

# Tritium Release from Molten FLiNaBe under Low Flux Neutron Irradiation

Kohki KUMAGAI<sup>1)</sup>, Teruya TANAKA<sup>1,2)</sup>, Takuya NAGASAKA<sup>1,2)</sup>, Juro YAGI<sup>3)</sup>, Takashi WATANABE<sup>2)</sup>, Gaku YAMAZAKI<sup>1)</sup>, Fuminobu SATO<sup>4)</sup>, Shingo TAMAKI<sup>4)</sup>, Isao MURATA<sup>4)</sup>, Satoshi FUKADA<sup>5)</sup>, Kazunari KATAYAMA<sup>5)</sup> and Akio SAGARA<sup>1,2)</sup>

<sup>1)</sup>*SOKENDAI (The Graduate University for Advanced Studies), Toki 509-5292, Japan*

<sup>2)</sup>*National Institute for Fusion Science, Toki 509-5292, Japan*

<sup>3)</sup>*Institute of Advanced Energy, Kyoto University, Uji 611-0011, Japan*

<sup>4)</sup>*Graduate School of Engineering, Osaka University, Suita 565-0871, Japan*

<sup>5)</sup>*Interdisciplinary Graduate School of Engineering Science, Kyushu University, Kasuga 816-8580, Japan*

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Release behaviors of tritium from molten FLiNaBe (LiF–NaF–BeF<sub>2</sub>) have been examined in a series of low flux neutron irradiation experiments with an AmBe neutron source at the OKTAVIAN facility of Osaka University. Tritium released from FLiNaBe is swept by He and Ar gases, and recovered by water bubblers. The recovery rate of tritium released from 300 cm<sup>3</sup> of FLiNaBe was 58.0 mBq/h in a steady state, and it was close to the tritium production rate of 58.0 mBq/h calculated by the MCNP6 code. The values of the overall mass transfer coefficients for tritium release from FLiNaBe were close to those for FLiNaK (LiF–NaF–KF) at 773 K. The influence of the diffusivity and the solubility of tritium on the overall mass transfer coefficients has been examined on molten FLiNaBe.

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## 1. Introduction

Fluoride molten salts, for example, FLiNaBe, FLiBe (LiF–BeF<sub>2</sub>), and FLiNaK, are candidates of tritium breeder/coolants for a fusion blanket system because they have advantages in chemical stability and small MHD pressure drop under magnetic fields [1, 2]. Because FLiNaBe has the lower melting point (578 K) among the fluoride salts, efficient heat recovery is expected [3, 4]. Therefore, molten FLiNaBe has been selected as the breeder/coolant of the self-cooled blanket system in the design of the helical fusion reactor FFHR [1].

Understanding release behaviors of tritium from the molten salts is significantly important for tritium transport and tritium recovery in the blanket systems. Hydrogen transport behaviors in molten FLiNaBe were examined using H<sub>2</sub> gas as a simulant for tritium in a previous study [5]. However, release behaviors of tritium produced by nuclear reactions in molten FLiNaBe have not been clarified. In the present study, the low flux neutron irradiation experiments with an AmBe neutron source are conducted on molten FLiNaBe to examine the tritium release behaviors at the OKTAVIAN facility of Osaka University. This paper presents the first results obtained from molten FLiNaBe under the neutron irradiation.

In the present study, the overall mass transfer coeffi-

cients for tritium release from molten FLiNaBe at 673 K and 773 K were evaluated by conducting the neutron irradiation until the tritium release rate reached a steady state. In our previous study, the overall mass transfer coefficients for molten FLiNaK have already been evaluated at 773 K with the AmBe neutron source [6]. The overall mass transfer coefficient for tritium release from a molten salt is considered to be related to the diffusivity of tritium in a molten salt and a gas phase, and the solubility of tritium in a molten salt [7]. The influence of the diffusivity and the solubility of tritium in the molten salts on the overall mass transfer coefficients was examined in comparison with the results for FLiNaBe and FLiNaK. In addition, the overall mass transfer coefficients for FLiNaBe at 673 K were evaluated and compared with the results at 773 K for further discussions regarding mass transfer of tritium in molten FLiNaBe and a gas phase.

## 2. Experimental Setup

### 2.1 Neutron irradiation experiments

Figure 1 shows a schematic diagram of the neutron irradiation experiments on FLiNaBe. An Inconel 600 crucible containing FLiNaBe was irradiated with an AmBe neutron source (3 cm diameter × 6 cm). The volume of molten FLiNaBe was approximately 300 cm<sup>3</sup>. The neutron intensity of the AmBe neutron source was approxi-

author's e-mail: kumagai.kohki@nifs.ac.jp

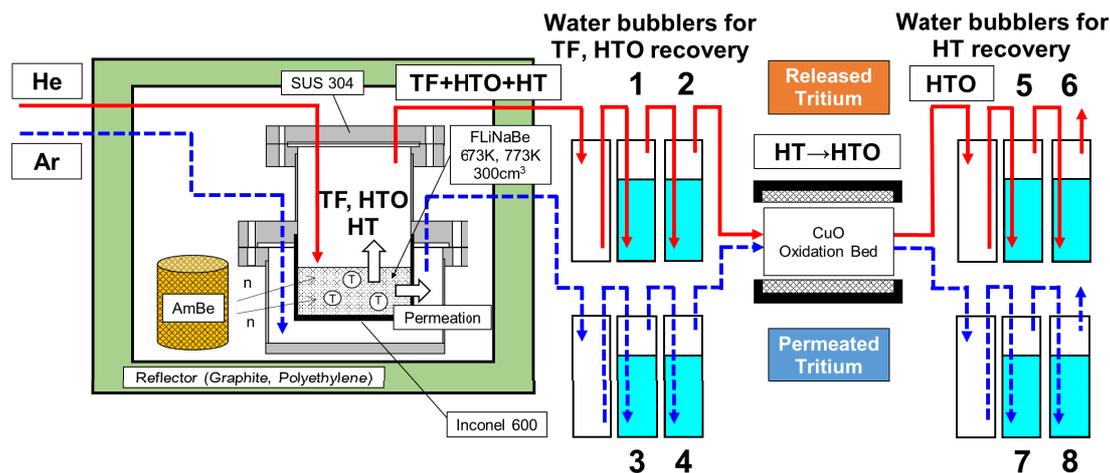


Fig. 1 Schematic diagram of irradiation experiments with an AmBe neutron source on FLiNaBe.

mately  $2 \times 10^6$  n/s. The composition of FLiNaBe was 1:1:1 mixture of LiF–NaF–BeF<sub>2</sub>. Each material was mixed and melted in the Inconel 600 crucible in Ar (5N) atmosphere in a glove box at Kyushu University. The details of the molten salt container and the setting position of the AmBe neutron source are described in the previous paper [6]. The FLiNaBe container consisted of an Inconel 600 crucible, Inconel 600 sheathed thermocouples, a flexible heater, a heat transfer compound, and a thermal insulator. The tips of the thermocouples were immersed in FLiNaBe.

Tritium is produced by the nuclear reactions of  ${}^6\text{Li}(n, \alpha)\text{T}$  and  ${}^7\text{Li}(n, n'\alpha)\text{T}$  in molten FLiNaBe. As shown in Fig. 1, tritium released from the free surface of FLiNaBe (i.e., Released tritium) and tritium permeating through the Inconel 600 crucible (i.e., Permeated tritium) were recovered by 25 Ncm<sup>3</sup>/min of He and Ar gases, respectively. Each tritium was recovered by water bubblers. The first stage of the bubblers (No. 1–4 in Fig. 1) recovered soluble tritium (TF and HTO) and the second stage of the bubblers (No. 5–8) recovered insoluble tritium (HT) by changing HT into HTO using CuO oxidation beds. Each set of bubblers (e.g., bubbler No. 1 and 2, etc.) had a vacant bubbler for receiving backwater. All of the bubblers were cooled to around 273 K to suppress evaporation of water during bubbling. The integrated amounts of tritium recovered by the water bubblers were measured by a liquid scintillation counter.

## 2.2 Purification of FLiNaBe

Prepared FLiNaBe was considered to include BeO since BeF<sub>2</sub> can contain the impurity. The impurity BeO could also be produced in the melting process of FLiNaBe. In order to change BeO into BeF<sub>2</sub>, the following chemical reaction was applied by introducing HF gas into molten FLiNaBe [5]:



Figure 2 shows a schematic drawing of the HF bub-

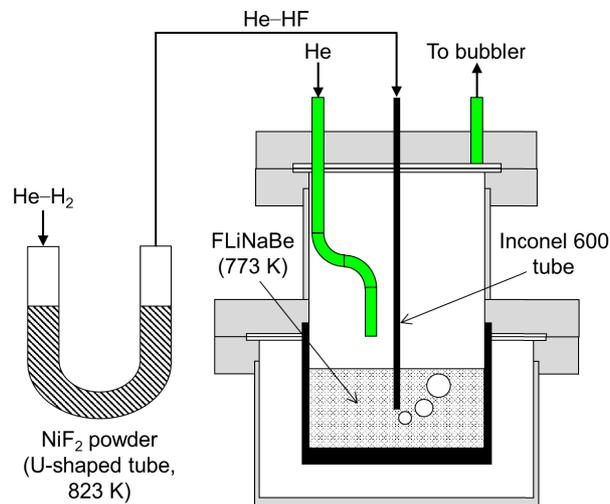
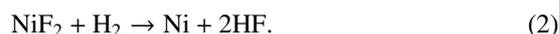


Fig. 2 Schematic drawing of the HF bubbling in FLiNaBe.

bling in FLiNaBe. The temperature of FLiNaBe was kept at 773 K during the HF bubbling. An Inconel 600 tube (3.2 mm outer diameter  $\times$  0.7 mm thickness) was inserted into FLiNaBe for the HF bubbling, as shown in Fig. 2. The following chemical reaction was applied to produce HF gas using He–3vol% H<sub>2</sub> gas and NiF<sub>2</sub> powder (25 g):



The HF production was conducted by flowing 30 Ncm<sup>3</sup>/min of the He–H<sub>2</sub> gas through a U-shaped tube made of Monel 400 filled with NiF<sub>2</sub> powder, as shown in Fig. 2. The U-shaped tube was heated at 823 K. The HF production rate is evaluated as  $3.9 \times 10^{-5}$  mol/(H<sub>2</sub>%  $\cdot$  min) by an autotitrator. The residual HF exhaust gas from the FLiNaBe container was recovered by water bubbling. If all BeO in FLiNaBe is changed into BeF<sub>2</sub> according to Eq. (1), the water bubbler is considered to recover the residual HF. The pH value of water in the bubbler was measured by a piece of pH indicator paper (ADVANTEC

Co., TEST PAPER pH 0–14) after introducing  $7 \times 10^{-3}$  mol of HF into FLiNaBe. The color of the paper indicated that the pH value of water was 3 after the HF bubbling. This value was sufficiently smaller than pH 6 for water before the HF bubbling. Since residual HF was recovered by the water bubbler, the impurity BeO in FLiNaBe is considered to be changed into  $\text{BeF}_2$ .

### 3. Tritium Production Analysis

The tritium production rate is estimated by the Monte Carlo neutron transport code MCNP6 for the neutron irradiation system shown in Fig. 1 with the ENDF/B-VII.1 nuclear data library [8]. Figure 3 shows the configuration in the simulation. The energy spectrum for neutrons from an AmBe neutron source is set in accordance with the literature [9, 10]. The neutron intensity is  $2 \times 10^6$  n/s of the AmBe neutron source. Also, the particle history number is about  $5 \times 10^7$  in the simulation.

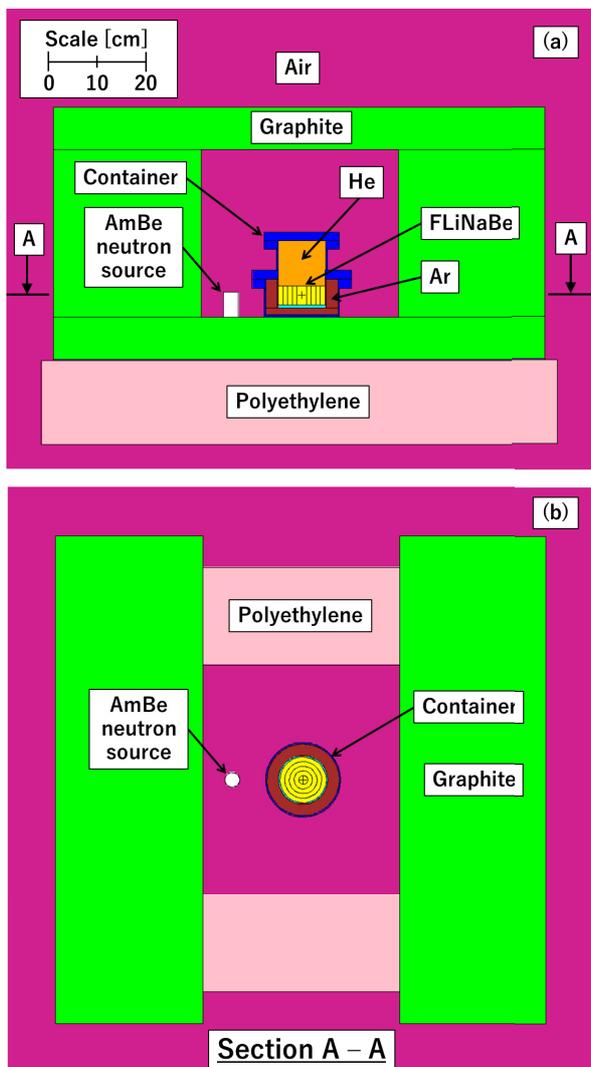


Fig. 3 Configuration in the MCNP calculation: (a) longitudinal sectional view, (b) cross-sectional view (Section A–A).

## 4. Results and Discussion

### 4.1 Tritium production and recovery rates

Figure 4 shows the two-dimensional distribution of the total neutron flux (i.e., the neutron flux in the whole energy range) calculated by MCNP6. In addition, Fig. 5 shows the volume-averaged energy spectrum for the neutron flux in the region of FLiNaBe. The tritium production rate calculated by MCNP6 was  $58.0 \pm 0.1$  Bq/h for FLiNaBe. The transient change in the tritium concentration in FLiNaBe is expressed by Eq. (3) in the Sec. 4.2. From the equation, the tritium production rate is considered to be balanced with the tritium release rate from FLiNaBe after the tritium concentration in the molten salt reached a steady state. The tritium recovery rate in a steady state obtained in the irradiation experiments on FLiNaBe at

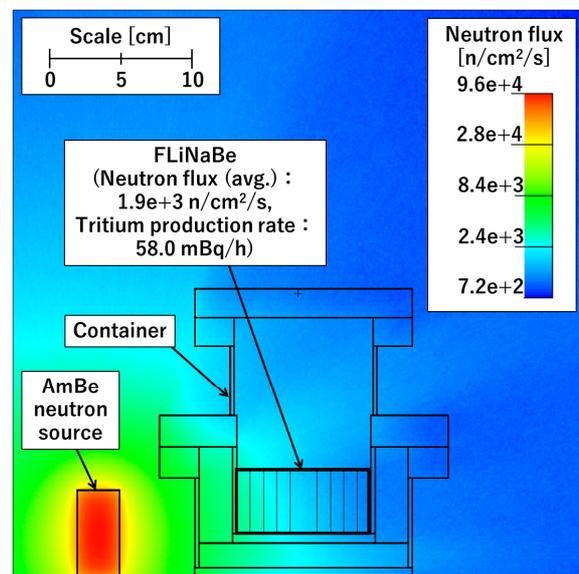


Fig. 4 Two-dimensional distribution of total neutron flux calculated by MCNP6.

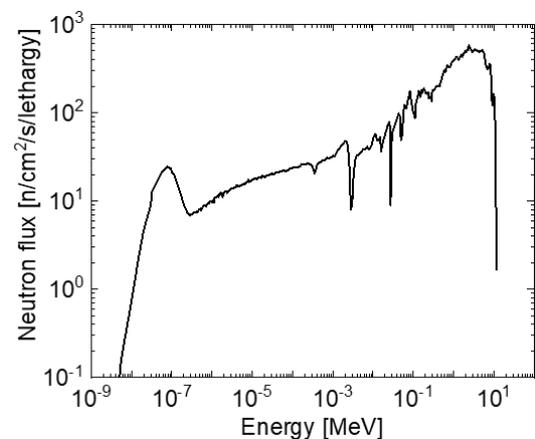


Fig. 5 Volume-averaged energy spectrum for the neutron flux in the region of FLiNaBe.

773 K was  $58.0 \pm 2.1$  mBq/h. The calculated tritium production rate is in good agreement with the tritium recovery rate in a steady state obtained experimentally.

## 4.2 Comparison of the overall mass transfer coefficients for tritium release from FLiNaBe and FLiNaK

The transient change in the tritium concentration in FLiNaBe  $C_F(t)$  in Bq/m<sup>3</sup> is expressed by Eq. (3) and Eq. (4) below [7]:

$$V_F \frac{dC_F(t)}{dt} = S - Q_1(t) - Q_2(t), \quad (3)$$

$$K_i = \frac{Q_i(t)}{A_i[C_F(t) - C_{G_i}(t)]} \approx \frac{Q_i(t)}{A_i C_F(t)} \quad (i = 1, 2), \quad (4)$$

where  $V_F$  is the volume of FLiNaBe ( $3 \times 10^{-4}$  m<sup>3</sup>),  $S$  is the tritium production rate in Bq/h,  $Q_i$  is the tritium release rate in Bq/h,  $K_i$  is the overall mass transfer coefficient in m/h, and  $A_i$  is the surface area contacting FLiNaBe with a gas phase (i.e., He or Ar). The subscript  $i$  refers to the released tritium ( $i = 1$ ) and the permeated tritium ( $i = 2$ ). The subscripts F and G refer to a fluoride molten salt and a gas phase, respectively. The tritium concentration in a gas phase  $C_{G_i}$  is considered to be much smaller than that in a molten salt  $C_F$ . The analytical solution of  $C_F$  is obtained by solving Eq. (3) and Eq. (4). The tritium release rate  $Q_i$  is expressed by the solution of  $C_F$ . Since tritium is recovered by the water bubblers as shown in Fig. 1, the integrated amount of recovered tritium  $m_i$  in Bq is obtained as experimental data. Assuming all the released tritium and the permeated tritium are recovered by the water bubblers, the integrated amount of recovered tritium  $m_i$  is obtained by integrating  $Q_i$  with respect to  $t$  as follows:

$$m_i = \int_0^t Q_i dt = \frac{K_i A_i}{K_1 A_1 + K_2 A_2} \times \left\{ S t - V_F \left( C_F(0) - \frac{S}{K_1 A_1 + K_2 A_2} \right) \times \left[ \exp \left( - \frac{K_1 A_1 + K_2 A_2}{V_F} t \right) - 1 \right] \right\}. \quad (5)$$

The initial concentration of tritium  $C_F(0)$  equals zero because the experiments have been started by unirradiated FLiNaBe. The overall mass transfer coefficient  $K_i$  and the tritium production rate  $S$  are obtained with curve fitting of the experimental data based on Eq. (5).

Figure 6 shows the increase in the integrated amounts of tritium recovered from FLiNaBe at 773 K. The plots are the integrated values of tritium recovered at 48, 118, 284, and 476 h from the start of the neutron irradiation. The error bars for the plots in Fig. 6 were evaluated with the error propagation formula. Those were evaluated by uncertainties of the volume of water in the bubblers and scintillation cocktails, and statistical uncertainties of three times of measurements by the liquid scintillation counter [9]. Figure 6 also shows the tritium release rates for the released

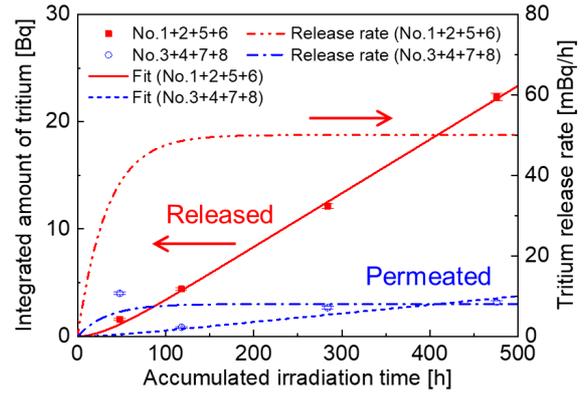


Fig. 6 Integrated amounts of tritium recovered from FLiNaBe at 773 K.

Table 1 Overall mass transfer coefficients for tritium release from FLiNaBe and FLiNaK at 773 K.

	$K_1$ [m/h]	$K_2$ [m/h]
FLiNaBe	$(1.1 \pm 0.4) \times 10^{-3}$	$(4.5 \pm 1.5) \times 10^{-5}$
FLiNaK [6]	$(1.8 \pm 0.8) \times 10^{-3}$	$(6.8 \pm 4.3) \times 10^{-5}$

tritium and the permeated tritium obtained by differentiating Eq. (5). The tritium release rates reach a steady state in about 200 h from the start of neutron irradiation.

The result for the permeated tritium at 48 h is removed from the curve fitting because the result was affected by contaminated tritium adsorbed on the inner surface of the pipes in the previous irradiation experiments on FLiNaK. Some amount of condensed water was found in the bubbler for receiving backwater from the bubblers No. 3 and 4 in Fig. 1, and that condensed water included a large amount of tritium. Tritium adsorbed on metal surfaces could be desorbed by moist gas [11, 12]. Moisture from the thermal insulator wound around the Inconel 600 crucible could be added to the Ar gas flow for the permeated tritium recovery. Such condensed water was not found on the released tritium.

Table 1 shows the overall mass transfer coefficients for FLiNaBe at 773 K obtained with the curve fitting, where  $K_1$  and  $K_2$  are the coefficients for the released tritium and the permeated tritium, respectively. The table also shows the results for FLiNaK at 773 K obtained in the previous study [6]. There was no significant difference between the overall mass transfer coefficients for FLiNaBe and FLiNaK.

While the evaluation of the overall mass transfer coefficients  $K_i$  was conducted once for FLiNaBe at 773 K, reproducibility of the experiment on the evaluation of  $K_i$  was evaluated by FLiNaK at 773 K using the similar ex-

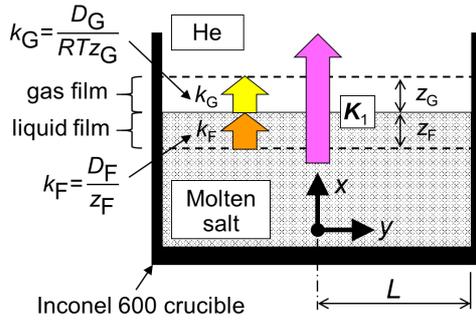


Fig. 7 Conceptual diagram of film theory.

perimental system shown in Fig. 1. The experiments on the evaluation of  $K_i$  for FLiNaK were conducted two times. Each value of  $K_1$  and  $K_2$  was almost the same in the two experiments.

Film theory is applied in order to discuss the result in which the overall mass transfer coefficients for FLiNaBe are approximately the same as those for FLiNaK at 773 K [7]. The two imaginary and stationary boundary films consisting of gas (i.e., He) and liquid (i.e., the molten salts), respectively, are assumed at the interface between gas and liquid, as shown in Fig. 7. The overall mass transfer coefficient  $K$  is expressed by the following equation [7]:

$$K = \frac{k_F k_G}{H k_F + k_G}, \quad (6)$$

where  $k_F$  is the mass transfer coefficient in the liquid film in m/h,  $k_G$  is the mass transfer coefficient in the gas film in mol/(m<sup>2</sup> · h · Pa), and  $H$  is the Henry's law solubility constant for tritium in mol/(m<sup>3</sup> · Pa).

The mass transfer coefficient  $k_F$  is expressed as  $D_F/z_F$ , where  $D_F$  is the diffusion coefficient of tritium in the liquid film, and  $z_F$  is the liquid film thickness, respectively [13]. The film thickness  $z_F$  is considered to be related to the velocity of natural convection in a molten salt. Assuming a steady flow in one direction of  $x$  in Fig. 7, and neglecting pressure difference in a molten salt, equilibrium between the viscous force and the buoyant force is expressed by the following equation:

$$\Delta\rho g = \mu \frac{\partial^2 u}{\partial y^2}, \quad (7)$$

where  $\Delta\rho$ ,  $\mu$ , and  $u$  are the difference of the density in a molten salt, the viscosity of a molten salt, and the velocity in the  $x$ -direction of a molten salt, respectively. The relationship between  $u$ ,  $\Delta\rho$ , and  $\mu$  is obtained as  $u \sim \Delta\rho/\mu$  by solving Eq. (7) under the boundary condition of  $u = 0$  at  $y = \pm L$ , where  $L$  is the inner radius of the Inconel 600 crucible, as shown in Fig. 7.

The liquid film thickness  $z_F$  is considered to be inversely proportional to the square root of the velocity  $u$  in Eq. (7) [13]. From the relationship  $u \sim \Delta\rho/\mu$ , the ratio of the liquid film thickness for FLiNaBe to that for FLiNaK

is below:

$$\frac{z_{F,FLiNaBe}}{z_{F,FLiNaK}} = \sqrt{\frac{u_{FLiNaK}}{u_{FLiNaBe}}} = \sqrt{\frac{\Delta\rho_{FLiNaK} \cdot \mu_{FLiNaBe}}{\Delta\rho_{FLiNaBe} \cdot \mu_{FLiNaK}}}. \quad (8)$$

The buoyant force is estimated by  $\Delta\rho g = (\rho_H - \rho_L)g$ , where  $\rho_H$  and  $\rho_L$  are the densities at the highest and the lowest temperatures of the molten salts in kg/m<sup>3</sup>. The heating temperature of 903 K is used as the highest temperature. The melting point of FLiNaK (727 K) is used as the lowest temperature. The differences of the density  $\Delta\rho_{FLiNaBe}$  and  $\Delta\rho_{FLiNaK}$  are calculated as 79.2 kg/m<sup>3</sup> and 110 kg/m<sup>3</sup>, respectively [4]. Also, the viscosities  $\mu_{FLiNaBe}$  and  $\mu_{FLiNaK}$  at 773 K are  $15.5 \times 10^{-3}$  Pa·s and  $8.15 \times 10^{-3}$  Pa·s, respectively [4]. The ratio of the liquid film thickness for FLiNaBe to that for FLiNaK is calculated as 1.6 from Eq. (8).

From the calculation result of Eq. (8), the ratio of the mass transfer coefficient  $k_F$  for FLiNaBe to that for FLiNaK is evaluated as follows:

$$\begin{aligned} \frac{k_{F,FLiNaBe}}{k_{F,FLiNaK}} &= \frac{D_{F,FLiNaBe} \cdot z_{F,FLiNaK}}{D_{F,FLiNaK} \cdot z_{F,FLiNaBe}} \\ &= \frac{D_{F,FLiNaBe}}{1.6 D_{F,FLiNaK}}. \end{aligned} \quad (9)$$

Since the overall mass transfer coefficients for FLiNaBe and FLiNaK were approximately the same at 773 K (i.e.,  $K_{1,FLiNaBe}/K_{1,FLiNaK} \approx 1$ ), the following relationship is obtained from Eq. (6):

$$H_{FLiNaBe} = H_{FLiNaK} - \frac{\left(1 - \frac{D_{F,FLiNaBe}}{1.6 D_{F,FLiNaK}}\right) k_G}{k_{F,FLiNaBe}}, \quad (10)$$

where the mass transfer coefficient in the gas film is assumed to be the same for FLiNaBe and FLiNaK (i.e.,  $k_{G,FLiNaBe} = k_{G,FLiNaK} = k_G$ ).

The earlier studies have reported that the diffusion coefficients of hydrogen molecules in FLiNaBe and FLiNaK at 773 K were  $2.3 \times 10^{-6}$  m<sup>2</sup>/h and  $4.5 \times 10^{-5}$  m<sup>2</sup>/h, respectively [5, 14]. Assuming the diffusion coefficient of tritium in FLiNaBe is smaller than that in FLiNaK as in the case of hydrogen molecules (i.e.,  $D_{F,FLiNaBe} < D_{F,FLiNaK}$ ), both the mass transfer coefficient  $k_F$  and the Henry's law solubility constant of tritium  $H$  in FLiNaBe are smaller than those in FLiNaK from Eq. (9) and Eq. (10).

### 4.3 Comparison of the overall mass transfer coefficients for tritium release from FLiNaBe at 673 K and 773 K

Figure 8 shows the increase in the integrated amounts of tritium recovered from FLiNaBe at 673 K. The plots are the integrated values of tritium recovered at 72, 171, and 238 h from the start of the neutron irradiation. Table 2 shows the results of the overall mass transfer coefficients for FLiNaBe at 673 K. The table also shows the results for FLiNaBe at 773 K obtained in the previous subsection. The overall mass transfer coefficients for FLiNaBe

at 673 K were approximately 1/5 in comparison with those at 773 K.

The ratio of the film thickness in FLiNaBe at 673 K to that at 773 K is calculated in the same way as Eq. (8), as follows:

$$\frac{z_{F,673K}}{z_{F,773K}} = \sqrt{\frac{\Delta\rho_{773K} \cdot \mu_{673K}}{\Delta\rho_{673K} \cdot \mu_{773K}}} = 1.9, \quad (11)$$

where the heater temperatures of 793 K and 903 K are used as the highest temperature for the calculations of  $\Delta\rho_{673K}$  and  $\Delta\rho_{773K}$ , respectively. The melting point of FLiNaBe (578 K) is used as the lowest temperature. The differences of the density  $\Delta\rho_{673K}$  and  $\Delta\rho_{773K}$  are calculated as 96.8 kg/m<sup>3</sup> and 146 kg/m<sup>3</sup>, respectively [4]. Also, the viscosities  $\mu_{673K}$  and  $\mu_{773K}$  are  $38.6 \times 10^{-3}$  Pa·s and  $15.5 \times 10^{-3}$  Pa·s, respectively [4].

The temperature dependence of the diffusion coefficient of tritium in FLiNaBe  $D_F$  is assumed to be according to the Stokes–Einstein relation,  $D_F = k_B T / (6\pi\mu r)$ , where  $T$  and  $r$  are the absolute temperature of a molten salt and the radius of tritium, respectively [15]. The earlier study has reported that the diffusion activation energy of tritium in molten FLiBe is consistent with the viscosity data [16]. Using the Stokes–Einstein relation, the ratio of the diffusion coefficient of tritium in FLiNaBe at 673 K to that at 773 K is calculated as follows:

$$\frac{D_{F,673K}}{D_{F,773K}} = \frac{673\mu_{773K}}{773\mu_{673K}} = \frac{673 \cdot 15.5 \times 10^{-3}}{773 \cdot 38.6 \times 10^{-3}} = 0.35. \quad (12)$$

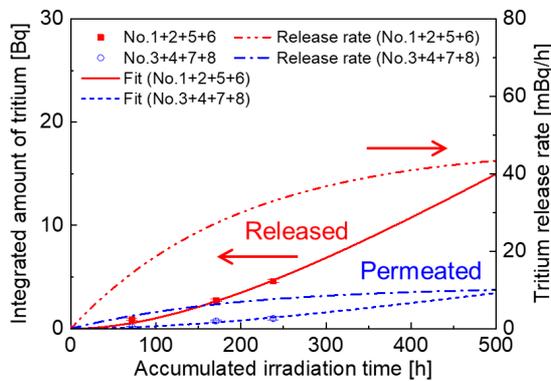


Fig. 8 Integrated amounts of tritium recovered from FLiNaBe at 673 K.

Table 2 Overall mass transfer coefficients from FLiNaBe at 673 K and 773 K.

	$K_1$ [m/h]	$K_2$ [m/h]
673 K	$(1.8 \pm 0.1) \times 10^{-4}$	$(1.0 \pm 0.1) \times 10^{-5}$
773 K	$(1.1 \pm 0.4) \times 10^{-3}$	$(4.5 \pm 1.5) \times 10^{-5}$

Thus, the ratio of the mass transfer coefficient  $k_F$  for FLiNaBe at 673 K to that at 773 K is calculated from Eq. (11) and Eq. (12) as follows:

$$\frac{k_{F,673K}}{k_{F,773K}} = \frac{D_{F,673K} \cdot z_{F,773K}}{D_{F,773K} \cdot z_{F,673K}} = \frac{0.35}{1.9} = 0.18 \approx \frac{1}{5}. \quad (13)$$

The following equation is obtained from Eq. (6) and Eq. (13) by the result for the ratio of the overall mass transfer coefficient  $K_1$  for FLiNaBe at 673 K to that at 773 K (i.e.,  $K_{1,673K}/K_{1,773K} \approx 1/5$ ):

$$\frac{K_{1,673K}}{K_{1,773K}} = \frac{k_{F,673K}}{k_{F,773K}} \cdot \frac{H_{773K} \cdot k_{F,773K} + k_G}{H_{673K} \cdot k_{F,673K} + k_G} \approx \frac{1}{5}, \quad (14)$$

where the mass transfer coefficients in the gas film are assumed to be the same for FLiNaBe at 673 K and 773 K (i.e.,  $k_{G,673K} = k_{G,773K} = k_G$ ). The following relationship is obtained from Eq. (14):

$$\frac{H_{773K} \cdot k_{F,773K} + k_G}{H_{673K} \cdot k_{F,673K} + k_G} \approx 1. \quad (15)$$

The earlier study has reported that the Henry's law solubility constants of hydrogen molecules in FLiNaBe increased with temperature, which suggests that the solution is an endothermic reaction [5]. When the solution of tritium in FLiNaBe is assumed to be an endothermic reaction (i.e.,  $H_{673K} < H_{773K}$ ), the relationships  $k_G \gg H_{673K} \cdot k_{F,673K}$  and  $k_G \gg H_{773K} \cdot k_{F,773K}$  are obtained from Eq. (13) and Eq. (15). Under the assumption, the following equation is obtained from Eq. (6):

$$K_1 \approx \frac{k_F k_G}{k_G} = k_F. \quad (16)$$

Then, the mass transfer coefficients in the liquid film  $k_{F,673K} \approx 2 \times 10^{-4}$  m/h and  $k_{F,773K} \approx 1 \times 10^{-3}$  m/h are obtained for FLiNaBe from the overall mass transfer coefficient  $K_1$  in Table 2.

#### 4.4 Chemical forms of tritium recovered from FLiNaBe

Table 3 shows the experimental results on percentages of soluble (TF and HTO) and insoluble (HT) tritium recovered from FLiNaBe at 773 K in a steady state. The majority of recovered tritium was the released tritium and

Table 3 Percentages of soluble and insoluble tritium recovered in a steady state.

	TF and HTO (Soluble)	HT (Insoluble)
Released tritium	93%	3%
Permeated tritium	3%	1%
Total	96%	4%

soluble. That was similar to the result for the previous irradiation experiments on FLiNaK in which most of the tritium recovered was the released tritium and soluble [6]. A larger amount of insoluble HT was expected to be recovered from FLiNaBe than from FLiNaK because of a smaller amount of free  $F^-$  ions in FLiNaBe forming clusters, e.g.,  $BeF_4^{2-}$  and  $Be_2F_7^{3-}$ , due to the high reducibility of Be [17, 18]. A possible reason for the soluble tritium production in FLiNaBe is due to the formation of HTO by chemical reactions with metal oxide on the crucible wall and/or with impurities in FLiNaBe (e.g.,  $H_2O$ ). Further investigation is necessary to clarify the chemical behaviors of tritium in the molten salts.

## 5. Summary

A series of neutron irradiation and tritium recovery experiments on molten FLiNaBe have been conducted with an AmBe neutron source for examining tritium release behaviors. The overall mass transfer coefficients for tritium release from FLiNaBe at 673 K and 773 K have been evaluated by conducting tritium recovery measurements.

The value of the overall mass transfer coefficient for FLiNaBe at 773 K was almost the same as that for FLiNaK. This result was analyzed using film theory. According to the theory, an overall mass transfer coefficient is obtained from a diffusion coefficient and a Henry's law solubility constant of tritium in a molten salt, and liquid film thickness at a free surface of a molten salt. The ratio of the film thickness in FLiNaBe to that in FLiNaK could be estimated from the difference in the velocity of natural convection in the molten salts. The diffusion coefficient of tritium in FLiNaBe is assumed to be smaller than that in FLiNaK, as in the case of hydrogen molecules reported in the earlier studies. As a result, it was derived that the Henry's law solubility constant of tritium and also the mass transfer coefficient of tritium in the liquid film

for FLiNaBe are smaller compared with those for FLiNaK under the assumption.

The overall mass transfer coefficient for FLiNaBe at 673 K were approximately 1/5 compared with that for FLiNaBe at 773 K. In the analysis using film theory, the ratio of the liquid film thickness at 673 K to that at 773 K was calculated from the Stokes–Einstein relation. From these two ratios and the assumption that the solution of tritium in FLiNaBe is an endothermic reaction, it was derived that the values of the overall mass transfer coefficients are almost equal to those of the mass transfer coefficients in the liquid film of FLiNaBe. In this case, the existence of the gas film could be ignored.

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