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A NEW SYSTEM FOR COMPLETE SEPARATION OF ^3He AND T_2 COMPOSED OF A FALLING LIQUID FILM CONDENSER AND A CRYOGENIC DISTILLATION COLUMN WITH A FEEDBACK STREAM

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A new system composed of a falling liquid film condenser and a cryogenic distillation column with a feedback stream, is developed for complete separation of ^3He and T_2. For accomplishment of the separation, a sufficient flow rate of protium is added to the feed mixture. The resultant stream of ^3He, H_2, HT and T_2 is fed to the falling liquid film condenser, and ^3He is removed almost completely. The H-T mixture from the bottom of the falling liquid film condenser is further processed by the cryogenic distillation column for complete separation of protium and tritium. The tritium recovery percentage of the system is 100 %, and the two top streams can be transferred to a tritium waste treatment system.

KEYWORDS : Helium, Tritium, Falling Liquid Film Condenser, Cryogenic Distillation, Feedback Stream, Tritium Recovery, Tritium Waste Treatment

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流下液膜式凝縮器及びフィードバック流れを持つ深冷蒸留塔で構成された^{3}He と T_2 の完全分離のための１つの新しいシステム

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流下液膜式凝縮器及びフィードバック流れを持つ深冷蒸留塔で構成された１つの新しいシステムを^{3}He と T_2 の完全分離用に提案する。完全分離を達成するため，充分な流量のプロチウムが原料流れに添加される。^{3}He，H_2，HT，T_2 の混合物が流下液膜式凝縮器に連続的に供給され，ここで ^{3}He が完全に除去される。塔底液（H－T混合物）は，次の深冷蒸留塔で処理され，HとTの分離が行われる。本システムのトリチウム回収率は100％であり，２つの塔頂ガスはそのままトリチウム廃棄物処理システムに送ることができる。

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1. Introduction

It is significant that tritium has a comparatively short half-life time (12.3 years). By decay of tritium, ^{3}He is produced ; if tritium has been stored for a significant period, a large percentage of ^{3}He is expected to be contained in the tritium. For this reason, removal of helium from tritium is needed in some situations for use of tritium.

One of the possible methods applicable to the present case is the Pd-alloy membrane diffuser, and it could be efficient in cases where the amount of tritium to be processed is relatively small. Some researchers,[1] however, have pointed out several disadvantages of the membrane. Besides, the method requires significantly high operating temperature and pressure, posing a serious problem of tritium permeation. Another feasible helium removal process is composed of a set of charcoal beds operated at liquid nitrogen temperature.[2] The process may be attractive in cases of the relatively small flow rate of tritium. Tritium is adsorbed on charcoal while helium goes through the beds without being adsorbed on charcoal. Tritium is recovered by warming beds to approximately 350 K, and T_2 and ^{3}He are thus separated. However, the process has a shortcoming that charcoal is combustible, and hence the process is not absolutely the best choice. For this reason, it is worth while to explore other processes particularly in cases of a large flow rate of tritium.

The authors[3]-[5] have analyzed separation characteristics of a falling liquid film condenser proposing this cryogenic process as the most attractive one for removing helium from hydrogen isotopes. By application of this process to the ^{3}He-T separation, however, the helium removed is unavoidably accompanied by approximately 10 % of tritium, while pure tritium almost completely free from helium is readily obtained. This requires another process for recovery of tritium from the helium waste. Though the set of charcoal beds may be applicable to recovery of tritium from the helium stream leaving the falling liquid film condenser,[2] the present study considers another method.

Our key proposal is addition of protium to the feed stream. A gas mixture of ^{3}He, H_2, HT and T_2 is continuously fed to the falling liquid film condenser and separated into two streams : a liquid of H_2-HT-T_2 withdrawn from the bottom and a gas of ^{3}He-H_2 leaving the top. The isotopic species, H_2, predominates over HT and T_2 in the hydrogen isotopes contained in the top gas, with the result that the tritium concentration in the top gas can be kept adequately low. The bottom product is separated into two pure streams of H_2 and T_2 by a cryogenic distillation column.

In the present paper, the conceptual flow diagram of the new helium removal system is described, and design specifications are determined in an example case.

2. Description of New Helium Removal System

Figure 1 shows a conceptual flow diagram of the new system composed of a falling liquid film condenser and a cryogenic distillation column.

The mixture of ^3He, H_2, HT and T_2 is fed to a point near the lower end of the packed section of the falling liquid film condenser. The tritium concentration in the top gas is sufficiently low ; the top gas is discarded to the environment or transferred to a tritium waste treatment system. This feature is ascribed to the simultaneous promotion of hydrogen isotope separation by the sufficiently long packed section of the falling liquid film condenser.

The hydrogen isotope mixture free from helium, which is obtained from the bottom, serves as the external feed to the cryogenic distillation column with a feedback stream and a catalytic equilibrator.[6] The vapor sidestream abundant in HT is withdrawn and transferred to the equilibrator for achievement of the equilibration, $H_2 + T_2 = 2HT$. The equilibrated gas mixture is then liquified and combined with the external feed. Separation of protium and tritium is thus performed and a stream of pure tritium is obtained from the bottom.

As previously described, if removal of ^3He from tritium was projected by a falling liquid film condenser alone without the idea of addition of protium to the feed, separation of ^3He and tritium would not be complete because a significant amount of tritium would be contained in the top gas.

3. Example of Design Calculation

3.1 Falling Liquid Film Condenser

The percentages of ^{3}He, H and T in the feed are assumed to be 10 %, 27 % and 63 %, respectively. The quantity of protium to be added greatly depends upon the helium percentage in tritium. The resultant percentage of protium must be adequately high in comparison to the helium percentage.

If it was not adequate, the tritium concentration in the top gas would be difficult to keep below a sufficiently low level.

The information obtained from our previous analyses[3]-[5] is useful in determination of input parameters for the design calculation. The input specifications are assumed as given in Table 1, and the corresponding column performance is analyzed by the computer code, FLFC.

Since no data on the vapor-liquid equilibrium of ^{3}He-H_2 or ^{3}He-T_2 system is available, the formulations previously reported by the authors for calculation of the vapor-liquid equilibrium of ^{4}He-hydrogen isotopes is also used in the present case. As a consequence, the helium concentration in the liquid phase and in the bottom product will be accompanied by an inaccuracy while the other variables will not be affected. However, the helium concentration in the bottom product is always very low, and hence it is not a critical parameter to be accurately estimated.

The simulation result is summarized in Table 2. The tritium percentage in the top gas is approximately 10^{-5} atom% which is adequately small. This number can be further decreased by addition of more amount of protium to the feed or by a longer packed section. The temperature at the top of the column is approximately 19.7 K : the inlet temperature of the refrigerant (helium gas) must be adequately lower than 19.7 K and higher than the freezing point of H_2 (~ 14 K). The pertinent inlet temperature of the refrigerant is expected to be in the range from 17 K to 18 K. The bottom product of liquid hydrogen isotopes free from ^3He is processed by the cryogenic distillation column for separation of protium and tritium.

3.2 Cryogenic Distillation Column

Kinoshita[6] performed in his previous work detailed computer analysis on separation characteristics of an H-T separating column with a feedback stream and a catalytic equilibrator. The information reported in his paper is sufficient to determine input parameters for the design calculation. Among these parameters, the pertinent sidestream location and the sidestream flow rate vary greatly depending on the external feed composition. The input parameters are assumed as given in Table 3, and the corresponding column performance or output specifications are calculated by means of the computer code, CRYDIS-B.[6]

The simulation result is summarized in Table 4. As observed from the table, protium and tritium are almost perfectly separated by the column.

The new system composed of a falling liquid film condenser and a cryogenic distillation column with a feedback stream presents a significantly high tritium recovery percentage (100 %) as proved below :

Flow rate of tritium fed to the system = 90 X 2 X 0.70
= 126 g-atom/h

Flow rate of tritium lost from the system =
$12.5 \times 2 \times (0.1594 \times 10^{-6}/2) + 24.5 \times 2 \times (0.4494 \times 10^{-5}/2)$
$= 1 \times 10^{-4}$ g-atom/h

The flow rate of tritium lost from the system is negligibly small in comparison to the flow rate of feed tritium.

4. Conclusion

A new system composed of a falling liquid film condenser and a cryogenic distillation column with a feedback stream, is developed for complete separation of ^3He and T_2. For accomplishment of the separation, a sufficient flow rate of protium is added to the feed stream. The resultant stream of ^3He, H_2, HT and T_2 is separated into two streams by the falling liquid film condenser : an essentially tritium-free stream of ^3He and H_2, and a stream of H_2, HT and T_2 almost completely free from ^3He. The latter stream is further processed by the cryogenic distillation column for separation of protium and tritium. Complete separation of ^3He and T_2 is thus achieved by the present system with the tritium recovery percentage of 100 %.

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References

(1) Kerr, E. C., J. R. Bartlit and R. H. Sherman : Proc. Tritium Technology in Fission and Isotopic Applications, Dayton, Ohio, April 29 - May 1, 1980, pp.115-118.

(2) Denton, W. H. and J. R. Bartlit : Private Communication, Los Alamos National Laboratory, 1978.

(3) Kinoshita, M., J. R. Bartlit and R. H. Sherman : "A Preliminary Design Model of Falling Liquid Film Condenser for Removal of Helium from Hydrogen Isotopes," Private Communication, Japan Atomic Energy Research Institute, 1981.

(4) Kinoshita, M., J. R. Bartlit and R. H. Sherman : Proc. 9th Symposium on Engineering Problems of Fusion Research, Palmer House, Chicago, Illinois, Oct.26-29, 1981, pp.1217 - 1220.

(5) Kinoshita, M., J. R. Bartlit and R. H. Sherman : JAERI-M 82-048, Japan Atomic Energy Research Institute, 1982.

(6) Kinoshita, M. and Y. Naruse : Nucl. Technol./Fusion, 2, 410 (1982).

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(6) Kinoshita, M. and Y. Naruse : Nucl. Technol./Fusion, 2, 410 (1982).

Table 1 Input Specifications of Falling Liquid Film Condenser

Flow rate of feed stream = 100 g-mol/h
Composition of feed stream : ^3He = 0.1000 , H_2 = 0.1000
HT = 0.3400 , T_2 = 0.4600
(The hydrogen isotopes are equilibrated by $H_2 + T_2 = 2HT$ at 25 ^oC)
Temperature of feed stream = 30.5 K (Gas)
Flow rate of top stream = 12.5 g-mol/h
Flow rate of bottom stream = 87.5 g-mol/h
Operating pressure = 4 atm
Number of total theoretical stages = 30
Cooled section = Stages 1 through 15
Packed section = Stages 16 through 29
Feed stage number = 28
Heat subtraction rate on cooled section = 6000 cal/h/stage

Table 2 Output Specifications of Falling Liquid Film Condenser

Composition of top stream : ^3He = 0.8000 , H_2 = 0.2000
HT = 0.1594×10^{-6}
T_2 = 0.3544×10^{-13}
Composition of bottom product : ^3He = 0.1735×10^{-7}
H_2 = 0.8573×10^{-1}
HT = 0.3886 , T_2 = 0.5257
Control heater duty = 76.9 W
Temperature at the top = 19.7 K
Temperature at the bottom = 29.5 K

Table 3 Input Specifications of Cryogenic Distillation Column

Flow rate of external feed = 87.5 g-mol/h
External feed composition : H_2 = 0.8573 X 10^{-1}
HT = 0.3886 , T_2 = 0.5257
Feed stream = Saturated liquid
Flow rate of top stream = 24.5 g-mol/h
Flow rate of bottom stream = 63 g-mol/h
Reflux ratio = 20
Operating pressure = 1 atm
Condenser = Partial condenser
Number of total theoretical stages = 60
Feed stage number = 30
Sidestream stage number = 25
Flow rate of sidestream (vapor) = 175 g-mol/h
Heat subtraction rate on the stripping section
(Stages 31 through 59) = 1500 cal/h/stage
Liquid hildups : 0.086 g-mol/condenser, 0.86 g-mol/stage
4.3 g-mol/reboiler

The differences in latent heat of vaporization among the three isotopic species, decay heat of tritium, and nonideality of the solutions are all incorporated in the simulation model.

Table 4 Output Specifications of Cryogenic Distillation Column

Composition of top stream :	H_2 = 1.0000 , HT = 0.4494 X 10^{-5}
	T_2 = 0.7802 X 10^{-12}
Composition of bottom stream :	H_2 = 0.8220 X 10^{-15}
	HT = 0.1753 X 10^{-5} , T_2 = 1.0000
Condenser duty = 108 W	
Reboiler duty = 151 W	
Tritium inventory :	183 g in packed section
	26 g in reboiler

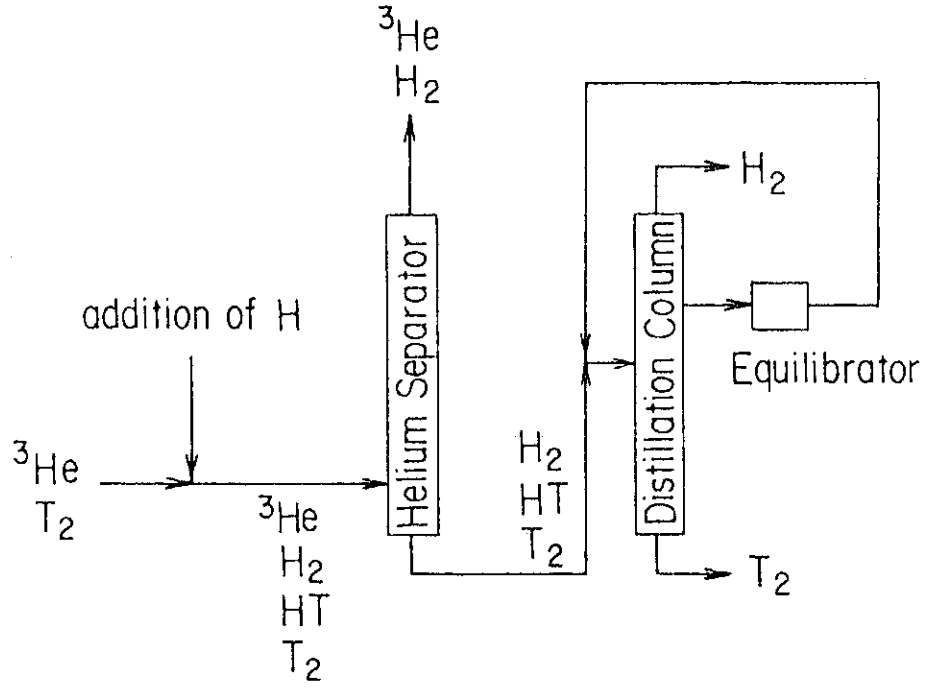

Fig.1 Conceptual Flow Diagram of New Helium Removal System