§73. Study on Dissolution and Diffusion Behavior of Hydrogen Isotopes in Oxide Ceramics Using Tritium

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Oxide materials are used in fusion reactors as plasma diagnosis windows, electric insulators and tritium permeation barriers. Tritium dissolution and permeation in the oxide materials are a significant issue from a stand point of tritium retention and leakage. However, compared with alloys, hydrogen solubility and diffusivity in oxides have not been studied well, because very low hydrogen solubility in oxides made difficult to detect hydrogen in them.

In this study, we have tried to examine the hydrogen solubility in three types of oxides (alpha-Al_2O_3, $MgAl_2O_4$ spinel and Y_2O_3 stabilized ZrO_2) using a rather high concentration tritium gas and an imaging plate (IP). The single crystals of these oxides were used in order to avoid the influence of grain boundary on hydrogen dissolution.

Mirror polished single crystals ($10x10x0.5mm^3$) of cubic zirconia (13%Y_2O_3-ZrO_2(100)), alumina (alpha-Al_2O_3(0001)), and spinel ($MgAl_2O_4$(111)) from Dalian Keri Optoelectronic technology Co., Ltd. were used as specimens. They were heated at 873K for 1h in a vacuum to remove an initially dissolved hydrogen, and then exposed to 133Pa of tritium-deuterium gas mixture (T/(T+D)~ 0.17) at temperatures ranging from 673 to 873 K for 5h. No apparent pressure change was observed during the exposure. After that, the specimens were quenched down to fix tritium distribution. The specimens were cut in halves with a diamond saw, and the cross sections of the specimens were also exposed to IP (TR2025, GE Health Care Co.) for 15 h, and photo-stimulated luminescence (PSL) intensities were obtained from IP reader (FLS7000, Fujifilm). The PLS values were converted to bulk tritium activities by calibration with a tritium standard sample (ART 123A, American Radiolabeled Chemicals, Inc.).

The present data on hydrogen solubility are compared with literature data [1-6] in Fig. 1. The all literature data have been converted to those at a pressure of 133 Pa of hydrogen containing gases (H_2 or H_2O), on an assumption of solubilities $s \propto p_{H_2}^{1/2}$ and $s \propto p_{H_2O}^{1/2}$. As shown in Fig. 1(a), hydrogen solubilities in zirconia were reported by Wagner [1], Park and Olander [2] and Yamanaka et al [3]. Since the crystal structures of zirconia and hydrogen containing gas of each study are different, the values of solubilities and its temperature dependence are rather scattered. Figure 1(b) shows the hydrogen solubilities for alpha-alumina [4,5], the data of which are also scattered but similar temperature dependence appears. Hydrogen solubility in spinel has been reported to be quite low, so that many experiments were previously conducted at high temperatures and/or high pressures of hydrogen containing gas. Nevertheless, the solubility data reported are very scarce. Figure 1(c) shows the data of high pressure experiment in water [6] and the present study, which much differ from each other. It is reported that hydrogen solubilities in several types of spinels depend on the ratio of Mg and Al and content of impurity elements but the reason that the hydrogen solubility is very high in this study is not clear at present.

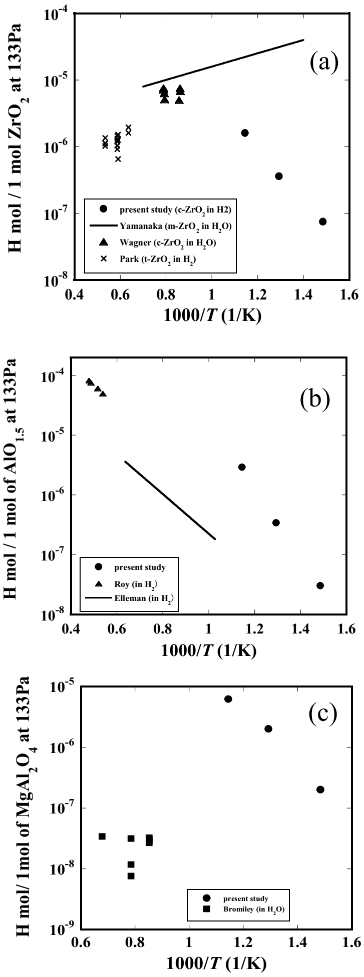

Fig. 1 Comparison of hydrogen solubilities in (a) ZrO_2, (b) Al_2O_3 and (c) $MgAl_2O_4$.

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