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Spectroscopic studies of swift heavy ion irradiated nanophase mullite

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Abstract

Photoluminescence (PL) studies of 100 MeV swift Ag^{8+} ion bombarded combustion synthesized nanophase mullite has been studied at room temperature (RT) and the results are reported here. A pair of PL bands, one broad band centres at ~ 550 nm and another sharp one at ~ 690 nm are observed with excitation by a 442 nm laser beam. However, when the sample is excited with 326 nm laser beam, three bands with peaks at ~ 460 , 550 and a well resolved one with peak at 760 nm are observed. It is observed that the PL intensity increases up to 5×10^{11} ions/cm² and thereafter it decreases with increase of ion fluence. The pristine as well as Ag^{8+} ion irradiated mullites are characterized by infrared spectroscopy (IR) and X-ray diffraction (XRD) techniques. The decrease in PL intensity is attributed to Al–O and Si–O bonds present as the surface getting amorphized. The effects of Ag^{8+} irradiation are compared to those obtained with Ni^{8+} ions and the results are discussed.

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1. Introduction

Radiation effects in inorganic insulators have been a subject of rapidly growing interest in the past few years. A large field of potential applications exists for these materials and the development of various new techniques is directly related to the progress in this field. Swift heavy ions (SHI) are very useful for modifying the properties of films, foils and surface of bulk solids. Ions penetrate deep into the material and produce a long and narrow disordered zone along their trajectory.

In the present paper, we report the PL, IR and XRD analysis of 100 MeV Ag^{8+} irradiation effects in combustion synthesized mullite bombarded, for fluences in the range 1×10^{11} – 5×10^{13} ions/cm². Mullite is an aluminum silicate

($3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$) crystallizes in orthorhombic. This is a promising, material for high temperature applications because of its low thermal conductivity. The silicate group of this mineral used in the production of non-fusion cast tank blocks in the manufacture of spark plugs and as a mineral specimens [1].

2. Experimental

Nanocrystallized mullite particles were synthesized by combustion technique using aluminum nitrate and dimethyl hydrazine dissolved in a minimum quantity of double distilled water in a cylindrical Pyrex dish of ≈ 300 ml capacity. To this, a silica fumes was added and dispersed well using a magnetic stirrer. The dish containing the heterogeneous mixture was introduced into a muffle furnace maintained at 500 ± 10 °C. Initially, the solution boils and undergoes dehydration followed by decomposition with evaporation of large amount of gasses. The mixture then froze and swells, forming a foam which ruptures with

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a flame and glows to incandescent with a flame temperature of 1200 °C. The final product is voluminous and weakly crystalline. The whole process lasts less than 5 min. The crystallinity and particle size of final product was confirmed by XRD and scanning electron microscope [2]. The particle size is observed to be in the range of 100 nm. Pellets of 1 mm thickness and 6 mm diameter were prepared using pestle and mortar and poly vinyl alcohol as binder and by applying a pressure of 6 ton per ram of 6 mm diameter using an home made pelletizer.

One sample was reserved as pristine for comparison with the irradiated ones. The irradiation was carried out on two identical samples. The mullite pellets were mounted on a vacuum shielded vertical sliding ladder having four rectangular faces. The samples are irradiated in Material Science chamber of the Pelletron accelerator at NSC under high vacuum (6×10^{-6} Torr) with 100 MeV Ag^{8+} ions with appropriate beam current of ~ 2 pna.

Photoluminescence measurements of Ag^+ ion irradiated mullites were carried out using an excitation source – Kimmon He–Cd laser (442 or 326 nm) with a power of 32 or 120 mW. CCD detector was used to record PL spectra in the 400–1200 nm range. Infrared spectra of pristine and ion bombarded samples were recorded in the range 500–4000 cm^{-1} using Pye Unicam SP3-300 spectrophotometer using nuzol solvent. The XRD study was carried out using a Rich–Siefert, XRD-3000-TT setup with ^6Cu $\text{K}\alpha$ radiation.

3. Results and discussion

Photoluminescence of combustion-mullite bombarded with 100 MeV swift Ag^{8+} ions at fluence in the range 1×10^{11} – 5×10^{13} ions/ cm^2 under excitation by 442 and 326 nm laser were recorded and the obtained results are presented in Figs. 1 and 2, respectively. Two PL bands, one centred at ~ 550 nm and a sharp peak at ~ 690 nm are observed with excitation at 442 nm. However, when the sample is excited with 326 nm laser beam, two additional PL bands centred at ~ 460 and 760 nm along with

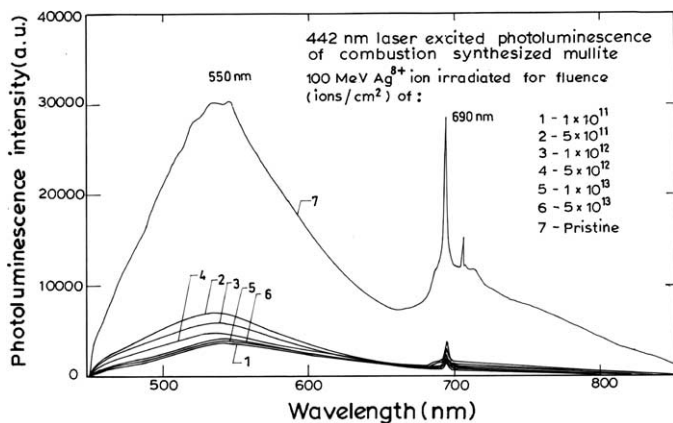


Fig. 1. Photoluminescence spectra ($\lambda_{\text{excitation}}$ 442 nm) of 100 MeV Ag^{8+} ion irradiated combustion synthesized mullite.

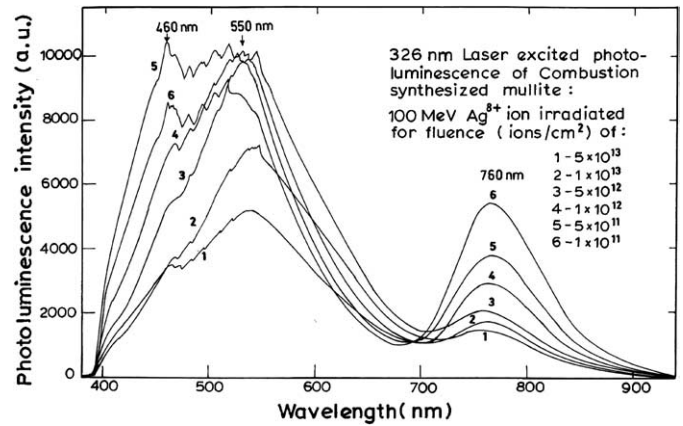


Fig. 2. Photoluminescence spectra ($\lambda_{\text{excitation}}$ 326 nm) of 100 MeV Ag^{8+} ion irradiated combustion synthesized mullite.

a broad and intense PL peak at 550 nm are observed. The 690 nm band is absent in the later case. It is observed that the intensity of these peaks increases up to a fluence of 5×10^{11} ions/ cm^2 and thereafter decreases with increase of Ag^{8+} ion fluence as can be seen from Figs. 1 and 2. The variation of PL intensity due to 442 and 326 nm excitation for various Ag^{8+} ion irradiated mullite are shown in Figs. 3 and 4, respectively. The increase in PL intensity up to 5×10^{11} ions/ cm^2 is attributed to creation of additional defect centers responsible for PL during the bombardment process [3]. The decrease in PL intensity with further increase of ion fluence ($> 1 \times 10^{11}$ ions/ cm^2) implies some changes in the energy levels of the crystal resulting from the perturbation of the luminescent sites and from transient defect states formed during the bombardment process [4]. At fluence larger than 5×10^{11} ions/ cm^2 , the PL intensity becomes dose independent as a result of more damage occurred already in the sample. The sample in this case is supposed to be amorphized as a result of cascade quenching with swift heavy ion irradiation [5–7]. The susceptibility of amorphization may be measured with irradiation either at fixed temperature or at critical temperature. However, in

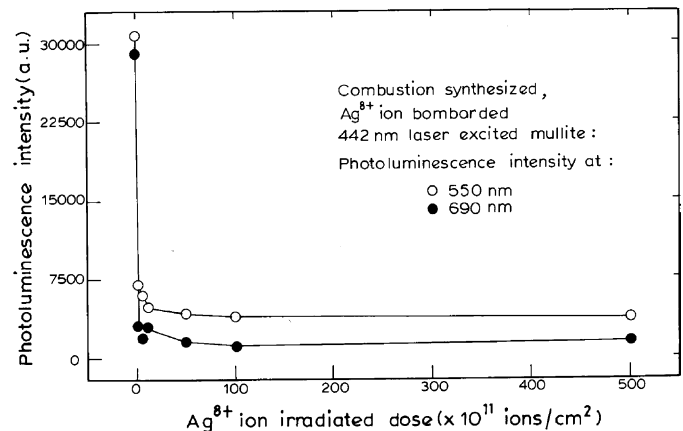


Fig. 3. Variation of PL intensity due to 442 nm laser beam excitation as a function of Ag^{8+} ion fluence in combustion synthesized mullite.

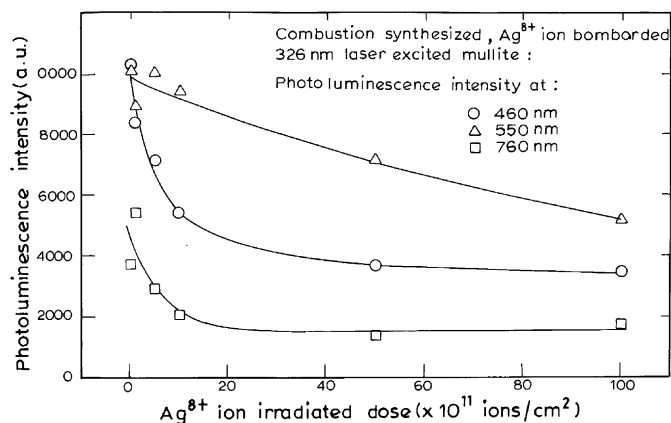


Fig. 4. Variation of PL intensity due to 326 nm laser beam excitation as a function of Ag⁸⁺ ion fluence in combustion synthesized mullite.

Table 1
Infrared absorption data of pristine and Ag⁸⁺ ion irradiated combustion synthesized mullite

Absorption (cm ⁻¹)	Pristine sample	Irradiated sample
467–472	Si–O (ν ₄)	Si–O (ν ₄)
566–571	Si–O–Al	Si–O–Al
747–829	Al–O	–
2376–2929	Si–O (2ν ₃)	Si–O (2ν ₃)
3441–3461	–OH (ν ₁ , ν ₃)	–OH (ν ₁ , ν ₃)

the case of swift 100 MeV Ni⁸⁺ ion bombarded mullite in the dose range 1×10^{11} – 5×10^{13} ions/cm² with excitation by 442 nm laser beam, a broad emission band with peak at ~ 557 nm besides a sharp emission band with peak at ~ 705 nm was observed. The PL peaks in Ag⁸⁺ ion bombarded mullite are found to be shifted towards lower wavelength side with respect to those in Ni⁸⁺ ion irradiated samples [8].

The surface amorphization is confirmed by XRD measurements. They show that the material is crystalline up to 5×10^{11} ions/cm² and thereafter, the decrease in peak intensity indicates the amorphization of the sample. Atomic force microscopy may throw some light on the formation of amorphization.

Infrared absorption spectra of pristine and Ag⁸⁺ ion irradiated mullite with fluences of 1×10^{11} – 5×10^{13} ions/cm² have been recorded and the obtained data are given in Table 1. They clearly indicate to the characteristic absorption bands of silicates and water before irradiation of the samples [8]. It is further observed that the sharpness of the peaks increases in ion irradiated samples. This may

be attributed to close packing units of tetrahedral SiO₄ and/or octahedral AlO₆ [8]. The decrease in PL intensity at 741–827 nm and 460 nm might be due to destruction of Al–O bonds [9] and, the destruction of these bonds with irradiation may further increase the amorphous nature of the sample. The irradiation is supposed to lead to reducing of packing units because of the energy deposited through electronic loss of swift heavy ions and of the formation of ion induced defects leading to non radiative recombination centers. These two processes are simultaneous consequences of irradiation and they compete with each other. Consequently, the enhancement or degradation of PL might be due to the balance between these effects [9].

4. Conclusions

PL of Ag⁸⁺ swift ion bombarded combustion synthesized mullite give rise to 550 and 690 nm bands and also 460, 550 and 760 nm bands when the sample is excited by 442 and 326 nm laser light, respectively. The increase in PL intensity up to 5×10^{11} ions/cm² is attributed to creation of new traps with bombardment process. The decrease in PL, XRD and IR intensity with increase of ion fluence is attributed to the amorphization of the material due to destruction of Al–O bonds.

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