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Analysis of diffusion and dissolution of two-component hydrogen (H + D) in lead lithium

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ABSTRACT

A lead–lithium eutectic alloy (Pb–Li) is one of the most promising candidate materials for the liquid blanket of an advanced fusion reactor. We have experimentally determined mass-transfer properties by an unsteady permeation method, which data are necessary to design a system to recover tritium (T) from a Pb–Li blanket. An experiment of simultaneous H and D permeation through $Li_{17}Pb_{83}$ is performed to clarify interactions between atoms in the two-component permeation process. The experimental results are analyzed by a model of one-dimensional or two-dimensional permeation through $Li_{17}Pb_{83}$. The major permeation proceeds in the longitudinal direction of the present system, and the ratio of hydrogen leak in the radial direction is evaluated using the simulation. As a result, it was found that H and D atoms permeate independently regardless of the H/D component ratio within the present experimental conditions. The permeability and diffusivity of H are 1.4 times higher than that of D. The solubility of H is close to that of D. The isotope effect in diffusivity is in proportion to the square root of the mass ratio of D to H. When these data can be extended to the case of T, T permeability and diffusivity is predicted as 1/1.7 times lower than that of H in the temperature range from 773 K to 973 K.

1. Introduction

Tritium (T) is a radioisotope used as a fuel in fusion reactors. Establishing a reliable T fuel cycle is an extremely important issue. Therefore, T should be recovered as much as possible and the amount of T leaks to the environment should be lowered below an acceptable level. Liquid lead–lithium (Pb–Li) eutectic alloy is one of the most promising candidate materials for the liquid blanket in a fusion reactor. Pb–Li is adopted as a material for the Test Blanket Module (TBM) of ITER [1] such as the He-cooled Pb–Li (HCLL) [2] and the dual coolant Pb–Li (DCLL) [2]. Moreover, a blanket concept of Pb–Li wet wall is adopted in a laser fusion reactor KOYO-Fast [3], which is proposed by ILE Osaka University in Japan.

From the viewpoints of T safety and fuel self-sufficiency for stable operation of a fusion reactor, properties on T transfer are necessary to construct a reliable T recovery system and to estimate rates of T leak through a Pb–Li blanket system. At least two necessary conditions of self-supplemental T production of TBR > 1 and authorized T leak to the outside of 1 g/year (≅10 Ci/day) should be achieved in blankets. Diffusivity and solubility of hydrogen isotopes including T in Pb–Li were determined by some researchers [4–8]. However, there was some scattering among the previous data of diffusivity and solubility.

We have experimentally determined the solubility and diffusivity necessary for the design of a system to recover T from Pb–Li by a permeation method [9]. Our earlier experiment to determine permeability, diffusivity and solubility is focused on a single component of H_2 or D_2 in $Li_{17}Pb_{83}$ [9].

It is important to clarify isotope effects in multi-component permeation process because T will be present along with other hydrogen isotopes of H and D. Isotope effects among H, D and T can be observed by investigating simultaneous permeation behavior when two-component or three-component hydrogen isotopes are present at the same time in a system. However, no datum has been reported on hydrogen isotopes permeation through $Li_{17}Pb_{83}$ in the two- or three-component system. An experiment of the two-component (H + D) permeation through $Li_{17}Pb_{83}$ is performed by means of the transient permeation method, and it is aimed to clarify interactions between H and D atoms in the simultaneous permeation process. Isotope effects between H and D are determined to estimate T data.

2. Experimental

The solubility and diffusivity of hydrogen isotopes in $Li_{17}Pb_{83}$ eutectic alloy are determined using the permeation apparatus as shown schematically in Fig. 1. A $Li_{17}Pb_{83}$ layer is put on an α-Fe

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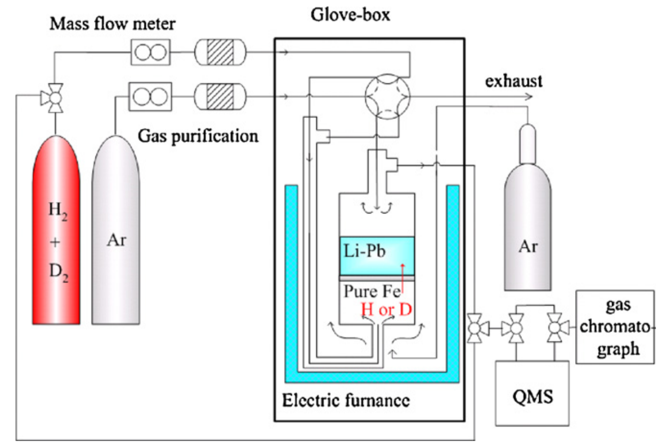

Fig. 1. Experimental apparatus for dual component H + D permeation through $Li_{17}Pb_{83}$.

plate with 1.0 mm in thickness in a SUS304 enclosure. The thickness of the $Li_{17}Pb_{83}$ layer is 1.0 cm. The permeation pot is put in a glovebox of Ar atmosphere in order to keep out of gaseous impurities including N_2, O_2, H_2O and so on. A $Li_{17}Pb_{83}$ eutectic alloy is melted in another Ar glovebox. The eutectic temperature of $Li_{17}Pb_{83}$ is confirmed at its melting point of 508 K. The whole pot is heated by an electric heater and controlled to a constant temperature. The bottom section of the pot is supplied with a gas mixture of $H_2 + D_2$, which is purified with molecular sieve 5A adsorbent. The following composition ratios of H–D mixtures are supplied; H_2:D_2 = 100:0, 90:10, 80:20, 66:34, 44:56, 34:66 and 0:100 in the temperature range from 773 K to 973 K. H_2, HD and D_2 permeated in the upper section are purged by Ar purified with SAES getters ST707 (Zr–V–Fe alloy). The atomic concentrations of H and D in Ar are detected by gas chromatography (GC-14B, SHIMADZU Co.) and a quadrupole mass spectrometer (Qulee BGM-102, ULVAC, Inc.). The flow rates of the upstream and downstream side in the pot are 5 cc(NTP)/min.

3. Model and analysis

3.1. Model of H and D permeation in $Li_{17}Pb_{83}$

H and D permeate from upstream to downstream with the following five fundamental processes.

1. Adsorption and dissociation of H_2, D_2 and HD molecules on the surface of an α-Fe plate.
2. Dissolution and diffusion of H and D atoms in the α-Fe plate.
3. Dissolution of H and D atoms from α-Fe to $Li_{17}Pb_{83}$.
4. Diffusion of H and D atoms in liquid $Li_{17}Pb_{83}$.
5. Recombination of H and D atoms into H_2, D_2 and HD molecules and desorption from the surface of $Li_{17}Pb_{83}$.

Hydrogen or deuterium permeates in the form of atomic H or D in $Li_{17}Pb_{83}$. The Sievert's law applies to an interface between gaseous phase and liquid one [10]. Without $Li_{17}Pb_{83}$ layer, the permeation rate of H or D through α-Fe is 10^3 times faster than that through Pb–Li. Therefore, the permeation resistance of H and D through α-Fe is considered to be negligibly small compared to that through $Li_{17}Pb_{83}$ [11]. The permeation flux of hydrogen isotopes, $J_{k,0}$ (mol/m^2 s), in the one-dimensional direction for the one-component and also two-component syetems is described by the following equation:

$$\frac{J_{k,0}L}{c_{\mathrm{PbLi}}D_{k,\mathrm{PbLi}}K_{k,\mathrm{PbLi}}(\sqrt{p_{k_2,\mathrm{up}}}-\sqrt{p_{k_2,\mathrm{down}}})} = 2\sqrt{\frac{L^2}{\pi D_{k,\mathrm{PbLi}}t}}(e^{-L^2/4D_{k,\mathrm{PbLi}}t}+e^{-9L^2/4D_{k,\mathrm{PbLi}}t}+e^{-25L^2/4D_{k,\mathrm{PbLi}}t}+\ldots) \quad (1)$$

k = H and D.

The H and D buildup curves of the one or two-component system are fitted by Eq. (1). Thus, diffusivity of H or D in Pb–Li is determined from its transient curve, and the permeability of H or D is determined from the steady-state concentration curve. Where J_k (mol/m^2 s) is a permeation flux of the k component (H or D), L (m) is the thickness of $Li_{17}Pb_{83}$, $c_{k,\mathrm{PbLi}}$ (mol/m^3) is a molar concentration of the k component, $D_{k,\mathrm{PbLi}}$ (m^2/s) is its diffusion coefficient in $Li_{17}Pb_{83}$, $K_{k,\mathrm{PbLi}}$ (1/Pa$^{0.5}$) is its solubility constant and p_{k_2} is the partial pressure of the k component. When the system includes only the single component hydrogen, the p_{k_2} value is equal to the total pressure p_t. For the two-component hydrogen isotopes, it is necessary to consider the contribution of HD. The $p_{k_{2,i}}$ value is calculated by taking into account of the isotopic exchange reaction of $H_2 + D_2$ = 2HD on the interface between the liquid Pb–Li and the gaseous phase:

$$p_{\mathrm{H},i} = p_{\mathrm{t}}y_{\mathrm{H},i} \quad (2)$$

$$p_{\mathrm{D},i} = p_{\mathrm{t}}y_{\mathrm{D},i} \quad (3)$$

$$p_{\mathrm{H},i} = p_{\mathrm{H}_2,i} + \frac{p_{\mathrm{HD},i}}{2} \quad (4)$$

$$p_{\mathrm{D},i} = p_{\mathrm{D}_2,i} + \frac{p_{\mathrm{HD},i}}{2} \quad (5)$$

$$p_{\mathrm{t},i} = p_{\mathrm{H}_2,i} + p_{\mathrm{D}_2,i} + p_{\mathrm{HD},i} \quad (6)$$

i = up or down

When the isotopic equilibrium is achieved among H_2, D_2 and HD, it follows:

$$K_{\mathrm{HD},i} = \frac{p_{\mathrm{HD},i}^2}{p_{\mathrm{H}_2,i}p_{\mathrm{D}_2,i}} \quad (7)$$

Eqs. (2)–(7) lead to the following relation between p_{H_2} and y_H

$$p_{\mathrm{H}_2,i} = \frac{y_{\mathrm{H},i}p_{\mathrm{t},i}}{\beta} \quad (8)$$

$$\beta = \frac{-K_{\mathrm{HD},i}/2 + K_{\mathrm{HD},i}/4y_{\mathrm{H},i} + 2 + \sqrt{\left(K_{\mathrm{HD},i}/2 - K_{\mathrm{HD},i}/4y_{\mathrm{H},i} - 2\right)^2 - 4\left(1 - K_{\mathrm{HD},i}/4\right)}}{2} \quad (9)$$

When K_{HD} = 4, the relation of $p_D/p_H = (p_{D_2}/p_{H_2})^{0.5}$ is held.

3.2. Two-dimensional permeation analysis through $Li_{17}Pb_{83}$

Fig. 2 shows the model for the two-dimensional permeation of H and/or D through $Li_{17}Pb_{83}$. The ratio of the hydrogen leak through the sidewall of the pot is estimated using the two-dimensional calculation. The diffusion equations are described as follows:

$$\frac{\partial c_{k,i}}{\partial t} = D_{k,i}\left[\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial c_{k,i}}{\partial r}\right) + \frac{\partial^2 c_{k,i}}{\partial z^2}\right], \quad (10)$$

$$j_{k,0} = -\frac{c_{\mathrm{LiPb}}D_{k,\mathrm{PbLi}}K_{k,\mathrm{PbLi}}}{L}(\sqrt{p_{k_2,\mathrm{up}}} - \sqrt{p_{k_2,\mathrm{down}}}), \quad (11)$$

$$j_k = -D_{k,\mathrm{PbLi}}\left.\frac{\partial c_{k,\mathrm{PbLi}}}{\partial z}\right|_{z=L+\delta\mathrm{Fe}}. \quad (12)$$

Here, the subscript i means Pb–Li, SUS304 or α-Fe. The values of each parameter used in the calculation are summarized in Table 1.