

Optical properties of Ce³⁺ doped fluorophosphates scintillation glasses



Yongxin Yao^a, Liwan Liu^{b,c}, Yu Zhang^b, Danping Chen^{a,b,*}, Yongzheng Fang^{a,*}, Guoying Zhao^a

^a School of Materials Science and Engineering, Shanghai Institute of Technology, Shanghai 201418, PR China

^b Key Laboratory of Materials for High Power Laser, Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, Shanghai 201800, PR China

^c Department of Physics, Shanghai University, Shanghai 200444, PR China

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ABSTRACT

Fluorophosphates (P₂O₅–BaO–BaF₂–Al₂O₃–Gd₂O₃–Ce₂O₃) glasses with different Gd₂O₃ and BaF₂ concentrations have been prepared by a melt quenching method. The effect of Gd₂O₃ and BaF₂ on the glass performance including the density, absorption as well as luminescence properties under both ultraviolet (UV) and X-ray excitation was studied systematically. Energy transfer from Gd³⁺ to Ce³⁺ plays an important role in the scintillation mechanism of these glasses and the optimum concentration of Gd₂O₃ is found to be approximately 3 mol%. The highest integrated light emission intensity of these glass samples excited by X-ray is 25% of BGO and the decay time constants are in the range of 25–35 ns, much shorter than the 300 ns decay time of BGO. Meanwhile, replacing lighter compound BaO with the BaF₂ can increase the density of the glasses and also improve the light yield.

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1. Introduction

Scintillating glasses can be used for the detection of X-rays, γ -rays and neutrons [1–3]. They are attractive scintillating material due to their advantages including low production cost, easy shaping of elements, possibility to incorporate activator ions at high concentrations and in the ease of manufacture in different sizes and shapes with respect to single crystal materials [4–9]. Many studies have been devoted to researching on borosilicate scintillating glasses, but the larger problem of these glass are low light yield and radiation-induced photo darkening problems. However, phosphate glasses are as regarded as suitable host for scintillator mainly because the structure of phosphate glass has a strong dispersing ability to rare-earth ions, high luminescence efficiency of rare earth ions, low darkening effect, then, there is low melting temperature [10,11]. In particular, the studies on phosphate scintillating glasses doped with rare earth ions have been reported by Nitsch et al. [12–15], while most of them contain alkali metal ions and they can result in large numbers of non-bridge oxygen and defects, which is not conducive to the scintillation light outputs.

Compared with the other glasses, oxyfluoride glasses can provide a desirable low phonon energy environment of fluoride for

activator ions, which results in an increased radiative emission rate of the incorporated RE³⁺ ions. At the same time, they can maintain the advantages of an oxide glass, such as high mechanical strength chemical durability and thermal stability [16]. In this paper, we prepared Ce³⁺ ions doped transparent fluorophosphates glasses containing Gd₂O₃ and BaF₂. The effects of Gd₂O₃ and BaF₂ on the glass performance, including the density, absorption as well as luminescent properties under both ultraviolet (UV) and X-ray excitation were investigated systematically.

2. Experimental

Glasses were prepared from high purity compounds (NH₄H₂PO₄, Al₂O₃, BaO, BaF₂, Gd₂O₃ and Ce₂O₃, 99.99%). The molar compositions of glass samples are listed in Table 1. Powders of these compounds taken into appropriate amounts were thoroughly mixed in an agate mortar and afterwards were melted in an alumina crucible within the temperature range 1200–1250 °C. The samples were subsequently annealed at 500 °C in another furnace and cooled to ambient temperature at the rate of about 1 °C/min. They were cut and polished with the dimension of 10 × 10 × 1 mm to achieve good optical quality.

Differential scanning calorimetry (DSC) measurements were done in a Netzsch calorimeter model STA Jupiter 449F3 in order to determine the glass transition temperatures (T_g). The emission and excitation spectra were measured by a JASCO FP-6500 spectrometer. Photoluminescence (PL) decay times were determined by time-correlated single photon counting on an Edinburgh

* Corresponding authors at: School of Materials Science and Engineering, Shanghai Institute of Technology, Shanghai 201418, PR China (D. Chen).

E-mail addresses: d-chen@mail.siom.ac.cn (D. Chen), fy21003@sina.com (Y. Fang).

Table 1
Composition, densities and transition temperature of glass samples.

Sample	Molar compositions	Density (g/cm ³)	T _g (°C)
G0	55.5P ₂ O ₅ –19.5BaO–15BaF ₂ –7Al ₂ O ₃ –3Gd ₂ O ₃	3.49	527
GG1	55.5P ₂ O ₅ –19.5BaO–15BaF ₂ –8Al ₂ O ₃ –1Gd ₂ O ₃ –1Ce ₂ O ₃	3.45	520
GG3	55.5P ₂ O ₅ –19.5BaO–15BaF ₂ –6Al ₂ O ₃ –3Gd ₂ O ₃ –1Ce ₂ O ₃	3.53	531
GG5	55.5P ₂ O ₅ –19.5BaO–15BaF ₂ –4Al ₂ O ₃ –5Gd ₂ O ₃ –1Ce ₂ O ₃	3.61	539
GG7	55.5P ₂ O ₅ –19.5BaO–15BaF ₂ –2Al ₂ O ₃ –7Gd ₂ O ₃ –1Ce ₂ O ₃	3.69	547
GB10	55.5P ₂ O ₅ –24.5BaO–10BaF ₂ –2Al ₂ O ₃ –7Gd ₂ O ₃ –1Ce ₂ O ₃	3.68	522
GB20	55.5P ₂ O ₅ –14.5BaO–20BaF ₂ –2Al ₂ O ₃ –7Gd ₂ O ₃ –1Ce ₂ O ₃	3.70	525

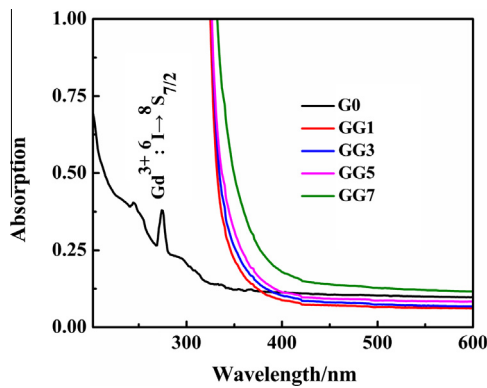


Fig. 1. Absorption spectrum for Ce³⁺ doped glass samples with different Gd₂O₃ concentrations.

Instruments FLSP 920 spectrophotometer equipped with a pulsed H₂ flash lamp nF900. Absorption spectrum in the UV/visible region was recorded on a JASCO V-570 spectrometer. The radio luminescence (RL) spectra were obtained with an X-ray tube (Mo anode, 50 kV, 40 mA). All the measurements were conducted at room temperature.

3. Results and discussions

Densities of all samples are shown in Table 1. It can be seen that the density have an obvious increasing trend with increasing Gd₂O₃ concentration, and maximize is about 3.7 g/cm³. One reason

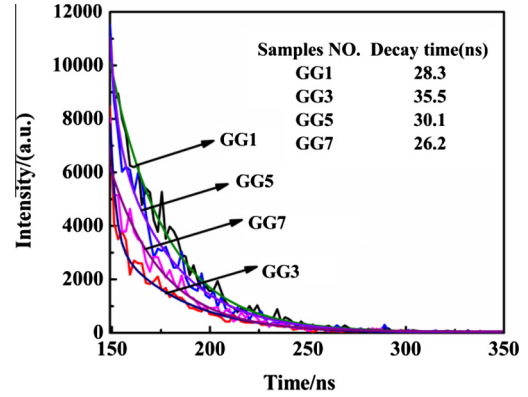


Fig. 3. PL decay curves ($\lambda_{ex} = 315 \text{ nm}$, $\lambda_{em} = 350 \text{ nm}$) of Ce³⁺ doped glass samples with different Gd₂O₃ concentrations.

for the increasing of density is the substitution of the lighter compounds Al₂O₃ by the greater relative atomic mass of Gd₂O₃. In addition, the ionic bond will augment when the concentrations of BaF₂ increase, and easy to form a close-packed structure so that the density increase.

The absorption spectra of glass samples with different Gd₂O₃ concentrations are shown in Fig. 1. In the near UV region and visible, the host glass (G0) without Ce³⁺ doping exhibits good violet transparency. Owing to the transition of Gd³⁺(⁶I → ⁸S_{7/2}) [17], there has a small absorption peak 275 nm in the sample G0 absorption spectrum. However, the Ce³⁺ doped glasses show the onset of a strong absorption at wavelength close to 350 nm, which can be attributed to Ce³⁺:4f → 5d. With increasing Gd₂O₃ concentration, the beginning of the cut-off is shift toward longer wavelength. Nephelantetic effect can be used to explain the red-shift. Owing to the high electron density of Gd³⁺, it can cause a large expansion of the electron cloud, which will influence the position of the absorption bands [18].

The excitation spectrum of 1.0 mol% Ce³⁺ doped glass samples monitoring emission at 340 nm (Fig. 2a). A sharp excitation peaks located at 315 nm, which can be attributable to transitions from the ground state, the ²F_{5/2} level to the excited 5d state in Ce³⁺ [19]. Meanwhile, the excitation spectrum of the sample (G0) is also shown in inset of Fig. 2(a) for reference.

The emission spectra for glass samples with different concentrations of Gd₂O₃ are shown in Fig. 2(b). The transitions from the lowest 5d levels in Ce³⁺ ions to the Ce:²F_{5/2} and Ce:²F_{7/2} levels of the ground configuration is well established [21]. In Fig. 2(b) illustration, the sample (G0) without Ce³⁺ doping is excited with a 275 nm UV light. Although the content of Gd³⁺ ions is increased

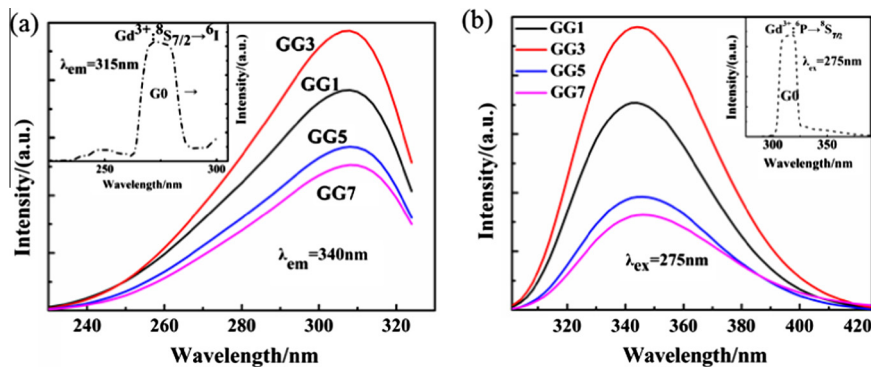


Fig. 2. Excitation ($\lambda_{em} = 340 \text{ nm}$ for figure (a) and emission ($\lambda_{ex} = 275 \text{ nm}$ for figure (b) spectra of glass samples with different Gd₂O₃ concentrations. The inset shows the characteristic excitation and emission of Gd³⁺ ions.

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