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JOURNAL OF RARE EARTHS
JOURNAL OF RARE EARTHS, Vol. 35, No. 8, Aug. 2017, P. 787
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Luminescent properties of Tb^{3+} doped high density borogermanate scintillating glasses

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Received 22 October 2016; revised 3 May 2017

Abstract: Tb^{3+}-doped high density borogermanate glasses were prepared by melt-quenching method. Their physical and luminescent properties including differential thermal analysis (DTA), density, transmittance spectra, photoluminescence, and X-ray excited luminescence spectra were investigated. The densities of the glasses doped with Tb^{3+} were in the range from 5.690 to 6.086 g/cm^3. Under UV and X-ray excitations, the glasses showed intense green emissions. The lifetimes of Tb^{3+} doped borogermanate glasses were in the range from 1.597 to 1.869 ms. The results indicated that Tb^{3+} doped borogermanate glasses could be scintillator candidate used in X-ray detection application.

Keywords: luminescence; Tb^{3+}; scintillating glass; high density; rare earths

In recent years, as one of the most attractive hosts for active optical devices and optical amplifiers, rare earth ion doped transparent germanate glasses have attracted considerable attention due to the advantages in high rare earth ion solubility, low phonon energy, excellent chemical and thermal stability. Yb^{3+} ion[1,2], Dy^{3+} ion[3], Eu^{3+} ion[4,5], and Tm^{3+} ion[6–8] have been doped in germanate glasses to reveal their luminescence properties. As we know, heavy germanium oxide is an important component for glass with high density. Thus, rare-earth ions doped germanate glasses show great promise as candidates for X-ray scintillating materials. Meanwhile, among the scintillating performance required in practice, such as X-ray computer tomography (X-CT), glass density close to or exceeding 6.0 g/cm^3 is particularly important because a high glass density increases the X-ray absorption cross-section, which results in significant increment of the image's signal-to-noise ratio[9,10].

Recently, many researchers reported high density glasses by the addition of rare-earth oxides RE_2O_3 (RE=Y, La, Gd and Lu) to glass scintillators with suitable light yield[11–14] and many silicate, borosilicate, germanate, borogermanate and tellurite glasses containing high Gd_2O_3 contents have been used for glass scintillators[10–15]. Among these RE_2O_3 oxides, Lu^{3+} is optically inert, which is suitabe for matrix material and borogermanate glass with a high Lu_2O_3 content was rarely studied. Besides, dense Lu_2O_3 is a favorite raw material to elevate the density of the glass. Regarding to the borogermanate system, Eu^{3+}-, Tb^{3+}-activated borogermanate scintillating glasses with a density of 5.6–5.8 g/cm^3 have been proposed to detect high energy X-rays[10,15,16]. Under X-ray excitation, Tb^{3+}-doped borogermanate glasses show strong greenish fluorescence which could be applied in X-ray detection for slow event, especially. To our knowledge, there are much work on rare earth ions doped borogermanate or germanate glass. However, Tb^{3+}-doped high density borogermanate scintillating glasses with a high Lu_2O_3 content are rarely studied and the strongest emission of Tb^{3+} is around 540 nm, which is convenient for direct coupling with silicon detector. Therefore, it is useful for the X-ray scintillation detection. In this paper, the physical and luminescent properties of Tb^{3+}-doped high density borogermanate scintillating glasses containing dense Lu_2O_3 were investigated in detail.

1 Experimental

Tb^{3+} doped borogermanate glasses samples with the composition of $20B_2O_3$-$40GeO_2$-$20Lu_2O_3$-$5La_2O_3$-$(15–x)BaF_2$-$xTbF_3$ (x=1, 2, 4, 6, 8, 10, 12 mol.%) were prepared by melt-quenching method. All raw materials were derived from H_3BO_3 (A.R.), GeO_2 (99.999%), Lu_2O_3 (99.99%), La_2O_3 (99.99%), BaF_2 (99.9%) and TbF_3 (99.99%). About 20 g weighed raw materials were mixed in an agate mortar and melted in a covered high purity alumina crucible at 1500 ºC for 1 h in an electric furnace in the air atmosphere so that a homogeneously mixed melt was obtained. Then, the melt was poured into a preheated stainless steel mould, subsequently annealed for 2 h below the glass temperature

Foundation item: Project supported by the National Natural Science Foundation of China (11375166, 51372236)

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DOI: 10.1016/S1002-0721(17)60977-3

and then slowly cooled down to room temperature. The glass samples were polished for optical measurements with a thickness of 1.80±0.02 mm.

DTA measurements were carried out by a Netzsch DTA 404PC at a heating rate of 10 K/min. The glass densities were measured by Archimedes' method using distilled water as an immersion liquid. Transmittance spectra were obtained with a Shimadzu UV-3600 spectrophotometer in the range of 200–710 nm. Photoluminescence (PL) spectra and luminescence decay curves were recorded on a Jobin-Yvon Fluorolog3 fluorescence spectrophotometer using Xe lamp as an excitation source. The X-ray excited luminescence (XEL) spectra were measured using an X-ray tube (Copper target, 70 kV, 1.5 mA) and an Ocean Optical QE65000 spectrometer based on charge coupled device (CCD) detector. The spectral sensitivity of the detection system as a function of wavelength was corrected with a standard radon lamp. All the measurements were performed at room temperature.

2 Results and discussion

Fig. 1 shows the DTA curve of 10 mol.% Tb^{3+} doped borogermanate glass. The glass transition temperature (T_g) is around 571 ºC. Two exothermic peaks are observed at 755 and 956 ºC. The former peak could be the precipitation of BaF_2 and the latter is due to the crystallization of the glass. There are few reports on the thermal analysis of germanate scintillation glasses. Meanwhile, the T_x-T_g value of glass is higher than other glasses[17,18], which indicates that the present glass has better thermal stability.

The densities of the borogermanate glasses doped with different Tb^{3+} concentrations (1, 2, 4, 6, 8, 10, 12 mol.%) are shown in Fig. 2. The density of the glass decreases slightly from 6.086 to 5.690 g/cm^3 with increasing the concentration of Tb^{3+}. This result may be attributed to the content of heavy Lu_2O_3 slightly decreased while that of light TbF_3 increased. It is worth noting that all the densities of the Tb^{3+} doped borogermanate glasses are around 6.0 g/cm^3. This indicates that the Tb^{3+} doped borogermanate glasses could be potential glass scintillator candidates for practical applications.

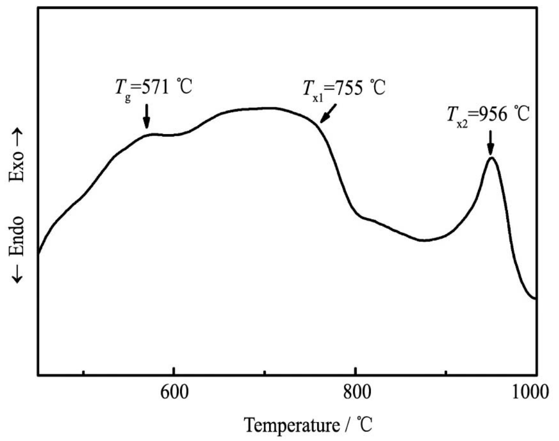

Fig. 1 DTA curve of 10 mol.% Tb^{3+}-doped borogermanate glass

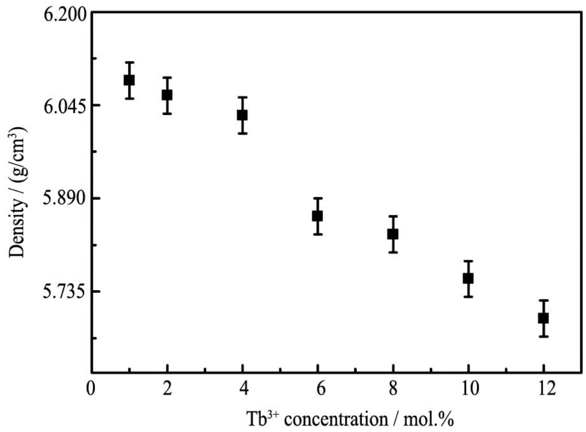

Fig. 2 Densities of borogermanate glasses doped with different Tb^{3+} concentrations

Fig. 3 shows the transmittance spectra of Tb^{3+}-doped borogermanate glass with a thickness of 1.80±0.02 mm. It is obvious that the transmittance of the glasses decreases with increasing Tb^{3+} concentration, but the overall has good transmittance in the visible spectrum region and the UV cut-off wavelength is around 265 nm. It indicates that the obtained glasses may be used as scintillator material. The absorption bands of Tb^{3+} ions with peaks at 317, 340, 352, 369, 378, 487 nm could be observed in the spectra as well, which are due to the transitions from the ground state $^7F_6$ to the $^5H_7$ and higher ^5D states of Tb^{3+}.

Under UV light, Tb^{3+}-doped borogermanate glasses show greenish fluorescence. Fig. 4 shows the emission spectra of the borogermanate glasses doped with 1, 2, 4, 6, 8, 10 and 12 mol.% Tb^{3+} under 376 nm light excitation. The emission spectra contain four emission bands peaked at 489, 543, 589, and 622 nm, which are attributed to the transitions $^5D_4\rightarrow{}^7F_6$, $^5D_4\rightarrow{}^7F_5$, $^5D_4\rightarrow{}^7F_4$, and $^5D_4\rightarrow{}^7F_3$ of Tb^{3+}, respectively. Besides, there is a shoulder peak at about 549 nm in Fig. 4. It may be caused by asymmetry of the glass matrix results in splitting of the energy level[19]. The emission at 543 nm is the strongest

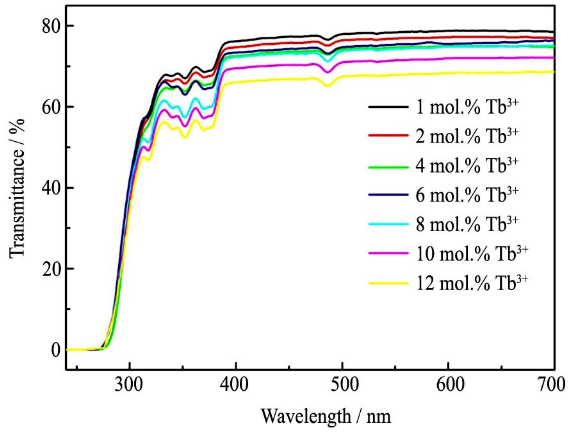

Fig. 3 Transmittance spectra of Tb^{3+} doped borogermanate glass with a thickness of 1.80±0.02 mm

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