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In-reactor experiment and the tritium diffusion coefficient in molten lithium–tin alloy

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Abstract

The measurement of the tritium diffusion coefficient in molten $Li_{20}Sn_{80}$ alloy was carried out using an in-reactor tritium release experiment. The analysis of the measurements is based on a diffusion-controlled steady state model. When the hydrogen partial pressure was higher than 1100 Pa in the purge gas, and the temperature was 873 K or higher, it satisfied the requirements of the model. At 873 K, the tritium diffusion coefficient in the alloy is of order 10^{-9} m^2/s, and the Sieverts' constant in $Li_{20}Sn_{80}$ is below 10^{-5} $Pa^{-0.5}$, based on the rough estimation which is supported by the relation of the tritium residence time to the diffusion coefficient and the Sieverts' constant in the model.

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1. Introduction

Lithium–tin (Li–Sn) alloy has some attractive properties, which make it a potential candidate for liquid tritium breeder or coolant in liquid wall concept fusion reactors [1]. $Li_{20}Sn_{80}$ is used in our research since it has the acceptable tritium breeding ratio [2], and as the coolant, its melting point is lower than the higher Li% Li–Sn alloy, which benefits the heat removal in the fusion reactor.

The tritium diffusivity and solubility in the alloy are important properties to evaluate the tritium distribution in the blanket and to determine the tritium recovery method from the alloy. Sometimes the diffusivity and the solubility of hydrogen are used to estimate the properties of tritium in the alloy. No data about the solubility and the diffusivity of hydrogen isotopes in molten Li–Sn alloy yet have been obtained. An in-reactor tritium release experiment is a good way to investigate these properties.

2. Experiments

2.1. Preparation of $Li_{20}Sn_{80}$ alloy

The $Li_{20}Sn_{80}$ was prepared from 99.999% tin and 99.9% lithium. The impurities in Li, from analysis provided by the supplier, are Na 0.010%, Fe less than 0.001%, K 0.002%, Ca 0.009%, Si 0.003%, N 0.001%, etc. All the operations were carried out in an argon atmosphere (99.9999%) glove box.

2.2. Apparatus

The measurements of the diffusion coefficient were performed through the in-reactor tritium release experiments, using the fast neutron source reactor 'YAYOI' of the University of Tokyo. The system, which consists of a gas supply system, an in-reactor component and a tritium monitoring and recovering system, has been described previously [3].

Alpha Fe is utilized as the material of the sample holder, and any surface oxide is easily reduced by H_2 in the He purge gas. The dimensions of the Fe sample holder are 90 mm depth, 6 mm inner diameter and 7 or 7.5 mm outer diameter. About 80 mm $Li_{20}Sn_{80}$ alloy was placed in a sample holder. During the irradiation,

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tritium is generated in the alloy. Tritium diffuses through the alloy and the wall of the sample holder, then is taken away by He purge gas with variable pressures of H_2. Tritium concentration in the gas phase is monitored with an ionization chamber.

Tritium residence time [3] is a key parameter for quantitative analysis of the diffusion, and it can be obtained from the continuous output signal of the ionization chamber.

3. Tritium migration model

For calculating the tritium diffusion coefficient, a diffusion-controlled steady state model is adopted, which had ever used in $Li_{17}Pb_{83}$ [3] system, because $Li_{20}Sn_{80}$ has the similar tritium generation mechanism under the irradiation and the similar Li content to $Li_{17}Pb_{83}$.

3.1. Diffusion-controlled steady state model

Compared to the tritium release through the side surface wall, the release from the top and the bottom of the sample holder can be neglected because of the configuration. Keeping the top surface of the alloy at the center of the heating section, the natural convection is expected to be small enough. Hydrogen isotopes are combined to HT or T_2 on the surface of the Fe sample holder. The tritium flux J_r (mol/m^2 s) for the recombination-controlled process is

$$J_r = J_r(T_2) + J_r(\mathrm{HT}) = K_r C_{2\text{-T}}(C_{2\text{-T}} + C_{2\text{-H}}), \tag{1}$$

where $C_{2\text{-H}}$ and $C_{2\text{-T}}$ are the hydrogen and tritium concentrations on the surface of Fe, K_r [4] is the recombination coefficient (m^4/s mol). Considering that the gaseous tritium is mainly in the form of HT,

$$J_r = K_r C_{2\text{-T}} C_{2\text{-H}} = \frac{K_r S R T_g}{2\upsilon}\left(\frac{\rho_2 K_{s2}}{W_2}\right)^2 \quad \text{for } r = b, \tag{2}$$

where K_s is Sieverts' constant (Pa$^{-0.5}$), ρ is density (g/m^3), W is molecular weight (g/mol) and subscripts '2' mean Fe. S, T_g and υ are the tritium release rate (mol/s), temperature (K) and the flowrate of the purge gas (m^3/s). R is the gas constant (8.31 J/mol K), and 'b' is the outer radius of the sample holder. On the other hand, the tritium flux J_D (mol/m^2 s) for the diffusion-controlled steady state process is

$$J_D = \frac{S}{2\pi b h} \quad \text{for } r = b, \tag{3}$$

where h is the height (m) of the $Li_{20}Sn_{80}$ alloy. Thus,

$$J_r/J_D = \frac{K_r \pi b h R T_g}{\upsilon}\left(\frac{\rho_2 K_{s2}}{W_2}\right)^2 \quad \text{for } r = b. \tag{4}$$

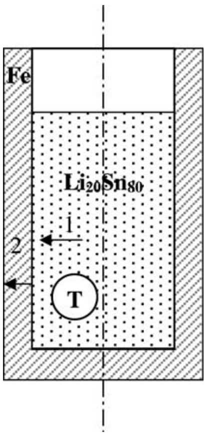

Fig. 1. Tritium migration in the system. (1) Tritium diffusion in the Li–Sn alloy, (2) tritium diffusion in the Fe wall.

In the present experimental conditions, J_r/J_D is in the order of 10^4. Hence the recombination is not the rate-determining process. Then the tritium release is dominated by the tritium diffusion in the radial direction when it reaches the steady state, as shown in Fig. 1.

3.2. Tritium residence time and the tritium diffusion coefficient

With the equations and initial and boundary conditions used in Ref. [3], the tritium residence time τ (s) is obtained:

$$\tau = \frac{a^2}{8D_1} + \left(\frac{W_2 K_{s1} \rho_1}{W_1 K_{s2} \rho_2} - 1\right)\frac{a^2}{2D_2}\ln\frac{b}{a} + \frac{b^2 - a^2}{4D_2}, \tag{5}$$

where D is the diffusion coefficient of tritium, subscripts '1' and '2' mean $Li_{20}Sn_{80}$ and Fe. The characters 'a' and 'b' are the inner and outer radii of the α-Fe sample holder, respectively. The Sieverts' constant for tritium in $Li_{20}Sn_{80}$ is not available.

Clearly, Eq. (5) applies only to the cases where the conditions for the diffusion-controlled steady state model are fulfilled.

4. Results and discussion

4.1. Effect of temperature

At temperatures higher than 873 K, steady state is reached in less than 2 h. Below 873 K, on the other hand, the irradiation time is set to be the maximum value, 5 h though a longer time is needed to reach steady state. At each temperature, the apparent steady state is observed, where the tritium release rate does not change significantly in a short time at the end of the irradiation time.

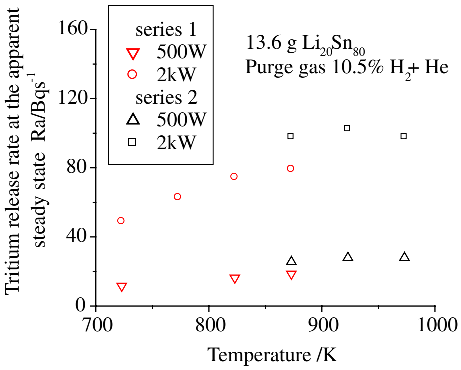

Fig. 2. Dependence of R_a on temperature.

Fig. 2 shows the tritium release rate R_a (Bq/s) in the apparent steady state at different temperatures with the Fe sample holder thickness of 0.75 mm. The data in the range 723–873 and 873–973 K were obtained in two weeks with the system apparatus set at different positions in the irradiation cell. This is the reason why R_a shows the different values at 873 K in the two series.

When the temperature is below 873 K, the R_a values differ markedly, and increase with temperature. When the temperature is higher than 873 K, the R_a is nearly temperature independent. This shows that when temperature is below 873 K, the experiment does not reach the steady state. We also confirmed the steady state at 873 K by comparing the tritium inventory calculated by the assumption that R_a is tritium generation rate with the real tritium inventory released out after the end of the irradiation. The reason of unsteady state at low temperature is not yet clear. But probably, it is not the recombination based on the conclusion deduced in Section 3.1 and the results of Ref. [4], in which the hydrogen recombination coefficient increases with the decrease of the temperature. One possible explanation is that the diffusion coefficient is so small at low temperatures that steady state is not established in the limited irradiation time.

Eq. (5) should only be used to obtain the diffusion coefficient if the experimental temperature is not lower than 873 K.

4.2. Effect of the hydrogen partial pressure at 873 K

The influence of the H_2 partial pressure on the measurement was investigated at 873 K with the Fe sample holder thickness 0.5 mm. The results are shown in Fig. 3. Tritium residence time is similar with H_2 partial pressure varying from 1.1 to 101 kPa in the purge gas. This indicates that the hydrogen pressure is high enough to reduce any possible oxide layer formed in situ by moisture in the gas. This removes the concern that an oxide layer can limit the tritium release on the surface.

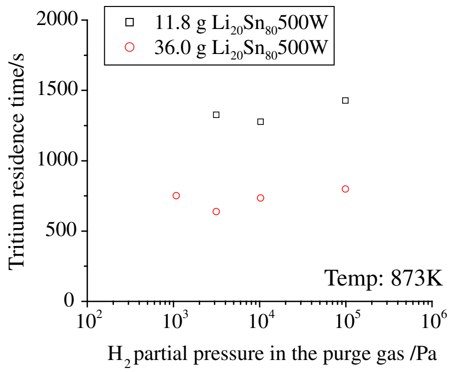

Fig. 3. Dependence of tritium residence time on P_{H2}.

For using Eq. (5) to obtain the diffusion coefficient, the hydrogen partial pressure should not be lower than 1.1 kPa at 873 K.

4.3. Tritium diffusion coefficient and solubility in molten $Li_{20}Sn_{80}$ alloy at 873 K

At 873 K, with pure hydrogen as the purge gas, three series of experiments were carried out at different reactor power levels. Fe sample holder thickness is 0.5 mm.

The Sieverts' constant of tritium or hydrogen in the $Li_{20}Sn_{80}$ alloy is not available. The Sieverts' constant is very high in Li, which is 7.49×10^{-3} Pa$^{-0.5}$ at 873 K [5], while it is 1.76×10^{-7} Pa$^{-0.5}$ at 873 K in Sn [6]. The K_{s1} in $Li_{20}Sn_{80}$ should be between the two values.

We evaluate D_1 and K_{s1} by the relationship between them, which are effective in the diffusion-controlled steady state model, namely Eq. (5). The K_{s1} in the range 10^{-7}–10^{-2} Pa$^{-0.5}$ is examined in the calculation of the tritium diffusion coefficient at 873 K. The K_{s2} and D_2 are taken from Ref. [5]. The results obtained are plotted in Figs. 4 and 5.

With the increase of Sieverts' constant in the range of 10^{-7} to 6×10^{-6} Pa$^{-0.5}$, the diffusion coefficient increases.

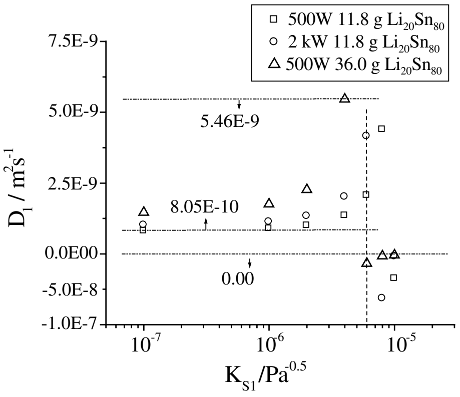

Fig. 4. Dependence of D_1 on K_{s1} in low K_{s1} range.

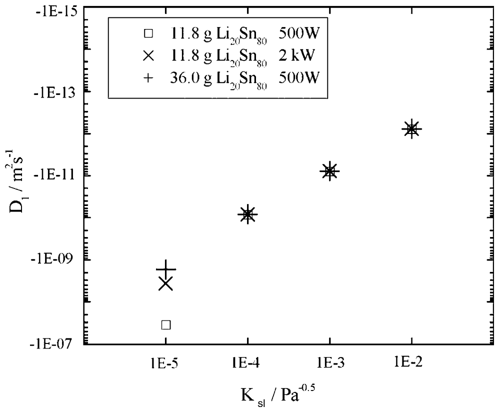

Fig. 5. Dependence of D_l on K_{sl} in high K_{sl} range.

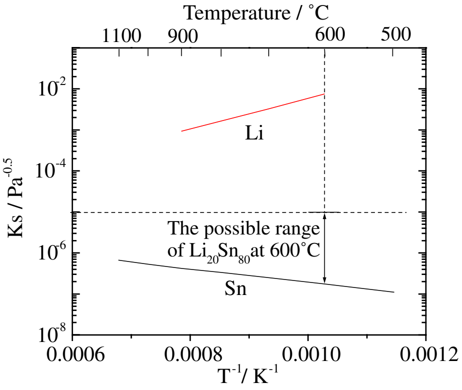

Fig. 6. The possible K_s range for $Li_{20}Sn_{80}$ at 873 K.

But from 6×10^{-6} $Pa^{-0.5}$ K_{sl} the negative diffusion coefficient appears. If K_{sl} is between 10^{-5} and 10^{-2} $Pa^{-0.5}$, all of the D_l in the three experiments are negative, which is unreasonable. Hence, at least the K_{sl} should be lower than 10^{-5} $Pa^{-0.5}$.

The lowest D_l value is 8.05×10^{-10} m^2/s while the K_{sl} is 10^{-7} $Pa^{-0.5}$, which is lower than the K_s of Sn. The highest D_l value is 5.46×10^{-9} m^2/s while the K_{sl} is 4×10^{-6} $Pa^{-0.5}$. Most values are of order 10^{-9} m^2/s. Hence, we think the D_l should be of order 10^{-9} m^2/s.

The comparison of the K_s in Li [5], in Sn [6] and the possible K_s range in $Li_{20}Sn_{80}$ is shown in Fig. 6. The possible K_s in $Li_{20}Sn_{80}$ is closer to Sn, compared with Li.

5. Conclusions

The tritium diffusion coefficient in liquid $Li_{20}Sn_{80}$ was measured, using an in-reactor tritium release experiment. The conclusions that can be drawn are:

1. In order to obtain the tritium diffusion coefficient in the molten $Li_{20}Sn_{80}$ alloy, a diffusion-controlled steady state model is used for analysis.
2. For the present experimental conditions, the recombination on the sample holder surface is not the rate-determining process, which is deduced by the recombination coefficient of Ref. [4].
3. When temperature is 873 K or higher and the hydrogen partial pressure is higher than 1100 Pa, the experimental conditions match the requirements of the diffusion-controlled steady state model.
4. At the proper experimental conditions, the tritium diffusion coefficient measurement was performed. From the relation of tritium residence time with the diffusion coefficient and the Sieverts' constant in the model, we evaluated both. At 873 K, the order of the diffusion coefficient is 10^{-9} m^2/s, and the Sieverts' constant in the $Li_{20}Sn_{80}$ is below 10^{-5} $Pa^{-0.5}$ by this rough estimation.

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