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Impedance measurements of thin film ceramics under ion beam irradiation

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Abstract

High-purity alumina films were made by a laser ablation method, and its impedance was measured under ion beam irradiation. Change of radiation induced conductivity per the dose rate under ion beam irradiation was a few orders of magnitude lower than that under gamma-ray irradiation. Pulsed X-ray irradiation experiments were performed to determine the lifetime of the charge carrier in the crystalline alumina. The carrier lifetime was $\sim 10^{-7}$ s and the mobility of the charge carriers responsible for RIC in the crystalline alumina was estimated to be $\sim 10^{-5}$ $m^2/V/s$.

1. Introduction

Radiation-induced conductivity (RIC) and radiation-induced electrical degradation (RIED) of insulator materials are very important issues for the design of a future nuclear fusion device [1]. Many electrical components such as feedthroughs, mineral insulated cables and sensors with a high voltage require the high electrical resistivity for the insulation under severe conditions of a high radiation dose and high temperature. Thus, radiation effects on insulator materials have been investigated with neutron [2–6], electron beam [7–11] and ion beam irradiation [12–16]. The RIC of insulators increases with radiation dose rate, and the mechanism of the RIC is related to the production of electron–hole pairs and the movement, trapping and recombination of such charge carriers. This paper discusses the lifetime and mobility of charge carriers responsible for the RIC of Al_2O_3 sample. Proton experiments were performed to measure the impedance of the thin alumina films and the lifetime of charge carriers in the crystalline alumina was obtained with pulsed X-ray irradiation.

2. Experimental methods

Measurements of the impedance of a thin alumina films were performed under 100 keV proton beam irradiation [17]. The measurement system was composed of a sample holder, a sine-wave generator, an impedance analyzer and related electronic devices. The sample holder was designed for impedance measurement over a wide frequency range, and it was set in the center of a target vacuum chamber at room temperature. The target vacuum chamber was evacuated with a turbo-molecular pump and its vacuum level was kept better than 3×10^{-5} Pa. The impedances of the alumina films were measured in the frequency range of 0.1–100 kHz.

Fig. 1 shows a cross-section view of the thin alumina sample. An aluminum electrode of about 100 nm in thickness was at first deposited on a sapphire substrate using an evaporation process, and then the thin alumina layer was deposited on the substrate at room temperature using a pulsed laser ablation method [18,19]. A Nd:YAG laser (266 nm) was used for the laser irradiation. A high-purity crystalline alumina (SA-100;

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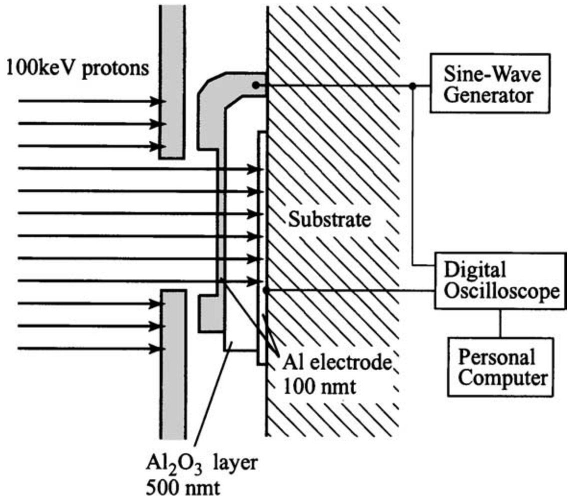

Fig. 1. Cross-section view of alumina films for proton irradiation and the electrical measurement system.

Kyocera Inc.) was ablated at a laser beam intensity of 3 J/cm^2 on the target and the vacuum pressure during the ablation was 10^{-5} Pa. Its major impurities were Si (10 ppm), S (4 ppm), Fe (2 ppm), Na, K, Ca (1 ppm), and Ti, Y, Zr, Cr (<1 ppm). This method was effective in preventing impurities from mixing. The thickness of the alumina layer was adjusted by means of the number of laser shots. The thickness of the alumina layer was also measured with ellipsometery, and it was determined to be about 500 nm. Moreover, an aluminum electrode of about 100 nm in thickness was deposited on the alumina layer using the evaporation process. The size of the electrodes was 4 mm in diameter and was equal to that of a beam collimator for the ion irradiation. The capacitance of the sample was 2.1 nF (100 kHz) and the dielectric constant of the alumina sample was estimated to be 9.8. The RIC for the thin alumina films was measured using 200 TBq ^{60}Co gamma-ray source, and the electrical conductivity was compared to SA-100 crystalline alumina samples with 50 μm thickness. The samples were supplied with AC and DC bias voltages, which strength of the electric field was 200 ± 50 kV/m. For the thin alumina films, negative current at 0 V was observed during gamma-ray irradiation. In AC measurement, the RIC coefficient c is defined as the conductivity σ divided by the ionization dose rate D. The RIC coefficient for the alumina films was a factor of 6 smaller than that for the crystalline alumina sample. The RIC coefficient for the crystalline alumina sample is estimated to be $\sim 10^{-10}$ (S/m)/(Gy/s) from results of gamma-ray irradiation experiments [20].

The sample was irradiated with 100 keV protons at room temperature. In this experiment, typical beam intensity was 5×10^{12} p/cm^2/s. The ionization dose rate for the sample was calculated with the TRIM code [21].

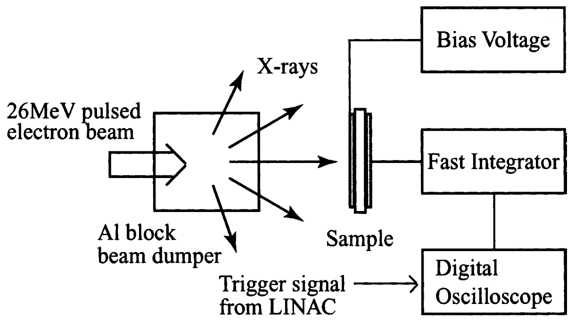

Fig. 2. Schematic drawing of experimental arrangement and block diagram of electrical measurement system in pulsed X-ray irradiation experiment.

Electronic and nuclear energy loss for 100 keV protons in Al_2O_3 are 200 and 0.4 keV/μm, respectively. The fraction of the protons passing through the alumina layer was ~79%. The average ionization dose rate for the alumina layer was estimated to be 330 kGy/s for the 100 keV protons flux of 5×10^{12} p/cm^2/s.

In order to examine the behavior of the electric charge induced in the crystalline alumina (SA-100), a pulsed X-ray irradiation experiment was performed with an electron linear accelerator (LINAC) at the Institute for Science and Industrial Research of Osaka University [22]. Fig. 2 shows a schematic drawing of the experimental arrangement and a block diagram of the electrical measurement system. X-ray pulses with an 8 ns time width were generated by means of the injection of a pulsed 26 MeV electron beam into a beam dump. The intensity of the pulsed X-rays was monitored with an ionization chamber. The crystalline alumina sample was 50 μm in thickness. The response time of the measuring system was less than 10 ns. The strength of the electric field was 200 kV/m in the sample. The response of the crystalline alumina sample to the pulsed X-rays, in other words, the pulse shape of the current flowing in the alumina sample was measured with a digital oscilloscope, which was triggered by the pulse generator for the electron accelerator. An averaging method with the same timing was introduced for the reduction of random noise.

3. Results and discussion

The impedance of the thin alumina sample was determined from the ratio of the input sine-wave voltage to the output sine-wave current. The impedance measurement was performed in situ during the proton irradiation. The incident proton beam was observed as DC current increase of the sample. Equivalent circuit model of parallel resistance–capacitor for the thin sample was obtained from the impedance analysis. The electrical conductivity and permittivity during proton irradiation are shown in Fig. 3. In the present experiment, the electrical conductivity without proton irradiation was $\sim 10^{-13}$ S/m and the RIC of 10^{-10} S/m was observed during beam irradiation at dose rate of 330 kGy/s. There was no large difference of the permittivity between with and without proton irradiation. The magnitude of the RIC coefficient of the alumina films was several orders of magnitude lower than that obtained under gamma-ray irradiation. Further discussions for difference in the RIC coefficient between protons and gamma-rays are necessary for detailed experiments. It may be presumed that RIC coefficient for protons is concerned with energy loss per length of charged particles in materials. For heavy charged particle, the density of production of electron and hole pairs in the ion path is high, and probability of electron–hole recombination is also higher than that for the light charged particle with the same energy. For semiconductor and scintillation radiation detectors, it is known that charge carrier collection and scintillation efficiency are deficient for heavy charged particles.

The RIC of insulators is determined by the density, mobility and lifetime of charge carriers. In this paper the RIC coefficient for the pure crystalline alumina is assumed to be $\sim 10^{-10}$ (S/m)/(Gy/s) from results of gamma-ray irradiation experiments. According to studies of the electrical properties of ceramics [23], the conductivity is proportional to the product of the density n and mobility μ of charge carriers. Thus the RIC coefficient c is expressed by

$$c \approx nq\mu/D, \tag{1}$$

where q is elementary electric charge.

Radiation produces electron–hole pairs in ceramics, and the number of produced electron–hole pairs (charge carriers) is approximately estimated from the ionization dose and W, which is the energy necessary for the production of an electron–hole pair in a material [24]. In ceramics under an electric field, electrons drift slightly and recombine with holes and/or are captured in trapping sites such as lattice defects. Thus, the density of charge carriers in ceramics under irradiation is roughly expressed by

$$n \approx \rho\tau D/W, \tag{2}$$

where ρ is density of the ceramic sample, τ the lifetime of charge carriers and W. The value of W is said to be roughly three times of the band-gap energy of the material.

In the present experiment, the lifetime of charge carriers in the crystalline alumina was estimated from the measurement of the decay time of the currents induced by the pulsed X-ray irradiation. Fig. 4 shows an example of analyzed data on output signals for the crystalline obtained from a fast integrator with a response time less than 10 ns. It was assumed from the figure that the lifetime of the charge carrier for the crystalline samples alumina was $\sim 10^{-7}$ s. For thin alumina films, the lifetime of carriers was not undoubtedly observed, possibly due to the sample thickness was short compared to mean free path of carriers within lifetime.

As for the mobility of charge carriers in the alumina, we obtained $\sim 10^{-5}$ m^2/V/s for the crystalline alumina from the RIC data and Eqs. (1) and (2). This value is similar to that of mobility estimated with temperature-dependence RIC model of Al_2O_3 crystals ($\sim 10^{-4}$ m^2/V/s at RT) [25], and a few orders of magnitude larger than the mobility for Al_2O_3 powder samples which has been obtained from pulsed electron irradiation experiments ($<10^{-8}$ m^2/V/s) [26]. The smaller carrier mobility may have much to do with excited state molecules. It is known that the mobility in several ceramic materials

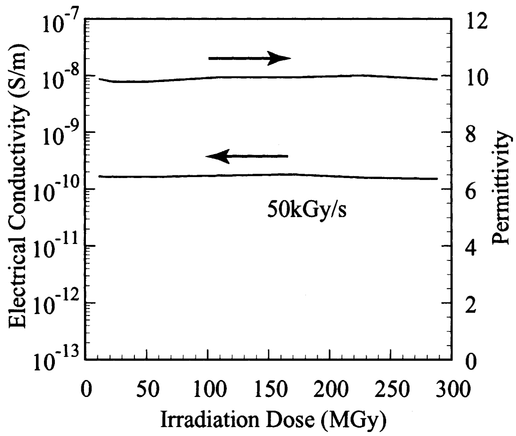

Fig. 3. Electrical conductivity and permittivity during proton irradiation.

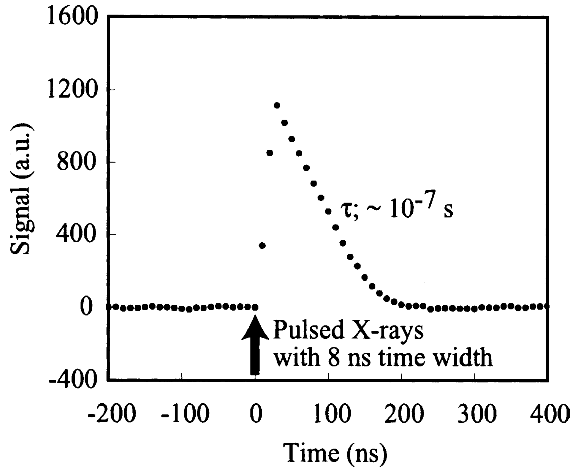

Fig. 4. Examples of output signals from a fast integrator for the crystalline alumina (SA-100) irradiated with pulsed X-rays.

without radiation is less than 10^{-4} $m^2/V/s$. Detailed experiments and further discussions are necessary for the clarification of the electrical characteristics of ceramics under irradiation.

4. Conclusions

Impedance measurements of the thin alumina films grown by pulsed laser deposition were performed under 100 keV proton beam irradiation. The electrical conductivity and permittivity estimated from parallel resistance–capacitor circuit model for the sample were obtained. The RIC increase for the thin films was observed during beam irradiation, and no large difference of the permittivity between with and without proton irradiation was observed. The magnitude of the RIC coefficient of the thin alumina films per dose rate was several orders of magnitude lower than that obtained under gamma-ray irradiation.

The lifetime of charge carriers for RIC was measured with a pulsed X-ray irradiation experiment, and the lifetime of the charge carrier for the crystalline alumina samples was $\sim 10^{-7}$ s. The mobility of the charge carriers responsible for RIC in the crystalline alumina was estimated to be $\sim 10^{-5}$ $m^2/V/s$.

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