RADIATION INDUCED PHENOMENA ON ELECTRICAL AND PROTONIC PROPERTIES OF COMPACT METAL-ELECTROLYTE FUEL CELL

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Introduction

It has been already proposed that proton conductive ceramics are applied as an electrolyte for compact fuel cells combined with metal-hydride and are expected for use under radiation environments such as fission and fusion reactors or cosmic [1].

In the present study, to investigate radiation induced phenomena of proton conduction in radiation environments, the electrical conductivity of typical proton conducting perovskite-type oxide ceramics ($SrCe_{0.95}Yb_{0.05}O_{3-\delta}$) with zirconium metal containing hydrogen was measured in-situ at irradiation temperatures below 473 K (first cycle) and 673 K (second cycle) under fission reactor irradiation.

Results and discussion

The specimen used in the present experiments was $SrCe_{0.95}Yb_{0.05}O_{3-\delta}$ which had orthorhombic perovskite-type and exhibits high protonic conductivity under a hydrogen-containing atmosphere at temperatures above 573 K [2]. Dimension of the specimen was ϕ8 mm diameter and 1 mm thick. After etching on the surface of specimen by Ar^+ ion sputtering in a high vacuum, 1.0 μm thick zirconium metal (Zr) was deposited on one side of the specimen using an electron beam-sputtering device, and hydrogen was implanted into the Zr film with 10 keV H_2^+ ions at 473 K. The fluence was about 5.0×10^{21} H/m^2. In order to make hydrogen atoms transported by chemical reaction easily, zirconium oxide (ZrO_2) was deposited on another side of the specimen. The specimens with and without H were accommodated in a fixture (sub capsule) made of copper and alumina, used as a guard ring geometry, and were installed in a specially designed irradiation rig. The irradiation rig was filled with helium gas at a pressure of 1.0×10^5 Pa and inserted into a fission reactor core of Japan Materials Testing Reactor (JMTR) in oarai research establishment of Japan Atomic Energy Research Institute (JAERI) [3, 4].

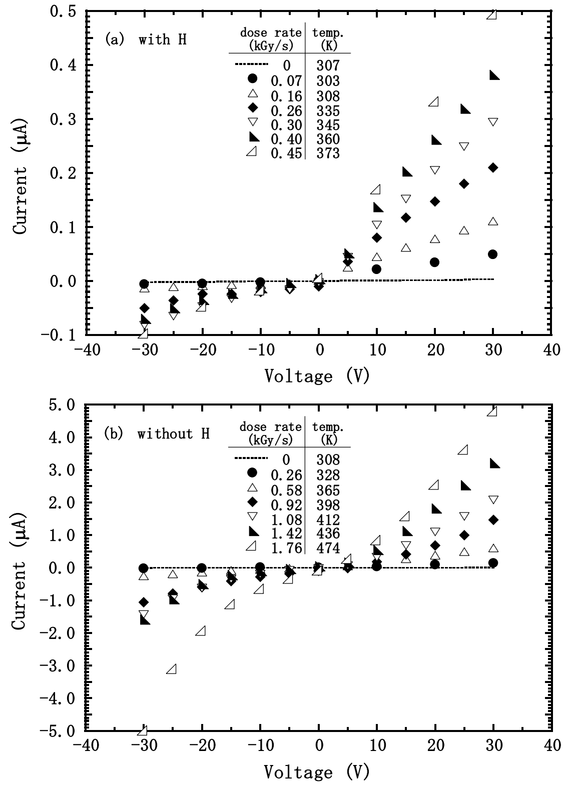

Fig. 1 Relation between the measured current and the applied voltage for $SrCe_{0.95}Yb_{0.05}O_{3-\delta}$ specimens (a) with and (b) without H during reactor irradiation.

Figs. 1(a) and (b) show relations between the currents measured and the voltages applied for the specimens with and without H, respectively, at several ionizing dose rates. The currents at the applied voltage increased as the ionizing dose rate increased. Moreover, the irradiation temperature also increased by gamma-heating. The increment of the irradiation temperature has little influence on the current.

Figs. 2(a) and (b) show radiation induced conductivity (RIC) for the specimens with and without H as a function of ionizing dose rate at first and second cycles, which were calculated using Ohm's law from the experimental data from 0 to +10 V. It was found that the RIC for the specimen with H at 0.5 kGy/s was higher by about two orders of magnitude than the base conductivity, and higher than that without H. The RIC in the

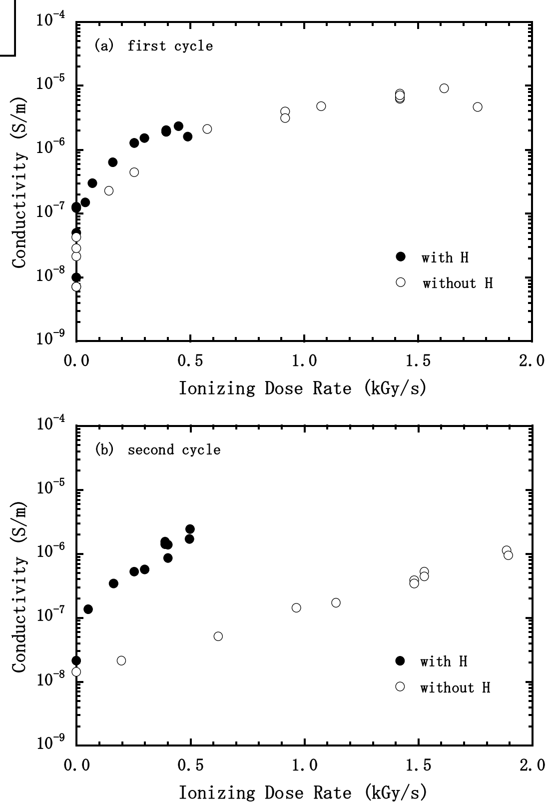

Fig. 2 Conductivities for $SrCe_{0.95}Yb_{0.05}O_{3-\delta}$ specimens with and without H as a function of ionizing dose rate at (a) first and (b) second cycles.

present study is attributed to the electronic excitation as well as enhanced diffusion of hydrogen due to ionizing irradiation. After irradiation for 27 reactor full power days, the RIC with H at first cycle was almost same as that at second cycle, while the RIC without H at several ionizing dose rate reduced lower by about one order of magnitude. According to the references [1, 5, 6], the protonic conduction has no influence on the irradiation. Moreover, the electronic conduction is modified by sub-band annihilation between valence and conduction bands after neutron and electron irradiations.

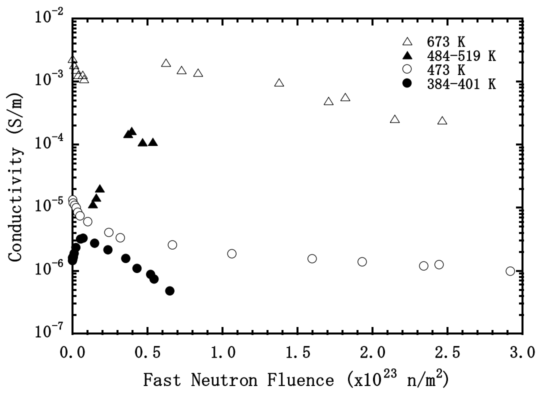

Fig. 3 Fast neutron fluence dependence of conductivities for $SrCe_{0.95}Yb_{0.05}O_{3-\delta}$ specimens with H at 384-401 and 484-519 K and without H at 473 and 673 K, respectively.

Fig. 3 shows fast neutron fluence dependence of conductivities for the specimens with and without H at ionizing dose rates of 0.5 and 1.8 Gy/s, respectively. The irradiation temperatures for the specimens with and without H were 384-401 and 473 K at first cycle and 484-519 and 673 K at second cycle. The RIC depends on irradiation temperature. Particularly, the RIC with H greatly corresponds to the change of the irradiation temperature. The decrement of the RIC with H may show to trapping of H at some point defects. The RIC without H decreased gradually as the fast neutron fluence increased. The decrement of the RIC shows the annihilation of some sub-bands, caused by neutron cascade collisions [5].

Conclusions

The RIC of $SrCe_{0.95}Yb_{0.05}O_{3-\delta}$ specimens with and without H was observed under fission reactor irradiation. The RIC for the both of them increased with increasing ionizing dose rate, and those with H at the maximum ionizing dose rate of 0.5 kGy/s, by 50 MW reactor power, and without H at 1.8 kGy/s were higher by about two and three, respectively, orders of magnitude than the base conductivity without radiation at 0 MW. The RIC with H in the dose rate range of 0.5 kGy/s was higher than that without H. The result may show that the RIC is caused by electronic excitation as well as enhanced diffusion of hydrogen due to gamma irradiation. After irradiation for 27 reactor full power days, the RIC for second cycle was measured again. The RIC with H was almost same as the result for the first cycle, while that without H reduced lower by about one order of magnitude. It may indicate that the protonic conduction has no influence on the irradiation, while the electronic conduction is modified by radiation induced defects.

References

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