



Hydrogen transport through interface between gas bubbling and liquid breeders

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ABSTRACT

An experimental study was performed to investigate hydrogen transport under conditions of gas bubbling into the liquid breeders of Li, Pb–17Li and Flinak. Gas of H₂ or high-purity Ar at 0.1 MPa was injected into the liquid breeders using an I-shape nozzle. Then, the H₂ partial pressure in the cover gas under transition state was measured by a solid electrolyte sensor. Variations of the partial pressure in the exhaust gas were compared with a numerical model to describe hydrogen transfer through the bubble–liquid interface. The experimental rates of hydrogen transport in the liquid breeders were correlated in terms of a mass-transfer coefficient for the hydrogen transport under gas bubbling. The overall mass-transfer coefficient for H₂ charging was compared with that for Ar purging. The coefficient for the recovery was about 10 times lower than that of the charging in the respective cases of the Pb–17Li and the Flinak.

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1. Introduction

An efficient processing system of tritium recovery from the liquid breeders of Li, Pb–17Li and LiF–BeF₂ (Flibe) is one of the critical issues for self-sustaining D-T fueling of fusion reactors. The tritium recovery systems with a molten salt extraction, a permeation window, a trapping and a liquid gas contactor have been studied [1]. The efficiency of the hydrogen recovery by the permeation window depends on the surface condition. For example, when oxides formed on the surface, the efficiency decreased due to the barrier effect [2]. In the molten salt extraction methods, the dissolution of molten salt in liquid lithium is significant concern [3].

The tritium recovery methods through a bubble–liquid interface have been studied [4–6]. Gas bubbling has beneficial influence on the hydrogen transport through the bubble–liquid interface. The mass transfer coefficient by the gas bubbling was 10–100 times higher than that by a sweep gas, because the bubble surface was clean more than the free surface of the liquids. The bubbling made the convection of the fluids, and the effect of the convection also promoted the hydrogen transport. However, these characteristics in the liquid breeders were limited so far. The method of hydrogen isotopes charging into the liquid breeders is an essential technology

to adjust the hydrogen concentration in the fluids for the investigation on the hydrogen recovery.

In the present study, an experimental study was performed to investigate hydrogen transport under conditions of gas bubbling into the liquid breeders of Li, Pb–17Li and LiF–NaF–KF (Flinak). Gas of H₂ or high-purity Ar at 0.1 MPa was injected into the liquid breeders using an I-shape nozzle. The experimental rates of hydrogen transport in the liquid breeders were correlated in terms of a mass-transfer coefficient for the hydrogen transport under gas bubbling.

2. Experimental condition

2.1. Test materials

An initial impurity of the liquid breeders of Li, Pb–17Li and Flinak used is presented in Table 1.

Flinak has similar characteristics to Flibe, which is considered as a tritium breeder in FFHR [7] with respect to hydrogen transport [8], chemical reaction [9,10] and thermofluid properties. Flinak has a potential not only for a simulant fluid but a candidate of the tritium breeder for the blankets.

The hydrogen solubility is summarized in Table 2 [11–13]. It is known that the solubility in the Pb–17Li has large scattering possibly because the measurement method was not standardized.

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Nomenclature

A	atomic weight [g/mol]
σ	surface tension [mN/m]
C	concentration of hydrogen in liquid breeders [wppm]
C_S	solubility of hydrogen in liquid breeders [wppm]
C_D	drag coefficient of bubble in the liquid breeder
D	diameter of injection tube [m]
d	diameter of bubble [m]
D_H	diffusion coefficient of hydrogen isotopes in liquid breeders [m ² /s]
E	electromotive force of hydrogen sensor [V]
F	Faraday constant, 96,485 [C/mol]
g	gravitational acceleration [m ² /s]
h	overall mass transfer coefficient [m/s]
k	Sievert's constant [$\frac{H}{Li} Pa^{-0.5}$]
k'	Henry's constant [$\frac{H_2}{Flinak} Pa^{-1}$]
ρ_L	density of liquid breeder [g/m ³]
ρ_b	density of gas [g/m ³]
L	bubbling depth [m]
h	overall mass coefficient [m/s]
h_t	overall mass transfer ratio by Eq. (15) [m/s]
P	hydrogen partial pressure in cover gas
$P_{H_2, reference}$	hydrogen partial pressure of reference compartment, 0.1
P_t	total pressure in the experiment, 0.1×10^6 [Pa]
$P_{equilibrium}$	equilibrium pressure [Pa]
Q	flow rate of gases [m ³ /s]
Q_H	flow rate of hydrogen gas [m ³ /s]
Q_{Ar}	flow rate of argon gas [m ³ /s]
R	gas constant, 8.31 [J/mol]
S_b	total surface area of bubbles [m ²]
T	temperature of hydrogen sensor = 873 K [K]
t	time [s]
u	terminal rising velocity of bubbles [m/s]
V	volume of liquid breeders [m ³]
$\Delta_{adsorption}$	difference between the total H ₂ adsorption amount to the Pb–17Li and that to the Flinak [m ³]
$\Delta_{desorption}$	difference between the total H ₂ desorption amount from the Pb–17Li and that from the Flinak [m ³]

Subscript

Pb–17Li	properties of Pb–17Li
Flinak	properties of Flinak

Numbers

Re	Reynolds number
Eo	Etovos number
M	Morton number

2.2. Experimental apparatus

Fig. 1 shows the schematic diagram of the experimental apparatus.

The liquid breeder was filled in the crucible made of SUS316. The inner diameter and the height of the crucible used in the Li experiment were 40 mm and 90 mm, respectively. The inner diameter and the height of the crucible used in the Flinak and Pb–17Li experiments were 36 mm and 160 mm, respectively. The crucible was installed in the vessel 1. The lid of the vessel 1 equipped an I shape nozzle for the gas injection [14]. The nozzle was inserted into the fluids. The immersion depth equaled to a bubbling depth. The

Table 1

Impurity of liquid breeders (unit: wppm).

	Fe	Cr	W	Non metal
Li	0.85	0.12	0.31	Nitrogen: 65
Pb–17Li	2.2	0.17	0.52	–
Flinak	52	2.6	–	H ₂ O: 42.6

inner diameter of the nozzle was 0.6 mm. The gas inlet line and the gas outlet line were preheated. The length of the inlet and outlet lines was 3100 mm and 450 mm, respectively. The inner diameter of the lines was 4.4 mm. The system pressure in the apparatus was fixed at 1 atm (0.1 MPa).

The vessel 2 was installed at downstream of the vessel 1 for the measurement of hydrogen partial pressure in the cover gas with solid electrolyte hydrogen sensor [15,16]. The outer diameter of the sensor was 8 mm.

2.3. Principle of solid electrolyte hydrogen sensor

The on-line measurement of the hydrogen partial pressure in the cover gas was performed with a solid electrolyte type hydrogen sensor made of a perovskite-type oxide. The perovskite-type oxide has proton conduction in hydrogen containing atmosphere at elevated temperature. The oxide used in the present work was CaZr_{0.9}In_{0.1}O_{3-a}. The principle of the hydrogen measurement by the proton conductive solid electrolyte sensor is shown in Fig. 2 [15]. The sensor was composed of two gas compartments. One was filled with the gas for the measurement and the other was filled with the reference gas with known hydrogen partial pressure. Pd electrode was coated on the surface of the solid electrolyte [17]. The difference in hydrogen partial pressure between the two compartments converts into an electromotive force (EMF) in unit of V. The EMF output was evaluated according to the Nernst equation as

$$E = \frac{RT}{2F} \ln \frac{P}{P_{H_2, reference}} \quad (1)$$

All the notations in the equations are listed in Nomenclature. Unknown partial pressure of gas in the measurement compartment is estimated by Eq. (1) with the measured electro motive force in the experiments and the known hydrogen partial pressure of the reference gas. In the present work, $P_{H_2, reference}$ was 0.1. The diameter of the sensor was 8 mm.

2.4. Experimental procedure

The experimental conditions are presented in Table 3. The test temperature was 873 K. The hydrogen partial pressure in the vessel 2 was measured by the sensor at 873 K. The total pressure in the apparatus was 1 atm (0.1 MPa). The experimental procedure was follows: The vessels 1 and 2 were heated up to 873 K under Ar atmosphere. Then, the gas line in the apparatus also pre-heated. After the melting of the Li, Pb–17Li or Flinak, the injection nozzle was inserted into the melts, and H₂ gas (0.1 MPa) was injected into the melts. After the sensor output indicated that the hydrogen partial pressure in the cover gas reached to the total pressure of 1, the injection of hydrogen gas was stopped. Then, Ar gas (99.999%

Table 2

Hydrogen solubility in liquid breeders at 873 K.

	Li [11]	Pb–17Li [12]	Flinak [13]
Sievert's or Henry's coefficient	$\frac{H}{Li} Pa^{-0.5}$ 4.0×10^{-4}	$\frac{H}{Pb17Li} Pa^{-0.5}$ 1.1×10^{-6}	$\frac{H_2}{Flinak} Pa^{-1}$ 9.1×10^{-10}
Hydrogen solubility at 0.1 MPa	$\frac{H}{Li}$ 1.3×10^{-1}	$\frac{H}{Pb17Li}$ 3.6×10^{-4}	$\frac{H_2}{Flinak}$ 9.1×10^{-5}

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