Electrical insulating performances of SiC materials under gamma-ray irradiation

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For evaluation of electrical insulating performances of SiC materials in radiation and high temperature environments, electrical conductivities of single crystal and sintered SiC materials were measured under gamma-ray irradiations at temperatures of up to ~450 ^{o}C. The radiation induced conductivities (RICs) evaluated for single crystal SiC plates were 2.8 x 10^{-8} – 1.3 x 10^{-7} S/m under the irradiations of 2.3 Gy/s at room temperature. The RICs in the sintered SiC plates were almost below measurable level due to their high inherent conductivities. The magnitudes of the RICs did not increase significantly with temperature. Extrapolation from the present data indicates that the maximum magnitude of RIC in SiC materials would be the order of 10^{-4} S/m for a dose rate of several kGy/s at a first wall of a fusion reactor. The magnitude is adequately lower than the allowable electrical conductivities in the SiC flow channel inserts (FCIs) for the dual-cooled lithium lead (DCLL) blanket design.

Keywords: Silicon carbide, Electrical conductivity, Radiation induced conductivity, Gamma-ray irradiation

1. Introduction

Silicon carbide (SiC) materials have been studied in the fusion reactor development especially as a blanket material for the superior properties of high-temperature strength, irradiation resistance, chemical stability, low activation etc [1, 2]. In the conceptual design of the Dual Cooled Li-Pd (DCLL) blanket system, install of SiC flow channel inserts (FCIs) has been proposed for high efficiency operation at higher temperature region. The design indicates that the maximum coolant temperature of ~800 ^{o}C could be achieved by attenuating the temperature of ferritic steel blanket walls with the SiC FCIs. In this concept, electrical insulation is also a required function of the SiC FCIs for suppression of MHD pressure drop, since electrically conductive liquid Li-Pb coolant flows through the FCIs across strong magnet fields. The maximum allowable electrical conductivity for the FCIs has been limited to 100 S/m [3].

Pure ceramic materials generally indicate adequate electrical insulating performances. However, the SiC materials studied for the FCIs contain sintering additives in the fabrication process. While the additives improve the mechanical properties, fabrication process etc. [9], they might affect the electrical insulating performance significantly. In addition, the electrical insulating performances will be degraded by radiation and temperature effects under a fusion reactor environment. Radiation effects on the electrical performances have been studied profoundly for ceramic materials of Al_2O_3, MgO etc. used in diagnostics systems [4-7]. Although a few data for sintered SiC materials have been reported [8], similar database is required to be constructed also for SiC materials.

In the present study, degradation of the electrical insulating performances of SiC materials due to radiation induced conductivities (RICs) and the temperature dependences were examined by gamma-ray irradiations at temperatures of up to ~450 ^{o}C. The irradiations were performed on monolithic sintered plates studied for the FCIs and especially on high-purity single crystals to understanding the fundamental electrical properties of SiC under a high temperature and radiation environment.

2. Experiment

Measurements of electrical conductivities were performed for two types of single crystal SiC plates and three types of sintered SiC plates. The single crystal SiC plates of 10 mm x 10 mm were prepared by cutting a 50 mmϕ high-purity semi-insulating wafer and a 75 mmϕ n-type semiconductor wafer supplied from Cree, Inc.

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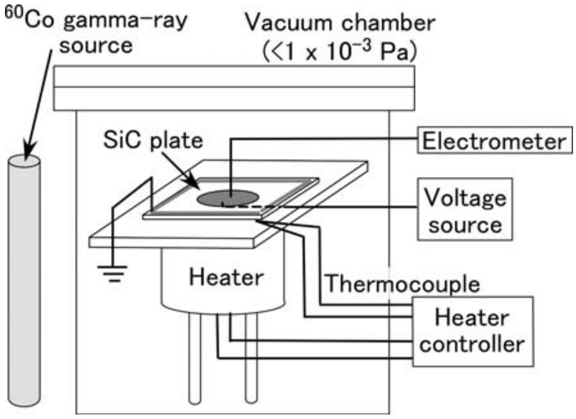

Fig.1 Schematic drawing of electrical conductivity measurement of SiC materials under gamma-ray irradiation.

The thickness of the plates was 0.39 mm and the crystal structure was 4H-SiC. Sintered SiC plates were monolithic Hexoloy-SA® commercially obtained from Hitachi chemical Co., Ltd. and monolithic NITE-SiC (NITE; Nano-Infiltration Transient-Eutectic Phase) fabricated in Kyoto University [9]. The dimensions of the sintered plates were 10 mm x 10 mm x 1 mm. In addition to the above monolithic sintered plates, sandwiched plates consisting of Hexoloy-SA (0.45 mm in thickness) / NITE-SiC (0.1 mm) / Hexoloy-SA (0.45 mm) layers were also prepared with the same dimensions.

Schematic drawing of the conductivity measurements is shown in Fig. 1. On the single crystal SiC plates, Al or Pt electrodes were deposited by resistance heating or DC sputtering. The diameters were 5 mm for the current measurement side and 8 mm for the bias voltage side. On the sintered monolithic and sandwiched SiC plates, Pt electrodes were deposited by DC sputtering. The sizes were 5 x 5 mm^2 for the current measurement side and 9 x 9 mm^2 for the bias voltage side. Guard electrodes were also deposited around the current measurement electrodes to avoid surface currents flowing across the edges of the plates. Electrical conductivities of the SiC plates were evaluated from relations between bias voltages and currents flowing through the plates.

Examination of electrical insulating properties under gamma-ray irradiations were performed at the ^{60}Co gamma-ray irradiation facility of the Institute of Scientific and Industrial Research, Osaka University. The SiC plate was set in a vacuum chamber evacuated to < 10^{-3} Pa. The averaged dose rate estimated with the MCNP Monte Carlo code [10] was 2.3 Gy/s in a SiC plate at the specimen position. Magnitudes of radiation induced conductivities (RICs) of all single crystal and sintered SiC plates were evaluated from differences between conductivities measured with and without gamma-ray irradiations. By changing the position of the gamma-ray source, the dose rate dependence of the RIC was examined for a semi-insulating single crystal SiC sample in the region of 0.01–2.3 Gy/s at room temperature. To examine the temperature effects on RICs, the semi-insulating single crystal and Hexoloy/NITE/Hexoloy sandwiched SiC plates were heated up to ~450 ^{o}C and the conductivities were measured under the gamma-ray irradiations.

Table 1 lists the specimen names and examinations performed in the present study.

3. Results

Fig. 2 shows the current-voltage (I-V) curves of the single crystal SiC plates measured without and under the gamma-ray irradiations at room temperature. In the semi-insulating #1 and #2 plates, both the currents without and under irradiations showed the ohmic characteristics (Fig. 2 (a)). The inherent electrical conductivities of the semi-insulating plates without gamma-ray irradiations were 4.0 x 10^{-14} and 1.0 x 10^{-13} S/m, respectively. The orders of the inherent conductivities were almost similar to those of oxide ceramic materials such as Er_2O_3 and Y_2O_3 examined in our previous studies [11]. Under the gamma-ray irradiations of 2.3 Gy/s, the electrical

Table 1 SiC specimens and examinations performed in present study.

Specimen		Without irradiation	Under gamma-ray irradiation		
		Inherent conductivity	RIC evaluation for 2.3 Gy/s	Dose rate dependence	Temperature Dependence
High-purity single crystal	Semi-insulating #1	O	O	O	RT - 450 ^{o}C
	Semi-insulating #2	O	O	-	-
	n-type	O	O	-	-
Sintered	Hexoloy-SA	O	-	-	-
	NITE-SiC	O	O	-	-
	Hexoloy/NITE/Hexoloy	O	O	-	RT - 450 ^{o}C

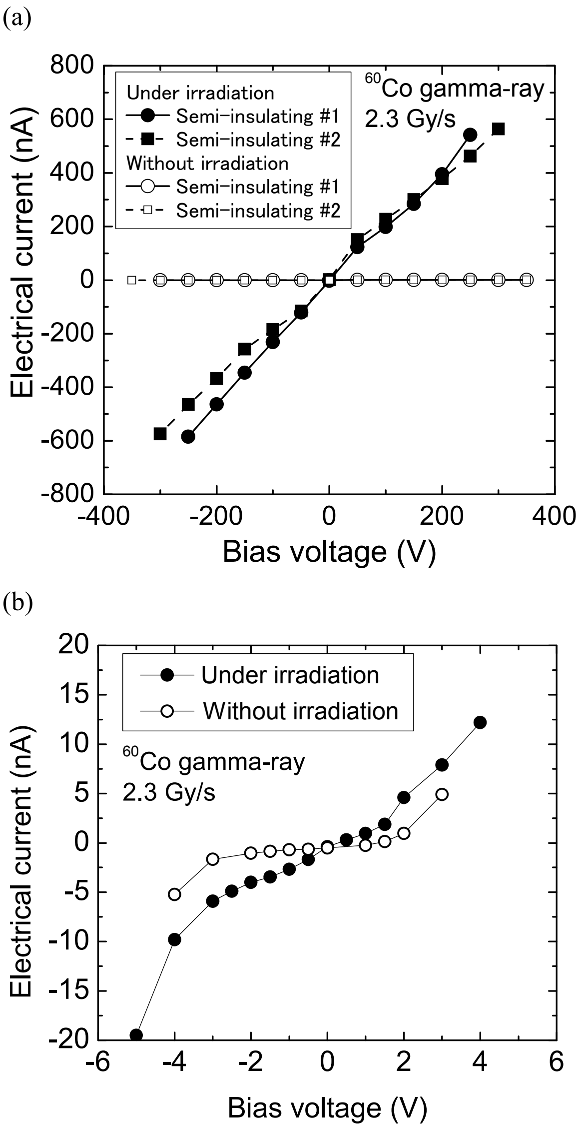

Fig.2 Current-voltage (I-V) curves of (a) semi-insulating and (b) n-type single crystal SiC plates measured under gamma-ray irradiation and without irradiation at room temperature.

conductivities of the semi-insulating plates increased to 4.3 x 10^{-8} S/m and 3.7 x 10^{-8} S/m. From the difference in conductivities measured without and under the irradiations, the magnitudes of the radiation induced conductivities (RICs) in the semi-insulating plates were evaluated to be 3.7 – 4.3 x 10^{-8} S/m for the dose rate of 2.3 Gy/s. In the measurement of the I-V curve of the n-type single crystal SiC plate, the current increased rapidly with the bias voltages of higher than +2 V and lower than -3 V (Fig. 2 (b)). It is considered that a rectification behavior of the n-type semiconductor did not appear for the low bias voltages due to the electrode material selected in the present evaluation. Increase of the electrical current in the n-type plate was observed under the gamma-ray irradiation and the shape of the I-V curve under the irradiation was similar to that obtained without irradiation. The conductivities for the bias voltage of -3 V were 1.1 x 10^{-8} S/m without irradiations and 3.9 x 10^{-8} S/m under the irradiation, respectively. From the difference, the magnitude of the RIC in the n-type plate was evaluated to be 2.8 x 10^{-8} S/m for the dose rate of 2.3 Gy/s.

The inherent electrical conductivities of the sintered monolithic SiC plates without irradiations were 1.1 x 10^{-6} S/m in the Hexoloy-SA plate and 3.2 x 10^{-2} S/m in the NITE-SiC plate at room temperature. In the Hexoloy/NITE/Hexoloy sandwiched plate, the inherent conductivity was dominated by the Hexoloy-SA layers with a higher insulating performance and the value was 1.3 x 10^{-6} S/m at room temperature. Magnitudes of RICs were examined for the NITE-SiC and Hexoloy/NITE/Hexoloy sandwiched plates at room temperature. In the case of the NITE-SiC plate, an increase in the conductivity under the gamma-ray irradiation could not observed since the inherent conductivity was relatively high and the conductivity increased easily by a thermal conduction from the radiation source. In the measurement for the sandwiched plate, the RIC was evaluated to be 1.6 x 10^{-8} S/m under the irradiation of 2.3 Gy/s. However, since the magnitude of the RIC was ~1/100 of the inherent conductivity, the measurements with more accurate temperature control might be required for evaluation of the factor of the value. Table 2 summarizes the above measurement results at room temperature.

The dose rate dependence of the RIC in the semi-insulating #1 plate is plotted in the Fig. 3 with the results described in the above paragraphs. While the same semi-insulating sample (semi-insulating #1) was examined, the magnitude of RIC in the dose rate dependence measurement was 1.3 x 10^{-7} S/m for the dose rate of 2.3 Gy/s, which was three times larger than the result of the I-V curve measurement described in the above paragraph. The magnitude of the RICs might be affected by a condition and behavior of electron trapping levels in the crystal. In Fig. 3, the RICs of the SiC plates are compared with those of several oxide ceramic materials obtained in our previous experiments [11]. Magnitudes of RICs of Al_2O_3 materials, which have been studied most profoundly, were almost the same order as those of the Y_2O_3 material plotted in Fig. 3 [12]. In the present evaluation, the RICs of the SiC plates were 2-3 orders higher than those of the oxide ceramic materials. It is considered that the drift distance of radiation excited charge carriers in the SiC materials is 2-3 orders larger compared with that in the oxide ceramic materials.

The temperature dependences of the electrical conductivities of the SiC plates are shown in Fig. 4. The applied bias voltage was 250 V or -250 V for the semi-insulating #1 plate and +1 V for the Hexoloy/NITE/Hexoloy plate. The conductivity of the semi-insulating #1 plate without a gamma-ray irradiation

Table 2 Electrical conductivities of SiC specimens before gamma-ray irradiation and magnitudes of radiation induced conductivities (RICs) for dose rate of 2.3 Gy at room temperature.

Specimen		Inherent conductivity (S/m)	RIC for 2.3 Gy/s at room temperature (S/m)
High-purity single crystal	Semi-insulating #1	4.0×10^{-14}	4.3×10^{-8}
	Semi-insulating #2	1.0×10^{-13}	3.7×10^{-8}
	n-type	1.1×10^{-8}	2.8×10^{-8}
Sintered	Hexoloy-SA	1.1×10^{-6}	-
	NITE-SiC	3.2×10^{-2}	Under measurable level
	Hexoloy/NITE/Hexoloy	1.3×10^{-6}	1.6×10^{-8}

increased drastically with temperature. In contrast, significant increase in the conductivity was not observed up to ~300 °C under the irradiation. The conductivity under the irradiation was 1.3×10^{-7} S/m at room temperature and decreased gradually to 2.6×10^{-8} S/m at ~200 °C. It could be considered that shallow trapping levels became vacant by thermal excitation and the drift distance of radiation excited charge carriers decreased due to trapping to the vacant levels. At temperatures of >300 °C, the conductivities obtained without and under the irradiation were almost same magnitudes and increased with temperature. The results indicate that the magnitude of RIC of the high-purity single crystal SiC material does not increase significantly with temperature. This temperature dependence of the RIC is similar to that of other oxide ceramic materials [11]. From exponential curve fitting for the temperature dependences, the electrical conductivity in the semi-insulating #1 plate was considered to be dominated by a shallow energy level under 200 °C. The estimated donor (or acceptor) energy level using the equation of $\sigma = \sigma_0 \exp(-E_d/kT)$ was 0.2-0.4 eV due to scattering in the plots. In this equation, σ is conductivity and E_d is the dominant energy level in the material. For the region higher than 200 °C, the dominant energy level was calculated by using the equation for an intrinsic semiconductor, i.e. $\sigma = \sigma_0 \exp(-E_g/2kT)$, and estimated to be 3.4 eV. E_g is the band gap energy of the material. The value was close to 3.3 eV in literature [13].

The temperature dependence of the conductivity was examined also for the sandwiched plate of Hexoloy/NITE/Hexoloy, whose insulating property is dominated by the Hexoloy-SA layers. Since the inherent conductivity without the irradiation was relatively high, significant increase due to the gamma-ray irradiation was not observed up to ~450 °C except for 1.6×10^{-8} S/m measured at room temperature described above. The temperature dependence of the conductivity of the NITE-SiC plates were also plotted in the Fig. 3 for

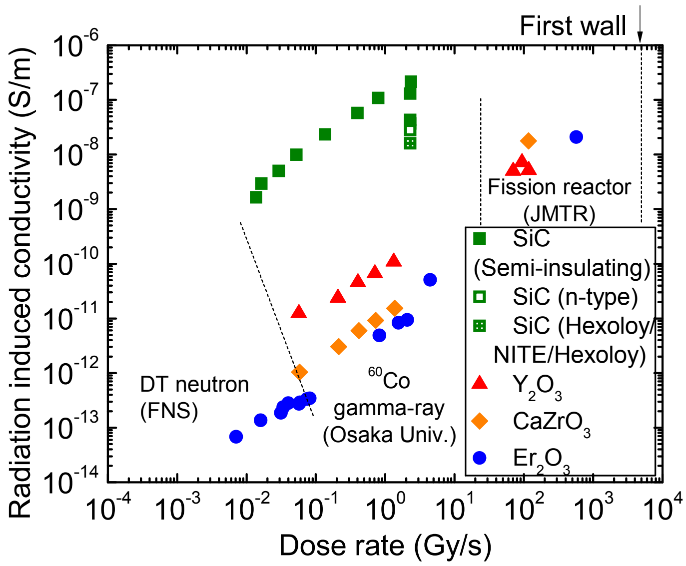

Fig.3 Magnitudes of radiation induced conductivities (RICs) in the SiC materials obtained at room temperature and comparison with those of oxide ceramic materials [11]. All data for the oxide ceramic materials have been obtained at low temperatures from room temperature to 50 °C.

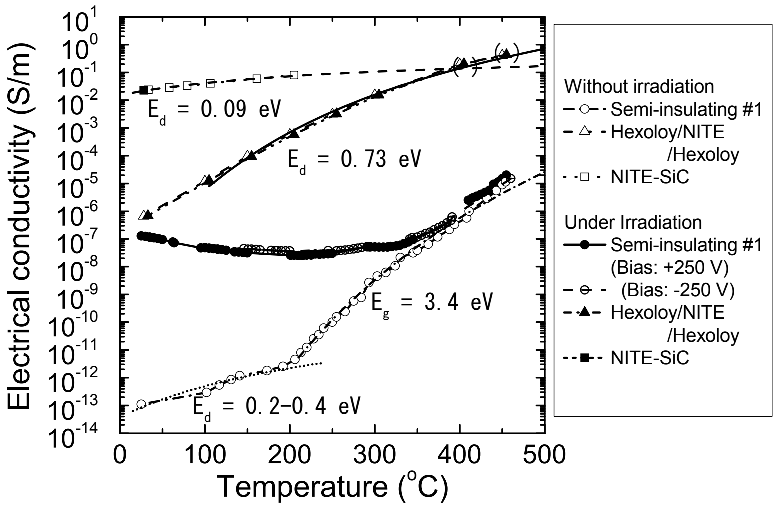

Fig.4 Temperature dependence of electrical conductivities of SiC materials under the gamma-ray irradiations and without irradiation. Plots of the Hexoloy/NITE/Hexoloy specimen at 400 and 450 °C might be several tens percent smaller due to the electrical resistance of the electrode. E_d: Estimated donor (or acceptor) level dominating conductivity. E_g: Estimated band gap energy.

comparison. For the NITE-SiC plate, the conductivity measurement under the irradiation was performed only at room temperature and significant increase could not be observed. In the measurement circuit, a maximum electrical resistance of the thin Pt electrode was ~25 Ω. Therefore, it is considered that the plots of the Hexoloy/NITE/Hexoloy sample at 400 and 450 °C in Fig. 3 were several tens percent lower than actual magnitudes, while they indicate that the RIC does not increase significantly at the high temperatures. The energy levels dominating the conductivities of the Hexoloy/NITE/Hexoloy and NITE-SiC plates were estimated to be ~0.73 eV and ~0.09 eV by using the equation of $\sigma = \sigma_0 \exp(-E_d/kT)$.

4. Discussions

The magnitude of radiation induced conductivity (RIC) is considered to be almost proportional to the drift distance of radiation excited charge carriers of electrons and holes. Assuming the energy for an electron-hole pair production is three times larger than the band gap energy, the density of the pairs are calculated to be ~5 x 10^{21} pairs/m^3/s under the gamma-ray irradiation of 2.3 Gy/s. If all the electrons and holes are collected by electrodes in the semi-insulating #1 plate with the thickness of 0.39 mm, the conductivity will be 4.9 x 10^{-7} S/m for the bias voltage of +250 V. However, the maximum RIC was 1.3 x 10^{-7} S/m in the present measurement. Also assuming that a drift distance of electrons is significantly longer than that of holes and dominates the magnitude of RIC, this indicates that the drift distance of electrons is almost 0.39 x (1.3 x 10^{-7} / 4.9 x 10^{-7}) mm, i.e. ~100 nm for the bias voltage of +250 V. The drift distance is considered to be longest in the high-purity single crystal due to the minimum trapping rate by impurities, defects and grain boundaries, and the magnitude of RIC would be largest.

Although the maximum temperature was ~450 °C in the present experiments due to the heater performance and limitation of irradiation time, it is considered possible to extrapolate the present results to a fusion blanket condition with a higher dose rate and temperature. In a fusion blanket, the highest dose rate is estimated to be several kGy/s at the fist wall [14]. Assuming that the RICs are almost proportional to a dose rate and weakly depend on radiation species, the magnitude at the first wall is extrapolated to be order of 10^{-4} S/m from the present evaluation for the high-purity single crystal plates. This magnitude is adequately small compared with the requirement on the SiC FCIs for the DCLL blanket, i.e. 100 S/m. Under the radiation environment of the blanket, the RIC would dominate the electrical insulating performance of the semi-insulating single crystal plates up to ~500 °C. In the case of the Hexoloy/NITE/Hexoloy plate, the RIC would be dominant up to ~100 °C. In a higher temperature region, the insulating performances would be dominated by the inherent electrical conductivities, i.e. thermal excitation of electrons.

5. Conclusion

Electrical insulating performances of single crystal and sintered SiC materials were examined by conductivity measurements under gamma-ray irradiations at temperatures up to ~450 °C. The radiation induced conductivities (RICs) evaluated for the semi-insulating single crystal plates, in which the magnitudes of RICs are considered to be largest, were 2.8 x 10^{-8}-1.3 x 10^{-7} S/m under the irradiations of 2.3 Gy/s at room temperature. For the sandwiched plate of sintered SiC layers (Hexoloy-SA/ NITE-SiC/ Hexoloy-SA), the RIC was 1.6 x 10^{-8} S/m. In the NITE-SiC plate, the RIC was under measurable level due to the high inherent conductivity. The maximum magnitude of RIC extrapolated from the present results would be the order of 10^{-4} S/m for the dose rate of several kGy/s at the first wall of a fusion blanket. The value is adequately lower than the allowable electrical conductivities in the SiC FCIs for the DCLL blanket design, i.e. 100 S/m. Significant increases in the magnitudes of the RICs were not observed at temperatures of up to 450 ^{o}C. At a high temperature region in a fusion blanket, thermal excitation of electrons would dominate the insulating performances of SiC materials. Influences of radiation damages on electrical properties will be issues in irradiation effects to be studied for the electrical applications of SiC materials in a fusion blanket.

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