TECHNICAL REPORT

Measurement of HETP of SUS Dixon Ring and Porcelain Packing in Small-Scale Water Distillation Column for $\text{H}_2\text{O-}{\text{HTO}}$ Isotope Separation

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Series of water distillations in a total reflux mode have been performed in a 100 cm height column of 1.6 cm I.D. in order to measure values of HETP for various packings, that is, one brass or three SUS Dixon rings and three porcelain packings. The HETP were measured by changing the vapor flow rate within the column. The SUS Dixon ring of 1.5 mm diameter and the porcelain packing of 1.2 mm O.D. had a small HETP (~5 cm), but could not meet a large vapor flow rate because of a large pressure drop. The SUS Dixon ring of 6.0 mm diameter had a small HETP (~6 cm) in the vapor flow rate under 2 g/min, but the HETP value increased with increasing the vapor flow rate. The pressure drop for the ring, however, was almost constant in the range of these measurements.

KEYWORDS: isotope separation, distillation, packed column, separation factor, HETP, total reflux, tritiated water, tritium, Dixon ring, porcelain packing, pressure drop

I. INTRODUCTION

Among various methods for separation of hydrogen isotopes(1)~(5), distillation of water has attractive possibilities in spite of its small separation factor because the process has inherent advantages such as: (1) simplicity of operation, (2) absence of corrosive and toxic compound $\text{H}_2\text{S}$ (used in the dual-temperature water-$\text{H}_2\text{S}$ exchange process for heavy water production), (3) absence of water-hydrogen gas conversion by electrolysis (in the water-hydrogen isotope exchange process), and (4) absence of catalyst for isotope exchange. Distillation of water has been, therefore, applied to the de-tritiation of drainage from a nuclear fuel reprocessing plant, and will be adopted for the volume reduction of tritiated water, based on tritium enrichment, from the cooling and safety systems of a nuclear fusion reactor.

A packed column will be used in a practical plant for tritium isotope separation, but available data are scarce for designing the optimal dimensions and operating conditions. A conceptual column design usually requires a value of HETP (Height Equivalent to a Theoretical Plate), to which an effective packed height of the column is directly proportional. The HETP value is affected by many parameters such as: (1) physical properties of gas and liquid, (2) dimension and material of packings, (3) flow rates of gas and liquid, (4) pressure and temperature within the column, and (5) structure and dimension of the column. The dependences of HETP on these parameters,

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however, have not sufficiently been studied theoretically nor experimentally, except for a preliminary experimental study for a cryogenic distillation column with small inner diameter by Yamanishi & Kinoshita(6)(7).

The purpose of the present report is to show measured values of HETP for tritium isotope separation using SUS Dixon rings and porcelain packings in a small-scale water distillation column, which was previously used to obtain the value of HETP for Dixon ring made of brass(8). The results, not treated theoretically here, are shown for practical use and for future's analysis of the dependence of HETP on various parameters.

II. EXPERIMENTAL

1. Apparatus of Water Distillation Column

The apparatus used for water distillation consisted of four sections; the reboiler, the column, the condenser and the associated reflux regulator, as shown in Fig. 1.

![Fig. 1 Schematic diagram of apparatus for water distillation](image)

The reboiler was a round-bottom 500 cm³ heat-resistant glass flask and was heated by a constant-temperature silicon oil bath (Toyo Seisaku-sho Co., VC-250). The vapor flow rate was controlled by changing the temperature of the oil. The heat-resistant glass column was 100 cm in length and 1.6 cm I.D. (~200 cm³). The column was insulated with a silver plated vacuum jacket (Kiriyama Seisaku-sho Co.). The vertical vacuum jacketed reflux condenser was cooled by water at 293 K. The reflux regulator consisted of two timers and a solenoid valve, dividing the condensed liquid intermittently (Kiriyama Seisaku-sho).

2. Experimental Procedure

After wetting the packings with the distilled water and feeding an initial charge into the reboiler, a total reflux distillation was performed under the atmospheric pressure. The volume and the tritium concentration of the charge were about 430 cm³ and approximately 1.0×10⁻⁸ Ci/cm³, respectively. The tritium concentration at the top of the column was measured every 30 or 60 min after the temperature of the water in the reboiler became 100°C. The condensed water of 1 cm³ was sampled by several extractions from the condenser with the reflux regulator, so that the extraction would cause no change in the flow rates of vapor and liquid within the column. In practice, 2 s extraction every 60 s was repeated till the volume of the sample exceeded 1 cm³. The tritium concentration of the sample was measured by a liquid scintillation spectrometer (Aloka model LSC602). Distillation was carried out to equilibrium, as shown in Fig. 2. The vapor flow rate at the top of the column was estimated by measuring the volume of condensed water within a certain period time, for example 60 s, at the condenser.

When the tritium concentration is very small, a total separation factor $\alpha_T$ of the column is defined and approximated as

$$\alpha_T = \frac{N_{\text{reboiler}}}{1-N_{\text{reboiler}}}/\frac{N_{\text{top}}}{1-N_{\text{top}}}$$

$$\approx N_{\text{reboiler}}/N_{\text{top}}$$

$$= C_{\text{reboiler}}/C_{\text{top}},$$

where $N$ and $C$ are mole fraction and concentration of the tritiated water, respectively.
Since an equilibrium separation factor $a_0$ of water distillation is expressed in terms of the vapor pressure $p$ of H$_2$O and T$_2$O by

$$a_0 = (p_{H_2O}/p_{T_2O})^{1/2}, \quad (2)$$

the HETP value and the number of theoretical plates $n$ are calculated in the following manner:

$$n = (\ln \alpha_T/\ln \alpha_0) - 1, \quad (3)$$

$$\text{HETP} = 100/n \, (\text{cm}). \quad (4)$$

As vapor pressure ratio obtained by Jones$^{(9)}$ is expressed in terms of temperature $T$ (K):

$$(p_{H_2O}/p_{T_2O}) = \exp\left(- \frac{103.87}{T} + \frac{46.480}{T^2}\right), \quad (5)$$

the equilibrium separation factor at 100°C is 1.0281. In addition, $F$-factor connected with a vapor loading is defined as

$$F\text{-factor} = (\text{Vapor velocity}) \cdot (\text{Vapor density})^{1/2} /
\sqrt{((\text{m/s})(\text{kg/cm}^3))^{1/2}}. \quad (6)$$

**III. RESULTS AND DISCUSSION**

**1. Porcelain Packings**

Specifications of three porcelain packings are summarized in Table 1. The present results of total reflux distillations using the porcelain packings are summarized in Table 2.

![Fig. 2 Variation of concentration of tritiated water at top of column](image)

The pressure drop in the table is calculated as

$$\text{Pressure drop} = (\text{Pressure drop across column}) \div 100$$

and the pressure drop across the column was estimated from the temperature of the water in the reboiler. The void fraction is calculated as

$$\text{Void fraction} = 100 \times \left(\frac{\text{Total volume of voids}}{\text{Volume of packed section of column}}\right) \times (\%)$$

The porcelain packing 1.2 mmφ had the small HETP. The packing, however, had a large pressure drop and could not meet a large vapor flow rate, because it would bring about a flooding phenomena. In the case where the oil bath’s temperature was 130°C, the packing had a large pressure drop (>2 Torr/cm) and the operation of the column could not be carried on due to the flooding except the case given in Table 2. If the inner void of all the packing is filled with water, the void fraction is reduced from 73.5 to 33.4%. It would be surmised that this reduction of voids brings about the high pressure drop for the packing.
The porcelain packings 3.0 and 5.0 mmφ had large values of HETP, so that they will not be suitable for tritium isotope separation by water distillation.

The vapor flow rate will be directly proportional to the difference between the oil bath's temperature and 100°C in the range of these measurements.

2. Brass Dixon Ring

A vapor flow rate dependence of the HETP value is shown in Fig. 3 for the brass Dixon ring, which was reported in a previous paper(8).

An effect of the vapor flow rate on the pressure drop for the ring is shown in Fig. 4.

The values of pressure drop were estimated from the temperature of the water in the reboiler, so they have errors due to (1) a limit of reading temperature, (2) an indirect correspondence of the temperature to the pressure drop because of other factors such as heat loss along the column. Moreover, the pressure drops were not reproducible in the series of measurements. Thereupon, Fig. 4 shows only a rough magnitude of the pressure drop and an example of the dependence on the vapor flow rate. The average diameter

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Table 1 Specifications of porcelain packings

<table>
<thead>
<tr>
<th>Porcelain packing</th>
<th>Length (mm)</th>
<th>Void fraction (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.2 mmφ</td>
<td>1.2 mm O.D.</td>
<td>4</td>
</tr>
<tr>
<td>0.9 mm I.D.</td>
<td></td>
<td>73.5</td>
</tr>
<tr>
<td>3.0 mmφ</td>
<td>3.0</td>
<td>5</td>
</tr>
<tr>
<td>1.0</td>
<td></td>
<td>53.0</td>
</tr>
<tr>
<td>5.0 mmφ</td>
<td>5.0</td>
<td>5</td>
</tr>
<tr>
<td>2.5</td>
<td></td>
<td>61.4</td>
</tr>
</tbody>
</table>

Table 2 Results of distillations using porcelain packings

<table>
<thead>
<tr>
<th>Porcelain packing</th>
<th>Oil bath's temperature (°C)</th>
<th>Vapor flow rate (g/min)</th>
<th>Pressure drop (Torr/cm)</th>
<th>αr</th>
<th>HETP (cm)</th>
<th>Equilibrium time (h)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.2 mmφ</td>
<td>121 ± 1</td>
<td>0.85</td>
<td>1.46 ± 0.34</td>
<td>1.86 ± 0.14</td>
<td>4.73 ± 0.61</td>
<td>8~9</td>
</tr>
<tr>
<td>1.0</td>
<td>129.4</td>
<td>2.7</td>
<td>0.42</td>
<td>1.77</td>
<td>5.1</td>
<td>~5</td>
</tr>
<tr>
<td>3.0 mmφ</td>
<td>128.8</td>
<td>2.4</td>
<td>0.28</td>
<td>1.28</td>
<td>12.5</td>
<td>3~4</td>
</tr>
<tr>
<td>1.0</td>
<td>120.8</td>
<td>—</td>
<td>0.28</td>
<td>1.27</td>
<td>13.0</td>
<td>3~4</td>
</tr>
<tr>
<td>5.0 mmφ</td>
<td>130.2</td>
<td>3.0</td>
<td>0.28</td>
<td>1.21</td>
<td>16.8</td>
<td>~3</td>
</tr>
</tbody>
</table>

Fig. 3 Effect of vapor flow rate on total separation factor and HETP for brass Dixon ring(8)

An effect of the vapor flow rate on the pressure drop for the ring is shown in Fig. 4.

Fig. 4 Effect of vapor flow rate on pressure drop for brass Dixon ring
and length of the rings were 4.8 and 4 mm, respectively. The void fraction for the ring was 93.4%.

The vapor flow rates of 1.3 and 6.5 g/min were obtained by the oil heated in 120 and 150°C, respectively. The HETP value was approximately 5.0–5.5 cm, and was not affected by the vapor flow rate in the range of these measurements. As shown in Fig. 4, the ring can meet a larger vapor flow rate due to a small pressure drop. The brass, however, is not corrosion-resistant in the atmosphere of water distilled column, and durability of the ring is a subject to solve.

3. SUS Dixon Rings

In Table 3, the dimension, material and void fraction are summarized for three Dixon rings, which are made of SUS and are resistant to corrosion. Figure 5 shows an effect of vapor flow rate on the HETP. In this figure, an open circle (○), triangle (△) and square (□) denote measured values in the case where the rings were packed only by the gravitational force into the column, while a closed circle (●), triangle (▲) and square (■) those in the case where the rings were packed as close as possible. A packing density, for example 0.93 g/cm³ in the figure, is calculated as

\[
\text{Packing density} = \frac{(\text{Total mass of packings } (g))}{(\text{Volume of packed section of column } (cm^3))}.
\]

An effect of the vapor flow rate on the pressure drop for the SUS Dixon rings is shown in Fig. 6.

<table>
<thead>
<tr>
<th>Dixon ring</th>
<th>Dimension (mm)</th>
<th>Material</th>
<th>Void fraction (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.5 mmφ</td>
<td>1.5 φ×1.5</td>
<td>SUS316</td>
<td>80.7±1.4</td>
</tr>
<tr>
<td>3.0 mmφ</td>
<td>3.0 φ×3.0</td>
<td>SUS316</td>
<td>88.9±0.6</td>
</tr>
<tr>
<td>6.0 mmφ</td>
<td>6.0 φ×6.0</td>
<td>SUS304</td>
<td>95.5</td>
</tr>
</tbody>
</table>

* Measured values in the case where the Dixon rings were packed as close as possible.

![Fig. 5 Effect of vapor flow rate on HETP for SUS Dixon rings](image_url)

In the case of the Dixon ring 1.5 mmφ, the HETP value is 5.3±0.7 cm and is not affected by the vapor flow rate in the range of these measurements. As shown in Fig. 6, the pressure drop for the ring 1.5 mmφ is little higher than that for the rings of 3.0 and 6.0 mmφ in the vapor flow rate under 1.26 g/min. The pressure drop for the ring

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1.5 mmϕ, however, increased extremely in the vapor flow rate over 1.26 g/min. Consequently, the ring could not meet a large vapor flow rate, because it would bring about the flooding phenomena.

The Dixon ring 3.0 mmϕ had the largest HETP among the three Dixon rings. The HETP value was approximately 9 cm in the vapor flow rate under 3 g/min, and increased with increasing the vapor flow rate in the range of 3~7 g/min.

The HETP value for the Dixon ring 6.0 mmϕ was approximately 6 cm in the vapor flow rate under 2 g/min. The HETP value increased with increasing the vapor flow rate in the range of 2~7 g/min, and was nearly equal to that for the ring 3.0 mmϕ in the vapor flow rate over 6 g/min.

A total surface area, which is a product of a surface area of one packing and a number of packings, for the SUS Dixon rings of 1.5, 3.0 and 6.0 mmϕ are 3,550, 2,300 and 900 cm², respectively. In the vapor flow rate under 2 g/min, the ring 3.0 mmϕ has a larger HETP value than the ring 6.0 mmϕ, although the ring 3.0 mmϕ have two and a half times the total surface area of the ring 6.0 mmϕ. The larger HETP of the ring 3.0 mmϕ is attributable to unused inner surfaces because of filling and/or blocking of the inner void of the ring by water drip and/or film at the bases of the cylindrical ring. It is because the surface tension of water is so large compared with the diameter of the ring that water drip and/or film sometimes cannot enter into the inner void area. The effective surface area for bringing vapor into contact with the liquid hold-up, therefore, decreases, and the HETP value increases.

The HETP value for the brass Dixon ring of 4.8 mmϕ was smaller than that for the SUS Dixon rings of 3.0 and 6.0 mmϕ. We suppose that the smaller HETP of the brass is due to its wettability for water; the brass ring is wet with water more easily than the SUS rings. It is because ① a wettability of brass with water is larger than that of SUS, ② oxidation of the brass makes the rings wet more easily, and ③ contamination of the SUS ring with organic matter makes the rings hard to be wet.

IV. CONCLUSION

Experiments of tritium isotope separation have been performed by water distillation. The HETP values were measured in the distillation column of a small inner diameter at a total reflux mode. Three porcelain packings (1.2, 3.0 and 5.0 mm O.D.), brass Dixon ring (4.8 mm diameter), and three SUS Dixon rings (1.5, 3.0, and 6.0 mm diameter) were tested by changing the vapor flow rate within the column. The results are summarized as follows:

1. The HETP value for the porcelain packing 1.2 mmϕ was small (≈5 cm), but the pressure drop for the ring was large. The HETP values for the porcelain packings 3.0 and 6.0 mmϕ were very large.

2. The brass Dixon ring can meet a larger vapor flow rate due to a small pressure drop, and had a small HETP (5.0~5.5 cm). The ring, however, is not resistant to corrosion.

3. The SUS Dixon ring 1.5 mmϕ had a small HETP (≈5.3 cm), but a large pressure drop. The ring 3.0 mmϕ had a large HETP (≈9 cm), and the HETP was affected by the vapor flow rate in the range over 3 g/min. The HETP value for the ring 6.0 mmϕ was small (≈6 cm) in the vapor flow rate under 2 g/min. The HETP, its value was smaller than that for the ring 3.0 mmϕ, was affected by the vapor flow rate in the range over 2 g/min.

REFERENCES

(7) idem: ibid., 21[11], 853.