

Studies of radiation defects in cerium, europium and terbium activated oxyfluoride glasses and glass ceramics



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ABSTRACT

Terbium, cerium and europium activated oxyfluoride glasses and glass ceramics have been studied by thermally stimulated luminescence (TSL) and optical absorption techniques after the X-ray irradiation. A creation of colour centres in oxyfluoride glass matrix and TSL peaks depending on the activator type were observed. LaF₃ and rare earth activators were analysed by SEM–EDS.

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1. Introduction

Glass ceramics scintillators' advantages over single crystals are better uniformity and simplest production [1–3]. It would be a good material for applications where cheap and simple fabrication is more important than very high intensity or very short decay times.

First discovery of rare earth activator (Eu³⁺) partitioning into LaF₃ crystals in the oxyfluoride glass–ceramic was published in a paper [4].

Many studies have been done of rare earth activator incorporation into LaF₃ crystals and its spectroscopy: europium [5], thulium [6], cerium and dysprosium [7], terbium [8] and other.

LaF₃ has relatively low phonon energies (300 cm⁻¹) [9,10], that reduce the quenching of the excited-states of lanthanide ions by lattice vibrations. LaF₃ shell does not change the luminescent properties of the luminescent ion [10]. Therefore, LaF₃ is a good host for rare-earth ions.

In our previous researches [11,12] we have studied terbium, cerium and europium activated oxyfluoride glasses and glass ceramics using various methods: differential thermal analysis, X-ray diffraction, cathodoluminescence and X-ray induced luminescence. X-ray induced luminescence showed that among the studied samples the most intense was a terbium activated glass ceramics sample – the integrated area of the spectral curve was about 10 times smaller than that of etalon sample CsI:Tl. Cathodoluminescence measurements showed that a cerium activated glass

ceramics sample had the fastest decay times – average decay time was approximately 1.7 μs [12].

In this work, we have discussed creation of radiation defects in glass and glass ceramic samples after their X-ray irradiation. Experimental results have been obtained by thermally stimulated luminescence (TSL), optical absorption and scanning electron microscopy dispersive X-ray spectroscopy (SEM–EDS) techniques.

2. Experimental

2.1. Composition of samples

The investigated samples have the following molar composition: 48% SiO₂, 6% Al₂O₃, 24% Li₂O, 20% LaF₃ with different activators. The samples without activator hereinafter will be referred as SALL, those activated with 2% TbF₃ – SALL:Tb, 2% Eu₂O₃ – SALL:Eu and with 2% CeO₂ – SALL:Ce. After a heat treatment at 750–800 °C for 1 h glass ceramics have been formed [11].

2.2. Experimental equipment

Optical absorption spectra at room temperature were measured by the spectrometer Specord 210-2 using the integrating sphere with spectral range 380–1100 nm. The samples were X-ray irradiated at room temperature for 1 h.

Thermally stimulated luminescence spectra were measured by the Andor Shamrock B303-I spectrograph. For TSL measurements the samples were X-ray irradiated at liquid nitrogen temperature and then heated.

SEM (scanning electron microscopy) and EDS (energy dispersive X-ray spectroscopy) results were obtained by the SEM-FIB Tescan

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Lyra equipped with Oxford Aztec spectrometer. Samples for SEM and EDS studies were cut and polished.

3. Results

3.1. Optical absorption for the SALL (non activated) glass and glass ceramics samples

Fig. 1 shows absorption spectra of the SALL sample before and after irradiation with X-rays. As the glass ceramic samples were not transparent, we used integrating sphere, which limits the minimum measurable wavelength to 380 nm. The glass ceramic sample has a broad absorption bands around 550 nm and 750 nm. After X-ray irradiation the glasses and glass ceramics have a higher absorption than before.

3.2. Activated SALL samples

Figs. 2 and 3 show absorption spectra of the glass and glass ceramics samples before and after X-ray irradiation, respectively. Generally the optical density of the activated glasses and glass ceramics after X-ray irradiation increases. All the activated glass ceramics (SALL:Eu, SALL:Ce, SALL:Tb) have higher absorption than the non activated glass ceramics (SALL) and glass samples. The SALL:Tb glass ceramics has the highest absorption among the investigated samples. We did not observe any new absorption bands in the SALL:Tb spectra, that would indicate to the formation of the same defects as in other samples.

Optical density of X-ray irradiated samples decreases with time after a heat treatment and defect concentration decreases. Absorption reduction after the heating is faster for the SALL:Tb than that of the SALL:Eu and the SALL:Ce glass ceramic samples.

Summarizing the results of the induced optical absorption, we have observed that very broad induced absorption bands could be created by the X-ray irradiation and that they are not very dependent from the activator type or from the presence of crystallites.

3.3. Thermally stimulated luminescence measurements

3.3.1. TSL for the SALL glass and glass for the SALL (non activated) samples

We performed measurements of TSL curves in the temperature range 77–350 K. Figs. 4 and 5 show that the SALL glass and the

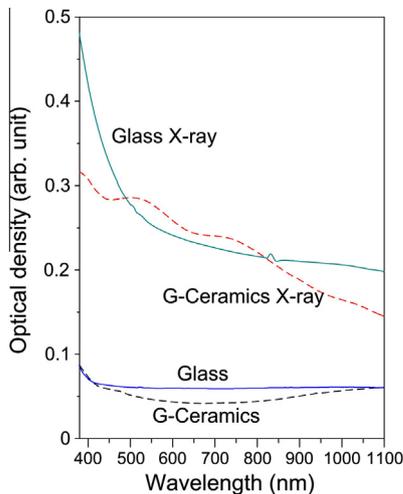


Fig. 1. Optical absorption spectra before and after irradiation with X-rays.

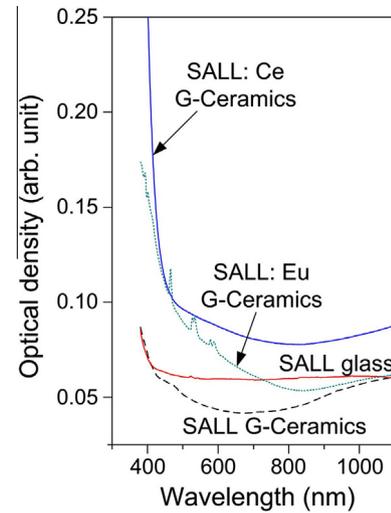


Fig. 2. Absorption spectra of the glass and glass ceramics samples before X-ray irradiation.

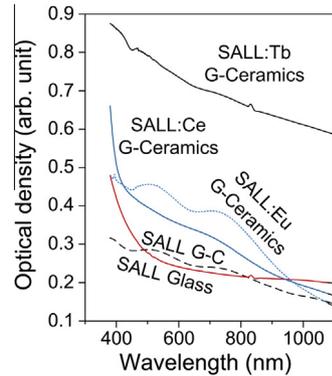


Fig. 3. Absorption spectra of the glass and glass ceramics samples after X-ray irradiation.

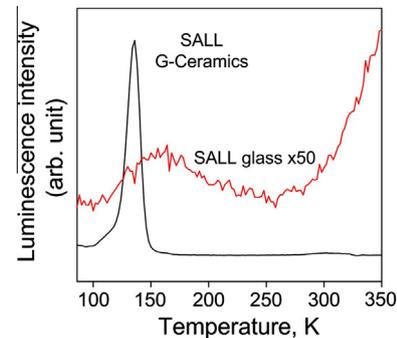


Fig. 4. TSL curves of the SALL glass and the SALL glass ceramic, measured at the luminescence wavelength 734 nm.

SALL glass ceramic TSL curves and spectra intensity of glass ceramic samples curves are about 50 times more intense than the intensities of the glass samples.

3.3.2. TSL for the SALL and activated glass ceramics

To show TSL curves and TSL luminescence spectra we selected one wavelength – 700 nm (Fig. 6) and one temperature – 140 K (Fig. 7) for all samples.

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