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Production and reaction kinetics of radiation-induced defects in α-alumina and sapphire under ion beam irradiation

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Abstract

The production behavior of radiation-induced defects in α-alumina and sapphire was studied by in situ luminescence measurement technique under ion beam irradiation of He^+. The irradiation time dependence of the luminescence intensities of the F^+ centers and F^0 centers at 330 nm and 420 nm, respectively, was measured at each temperature from 298 to 523 K. By considering that the luminescence intensities represent the accumulated F^+ and F^0 centers, the observed irradiation time dependence was analyzed to obtain the rate constants for the production and reaction kinetics of radiation-induced defects of F-type centers.

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1. Introduction

Irradiation behavior of ceramic materials has been studied for many years [1]. However, the dynamic production behavior of radiation-induced defects under irradiation has not been fully clarified in spite of the considerable progress in understanding many aspects of radiation-induced defects in these ceramic materials. It is thus important to know the production behavior of radiation-induced defects by such an in situ luminescence measurement technique, which may be applied to monitor radiation-induced defects as reported previously [2].

In our previous study [2], the production behavior of radiation-induced defects in α-alumina and sapphire was studied by in situ luminescence measurement technique under ion beam irradiation of H^+ and He^+. An irradiation history was observed repeatedly with annealed specimens, in which the luminescence intensities increased with irradiation time and reached the steady state ones, and the possibility of using this technique to monitor radiation-induced defects was indicated. The steady state luminescence intensity of the F^+ centers at 330 nm was observed to decrease monotonically with the increasing temperature up to 800 K, while the intensity of the F^0 centers at 410 nm to show non-monotonic temperature-dependence. In the latter case, the intensity decreased with the temperature up to 600 K, and then increased above this temperature. These observations were successfully analyzed by considering the production mechanisms and reaction kinetics of the radiation-induced defects of F-type centers. In spite of successful analysis, however, it is still needed to improve our knowledge on the reaction kinetics since the first-order reactions have been assumed for all the reactions for simplicity.

Following our previous study [2], the irradiation time dependence of the luminescence intensities of the F^+ and F^0 centers was more extensively measured in the temperature range from 298 to 523 K in the present study. The measurement was made at the lower temperatures to decrease such effects as of thermal quenching. By considering that the luminescence intensities represent the accumulated F^+ and F^0 centers and by considering the second-order reactions, the observed irradiation time dependence was analyzed to

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obtain the reaction rate constants for radiation-induced defects of F-type centers together with those for oxygen interstitials.

2. Experimental

Procedures were mostly the same as those in the previous study [2]. The pellet-type specimens of α-alumina (Al_2O_3: 99.9%, SiO_2: 0.04%, Na_2O: 0.02%, MgO: 0.01%, CaO: 0.01%, Fe_2O_3: <0.01% [3]) and sapphire were obtained from Kyocera Co. Ltd., which were of 10 mm in diameter and about 0.5 mm in thickness. Each specimen was irradiated with a He^+ ion beam, accelerated to 2 MeV with a Van de Graaff accelerator. The range and total number of displacements were estimated to be about 4 μm and 170 displacements/ion, respectively, by using the TRIM code (SRIM-98) with the displacement energies of 20 eV for Al and 50 eV for O. The size of the ion beam was about 2 mm × 2 mm and its current was monitored. A photonic multichannel analyzer, Hamamatsu PMA-11 was used to measure the irradiation time dependence of the luminescence. The temperature of the sample holder was controlled with an electric heater and a thermocouple while another thermocouple was attached to the sample surface to monitor its temperature at a distance close to beam area. In the room temperature irradiation, the temperature rise by beam heating was observed to be within a few degrees as estimated by taking the values of thermal diffusion coefficient, density and heat capacity of alumina. The experimental conditions are summarized in Table 1.

3. Results and discussion

3.1. Irradiation time dependence

The observed luminescence spectra are composed of a number of luminescence bands, namely 330 nm, 420 nm and others, as already reported [2], of which the luminescence bands centered at 330 and 420 nm can be attributed to the F^+ and F^0 centers, respectively [4].

Figs. 1 and 2 show typical results of the measurement of α-alumina and sapphire, respectively, in which the luminescence intensity is given in cps for a fixed window of 0.75 nm. It is shown that the luminescence intensities at 330 and 420 nm show an irradiation history in which the intensities increase with irradiation time and reach the maximum ones, and that the intensities decrease at the longer irradiation in some cases. By annealing the irradiated specimens above 1073 K for some 10 min, such an irradiation history has been observed repeatedly [2]. This fact suggests that a considerable part of the luminescence comes from the defects accumulated by irradiation and that the intensities at these bands may reflect the amounts of the F^+ and F^0 centers. A similar irradiation history for the ion beam induced luminescence of sapphire has been reported by Al Ghamdi and Townsend [5], who have observed non-zero values for the luminescence intensity at a low fluence and have suggested that the luminescence is from intrinsic color centers activated by excited electrons. In the present case, on the other hand, nearly zero values have been observed with annealed specimens, suggesting that the luminescence originates from the F^+ and F^0 centers accumulated by irradiation.

In the case of electron irradiated sapphire [6], it has been found that the peak height ratio between the F^+ and F^0 is strongly dependent on the dose rate and that the F^+ luminescence increases with the dose rate while the F^0 luminescence decreases. Since the F^+ is hardly accumulated in the electron irradiated sapphire, this dose rate effect has been explained by considering the conversion of the F^0 to the F^+; the F^0 is excited by electron and hole capture and this excited F^0 is ionized by hole capture giving the F^+ [6]. As already reported [2], on contrast, such a dose rate effect was hardly observed in the studied region, possibly due

Table 1
Experimental conditions and steady state luminescence intensity

Specimen	Irradiation condition			Steady state luminescence intensity (cps) at	
	Projectile	Temperature[a] (K)	Beam current (nA/cm^2)	330 nm	420 nm
α-Alumina	2 MeV He^+	298	63	⩾6000	≈2000
		298	81	⩾6000	≈1000
		423	164	⩾14000	≈2800
		423	238	≈8000	≈2000
		448	67	≈3500	≈600
		473	81	>4000	≈100
		473	131	≈13000	≈200
		523	77	≈4500	≈200
Sapphire	2 MeV He^+	298	66	≈8000	≈2800
		300	133	⩾6000	≈1700
		323	119	≈11000	≈2000
		373	514	≈25000	≈6000
		423	133	≈17000	≈6000
		448	75	⩾1400	≈500
		523	140	≈7000	≈300

[a] Temperature of the thermocouple attached to the sample surface to monitor its temperature.