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EFFECT OF HELIUM ON SEPARATION
PERFORMANCE OF CRYOGENIC DISTILLATION
COLUMN CASCADE FOR FUSION REACTOR

July 1993

Toshihiko YAMANISHI and Kenji OKUNO

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Effect of Helium on Separation Performance of
Cryogenic Distillation
Column Cascade for Fusion Reactor

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The effect of helium in the feed stream on separation performance of the cryogenic distillation columns was discussed in a probable case. The column cascade at Los Alamos National Laboratory was chosen in the present study. The new data for solubility of helium in liquid hydrogen isotopes was used in the present study. Column (2) is mainly affected by the presence of helium in the fuel stream. If the helium percentage in the feed stream is 1%, the column performance can be assured by increasing the condenser load: a larger inner diameter of the column; larger flow rate of the refrigerant gas; and larger heat transfer area at the condenser should be considered. If the percentage is 5%, both the column pressure and condenser load must be doubled in the steady-state operation. These results qualitatively agreed with the simulation results by Kinoshita in which the old data for solubility of helium were used.

Keywords: Cryogenic Distillation, Helium, Cascade, Solubility,
Hydrogen Isotopes, Tritium, Computer Simulation,
Condenser Load, Control Operation

核融合炉における深冷蒸留塔カスケードに対するヘリウムの影響

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深冷蒸留塔の分離特性に対する、フィード中に含まれるヘリウムの影響を幾つかのヘリウム濃度に対して検討した。選んだ塔カスケードは、ロスアラモス国立研究所におけるものである。検討に際し、著書らが得た新しいヘリウムの水素同位体に対する溶解度データを使用した。ロスアラモス国立研究所の 4 塔カスケードにおいて、ヘリウムの影響を受けるのはほとんど Column (2) である。フィード中のヘリウム濃度が 1 % の場合、凝縮器の冷却容量を増加させることで蒸留塔の分離性能を維持することが可能である。この結果は、塔径の増大、冷媒ヘリウムガスの流量あるいは凝縮器内の熱交換面積を増大を考慮しなければならないことを意味する。ヘリウム濃度が 5 % の場合、凝縮器容量及び塔圧力を 2 倍にすることが要求される。以上本研究で得られた結果は、従来のヘリウム溶解度データを用いてヘリウムの影響を検討した木下の報告と、定性的には一致した。

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1. Introduction

In the fuel circulation system for a fusion reactor, it is necessary to remove impurities such as protium; helium; carbon; nitrogen; oxygen and argon from the spent fuel (the deuterium-tritium mixture). After the removal of these high molecular weight atoms and helium, the mixture of hydrogen isotopes is fed to a cryogenic distillation column cascade. It is quite obvious that all the impurities other than helium must be removed from the mixture before feeding it to the cascade.

The extent of the helium of the helium effects may largely depend on its percentage in the feed stream to the ISS (Isotope Separation System). On the basis of design study of the presently conceived tokamak-type reactor, the helium percentage is expected to be in the range from 1 to 10%. Steiner and Flanagan¹⁾ proposed the column cascade at Los Alamos National Laboratory^{2),3)} for the isotope separation system of the fusion reactor. Kinoshita⁴⁾ has discussed the effect of helium in the feed stream on separation performance of this cascade. He concluded that the helium effects could be minor, for instance, if the percentage of helium is no more than 1%. He used the data for solubility of helium in liquid protium and deuterium measured by Sherman⁵⁾; for HD, the geometric mean value between the solubility for H₂ and that of D₂ was applied. Recently, the author measured the solubility of helium in liquid H₂; HD and D₂⁶⁾. Especially for HD, our solubility data is 10% greater than that estimated from the data by Sherman (Henry's law constant is 10% smaller).

The present paper reports the results of computer simulation on the effects of helium on the separation performance of the column cascade. Our new data for the solubility of helium are used in the computer simulation.

2. Simulation Models

In the present study, essential information is on the extent of the helium effects on separation performance of the column. Helium has the exceptionally large volatility in comparison with those of the hydrogen isotopes (H_2 ; HD; HT; D_2 ; DT and T_2): The volatility of helium in the hydrogen isotope solutions is the most important parameter in the attainment of the purpose of the present study. Hence, the simplified model in which the heat balances and nonideality of the hydrogen isotope solutions are neglected is used for simulation of the cryogenic distillation columns, since useful information can be obtained with a minimum computational effort.

The model column for computer simulation is illustrated in Fig. 1. The component material balances^{4),7)} are expressed by the following matrix equations. Since, the nonideality is not incorporated in the model, $K_{i,j}$ is a function of temperature only.

$$\begin{bmatrix} B_{i,1} & C_{i,1} & 0 & \dots & \dots \\ A_{i,2} & B_{i,2} & C_{i,2} & 0 & \dots \\ \dots & \dots & \dots & \dots & \dots \\ & A_{i,j} & B_{i,j} & C_{i,j} & \\ 0 & \dots & \dots & A_{i,N} & B_{i,N} \end{bmatrix} \cdot \begin{bmatrix} x_{i,1} \\ x_{i,2} \\ \vdots \\ x_{i,j} \\ x_{i,N} \end{bmatrix} = \begin{bmatrix} D_{i,1} \\ D_{i,2} \\ \vdots \\ D_{i,j} \\ D_{i,N} \end{bmatrix} \quad (1)$$

$$A_{i,j} = L_{j-1}, \quad B_{i,j} = -(V_j + W_j)K_{i,j} - (L_j + U_j), \quad C_{i,j} = V_{j+1}K_{i,j+1}, \quad D_{i,j} = -F_j z_{i,j}. \quad (2)$$

Equations (1) and (2) can be solved by the tridiagonal matrix method^{7),8)}.

The expression used for computing the vapor-liquid equilibrium ratio of helium⁴⁾ is given by

$$K_{\text{He},j} = \frac{C_{\text{He},j}}{P_j}, \quad (3)$$

where $C_{\text{He},j}$ is calculated from the Henry's law constants of the six systems, $C_{\text{He-H}_2}$, $C_{\text{He-HD}}$, $C_{\text{He-HT}}$, $C_{\text{He-D}_2}$, $C_{\text{He-DT}}$, and $C_{\text{He-T}_2}$, by using

$$C_{\text{He},j} = \sum_{k=1}^6 C_{\text{He-k},j} x'_{k,j}, \quad x'_{k,j} = \frac{x_{k,j}}{\sum_{n=1}^6 x_{n,j}}. \quad (4)$$

The expression for $C_{\text{He-k},j}$ is written by

$$C_{\text{He-k},j} = F_1 \exp(F_2/RT), \quad F_1 = f_1 + f_2T, \quad F_2 = f_3 + f_4T. \quad (5)$$

for H_2 , HD, D_2 . The values of four parameters are listed in Table 1⁶⁾. In the cryogenic distillation columns, helium exists with H_2 and HD. The Henry's law constants of helium for HT, DT and T_2 have little influence on simulation results. Hence, the Henry's law constants for these three molecular species are estimated from the data by Sherman⁴⁾:

$$C_{\text{He-k},j} = a_k \exp(b_k/RT),$$

$$C_{\text{He-HT},j} = (C_{\text{He-H}_2,j} C_{\text{He-T}_2,j})^{1/2},$$

$$C_{\text{He-DT},j} = (C_{\text{He-D}_2,j} C_{\text{He-T}_2,j})^{1/2}. \quad (6)$$

The coefficients are determined by Kinoshita⁴⁾ so that the computed values fit Sherman's

data⁵⁾ : $a_k=0.445$ and $b_k=94.6$ for H_2 , $a_k=0.928$ and $b_k=108.6$ for D_2 , and $a_k=1.82$ and $b_k=114.3$ for T_2 . The equilibrium constant for helium, $K_{He,j}$, only depends on liquid composition.

3. Result and Discussion

3.1 Extent of helium effect to helium percentage in feed stream

The cascade configuration chosen is shown in Fig. 2. By assuming that helium is not present in the feed stream, appropriate design specifications for columns (1) and (2) and the resultant product compositions are calculated as summarized in Table 2. Then, the two columns are simulated by incorporating the presence of helium.

First, the helium percentage in the feed stream is assumed to be 1%. The design specifications and the resultant product compositions in the case of 1% helium are given in Table 3. No appreciable deterioration in the separation performance is found for Column (1). The most significant function of Column (2) is to remove protium (Molecular forms are H_2 and HD) from top of the column without considerable loss of tritium (HT). The performance of Column (2) can be assured as observed from the table, and the column can be operated at ordinary pressure. However, the temperature of the condenser appreciably decreases. We need to increase the condenser load appreciably.

The effect of helium on the separation performance of the column cascade is examined for the case where the helium percentage is 5% also. In this case, it is impossible to liquefy the mixture completely under the usual conditions, so the feed is supplied to each column in the gas state. In Table 4, the design specifications and the resultant product compositions in the case of 5% helium are presented. Similarly to the case of 1% helium, there is no appreciable

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effect on the separation performance of Column (1). However, the mole fraction of HT in the top product from Column (2) increases significantly, causing an unacceptably high tritium level in the H_2 -HD waste gas. To decrease the flow rate of top stream is effective to decrease the tritium level in the top stream; the protium level in the bottom stream would become an unacceptably high. Decreasing the flow rate of top stream would result in deterioration of purity of D_2 produced by Column (4). One of possible methods to overcome this problem may be to increase the liquid flow rate from the condenser or, equivalently, the reflux ratio. However, an increase in the reflux ratio up to 39.0 produces no improvement at all. When we try to further increase the reflux ratio, it is found that there are no solutions of the basic equations as long as the column pressure is set at 101.3 kPa. This means that higher column performance presented by a larger reflux ratio is unavoidably accompanied by the increase in the column pressure. Indeed, the basic equations have a set of solutions if the column pressure is increased. Choosing 202.6 kPa for the column pressure and 73.89 for the reflux ratio, the column performance can be assured, as proved in Table 4. Thus, it is true that the column performance can be assured, but significant increases in the column pressure and condenser load are needed.

It is obvious that a higher helium percentage in the feed stream requires a higher column pressure and heavier condenser load. It is well known that the relative volatilities for the six molecular species of hydrogen isotopes decrease with the increase in pressure. Hence, the high column pressure causes significant deterioration in separation performance. Note that the condenser temperature in cases where helium is present is appreciably lower than in cases of no helium (See Table 2~4). Nevertheless, the heavier condenser load is needed. Hence, the flow rate of the refrigerant gas or the heat transfer area at the condenser must be significantly larger. One of feasible methods to avoid a relatively high column pressure may be to add protium to the top flow from Column (1). This addition results in the increase in the flow rate of H_2 -HD fed to Column (2); thus, the percentage of helium in the top gas from this column is decreased. Although the column should be operated at a lower pressure, the protium addition

deteriorates the purity of the D_2 stream from Column (4) to some extent. Thus, the protium addition does not yield very successful results.

3.2 Comparison with the simulation results by Kinoshita⁴⁾

Kinoshita examined the effect of helium on the separation performance of the same column cascade selected in the present study, using the data measured by Sherman⁵⁾. Qualitative conclusions reported by Kinoshita agreed with those in the present study: In the column cascade chosen, Column (2) is mainly affected by the presence of helium. A higher percentage of helium in the feed stream requires a higher column pressure and heavier condenser load. Effect of helium on separation performance of the column was summarized in Table 5. The simulation results by Kinoshita are also presented in the table. In the case where the helium percentage is 1%, the simulation results by Kinoshita are almost equal to those of the present study. From the results for the case of 5% helium, it is found from our new data for the solubility of helium that the effect of helium on the separation performance of the column is slightly more serious for the case of the higher helium percentage than those expected by Kinoshita: the heavier condenser load and the higher reflux ratio are required.

Kinoshita discussed the helium effect on the dynamic behavior of the column also. His conclusion was that the presence of helium produced more serious problem for the dynamic column behavior. If the tritium level in the top stream of Column (2) increases, its flow rate must be decreased to maintain the tritium level at a desired value. In the helium percentage is 5%, this operation causes a serious pressure rise. As mentioned above, the effect of helium is slightly more serious for the case of the higher helium percentage than those expected by Kinoshita. We can conclude that only 5% of helium percentage in the feed stream is not acceptable to avoid a serious effect of helium on the separation performance of the column cascade.

4. Conclusion

The effects of helium on the separation performance of the column cascade are as follows (See also Table 5).

- 1) A higher percentage of helium in the feed stream causes a higher column pressure and heavier condenser load especially for Column (2) in the column cascade chosen.
- 2) If the helium percentage is 5%, for example, both the pressure and condenser load of Column (2) are doubled. If it is 1%, considerable increase of the condenser load is not required; the temperature of the refrigerant gas should be decreased.
- 3) The presence of helium in the feed stream has serious impact on the dynamic column behavior even if the helium percentage is only 5%.

The extent of the helium effects depends on the helium percentage in the feed stream. If the percentage has a significant value ($>1\%$), the helium removal should be completed before feeding the isotope mixture to the column cascade. The authors propose a falling liquid film separator as one of the most feasible system for helium separation⁹⁾. The conceptual design of the helium separator is shown in Fig. 3. This separator has an advantage that it can easily be installed in a cryogenic package with distillation columns. If the helium percentage is no more than 1%, no special helium separator may be required; a larger condenser should be considered in the design stage of the columns.

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NOMENCLATURE

$C_{He,j}$ = Henry's law constant of He in liquid hydrogen isotopes on j-th stage (Pa)

D = flow rate of top product (mol/h)

F_j = flow rate of feed steam supplied to j-th stage (mol/h)

$K_{i,j}$ = vapor-liquid equilibrium ratio of i-component on j-th stage (-)

L_C = flow rate of liquid stream leaving condenser (mol/h)

L_j = flow rate of liquid stream leaving j-th stage (mol/h)

m = total number of components (-)

N = number of total theoretical stages (-)

N_F = feed stage number (-)

P_j = pressure on j-th stage (Pa)

R = reflux ratio (-)

R' = gas constant (Pa.m/mol.K) or (J/mol.K)

T_C = condenser temperature = T_1 (K)

T_j = temperature on j-th stage (K)

U_j = flow rate of liquid sidestream from j-th stage (mol/h)

V_j = flow rate of vapor stream leaving j-th stage (mol/h)

W_j = flow rate of vapor sidestream from j-th stage (mol/h)

$x_{i,j}$ = mole fraction of i-component in liquid stream leaving j-th stage (-)

$y_{i,j}$ = mole fraction of i-component in vapor stream leaving j-th stage (-)

$z_{i,j}$ = mole fraction of i-component in feed stream supplied to j-th stage (-)

Subscript

i = component index (1 through 6: H_2 , HD, HT, D_2 , DT and T_2 ; 7: helium)

j = stage index

Table 1 Experimentally determined parameters of Henr's law constants

System	$f_1(\text{MPa})$	$f_2(\text{MPa/K})$	$f_3(\text{J/mol})$	$f_4(\text{J/mol})$
He-H ₂	1.0×10^{-7}	0.17	630	-9.2
He-HD	1.9×10^{-6}	0.57	620	-14
He-D ₂	1.1×10^{-6}	0.50	790	-16

Table 2 Input and output specifications of Columns (1) and (2) in cases of no helium in fuel input

	Column (1)	Column (2)
F (mol/h)	100	75
D (mol/h)	25	2.08
Reflux ratio	25	130
L _c (mol/h)	625	270.4
N	80	80
N _F	50	55
Pressure (kPa)	101.3	101.3
T _c (K)	23.59	22.13
Feed composition		
H ₂	1.368×10^{-4}	2.527×10^{-4}
HD	1.048×10^{-2}	2.798×10^{-2}
HT	9.248×10^{-3}	1.775×10^{-4}
D ₂	0.2481	0.9581
DT	0.4832×10^{-2}	1.348×10^{-2}
T ₂	0.2488	5.010×10^{-5}
Top composition		
H ₂	5.472×10^{-4}	9.111×10^{-3}
HD	4.192×10^{-2}	0.9909
HT	3.699×10^{-2}	2.947×10^{-7}
D ₂	0.9203	2.048×10^{-9}
DT	2.655×10^{-4}	0.0
T ₂	2.244×10^{-9}	0.0

Table 3 Input and output specifications of Columns (1)
and (2) in cases of 1% helium in fuel input

	Column (1)*	Column (2)*
F (mol/h)	101.0101	76.0101
D (mol/h)	26.0101	3.08
Reflux ratio	24.03	87.99
L _c (mol/h)	625	270.8
N	80	80
N _F	50	55
Pressure (kPa)	101.3	101.3
T _c (K)	23.46	20.84
Feed composition		
He	1.000x10 ⁻²	1.329x10 ⁻²
H ₂	1.354x10 ⁻⁴	2.494x10 ⁻⁴
HD	1.038x10 ⁻²	2.761x10 ⁻²
HT	9.156x10 ⁻³	1.742x10 ⁻⁴
D ₂	0.2456	0.9453
DT	0.4784	1.330x10 ⁻²
T ₂	0.2463	4.944x10 ⁻⁵
Top composition		
He	3.884x10 ⁻²	0.3274
H ₂	5.260x10 ⁻⁴	6.144x10 ⁻³
HD	4.030x10 ⁻²	0.6664
HT	3.555x10 ⁻²	1.287x10 ⁻⁷
D ₂	0.8845	0.0
DT	2.534x10 ⁻⁴	0.0
T ₂	2.127x10 ⁻⁹	0.0

* The reflux ratio is established so that the liquid flow from the condenser (or, equivalently, the condenser load) is equal to that in cases of no helium.

Table 4 Input and output specifications of Columns (1)
and (2) in cases of 5% helium in fuel input

	Column (1)*1	Column (2)*1	Column (2)*2
F (mol/h)	105.263	80.2631	
D (mol/h)	30.2631	7.33	
Reflux ratio	20.65	37.00	73.89
L_c (mol)	625	271.2	541.6
N	80	80	
N_F	50	55	
Pressure (kPa)	101.3	101.3	202.6
T_c (K)	22.95	18.54	20.32
Feed composition			
He	5.000×10^{-2}	6.557×10^{-2}	
H ₂	1.230×10^{-4}	2.362×10^{-4}	
HD	9.955×10^{-3}	2.615×10^{-2}	
HT	8.786×10^{-3}	1.650×10^{-4}	
D ₂	0.2357	0.8952	
DT	0.4591	1.259×10^{-2}	
T ₂	0.2363	4.678×10^{-5}	
Top composition			
He	0.1739	0.7154	0.7180
H ₂	4.278×10^{-4}	2.577×10^{-3}	2.586×10^{-3}
HD	3.463×10^{-2}	0.2820	0.2794
HT	3.056×10^{-2}	1.484×10^{-6}	7.049×10^{-8}
D ₂	7.603×10^{-4}	1.085×10^{-8}	0.0
DT	2.122×10^{-4}	0.0	0.0
T ₂	1.733×10^{-9}	0.0	0.0

*1 The reflux ratio is established so that the liquid flow from the condenser (or, equivalently, the condenser load) is equal to that in cases of no helium.

*2 Both the column pressure and condenser load are doubled.

Table 5 Summary of helium effects on specifications of Column (2)

Helium percentage in feed stream (%)	1		5	
	This work	Kinoshita's work	This work	Kinoshita's work
D (mol/h)	3.08	3.10	7.33	7.35
R (-)	88.0	87.2	73.9	61.2
L _c (mol/h)	270.8	270.0	541.6	450.0
T _c (K)	20.84	20.85	20.32	20.34
P (kPa)	101.3	101.3	202.6	202.6

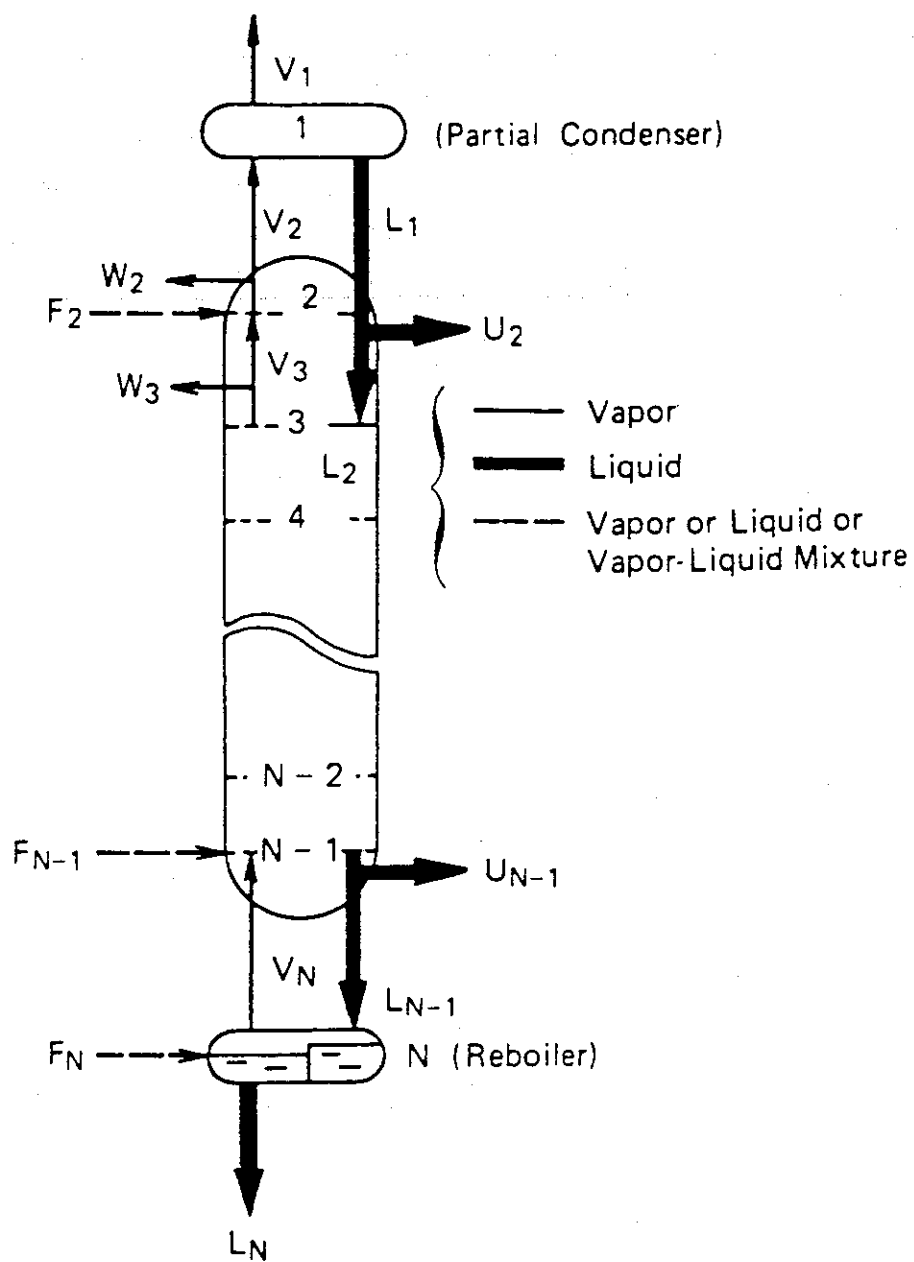


Figure 1 Model column for computer simulation.

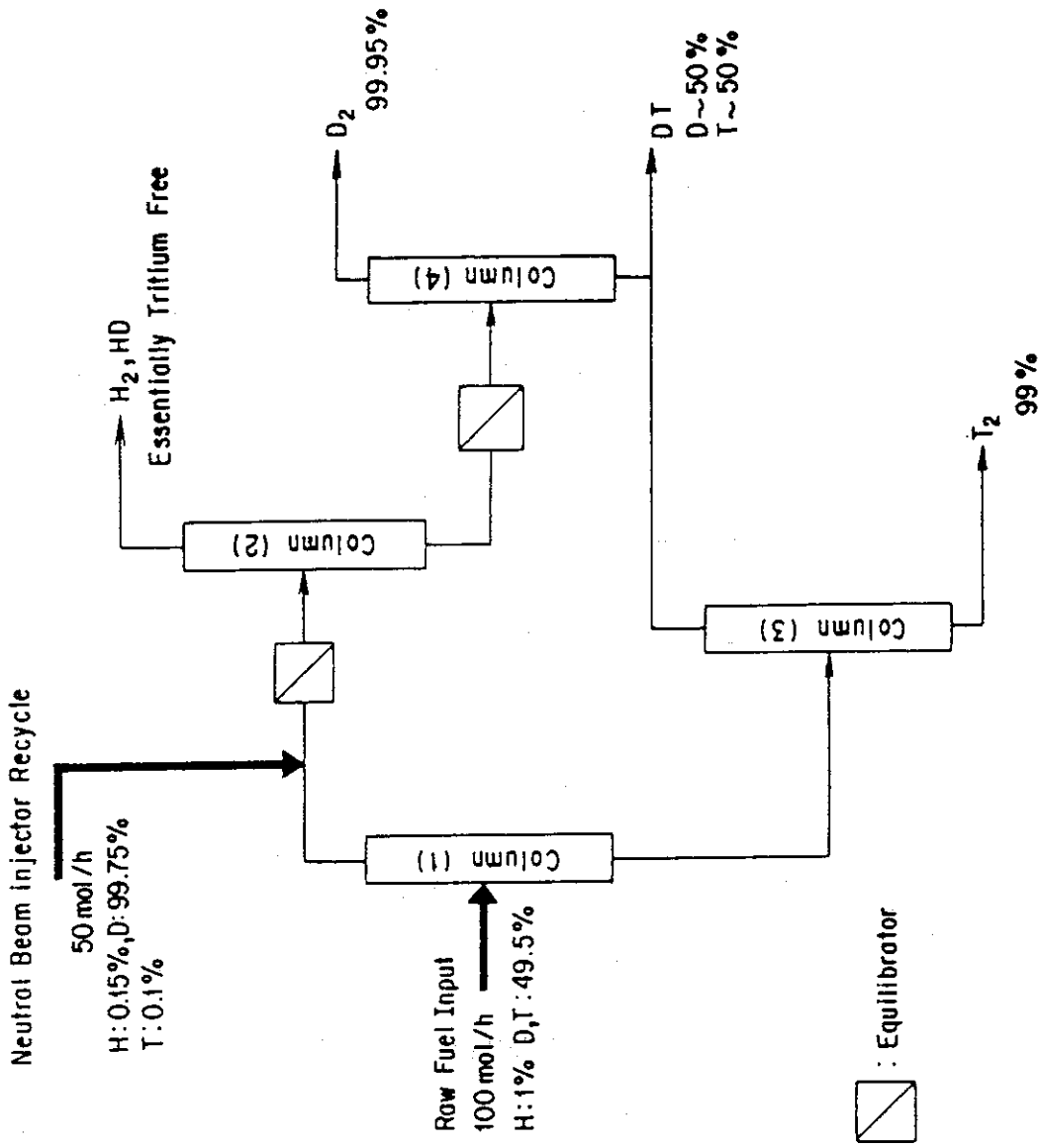


Figure 2 Cryogenic distillation column cascade chosen for the present study. The input specifications assumed are not exactly equal to those in the design at LANL, but the qualitative aspects are essentially the same.

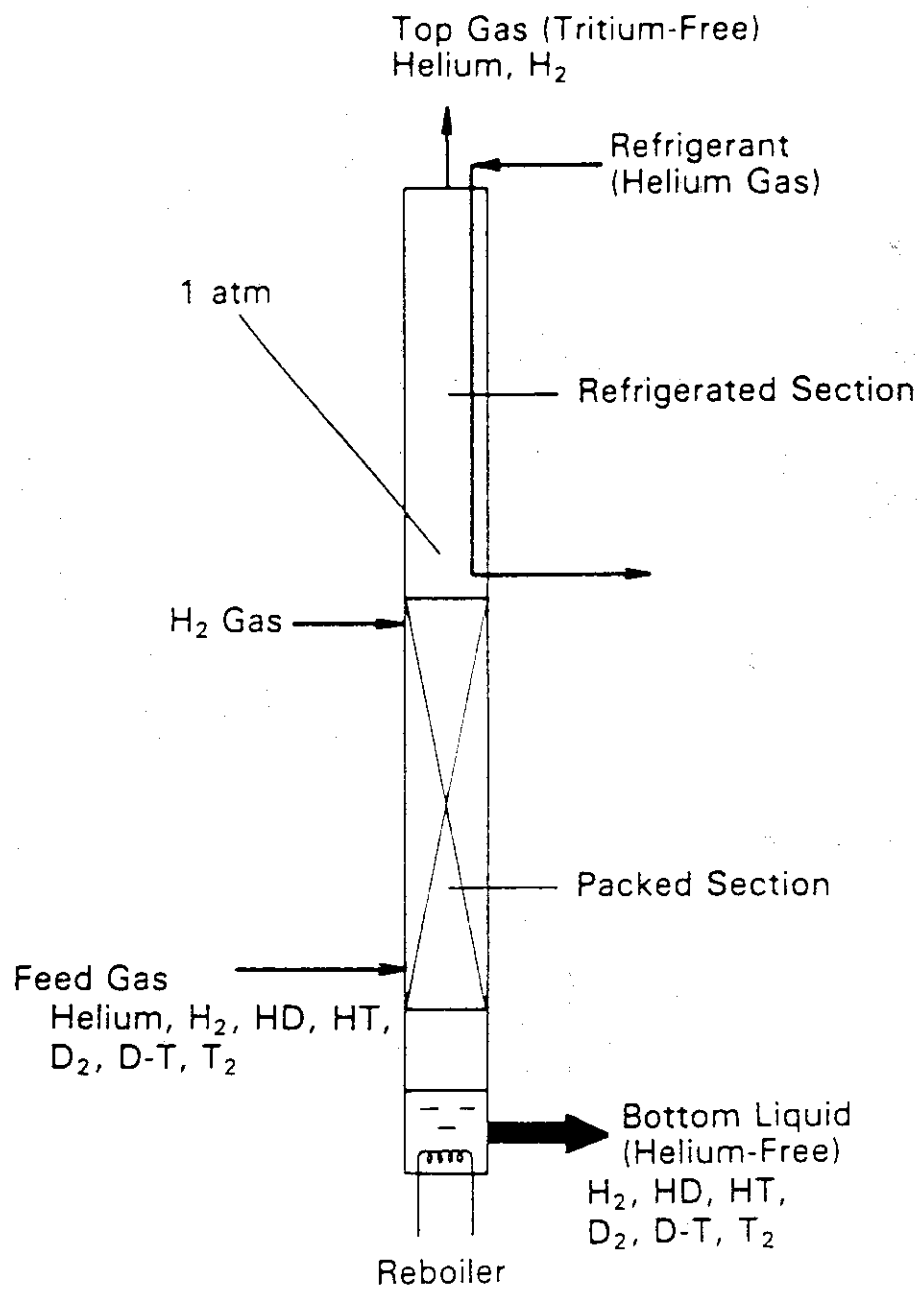


Figure 3 Conceptual flow diagram of cryogenic falling liquid film helium separator.