§6. Radiation Induced Phenomena of Hydrogen-doped Perovskite-type Oxide Ceramics

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So far, it has been reported that the electrical properties of the insulating materials such as oxide ceramics are dynamically changed by several radiation induced phenomena.[1, 2] It will be predicted that the radiation induced phenomena are further enhanced by behavior of hydrogen isotopes trapped in the insulating materials during long term D-T discharge. Our groups have carried out the electrical conductivity in-situ measurements under reactor irradiation using Japan Materials Testing Reactor at Japan Atomic Energy Agency (JAEA), in order to investigate the hydrogen effects on the radiation phenomena for the typical proton conducting perovskite-type oxide ceramics ($BaCe_{0.9}Y_{0.1}O_{3-\delta}$).[2] As the results, the increment of the electrical conductivity, called radiation induced conductivity (RIC), was observed as the reactor full nuclear power increased up to 50 MW. The maximum ionizing dose rates were 1.1 and 2.0 kGy/s for hydrogen (H)-doped and undoped $BaCe_{0.9}Y_{0.1}O_{3-\delta}$. The irradiation temperatures for the H-doped and undoped specimens were 426-448 and 473 K during the first cycle at the full power for 29 days and 421-572 and 673 K during the second cycle for 27 days. For 1.1 kGy/s, the RIC of H-doped $BaCe_{0.9}Y_{0.1}O_{3-\delta}$ was about two orders of magnitude higher than that of H-undoped one. The result is attributed to hydrogen enhanced diffusion. Moreover, the RIC for H-doped $BaCe_{0.9}Y_{0.1}O_{3-\delta}$ in first cycle, which initially decreased quickly for a brief period, and hereafter reached a constant level which was kept in second cycle. However, the correlation between the hydrogen behavior and the radiation phenomena is not yet fully understood, since there are some effects such as temperature, neutron elastic cascade collision and electronic excitation by gamma rays in the reactor. It is necessary to separate their effects and make out some experiments, in order to understand the results on the reactor radiation.

To investigate the role of hydrogen in the perovskite-type insulating materials under irradiation, the conductivities of the H-doped and undoped $BaCe_{0.9}Y_{0.1}O_{3-\delta}$ were in-situ measured during electron irradiation with 10-1000 Gy/s at 473 K. It can be seen in Fig. 1 that the conductivity in the final dose for irradiation time of 60 min increased as the dose rate increased up to 300 Gy/s, and those at 100 and 300 Gy/s became about two and three, respectively, orders of magnitude higher than the base conductivity without radiation, where it is called RIC. It was found that the RIC for the H-doped specimen was higher by one order of magnitude than that for the H-undoped one. The results may show that the RIC takes place due to hydrogen diffusion as well as electronic excitation, enhanced by ionizing effects. From 300 to 1000 Gy/s, the RIC in the final dose hardly depended on the dose rate and the hydrogen concentration. The base conductivity due to protonic conduction after irradiation was reduced to about one order of magnitude lower before one. Namely RIED was observed. The formations of the Ba and Ce hydroxides were observed in XPS and optical absorption spectra. The dose rate dependence of the RIC above 300 Gy/s may probably show that the ionic conductions by constituent oxygen elements in $BaCe_{0.9}Y_{0.1}O_{3-\delta}$, were caused by the ionizing effects, and the behavior of the excited hydrogen atoms was suppressed by them.

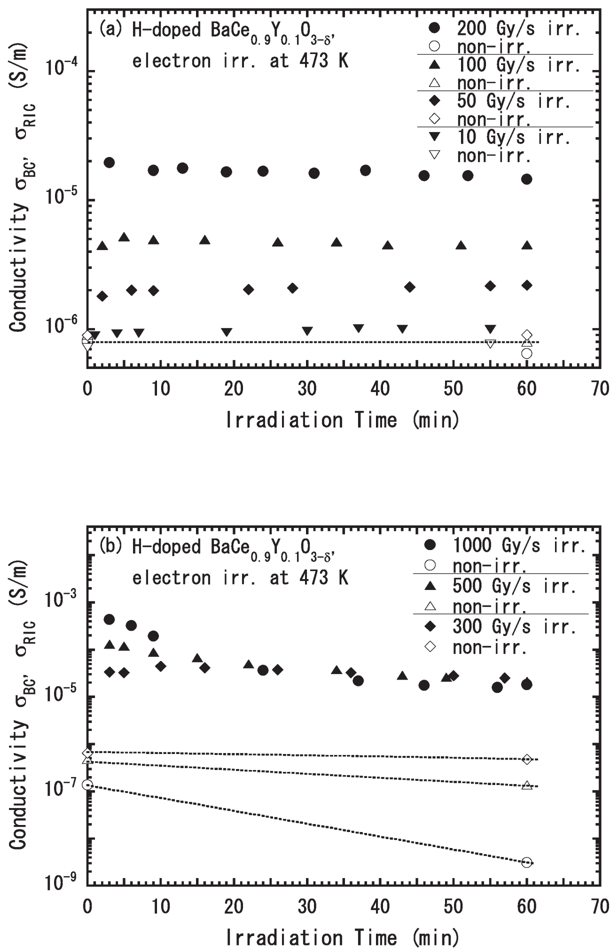

Fig. 1. Radiation induced conductivity, σ_{RIC}, of H-doped $BaCe_{0.9}Y_{0.1}O_{3-\delta}$ as a function of electron irradiation time at temperature of 473 K and ionizing dose rates of (a) 10-200 and (b) 300-1000 Gy/s, as compared with base conductivity, σ_{BC}, before and after irradiation.

1) Hodgson, E. R., Nucl. Instr. and Meth. in Phys. Res. B 191 (2002) 744.
2) Tsuchiya, B., Shikama, T., Nagata, S., Toh, K., Narui, M. and Yamazaki, M., J. Nucl. Mater. 367-370 (2007) 1073.