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Experimental Study for Cryogenic Distillation Column with N_2-Ar System

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Characteristics of cryogenic distillation columns having small inner diameters (1 and 2cm) were studied by separating N_2 and Ar. The columns were packed with Dixon Rings whose sizes were 1.5, 3.0, and 6.0mm. No significant effect of the vapor velocity within the column on the HETP was observed for the sizes of the packings and the column diameters tested. For both of the columns, the 1.5mm and 3mm Dixon Rings gave almost the same HETP values (5.5cm), whereas larger HETP values (8∿12cm) were obtained for thr 6mm Dixon Rings. A discrepancy between the calculated and experimental values of the Ar concentration at the middle of the column was observed in cases where the vapor velocity was small; this tendency was enhanced by the decrease in the sizes of the packings. The 3mm Dixon Rings presented the most excellent separation characteristics for both of the columns in terms of the HETP and the suitability of predicting column behavior by the stage model.

Keywords: Cryogenic Distillation, Dixon Rings, Inner Diameter, Packings, HETP, Stage Model

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窒素－アルゴン系を用いた深冷蒸留塔に関する実験研究

日本原子力研究所那珂研究所核融合工学部
山西　敏彦・木下　正弘*・奥野　健二・成瀬　雄二

（1991年8月22日受理）

深冷蒸留塔の分離特性を，N_2－Ar 系で，内径の小さい（1，2cm）塔及びサイズが1.5，3.0，6.0mm のディクソンリングを用いて測定した。充塡物のサイズ及び塔内径によらず，ＨＥＴＰに対する塔内蒸気速度の明確な影響は認められなかった。内径の異なる両方の塔に対して，1.5mm と3mm のディクソンリングはほぼ同じＨＥＴＰ（～5.5cm）の値を与えたが，一方，6mm のディクソンリングでは大きなＨＥＴＰ（8～12cm）を得た。塔中央部での Ar 濃度の実験値と計算値の不一致が蒸気速度の小さい範囲で認められたが，この傾向は，充塡物サイズが小さくなると共に顕著になることが認められた。3mm のディクソンリングが，結果として，ＨＥＴＰの値が小さい，段モデルによる塔挙動の予測が容易であるという点で，最もすぐれた分離特性を与えた。

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1. Introduction

Cryogenic distillation is a promising method for hydrogen isotope separation systems in the fuel cycle of nuclear fusion reactors. For analysis of separation characteristics of cryogenic distillation columns, the HETP (Height Equivalent to a Theoretical Plate) is one of the most significant parameters. Factors which can possibly affect the HETP values are as follows : the packing species, size of packings, vapor velocity within the column, column diameter, and physicochemical properties of the fluid. The authors have performed preliminary experimental studies for the cryogenic distillation(1)(2) with the N_2-Ar system to examine the above-mentioned factors affecting the HETP. A significant result obtained was that the HETP value was almost independent of the vapor velocity within the column(1). Four packing species, Dixon Rings, Heli-Pak, Helix, and Coil Pack were tested, and our conclusion was that the Dixon Rings presented the best set of results of the lower pressure drop and the smaller HETP(2). The authors have performed distillation experiments with the H-D system(3), and the above-mentioned conclusion for the dependency of the HETP on the vapor velocity was also verified for the hydrogen isotope separation column.

Since the effect of the packing species and the vapor velocity within the column on the HETP was reported in our previous papers, the next subject to be studied is the effect of the size of packings and the column diameter on the separation characteristics. Bartlit et al(5)(6) have reported experimental results, using a hydrogen isotope distillation column 0.95 cm in inner diameter and Heli-Pak and Eglin whose size was ~1.5 mm. Wilkes[8][9] have measured the HETP values for a hydrogen isotope distillation column (0.6 cm in inner diameter) packed with 3 mm Eglin. Thus, the previous experimental studies for the cryogenic distillation were not performed by changing the column diameter and the size of the packings. The HETP values for the packings differing in the size have been reported in a study for a water distillation column by Yamamoto et al.[10] Although their report produced useful information, they did not discuss the effect of the column diameter. There has been no effective information in selecting the size of packings and the column diameter for the cryogenic distillation column. For the above reason, in Tritium Systems Test Assembly at Los Alamos National Laboratory[5], the same packings (4.4 mm Heli-Pak) are used for four hydrogen isotope distillation columns different in the column diameter (The inner diameter of the four columns are 2.84 cm, 1.93 cm, 2.50 cm, and 3.80 cm, respectively). Further experimental studies are desired to overcome this problem.

The principal objective of the present study is to examine the effect of the size of packings and the column diameter on separation characteristics of the cryogenic distillation columns. Although the columns used in the present study separate N_2 and Ar, they simulate some significant features of a hydrogen isotope distillation column : the inner diameters and the sizes of packings are both very small ; the packing species used for the hydrogen isotope distillation is chosen for the measurement ; and the columns are operated by using liquefied gas at cryogenic temperature.

2. Experimental

Two columns 0.97 and 1.94 cm in inner diameter were packed with 1.5, 3, and 6 mm Dixon Rings to the height of 50 cm. The specifications of these Dixon Rings are summarized in Table 1.

The experimental apparatus and procedure were already described in Ref. (1). The experiments were performed at a total reflux operation mode, and dynamic variations of Ar concentrations at the top, middle, and bottom of the columns were measured by the gaschromatography.

3. Result and Discussion

In the experiment, the liquid holdups in the reboiler and that in the rest of the column can be measured. In computer simulation, however, separative values of the liquid holdups in the packed section and that in the condenser are also needed. Since it was verified by the computer simulation that the amount of the liquid holdup in the packed section had only a small effect on calculated results(1), the value used for the larger column packed with the 3 mm Dixon Rings in Ref. (1) and (2) (The amount is 15 % of the superficial volume of the packed section.) was applied in the present study. The detailed information on the computer simulation procedure was given in Ref. (1).

The distillation experiments were carried out under the vapor velocity range 10 ~ 30 cm/s for the larger column and 18 ~ 46 cm/s for the smaller column. The pressure drops across the

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The distillation experiments were carried out under the vapor velocity range 10 ~ 30 cm/s for the larger column and 18 ~ 46 cm/s for the smaller column. The pressure drops across the columns measured for the 6 mm, 3 mm, and 1.5 mm Dixon Rings increased in that order. The smaller column packed with the 1.5 mm Dixon Rings caused the flooding at the vapor velocity of 35 cm/s. The column operation was successful for the other cases.

3.1 Effect of the size of packings and the column diameter on the HETP

Figure 1 shows the HETP values measured under a variety of vapor velocity conditions. No significant effect of the vapor velocity on the HETP value is observed in all the cases tested. In addition, no considerable difference between the larger and the smaller columns in the HETP value is observed. The HETP value is 5.5 cm for the 1.5 and 3 mm Dixon Rings, whereas the 6 mm Dixon Rings gives appreciably larger HETP values (8~12 cm). The surface area of the Dixon Rings increases with decreasing the size as shown in Table 1. The smallest surface area of the 6 mm Dixon Rings would result in the largest HETP value. For the water distillation column, Yamamoto et al.[10] reported that the HETP values were 5~6 cm for the 1.5 mm Dixon Rings, 8~10 cm for the 3 mm Dixon Rings, and 6~7 cm for the 6 mm Dixon Rings : the effect of the size of the packings on the HETP was not clear for the water distillation column. The HETP is closely related to the effective vapor/liquid interface area within the column[2]. The effective interface area depends on wetting characteristics between liquid and the packings as well as the surface area of the packings. Since surface tension of water (60~70 dyn/cm) is much larger than that of N_2-Ar system (6~10 dyn/cm), it appears that for the water distillation column the increase in the surface area of the packings did not necessarily lead to that in the effective interface area. The hydrogen isotopes have small surface tension (2~6 dyn/cm) similar to that of the N_2-Ar system. The qualitative aspect for the effect of the size of packings on the HETP is expected to be applicable to hydrogen isotope distillation columns.

Figure 2 shows a comparison between the calculated result and the experimental observation for the smaller column packed with the 3 mm Dixon Rings. Figure 3 shows the experimental observation in which the 1.5 mm Dixon Rings was used. The experimental and calculated conditions of these representative runs are presented in Table 2. For the 3.0 mm Dixon Rings, the dynamic column behavior is adequately described by the computer simulation, and all the three calculated lines are in very close agreement with the experimental observation. In spite of almost the same experimental conditions as seen in Table 2, the Ar concentration at the middle did not agree with the calculated results for the 1.5 mm Dixon Rings. Table 3 shows approximate ratios of the experimentally observed values to the calculated values for the Ar concentrations at the middle of the columns. It should be noted that a smaller ratio means larger difference between the local HETP value defined for the upper half of the column and that for the lower half. In our previous study testing the four packing species[2], it was observed that the difference in the local HETP increased with the decrease in the vapor velocity. The difference was mainly due to the hydraulic effect caused by such factors as the variation in the amount of free liquid held near the wire supporting the packings and fluid mechanism of the liquid falling from the condenser. In addition, the hydraulic effect was significantly large for the Coil Pack having a densely coiled geometry. In the present study, as indicated in Table 3, the difference in the local HETP increased with decreasing the size of the packings and the vapor velocity. The smaller packings are more densely packed into the column. The dense packing appears to enhance the hydraulic effect. The inner diameter of the column can also affect the packed conditions. However, marked difference between the larger and smaller columns in the local HETP values was not observed under the present experimental conditions.

3.2 Information useful in selecting the size of packings and the column diameter

The size of packings is selected in order that the highest separation performance is obtained. In addition, to predict column behavior by the stage model, it is desired that the HETP value depends little on the column height. As a tendency common in the columns differing in the inner diameter, the 6 mm Dixon Rings gave lower separation performance than the others. The 1.5 mm Dixon Rings was unfavorable because of the higher pressure drop and the variation of the HETP across the column. For the above reasons, the 3 mm Dixon Rings appears to be best suited to cryogenic distillation columns having small inner diameters (1~2 cm). As described in the preceding section, the hydrogen isotopes have small surface tension, which is closely related to the HETP, similar to that of the N_2 -Ar system. The specific information obtained in the present study can be applied to hydrogen isotope distillation columns.

4. Conclusion

The HETP value was almost independent of the vapor velocity under the conditions of the very small inner diameters (1~2 cm) and the sizes of the packings (1.5~6 mm). For four combinations of the 1.5 and 3 mm Dixon Rings and the two columns used, almost the same HETP values were obtained. The 6 mm Dixon Rings gave larger HETP values than the others.

The difference between the local HETP values defined for the upper half of the column and that for the lower half increased with decreasing the size of the packings and the vapor velocity. The hydraulic effect (i.e. the variation in the amount of free liquid held near the wire supporting the packings and fluid mechanism of the liquid falling from the condenser) causes this difference, and the degree of the hydraulic effect depends on the vapor velocity and the size of packings.

The 3 mm Dixon Rings was best suited to the cryogenic distillation columns having small inner diameters (1~2 cm) with respect to the HETP, pressure drop, and constancy of the HETP along the column height.

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Table 1 Specifications of packing materials

Material	Dimension	Surface area	Description
SUS-316	1.5 mm x 1.5 mm	4140 m^2/m^3	42 x 100 mesh
SUS-316	3.0 mm x 3.0 mm	2378 m^2/m^3	42 x 100 mesh
SUS-316	6.0 mm x 6.0 mm	996 m^2/m^3	38 x 60 mesh

Table 2 Experimental and calculational conditions

	Run 1	Run 2
Initial Percentage of Ar in liquid phase (%)	34.4	35.5
Operating pressure (kPa)	134	136
Liquid holdups (mol)		
Condenser	0.7	0.7
Stage	0.022	0.022
Reboiler	3.5	2.6
Number of total theoretical stages	11	11
Vapor velocity (cm/s)	35.0	33.0
Size of packings (mm)	3.0	1.5
Inner diameter of the column (cm)	0.97	0.97

Table 3 Approximate rations of observed values to calculated values for Ar concentrations at middle of the column

Inner diameter of the column (cm)	Size of packings (mm)	Vapor velocity (cm/s) 20	30	35
0.97	1.5	0.2	0.4	0.5
1.94	1.5	0.2	0.3	—
0.97	3.0	0.6	0.9	1.0
1.94	3.0	0.9	1.0	—
0.97	6.0	—	1.0	1.0*
1.94	6.0	1.0**	1.0	—

* The vapor velocity is 46 cm/s.

** The vapor velocity is 9 cm/s.

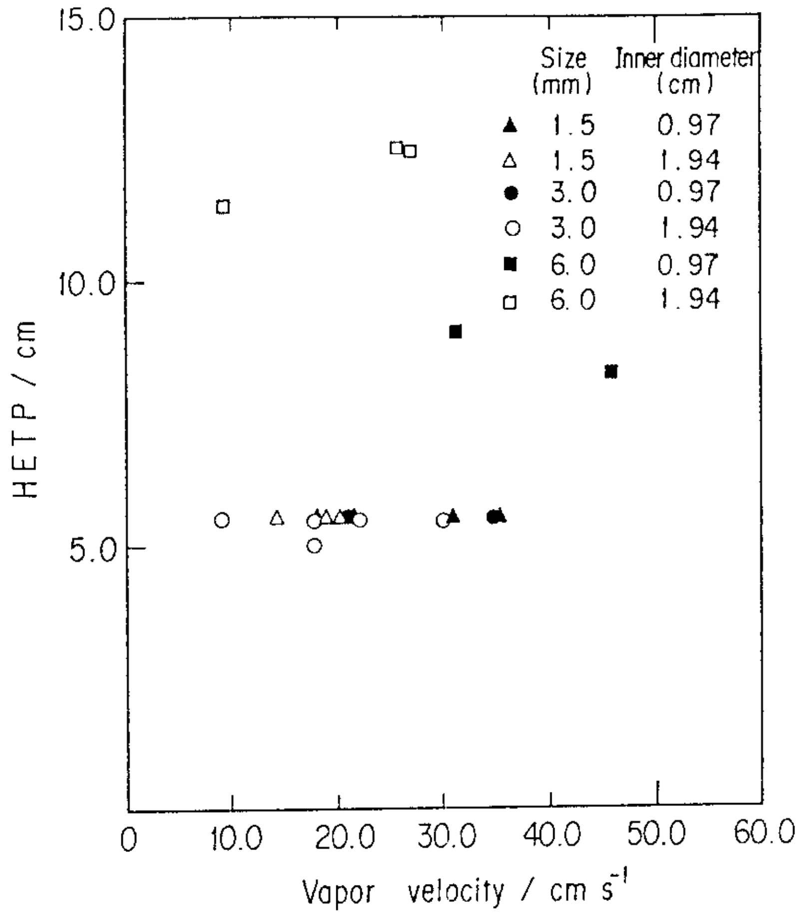

Fig. 1 Effect of the size of packings and the inner diameter of the column on the HETP

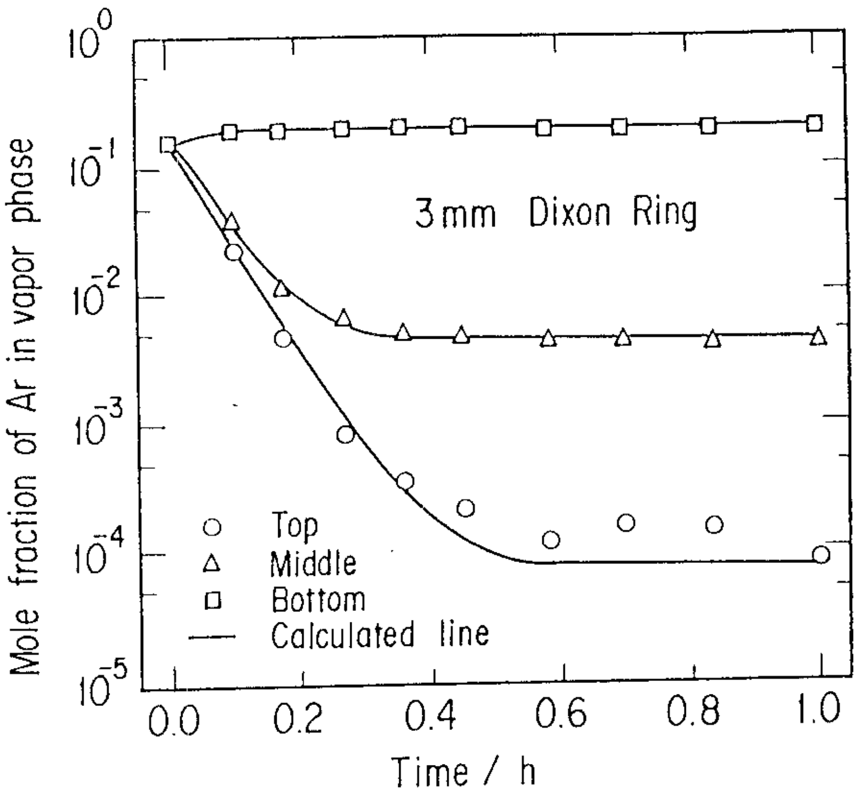

Fig. 2 Comparison between the experimental observation and the calculated result for Run 1

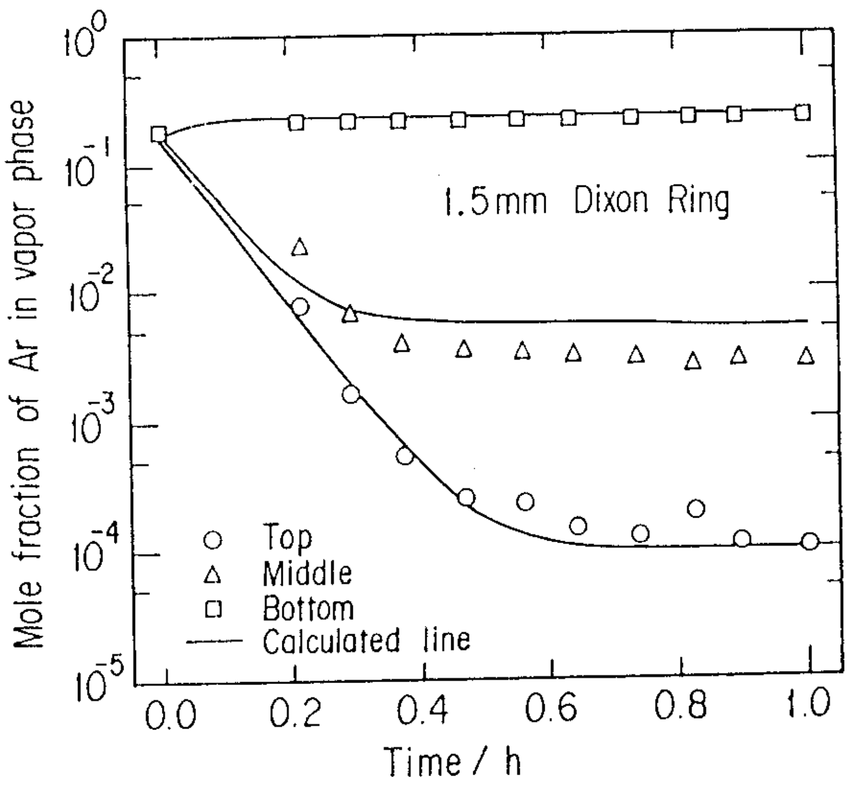

Fig. 3 Comparison between the experimentak observation and the calculated result for Run 2