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Electrical conductivities of dense and porous alumina under reactor irradiation

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Abstract

Radiation induced changes of electrical conductivity for alumina varying density were investigated during fission reactor irradiation. Radiation induced conductivity (RIC) for dense and porous alumina increased due to electronic excitation with increasing the reactor power. The RIC at the reactor full power of 50 MW with ionization dose rate of 2.3 kGy/s and fast neutron flux of $1.4–1.6\times10^{17}$ n/m^2 s was higher by about two orders of magnitude than base conductivities at 0 MW. The RIC for porous alumina was about 3.3 times as much as that for dense alumina. The RIC of dense and porous alumina at 50 MW decreased at the initial fluence and reached a constant at fast neutron fluences of about 5.5×10^{22} and 2.5×10^{22} n/m^2, respectively and subsequently kept up to the resultant ionization dose of 4.4×10^3 MGy and the fast neutron fluence of about 2.9×10^{23} n/m^2.

1. Introduction

Ceramics will be used as electrical insulator materials for burning-plasma diagnostics components in nuclear fusion reactors. However, during the plasma operation, radiation effects have a significant impact on the electrical signals obtained using mineral insulated (MI) cables. It is necessary to compensate the modified electrical signals in order to estimate the conditions of the plasma in the reactor. Therefore, it is essential to understand the radiation induced change in the electrical conductivity of ceramics under irradiation.

So far, it has been reported that the electrical property of some ceramics such as single- and poly-crystalline oxides is dynamically modified by radiation induced phenomena such as radiation induced conductivity (RIC) and radiation induced electromotive force (RIEMF) due to electronic excitation [1–9]. For high neutron fluence, their radiation phenomena are greatly changed by atomic displacements, caused by neutron cascade collisions.

In the present study, dense and porous aluminum oxide (alumina) specimens which had different densities were fabricated. The porous specimens may provide insight on the expected behavior due to aluminum and oxygen interstitial atoms or vacancies produced due to atomic displacements during high fluence reactor irradiation. Also, the RIC and RIEMF of dense and porous alumina were in situ studied using Japan Materials Testing Reactor (JMTR) in Oarai research establishment of Japan Atomic Energy Research Institute (JAERI).

2. Experiments

Dense and porous polycrystalline alumina specimens, have a gradient of density at one side and density of 3.8×10^3 kg/m^3 and porosity of 97% at another side, were prepared by NGK INSURATORS, LTD. The porosity and density of dense alumina were 99% and 3.9×10^3 kg/m^3. The porosity and density of porous

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alumina were 60% and 3.6×10^3 kg/m^3. The dimension for both specimens was ϕ 8 mm diameter and 1 mm thickness. Two platinum electrodes of ϕ 3 mm diameter and 1 mm thickness were contacted with both sides of the specimen by silver conductive paints. A guard ring geometry was not used in this experiment. They were accommodated in a fixture (sub capsule) made of copper and alumina 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 was inserted into a fission reactor core of JMTR in Oarai research establishment of JAERI.

Electrical conductivity measurements were carried out by measuring DC-currents associated with applying DC-voltages in the range from +100 to −100 V under the reactor irradiation. The DC electric field was disconnected during irradiation, except for the short times needed for the measurements. Magnesia insulating electrical triaxial cables with 1.6 mm in diameter were used to carry the electrical signals. The inner conductor and outer sheath materials were made from nickel and stainless steel, respectively. The length of the cable was about 10 m [10]. The reactor power was raised sequentially with steps at 2, 5, 16, 25, 30, 40, 45 and 50 MW for 21.9 h. When the reactor power reached 50 MW, the irradiation temperature increased up to 620 K, measured using a thermocouple, due to gamma-ray heating (ionization dose rate) of 2.3 kGy/s. The 50 MW fast ($E > 1.0$ MeV) and thermal ($E < 0.683$ eV) neutron fluxes were $1.4–1.6\times10^{17}$ and $1.1–1.3\times10^{18}$ n/m^2 s, respectively. The specimens were irradiated for 22 reactor full power days. The resultant ionization dose was 4.4×10^3 MGy and the fast and thermal neutron fluences were about 2.9×10^{23} and 2.3×10^{24} n/m^2.

3. Experimental results and discussion

Relations between the currents measured and the voltages applied at reactor powers of 16, 25, 30 and 40 MW are plotted in Fig. 1(a) and (b), respectively, where (a) and (b) represent experimental data for dense and porous alumina, respectively. Fig. 2(a) and (b) show the similar voltage–current (*V–I*) relations for dense and porous alumina at different full power irradiation times. For the reactor powers below 40 MW in Fig. 1(a) and (b), the current is sigmoidal to the applied voltage. The result seems to show the typical well known gas conductivity [3]. Therefore, the behavior for high enough voltages can be explained as all the electron/ion pairs, produced by ionizing radiation, reach the corresponding electrodes. For the irradiation at 50 MW and about 620 K in Fig. 2(a) and (b), the current is mostly proportional to the voltage within ±100 V. As the irradiation time increased, the currents for dense and porous alumina reduced at several voltages and thereafter did not

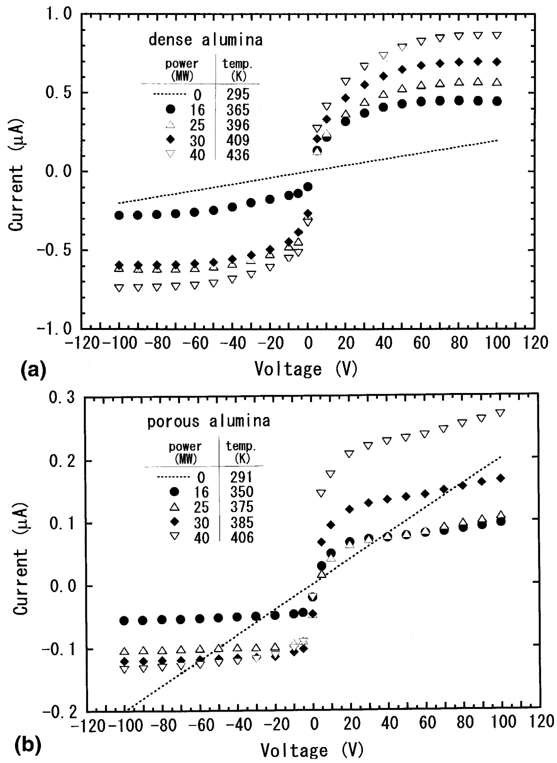

Fig. 1. Relation between the current and the voltage for (a) dense and (b) porous alumina, respectively, at the reactor powers of 16, 25, 30 and 40 MW.

change. Those *V–I* relations are not symmetric for positive and negative voltages, because a slight increase of the current without the applied voltage, RIEMF, namely offset electrical current at zero applied voltage, is occurred during irradiation. Fig. 3 shows RIEMF current at full power of 50 MW at about 620 K. The RIEMF seems to have a constant value, except for the irradiation time above 500 h. The RIEMF observed at full power of 50 MW may be strongly influenced by cabling, since it has been demonstrated by Vila and Hodgson [7]. However, the RIEMF of dense alumina was higher than that of porous one. The result may contribute to density of conducting electrons or different potential due to gradient of atomic density. The differences between the *V–I* relations and RIEMF for dense and porous alumina cannot be clearly explained in the present experiment.

Fig. 4 shows RIC of dense and porous alumina as a function of the reactor power from 0 to 40 MW. The RIC was calculated using Ohm's law from the volume of the specimen at electrode part and the experimental data of 0 to +5 V in Figs. 1 and 2. The RIC increased with increasing the reactor power. The solid and dotted curves in Fig. 4 represent the theoretical values of the

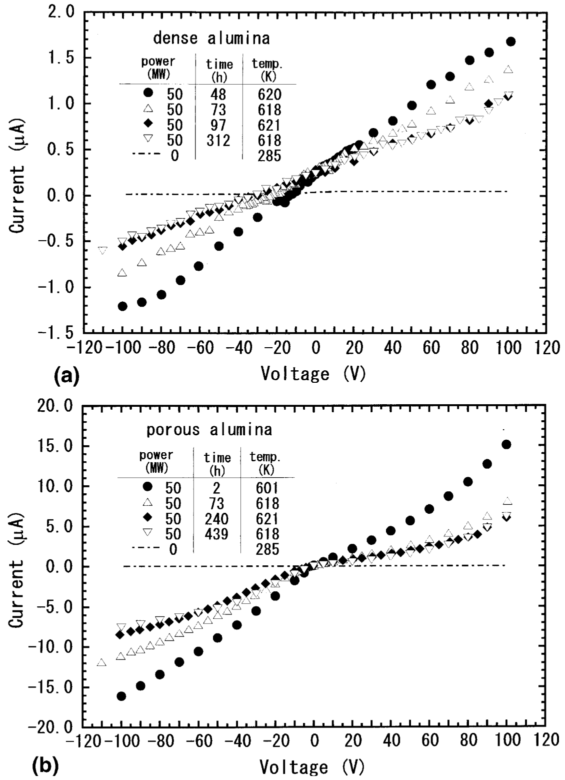

Fig. 2. Relation between the current and the voltage for (a) dense and (b) porous alumina, respectively, with increasing irradiation time at the reactor power of 50 MW.

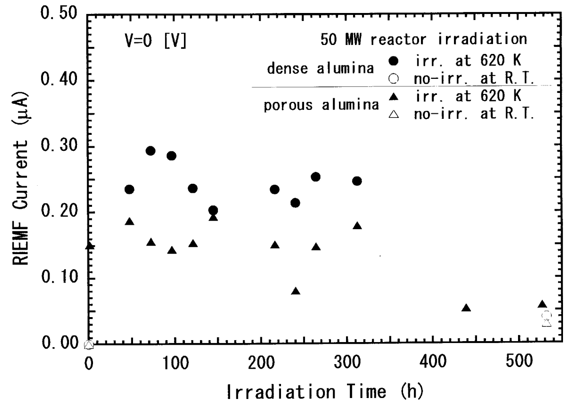

Fig. 3. RIEMF, offset electrical current at zero applied voltage, for dense and porous alumina at the reactor power of 50 MW.

RIC, σ_{RIC}, which is estimated using the equation of $\sigma_{\mathrm{RIC}} = \sigma_0 + KR^d$, based on electronic excitation model [4,5]. σ_0, K, R and d are the base conductivity, the value of the proportionality constant, the ionization dose rate which corresponds to reactor power and the value of the dose rate exponent, respectively. Their parameters were

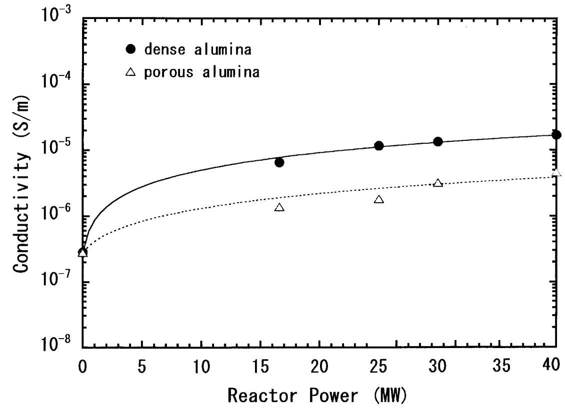

Fig. 4. RIC of dense and porous alumina at the reactor powers of 16, 25, 30 and 40 MW. The curves represent the theoretical values of RIC, which were based on electronic excitation model.

determined by fitting to the experimental data as a function of the reactor power, as follows; $\sigma_0 = 2.83 \times 10^{-7}$ S/m, $K = 6.0 \times 10^{-7}$ for dense alumina ($K = 1.3 \times 10^{-7}$ for porous alumina) and $d = 0.9$. The meaning of the experimental values of the RIC can be analyzed by considering the theoretical conductivity terms. The value of $\sigma_0 = 2.83 \times 10^{-7}$ S/m, measured in the reactor at the reactor power of 0 MW and room temperature, is higher by about five orders of magnitude than $>10^{-12}$ S/m at 293 K measured outside the reactor. The ionization dose rate in the shut-down reactor core is estimated to be around 10^6–10^7 R/h (28–280 Gy/s). Therefore, the measured base conductivity may include the RIC of the specimens as well as the cable and the sub capsule [10]. The difference for the value of K may be due to the band energy of dense versus porous alumina. There are shallow trapping states for electrons and holes near the conduction and valence bands [4]. The electrons falling into these states are thermally re-excited into the conduction band in order to maintain electrical charge balance. The thermal re-excitations from steady state Fermi levels for electrons and holes, respectively, dominate for the RIC of dense and porous alumina. Fig. 5 shows RIC of dense and porous alumina as functions of the irradiation time at 50 MW at about 620 K, as compared with the base conductivity at room temperature before and after the irradiation. The RIC of dense (porous) alumina at 50 MW decreased at the initial fluence and hereafter reached a constant at fast neutron fluences of about 5.5×10^{22} (2.5×10^{22}) n/m^2. It has been reported that the RIC of polycrystalline alumina is studied at 713–723 K during reactor irradiation and the decrease in conductivity at 3×10^{22} n/m^2 can be associated with the nucleation and growth of defect aggregates [8]. Moreover, it has been also reported that polycrystalline alumina of 99.5% purity in vacuum of 10^{-4} Pa is irradiated at 770–800 K under reactor irradiation and

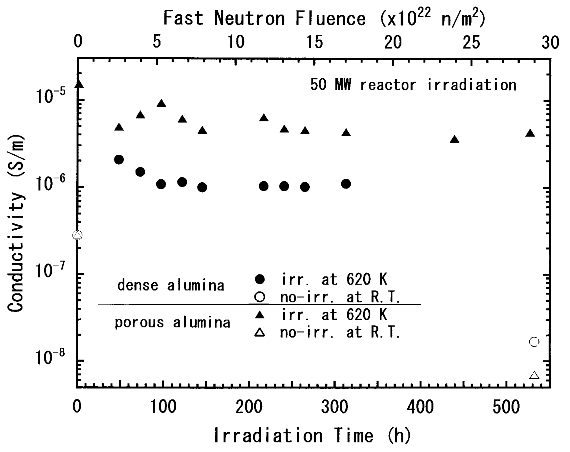

Fig. 5. Fluence dependence of RIC for dense and porous alumina at the reactor power of 50 MW at 620 K, as compared with those at room temperature before and after the reactor irradiations.

the RIC without electric field has a constant value in the fast neutron fluence of about 6.5×10^{24} n/m^2 [2]. The present result is similar to their previously reported results. However, the RIC of porous alumina was higher by about 3.3 times than that of dense alumina. As the guard ring geometry was not used in the present study, all the measured RIC and base conductivity may include leakage currents. Moreover, it has been reported by Moroño et al. that Wesgo alumina, has an extremely large grain size and is a more porous material with density of 3.86×10^3 kg/m^3, is highly susceptible to surface electrical degradation due to impurity segregation when irradiating in high vacuum, while Vitox alumina, is a far more pure material with density of 3.97×10^3 kg/m^3, is not [9]. Therefore, the conductivity of porous alumina at 50 MW and 620 K may be higher than that of dense alumina. Also, the difference between *V–I* behavior up to 40 and at 50 MW may be attributed to increases of electronic excitation (free electron carrier densities) as well as surface leakage currents with increasing the ionization dose rate and the irradiation temperature.

The base conductivity at room temperature after the irradiation became lower by about one order of magnitude than the initial base conductivity. The decreases of the base conductivity and the RIC at the initial fluence suggest that the band energy for the alumina is modified by changes in the levels between the valence and conduction bands, possibly due to point defects, dislocation loops, aggregates, impurity segregation, produced by atomic displacements, caused by neutron cascade collisions. The subsequent constant values of the RIC above 2.5–5.5×10^{22} n/m^2 may be due to the attainment of dynamic equilibrium between the formation and recombination of the defects.

4. Summary

The combined effect of gamma and neutron irradiation on RIC and RIEMF for alumina varying density was studied in situ using JMTR fission reactor in Oarai research establishment of JAERI. The dense and porous alumina specimens were irradiated for 22 reactor full power days under 50 MW reactor irradiation with ionization dose rate of 2.3 kGy/s, fast and thermal neutron fluxes of 1.4–1.6×10^{17} and 1.1–1.3×10^{18} n/m^2 s, respectively. Then the irradiation temperature increased up to 620 K. The RIC for both of them became higher by about two orders of magnitude than the base conductivity without radiation at 0 MW. Moreover, the RIEMF also occurred during irradiation. The RIEMF observed may be strongly influenced by magnesia insulating cables which are used to carry the electrical signals for in situ electrical conductivity measurement.

The RIC for dense and porous alumina increased, as the reactor power increased and the electronic excitation increased. However, the RIC at the first neutron fluence of above $\sim 10^{22}$ n/m^2 was modified due to the radiation induced defects and had a constant value in the fast neutron fluence range of 2.9×10^{23} n/m^2. The RIC of porous alumina was higher by about 3.3 times than that of dense alumina. It is supposed that the RIC may include surface leakage currents which are caused by no guard ring geometry and the porous alumina is highly more susceptible to surface electrical degradation than dense alumina.

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