# Selection of Tritium Permeable Membranes of Measuring the Tritium Concentration in Liquid Breeders

B. G. Choi<sup>1\*</sup>, D. W. Lee<sup>2</sup>, E. H. Lee<sup>2</sup>, K. I. Shin<sup>2</sup>, H. G. Jin<sup>2</sup>, H. G. Lee<sup>3</sup> <sup>1</sup>Univercity of Science & Technology, Daejeon, Republic of Korea <sup>2</sup>Korea Atomic Energy Research Institute, Daejeon, Republic of Korea <sup>3</sup>National Fusion Research Institute, Daejeon, Republic of Korea <sup>\*</sup>Corresponding author: ehl@kaeri.re.kr

## 1. Introduction

One of the important issues in the liquid breeder blanket systems is the tritium extraction from the breeder material such as lead lithium eutectic. The measurement of the amount of tritium in liquid breeder is essential technology, but it is difficult to develop the tritium measurement system. As a consequence, a study of a measuring instrument of hydrogen isotopes concentration in the liquid breeder should be given priority [1-5]. The flange type permeation sensor has been proposed because of the easiness of the fabrication and the maximized ratio of the surface per the volume [6-7]. In this study, the suitable materials for permeation membranes were studied and surveyed for the technology of the breeders.

### 2. Basic considerations for permeation sensor

Generally, the concentration of the tritium dissolved in liquid metal could be measured using the process of the hydrogen permeation in the membrane. It is meaningful study to understand the hydrogen behavior in metals. One H<sub>2</sub> molecule was separated into two atoms when the hydrogen molecule (gas-phase) was adsorbed on the metal. And the absorbed H<sub>2</sub> atoms were moved through the thickness in the metal by diffusion. Finally H<sub>2</sub> molecules (gas-phase) came out of the metal via the recombination process of H<sub>2</sub> atoms when H<sub>2</sub> atoms arrived at the other side in the metal.

In this process, the membrane of the hydrogen permeation should need two requirements for the qualification of the sensor in Pb-Li. First, the membrane must be able to quickly follow the rapid changes of the hydrogen concentration in the liquid metal. This means that the membrane material should have a high permeability. In addition, the membranes with a higher diffusivity and a lower solubility are the most consideration factors for the permeation materials [8-9].

Second, the membrane must chemically have the strong corrosion resistance and the durability under the operation with the high temperature such as the eutectic lead lithium.

#### 3. Investigation for the qualified membrane

The membrane materials with high permeability are investigated in the literature. The candidate materials were Nb, Ta, Pd and V which have a higher permeability characteristic. the permeability values (the product of the diffusivity and solubility) were considered and investigated The permeability of materials were measured to obtain the values between 450  $^{\circ}$ C and 550  $^{\circ}$ C in liquid metal. The same data can be checked in another reference [10-11]. The results are showed in Table 1.

Table I. Permeability values for metals between 450  $\,^\circ\!\!\mathbb{C}$  and 550  $\,^\circ\!\!\mathbb{C}$ 

Metal	Permeability (mol/m·s·Pa <sup>1/2</sup> )
Aluminum	$7.47 \times 10^{-14} \sim 8.98 \times 10^{-13} [12]$
Germanium	$4.60 \times 10^{-20} \sim 2.60 \times 10^{-18} [13]$
Iron	$1.23 \times 10^{-10} \sim 2.49 \times 10^{-10} [14]$
Molybdenum	$3.38 \times 10^{-13} \sim 1.73 \times 10^{-12} [15]$
Niobium	$4.05 \times 10^{-7} \sim 2.44 \times 10^{-7}$ [16]
Palladium	$1.62 \times 10^{-8} \sim 2.23 \times 10^{-8} [16]$
Silver	$1.15 \times 10^{-12} \sim 3.97 \times 10^{-12} [18]$
Tantalum	$1.67 \times 10^{-7} \sim 1.11 \times 10^{-7}$ [19]
Tungsten	$1.60 \times 10^{-14} \sim 1.37 \times 10^{-13} [20]$
Vanadium	$2.50 \times 10^{-7} \sim 1.51 \times 10^{-7} [16,17]$

The metals of the compatibility character in Pb-Li were also investigated by the literature in fusion materials. The possible candidates were Fe, Nb, Ta, Mo and Re to be qualified for the corrosion from the condition of liquid metal [21-22]. Particularly, the materials like Nb, Ta were very strong from the point view of the corrosion.

On the basis of the researches, Nb, Ta, and Iron were chosen the most possible permeation membrane. That was why these materials satisfied the requirements of the qualification of the permeation membrane.

#### 4. Conclusion and future works

The tritium extraction from the breeding blanket systems was one of the main technologies in the fusion reactor. The tritium extraction method has been developed in parallel with the development of the sensor for measuring the tritium amount in the liquid metal.

The permeation sensor has developed and the flange type sensor was recently proposed in Korea. To find the appropriate membranes for hydrogen permeation in liquid metal, the compatible materials for the permeation sensor was investigated by the literature data.

Metal	Durable time	Chemical reactions	Remark
Vanadium [21]	907 h at 500 ℃ 1220 h at 600 ℃	Thin gray surface Week corrosion attack	A thin chemical reaction zone was found at high temperature
Iron [21]	1776 h at 500 ℃ 1560 h at 600 ℃	The grain boundaries are attacked at $600^\circ\!C$	Relatively stable up to $600^\circ\!\!\mathbb{C}$
Niobium [22] Tantalum [22]	5280 h at 600 $^\circ C$	Evident corrosion at about $1000 \ ^{\circ}{\mathbb{C}}$	Observed Intergranular reaction after 5280 h Both extremely stable
Molybdenum [21]	5280 h at 600 ℃ 1880 h at 645 ℃	No embrittlement but attack of the surface	Very stable in the Pb-Li Modified surface structure was observed at higher temperature
Rhenium [22]	1600 h at 450 ℃ 2750 h at 500 ℃	Only a boundary value for the solubility in Pb-Li	Tested in form of two Mo-Re alloys
Titanium [21]	476 h at 500 ℃ 634 h at 550 ℃	Only reaction zone Reaction zone ad formed crystals	Because of reactions between the metals, the compound $Ti_2Pb$ is formed at the surface in a reaction zone, destroying the sound metal
Yttrium [21]	60 h at 400 $\degree$ C	Strong reaction with Pb-Li	Not considered as membrane

Table II. Durable and corrosion-resistant metals in Pb-17Li [21-22]

Therefore, the membranes to have the comparatively high permeability and the strong corrosion resistance against liquid metal were selected in this study.

With a flange type sensor applied to the selected membranes, the performance test for the hydrogen permeation will be evaluated under the operational environment of liquid breeder.

#### REFERENCES

[1] D.W. Lee, et al., "Current status and R&D plan on ITER TBMs of Korea" Journal of Korean Physical Society 49 (2006) S340-S344.

[2] D.W. Lee, et al., "Preliminary design of a helium cooled molten lithium test blanket module for the ITER test in Korea," Fusion Eng. Des. 82 (2007) 381-388.

[3] D.W. Lee, et al., "Helium cooled molten lithium TBM for the ITER in Korea," Fusion Sci. Tech. 52 (2007) 844-848

[4] D.W. Lee, et al., "Design and preliminary safety analysis of a helium cooled molten lithium test blanket module for the ITER in Korea," Fusion Eng. Des., Vol. 83 (2008) 1217-1221.
[5] D.W. Lee, et al., "Progress and Strategy of ITER Liquid-type TBM Development," Proc. of the KNS Spring Meeting, Gwangju, Korea, May 2013

[6] E.H. Lee, et al., Development of a permeation sensor made of vacuum flanges to measure hydrogen isotopes in liquid metal breeder, Tran. of Korean Nuclear Society Autumn Meeting, Gyeongju, Korea, 2013.

[7] B.G. Choi, et al., performance tests of a permeation sensor for test blanket modules using liquid metal, Tran. of Korean Nuclear Society Spring Meeting, Gwangju, Korea, 2013

[8] A. Aiello, et al., "TRIEX hydrogen sensor qualification" ENEA division FPN FIS ING (2007), 1-23.

[9] A. Ciampichetti, et al., "Performance of a hydrogen in Pb-17Li," Journal of Nuclear Materials (2007) 367-370

[10] S.A. Steward, Review of hydrogen isotope permeability through the materials. (1983)

[11] N.P. Kherani, W.T. Shmayda, "Tritium-Materials Interactions", Nuclear Science & Technology, Safety in Tritium Handling Technology, Euro Course Series (1993) 85-105. [12] H. Ihle, et al., TN, 1975, National Technical Information Service, U.S. Dept. of Commerce, Springfield, VA, CONF-750989 (1976), p. IV-414.

[13] A. Van Wieringen, N. Warmoltz, Physica (*Utrecht*) 22, 849 (1956).

[14] O. D. Gonzalez, Trans. Metall. Soc. AIME 239, 929 (1967).

[15] W. T. Chandler and R. J. Walter, in *Refractory Metal Alloys*, I. Machlin, R. T. Begley, and E. D. Weisert, Eds. (Plenum, New York, 1968), 197-249.

[16] E. Veleckis, R. K. Edwards, J, Phys. Chem. 73, 683 (1969).

[17] J. Volkl and G. Alefeld, in Diffusion in Solids: Recent Developments, A. S. Nowick and J. J. Burton, Eds.

(Academic Press, New York, 1975), 232-302.

[18] Stephen A. Koffler, John B. Hudson, and George S. Ansell, Trans. Metall. Soc. *AIME* 245, 1735 (1969).

[19] Y. I. Zvedin, Y. I. Belyakov, *Sou. Mater.* Sci. (Eng. Trans.) 3, 255 (1967).

[20] E. A. Aitken, et al., Trans. Metall. SOC. AIME 239

[21] H. Feuerstein, et al., "Compatibility of 31 metals, alloy and coatings with static Pb-17Li eutectic mixture" Report FZKA 5596 (1995)

[22] H. Feuerstein, et al., Compatiblilty of refractory metals and beryllium with molten Pb-17Li, "Journal of Nuclear Materials" (1996)