

Supplementary Information for

Transparent $\text{YPO}_4\text{:Eu}^{2+}$ glass-ceramic scintillator for high-resolution X-ray imaging

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A. Experiment

(1) Preparation of samples

The samples with the molar composition of $3\text{NaF}-22\text{Al}_2\text{O}_3-43\text{SiO}_2-10\text{YF}_3-18\text{Na}_2\text{CO}_3-4\text{P}_2\text{O}_5-0.5\text{EuF}_3$ were prepared using the conventional melt quenching method. NaF, Na_2CO_3 , P_2O_5 , Al_2O_3 , SiO_2 (A.R., from Sinopharm Chemical Reagent Co., Ltd.), YF_3 , EuF_3 (99.99%, from A&C Rare Earth Materials Center) were used as raw materials, and graphite powder (carbon, 99%, from Shanghai Zhanyun Chemical Co., Ltd.) was used as a reducing agent to reduce Eu^{3+} to Eu^{2+} . Firstly, raw materials with a total mass of 15 g were weighed according to the stoichiometric ratio and then ground in an agate mortar for 1 h. Milled raw materials were placed into corundum crucible and melted at 1500 °C for 1 h. After that, the molten liquid was poured onto a copper plate preheated to 350 °C and pressed by another copper plate. The resulting glasses were annealed in a Muffle furnace at 400 °C for 4 h to release internal stress. These precursor glasses (PG) were heat-treated at 680–700 °C for 3 h to grow NCs. The obtained GC samples are named GC680, GC690, GC695, GC700, respectively. Finally, all samples were cut to suitable shape and optically polished to a thickness of 2 mm for subsequent characterization.

The optimal concentration of EuF_3 is 0.5 mol%, and the specific optimization process is described in Part B in ESI file.

(2) Characterization of samples

The X-ray diffraction (XRD) patterns were obtained using a Rigaku MiniFlex/600 (Tokyo, Japan) X-ray diffractometer with radiation of $\text{CuK}_{\alpha 1}$ ($\lambda = 0.154056$ nm, 45 kV, 15 mA). The transmission spectra in 200–800 nm range were measured by a Hitachi U-3900 ultraviolet-visible (UV-Vis) spectroscopy (Tokyo, Japan). Transmission electron microscopy (TEM), High-resolution TEM (HRTEM), and selected area electron diffraction (SAED) images were tested on a JEOL JEM-2010F transmission electron microscope (Tokyo, Japan). An Edinburgh FS5 spectrofluorometer (Livingston, UK), using a Xenon lamp (150 W) as light source, was used to get photoluminescent excitation (PLE) spectra, photoluminescent (PL) spectra, decay curves and quantum

efficiency. A Zolix OmniFluo960-X-ray spectrometer (Beijing, China) with TUB00154-9I-W06 X-ray tube (Moxtek) was used to obtain XEL spectra. The X-ray imaging photographs were captured by a Canon EOS 600D camera with the same X-ray source. The image acquisition time of samples is 5 s. The density (ρ) was measured according to Archimedes' principle.

(3) The information of $\text{Bi}_4\text{Ge}_3\text{O}_{12}$ crystal

The BGO crystal was purchased from Shanghai Institute of Ceramics, Chinese Academy of Sciences, which is one of best quality suppliers of BGO crystal. The specific parameters were listed in following Table S1.

Table S1 Parameters of BGO crystal from Shanghai Institute of Ceramics, Chinese Academy of Sciences.

Parameter	Value
Density (g/cm^3)	7.13
Radiation length (cm)	1.12
Decay constant (ns)	300
Emission peak (nm)	480
Light output (ph/MeV)	8000-9000
Melting point ($^{\circ}\text{C}$)	1050
Hardness (Mho)	5
Refractive Index	2.15
Size (mm^3)	20*20*2
Hygroscopicity	none
Cleavage	none

B. The optimal doping concentration of EuF_3

The XEL spectra of PG samples doped with $x\%$ EuF_3 (in mol ratio, labeled as PG x) and BGO are displayed in Fig. S1. The integrated XEL intensity of PG x ($x = 0.4, 0.5, 0.6, 0.7$) samples are 32%, 41%, 36%, and 31% of that of BGO, respectively. The result indicates that the optimal doping concentration of EuF_3 is 0.5 mol%.

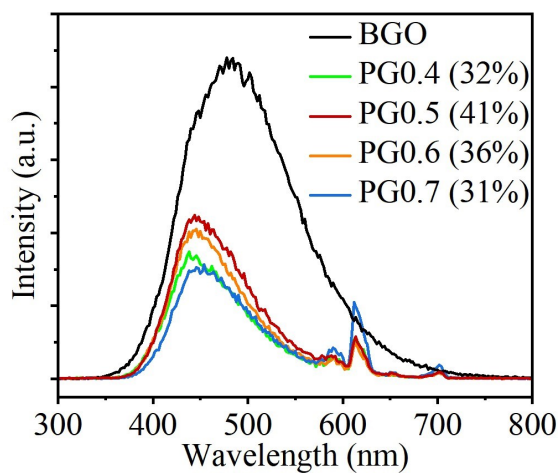


Fig. S1 XEL spectra of BGO and PG x ($x = 0.4, 0.5, 0.6, 0.7$) samples.

C. X-ray attenuation efficiency of $\text{YPO}_4:\text{Eu}^{2+}$ glass-ceramic

Fig. S2 shows function of X-ray attenuation efficiency with thickness of $\text{YPO}_4:\text{Eu}^{2+}$ glass-ceramic. X-ray attenuation efficiency of $\text{YPO}_4:\text{Eu}^{2+}$ glass-ceramic can reach 99.5% when its thickness is 2 mm.

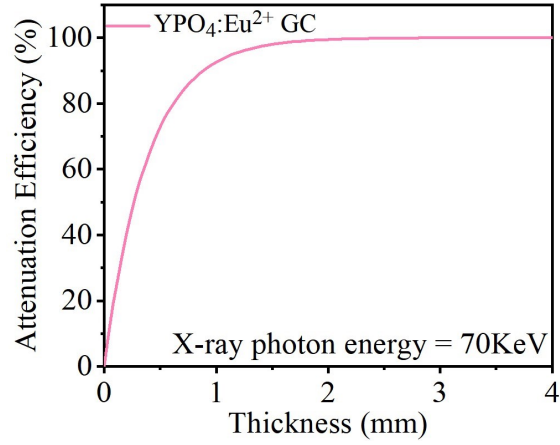


Fig. S2 Function of X-ray attenuation efficiency with thickness of $\text{YPO}_4:\text{Eu}^{2+}$ glass-ceramic.

D. Change of transmittance of GC695 sample after irradiation

Fig. S3 exhibits transmission spectra of GC695 sample without irradiation and after irradiation for 120 mins. Transmittance of GC695 sample hardly changes after being irradiated by X-ray for 120 mins, which indicates that GC695 sample has excellent irradiation stability.

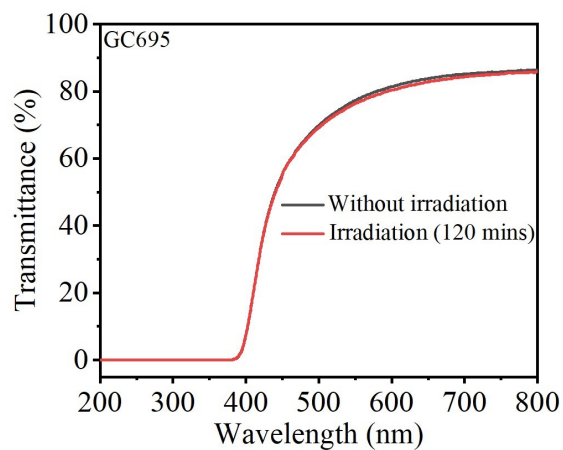


Fig. S3 Transmission spectra of GC695 sample without irradiation and after irradiation for 120 mins.

E. Thermal stability of $\text{YPO}_4\text{:Eu}^{2+}$ glass-ceramic

The thermal stability of GC695 sample and BGO crystal can be demonstrated by Fig. S4. XEL intensity of GC695 sample at 423 K remains 79.6% of that at 303 K. While XEL intensity of BGO at 423 K is only 13.0% of that at 303 K. This result indicates that the thermal stability of $\text{YPO}_4\text{:Eu}^{2+}$ glass-ceramic under X-ray irradiation is far superior to that of BGO crystal.

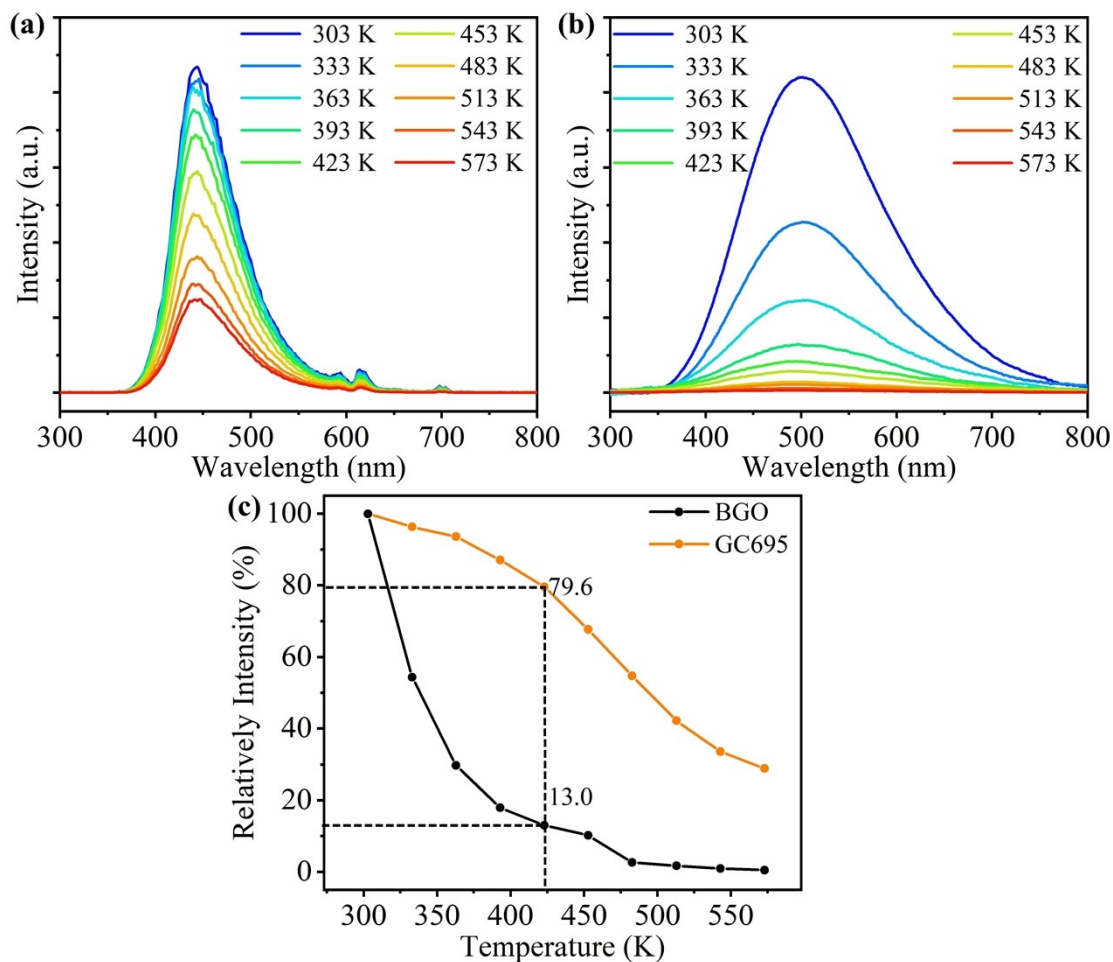


Fig. S4 XEL spectra of (a) GC695 sample and (b) BGO crystal at different temperatures. (c) Relatively XEL intensity of GC695 sample and BGO crystal at different temperature.

F. Density and effective atomic number of YPO₄:Eu²⁺ glass-ceramic

Density (ρ) and effective atomic number (Z_{eff}) are fundamental and important parameters for glass scintillators. ρ of YPO₄:Eu²⁺ glass-ceramic measured according to Archimedes' principle is 3.03 g/cm³. Z_{eff} values are estimated by using the following formula,¹

$$Z_{eff} = 2.94 \sqrt[2.94]{f_1(Z_1)^{2.94} + f_2(Z_2)^{2.94} + \dots + f_i(Z_i)^{2.94}} \quad (S1)$$

where f_i is fraction of total number of electrons associated with each element, and Z_i is atomic number of each element. Z_{eff} of YPO₄:Eu²⁺ glass-ceramic is 21.3.

Reference

1. M. P. Singh, B. S. Sandhu and B. Singh, *Phys. Scripta*, 2007, **76**, 281.