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Gamma-irradiation effects on Ce-doped YAG crystals grown by Cz and TGT method

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Abstract

Gamma-rays radiation effects on Ce:YAG crystals grown by Czochralski (Cz) and temperature gradient techniques (TGT) have been studied by means of optical absorption and luminescence spectra. Valence of Ce^{3+} ion changes during the gamma-ray irradiation process and this result indicates Ce^{4+} ion may exist in both Cz-Ce:YAG and TGT-Ce:YAG crystals. Thermally stimulated luminescence measurements reveal intense thermoluminescence peaks in gamma-irradiated Ce:YAG crystals and trap parameters were calculated by general-order kinetics expression.

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1. Introduction

Cerium activated yttrium aluminum garnet crystal ($Ce:Y_3Al_5O_{12}$), which show extremely high mechanical and chemical stability, high light yield and short luminescence decay times, have proved to be an excellent scintillator. The peak emission due to the allowed 5d–4f transition of Ce^{3+} ions peaking at 500–550 nm fits well to the sensitivity spectrum of Si photodiodes [1–5]. Hence, Ce:YAG has its potential applications in various fields, such as the detection of light charged particles and high-count-rate applications [6,7].

The stability of Ce:YAG crystals in a radiation environment are one of the most important characteristics for their application in precise electromagnetic calorimetric detectors and in space. One hand, a change of the balance between the fractions under irradiation, especially valence changes of the Ce^{3+} ions, could induce a change of the luminescent centers concentration under irradiation. On the other hand, color centers and impurity defects in the crystals have great influence on the energy transfer efficiency from the host lattice to Ce^{3+} ions [8]. In addition, another factor may affect scintillation parameters, particular the scintillation light yield [9], is the afterglow phenomenon. It has been shown that light pulse of Ce:YAG produced by gamma-ray consists of two components with the decay time constant of 88 ns and 302 ns rather than the single component of 25 ns expected from Ce^{3+} ions [1]. Slow components in the scintillation decay, possibly related to trap levels could be detected by thermally stimulated luminescence (TSL). It has been concluded that the existence of intrinsic, metastable trapping sites that capture and release electrons in oxyorthosilcate contribute to afterglow [10,11]. In the present work we examine the radiation-induced valence changes of Ce^{3+} ions and trapping sites in Ce:YAG by means of optical spectrum and TSL. Our attention is

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focused on the glow peak observed just above room temperature, which is closely linked to the afterglow phenomenon.

2. Experimental

Ce-doped YAG crystals were grown by temperature gradient techniques (TGT) under strong reducing atmosphere produced by the graphite heating element. A detailed description of the TGT furnace is given by Xu et al. [12]. The highly pure oxide powders Y_2O_3 ($\geqslant$99.999%), CeO_2 ($\geqslant$99.99%) and Al_2O_3 ($\geqslant$99.95%) were used and final chemical formula is $(Y_{0.992}Ce_{0.008})_3Al_5O_{12}$. The seed is [1 1 1] orientation and the whole growth atmosphere was performed under high-purity Ar gas. After growth process, Ce:YAG crystal was annealed in situ with the rate of (30–50) °C/h. More detail of the growth process can be found in [13]. For comparison, Ce:YAG crystals with the same concentration also were grown by Cz method with radio frequency (RF) induction heating in iridium crucibles. The raw materials were the same as those used in the TGT. A high-purity nitrogen atmosphere was used for the crystal growth. The heat shield was made of oxide such as ZrO_2 and Al_2O_3, so weak oxidizing atmosphere was maintained during the growth process. The growth apparatus was described elsewhere [14].

Samples of approximate dimensions $10 \times 10 \times 0.68$ mm thickness for spectroscopic measurements were cut out of the boules and surfaces perpendicular to (1 1 1)-growth axis were polished on both sides. The samples were performed twice irradiations by means of a ^{60}Co gamma source (average gamma energy 1.25 MeV) up to an absorbed dose of 2×10^4 and 2×10^5 Gy with dose rate of about 170 Gy/min at room temperature. The absorption and emission spectra were recorded by means of a V-570 UV/VIS/NIR spectrophotometer and FP-6500/6600 fluorescence spectrophotometer. The light sources were a deuterium lamp (190–350 nm) and a halogen lamp (340–1200 nm), and the spectral resolution was 1 nm. To make accurate comparison, three times irradiations performed on the same piece of sample. The thermally stimulated luminescence (TSL) glow curves measurements after gamma-irradiation were made over the temperature range from RT to 700 K by FJ-427A TL spectrometer with heating rate of 2 °C/s. Values of induced additional absorption (AA) due to the irradiation or thermal processing were calculated from the formula:

$$\Delta k = \frac{1}{d} \ln \frac{T_1}{T_2}$$

where k is the absorption, d is the sample thickness and T_1 and T_2 are the transmissions of the sample obtained before and after gamma-irradiation or thermal treatment, respectively.

3. Results and discussion

Fig. 1 shows absorption spectrum of Ce:YAG crystals grown by Cz method before gamma-irradiation and AA spectrum after twice gamma-irradiation, respectively. In YAG, Ce^{3+} substitutes for Y^{3+} at sites of D_2 symmetry and the excited 5d states is split into five levels. Three absorption peaks at about 227, 340 and 460 nm are due to the $4f^1 \rightarrow 5d^1$ transition in the Ce^{3+} ions of Ce:YAG crystals [15,16]. It can be seen from the AA spectra that the absorption coefficient of the Ce^{3+} ions absorption bands increases in the case of the first (2×10^4 Gy) and second dose (2×10^5 Gy). This result indicates that the Ce^{3+} ions concentration increases as a result of valence variation of Ce^{4+} ions after gamma-irradiation. During the growth process, Ce^{4+} ions in raw material should pass to trivalent state. It is not excluded, however, that some of these ions remain in the Ce^{4+} state. Presence of free electrons emitted during the interaction of gamma-rays with the Ce:YAG crystal, (e.g. by Compton effect), leads to free electrons trapping by Ce^{4+} ion and formed Ce^{3+} ions. Therefore, the increase of the AA values is due to the reaction of $Ce^{4+} \rightarrow Ce^{3+}$.

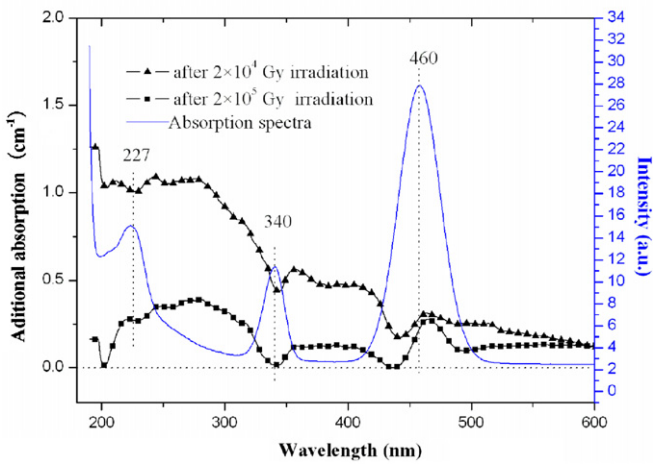

Fig. 1. Absorption and AA spectra of gamma-irradiated Cz-Ce:YAG with different dose.

Fig. 2 presents the emission spectra of as grown and gamma-irradiated Ce:YAG crystals grown by Cz method excited at 230 nm. The fluorescence emission of Ce:YAG originates from a transition from one or more of the 5d levels (often only the lowest level) to the ^{2}F ground state of the Ce^{3+} ion. Therefore, changes in fluorescence intensity after gamma-irradiation are closely related to the variations of Ce^{3+} ions concentration. Similarity in the changes of AA value, the changes of emission intensity clearly confirms the recharging process of the Ce^{3+} ions.

Fig. 3 presents absorption spectrum of Ce:YAG crystals grown by TGT method before gamma-irradiation and AA spectrum after twice gamma-irradiation, respectively. The AA values changes of the 227, 340 and 460 nm absorption bands after gamma-irradiation is similar to that of the Cz-Ce:YAG. However, it should be noticed that the magnitude of the AA values changes is much large than that of the Cz-Ce:YAG, indicating a large amount of Ce^{4+} ions exist in TGT-Ce:YAG crystals. The TGT-Ce:YAG crystals