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Radiation induced conductivity of ceramic coating materials under 14 MeV neutron irradiation

T. Tanaka [a,*], A. Suzuki [a], T. Muroga [a], F. Sato [b], T. Iida [b], T. Nishitani [c]

[a] Fusion Engineering Research Center, National Institute for Fusion Science, 322-6 Oroshi, Toki-shi, Gifu 509-5292, Japan
[b] Department of Electronic, Information Systems and Energy Engineering, Graduate School of Osaka University, 2-1 Yamada-oka, Suita, Osaka 565-0871, Japan
[c] Department of Fusion Engineering Research, Japan Atomic Research Institute, 2-4 Shirakata-shirane, Tokai-mura, Naka, Ibaraki 319-1195, Japan

Abstract

Irradiation with 14 MeV neutrons was performed on ceramic coating materials developed for V/Li blanket system to examine the electrical insulating properties in a radiation environment. Changes in the electrical conductivity of bulk specimens (Y_2O_3, $CaZrO_3$, Er_2O_3, AlN and $CaZr_{0.95}Sc_{0.05}O_3$) and coated specimens (stainless steel coated with AlN and Y_2O_3) were examined at room temperature by leakage current measurements under irradiation. Radiation induced conductivities of the bulk and coated specimens were 3.1×10^{-11}–1.3×10^{-10} and 6.3×10^{-12}–1.9×10^{-11} S/m for dose rate of 0.01 Gy/s, respectively. The values were the same level or one order higher compared with Al_2O_3 bulk specimens evaluated by previous experiments with various radiation species. The results indicate that the degradation of insulating properties under irradiation were within allowable levels for application to V/Li blanket system in the present experimental condition. Further evaluation is necessary at higher dose rate and higher temperature.

1. Introduction

The development of electrically insulating coating for the reduction of MHD pressure drop is an important issue for V/Li blanket systems [1]. Since the coating material will be exposed to highly corrosive liquid Li, fabrication and characterization of nonconventional ceramic materials have been carried out focusing on their stability in liquid Li [2–4]. Recently, it was confirmed that candidate ceramic materials of Y_2O_3, Er_2O_3, $CaZrO_3$ and AlN were stable in liquid Li of 1073 K for 1000 h [5,6].

For effective reduction of MHD pressure drop, the required electrical conductivity of insulating coating was estimated to be $<1.0\times10^{-2}$ S/m [6]. However, in V/Li blanket of a fusion reactor, ceramic insulators may be degraded by various factors including neutron and gamma-ray radiation, high temperature, corrosion by lithium and vanadium alloy substrate, erosion by Li flow, aging, etc. As to irradiation effects, degradation of insulating properties due to radiation induced conductivity is unavoidable and one of the most important issues for insulating materials to be used in intense radiation environment such as fusion blanket. Radiation induced conductivity is caused by excitation of electrons into the conduction band and observed as increase of leakage current flowing through the insulating material [7].

For ceramic materials such as Al_2O_3 and MgO, various irradiation experiments have been performed to examine degradation of the electrical properties in an irradiation environment. However, almost no data is available for ceramic materials developed for V/Li blanket system, much less for coated materials. In the present study, radiation induced conductivities of the candidate ceramic materials, both as bulk and as coated, were examined under 14 MeV neutron irradiation.

* Corresponding author. Tel.: +81-572 58 2319; fax: +81-572 58 2676.
E-mail address: teru@nifs.ac.jp (T. Tanaka).

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2. Experiment

Bulk specimens included an Y_2O_3 plate of $5.0\times5.0\times1.0$ mm^3 (3.3×10^{-13} S/m, supplied from TEP Corp.), an Er_2O_3 disc of 7.5 mm∅ × 1.0 mm (5.0×10^{-14} S/m, TYK Corp.), a $CaZrO_3$ disc of 15 mm∅ × 3.5 mm (8.8×10^{-13} S/m, TYK Corp.), an AlN plate of $10\times9.0\times1.0$ mm^3 (2.3×10^{-10} S/m, Tokuyama Corp.) and a $CaZr_{0.95}Sc_{0.05}O_3$ disc of 14 mm∅ × 3.0 mm (4.6×10^{-10} S/m, TYK Corp.) made by sintering method. As shown in Fig. 1, a sputtered Pt (~200 nm) guarded electrode configuration was used for prevention of surface leakage current. An electrode for bias voltage supply was made on the opposite side. The higher conductivities of the AlN and $CaZr_{0.95}Sc_{0.05}O_3$ specimens were likely due to large leakage currents flowing along grain boundaries. AlN and Y_2O_3 coated specimens were fabricated on stainless steel plates by an RF sputtering method [8]. Thickness of coating layers was 2.5 and 7.0 μm for AlN specimens (1.1×10^{-13} and 1.0×10^{-13} S/m) and 2.5 μm for Y_2O_3 specimen (8.0×10^{-13} S/m). On the coating layer, a central electrode of 4×4 mm^2 and a guard electrode were made by sputter deposition of Pt or painting of silver paste. Bias voltages were applied to the stainless steel plates.

Irradiation with 14 MeV neutrons was performed using the Fusion Neutronics Source (FNS) facility of Japan Atomic Research Institute. Fig. 2 shows a schematic arrangement of the measurement system. Specimens were located close to the tritium target bombarded with a 350 keV D^+ beam. A lead wire for leakage current measurement was connected to the specimen through acrylic pipe to prevent the influence of charges accumulated in a coaxial cable. A high resistance meter (Keithley 6517A) was used for both of voltage supply and leakage current measurement. The maximum neutron flux was 8.0×10^9 n/cm^2/s for the $CaZrO_3$ specimen and 4.2–7.8×10^8 n/cm^2/s for the other specimens. Neutron flux was varied by adjustment of the beam current and monitored with a ^{238}U fission chamber. From results of neutron transport calculations using MCNP code [9], contribution of induced gamma rays to total dose rate (Gy/s) were estimated to be ~30% for the Er_2O_3 specimen and ~10% for the other specimens. The radiation induced conductivities were examined from changes in bulk leakage currents flowing through the specimens at room temperature.

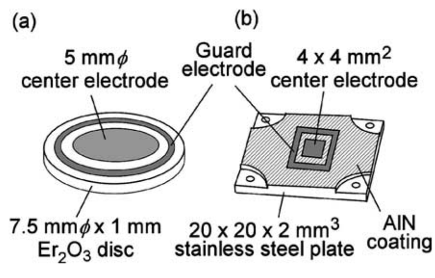

Fig. 1. Schematic drawing of ceramic specimens. (a) Er_2O_3 bulk specimen and (b) AlN coated specimen.

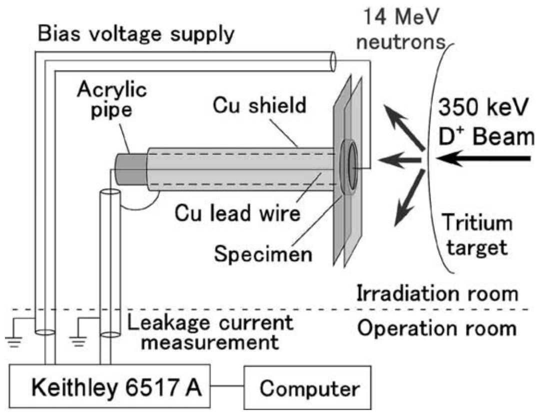

Fig. 2. Schematic arrangement of measurement system for examination of radiation induced conductivities.

3. Results and discussion

Fig. 3 shows an example of leakage current measurements under 14 MeV neutron irradiation. Changes in neutron flux and bulk leakage current flowing through the AlN coated specimen are plotted in Fig. 3(a) and (b). Time intervals of data acquisition are 10 s for neutron flux monitoring and 1 s for leakage current measurement. The leakage current changed coincidentally with the neutron flux. Similar responses of leakage currents were observed for all bulk and coated specimens. Bias voltage and flux dependence of the leakage current were plotted in Fig. 4(a) and (b). For all specimens, ohmic characteristics were observed in the relations between bias voltages and radiation induced currents. Radiation induced conductivities were evaluated from the flux dependence of leakage currents. The values extrapolated or interpolated to dose rate of 0.01 Gy/s were 1.3×10^{-10}, 5.3×10^{-11} and 3.1×10^{-11} S/m for the Y_2O_3, Er_2O_3 and $CaZrO_3$ bulk specimens, and 7.6×10^{-12}, 1.9×10^{-11} and 6.3 x 10^{-12} S/m for the AlN (2.5 μm), AlN (7.0 μm) and Y_2O_3 (2.5 μm) coated specimens, respectively.

Electrical properties of Al_2O_3 under radiation environment have been studied by various irradiation experiments [10–21]. Radiation induced conductivities for Al_2O_3 specimens evaluated previously are shown in Fig. 5 [17]. The figure indicates that the radiation induced conductivity is proportional to the dose rate and weakly dependent on radiation species. Results of the present 14 MeV neutron irradiation experiment were also plotted on the figure with larger symbols. Close and

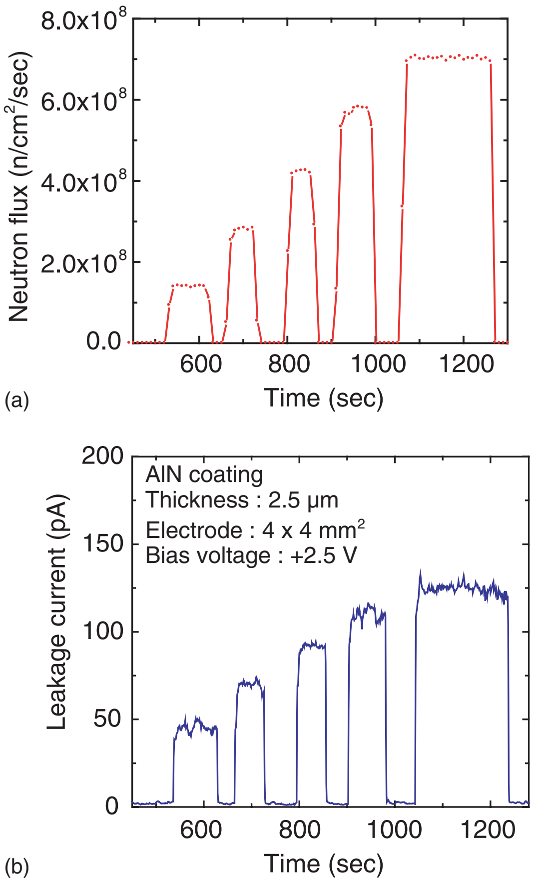

Fig. 3. Example of leakage current measurements under 14 MeV neutron irradiations. (a) Change in neutron flux. (b) Observed leakage current flowing through AlN coated specimen.

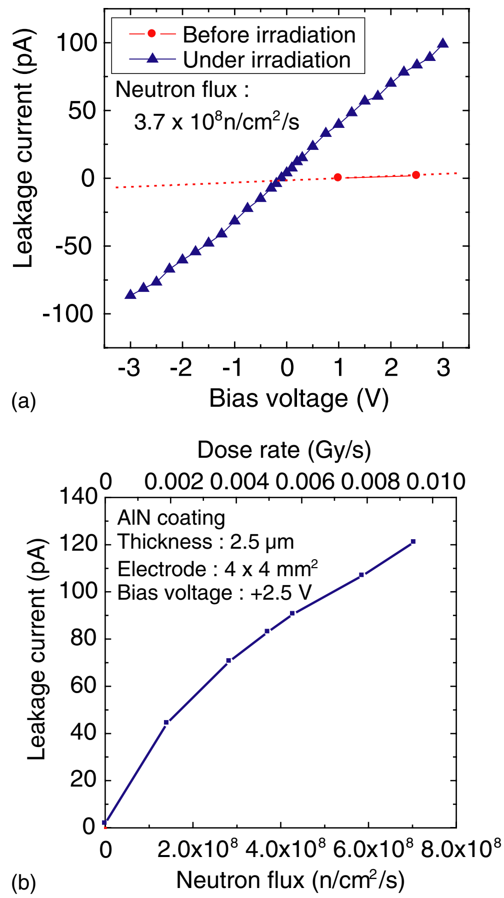

Fig. 4. (a) Bias voltage dependence and (b) neutron flux dependence of leakage current under irradiation obtained for AlN coated specimen.

open symbols are the results for the bulk and coated specimens, respectively. Radiation induced conductivities for the ceramic coating materials were the same level or one order higher compared with Al_2O_3 specimens. Around first wall in a fusion reactor with V/Li blanket system, dose rate is considered to be several kGy/s for

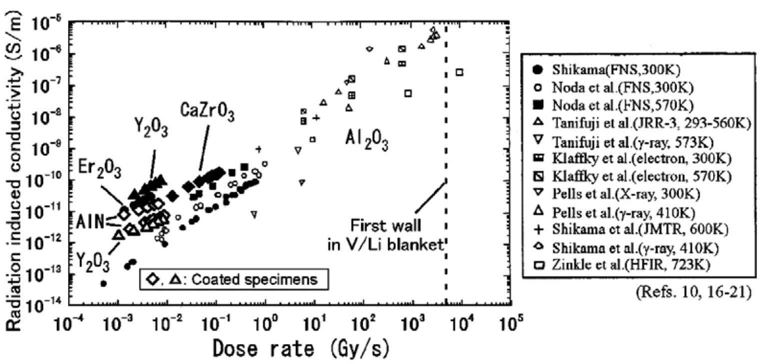

Fig. 5. Comparison of radiation induced conductivities. Plots for Al_2O_3 are extracted from Ref. [17]. The present results for ceramic coating materials are plotted with larger symbols. Close and open symbols are results for bulk and coated specimens, respectively.

ceramic materials [22]. The extrapolation of the present results indicates that the degradation of insulating properties under irradiation should be within allowable levels of 10^{-2} S/m [6] for application to V/Li blanket system in the present experimental condition.

Degradation of insulating properties was examined also for the AlN and $CaZr_{0.95}Sc_{0.05}O_3$ bulk specimens of 2.3×10^{-10} and 4.6×10^{-10} S/m. Under neutron irradiation, radiation induced conductivities extrapolated to 0.01 Gy/s were 1.5×10^{-10} and 5.5×10^{-12} S/m, respectively. The obtained values were almost same order as the other three bulk specimens. It is thus possible that influence of irradiation on leakage currents flowing along grain boundaries was considerably small. Further studies focusing on the irradiation effects on currents flowing along grain boundaries are necessary for the development of thin insulating coating.

4. Conclusion

Irradiation with 14 MeV neutrons was performed on bulk specimens (Y_2O_3, $CaZrO_3$, Er_2O_3, AlN and $CaZr_{0.95}Sc_{0.05}O_3$) and coated specimens (AlN and Y_2O_3) to examine the electrical insulating properties of candidate MHD coating materials in a radiation environment. During irradiation, bulk leakage currents flowing through the specimens changed coincidentally with neutron flux. Ohmic characteristics were observed in relations between bias voltages and increase of leakage currents induced by irradiation. Radiation induced conductivities of specimens evaluated from leakage current measurements were 3.1×10^{-11} – 1.3×10^{-10} and 6.3×10^{-12} – 1.9×10^{-11} S/m for dose rate of 0.01 Gy/s, respectively. The extrapolation of the present data indicates that the degradation of insulating properties under irradiation were within allowable levels for application to V/Li blanket system. Further evaluation is necessary at higher dose rate and higher temperature. Also necessary to be investigated are the influence of material parameters such as grain boundaries, impurities and cracks, and environmental parameters such as electric field.

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