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MATHEMATICAL SIMULATION PROCEDURE FOR SINGLE CRYOGENIC DISTILLATION COLUMN WITH FEEDBACK STREAM PROCESSING SIX ISOTOPIC SPECIES OF MOLECULAR HYDROGEN

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Mathematical Simulation Procedure for Single Cryogenic Distillation Column with Feedback Stream Processing Six Isotopic Species of Molecular Hydrogen

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This report deals with a single cryogenic distillation column with a feedback stream and a catalytic equilibrator for recovering tritium from glovebox atmospheres and other similar sources. Although it processes the six isotopic species of molecular hydrogen, the purpose is separation of protium and tritium without any regard to deuterium.

A mathematical simulation procedure is proposed to overcome difficulty in rigorous simulation caused by presence of the feedback stream. The effect of the sidestream location on column performance is explored and the optimum location is approximately determined fixing the feed location under an assumed external feed composition.

KEYWORDS : Isotope Separation, Hydrogen Isotopes, Tritium, Cryogenic Distillation, Feedback Stream, Mathematical Simulation, Convergence, Multicomponent Distillation, Multi-dimensional Newton-Raphson Method, Sidestream Location

水素同位体6成分系を処理するフィードバック流れを持った深冷蒸留塔の数学的シミュレーション手法

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（1982年4月24日受理）

グローブボックスの雰囲気ガス中に漏洩してくるトリチウムを回収するためのシステムの1ユニットとして，フィードバック流れと同位体平衡器を持った単一の深冷蒸留塔が研究対象となっている。この場合，塔は6成分（H_2，HD ，HT，D_2，DT，T_2）すべてを処理するが，目的はHとTの分離である。

フィードバック流れが存在するために生じる数学的シミュレーションの困難性を克服する1つの手法が示され，かつ，サイドカット段が塔の分離特性にいかなる影響を及ぼすかについて調べられている。

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1. Introduction

Cryogenic distillation is doudtlessly the most promising candidate for hydrogen isotope separation process in the mainstream fuel circulation system for fusion reactors. There are a number of other situations in which hydrogen isotope separation is needed, and cryogenic distillation is being studied as a possible method for recovering tritium from glovebox atmospheres and other similar sources.[1]~[3] The input conditions and the separation requirements in this case are expected to be different from those for the fuel circulation system : 1) the percentage of protium in the feed is much larger[4]; and 2) protium and tritium will be separated without any regard to deuterium. Deuterium is not the impurity for the fusion reaction, and it does not have any radioactive hazard. Hence, deuterium is allowed to be contained both in the protium waste and in the recovered tritium.

The column which could meet these requirements is shown in Fig. 1. The gas stream withdrawn from the column is transferred to the equilibrator which is operated at room temperature. After achievement of equilibration of the six isotopic species of molecular hydrogen (H_2, HD, HT, D_2, DT and T_2), the gas is liquified and recycled to the feed stream. Because of this feedback stream, rigorous simulation of the column is not easy to perform.

Kinoshita[5] made a detailed analysis of a cryogenic distillation column with a feedback stream which processes the three isotopic species : H_2, HT and T_2. He solved the simulation

problem by finding out the solution of a specific single-variable nonlinear equation. In the present case, however, the six isotopic species must be treated.

The main purpose of the present work is to propose a simulation procedure for the column with a feedback stream processing the six isotopic species, which is illustrated in Fig. 1. The effect of the sidestream location, a key parameter, on column performance is also investigated.

2. Mathematical Simulation Procedure

If the atom fraction of protium in the sidestream, z_H, and that of deuterium, z_D, are given, the composition and temperature of the stream fed to the column can be determined. Then, the composition and temperature distributions within the column are computed by the main distillation calculation. The computed values of the atom fraction of protium and deuterium in the sidestream, z'_H and z'_D, must be equal to z_H and z_D, respectively. However, since the former estimates are arbitrary ones, the errors espressed by

$$f_H = z'_H - z_H \quad , \quad f_D = z'_D - z_D \tag{1}$$

will not be zero. The most important feature in the present case is that the values of f_H and f_D are usually very small ($z'_H \simeq z_H$ and $z'_D \simeq z_D$) even if z_H and z_D are far from the solutions. For this reason, if the successive iteration method was used applying the criterion

$$|z'_H - z_H|/z'_H + |z'_D - z_D|/z'_D < \text{convergence tolerance } (\varepsilon), \tag{2}$$

many iterations would be needed requiring sufficiently small convergence tolerance.

In the rigorous simulation, all the factors inherent in hydrogen isotope separation by cryogenic distillation must be incorporated, such as differences in latent heat of vaporization among the six isotopic species, decay heat of tritium and nonideality of the hydrogen isotope solutions. As a consequence,

the simulation model for the main distillation calculation is complicated and the computation time needed is long. Therefore, the large number of total iterations due to application of the successive iteration method is not acceptable.

One of the methods which could overcome this problem is to solve the following nonlinear simultaneous equations :

$$f_H(z_H, z_D) = z'_H - z_H \ , \quad f_D(z_H, z_D) = z'_D - z_D \ , \tag{3}$$

by using the two-dimensional Newton-Raphson method. After initial estimation of the two independent variables (z_H and z_D), their values are repeatedly improved by

$$z_H^{i+1} := z_H^i - \{f_H(\partial f_D/\partial z_D) - f_D(\partial f_H/\partial z_D)\}/A \ ,$$

$$z_D^{i+1} := z_D^i - \{f_D(\partial f_H/\partial z_H) - f_H(\partial f_D/\partial z_H)\}/A \ , \tag{4}$$

$$A = (\partial f_H/\partial z_H)(\partial f_D/\partial z_D) - (\partial f_D/\partial z_H)(\partial f_H/\partial z_D) \ , \tag{5}$$

until Criterion (2) is satisfied, where i denotes the iteration number. The partial derivative is numerically calculated by using the approximation :

$$(\partial f_j/\partial z_k)\big|_{z_k} \simeq \frac{f_j(z_k + \Delta z_k) - f_j(z_k)}{\Delta z_k} \ . \tag{6}$$

This numerical calculation can be made with minor effort, because the values of the column variables for the main distillation calculation in the case of $z_k + \Delta z_k$ are very close to those in the case of Δz_k.[6]

3. Numerical Experiments

The effectiveness of the simulation procedure proposed in the previous chapter is now examined. The assumed input specifications and calculational conditions are given in Table 1. In the simulation model, all the factors previously described are incorporated. The nonideality of the hydrogen isotope solutions is estimated by using Souers' procedure.[7] The liquid holdups are approximately estimated by employing the technical data reported by Bartlit et al.[8] The equilibrium composition at 25 °C established by $H_2 + D_2 = 2HD$, $H_2 + T_2 = 2HT$ and $D_2 + T_2 = 2DT$ is computed by using the equilibrium constants of 3.234, 2.512 and 3.783, respectively.[9]

The simulation is made with the following initial estimates :

$$z_H = 0.4 \ , \quad z_D = 0.5 \ .$$

The calculation converges in five iterations as shown in Table 2. The simulation result is summarized in Table 3 and the liquid composition distribution within the column is shown in Fig. 2. It should be noted that the material balances around the column do not hold for the molecular species while they hold for the atomic species.

The conclusion is that the simulation procedure proposed presents rapid achievement of convergence.

Since the sidestream location is a key parameter, several calculations are made changing this parameter and keeping the other parameters unchanged. Mole fractions of HT in the two products are criteria of column performance. The relation between the sidestream stage number and column performance is

shown in Fig. 3. Although the top product is transferred to a tritium waste treatment system before being discarded to the environment, the tritium concentration in the top product needs to be kept below a certain level. On the other hand, the protium percentage in the bottom product must be sufficiently small (less than 0.01 atom%). This means that mole fractions of HT in both of the two products need to be sufficiently small. However, from Fig. 3, it is observed that the mole fraction of HT in the top product has an opposite dependency on the sidestream location if it is compared with the dependency of that in the bottom product. For this reason, determination of the optimum sidestream location is not easy to make. Nevertheless, the small number of the sidestream stage may be unacceptable due to high tritium concentration in the top product, while the large number results in too large protium percentage in the bottom product.
The optimum sidestream location may be approximately determined from the criterion, $N_S \simeq N_F$ (N_S and N_F denote the sidestream stage number and the feed stage number, respectively).

4. Conclusion

(1) A rigorous mathematical simulation procedure is proposed for a single cryogenic distillation column with a feedback stream, which processes the six isotopic species of molecular hydrogen. It is verified by numerical experiments that this procedure presents rapid achievement of convergence with acceptable iteration number of the main distillation calculations.

(2) The effect of the sidestream location on column performance is examined under an assumed external feed composition. The larger number of the sidestream stage results in smaller mole fraction of HT in the top product, whereas it results in larger mole fraction of HT in the bottom product. However, from the two curves of these two mole fractions plotted against the sidestream stage number, the ratio of N_S/N (N denotes the number of total theoretical stages) which could be proposed is 1/2. The feed stage number is fixed to be N/2 in the parametric survey for N_S, therefore, this criterion may be written by $N_S \simeq N_F$.

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The author wishes to express his sincere thanks to Prof. T. Takamatsu, Prof. I. Hashimoto (Kyoto University) and Mr. Y. Naruse (JAERI) for their comments on the present paper. Acknowledgment is also due to Dr. Y. Obata (JAERI) for his continuous encouragement.

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Table 1 Input specifications and calculational conditions

Flow rate of external feed = 100 g-mol/h
Composition of external feed : H = 20 atom%, D = T = 40 atom%
Flow rate of top product (vapor) = 25 g-mol/h
Reflux ratio = 25
Flow rate of sidestream (vapor) = 200 g-mol/h
Number of total theoretical stages = 80
Feed stage number = 40
Sidestream stage number = 30
Operating pressure = 760 Torr
Pressure drop : negligible
Condenser : partial condenser
Stream fed to column : liquid state
Liquid holdup : 0.1 g-mol/condenser, 1.0 g-mol/stage, 5.0 g-mol/reboiler
The superficial vapor velocity within the column is approximately 10 cm/sec.

Table 2 Rapid achievement of convergence in iterative calculation

Iteration number	z_H	z_D	z_T
1	0.40000	0.50000	0.10000
2	0.31556	0.56920	0.11525
3	0.31088	0.57408	0.11504
4	0.31081	0.57419	0.11500
5*	0.31081	0.57419	0.11500

* Converged to prescribed tolerance .

Table 3 Compositions of two products

	Composition of top product*	Composition of bottom product**
H_2	0.6007	0.2733×10^{-16}
HD	0.3987	0.2862×10^{-7}
HT	0.5897×10^{-3}	0.4723×10^{-4}
D_2	0.4958×10^{-5}	0.1830
DT	0.4650×10^{-8}	0.5696
T_2	0.1234×10^{-11}	0.2474

* H = 80.03 atom%, D = 19.94 atom%, T = 0.0295 atom%

** H = 0.00236 atom%, D = 46.78 atom%, T = 53.22 atom%

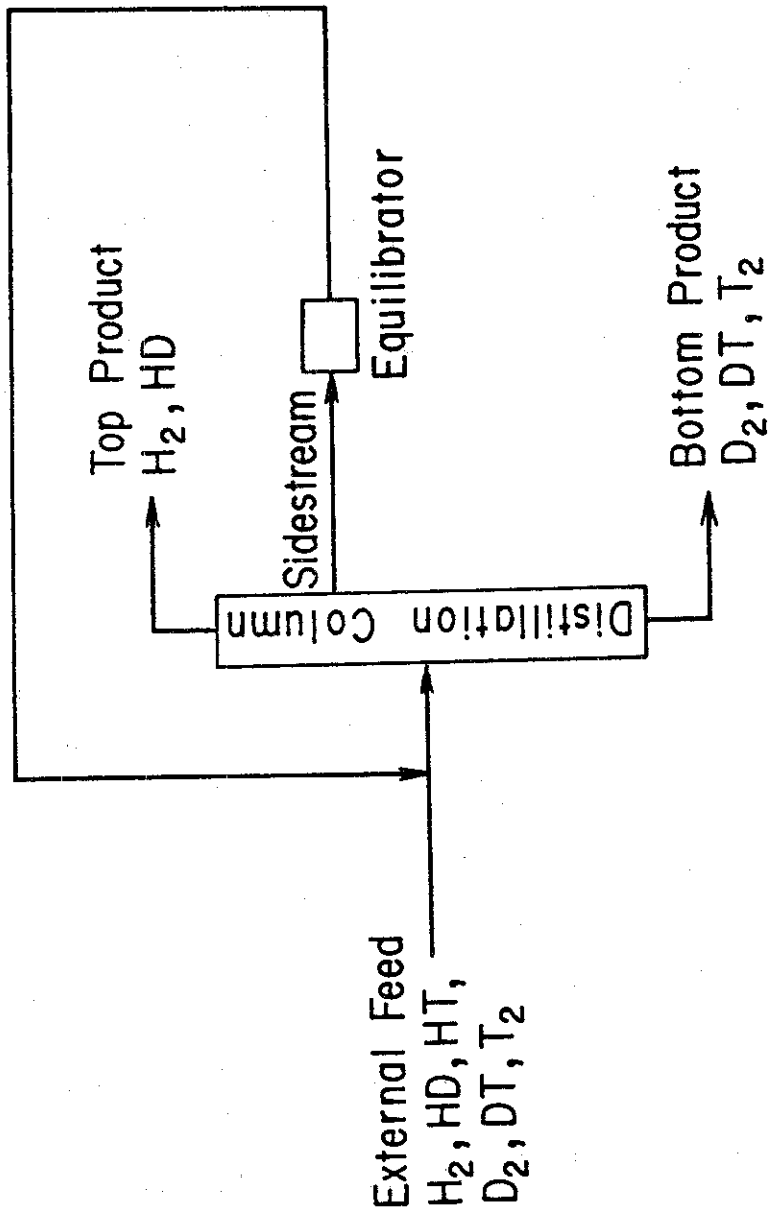

Fig. 1 Conceptual flow diagram of cryogenic distillation column as unit process of tritium system for recovering tritium from glovebox atmospheres and other similar sources

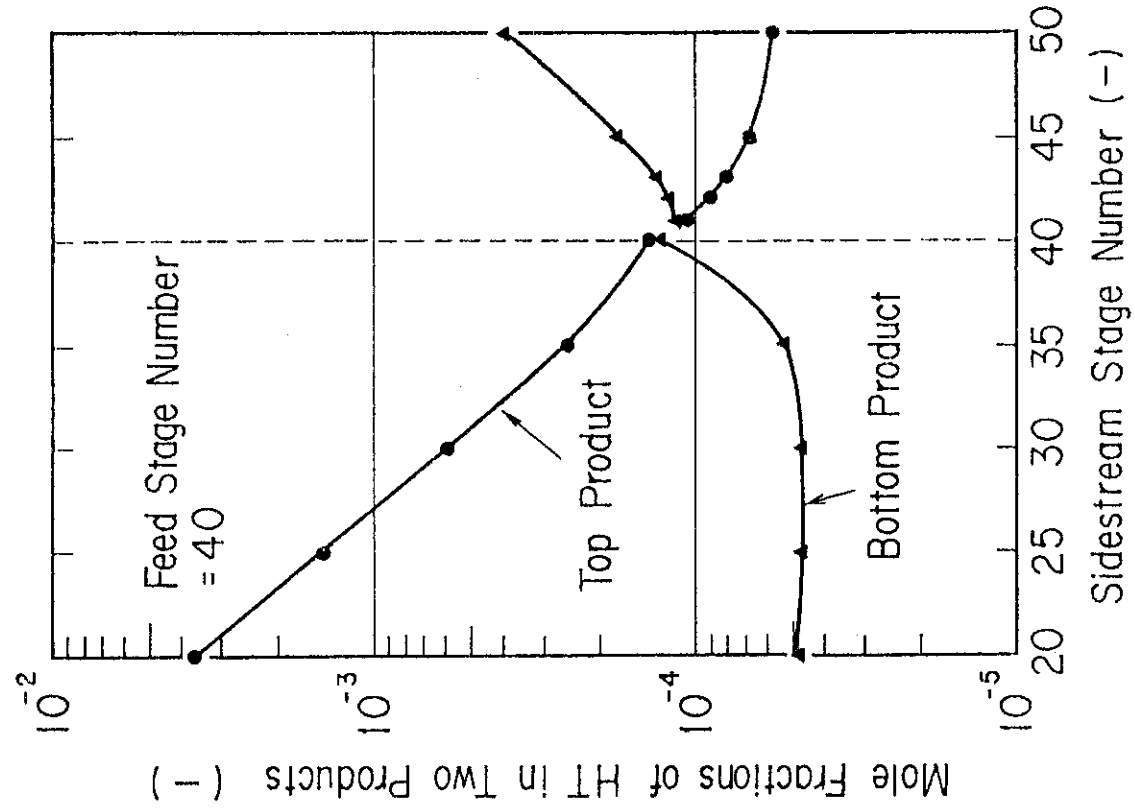

Fig. 3 Effect of sidestream location on column performance

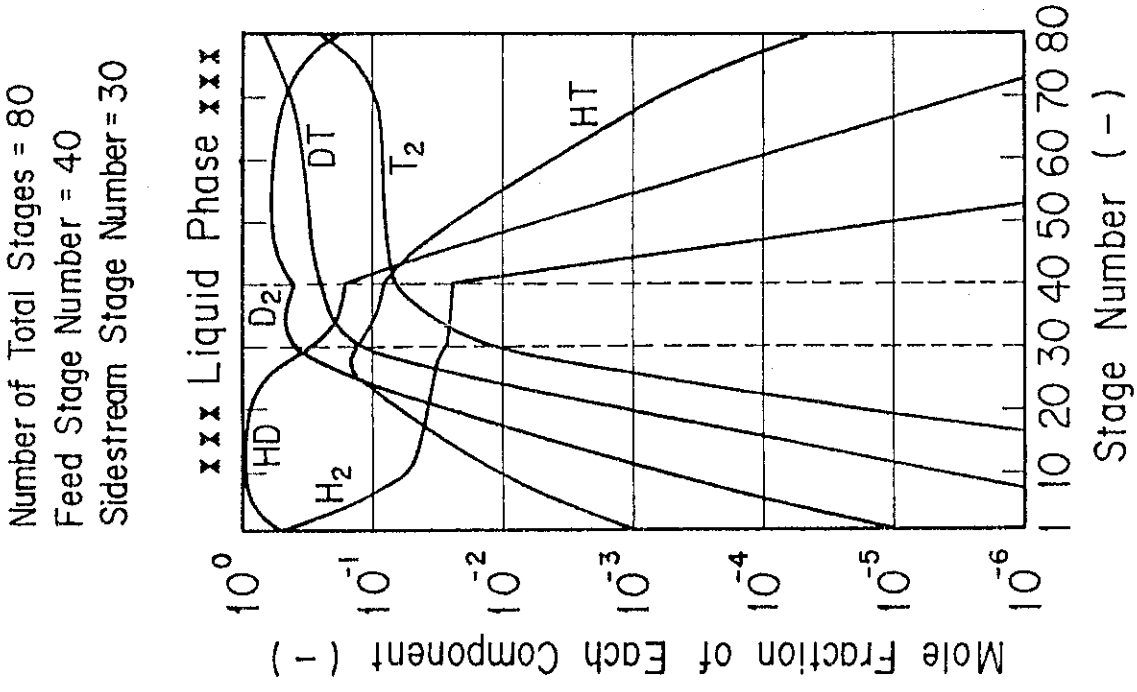

Fig. 2 Example of liquid composition distribution within distillation column