because it appears to have maximum affinity for tritium and is sufficiently abundant to be considered for fusion technology applications.

II. B. Thermodynamic Considerations for Yttrium-Tritium Reactions

A discussion of the yttrium-tritium-lithium system must be considered tentative on present knowledge, because no information exists regarding the ternary system. Furthermore, data for the binary systems, lithium-hydrogen and yttrium-hydrogen must be extrapolated to the UWMAC-I temperature range. Also, very little is known about the phase relationships which exist in the binary and ternary systems at the low tritium concentrations which are expected to be required in the lithium blanket to minimize tritium release to the steam system and to the environment. The properties of tritium and tritides must be deduced largely from published data for hydrogen and hydrides. The corrections generally are small compared to other uncertainties; however, some basis does exist for making corrections for isotopic differences where they are significant.⁽⁸⁾

The phase diagram for the yttrium-hydrogen system indicates three phases⁽⁹⁾: an alpha solid solution, YH_{2+x} , and YH_{3-x} . Comparisons of hydride stabilities^(c) at 300°C (extrapolated from 600-900°C) indicate that YH_2 is more stable than LiH.⁽⁵⁾ The equilibrium,

$$2LiT + Y \rightleftharpoons YT_2 + 2Li, \quad (1)$$

which is shifted strongly to the right, does not fully represent the reactants in the extraction system, because the tritium exists in liquid solution and a

(b) see figure 2

(c) see figure 3

range of yttrium-tritium solid solutions exist prior to YT_2 formation. The dihydride tends to be brittle, but otherwise has good mechanical stability. The trihydride tends to revert to powder and must be avoided.

At the UWMAK-I extraction system temperature ($\sim 285^\circ\text{C}$), the equilibrium hydrogen pressure over $Y-YH_2$ is 1.5×10^{-11} torr, calculated from the relationship⁽⁹⁾

$$\log P_{H_2} \text{ (torr)} = 10.52 - 11.9 \times 10^3/T \quad (2)$$

where T is $^\circ\text{K}$. At equilibrium tritium pressure calculated from Eqn. 1 is also the tritium pressure in the lithium system so that the atom fraction of tritium in the lithium can be calculated from Sieverts' Law:

$$P^{1/2} = K_s N \quad (3)$$

where P is tritium pressure in torr, K_s is the Sieverts' Law constant and N is the atom fraction of tritium in lithium. The value of K_s at 285°C , $0.346 \text{ torr}^{1/2}$ (atom fraction)⁽¹⁰⁾ was extrapolated from the high temperature data of lithium hydride.⁽¹⁰⁾ The calculated concentration of tritium in lithium is 1.1×10^{-5} atom fraction or 5 ppm (wt).

This base line concentration must be compared with the increment resulting from generation of new tritium in the blanket. The concentration, C , of T in Li as it exits from the blanket is

$$C = N + \Delta C \quad (4)$$

where $\Delta C = \frac{\text{tritium generation rate}}{\text{lithium flow rate}} = 4.9 \times 10^{-9}$ atom fraction.

Thus, ΔC is negligible compared to N . The solubility of LiH in lithium at 285°C is $N_s = 5.7 \times 10^{-4}$ mole fraction⁽¹⁰⁾, which is ~ 50 times larger than N , indicating that LiT will not deposit in the heat exchanger.

The amount of yttrium required to absorb the tritium generated daily in the blanket (1.05 kg) is 17.3 kg, assuming that Y is converted to $YT_{1.8}$. An efficiency of 20% is typical for absorption beds⁽¹¹⁾, suggesting that the total yttrium weight in an extraction bed be ~ 90 kg. Assuming a cost of $\$330/\text{kg}$ for yttrium foil, the total cost of one bed for the primary lithium system is $\$3 \times 10^4$. Because two beds are required, one to be on-line while the other is being regenerated, the total cost is $\$6 \times 10^4$ for yttrium in the primary system.

II. C. Kinetic Considerations of Tritium Absorption in Yttrium

The foregoing thermodynamic considerations require that ideal conditions exist. Under plant conditions the yttrium surface may become fouled by impurities in the lithium, such as oxygen, nitrogen and stainless steel corrosion products. Experience with zirconium alloys has demonstrated that very thin ($\sim 50 \text{ \AA}$) oxide films markedly decrease hydriding rates.⁽¹²⁾ Thermodynamic data cited earlier suggest that oxide films are likely to occur on yttrium if sufficient oxygen impurity is present in the lithium.

A second important kinetic consideration is the rate of tritium diffusion in yttrium, but such information does not exist. For diffusion of T in αY , diffusion data for tritium in αZr ⁽¹³⁾ were used. The approximation should be reasonable because both have hexagonal close-packed structures and similar melting points. For the purpose of this discussion, an estimated time of ~ 80 min. was obtained for 95% saturation of an yttrium foil 0.01 cm thick with a saturated boundary condition and a slab geometry⁽¹⁴⁾, based on tritium in αZr at 300°C ($5.3 \times 10^{-7} \text{ cm}^2/\text{sec}$). A thin tritide layer would almost certainly be required to assure the saturated boundary condition. With such a short time constant, the αY would quickly transport tritium from this layer into the bulk of the metal so that YT_2 would begin to form throughout the metal foil. Metallographic examinations⁽⁹⁾ indicate that YH_2 does form rapidly in the grain boundaries of yttrium and penetrate deeply into the metal. The formation of the increased volume (4%) of YT_2 in the grain boundaries should open up the structure and enhance diffusion especially after the initial absorption and desorption cycle.

II. D. Tritium Recovery from Yttrium

Tritium recovery will follow the procedure of valving-out the saturated yttrium bed, draining the lithium and heating the yttrium to the minimum temperature consistent with practical tritium extraction kinetics. A second consideration is that the yttrium surface activity may need to be regenerated by dissolution of thin surface films into the yttrium substrate. Hydrogen absorption by yttrium occurs rapidly at 900°C.⁽¹⁵⁾ Calculations based on a slab geometry^(13,14) indicate that the extraction of tritium from zirconium foils 0.01 cm thick should be ~95% complete in 200 sec at 700°C. Experimental evidence suggests, however, that oxide films inhibited tritium release from niobium and Zircaloy; eventually, the films dissolved, as a function of temperature and time, so that the tritium release rate increased.⁽¹⁶⁾ At 700°C an oxide film 4000 Å thick on Zircaloy dissolved in 10 min.⁽¹⁷⁾ Surface oxides have been observed to dissolve into⁽¹⁸⁾ yttrium at 900°C, but no information exists for dissolution at a lower temperature. If similar evolution and dissolution kinetics apply for yttrium as noted for zirconium alloys, then a 700°C tritium extraction cycle appears feasible. In order to minimize power requirements, technology studies should explore the possibility of longer cycles at lower temperature for the yttrium extraction bed.

III. Tritium Release and Total Inventory

The tritium inventory in the reactor system is a function of its concentration in the lithium and the total volume of lithium in the blanket, heat exchanger and connecting pipes, which has been estimated to be approximately 1.7×10^6 kg. Consequently, although the tritium concentration is low, 5 ppm(wt), the total tritium inventory in the blanket is nearly 8.5 kg. To this must be added nearly 1.05 kg of tritium being absorbed or desorbed from the yttrium extraction beds, for a total tritium inventory of nearly 9.5 kg. This tritium represents approximately two percent of the total radioactivity in the plant after ten years operation, although the biological health hazard of the tritium is low compared with the other radioisotopes in the plant structure.

The routine tritium release into the steam systems by diffusion through the power cycle was determined based upon the equipment specified, the tritium pressure in the lithium and the appropriate permeability relationships. The proposed intermediate heat exchanger⁽¹⁹⁾ consists of 1.86×10^4 m² surface area of stainless steel tubes, 1.07 mm thick, and operates at an average temperature of 360°C. The sodium-steam heat exchanger,⁽¹⁹⁾ incorporating 3.2×10^4 m² surface area, with a tube wall equivalent to 1.00 mm thick stainless steel and 0.63 mm of the ferritic alloy, Croloy T22, operates at an average temperature of 296°C. At steady state the volumes of tritium diffusing through the two heat exchangers are equal so that their total permeability equations can be set equal to each other. Based upon this relationship, the appropriate permeability values for each material^(20,21) and the assumption that no back pressure of tritiated water exists in the steam, the rate of release of tritium to the steam is a modest 10 ci/day, without the use of any barrier coating on the heat exchanger tubes.

The partial pressure of tritium in the intermediate sodium cycle was calculated to be 3.8×10^{-11} torr. Based upon the pressure of tritium, the estimated quantity of sodium, 7.6×10^5 kg, and the recently reported Sieverts' Law constant⁽²²⁾ for the hydrogen-sodium system, $4.43 \text{ ppm torr}^{-1/2}$, the total inventory of tritium in the sodium was only 0.02g. This quantity is negligible compared to the lithium system and indicates that it is not practical to incorporate a tritium extraction system in the sodium loop.

IV. A. Isotopic Dilution Techniques

The UWMAK-I design employs a maximum lithium temperature of 483°C in order to avoid potentially excessive corrosion of the stainless steel; consequently, the tritium diffusion through the heat exchangers to the steam cycle is a modest 10 ci/day. Recent experiments suggest, however, that the corrosion of stainless steel by lithium⁽²³⁾ may not be excessive up to 600°C. The adoption of this high

temperature for the lithium would significantly increase the thermal efficiency of the plant, but it would also promote permeation of the tritium through the heat exchangers. If the maximum lithium temperature is 600°C with a ΔT of 200°C and the yttrium bed is maintained at 283°C, an estimation discloses that nearly 2.4×10^3 ci/day of tritium diffuses into the steam cycle. This would be an unacceptable amount of tritiated water in the steam cycle and must be decreased by a factor of approximately 10^3 .

In order to decrease the tritium diffusion rate, the yttrium extraction system was retained, but the feasibility of the tritium being diluted with deuterium was studied.^(d) Deuterium was chosen rather than protium as the diluent because the molecules DT and HT are formed and the molecule DT can be used directly as plasma fuel while protium would have to be removed to a very low concentration in the plasma fuel. For this study it was assumed that the combined concentration of D+T in the lithium would be the same as previously calculated (1.1×10^{-5} atom fraction). If the atomic concentration of D is 10^3 greater than T, then the concentration of tritium is approximately 1.1×10^{-8} atom fraction tritium in lithium for a total of 9g tritium inventory. This represents the lowest practical tritium level, because the generation rate of tritium produces a concentration of 5×10^{-9} atom fraction tritium in the lithium during each cycle through the blanket. Because the volume of tritium diffusing through the heat exchangers is proportional to the square root of the tritium pressure and the mole fraction is also proportional to the square root of the tritium pressure, a reduction by a factor of 10^3 in concentration also results in a reduction of the release rate by the same amount so that the release rate becomes a manageable 2.4 Ci/day.

The deuterium would be supplied when a portion of the circulatory lithium stream flows through a bed containing YD₂ operating at a higher temperature than the tritium extraction bed. The yttrium absorption bed, which was originally designed to absorb 167 moles of tritium per day, would need to be 10^3 times larger to handle the additional deuterium. Because the yttrium requirements and the physical size of such a bed would be excessive, the possibility for the development of a moving bed of yttrium particles would reside for one hour in the absorption column and one hour in a regenerating furnace needs to be considered. Such an arrangement would only increase the amount of yttrium by $10^3/12$ to a total of 7.2×10^3 kg yttrium.

IV. B. Isotopic Separation

The introduction of deuterium into the tritium cycle makes it necessary to include an isotopic separation technique in the plant, and the tritium inventory in this system becomes of importance also. A cryogenic distillation column operating at approximately 25°K and two atmospheres pressure was selected for this study. At this temperature the relative volatility factor, α , for D₂ compared with DT⁽²⁴⁾ is nearly 1.2. The theoretical number of plates, N_p , which are required for such a distillation column operating under total reflux, can be estimated by use of the Fenske relationship,⁽²⁵⁾

$$(N_p + 1) \log \alpha = \log [X_B(1 - X_D)/X_D(1 - X_B)],$$

in which X_B and X_D are the mole fractions of DT in the boiler and distillate, respectively. A large number of theoretical plates, implying a large column with a large liquid inventory, is obtained if a single column is used to obtain reasonably high values for X_B and low values for X_D . As suggested by Wilkes⁽²⁶⁾, however, the distillation should be accomplished in stages in which the first stage column has a large diameter but few stages (short height) and is used to remove a large volume of D₂. The boiler removal from the first column, which is greatly reduced in volume, is used to feed a second column, which is smaller and has a small volume of entrained liquid.^(e)

The size of a theoretical plate was estimated from the recent work of Wilkes⁽²⁷⁾ in which a distillation column, 0.8 cm diameter, with a feed rate of 60-90 cm³ (STP)/min had a height-equivalent-theoretical-plate (HETP) of 1.4 cm

(d) see figure 4

(e) see figure 5

for a packed column used in a H_2, HD, D_2 separation at a reflux ratio of 32. When this size column was scaled enormously for the present feed rate, $2.6 \times 10^6 \text{ cm}^3$ (STP)/min, the size of the first column was estimated, shown in Table I, based upon a linear relationship between the cross-sectional area and the height of the plate. The equilibrium liquid inventory on this column, at 18 volume percent liquid phase, contains approximately 70 g tritium. The second column, Table I, has a boiler draw-off containing 90 mole percent DT and 10 mole percent D_2 which may be sufficiently pure for the plasma fuel. The distillates from each column would be used to prepare fresh YD_2 for introduction into the lithium blanket so that the tritium in these distillates would not be lost.

Based upon these approximations, the total tritium inventory, Table II, in the system diluted with deuterium is 250 g compared to the original system which had a tritium inventory of 8.6 kg without dilution.

V. Summary

The following conclusions can be drawn from the studies reported here, namely:

1. An yttrium extraction bed is capable of extracting tritium from lithium to the low levels required for UWMAK-I based upon thermodynamic considerations. Experimental information on the kinetics and thermodynamics in the temperature range of interest is needed. Also, the effect of impurities, especially oxygen, upon the performance of the extraction system is required.

2. The initial survey of the use of deuterium to dilute the tritium in the lithium blanket appears encouraging. More in-depth studies and experimental information on the cryogenics distillation system for isotopic separation of D_2 and T_2 mixtures are essential.

Table I
Preliminary Cryogenic Distillation System
for D_2/DT Isotopic Separation

| Column No. | Dia. cm | Ht. m | Stream | Rate 1 (STP)/h | Conc. (m. frac.) | |
|------------|---------|-------|----------|--------------------|------------------|-----------|
| | | | | | D_2 | DT |
| 1 | 60 | 6 | Feed | 1.56×10^5 | ~1 | 10^{-3} |
| | | | Distill. | 1.54×10^5 | ~1 | 10^{-4} |
| | | | Bottom | 1.56×10^3 | 0.9 | 0.1 |
| 2 | 6 | 6 | Feed | 1.56×10^3 | 0.9 | 0.1 |
| | | | Distill. | 1.34×10^3 | 0.99 | 0.01 |
| | | | Bottom | 205 | 0.1 | 0.9 |

Table II
Comparison of Tritium Inventories
(One-Hour Recycle)

| Location | w/ D_2 (g) | w/o D_2 (kg) |
|--------------|--------------|----------------|
| In Li | 10 | 8.5 |
| On Y | 50 | .05 |
| Distillation | | |
| 1st Column | 70 | --- |
| 2nd Column | 70 | --- |
| In Stream | 50 | --- |
| Total | 250 g | 8.55 kg |

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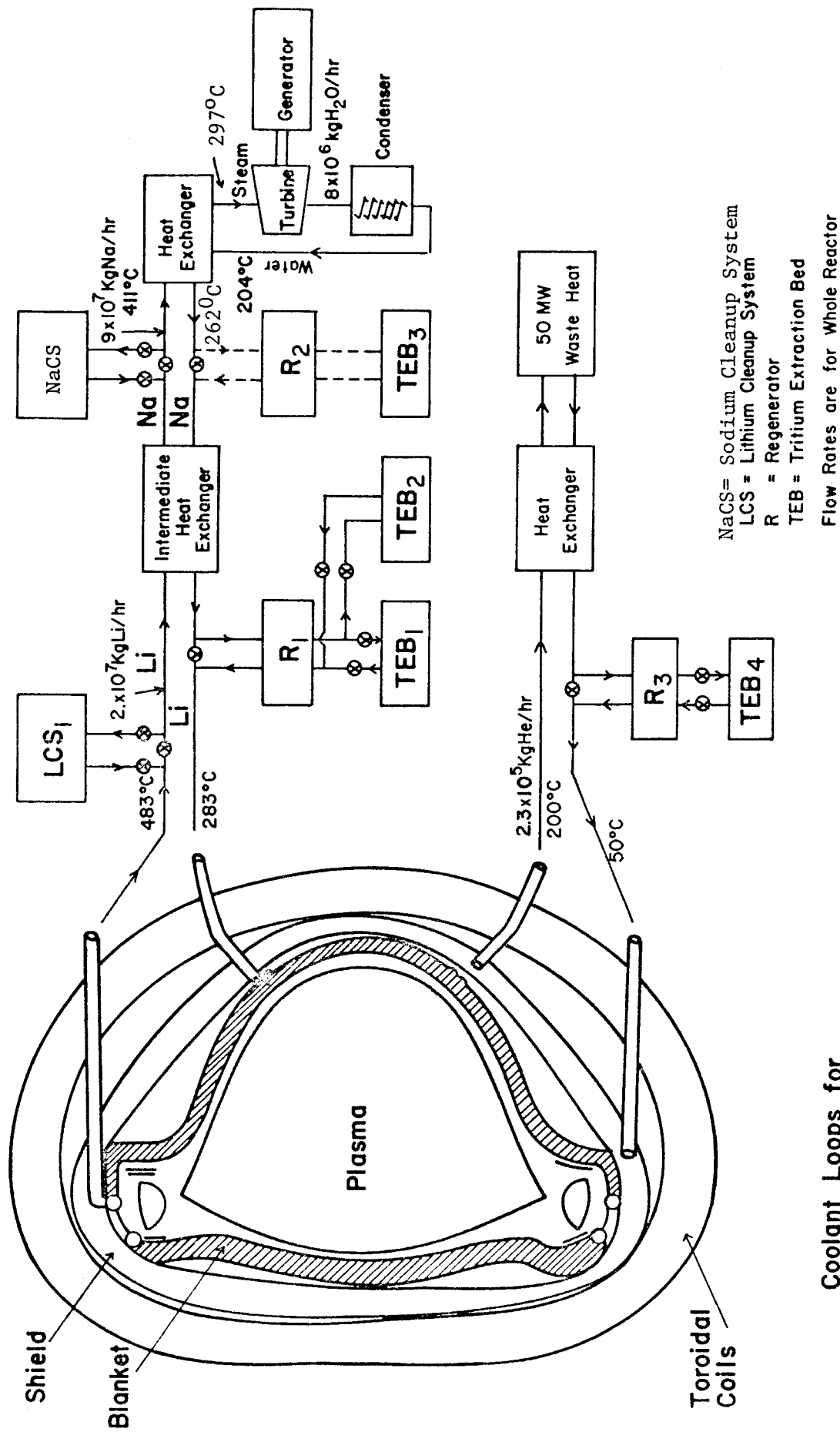
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Title of Illustrations

1. Coolant Loops for Wisconsin Toroidal Fusion Reactor - UWMAK-I.
2. Standard Free Energies of Formation per g - mole of O_2 (gas) of Metals of Importance in Lithium Blankets.
3. Hydrogen Pressure in Equilibrium with Hydrogen - Metal Systems of Importance in Lithium Blankets at 300°C.
4. Isotopic Delution Scheme for Tritium with Deuterium in a Lithium Blanket.
5. Schematic Flow Diagram of Cryogenic Distillation System for Isotopic Separation of $D_2 + DT$.



Coolant Loops for
Wisconsin Toroidal Fusion Reactor - UWMAK-I

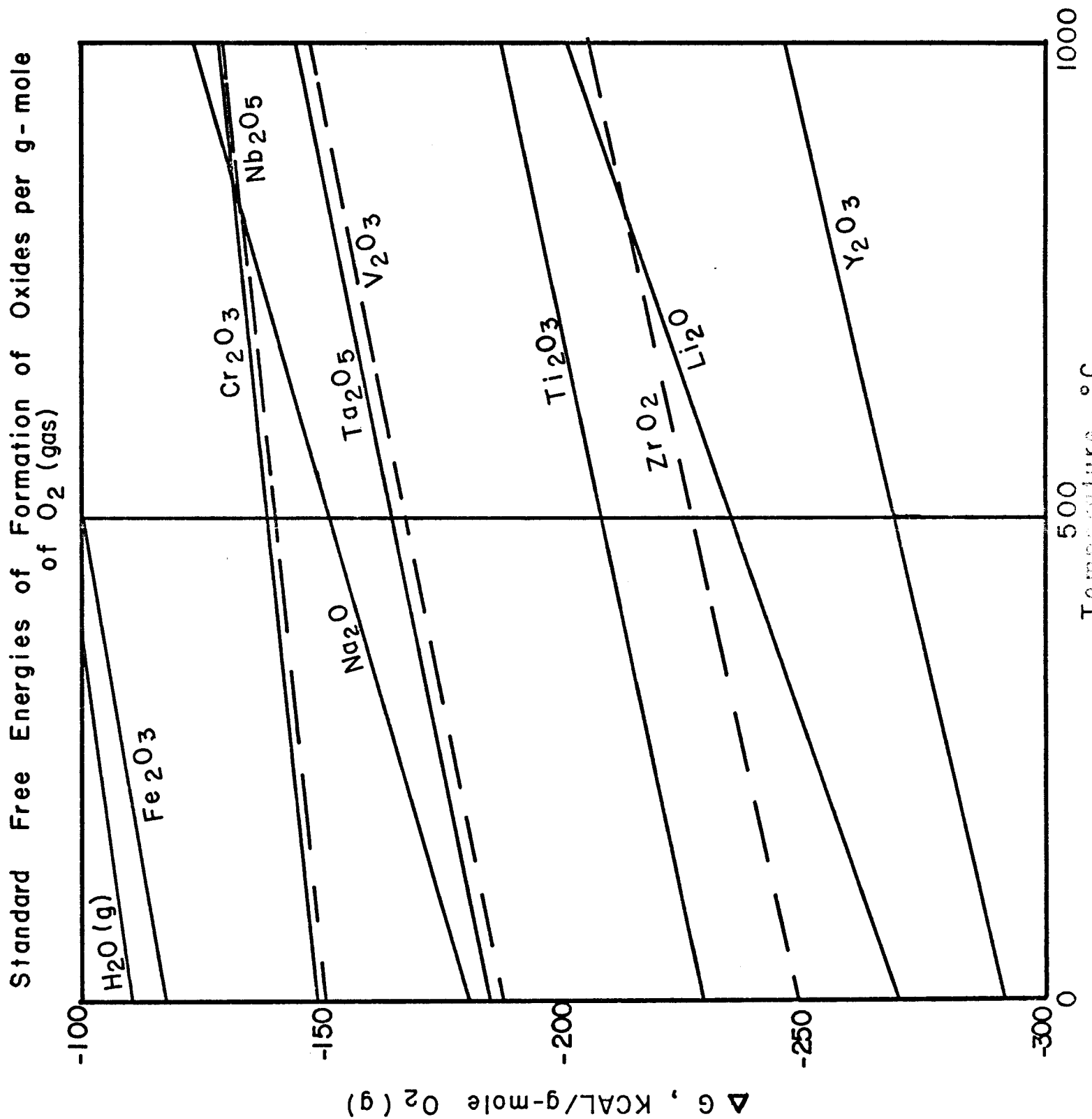


Figure 2

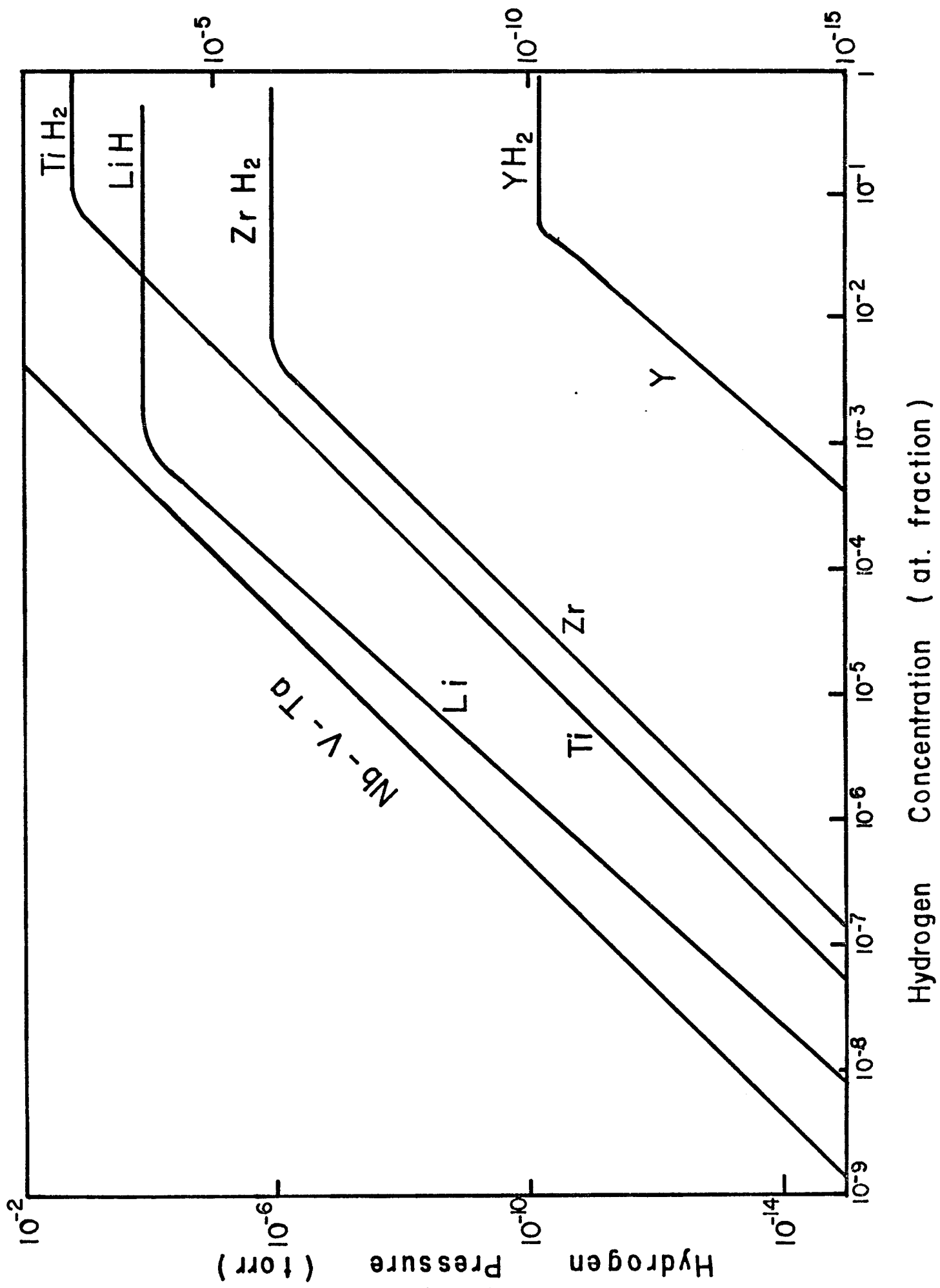


Figure 3

ISOTOPIC DILUTION OF TRITIUM IN LITHIUM BLANKET

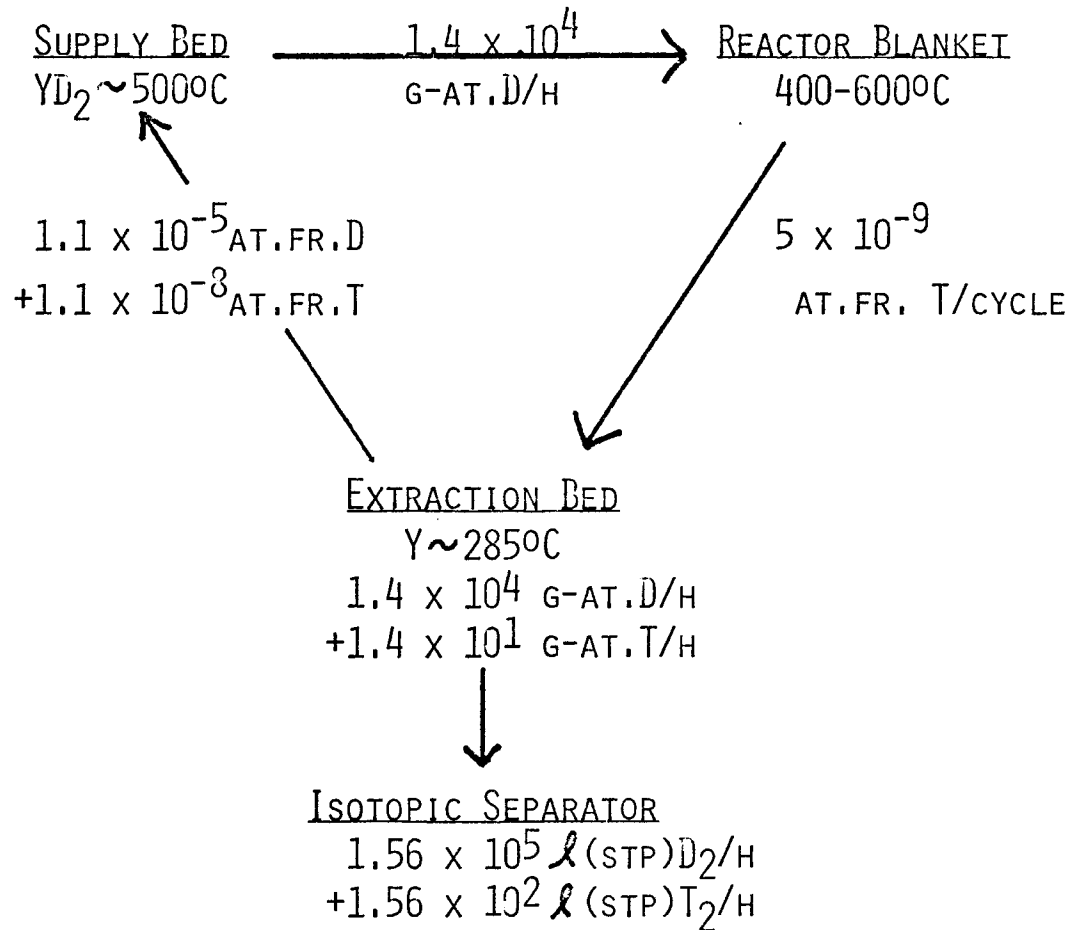


Figure 4

Schematic Flow Diagram of Cryogenic Distillation System

For Isotopic Separation of $D_2 + DT$

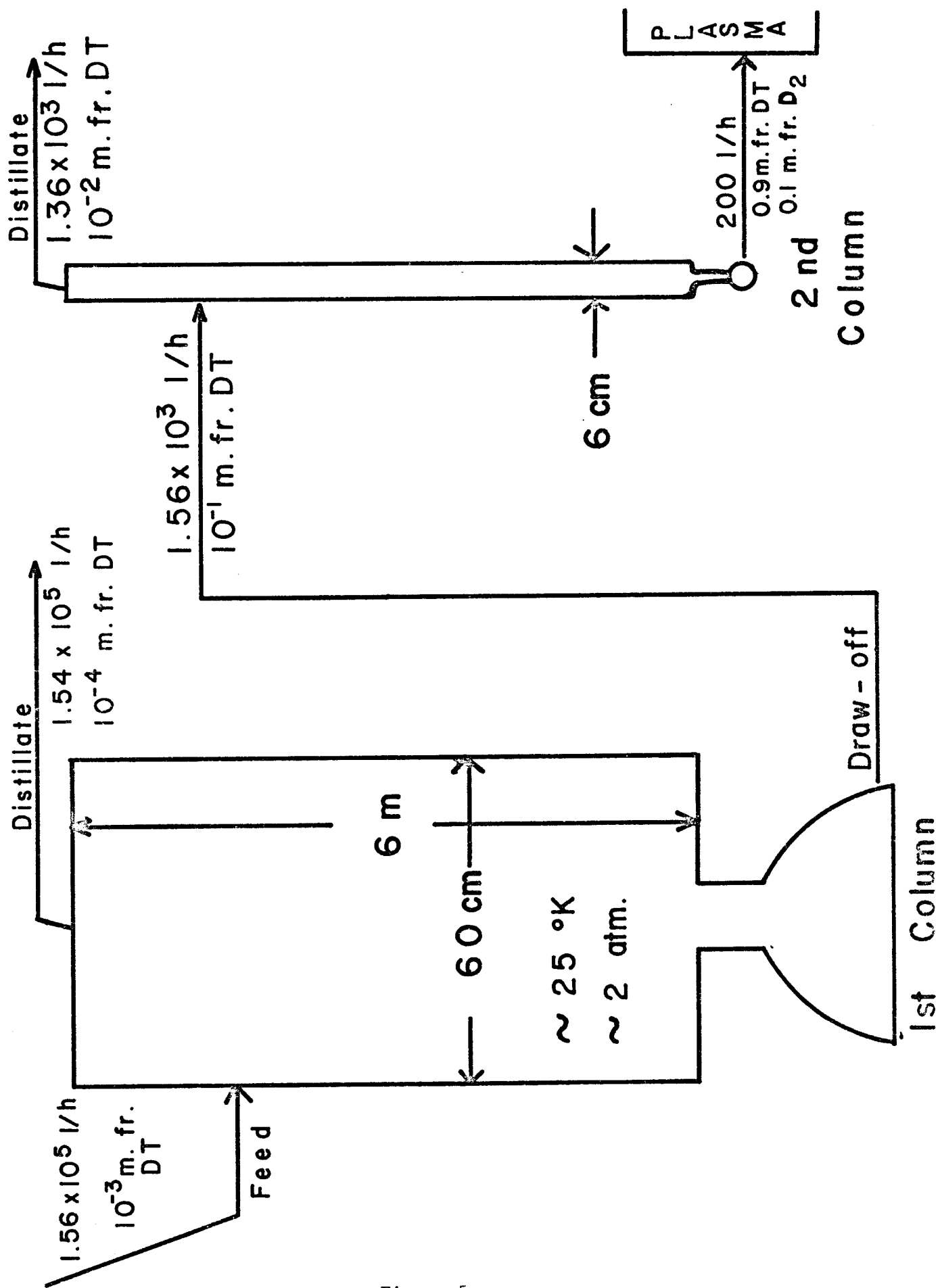


Figure 5