

Numerical Estimation Method of the Hydrogen Isotope Inventory in the Hydrogen Isotope Separation System for Fusion Reactor

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In the fuel cycle system of the ITER, a large fraction of tritium inventory is expected to be in the cryogenic distillation columns of the hydrogen isotope separation system (ISS). Therefore, the numerical estimation method of hydrogen isotopes inventory in the ISS with high precision is strongly required from safety point of view. Two series of experiments were performed to establish the numerical estimation method of the overall hydrogen isotope inventory in the ISS at steady state using ITER-scale large cryogenic distillation columns at the Tritium Systems Test Assembly in the Los Alamos National Laboratory under the US-Japan collaboration on tritium safety engineering. As a result of experiments, it was confirmed that the hydrogen isotope inventory in a cryogenic distillation column was estimated by the numerical estimation method proposed in this work with enough high precision from the engineering point of view, and it was proved that this method was applied for the ITER-scale cryogenic distillation columns. The precision of this estimation method was found to be almost independent on the composition profile in columns, and especially the liquid holdup ratio of deuterium to the volume of the column was less influential in the inner diameter of the packed section. In addition, the gaseous inventory in an ITER-scale cryogenic distillation column was found to have considerable impact on the total amount of holdup of the column.

KEYWORDS: ITER, fusion reactors, tritium, inventory, cryogenic distillation, isotope separation, liquid holdup, tritium handling, numerical estimation, fuel cycle, hydrogen isotopes, ISS

I. Introduction

In the fuel cycle system of the International Thermonuclear Experimental Reactor (ITER),¹⁾ a fusion experimental reactor under designing internationally, a large fraction of tritium inventory is expected to be in the cryogenic distillation columns of the hydrogen isotope separation system (ISS).¹⁻⁵⁾ Tritium inventory in the ISS is much larger than other sub-systems in the fuel cycle system because hydrogen isotope is held mainly as liquid in it. The estimated tritium inventory in the ISS of the ITER designed in the period of 1992–1998 (FDR-ITER) was 230 g.⁶⁾ The value of 230 g was approximately estimated because the inventory estimation method with enough high precision was not established, and it was done mainly considering the amount of liquid hydrogen in the packed section by using some experimental data for a hydrogen liquid holdup ratio per unit volume of the column. The liquid inventories in the reboiler and in the condenser, which depend on the operation conditions, were generally calculated with some assumptions such as ignoring gas holdup.⁶⁾ Design study of the ISS for the ITER is ongoing and its evaluation method is one of the disputable points in the design work. The concept adopted in it is that the tritium inventory in the ISS should be as low as achievable from safety point of view. Therefore, the numerical estimation method of inventory of hydrogen isotopes in the ISS with high precision is strongly required. In addition, the grasp of the amounts of inventory in the reboiler, packed section and condenser is essential to simulate the dynamic behavior of the ISS with high precision.

There are still future subjects to estimate overall hydrogen isotopic inventory in an ITER-scaled cryogenic distillation column with high precision. The inventory in a column consists of gaseous inventory and liquid inventory. Gaseous inventory (gas holdup) can be evaluated by the isotopic distribution profile in a column and geometrical information of a column. The compositions are changing along column height, however numerical code has been developed to evaluate it by measurable parameters such as flow rates at strategic locations (feed, top, and bottom), pressure, liquid level in reboiler, pressure drop in column, and reboiler power.⁷⁾ Liquid inventory (liquid holdup) in the reboiler can be directly measured. As long as the distillation system is closed, that is, there are no input flow from other systems and no output flow to other systems, the liquid holdup in the other sections (packed section and condenser) can be estimated by subtracting the gas holdup and liquid holdup in the reboiler from the loading amount in a column. However, the ISS would not be a closed system in the planned operation scenario of ITER tritium plant, the establishment of evaluation method of liquid holdup is required. There are some fragmentary data concerning liquid holdup ratios for H₂ and D₂ in the packed section to its superficial linear velocity, which is the apparent velocity defined as volumetric flow rate divided by cross section.⁸⁾ The data for HD, HT, DT and T₂ are very difficult to be obtained by experiments, since the productions of pure HD gas, HT gas and DT gas are not actual and a lot of amount of pure T₂ gas is needed to measure the liquid holdup ratios for T₂. It is one of the most significant subjects to establish a reliable evaluation method of liquid holdup ratio of each hydrogen isotope in the packed section, which has been found to be a function

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of the superficial linear velocity in the column and of the isotopic composition, under a wide range of conditions. There are some data concerning the hydrogen isotopic holdup of liquid phase in condenser section from experiments with small cryogenic distillation columns (10 theoretical plates each), where the standard value of height equivalent to a theoretical plate (H.E.T.P.) for the hydrogen isotope distillation column is 0.05 m.⁹⁾ It has been reported that this holdup is not strongly dependent on the refrigeration duty but the condensed amount of hydrogen isotope can be estimated as 1–3% of condenser volume.¹⁰⁾ It is one of the subjects to be considered that this value could be adapted to the ITER-scaled cryogenic distillation columns (about 100 theoretical plates each) or not. Tritium is held mainly as the liquid in a lower part of the column and the reboiler, so that the amount of liquid in the reboiler is a key parameter for the reduction of tritium holdup. Whereas, some extent of liquid is needed in the reboiler to avoid dry run. The minimum amount of the liquid in the reboiler is a function of reboiler heater power.

To discuss the subjects mentioned above, some experiments were carried out by using an actual ITER-scaled ISS of the Tritium Systems Test Assembly in the Los Alamos National Laboratory under the US-Japan collaboration. The aim of this study is to establish a numerical model to evaluate the hydrogen isotope holdup in an ITER-scaled cryogenic distillation column at the steady state under various cases, even where performing of experiment is actually restricted by the limitation of tritium loading amount.

II. Experiments

There is no experimental study concerning holdup profile in a distillation column of ISS before this work, so that two series of experiments were planned and performed in order to establish the estimation method of the overall hydrogen isotope holdup at steady state by using cryogenic distillation columns with about 80 theoretical plates, there are called as Column H and Column I at the Tritium System Test Assembly in the Los Alamos National Laboratory.¹¹⁾ **Table 1** shows the specifications of Column H and Column I.¹¹⁾ The series of experiments using Column H and Column I are called hereinafter Series A and Series B, respectively.

1. Series of Experiments by Column H (Series A)

This series of cryogenic distillation experiments was carried out using three hydrogen isotopes; H₂, HD and D₂. **Figure 1(a)** shows the flow diagram for Series A. The experiments were carried out as the following steps. The known volume of hydrogen isotopes was preliminary loaded into a standard volume. In Series A, the amount of 6.93 mol each of pure H₂ and D₂ was loaded into it from gas cylinders. The amount of each hydrogen isotope loaded into the column was confirmed by using a combination of Pressure-Volume-Temperature measurements and Mass Spectrometry analysis. Distillation was initiated. All measurable parameters such as column temperature, column pressure, column differential pressure, reboiler heat, helium refrigerator temperature, flow rates of feed and products, liquid level and compositions at all locations were recorded. During the experiment, flow rates of

Table 1 Specifications of Column I and Column H

| | |
|---|---|
| Column I (Series A) | |
| Inner diameter | 28.4 mm |
| Packed height | 4.11 m |
| Volume | |
| Condenser | 1.9 × 10 ⁻³ m ³ |
| Packed section | 2.494 × 10 ⁻³ m ³ |
| Reboiler | 5 × 10 ⁻⁴ m ³ |
| Feed position from bottom of the column | 1.45 m |
| Packed material | Heli-Pak (316SS) |
| | 4.4 × 4.4 × 2.3 mm |
| Column H (Series B) | |
| Inner diameter | 19.3 mm |
| Packed height | 4.06 m |
| Volume | |
| Condenser | 1.7 × 10 ⁻³ m ³ |
| Packed section | 1.134 × 10 ⁻³ m ³ |
| Reboiler | 5 × 10 ⁻⁴ m ³ |
| Feed position from bottom of the column | 1.25 m |
| Packed material | Heli-Pak (316SS) |
| | 4.4 × 4.4 × 2.3 mm |
| Standard volume | |
| Volume | 0.265 m ³ |

feed and products were controlled by Mass Flow Controller and the compositions at all locations were measured by Mass Spectrometry. The gas at each location was sampled a few times into sampling cylinders at regular intervals. The steady state was experimentally judged by the agreement of analysed compositions of each location.

Table 2 shows the summary of loaded amount of hydrogen isotopes, column conditions and observed liquid level at steady state of Series A. In the table, each amount of hydrogen isotopes loaded in the column is summarized by the following equations;

$$[H] = [H_2] + \frac{[HD]}{2}, \quad [D] = [D_2] + \frac{[HD]}{2}, \quad (1)$$

where [Q] means the loaded amount of Q species.

2. Series of Experiments by Column I (Series B)

This series of cryogenic distillation experiments was carried out using six hydrogen isotopes; H₂, HD, HT, D₂, DT

Table 2 List of loaded amount into Column H and operation conditions

| | | | |
|--|---------|-------------------------|-------------------------|
| Loaded amount of protium ^{a)} | (mol) | 6.93 | 6.93 |
| Loaded amount of deuterium ^{b)} | (mol) | 6.93 | 6.93 |
| Top flow rate | (mol/s) | 7.44 × 10 ⁻⁴ | 7.44 × 10 ⁻⁴ |
| Bottom flow rate | (mol/s) | 7.44 × 10 ⁻⁴ | 1.49 × 10 ⁻³ |
| Pressure inside column | (kPa) | 106.9 | 109.7 |
| Reboiler power | (W) | 10.7 | 18.0 |
| Liquid level | (mm) | 54.00 | 41.00 |
| Reflux ratio | (—) | 17.0 | 29.0 |

^{a)} This equals [H] in Eq. (1).

^{b)} This equals [D] in Eq. (1).

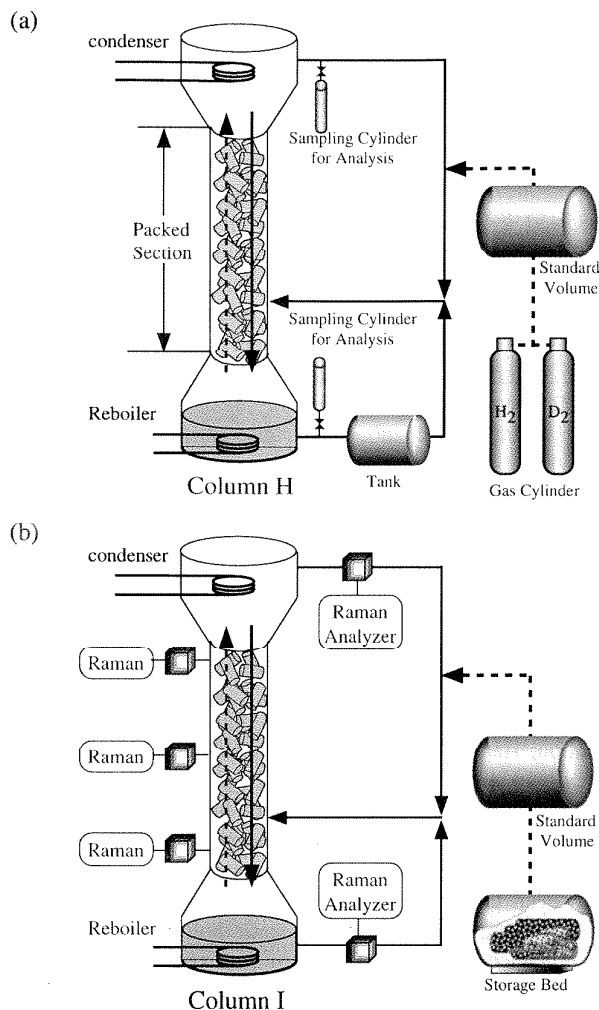


Fig. 1 Flow diagram of the experimental apparatus: (a) Series A and (b) Series B

and T_2 . The Column I is geometrically different from Column H, so that we can discuss the geometrical dependency upon our proposed method. Figure 1(b) shows the flow diagram for Series B. The experiments were carried out as the following steps. The known volume of hydrogen isotopes was preliminary loaded into the standard volume; 7.33 mol of H_2 , 10.88 mol of D_2 and 1.39 mol of T_2 . The amount of each hydrogen isotope loaded into the column was confirmed by using a combination of Pressure-Volume-Temperature measurements and Raman Spectroscopy analysis. All measurable parameters such as column temperature, column pressure, column differential pressure, reboiler heat, helium refrigerator temperature, flow rates of feed and products, liquid level and compositions at all locations were recorded. During the experiment, flow rates of feed and products were controlled by Mass Flow Controller and the compositions at all locations were measured by Raman Spectroscopy. The gas at each location was analysed at regular intervals. The steady state was experimentally judged by the agreement of analysed compositions of each location. Some amounts of hydrogen isotope were then intentionally removed from the col-

umn to consider the impact of loaded amount on the proposed estimation method. The amount of each hydrogen isotope remained into the column was confirmed by using a combination of Pressure-Volume-Temperature measurements and Raman Spectroscopy analysis.

Table 3 shows the summary of loaded amount of hydrogen isotopes, column conditions and observed liquid level at steady state of Series B. Each amount of hydrogen isotopes loaded in the column is summarized by the following equations;

$$\begin{aligned} [H] &= [H_2] + \frac{[HD]}{2} + \frac{[HT]}{2}, \\ [D] &= [D_2] + \frac{[HD]}{2} + \frac{[DT]}{2}, \\ [T] &= [T_2] + \frac{[HT]}{2} + \frac{[DT]}{2}, \end{aligned} \quad (2)$$

where $[Q]$ means the loaded amount of Q species.

III. Procedure of Numerical Estimation of the Hydrogen Isotope Inventory in the Hydrogen Isotope Separation System

The purpose of this study is to suggest the procedure of numerical estimation of holdup profile in a distillation column of the ISS, which is not formulated before this work. To evaluate the holdup profile, the experimental information we can observe in operating ISS is indirect parameters such as temperatures, pressure, flow rates of feed and product streams, and compositions of feed and product streams.

The following approach is employed. First, the hydrogen distribution profile in a column was estimated by the numerical code.⁷⁾ The decay heat of tritium and the nonideality of hydrogen isotopes are not taken into account in this study, because these effects on the design are not expected to be dominant. The code can describe the isotopic distribution profile in each theoretical stage. The agreement of calculated hydrogen isotopic distribution profile with experimental results was then checked. Figure 2 shows a typical example. The value of 0.05 m for the height equivalent to a theoretical plate (H.E.T.P.) was adopted in this analysis, so that the number of total stages is 83 which consist of the condenser (1st stage), the packed section (2nd–83rd stages), and the reboiler (84th stage). The value of 0.05 m is adopted as the standard for the ITER-ISS design. As seen in this figure, distribution of the hydrogen isotopes agrees well with the results of the analysis.

Secondly, the total holdup in a column (H_{te}) was estimated by dividing it into three sections that are the condenser section, the packed section, and the reboiler section. Each section is also divided into gas phase and liquid phase.

$$H_{te} = H_{VC} + H_{VP} + H_{VR} + H_{LC} + H_{LP} + H_{LR}. \quad (3)$$

The meaning of each symbol is summarized in **Nomenclature**. The hydrogen isotope holdups of gas phase at condenser (H_{VC}), packed section (H_{VP}), and reboiler (H_{VR}) are calculated from their volumes and their compositions as follows:

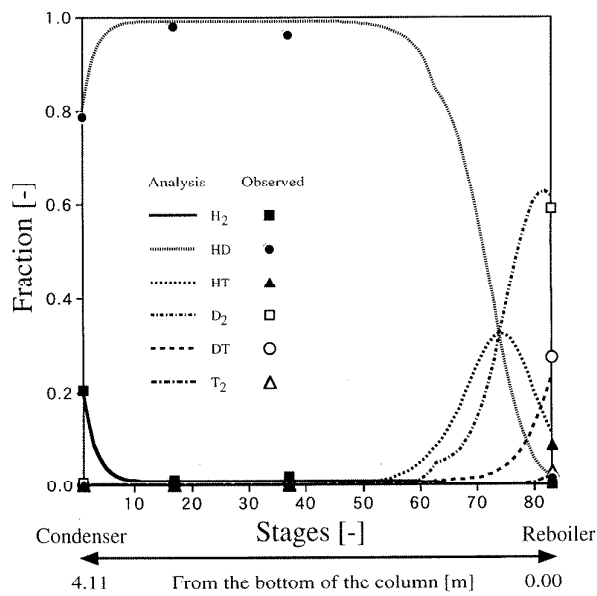
Table 3 List of loaded amount into Column 1 and operation conditions

| | | Run 3 | Run 4 | Run 5 | Run 6 | Run 7 |
|--|---------|-----------------------|-----------------------|-----------------------|-----------------------|-----------------------|
| Loaded amount of protium ^{a)} | (mol) | 7.33 | 7.33 | 7.33 | 4.28 | 4.28 |
| Loaded amount of deuterium ^{b)} | (mol) | 10.88 | 10.88 | 10.88 | 8.88 | 8.88 |
| Loaded amount of tritium ^{c)} | (mol) | 1.39 | 1.39 | 1.39 | 1.39 | 1.39 |
| Top flow rate | (mol/s) | 1.92×10^{-3} | 3.85×10^{-3} | 7.64×10^{-4} | 7.64×10^{-4} | 1.53×10^{-3} |
| Bottom flow rate | (mol/s) | 3.83×10^{-3} | 1.77×10^{-3} | 1.45×10^{-3} | 1.51×10^{-3} | 1.34×10^{-3} |
| Pressure inside column | (kPa) | 108.4 | 114.3 | 114.9 | 114.3 | 124.4 |
| Reboiler power | (W) | 34.3 | 34.3 | 36.2 | 17.8 | 17.6 |
| Liquid level | (mm) | 48.20 | 42.00 | 57.25 | 18.30 | 15.70 |
| Reflux ratio | (-) | 23.0 | 10.9 | 54.6 | 29.3 | 14.6 |

a) This equals [H] in Eq. (2).

b) This equals [D] in Eq. (2).

c) This equals [T] in Eq. (2).

**Fig. 2** Comparison of experimental and numerical column profile (Run 5)

$$H_{VC} = \sum_{i=1}^6 \frac{P_1 V_1 (1 - \varepsilon_1)}{RT_1} y_{1,i} \quad (4)$$

$$H_{VP} = \sum_{j=2}^{N-1} \sum_{i=1}^6 \frac{P_j V (1 - \varepsilon_j - \varepsilon)}{RT_j} y_{j,i} \quad (5)$$

$$H_{VR} = \sum_{i=1}^6 \frac{P_N (V_N - V_{LN})}{RT_N} y_{N,i} \quad (6)$$

The hydrogen holdup of liquid phase at reboiler (H_{LR}) is directly calculated from the liquid volume and reboiler composition:

$$H_{LR} = \frac{V_{LN}}{\sum_{i=1}^6 x_{N,i} v_{N,i}} \quad (7)$$

The hydrogen holdup of liquid phase at condenser (H_{LC}) is calculated from the composition of liquid phase and amount of liquid that is experimentally estimated as 1% of condenser

volume:¹⁰⁾

$$H_{LC} = \frac{V_1 \varepsilon_1}{\sum_{i=1}^6 x_{1,i} v_{1,i}} \quad (8)$$

The temperature dependency of liquid mole density (v_{ji}) was considered in this work.¹²⁾

There are two different types of liquid holdup in a packed section: static and operating. Static liquid holdup represents the liquid amount that remains in the packed section after the gas and liquid flows stop. This static holdup is dependent on the packed surface area, the roughness of packing surface and the contact angle between the packing surface and the liquid. However, this static holdup is normally not of great significance. The evidence that the static holdup is not of great significance can be derived from the experimental observation using the ISS in the TPL that liquid level of the reboiler was observed to be little changed before the reboiler heater was turned on and after it was turned off. When the reboiler heater is turned on, liquid level of the reboiler is observed to be lowered because some liquid in the reboiler is transferred to the packed section as liquid holdup. And the reboiler heater is turned off, operating holdup is down to the reboiler but static holdup will remain in the packed section. So if the static holdup is of great significance, liquid level of the reboiler after the reboiler heater is turned off is lower than that before the reboiler heater is turned on. However it was not observed. Hence, only operating liquid holdup was considered in this study. The operating liquid holdup at packed section (H_{LP}) is calculated from the following procedures. Packed section can be divided by each theoretical stage. The holdup of hydrogen isotopes in each theoretical stage is calculated from the holdup ratio, the composition and the volume of each theoretical stage. Total holdup of liquid phase at packed section is sum of these inventories in each theoretical stage:

$$H_{LP} = \sum_{j=2}^{N-1} \frac{V \varepsilon_j}{\sum_{i=1}^6 x_{j,i} v_{j,i}} \quad (9)$$

where the liquid holdup ratio of the packed section (ε_j) can

be expressed as

$$\varepsilon_j = \sum_{i=1}^6 x_{j,i} \gamma_{i,s_j} \quad (10)$$

The liquid holdup ratio of isotopic species i (γ_j) is reported as a function of the superficial velocity (S_j).⁸⁾ To obtain the superficial velocity of the column, at first, the vapor flow rate is calculated from the reboiler power, composition, and latent heat. The superficial velocity is then calculated from the vapor flow rate and cross section of the column. Strictly speaking, superficial velocity is also functions of pressure and temperature, so that in the calculation of superficial velocity of each theoretical stage, pressure and temperature behavior profile evaluated by the code⁷⁾ were considered. The data presented in Sherman's paper on liquid holdup ratios for H₂ and D₂ measured by Column-I were the only data measured by an ITER-scaled column before this work.⁸⁾ In this study, liquid holdup ratio for H₂ and D₂ both in Column-I and in Column-H is estimated based on the data presented in Sherman's paper. However, there is no data for HD, HT, DT and T₂. The subject is how to correlate these unknown data by using those for H₂ and D₂. The liquid holdup ratio would strictly be functions of viscosity, surface tension and density. However, it is difficult to derive an equation, in which above physical properties are directly used. Hence, let us suppose that each liquid holdup ratio for HD, HT, DT and T₂ can be expressed by the following linear model:

$$\gamma_{i,s_j} = \alpha_i \gamma_{1,s_j} + (1 - \alpha_i) \gamma_{4,s_j} \quad (i = 2, 3, 5, 6). \quad (11)$$

We correlate these unknown ratios (α_i) by trial and error method using our experiment data to minimize the error (F) of holdup between loaded (H_{te}) and estimated (H_{tm}):

$$F = \frac{|H_{tm} - H_{te}|}{H_{te}} \quad (12)$$

The liquid holdup ratios of isotopic species i as a function of the superficial velocity adopted in this study thus obtained are shown in Fig. 3. The results shown in Fig. 3 can be applied to the following cubic function model of superficial velocity:

$$\gamma_{i,s_j} = C_{1,i}(S_j)^3 + C_{2,i}(S_j)^2 + C_{3,i}S_j + C_{4,i} \quad (13)$$

Each constant was determined based on the results in Fig. 3 by the method of least squares. The estimated constants are listed in Table 4. Equation (13) can be applied with the range of $0.02 < S_j < 0.08$ m/s because liquid holdup ratios of H₂ and D₂ were experimentally obtained with this range.⁸⁾

The validity of this method for holdup estimation is discussed by comparison the calculated total holdup with the preliminary loaded volume of hydrogen isotopes.

IV. Results and Discussion

1. Operating Liquid Holdup in Packed Section

Large difference of operating liquid holdup ratios for each species is observed and estimated as shown in Fig. 3. It may be presumed that the operating liquid holdup would strongly depend on density, while the static liquid holdup would depend on some properties concerning the contact between the packing surface and liquid such as viscosity and surface ten-

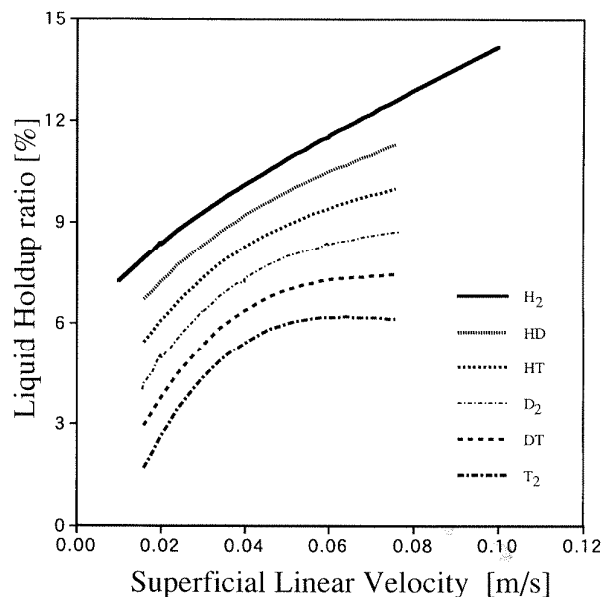


Fig. 3 Liquid holdup ratio of the packed section as function of the superficial linear velocity

The liquid holdup ratios of HD, HT, DT and T₂ adopted in this work were estimated from the observed results of H₂ and D₂.

Table 4 Constants for the liquid holdup ratios of isotopic species i

| i | $C_{1,i}$ | $C_{2,i}$ | $C_{3,i}$ | $C_{4,i}$ |
|-----|------------------------|-------------------------|-----------|-----------|
| 1 | 4.029×10^{-3} | -9.157×10^{-2} | 1.328 | 6.012 |
| 2 | 8.463×10^{-3} | -1.879×10^{-1} | 1.885 | 4.098 |
| 3 | 1.290×10^{-2} | -2.842×10^{-1} | 2.443 | 2.185 |
| 4 | 1.733×10^{-2} | -3.805×10^{-1} | 3.001 | 0.381 |
| 5 | 2.174×10^{-2} | -4.765×10^{-1} | 3.557 | -1.674 |
| 6 | 2.620×10^{-2} | -5.732×10^{-1} | 4.116 | -3.555 |

sion as mentioned earlier. Let us consider the effect of these properties on operating liquid holdup in packed section to clear what makes this difference. Table 5 shows the liquid hydrogen densities, viscosities and surface tensions at the temperature 22 K, which is the typical temperature in a column.¹²⁾ As for the viscosity, there are large differences among six species. There are little systematic data concerning the effect of viscosity on the liquid holdup in packed section. It was reported in them that liquid holdup is not strongly dependent on liquid viscosity and increases slightly with increasing it.¹³⁾ It can be explained qualitatively that viscous liquid tends to bridge the small void opening in packed section. The reported viscosity of D₂ is 2.8 times larger than that of H₂, while it was observed that the operating liquid holdup of D₂ is smaller than that of H₂.⁸⁾ This contradiction suggests that the effect of viscosity on operating liquid holdup is not high in hydrogen isotope distillation. Large differences of liquid surface tension are reported among six species as seen in Table 5. Liquid surface tension has practically slight effect on liquid holdup for high surface tension liquid such as water which surface tension is 7.275×10^{-2} N/m at the temperature of 20°C.¹³⁾ The

Table 5 Liquid hydrogen density, viscosity and surface tension at the temperature of 22 K

| | Density | | Viscosity (10^{-5} Pa·s) | Surface tension (10^{-3} N/m) |
|-------------------------|-----------------------|----------------------|--------------------------------|-------------------------------------|
| | (mol/m ³) | (kg/m ³) | | |
| <i>n</i> H ₂ | 34,192.0 | 68.9 | 1.14 | 1.655 |
| HD | 37,947.7 | 114.7 | 2.00 | 2.394 |
| HT | 39,587.0 | 159.3 | | |
| <i>n</i> D ₂ | 41,415.4 | 166.8 | 3.18 | 3.104 |
| DT | 43,042.8 | 216.5 | 3.8–4.3 | 3.826 |
| <i>n</i> T ₂ | 44,540.2 | 268.7 | 4.4–5.7 | 4.526 |

* *n*Q₂: para-Q₂25%+ortho-Q₂75%, Q=H, D, T

values of surface tension of hydrogen isotopes are smaller than that of water. There is no systematic data concerning the effect of such a low surface tension on liquid holdup. Even so, judging from such a small surface tensions of hydrogen isotopes and from the reported results that liquid surface tension has slight effect on liquid holdup,¹³⁾ the large difference of the liquid holdup ratios for each species cannot be explained by those differences of liquid surface tension. As a result, the effect of surface tension on operating liquid holdup is not high in hydrogen isotope distillation. As for the weight density, it is worth noticing that the weight density of D₂ is 2.42 times larger than that of H₂. There are large differences of weight density among six species. It can be explained qualitatively that the internal flow lifts the liquid isotopes against its weight in the packed section while the ISS is in operation, so that the species with smaller weight density would be lifted more in the column, that is, the operating liquid holdup ratio becomes larger value. As a result, large difference of each weight density is estimated to be a key parameter to explain the large difference among operating liquid holdup ratios of species shown in Fig. 3. Although the large difference among operating liquid holdup ratios of species will be caused by combined factors such as viscosity, surface tension and weight density, we interpret that it is mainly caused by large difference of each weight density of hydrogen isotope.

2. Series of Experiments by Column H (Series A)

We would like to examine the validity of the proposed method here. **Table 6** shows the estimated inventories in each part of the column H for Run 1. What is significant in this table is that the total estimated holdup is consistent with the total amount of hydrogen isotopes loaded in the column. As to the isotopic effect, a larger difference was seen between the estimated total and measured total of H (difference: 17.7%) in comparison with those of D (difference: -1.7%). Large amounts of H and D were evaluated to be existed in the packed section as liquid, so that the liquid holdup ratio is a key parameter to discuss it. The adopted liquid holdups for H₂ and D₂ shown in Fig. 3 are not measured with column H itself. It was reported that the liquid holdup ratio for H₂ is dependent on the column geometry to some extent.⁸⁾ The species with smaller weight density such as H₂ would be lifted more in packed section and the state of liquid dripping in packed section would have a tendency to effect on the

Table 6 Estimated hydrogen inventory in each part of Column H (Run 1)

| | H | D |
|-----------------------|-------|-------|
| Phase | | |
| Loaded amount (mol) | 6.929 | 6.929 |
| Liquid | | |
| Condenser (mol) | 0.625 | 0.075 |
| Packed section (mol) | 3.779 | 3.220 |
| Reboiler (mol) | 1.038 | 1.892 |
| Gas | | |
| Condenser (mol) | 1.267 | 0.092 |
| Packed section (mol) | 1.008 | 0.818 |
| Reboiler (mol) | 0.224 | 0.334 |
| Piping, etc. (mol) | 0.216 | 0.378 |
| Estimated total (mol) | 8.157 | 6.811 |
| Difference (%) | 17.71 | -1.71 |

arrangement of packing. Measuring the liquid holdup ratio for H₂ of each column is important for estimating the total holdup with high precision. Whereas, the weight density of D₂ is larger so that the amount of liquid holdup of D₂ per volume is not so different for column geometry. Good agreement between the estimated and loaded values for D is thus explained. The liquid holdup ratio for deuterium is less influential in column geometry. This fact would be applied on the liquid holdup ratio for tritium.

From the viewpoint of tritium accountancy in the ISS, it means that the tritium holdup in the packed section of an ITER-scaled cryogenic distillation column will be estimated with high precision by using the values shown in Fig. 3. However, the tritium is held as liquid not only in the lower part of the column but mostly in reboiler, so that the amount of liquid in the reboiler is important for tritium accountancy in the ISS, especially in case that the production of high purity T₂ gas is required. As for the liquid holdup in the reboiler, some amount of liquid is essential to prevent the "dry run" and to keep a steady state. The 21.1% of total holdup is in reboiler as liquid at the experimental conditions (see Table 6). The value of about 20% is typical in this work. To reduce the tritium holdup in a column, the control of liquid level is one of the efficient ways. The compatibility of this liquid-level control and other controls for product-compositions and for operation conditions such as pressure is one of the future subjects to study. It was confirmed that the amount of liquid holdup in the condenser is negligibly small as compared with those in other sections. Whereas, the percentages of gas holdup of H and D against the total loading amount (except in piping) reached to 31.5% and 19.4%, respectively. The main reason of such large amounts of gas holdup in the condenser is that the volume of condenser for the hydrogen isotope distillation column becomes inevitably large as seen in Table 1. This is because (1) large contact area is required in the condenser for the small heat transfer coefficient and small heat capacity of gaseous helium coolant, and (2) large temperature difference is not expected for the lower limit of gaseous

helium coolant temperature to avoid the solidification of hydrogen. The condensed amount of hydrogen isotope against condenser volume was estimated as 1% from our R&D result with small cryogenic distillation column. Judging from the result that total estimated inventory agreed with the loaded amount, this value is applied to the ITER-scaled cryogenic distillation columns. As seen in Table 6, it is worth noticing that the gas holdup in an ITER-scale cryogenic distillation column was found to have a considerable impact that should not be neglected mainly because of the large volume of condenser.

To verify the validity of the proposed estimation method of the holdup of the column, one more run (Run 2) was carried out under another conditions to make another composition profile in a column. Table 7 summarizes the comparison of the total estimated holdup with the loaded amount of hydrogen isotopes. As seen in this table, it can be concluded that both estimated amounts of D₂ for Run 1 and Run 2 are agreed well with the loaded amount under different column conditions. As for the H, some extent of disagreement was observed again. This disagreement was mainly caused by the same reasons discussed above. The tendency that the liquid holdup ratio for H₂ is more influential in column geometry than that for D₂ was observed by the experiment. The most important part of this argument is the total estimated holdup of Run 1 and Run 2 by using the liquid holdup ratio shown in Fig. 3 is consistent with the total amount of hydrogen isotopes loaded in the column.

3. Series of Experiments by Column I (Series B)

Table 8 shows the estimated inventories in each part of the column for Run 5 which column profile is shown in Fig. 2. The estimated amounts of D and T are consistent with the amounts of hydrogen isotopes loaded in the column, respectively. Furthermore, it is worth noticing that the estimated amount of H is also consistent with the loaded H in Run 5. This is because the adopted liquid holdup for H₂ shown in Fig. 3 is based on the experimental observation by using column I itself. From this result using six hydrogen isotopes, it is found that the hydrogen isotope holdup in a cryogenic distillation column was estimated by the proposed numerical estimation method with high precision. The 55.3% of tritium holdup is in reboiler as liquid at the conditions in the present case. As for the tritium inventory, the ratio of amount in reboiler against total increases more as product with higher tritium purity is required. The percentages of gas holdup of H, D and T against the total loaded amount are reached to 30.5%, 24.4% and 19.5%, respectively.

Table 7 Summary of overall hydrogen inventory estimation (Series A)

| | H | D |
|---------------------------|------|------|
| Loaded amount (mol) | 6.93 | 6.93 |
| Run 1 ^{a)} (mol) | 8.16 | 6.81 |
| Run 2 ^{a)} (mol) | 8.92 | 6.75 |

^{a)} Including the inventory in piping

Table 8 Estimated hydrogen inventory in each part of Column I (Run 5)

| | H | D | T |
|-----------------------|-------|-------|-------|
| Phase | | | |
| Loaded amount (mol) | 7.325 | 10.88 | 1.39 |
| Liquid | | | |
| Condenser (mol) | 0.503 | 0.38 | 0.00 |
| Packed section (mol) | 4.450 | 4.83 | 0.33 |
| Reboiler (mol) | 0.157 | 2.42 | 0.71 |
| Gas | | | |
| Condenser (mol) | 1.229 | 0.81 | 0.00 |
| Packed section (mol) | 0.959 | 1.07 | 0.09 |
| Reboiler (mol) | 0.054 | 0.60 | 0.16 |
| Estimated total (mol) | 7.351 | 10.12 | 1.29 |
| Difference (%) | 0.35 | -7.04 | -7.44 |

Table 9 Summary of overall hydrogen inventory estimation (Series B)

| | H | D | T |
|---------------------|------|-------|------|
| Loaded amount (mol) | | | |
| Run 3 (mol) | 7.07 | 9.88 | 1.18 |
| Run 4 (mol) | 6.81 | 8.38 | 2.13 |
| Run 5 (mol) | 7.35 | 10.12 | 1.29 |
| Loaded amount (mol) | | | |
| Run 6 (mol) | 4.51 | 6.77 | 2.00 |
| Run 7 (mol) | 4.26 | 7.69 | 1.24 |

Table 9 summarizes the results of the holdup estimation in five runs. It can be concluded that the holdup calculations agreed with experiments in most of the cases. The effect of the total loaded amount on the precision by this method is negligible. The precision of this estimation method is also found to be almost independent on the composition profile in a column as seen in Fig. 4, since no difference was observed between the results for H rich condition and that for T rich condition. And then, it is cleared that the liquid holdup ratio for D₂ is less influential in the inner diameter of the packed section. The reason for this is that each total estimated holdup of D not only in Table 9 using Column I (Runs 3-7) but in Table 7 using Column H (Runs 1-2) is well consistent with its total loaded amount of D. In some experiments in Table 9, disagreements were observed because of the following analytical reasons. (1) It spent about one hour to measure the compositions of all strategic locations. The steady state in Column I had not been maintained and the column profile was changed. The transit conditions of the column that cannot be well described with the simulation would result in this disagreement. (2) The trace amount of species was not detected by the Raman Spectroscopy. The disagreement was considered to be attributed for this error, especially for the tritium holdup. In a future work, the improvement for the analysis limit and time is desired to measure the compositions of feed

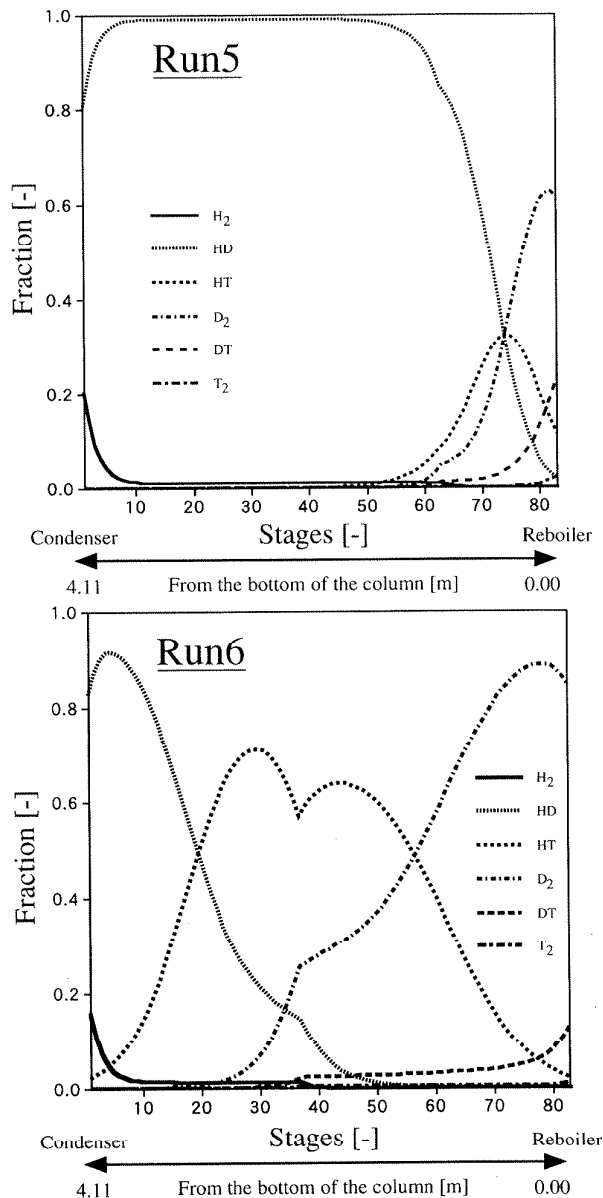


Fig. 4 Examples of isotopic distribution profile in Column H

and products of the column.

Consider lastly the validity of Eq. (11). Whereas we recognize that the ground why each liquid holdup ratio for HD, HT, DT and T_2 can be expressed by the linear model is not directly proven by experiments, from the evaluation using Eq. (11), enough examples of consistency between evaluated total holdup and the loading amount are presented in this work. For that reason, it seems valuable to suppose that each liquid holdup ratio for HD, HT, DT and T_2 can be expressed by the linear model.

V. Conclusions

The summaries of this study are as follows:

- (1) The hydrogen isotope holdup in a cryogenic distillation column is estimated by the proposed numerical estimation

method with high precision. The validity of this method was proved in the ITER-scale large cryogenic distillation columns.

- (2) The precision of this estimation method is almost independent on the composition profile in a column, since no difference was observed between the results for H rich condition and that for T rich condition.
- (3) The condensed amount of hydrogen isotope against condenser volume was estimated as 1% from our R&D result with small cryogenic distillation column. This value is applied to the ITER-scaled cryogenic distillation columns.
- (4) The liquid holdup ratio for deuterium is less influential than that for protium in the inner diameter of the packed section. This fact is applied on the liquid holdup ratio for tritium.
- (5) The gas holdup in an ITER-scaled cryogenic distillation column has a considerable impact on the total amount of holdup of the column.

The proposed numerical estimation method contributes the ISS design for the ITER. The goal of this study is to realize the on-line (dynamical) inventory measurement by this method. To achieve this goal, further experiments under a wider range of conditions should be performed.

Nomenclature

- F : Judgment index
 H_{te} : Total amount of hydrogen isotopes evaluated by the proposal method (mol)
 H_{im} : Total amount of hydrogen isotopes loaded in the column (mol)
 H_{LC} : Liquid holdup in the condenser (mol)
 H_{LP} : Liquid holdup in the packed section (mol)
 H_{LR} : Liquid holdup in the reboiler (mol)
 H_{VC} : Gas holdup in the condenser (mol)
 H_{VP} : Gas holdup in the packed section (mol)
 H_{VR} : Gas holdup in the reboiler (mol)
 P_j : Pressure at the j^{th} theoretical stage (Pa); Observed data
 R : Gas constant (Pa·m³/mol·K)
 S_j : Superficial velocity at the j^{th} theoretical stage (m/s)
 T_j : Temperature at the j^{th} theoretical stage (K); Observed data
 V : Superficial volume per theoretical stage in the packed section (m³)
 V_1 : Volume of condenser (m³); see Table 1
 V_{LN} : Liquid volume in the reboiler (m³); Observed data
 V_N : Volume of reboiler (m³); see Table 1
 x_{ji} : Liquid mole fraction of species i at the j^{th} theoretical stage
 y_{ji} : Vapor mole fraction of species i at the j^{th} theoretical stage
 α_i : Coefficient of correlation of species i
 γ_i : Liquid holdup ratio species i
 ε : Ratio of packing (20%)
 ε_1 : Liquid ratio against the condenser volume (1%)
 v_{ji} : Liquid mole density of species i at the j^{th} theoretical stage (mol/m³)
 ε_j : Liquid ratio against the volume at the j^{th} theoretical stage

(Subscripts)

- 1: Condenser
 i : Hydrogen isotopes (1=H₂, 2=HD, 3=HT, 4=D₂, 5=DT, 6=T₂)
 N : Reboiler

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