Research on Enhancing the Performance of Glass Scintillator

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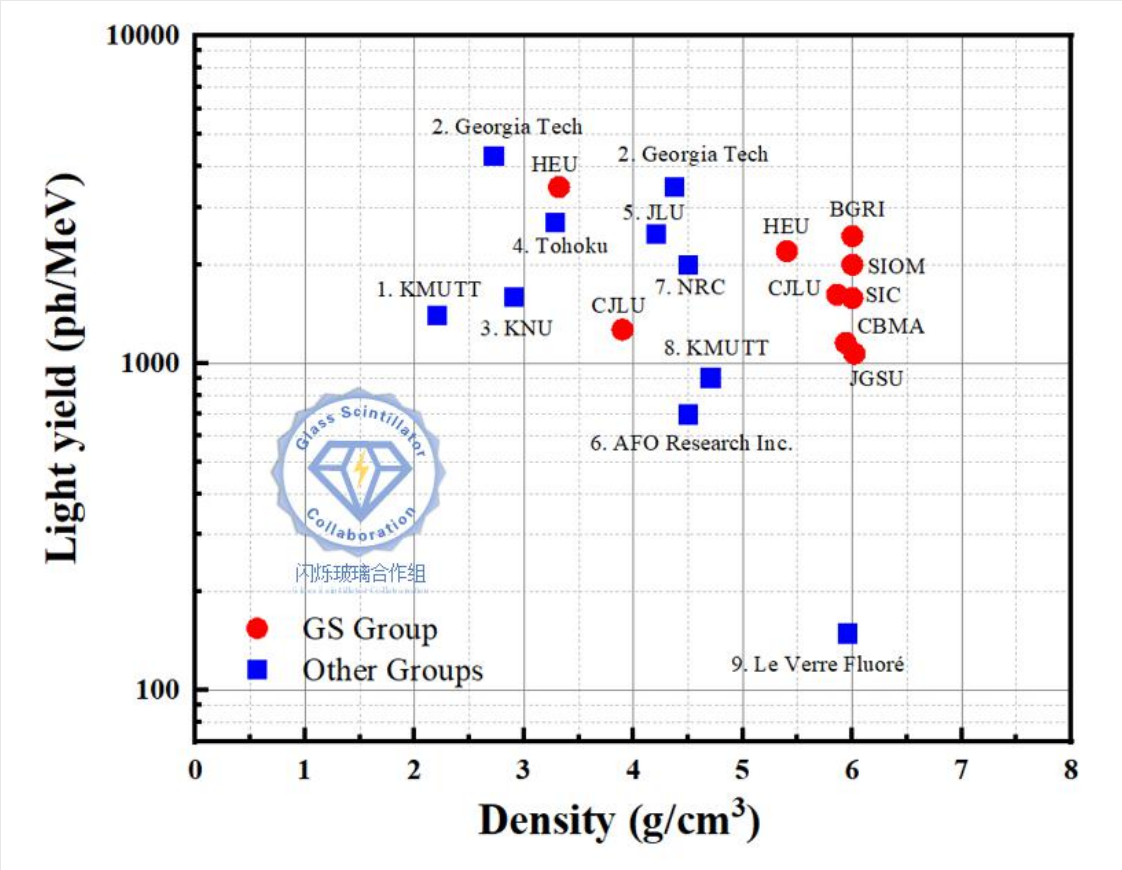
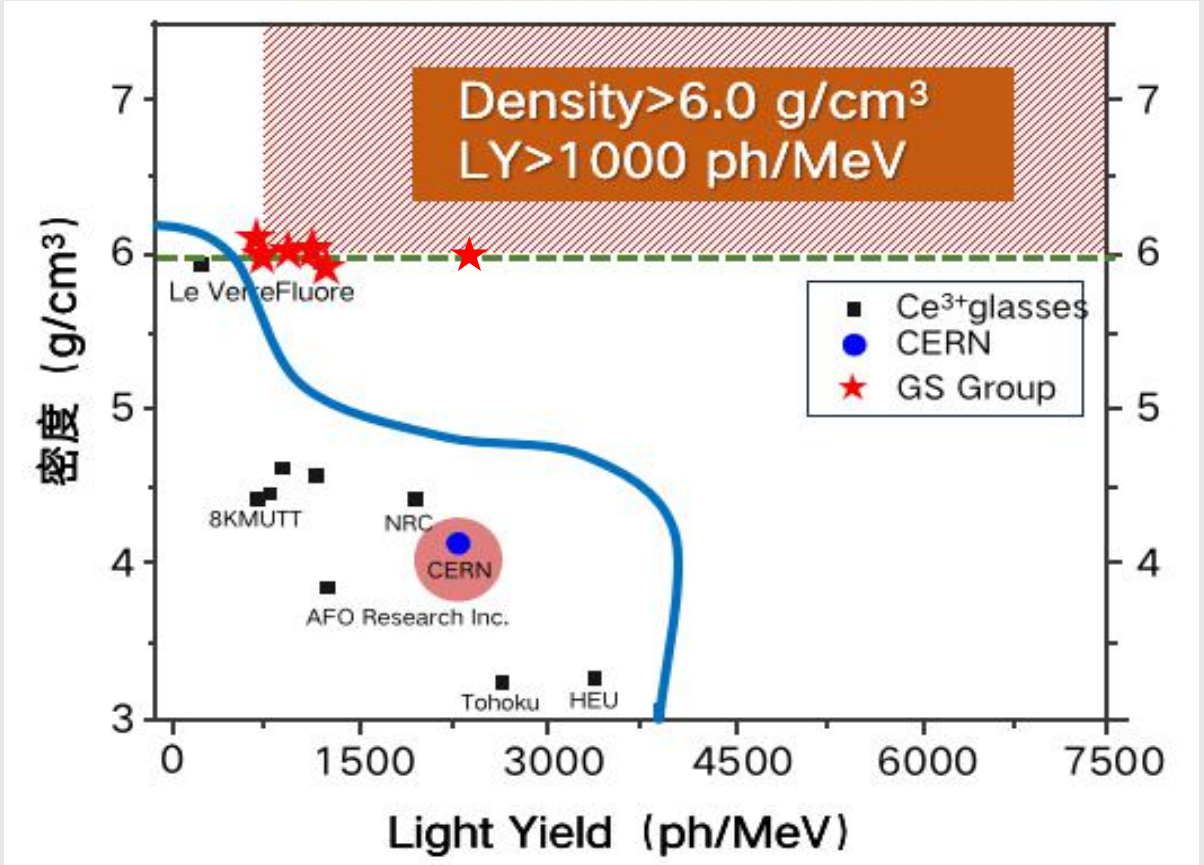
Outline

- 1. Performance target of Glass Scintillator（GFO）
- 2. Theoretical feasibility of Light Yield Enhancement
- 3. Challenges in Enhancing Light Yield
  - 3.1. Density
  - 3.2. Scintillation Decay Time
  - 3.3. Wavelength Shift
  - 3.4. Co-doping

1.0 Research Status of GS





- The GS group has carried out a comprehensive and complete study;
- For high density glass scintillator, the light yield of GS group samples is in the absolute lead.

1.1 Performance target of GS

	2021.09 (1st Meeting)	2024.05 (7th Meeting)	2024.10 (8th Meeting)	2025.09 (9th Meeting)	2026
	Target	Realization	Target	Realization	Target
Dimension (mm^3)	/	5 × 5 × 5	40 × 40 × 10	40 × 40 × 10	40 × 40 × 10
Density (g/cm^3)	6.0	5.93	6.0	>6.0	6.0
Transmittance (% @400 nm)	>75	~70	>80	76	>80
Emission peak (nm)	~400	~400	~400	390	~400
Light yield (ph/MeV)	1000	985	1500	1408	2000
Energy resolution (% @662keV)	/	30.3	/	28.7	<25
Decay time (ns)	<100	35.8(10.7%), 105.1	<500	72.4 (10.5%),547.8	<500
Attenuation length (cm@400nm)	/	/	/	6.05	>10

Published Paper about Glass Scintillator

Nuclear Inst. and Methods in Physics Research, A 1015 (2021) 165762

Contents lists available at ScienceDirect

Nuclear Inst. and Methods in Physics Research, A

journal homepage: www.elsevier.com/locate/nima

Multipurpose Ce-doped Ba-Gd silica glass scintillator for radiation measurements

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ARTICLE INFO

ABSTRACT

A new inorganic scintillation material based on Ba-Gd silica glass doped with cerium (BGS) is fabricated and studied. With the highest light yield among heavy glasses at the level of 2500 ph/MeV and fast scintillation response, the new scintillator ensures a good coincidence time resolution of < 230 ps FWHM for 511 keV γ-quanta from a ^{22}Na source and SiPM readout. In addition to good performance in γ-quanta detection, the material demonstrates capability for efficient detection of low-energetic neutrons. The scintillator is produced by exploiting the standard industrial glass technology, which allows for an unlimited scaling up the conversion of raw material into a high-quality scintillator at a high rate. The glass can be casted in application-specific molds, so minimizing the material losses. The presented glass scintillator has potential for further improvement of its light output and scintillation response time.

1. Introduction

Oxide glasses and glass ceramics made from binary compounds and containing metal and silica oxides have been found to be a good matrix to develop scintillation materials doped with Ce ions [1]. Silica glasses produced by sol–gel method and doped with Ce or Pr were found to be promising scintillation materials for medical and high energy physics (HEP) applications [2–8]. Other light glasses, with a relatively high light yield, which, however, is a few times smaller than the typical light yields of single-crystal scintillation materials [6–7], are Li-based compounds. Nevertheless, they are successfully used to detect thermal neutrons when the glass is enriched with ^{6}Li ions. In contrast, the enrichment of the glass with ^{7}Li ions makes it insensitive to thermal neutrons. Moreover, there are ongoing attempts in replacing Pb by Bi to trigger a new turn of the development of heavy and possibly scintillating glass for the needs in HEP applications.

The glass based on barium di-silicate $BaO{\cdot}2SiO_2$ (DSB) might become a new scintillation material suitable for operation even in harsh radiation environments such as those in collider experiments [8–11]. This glass is produced from an inexpensive initial charge and allows manufacturing in large quantities by standard technology exploited in the glass industry. It exhibits relatively fast response; its light yield is lower than that of the most efficient scintillators but exceeds the light yield of PWO_4, which is nowadays the most extensively used scintillating material in HEP experiments [12]. However, the radiation length is by far not compatible and could be considered for a sampling calorimeter concept only. The initial DSB composition can be supplemented by loading with rare-earth oxides and the corresponding amount of silica oxide. As a result, the light yield of the glass can reach up to 30% of that in the well-known scintillator BGO [13]. In parallel, the density can be increased to 4.2 g/cm^3 or even higher, which enhances capabilities of detecting low-energy gamma quanta below 1 MeV with acceptable energy resolution and designing heterogeneous detection cells of "shashlik" or "spaghetti " type. Furthermore, the enrichment of barium-silica glass with gadolinium is preferable, because Gd oxide pulse height spectrum (PHS), and light output (LO) before and after irradiations by γ-rays. We also discuss possible applications in future HEP experiments.

2. Samples and measurements

Fig. 1 shows two batches of 10 × 10 × 10 mm^3 glass samples produced in July (APO-1607) and November (APO-1611), 2016. These samples have a cerium doping level ranged from 0.5 to 5 wt% and various co-doping. Table 2 lists detailed information.

PL and time-resolved PL were measured by an Edinburgh Instruments FLS 920 fluorescence spectrometer. For the XEL measurements, samples were placed in the sample compartment of a HITACHI F-4500 spectrophotometer. X-rays generated by an Amptek Eclipse-III X-ray tube were used to excite the sample. Transmittance was measured by a PerkinElmer Lambda 950 spectrophotometer with 0.15% precision. LO was measured by a Hamamatsu R2059 PMT with a grease coupling for shown significant progress in the improvement of the time resolution [8], further progress in the improvement of crystalline scintillators is proceeding at a much slower pace. One of the strategies to improve the time response of crystalline scintillation detectors is the composition complication in order to achieve compositionally disordered inorganic compounds. The transition from Lu_2SiO_5:Ce to $(Lu,Y)_2SiO_5$:Ce and from $Gd_3(Al,Ga)_5O_{12}$:Ce to $(Gd,Y)_3(Al,Ga)_5O_{12}$:Ce allows to improve the coincidence time resolution at the registration of annihilation gamma-quanta from 120 to 80 ps [9] and from 160 to 120 ps [10] correspondingly. Apparently, the achievable level of coincidence time resolution for the state-of-art detectors with classical scintillator – photodetector coupling is limited by ~50 ps.

An alternative solution is to combine the detection of various types of ionizing radiation arising in the scintillator when interacting with the annihilation gamma-quanta [11]. In addition to scintillation, Cherenkov radiation is generated in an inorganic transparent material upon a gamma-quanta absorption, which is caused by a motion of the photo-electron. Depositing a photocathode directly on the face of the hybrid scintillation and Cherenkov emitter allows making large-area detectors with coincidence time resolution (CTR) of annihilation gamma-quanta of less than 50 ps [12]. It worth noting, that for such an application the scintillation yield ceases to be a necessary requirement, while the parameters of scintillation kinetics become important – scintillation rise and decay times, which determine the number of photons in the front edge of the scintillation pulse.

Nuclear Inst. and Methods in Physics Research, A 954 (2020) 161665

Contents lists available at ScienceDirect

Nuclear Inst. and Methods in Physics Research, A

journal homepage: www.elsevier.com/locate/nima

Alkali-free Ce-doped and co-doped fluorophosphate glasses for future HEP experiments

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ARTICLE INFO

ABSTRACT

We report status of alkali-free cerium-doped and co-doped fluorophosphate glasses as a potential inorganic scintillator for future high energy physics experiments. Optical and scintillation properties, such as emission, transmittance, light output, decay time and their degradation after γ-ray irradiations, are measured for glass samples produced at APO Research Inc. Further developments are needed for this potential cost-effective glass scintillator to be used for the HHCAL detector concept.

Contents

1. Introduction
2. Samples and measurements
3. Experimental results
4. Conclusion
Acknowledgements
References

1. Introduction

Because of their relatively lower fabrication cost as compared to inorganic crystals, scintillating glasses have a potential for future high energy physics (HEP) experiment where a large volume of scintillators are needed. One example is the Homogeneous Hadronic Calorimeter (HHCAL) detector concept featured with unprecedented jet mass resolution by measuring both scintillation and Cherenkov light, where up to 100 m^3 scintillators are required [1–4]. Table 1 compares basic properties of alkali-free cerium-doped and co-doped fluorophosphate (Ba 44 wt%, P 13 wt%, Mg 5 wt%, Al 2 wt%, O 20 wt%, F 16 wt%) glasses [5–8] with commonly used inorganic and plastic scintillators. With a density of 4.6 g/cm^3 and a nuclear interaction length of 26.4 cm, this material provides a cost-effective solution for the HHCAL detector concept.

In this investigation, we measured optical and scintillation properties of alkali-free cerium-doped and co-doped fluorophosphate glasses fabricated at APO Research Inc. (APO glasses), including photo-luminescence (PL), PL pulse shape, X-ray excited luminescence (XEL), transmittance,

Journal of Non-Crystalline Solids 580 (2022) 121393

Contents lists available at ScienceDirect

Journal of Non-Crystalline Solids

journal homepage: www.elsevier.com/locate/jnoncrysol

$(Gd,Ce)_2O_3$-Al_2O_3-SiO_2 scintillation glass

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ARTICLE INFO

ABSTRACT

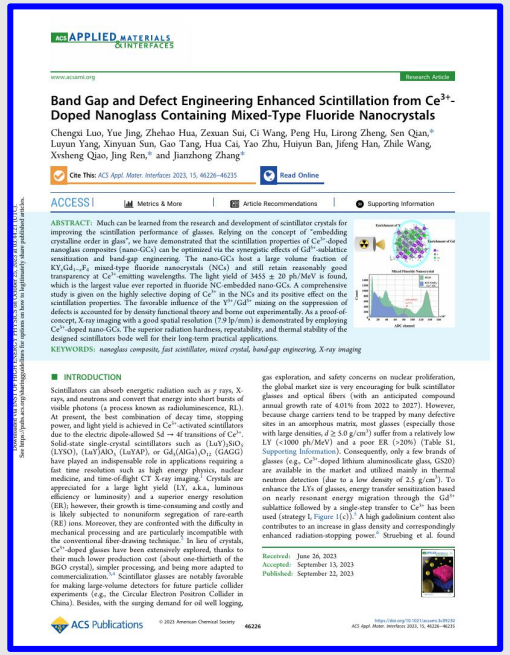
Heavy scintillation glass of Gd_2O_3-Al_2O_3-SiO_2 composition doped with Ce, combining high light yield and fast scintillation kinetics, was developed. The glass samples were prepared at 1500 °C by a melt-quench technique; the resulting glass was colorless and satisfactorily homogeneous. A photoluminescence study showed that a variety of sites for the Ce^{3+} ion stabilization exists in the glass, but the phenomenon was found less pronounced compared to BaO-Gd_2O_3-SiO_2 glass. The glass has demonstrated ~4 times faster scintillation kinetics and a minor drop of the light yield related to barium-gadolinium-silica glass, which makes this material of particular interest for large volume detectors in high energy physics, medical imaging, and other applications due to a low-cost production capability.

1. Introduction

Inorganic scintillation materials are widely used for ionizing radiation detection in medical imaging, high energy physics, homeland security, and other applications [1]. The most outstanding user parameters were shown for single-crystalline materials, which have certain technological drawbacks. Some of them require special encapsulation and caution on utilization due to their high hygroscopicity, such as bromides or iodides [2]. Oxides are produced at high temperatures from precious metal crucibles (Ir or Ir-Re alloy), such as yttrium and rare earth (RE)-based oxyorthosilicates, perovskites, and garnets. Moreover, most of them are manufactured with a low rate of transformation of raw material into a crystal. These features make detectors utilizing such materials quite expensive.

Nuclear instrumentation is on the verge of several important challenges, like creation of a new generation of computer tomography (CT) scanners with photon-counting [3], positron emission tomography (PET) scanners for the whole human body [4], as well as new detectors for use in high energy physics (HEP) experiments at the existing and future Colliders (including the Large Hadron Collider) [5–7]. The time resolution of a detector becomes a crucial consumer parameter for all these applications. As PET scanners and HEP detectors require large volumes of detector materials, it is preferable to use scintillators, which are cheap to manufacture.

While the electronic components of scintillation detectors have



ACS APPLIED MATERIALS & INTERFACES

Band Gap and Defect Engineering Enhanced Scintillation from Ce^{3+}-Doped Nanoglass Containing Mixed-Type Fluoride Nanocrystals

Chengxi Luo, Yue Jing, Zhehao Hua, Zexuan Sui, Ci Wang, Peng Hu, Lirong Zheng, Sen Qian, Luyun Yang, Xinyuan Sun, Gao Tang, Hua Cai, Yao Zhu, Huiyun Ban, Jifeng Han, Zhile Wang, Xvsheng Qiao, Jing Ren, and Jianzhong Zhang

Cite This: ACS Appl. Mater. Interfaces 2023, 15, 46226–46235

ABSTRACT: Much can be learned from the research and development of scintillator crystals for improving the scintillation performance of glasses. Relying on the concept of "embedding crystalline order in glass", we have demonstrated that the scintillation properties of Ce^{3+}-doped nanoglass composites (nano-GCs) can be optimized via the synergistic effects of Gd^{3+}-sublattice sensitization and band-gap engineering. The nano-GCs host a large volume fraction of $KY_xGd_{1-x}F_4$ mixed-type fluoride nanocrystals (NCs) and still retain reasonably good transparency at Ce^{3+}-emitting wavelengths. The light yield of 3455 ± 20 ph/MeV is found, which is the largest value ever reported in fluoride NC-embedded nano-GCs. A comprehensive study is given on the highly selective doping of Ce^{3+} in the NCs and its positive effect on the scintillation properties. The favorable influence of the Y^{3+}/Gd^{3+} mixing on the suppression of defects is accounted for by density functional theory and borne out experimentally. As a proof-of-concept, X-ray imaging with a good spatial resolution (7.9 lp/mm) is demonstrated by employing Ce^{3+}-doped nano-GCs. The superior radiation hardness, repeatability, and thermal stability of the designed scintillators bode well for their long-term practical applications.

KEYWORDS: nanoglass composite, fast scintillator, mixed crystal, band-gap engineering, X-ray imaging

INTRODUCTION

Scintillators can absorb energetic radiation such as γ rays, X-rays, and neutrons and convert that energy into short bursts of visible photons (a process known as radioluminescence, RL). At present, the best combination of decay time, stopping power, and light yield is achieved in Ce^{3+}-activated scintillators due to the electric dipole-allowed 5d → 4f transitions of Ce^{3+}. Solid-state single-crystal scintillators such as $(LuY)_2SiO_5$ (LYSO), (LuY)AlO_3 (LuYAP), or $Gd_3(Al,Ga)_5O_{12}$ (GAGG) have played an indispensable role in applications requiring a fast time resolution such as high energy physics, nuclear medicine, and time-of-flight CT X-ray imaging.[1] Crystals are appreciated for a large light yield (LY, a.k.a., luminous efficiency or luminosity) and a superior energy resolution (ER); however, their growth is time-consuming and costly and is likely subjected to nonuniform segregation of rare-earth (RE) ions. Moreover, they are confronted with the difficulty in mechanical processing and are particularly incompatible with the conventional fiber-drawing technique.[2] In lieu of crystals, Ce^{3+}-doped glasses have been extensively explored, thanks to their much lower production cost (about one-thirtieth of the BGO crystal), simpler processing, and being more adapted to commercialization.[3,4] Scintillator glasses are notably favorable for making large-volume detectors for future particle collider experiments (e.g., the Circular Electron Positron Collider in China). Besides, with the surging demand for oil well logging, gas exploration, and safety concerns on nuclear proliferation, the global market size is very encouraging for bulk scintillator glasses and optical fibers (with an anticipated compound annual growth rate of 4.01% from 2022 to 2027). However, because charge carriers tend to be trapped by many defective sites in an amorphous matrix, most glasses (especially those with large densities, $d \geq 5.0$ g/cm^3) suffer from a relatively low LY (<1000 ph/MeV) and a poor ER (>20%) (Table S1, Supporting Information). Consequently, only a few brands of glasses (e.g., Ce^{3+}-doped lithium aluminosilicate glass, GS20) are available in the market and utilized mainly in thermal neutron detection (due to a low density of 2.5 g/cm^3). To enhance the LYs of glasses, energy transfer sensitization based on nearly resonant energy migration through the Gd^{3+} sublattice followed by a single-step transfer to Ce^{3+} has been used (strategy I, Figure 1(c)).[5] A high gadolinium content also contributes to an increase in glass density and correspondingly enhanced radiation-stopping power.[6] Struebing et al. found

Received: June 26, 2023
Accepted: September 13, 2023
Published: September 22, 2023

ACS Publications

Ceramics International 51 (2025) 19780–19786

Contents lists available at ScienceDirect

Ceramics International

journal homepage: www.elsevier.com/locate/ceramint

Ce^{3+}-doped glass scintillator with high density and light yield for imaging application

Dong Yang, Kun Ge, Huiyun Ban, Huiping Yuan, Sen Qian, Zhehao Hua, Hua Cai, Danping Chen, Jifeng Han, Xvsheng Qiao, Jing Ren, Xinyuan Sun, Gao Tang, Shenghua Yin, Minghui Zhang, Jiawen Xiao

ARTICLE INFO

ABSTRACT

A series of Ce^{3+}-doped Gd_2O_3-GdF_3-Al_2O_3-B_2O_3-SiO_2 glass scintillators (GSC_s and GLC_s) were successfully synthesized in reducing atmosphere for future imaging application. The transmittance of GSC_s glasses exceeds 85 % in range of 500–800 nm, and the cut-off edge red shifts as the Ce^{3+} concentration increases. In photoluminescence (PL), GSC_s glass scintillators have a 360–600 nm broadband emission with a shoulder peak at around 490 nm. Meanwhile, the PL lifetime of GSC_s glasses is close to the CaF_2 crystals, and the PL QY of GSC_s glass scintillator reaches up 70.2 %. For scintillation application, the integral XEL intensity of GSC_8 and GSC_9 glasses is 31.3 % and 24.4 % of that of BGO crystal. GLC_8 and GLC_9 glasses exhibit similar spatial resolution of approximately 7 lp·mm^{-1}. Moreover, the light yields of GSC_8 and GSC_9 glass scintillator are 1702 and 1690 ph/MeV excited by γ-ray, respectively. The nonproportionality of GSC_s glasses is less than 3 % in the range of 100–1400 keV due to the large density and Z_{eff}. As Ce^{3+} concentration increases, both the fast and slow components of the glasses decrease (103.7 ns–89.7 ns, 1454.4 ns–1012.8 ns), and the proportion of fast component also increased from 3.3 % to 4.7 %. For γ-ray imaging, the scintillation light events of the glass can be observed from SiPM array, and the central position (−2, −1) have the highest count rate of the system. The results show that GSC_8 and GLC_8 glass scintillators deserve further development for future applications.

1. Introduction

Scintillator is a material that exhibits the luminescence property when it is excited by ionizing radiation or high-energy particles, which is commonly applied for detecting and measuring various types of radiation, incorporating X-, γ-rays, and charged, neutral particle (for instance,

- 2015 NIMA.
  - DSB:Ce glass
  - Density=4.2 g/cm^3
  - LY=2500 ph/MeV
- 2020 NIMA.
  - AFO glass
  - Density=4.6 g/cm^3
  - LY=700 ph/MeV
- 2022 J. Non-Cryst. Solids.
  - $(Gd,Ce)_2O_3$-Al_2O_3-SiO_2 glass
  - Density=4.5 g/cm^3
  - LY=2000 ph/MeV
- 2023 ACS Appl. Mater. Interfaces.
  - K(Y,Gd)F_4:Ce^{3+} GC
  - Density=3.32 g/cm^3
  - LY=3455 ph/MeV
- 2025 Ceram. Int.
  - GFO:Ce glass
  - Density=6.0 g/cm^3
  - LY=1700 ph/MeV

- Currently, more research on GS is directed towards X-ray imaging, which do not require high-density properties.
- Studies on high-density and light-yield GS remain virtually unexplored internationally.

1.2 Research Progress on GS

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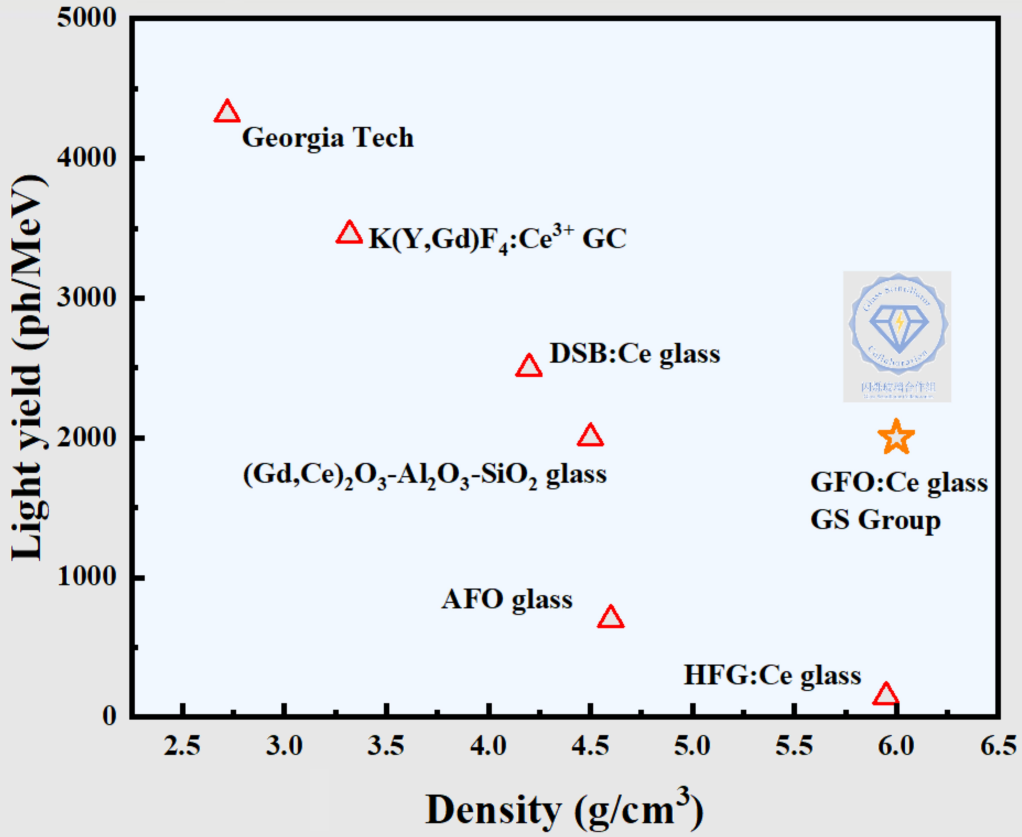
6. C.X. Luo, et al., ACS Appl. Mater. Interfaces, 2023, 15, 46226−46235.



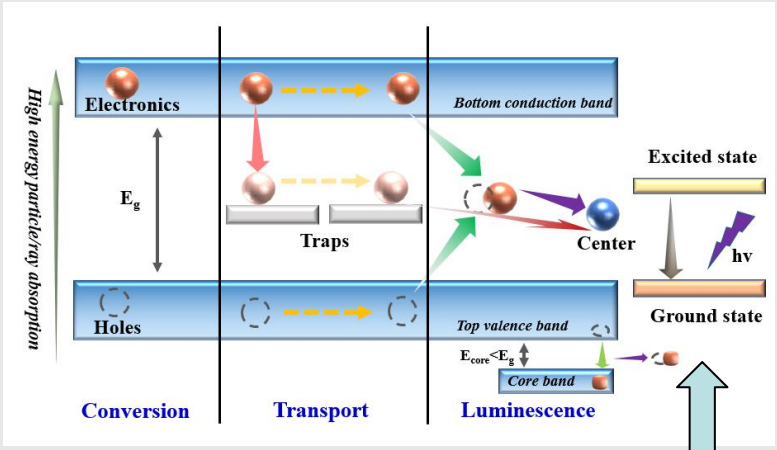
Year	Scintillator	Density (g/cm³)	Light yield (ph/MeV)
1996	HFG:Ce glass	5.95	150
2015	DSB:Ce glass	4.2	2500
2017	$Li(Al_y,Gd_{1-x-y},Ce_x)Si(O,F,Br)$ glass	2.72	4317
2020	AFO glass	4.6	700
2022	$(Gd,Ce)_2O_3$-Al_2O_3-SiO_2 glass	4.5	2000
2023	$K(Y,Gd)F_4:Ce^{3+}$ GC	3.32	3455
	GFO:Ce glass	6.0	1700

- The introduction of heavy elements and structural densification → high density → more severe self-absorption → difficult to balance density and light yield;
- GFO: Ce is the only type of GS which can achieve both high density and high light yield.

Outline

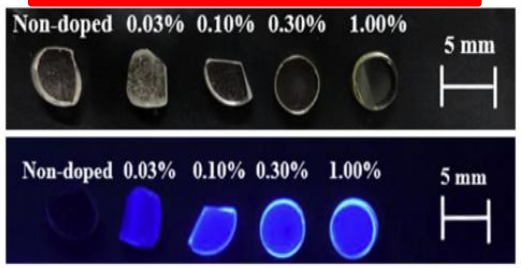
- 1. Performance target of Glass Scintillator（GFO）
- 2. Theoretical feasibility of Light Yield Enhancement
- 3. Challenges in Enhancing Light Yield
  - 3.1. Density
  - 3.2. Scintillation Decay Time
  - 3.3. Wavelength Shift
  - 3.4. Co-doping

2.0 The Design of the GS



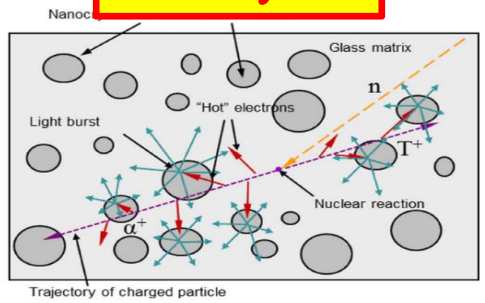
- Scintillation mechanism---- **Luminescence Center**
- **Conversion**—photoelectric effect and Compton scattering effect;
- **Transport**—electrons and holes migrate;
- **Luminescence**—captured by the luminescent center ions

Lanthanide elements



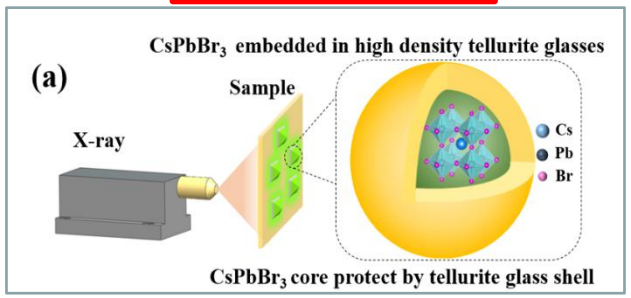
Journal of Alloys and Compounds 782 (2019) 859-864

Nanocrystals



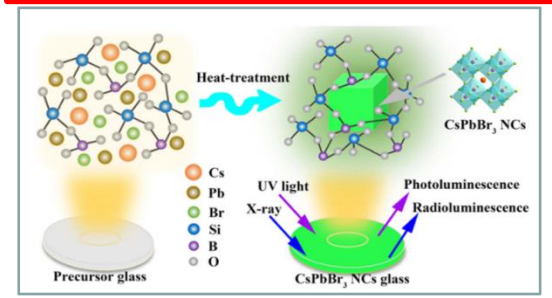
IEEE TNS 60 (2) 2013

Quantum Dots



Optics Letters 46(14) 3448-3451 (2021)

Lanthanide + Quantum Dots



Vol. 9, No. 12 / 2021 / Photonics Research

- High Light Yield (> 2000 ph/MeV): Lanthanide for the Luminescence Center: Cerium (Ce);
- High Density (> 6 g/cm^3) and Low radioactivity background: Gadolinium (Gd); ~~lutetium (Lu)~~

2.1 Theoretical feasibility

which then radiate scintillation photons. The light yield LY in terms of the number of scintillation photons produced per MeV of energy deposited in the crystal can be expressed as [64]

$$LY = 10^6 \, S \cdot Q/(\beta \cdot E_g), \tag{35.3}$$

where $\beta \cdot E_g$ is the energy required to create an e-h pair expressed as a multiple of the band gap energy Eg (eV), S is the efficiency of energy transfer to the luminescent center and Q is the quantum efficiency of the luminescent center. The values of β, S and Q are crystal dependent and

Cite by S.E. Derenzo, W.-S. Choong and W.W. Moses, Phys. Med. Biol.59, 3261 (2014).

For inorganic scintillation crystals, the light yield can be enhanced by improving the intrinsic properties of the material:

1. Reducing the bandgap energy (**Eg**);
2. Improving the energy transfer efficiency (**S**) from the sensitizer to luminescent center (activator) ;
3. Increasing the quantum yield (**Q**) of the luminescent center.

2.2 Enhance in light output

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Nuclear Instruments and Methods in Physics Research A 1072 (2025) 170182

Contents lists available at ScienceDirect

Nuclear Inst. and Methods in Physics Research, A

journal homepage: www.elsevier.com/locate/nima

Light attenuation and loss in Ce^{3+}-doped dense glass scintillator

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ARTICLE INFO

Keywords:
High energy physics
Glass scintillators
Light attenuation length
Light loss coefficient

ABSTRACT

For nuclear radiation detection and HEP experiments, glass scintillators with high transparency and high stability have been considered as potential alternatives. Due to the amorphous metastable structure and a large number of internal defects, the performance of glass scintillators is inferior to that of crystals. There is little research on the attenuation length and light loss coefficient of glass scintillators due to amorphous structure. Ce^{3+}-doped gadolinium aluminoborosilicate glass scintillator (GL_4 glass) was synthesized using high-temperature melting method in a N_2/H_2 reducing atmosphere and processed to different thicknesses. Optical and scintillation properties of the glasses under different thicknesses were investigated. As the thickness of the glasses increases, their cut-off edge of transmission spectra and X-ray excited luminescence (XEL) peak gradually red shift. The difference between theoretical and actual light attenuation lengths was compared. The actual light attenuation length around its luminescence peak is 2.3 ± 0.01 cm of GL_4 glass with a light yield of 1000 ph/MeV. The intrinsic light yield and light loss coefficient of the GL_4 glass were calculated. Its intrinsic light yield is 1583 ± 37 ph/MeV with a light loss coefficient of 0.57 ± 0.04 cm^{-1}. Besides, the scintillation characteristics of the glasses exhibits fast (in range of 160–200 ns) and slow (in range of 440–480 ns) components.

1. Introduction

Scintillation materials have been extensively applied in nuclear radiation detection, medical imaging and high energy physics (HEP), including crystal, plastic, liquid scintillator [1–3]. Glass with high density, high transparency and stable physical and chemical properties is a potential material for scintillation applications, has gradually attracted the attention. Moreover, glass scintillators have the advantages of low cost, large volume preparation and continuous tunable composition, which gives it a certain edge in some large-scale scientific projects.

In recent years, V. Dormenev et al. believed that Ce^{3+}-doped Ba–Gd based silica glass (BGS glass) may replace $PbWO_4$ (PWO) crystal as the next generation sampling calorimeter in Compact Muon Solenoid (CMS) of Large Hadron Collider (LHC) [4–7]. And barium di-silicate $BaO{-}2SiO_2$ (DSB) glass may also be applied in electromagnetic calorimeters at Electron Ion Collider (EIC) [8]. Two key aspects of glass application in EIC are homogeneity of the material and the improvement in glass density. Besides, G. Dosovitskiy et al. prepared the Ce^{3+}-doped $Gd_2O_3{-}Al_2O_3{-}SiO_2$ glass with a light yield ~2000 ph/MeV [6]. However, the density of glass is only 4.5 g/cm^3. In order to attain more accurate measurement of the Higgs boson, a Circular Electron Positron

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https://doi.org/10.1016/j.nima.2024.170182
Received 12 August 2024; Received in revised form 15 December 2024; Accepted 16 December 2024
Available online 17 December 2024
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Ceramics International 51 (2025) 23367–23373

Contents lists available at ScienceDirect

Ceramics International

journal homepage: www.elsevier.com/locate/ceramint

Scattering model of Ce^{3+}–doped high gadolinium fluoro–oxide glass in scintillation progress

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ARTICLE INFO

Handling Editor: Dr P. Vincenzini

Keywords:
Glass scintillator
Scattering model
Light yield
Light loss coefficient

ABSTRACT

High density $Gd_2O_3{-}GdF_3{-}Al_2O_3{-}B_2O_3{-}SiO_2{-}CeF_3$ (GFO_x) glass scintillators with different dimensions were synthesized. The glasses have a ~75 % transmittance in visible range with a cut off wavelength of 373 nm. With the dimension increases, the energy transfer efficiency of GFO_x glasses decreases from 77.0 % to 28.3 %. And the photoluminescence quantum yield (PL QY) of all glasses exceeds 56 % with a maximum of 68.4 %. GFO_x glass scintillators exhibit a X-ray excitation luminescence (XEL) spectrum in 300–600 nm, and the XEL intensity decreases with increasing dimension. Under γ-ray excitation, a scattering model was established based on the light yield and aspect ratio of the glasses. The accuracy of the model was verified by measurement of commercial BGO crystal. In GFO_x glass scintillator system, the intrinsic light yield of at 10 mm thickness is 1318 ph/MeV, while light loss coefficient is ~0.12. With the aspect ratio increases, the formation and transport efficiency of excitons improve due to a larger surface area and more interfaces, resulting in a faster rise time. Meanwhile, all glasses exhibit a double exponential decay, including a fast component (~100 ns) and a slow component (~520 ns), and shows same trend as the rise time. In addition, the photon collection efficiency of SiPM coupled with the glasses decreased from 96.2 % to 31.8 %. Because the scattered photons can hardly reach the SiPM due to poor actual light attenuation length.

1. Introduction

Scintillator materials are demanded for particle identification and energy measurements of particles and rays (such as α, β particles and γ-ray) in modern nuclear physics applications [1–5]. Novel glass scintillators have attracted plenty of attention because of low-cost and continuously adjustable composition, will be an alternative for future nuclear radiation detection and even high-energy physics (HEP) experiments [6–8]. For the large collider experiment, the research of glass scintillators has gone through decades of development. The concept of

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https://doi.org/10.1016/j.ceramint.2025.03.025
Received 2 January 2025; Received in revised form 18 February 2025; Accepted 1 March 2025
Available online 4 March 2025
0272-8842/© 2025 Published by Elsevier Ltd.

➢ 2025 NIMA.
Attenuation Length Test

➢ 2025 Ceram. Int.
Study on size effect

$$LY = LY_0 \times e^{-\frac{L}{LAL}},\ LY = \frac{LY_0 \times (1-e^{-2\mu L})}{2\mu L}$$

LY_0—Intrinsic light yield

L—The propagation distance of scintillation light

LAL—**Light attenuation length**

μ—Light loss coefficient (Reabsorption and reflection)

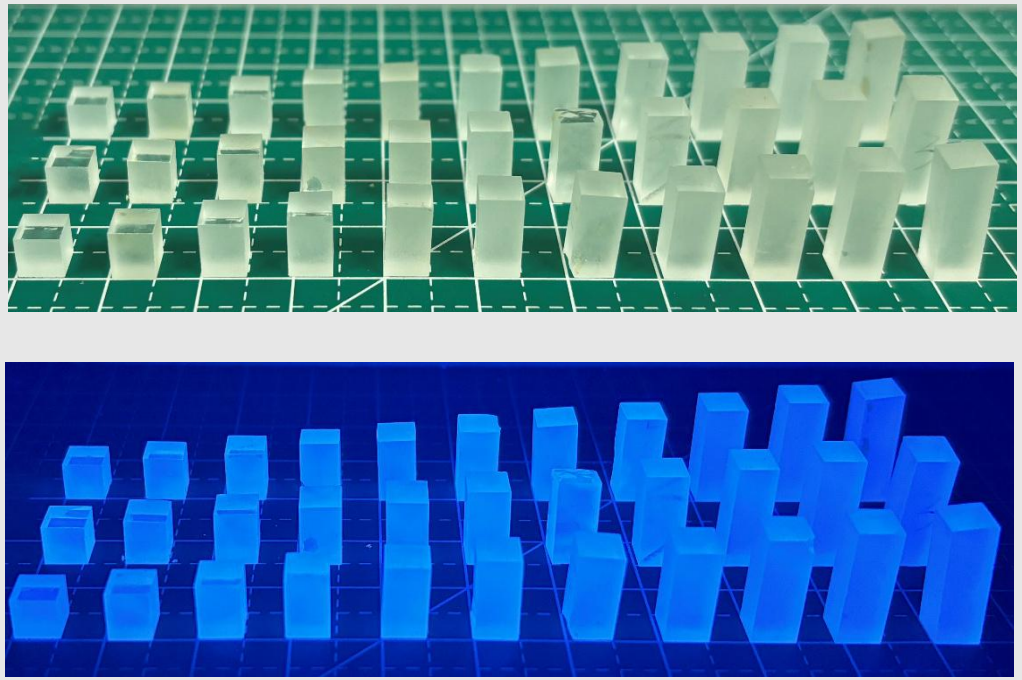
- Reducing losses during scintillation light transmission

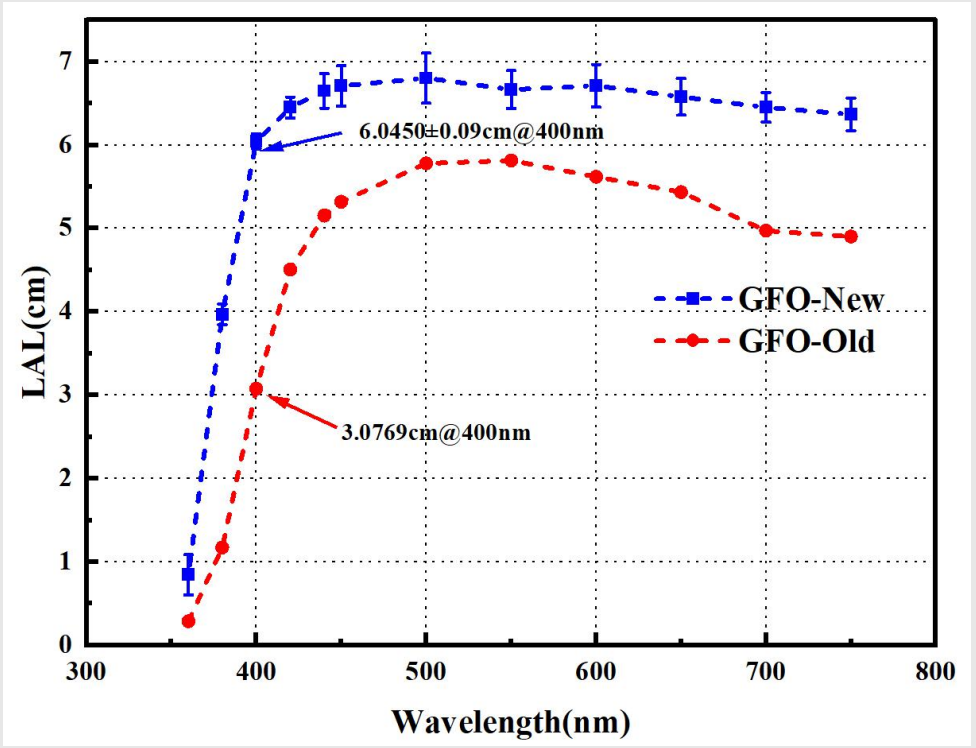
✓ **Improving optical uniformity:**
material purity, melting process (bubbles, striae);

✓ **Enhancing light collection efficiency:**
micro-nano structures, reflector selection, coupler;

✓ **Wavelength shifting:**
improving PDE of photoelectric detector.

2.3 LAL test



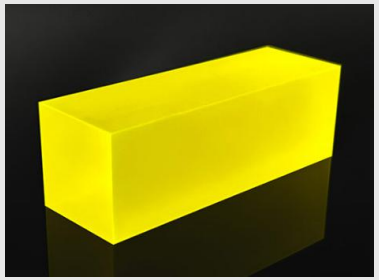


$$I = I_0 \cdot e^{-\frac{l}{L_0}} \quad \Rightarrow \quad L_0 = \frac{l_2 - l_1}{In(\frac{T_{l_1}}{T_{l_2}})}$$

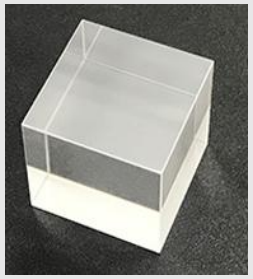
- Due to its high density and limited homogeneity, glass exhibits strong self-absorption, leading to a relatively short attenuation length.
- The uniformity of glass is related to the purity of the raw materials and the preparation process, which is challenging to control with lab-scale samples.

2.4 GFO:Ce glass to GAGG and GSO crystal

Parameters	Unit	GAGG	GSO	GFO:Ce
Density	g/cm^3	6.63	6.71	6.0
Melting point	℃	1850	1950	1150
Radiation Length, X_0	cm	1.63	1.38	1.64
Molière radius	cm	2.20	2.96	2.50
Nuclear interaction length	cm	21.5	20.6	24.1
Z_{eff}	--	50.6	58.0	56.9
dE/dX	MeV/cm	8.96	8.5	8.0
Emission peak	nm	540	430	390
Refractive Index	--	1.92	1.85	1.76
Light yield, LY	ph/MeV	60,000	8,000	~ 1,500
Energy resolution, ER	%662keV	5.0	9.2	25.8
Decay time	ns	100, 600	28, 260	101, 1456
d(LY)/dT	%/℃	/	/	/
Cost	$/cc	/	/	<1.0



GAGG



GSO

- $Gd_3(Al_xGa_{1-x})_5O_{12}$:Ce **(GAGG)**

- Gd_2SiO_5:Ce (GSO)
- GFO:Ce Glass

have the similar compositions;

represent the ultimate performance goal.

1. *Y. Zhu, et al., Opt. Mater., 2020, 105, 109964.*
2. *Z. Wang, et al., JINST, 2020, 15, C06031.*
3. *W. Chewpraditkul, et al., Radiat. Meas., 2016, 92, 49-53.*
4. *B.V. Grinyov, et al., IEEE TNS, 2010, 57, 1236-1240.*

GAGG

Optical Materials 105 (2020) 109964

Contents lists available at ScienceDirect

Optical Materials

journal homepage: www.elsevier.com/locate/optmat

Scintillation properties of GAGG:Ce ceramic and single crystal

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ARTICLE INFO

Keywords:
Scintillation property
Ceramic
Single crystal
GAGG:Ce

ABSTRACT

Recently developed ceramic and single crystal Cerium-doped gadolinium aluminum gallium garnet (GAGG:Ce) scintillators were studied. We compared their scintillation properties with samples of the same 5 × 5 × 5 mm³ size. In photoluminescence emission spectra, ceramic and single cryslut GAGG:Ce showed intense emission peak at 555 nm and 515 nm, respectively, ascribed to 5d–4f transitions of Ce^{3+}. And the single crystal GAGG:Ce had a weak emission peak appearing at 380 nm. The light yield induced by ^{137}Cs γ-ray of ceramic was higher than that of single crystal. The afterglow of ceramic GAGG:Ce was shorter than that of single crystal after illumination. At room temperature, single crystal GAGG:Ce was more severely affected by thermoluminescence than ceramic GAGG:Ce. The primary decay time of ceramic and single crystal under γ-ray was 182.9 ns and 50.1 ns, respectively. Furthermore, the fast component of decay time was found in single crystal. This research can lay the foundation for the preparation of better GAGG:Ce scintillators in the future.

1. Introduction

Inorganic scintillator is a kind of luminescent material which is able to absorb high energy ionizing radiation such as γ-rays and to emit near-ultraviolet or visible photons [1,2]. It is coupled with a photodetector as a scintillation detector. The scintillation detector is widely used in high energy physics [3,4], nuclear medicine [5,6], security inspection [7], non-destructive testing [8], geological exploration [9] and other fields. Scintillators are the core functional element in these applications. Among them, Cerium-doped gadolinium aluminum gallium garnet (GAGG:Ce) is one of the most promising scintillators. GAGG:Ce scintillator has high light yield, high density, fast decay time and so on, which is suitable for γ-ray detectors. The recently developed GAGG:Ce ceramic and single crystal exhibited excellent scintillation properties [10,11]. Therefore, it has attracted much attention especially in computer tomography (CT) and time-of-flight positron emission tomography (TOF-PET) [12,13].

In 2011, GAGG:Ce single crystal was grown by micro pulling down method with the light yield above 40000 photon/MeV and fast decay time about 53 ns [14]. However, the size of crystal grown by the micro pulling down method is small. A 2-inch GAGG:Ce single crystal grown by the Czochralski method demonstrated high light yield of 46000 photon/MeV [15]. Its energy resolution was 4.9%@662 keV and the fast decay time was 88 ns (91%).

Transparent single-phase garnet GAGG:Ce ceramic was successfully produced using ceramic processing technology and the sample displayed high light yield about 65 000 photon/MeV [16]. GAGG:Ce ceramic with high transmittance and light yields of 15500 photon/MeV was fabricated with the vacuum sintering method. The sample exhibited 5 ns scintillation decay time which was different from about 50 ns fast component of other ceramics attributed to the 5d-4f transitions of Ce^{3+}, probably due to instrumental response [17]. Ceramic GAGG:Ce samples were investigated by controlling the annealing temperature in the air which could improve the light yield and energy resolution [18].

In this work, we compared the scintillation properties of ceramic and single crystal GAGG:Ce samples with same size, which provided ideas for improvement and application direction for GAGG:Ce scintillators. We studied ultraviolet light induced emission spectra, transmittance, afterglow, thermoluminescence, γ-ray induced pulse height spectra and scintillation decay time though this work.

2. Samples and test facility

2.1. Sample preparation

The purity of raw materials has a great influence on the scintillation properties of the GAGG:Ce ceramics and single crystal. Therefore, the

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https://doi.org/10.1016/j.optmat.2020.109964
Received 9 March 2020; Received in revised form 24 April 2020; Accepted 25 April 2020
Available online 4 May 2020
0925-3467/© 2020 Published by Elsevier B.V.

➢ 2020 Opt. Mater.

LY=28244±10 ph/MeV

Decay= 50.1, 321.5 ns (60.8%).

GAGG

JINST — Published by IOP Publishing for Sissa Medialab

Received: February 13, 2020
Revised: March 25, 2020
Accepted: April 18, 2020
Published: June 11, 2020

CHEF2019 — Calorimetry for the High Energy Frontier
25–29 November 2019
Centennial Hall, Kyushu University, Fukuoka, Japan

Performance study of GAGG:Ce scintillator for gamma and neutron detection

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Abstract: Inorganic scintillators with high density and high light output are widely used for the detection of ionizing radiation in high energy physics, space exploration, modern medical imaging and industry. Recently developed cerium-doped $Gd_3Al_2Ga_3O_{12}$ (GAGG:Ce) crystal is a promising scintillator with high density, high light yield, fast scintillation decay time and non-hygroscopicity used for calorimetry. Furthermore, the presence of Gd isotopes with a very high thermal neutron capture cross section makes the GAGG:Ce crystal efficiently sensitive for neutron detection. In this manuscript, performance of gamma and neutron detection based on GAGG:Ce scintillator has been studied carefully. Scintillation properties of Single crystal and ceramic GAGG:Ce were investigated. Thanks to very high light yield, the photopeaks due to conversion electrons and low energy X-ray are successfully observed and can be used to do the neutron/gamma discrimination.

Keywords: Scintillators, scintillation and light emission processes (solid, gas and liquid scintillators); Neutron detectors (cold, thermal, fast neutrons); Gamma detectors (scintillators, CZT, HPGe, HgI etc)

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https://doi.org/10.1088/1748-0221/15/06/C06031

2020 JINST 15 C06031

➢ 2020 JINST.

LY=35000 ph/MeV

thermal neutron detection

GSO

Radiation Measurements 92 (2016) 49–53

Contents lists available at ScienceDirect

Radiation Measurements

journal homepage: www.elsevier.com/locate/radmeas

Scintillation timing characteristics of $(La,Gd)_2Si_2O_7$:Ce and Gd_2SiO_5:Ce single crystal scintillators: A comparative study

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HIGHLIGHTS

- Scintillation characteristics of GPSLa23.5%:Ce and GSO:Ce were compared.
- Both crystals showed slow rise-time characteristics in their scintillation pulses.
- Energy resolution of 5.4% @511 keV was obtained for GPSLa23.5%:Ce.
- GPSLa23.5%:Ce showed higher contribution of slow scintillation light.
- Coincidence time resolution of 660 ps was obtained for GPSLa23.5%:Ce.

ARTICLE INFO

Article history:
Received 11 May 2016
Received in revised form
15 June 2016
Accepted 22 June 2016
Available online 25 June 2016

Keywords:
Coincidence time resolution
Energy resolution
$(La,Gd)_2Si_2O_7$:Ce, Gd_2SiO_5:Ce
Scintillation decays

ABSTRACT

The scintillation timing characteristics of $(La,Gd)_2Si_2O_7$:Ce (GPSLa23.5%:Ce) single crystal were studied and compared with Gd_2SiO_5:Ce (GSO:Ce) single crystal. The photoelectron yield, scintillation decay times and coincidence time resolution were measured. At 511 keV γ-rays, the photoelectron yield of 10,770 ± 500 phe MeV^{-1} and energy resolution of 5.4 ± 0.2% obtained for GPSLa23.5%:Ce are much better than those of 3350 ± 160 phe MeV^{-1} and 7.8 ± 0.3% obtained for GSO:Ce. The scintillation decay time profile was measured by the time-correlated single photon counting technique using a fast-slow coincidence setup. In both materials the comparable rise times of several nanoseconds are present. The fast component decay time of 56 ns with relative intensity of 49% obtained for GPSLa23.5%:Ce is inferior to that of 32 ns(88%) obtained for GSO:Ce. Consequently, the coincidence time resolution of GPSLa23.5%:Ce is slightly worse than that of GSO:Ce. The normalized time resolution was also discussed in terms of a number of photoelectrons and decay time of the scintillation pulse.

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1. Introduction

Ce^{3+}-doped oxy-orthosilicate, namely, Gd_2SiO_5:Ce (GSO:Ce) (Takagi and Fukazawa, 1983), Lu_2SiO_5 (LSO:Ce) (Melcher and Schweitzer, 1992), and $(Lu,Y)_2SiO_5$:Ce (LYSO:Ce) (Cooke et al., 2000) are well-known scintillation materials applied in medical imaging, high-energy physics and advanced instruments. They offer favourable combination of high light yield, fast scintillation decay time, high detection efficiency and non-hygroscopic nature. Their luminescence and consequently also scintillation characteristics are influenced by the fact that there are two rare-earth cation sites with different symmetry and number of oxygen anions in the first coordination sphere, in which the Ce^{3+} is embedded (Takagi and Fukazawa, 1983; Melcher and Schweitzer, 1992; Cooke et al., 2000). In LSO:Ce the occupancy of the larger Lu (1) site with

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http://dx.doi.org/10.1016/j.radmeas.2016.06.008
1350-4487/© 2016 Elsevier Ltd. All rights reserved.

➢ 2016 Radiat. Meas.

LY=3350±160 phe/MeV

Decay= 32, 220 ns

GSO

1236 IEEE TRANSACTIONS ON NUCLEAR SCIENCE, VOL. 57, NO. 3, JUNE 2010

Absolute Light Yield Determination for LGSO:Ce, CWO, ZnSe:Al, and GSO:Ce Crystals

Boris V. Grinyov, *Member, IEEE*, Vladimir D. Ryzhikov, Oleg Ts. Sidletskiy, Gennadiy M. Onyshchenko, Sergey N. Galkin, Alexey I. Ivanov, Vladimir A. Tarasov, Olga V. Zelenskaya, Daniil A. Kurtsev, Leonid A. Piven, and Igor M. Zenya

Abstract—**This work has been aimed at measuring the absolute light yield of LGSO:Ce, CWO, ZnSe:Al, and GSO:Ce scintillators in photons/MeV using the PMT intrinsic resolution method. Special feature of this paper is simultaneous and self-consistent determination of absolute light yield and light collection coefficient of the detector.**

Index Terms—**Crystals, lutetium compounds, scintillation detectors, zinc compounds.**

I. Introduction

Light yield is a decisive factor at development of new scintillation materials, optimization of known material properties, and estimation of their applicability.

Relative light yield is usually estimated by comparison of output signal amplitudes from standard and studied scintillators. Therefore, measured amplitude value is sensitive to amplifier shaping time, time of RC load formation for PMT, and type of used spectrographic amplifier. In the case of different scintillators with different decay times, for ex., NaI(Tl) and CWO, accurate determination of section gain (that is, output signal amplitude) is complicated for significantly different shaping times—6 μs for NaI(Tl) and 30 μs for CWO. A generator of real signal shape for a concrete scintillator is necessary for calibration of this section. However, its fabrication may be a rather difficult task.

Consequently, methods of relative light yield estimation may be not suitable for materials with different decay times. It means that any method capable to obtain the absolute light yield (ALY) value is always more precise.

In the present paper, ALY of the series of scintillators was determined by the procedure based on the well-known photo-multitube (PMT) intrinsic resolution method [1]–[4]. The peculiarity of our approach is the possibility of simultaneous experimental determination of both light collection coefficient τ and technical light yield (TLY) of scintillator. Basing on these data, ALY may determined. The developed method is applicable for different types of scintillators, especially for those with slow luminescence decay.

Applicability of our self-consistent method is checked on several scintillators with different light output values. $Lu_{2x}Gd_{2(1-x)}SiO_5$: Ce (LGSO) single crystals with optimized characteristics have been obtained recently [5]. These crystals show good energy resolution and very low afterglow level. Relative light yield estimation shows that these crystals possess rather high light yield. For confirmation of these results, ALY values in photons/MeV for LGSO crystals and known scintillators (CWO, ZnSe:Al, GSO:Ce) was determined in this work by the same method. The ALY values are compared to known results [6]–[8].

II. Theoretical Background

For description of the core of our method, some theory should be recalled. At first, we must clearly distinguish ALY and TLY. ALY value η is determined by the ratio of full energy of scintillation photons E_{ph} to energy E_0 absorbed in scintillator:

$$\eta = E_{ph}/E_0. \quad (1)$$

Under the full energy of scintillation photons, we imply full energy of single-photon light flashes which originate in scintillator at absorption of photon with energy 661.66 KeV and its further conversion into photoelectrons.

ALY of scintillators is a material 'point' property, which is determined by scintillator physical properties and its quality, but not depends on scintillation light collection conditions.

TLY (T) is determined by the ratio of scintillation energy L_{ph} of photons passed through detector window to energy E_0 absorbed in scintillator:

$$T = L_{ph}/E_0. \quad (2)$$

Scintillator TLY depends on many external conditions. These are quality of crystal and reflector surfaces (light collection conditions), shape of scintillator element.

ALY η and TLY T are connected as:

$$T = \tau \cdot \eta = (L_{ph}/E_{ph})\eta, \quad (3)$$

Manuscript received June 25, 2009; revised December 11, 2009, February 04, 2010, and March 30, 2010; accepted March 30, 2010. Date of current version June 16, 2010. This work was supported in part by CRDF Grant UKE2-2958-KH-08.

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Color versions of one or more of the figures in this paper are available online at http://ieeexplore.ieee.org.

Digital Object Identifier 10.1109/TNS.2010.2047732

➢ 2010 IEEE TNS

LY=7900±800 phe/MeV

Outline

- 1. Performance target of Glass Scintillator（GFO）
- 2. Theoretical feasibility of Light Yield Enhancement
- **3. Challenges in Enhancing Light Yield**
  - 3.1. Density
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3.1 Density—（1）Gd_2O_3

Z.X. Sui, et al., Ceram. Int., 2025, 50, 42625-42631.

Ceramics International 50 (2024) 42625–42631

Contents lists available at ScienceDirect

Ceramics International

journal homepage: www.elsevier.com/locate/ceramint

Balancing high density and scintillation light yield in Ce^{3+}-doped gadolinium borosilicate glass

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ARTICLE INFO

Handling Editor: Dr P. Vincenzini

ABSTRACT

A series of Ce^{3+}-doped borosilicate glasses is prepared with high Gd_2O_3 contents, and their physical, optical, and scintillation properties are investigated. The incorporation of Gd_2O_3 results in a notable augmentation of the glass density, elevating it from 4.5 g/cm^3 to 6.0 g/cm^3. Notably, the heaviest glass, containing 72.5 wt% of Gd, exhibits simultaneously an impressively high density of 6.0 g/cm^3 and a reasonably good light yield of 1051 ph/MeV (under the γ-ray irradiation from a ^{137}Cs source), surpassing benchmarks documented for high-density glasses. The glass is colorless and remains highly transparent across the visible spectrum, exhibiting commendably good thermal stability and radiation endurance. Collectively, out results underscore the potential of the glass scintillators as a promising candidate material for high-energy particle calorimeters.

1. Introduction

The confirmation of the existence of the Higgs boson at the Large Hadron Collider (LHC) in 2012 [1,2], heralded a new epoch in particle physics. Subsequently, the precise measurements of Higgs boson properties become a natural and important step for the future exploration of particle physics. Addressing this imperative target, scientists have proposed the establishment of the Large Circular Electron Positron Collider (CEPC), envisioned as a future Higgs factory with remarkable capability of precision measurement on Higgs bosons (*e.g.*, the absolute Higgs couplings) [3,4]. In this context, the selection of scintillation materials assumes paramount significance as they play a pivotal role in the CEPC calorimeter, converting high-energy photons and particles into ultraviolet–visible light for photomultiplier tubes (PMTs) or Silicon photomultipliers (SiPMs), thereby directly influencing measurement precision. Presently, the electromagnetic calorimeter (ECAL) and the hadron calorimeter (HCAL) utilized in compact muon solenoid (CMS) of LHC are constructed from lead tungstate scintillating crystals and plastic

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https://doi.org/10.1016/j.ceramint.2024.08.106
Received 26 June 2024; Received in revised form 27 July 2024; Accepted 6 August 2024
Available online 6 August 2024
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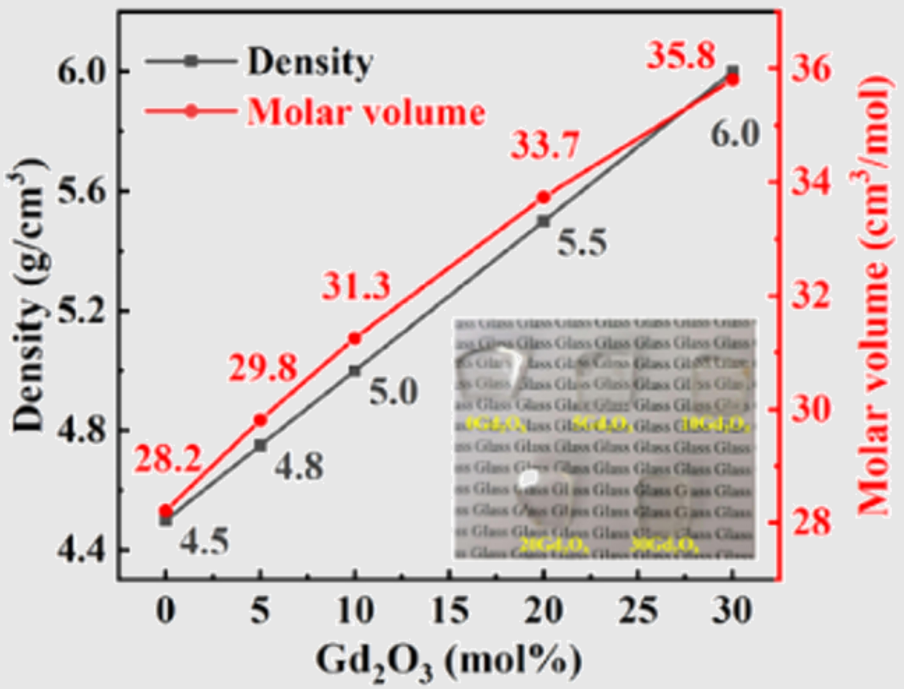
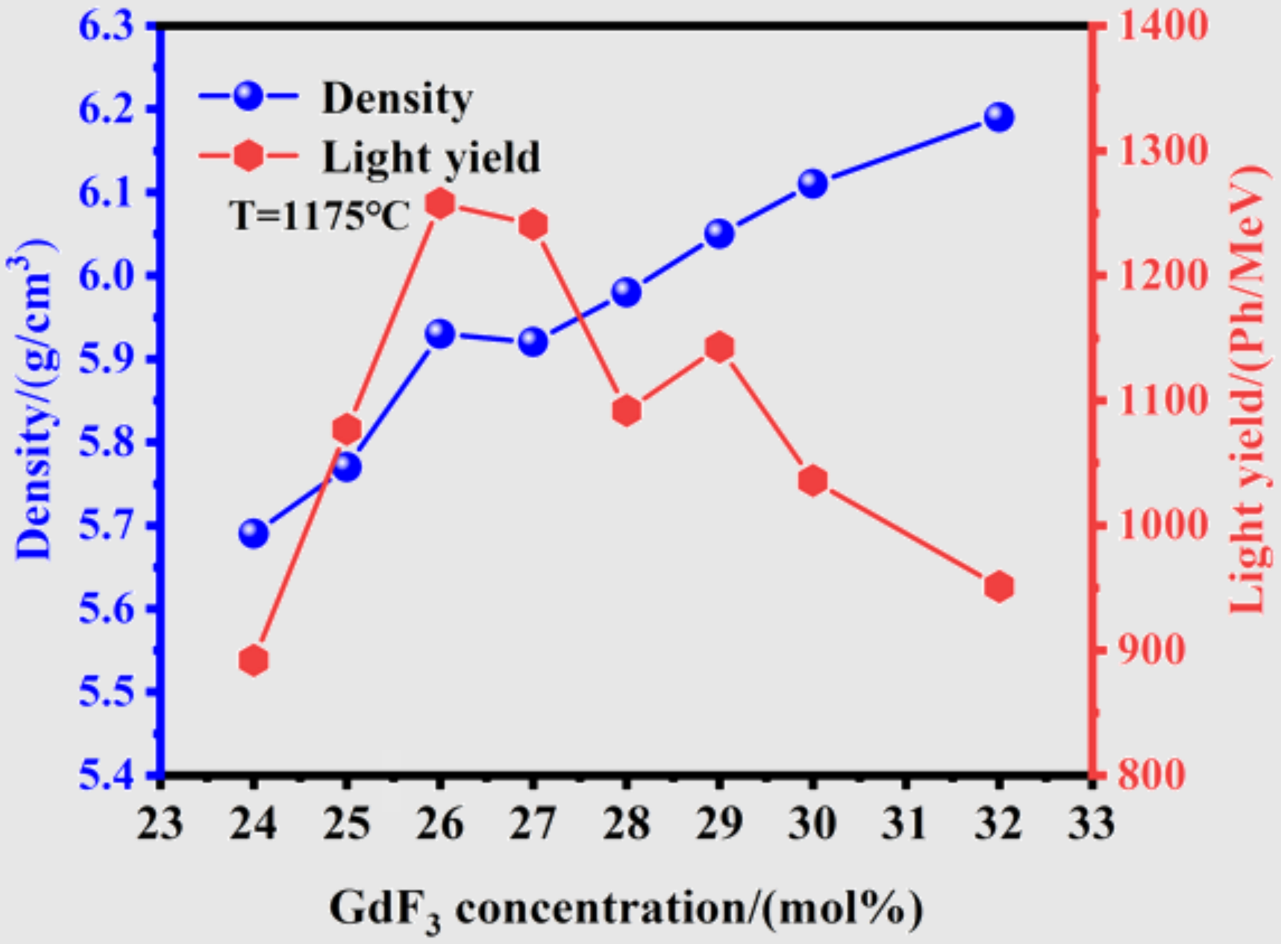


	Density (g/cm³)	LY (ph/MeV)
$0Gd_2O_3$	4.5	1669
$5Gd_2O_3$	4.8	1617
$10Gd_2O_3$	5.0	1547
$20Gd_2O_3$	5.5	1400
$30Gd_2O_3$	6.0	1051

- As the Gd_2O_3 content increases, the glass density rises to 6.0 g/cm³;
- While its light yield declines from around 1700 to ~1000 ph/MeV.

➢ 2025 Ceram. Int.
The influence Gd_2O_3 content

3.1 Density—（2）GdF_3



Recent research progress:

1. By increasing the GdF_3 content, the energy transfer efficiency from Gd to Ce has been enhanced, leading to an improvement in light yield;
2. However, excessive GdF_3 can create quenching centers, which degrade the scintillation performance;
3. By adjusting the ratio of Gd_2O_3 to GdF_3, the coordination structure of GS has been optimized, resulting in a further increase in light yield.

We really need the density of GFO larger than 6g/cc? What about 5.5? or others?

3.2 Decay Time —GAGG: Ce,Mg crystal

Journal of Crystal Growth 393 (2014) 134–137

Contents lists available at ScienceDirect

Journal of Crystal Growth

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Czochralski growth of $Gd_3(Al_{5-x}Ga_x)O_{12}$ (GAGG) single crystals and their scintillation properties

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ARTICLE INFO

Available online 7 November 2013

Keywords:
A2. Czochralski method
B1. Gadolinium compounds
B1. Gallium compounds
B1. Oxides
B2. Scintillator materials
B3. Scintillators

ABSTRACT

Ce:$Gd_3(Al_xGa_{1-x})_5O_{12}$ (x=2.5/5 and 3/5, Ce:GAGG-2.5 and Ce:GAGG-3) crystals were grown by the Czochralski process in order to reduce cost of the starting materials as compared with conventional Ce:$Gd_3Al_2Ga_3O_{12}$ (Ce:GAGG-2) crystal which have high light output. Although perovskite phase was detected in Ce:GAGG-3, Ce:GAGG-2.5 had single-phase garnet structure. Solidification fraction for the Ce:GAGG-2.5 growth was 0.52. Optical properties including transmittance, emission, and excitation spectra of 30 samples cut from the Ce:GAGG-2.5 bulk ingot did not depend on their original position along the growth axis. These samples had light outputs of approximately 58,000 ± 3000 photons/MeV. However, scintillation decay times varied from 140 to 200 ns and depended on the position clearly.

1. Introduction

Many oxide scintillators such as Ce:$(Lu,Y)_2SiO_5$ (Ce:LYSO) and $Bi_4Ge_3O_{12}$ (BGO) are not hygroscopic and demonstrate gamma-ray stopping power greater than that of halide scintillators as Ce:$LaBr_3$ and Tl:NaI [1–7]. On the other hand, corresponding oxide crystals have low light output and energy resolution when compared with the halides. However, Ce-doped $Gd_3Al_2Ga_3O_{12}$ scintillator has good light output of 46,000 photon/MeV [8]. It is non-hygroscopic, and it has high stopping power exceeding that observed in the halide group. Thus, the Ce:$Gd_3Al_2Ga_3O_{12}$ crystals are expected to be used in various highly demanded applications including radiation monitoring and medical imaging. Following these circumstances, development of the Czochralski (Cz) process for the growth of this type crystals is required to establish their mass production.

Ce:$Gd_3(Al_xGa_{1-x})_5O_{12}$ ($0<x<1$) crystals belong to the garnet structural family. Their optical and scintillation properties had been examined for only Ce:$Gd_3Al_2Ga_3O_{12}$ ($x=2/5$) crystal that was grown by the Cz process. Other crystals having different x ratios and grown by the Cz process have not been yet reported. Ga_2O_3 is more expensive than Al_2O_3. Therefore, purpose of the current project was to estimate maximum substitution of Ga with Al achievable in the Ce:$Gd_3(Al_xGa_{1-x})_5O_{12}$ crystals to reduce cost of the starting materials and the crystals. Moreover, the effective atomic number of Ce:$Gd_3(Al_xGa_{1-x})_5O_{12}$ ($2/5<x<4/5$) is similar to that of Ce:$Gd_3Al_2Ga_3O_{12}$, and gamma-ray stopping power is also similar. In this report, growth of these crystals with $x=2.5/5$ and 3/5 using the Cz process is demonstrated together with evaluation of their optical and scintillation properties such as light outputs and scintillation decay times.

2. Materials and experimental methods

The $(Ce_{0.01},Gd_{0.99})_3(Al_xGa_{1-x})_5O_{12}$ crystals ($x=2.5/5$ and 3/5) were grown by the Cz process from the melt produced from 99.99% pure starting oxides of α-Al_2O_3, β-Ga_2O_3, Gd_2O_3, and CeO_2. The growth was performed from Ir-crucible on the seed made of $Gd_3Al_2Ga_3O_{12}$ single crystal. The pulling and rotation rates were 0.8 mm/h and 12 rpm, respectively. The growth atmosphere was Ar (98%)+O_2(2%) mixture.

The chemical composition of the as grown crystals and distribution of their constituents along and perpendicular to the growth axis were examined using the electron probe microanalyzer (EPMA) (JXA-8621MX, JEOL). The measurements were made under accelerating voltage of 20 kV and current of 1.2×10^{-8} A.

The transmittance and reflectance spectra were recorded with spectrophotometers: V-530 (JASCO) and UV-2550 (SHIMADZU), respectively. Photo-luminescence (PL) spectra at room temperature were examined with a spectrofluorometer (FLS920, Edinburgh Instrument) using a Xenon lamp as an excitation source. Moreover,

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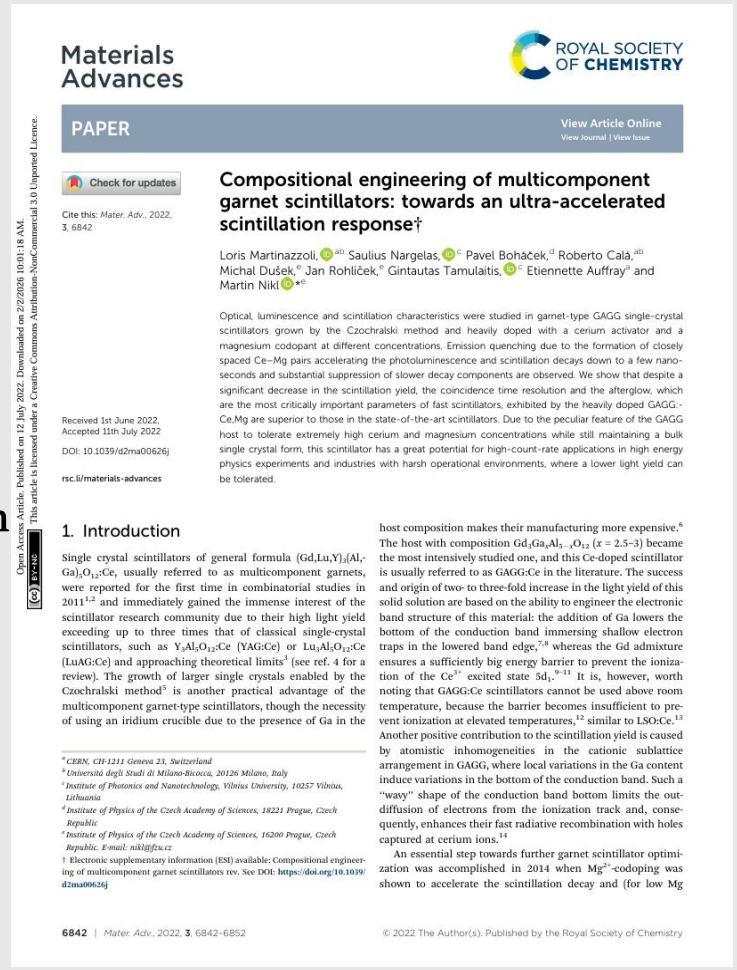
http://dx.doi.org/10.1016/j.jcrysgro.2013.10.059

Add Mg

Magnesium



Materials Advances

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Cite this: Mater. Adv., 2022, 3, 6842

Compositional engineering of multicomponent garnet scintillators: towards an ultra-accelerated scintillation response†

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Optical, luminescence and scintillation characteristics were studied in garnet-type GAGG single-crystal scintillators grown by the Czochralski method and heavily doped with a cerium activator and a magnesium codopant at different concentrations. Emission quenching due to the formation of closely spaced Ce–Mg pairs accelerating the photoluminescence and scintillation decays down to a few nanoseconds and substantial suppression of slower decay components are observed. We show that despite a significant decrease in the scintillation yield, the coincidence time resolution and the afterglow, which are the most critically important parameters of fast scintillators, exhibited by the heavily doped GAGG:Ce,Mg are superior to those in the state-of-the-art scintillators. Due to the peculiar feature of the GAGG host to tolerate extremely high cerium and magnesium concentrations while still maintaining a bulk single crystal form, this scintillator has a great potential for high-count-rate applications in high energy physics experiments and industries with harsh operational environments, where a lower light yield can be tolerated.

Received 1st June 2022,
Accepted 11th July 2022

DOI: 10.1039/d2ma00626j

rsc.li/materials-advances

1. Introduction

Single crystal scintillators of general formula $(Gd,Lu,Y)_3(Al,Ga)_5O_{12}$:Ce, usually referred to as multicomponent garnets, were reported for the first time in combinatorial studies in 2011[1,2] and immediately gained the immense interest of the scintillator research community due to their high light yield exceeding up to three times that of classical single-crystal scintillators, such as $Y_3Al_2O_{12}$:Ce (YAG:Ce) or $Lu_3Al_2O_{12}$:Ce (LuAG:Ce) and approaching theoretical limits[3] (see ref. 4 for a review). The growth of larger single crystals enabled by the Czochralski method[5] is another practical advantage of the multicomponent garnet-type scintillators, though the necessity of using an iridium crucible due to the presence of Ga in the host composition makes their manufacturing more expensive.[6] The host with composition $Gd_3Ga_xAl_{5-x}O_{12}$ (x = 2.5–3) became the most intensively studied one, and this Ce-doped scintillator is usually referred to as GAGG:Ce in the literature. The success and origin of two- to three-fold increase in the light yield of this solid solution are based on the ability to engineer the electronic band structure of this material: the addition of Ga lowers the bottom of the conduction band immersing shallow electron traps in the lowered band edge,[7,8] whereas the Gd admixture ensures a sufficiently big energy barrier to prevent the ionization of the Ce^{3+} excited state $5d_1$.[9–11] It is, however, worth noting that GAGG:Ce scintillators cannot be used above room temperature, because the barrier becomes insufficient to prevent ionization at elevated temperatures,[12] similar to LSO:Ce.[13] Another positive contribution to the scintillation yield is caused by atomistic inhomogeneities in the cationic sublattice arrangement in GAGG, where local variations in the Ga content induce variations in the bottom of the conduction band. Such a "wavy" shape of the conduction band bottom limits the out-diffusion of electrons from the ionization track and, consequently, enhances their fast radiative recombination with holes captured at cerium ions.[14]

An essential step towards further garnet scintillator optimization was accomplished in 2014 when Mg^{2+}-codoping was shown to accelerate the scintillation decay and (for low Mg

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† Electronic supplementary information (ESI) available: Compositional engineering of multicomponent garnet scintillators rev. See DOI: https://doi.org/10.1039/d2ma00626j

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2. L. Martinazzoli, et al., Mater. Adv., 2022, 3, 6842.

Sample	Light output (ph MeV^{-1})	Commercial sample	Light output (ph MeV^{-1})
PL1	12 000 ± 600	GFAG C&A	20 500 ± 1000
PL2	7600 ± 380	GAGG Fomos	28 300 ± 1400
PL3	5200 ± 260	GAGG ILM	21 000 ± 1100
PL4	3900 ± 190		
PL5	2300 ± 230		
PL6	500 ± 300		

Sample	τ_r (ps)	τ_{d1} (ns)	R_1 (%)	τ_{d2} (ns)	R_2%	τ_{d3} (ns)	R_3 (%)	$\tau_{d,eff}$ (ns)
C&A GFAG	32	6.0	4.6	44.5	69.2	222	26.3	41
ILM GAGG	37	4.0	3.2	40.4	56.4	138	40.4	40
Fomos GAGG	30	2.2	0.5	53.1	41.7	166	57.8	73
PL1	13	2.5	3.3	25.4	48.0	79.2	48.8	26
PL2	8	2.1	7.2	16.6	54.6	66.2	38.2	13.8
PL3	5	1.6	6.2	12.6	47.5	46.0	46.3	11.6
PL4	5	1.5	9.2	11.3	53.9	45.4	36.9	8.6
PL5	5	1.0	11.0	7.1	51.8	40.8	37.2	5.2
PL6	5	0.2	19.5	1.5	53.0	14.9	27.5	0.7

- LY=58000±3000 ph/MeV
- Decay=200 ns

- LY=500±300 ph/MeV
- Decay=0.2, 1.5, 14.9 ns

3.2 Decay—Ce doping concentration

X.Y. Sun, et al., Chin. Opt. Lett., 2025, 23(12), 121602.

CHINESE OPTICS LETTERS Vol. 23, No. 12 | December 2025

Ultra-high concentration Ce^{3+}-doped gadolinium-based borosilicate glass scintillators

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Received May 14, 2025 | Accepted July 16, 2025

Ce^{3+}-doped gadolinium-based borosilicate ($GBSC_x$) glass scintillators with an ultra-high concentration of 16 mol% were synthesized in ambient atmosphere for future calorimeter application. The valence state of Ce was preciously controlled in the glass by the X-ray absorption near edge structure (XANES) spectrum. With the increased Ce^{3+} concentration, the NBO/BO ratio decreases notably from 5.15 to 0.56. The $GBSC_x$ glass scintillators exhibit the broad photoluminescence (PL) band within 350–550 nm regions, with a maximum PL quantum yield (PL QY) of 60.6%. In X-ray excited luminescence (XEL), the integral intensity of the GBSC2 glass is 18.4% compared to the BGO crystal. Meanwhile, it has the highest light yield of 1043 ph/MeV with an energy resolution of 28.4% at 662 keV under γ-ray excitation. When the doped concentration of Ce^{3+} exceeds 4 mol%, the proportion of light yield within 1 μs integral gate exceeds 95%, which conforms to the requirement of fast time response. Interestingly, the concentration quenching effect of high concentration Ce^{3+} ($x \leq 14$) does not occur in the glass scintillators under γ-ray excitation. With the increase of Ce^{3+} concentration, both the fast (100–18 ns) and slow (1000–59 ns) components of scintillation decay time decrease dramatically. Therefore, the developed $GBSC_x$ glass scintillators, featured with the reasonable light yield and fast time response, have a promising application in future high energy physics (HEP) experiments.

Keywords: HEP experiment; glass scintillator; XANES; light yield; fast scintillation decay.
DOI: 10.3788/COL202523.121602

1. Introduction

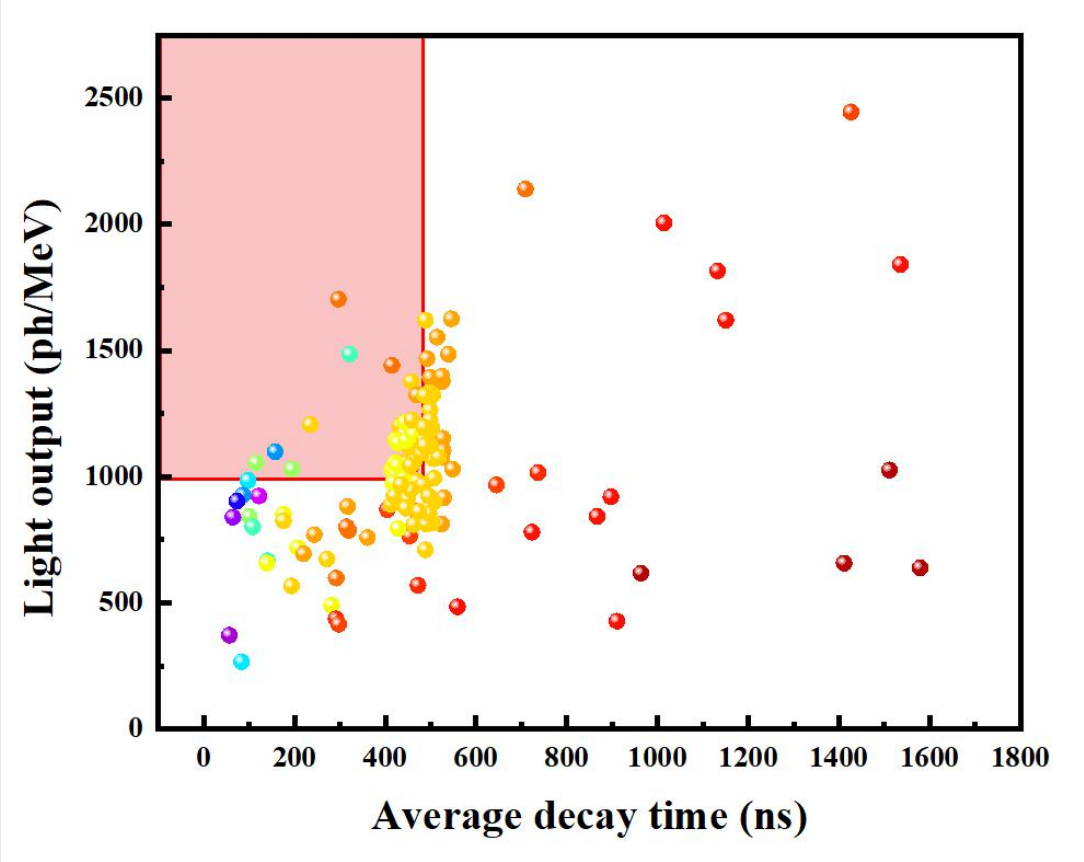
In recent decades, the purpose of scintillation materials for detectors and calorimeter systems has been a major factor in particle physics, nuclear medicine, and even astrophysics fields[1–3]. Currently, glass scintillators are targeted as static X-ray imaging [4–6], as they only pursue higher light yield with less care for scintillation time and glass density. However, HEP experiments will require a comprehensive advantage of scintillation materials, including low cost, high density, adequate energy resolution, and fast decay. It is still difficult to achieve

➢ 2025 Chin. Opt. Lett.
Different concentrations of Ce doping

	Density (g/cm³)	Light yield (ph/MeV)	Scintillation decay time (ns)
GFO1	6.1400	427	98.9 (1.6%), 924.2
GFO2	6.0494	1043	98.6 (8.2%), 490.7
GFO5	5.9736	825	82.0 (27.5%), 211.5
GFO9	5.9332	985	35.8 (10.7%), 105.1
GFO14	5.9152	839	25.3 (10.3%), 69.0
GFO16	5.8917	371	18.9 (7.2%), 59.7

- Optimizing the decay time of GS still relies on adjusting the concentration of Ce^{3+} centers.
- As the Ce doping concentration increases, the scintillation decay time gradually decreases.
- However, the light output first increases and then decreases.
- **It is necessary to balance light yield and decay time.**

Relationship between decay and light output



- Over the past four years, the GS group has prepared more than 1500 pieces of glass;
- Decay time of the GS, determined by physical properties: 100ns -> 300ns -> 500ns;
- Typical results show that decay time and light yield tend to be positively correlated: a higher light yield generally implies a slower decay time;
- Only a small number of GS samples meet all three performance criteria: density > 6.0 g/cm^3, light yield > 1000 ph/MeV, and decay time < 500 ns.

We really need the decay time of GFO less than 500ns? What about 1us? or others?

3.3 Peak emission wavelength shift

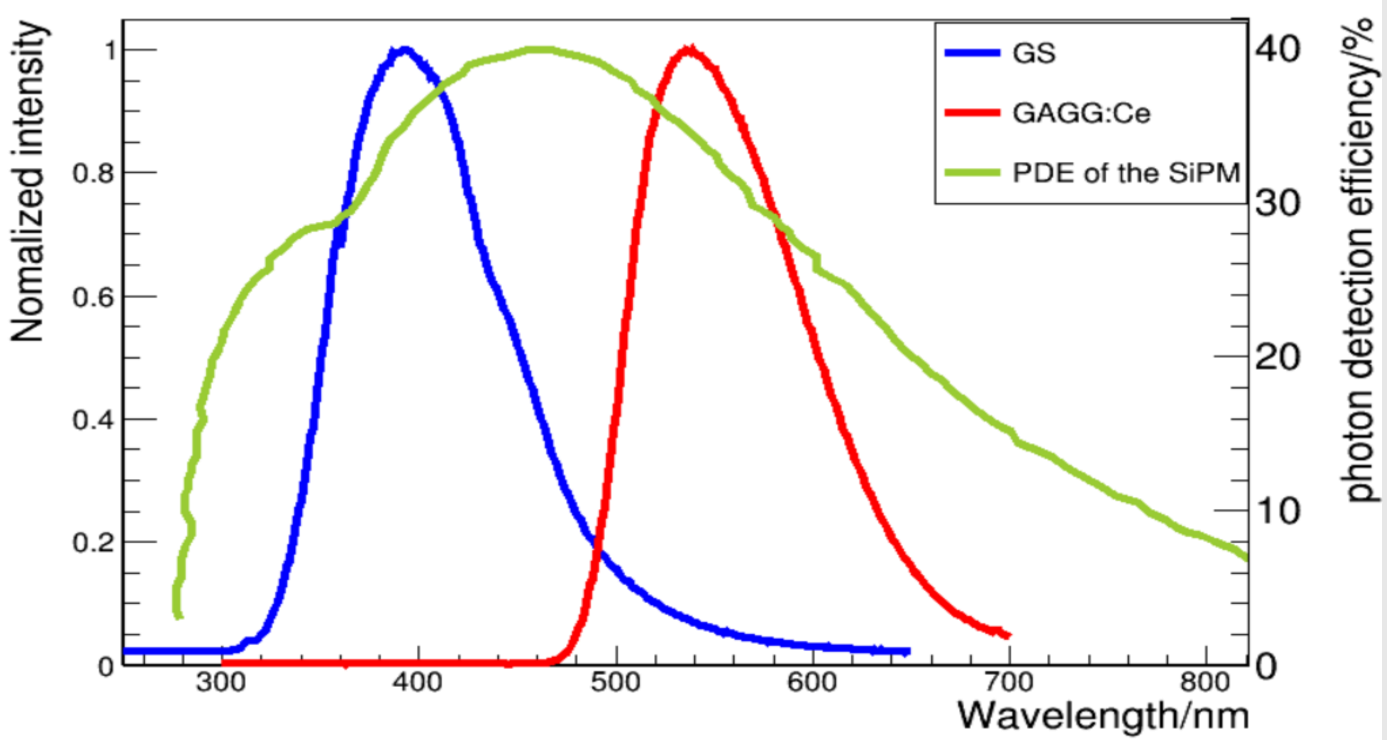
light yield (LY) in number of photons/MeV produced (N_{photons}/MeV) and the light output (LO) in number of photoelectrons/MeV detected involves the factors for the light collection efficiency (LCE) and the quantum efficiency (QE) of the photodetector:

$$LO = LY \cdot LCE \cdot QE. \tag{35.4}$$

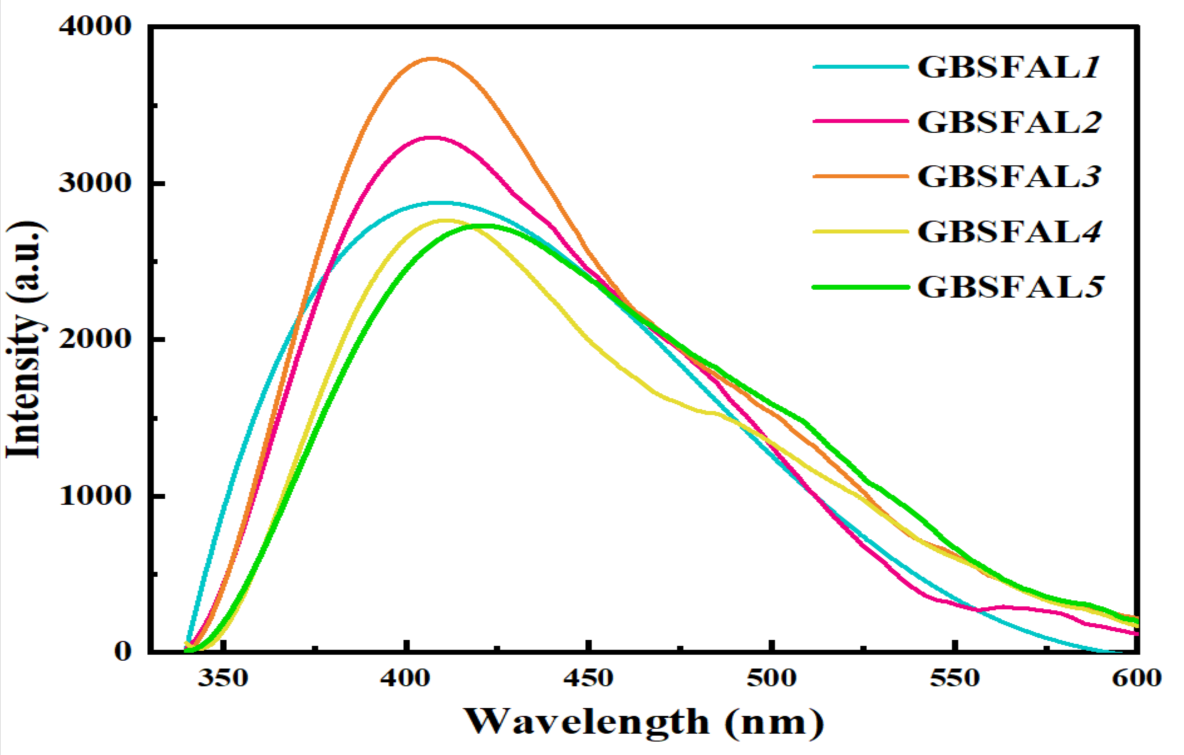
Cite by R.H. Mao, L.Y. Zhang and R.-Y. Zhu, IEEE Trans.NS55, 2425 (2008).

Enhancing the measured light output:

- Improving light collection efficiency
- Matching with the QE of photodetectors



3.3 Wavelength shift of GS（Unpublished）



Doping X into GFO Glass

	XEL peak (nm)	LO (ph/MeV)	ER (% @662keV)
GBSFAL1	407	1271	29.8
GBSFAL2	408	1360	26.3
GBSFAL3	409	1491	26.9
GBSFAL4	411	1116	29.7
GBSFAL5	420	1252	28.1

- After adding X^+, the glass XEL peak red-shifts from 390 nm to approximately 420 nm;
- As a strong network modifier, X^+ increases the coordination field around Ce^{3+}, lowers its luminescence energy levels, and ultimately leads to a redshift of the XEL peak and an increase in light output;
- However, excessive X^+ can disrupt the glass network structure and reduce the light yield.

3.4 Ion co-doping—$Cs_3Cu_2I_5$:Tl（CCI crystal）

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Zero-Dimensional $Cs_3Cu_2I_5$ Perovskite Single Crystal as Sensitive X-Ray and γ-Ray Scintillator

*Shuangliang Cheng, Alena Beitlerova, Romana Kucerkova, Martin Nikl, Guohao Ren, and Yuntao Wu**

Low-dimensional halide perovskites with highly efficient luminescent properties have attracted huge attention in optoelectronic and radiation detection applications in recent years. Herein, a highly efficient 0D $Cs_3Cu_2I_5$ perovskite crystal is presented for X-ray and γ-ray detection that can be easily grown from the melt by the Bridgman method. The crystal structure of $Cs_3Cu_2I_5$ belongs to an orthorhombic system with a space group of *Pnma*. The $Cs_3Cu_2I_5$ crystal is nonhygroscopic and self-absorption free due to a large Stokes shift of 120 nm originated from its self-trapped exciton emission. Its scintillation emission centers at 440 nm with a principal decay time of 967 ns. The $Cs_3Cu_2I_5$ crystal not only demonstrates a high scintillation yield of about 32 000 photons MeV^{-1} and an extremely low afterglow of 0.03% at 10 ms under X-ray radiation, but also possesses a high light yield of 29 000 photons MeV^{-1} with an excellent energy resolution of 3.4% at 662 keV under γ-ray radiation.

Organic–inorganic hybrid halide perovskites, represented by $CH_3NH_3PbX_3$ (X = Cl, Br, I), have demonstrated excellent optoelectronic and radiation detection properties.[1–3] All-inorganic $CsPbX_3$ perovskites (X = Cl, Br, I) with better long-term stability are also considered as promising materials for optoelectronic devices and semiconductor γ-ray detectors.[4–6] Recently, there has been a surge of interest in low-dimensional perovskites due to their high photoluminescence quantum yield (PLQY). For example, the crystal structure of Cs_4PbBr_6 that consist of spatially isolated $[PbBr_6]^{4-}$ octahedra surrounding with Cs^+ ions can be regarded as a 0D structure at molecular level, which leads to an intense quantum confinement effect. Excitons are strongly confined at each $[PbBr_6]^{4-}$ octahedron, enabling a high exciton binding energy of 353 meV and a high PLQY of between 42% and 45%.[7,8]

Quite a few low-dimensional all-inorganic perovskites with remarkable luminescent properties were also reported as sensitive and efficient scintillators. Bulk crystals of 0D Cs_4CaI_6:Eu, Cs_4SrI_6:Eu, and Cs_4EuBr_6 perovskites, isostructural to K_4CdCl_6 trigonal system, have excellent light yields from 51 800 to 78 000 photons MeV^{-1} and energy resolutions from 3.3% to 4.3% at 662 keV.[9,10] However, due to a small Stokes shift of Eu^{2+} ions, these materials suffer from strong self-absorption effect when scaling-up crystal size. Nanocrystals of 0D $CsPbBr_3/Cs_4PbBr_6$ perovskites show a high light yield of 64 000 photons MeV^{-1} and a fast decay time of <10 ns.[11] One-dimensional materials were also reported as sensitive X-ray scintillators, such as Rb_2CuBr_3 and Rb_2CuCl_3.[12,13] In particular, the Rb_2CuBr_3, that is self-absorption free and nonhygroscopic, was reported to achieve an ultrahigh scintillation yield of 90 000 photons MeV^{-1}.[12]

All-inorganic 0D perovskite $Cs_3Cu_2I_5$ was recently reported as a highly efficient blue-emitting material with a PLQY of 91.2%, and regarded as promising for application in photodetectors, light-emitting diodes, and memristors afterward.[14–17] In 2020, $Cs_3Cu_2I_5$ nanocrystals were developed for X-ray imaging with a light yield of 80 000 photons MeV^{-1}.[18] To the best of our knowledge, the X-ray and γ-ray detection capability of bulk $Cs_3Cu_2I_5$ single crystal has not been reported. Thus, the aim of this work is to study the physical and optical properties and the scintillation performance under X-ray and γ-ray radiation of high-quality $Cs_3Cu_2I_5$ perovskite single crystal grown by the Bridgman method.

The 7 mm diameter single crystal of $Cs_3Cu_2I_5$ was grown by the vertical Bridgman method. High-purity powders of CsI (99.99%, Grirem Advanced Materials) and CuI (99.999%, Sigma-Aldrich) were used as raw materials. These starting materials were mixed consistent with stoichiometric ratio and loaded into a quartz ampoule in a glovebox with <0.1 ppm moisture and oxygen. The loaded ampoule was sealed under a vacuum of 10^{-6} torr after drying at 180 °C for 16 h, and then placed into a Bridgman growth furnace. Prior to growth, the $Cs_3Cu_2I_5$ compound was synthesized under 630 °C (a few degrees above the melting points of all components) for 24 h. Then the growth process started at 390 °C[19] with a temperature gradient of 20–30 °C cm^{-1} and a translation rate of 0.5 mm h^{-1}. The furnace

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DOI: 10.1002/pssr.202000374

- LY=28000 photons/MeV
- E.R=3.4%@662KeV
- Decay=967 ns

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Highly Resolved X-Ray Imaging Enabled by In(I) Doped Perovskite-Like $Cs_3Cu_2I_5$ Single Crystal Scintillator

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Low-dimensional perovskite halides have shown a great potential as X-ray detection materials because of efficient exciton emissions originating from strongly spatially localized charge carriers. Nonetheless, most of them have a scintillation yield far below their theoretical limits. Here, it is found that the harvesting efficiency of produced charge carriers can be significantly enhanced via a small amount of In^+ doping in these highly localized structures. A bright and sensitive zero-dimensional $Cs_3Cu_2I_5$:In^+ halide with efficient and tunable dual emission is reported. The radioluminescence emission of $Cs_3Cu_2I_5$:In^+ crystals under X-ray excitation consists of a self-trapped exciton emission at 460 nm and an In^+-related emission at 620 nm at room temperature. In^+ doping enhances the photoluminescence quantum efficiency (PLQY) of $Cs_3Cu_2I_5$ from 68.1% to 88.4%. Benefiting from the higher PLQY, $Cs_3Cu_2I_5$:In^+ can achieve an excellent X-ray detection limit of 96.2 nGy_{air} s^{-1}, and a superior scintillation yield of 53 000 photons per MeV, which is comparable to commercial CsI:Tl single crystals. As a result, a remarkable X-ray imaging resolution of 18 line pairs mm^{-1} is demonstrated, which is so far a record resolution for single crystal perovskite-based flat-panel detectors. These results highlight the importance of efficient harvesting of carriers (and excitons) in low-dimensional perovskites for radiation detection applications.

1. Introduction

Metal halide perovskites are a promising platform for photovoltaics,[1] light-emitting applications,[2] photodetectors,[3] lasers,[4] and other optoelectronic applications owing to their extraordinary optoelectronic properties.[5] In particular, halide perovskites also show huge potential in scintillator-based high energy radiation detection due to their high effective atomic number (Z_{eff}) component and near-unity luminescence efficiency.[6] Since the first report on lead halide perovskite nanocrystal scintillators by Liu's group,[7] many efforts have been devoted to exploring the application of halide perovskite scintillators for radiation detection.[8,9] However, though the halide perovskite scintillators are highly promising, the light yield (LY, ~21 000 photons per MeV) is still far behind the commercial scintillators such as CsI:Tl (54000 photons per MeV), Gd_2O_2S:Tb (60000 photons per MeV).[8,10] Also, the stability of lead halide perovskite is still a great challenge for practical applications.[11]

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The ORCID identification number(s) for the author(s) of this article can be found under https://doi.org/10.1002/adom.202200304.
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DOI: 10.1002/adom.202200304

- LY=44000 photons/MeV
- Decay=3746.5 ns

Add Tl
Thallium

Nuclear Inst. and Methods in Physics Research, A 991 (2021) 164963

Contents lists available at ScienceDirect

Nuclear Inst. and Methods in Physics Research, A

journal homepage: www.elsevier.com/locate/nima

Crystal growth and scintillation properties of pure and Tl-doped $Cs_3Cu_2I_5$ ☆

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ARTICLE INFO

Keywords:
Bridgman technique
Single crystal growth
Scintillator
Halides

ABSTRACT

In this work the Bridgman crystal growth and scintillation properties of both undoped and Tl-doped $Cs_3Cu_2I_5$ are presented. This material is very attractive for gamma and X-ray detection applications, with a density of 4.53 g/cm³ and effective atomic number of 51.9. Undoped $Cs_3Cu_2I_5$ had a light yield of 41,500 photons/MeV, with an energy resolution of 4.4% at 662 keV. Thallium doping at 0.5 mol % resulted in a much-improved scintillation response, in which light yield increased to 98,200 photons/MeV and energy resolution reduced to 3.3% at 662 keV. The X-ray excited emission is centered at 442 nm for the undoped and 500 nm for the Tl-doped crystals. The undoped emission is broad, typical of excitonic emission, while thallium doping results in an even broader band with features of both the undoped and thallium defect-mediated emissions.

1. Introduction

The dearth in availability of high-performance, low cost scintillators has led to ongoing research by many groups in the search for new scintillators [1,2]. At the University of Tennessee, we have an active scintillator discovery program for materials for the detection of ionizing radiation. High density, high scintillation light yield, and good energy resolution are important for gamma-ray spectroscopy applications. In this work we are focusing on a promising new scintillator family, the alkali copper halides, specifically $Cs_3Cu_2I_5$. The existence of this material has been known for many years, and some of the properties have been investigated. Bigalke et al. synthesized $Cs_3Cu_2I_5$ in 1988, using both solid state synthesis and an organic solvent method, and reported that it has an orthorhombic structure with space group *Pbnm* [3]. Phase diagrams were published by Wojakowska, et al. in the early 2000s [4,5], reporting that the compound melts incongruently. The properties of polycrystalline powders and thin films have been reported [6–8] as have the properties of nanocrystals. Cheng, Lian, and Luo, et al. have all reported that undoped nanocrystals have a self-trapped exciton (STE) emission, photoluminescence and radioluminescence peaks around 445 nm, and a photoluminescence lifetime of about 1.9 μs; the bandgap was reported to be 4.40 eV [9–11].

There has been some interest in using this material in optoelectronic devices. Jun, et al. reported fabrication of a blue light-emitting diode (LED) as well as preparing white luminescence films by mixing $Cs_3Cu_2I_5$ with a yellow phosphor [12]. Zhang, et al. reported the use of a solvent evaporation crystallization technique to produce millimeter-size single crystals and powders with strong blue photoluminescence and quantum yield of up to 89% (crystal) and 60% (powder), with the aim of producing LEDs and fluorescent inks [13]. Mn^{2+}-activated yellow-emitting $Cs_3Cu_2I_5$ phosphors have been studied for use in white LEDs [14], while a $Cs_3Cu_2I_5$ thin film has been used as a component of a deep ultraviolet photodetector [15].

These reports suggest promise for $Cs_3Cu_2I_5$, as does its density of 4.53 g/cm³ and effective atomic number of 51.9 [8]. It has long been established that the performance of CsI is significantly improved by doping with thallium [16–21]. Our hypothesis, the basis for this current work, was that doping $Cs_3Cu_2I_5$ with thallium would result in similarly enhanced performance. In this manuscript we have established that $Cs_3Cu_2I_5$ single crystals are intrinsic scintillators, although they lack the high light yield and excellent energy resolution required for spectroscopy, and that doping $Cs_3Cu_2I_5$ with thallium does indeed result

☆ This manuscript has been co-authored by UT-Battelle, LLC under Contract No. DE-AC05-00OR22725 with the, US Department of Energy. The United States Government retains and the publisher, by accepting the article for publication, acknowledges that the United States Government retains a non-exclusive, paid-up, irrevocable, world-wide license to publish or reproduce the published form of this manuscript, or allow others to do so, for United States Government purposes. The Department of Energy will provide public access to these results of federally sponsored research in accordance with the DOE Public Access Plan (http://energy.gov/downloads/doe-public-access-plan).

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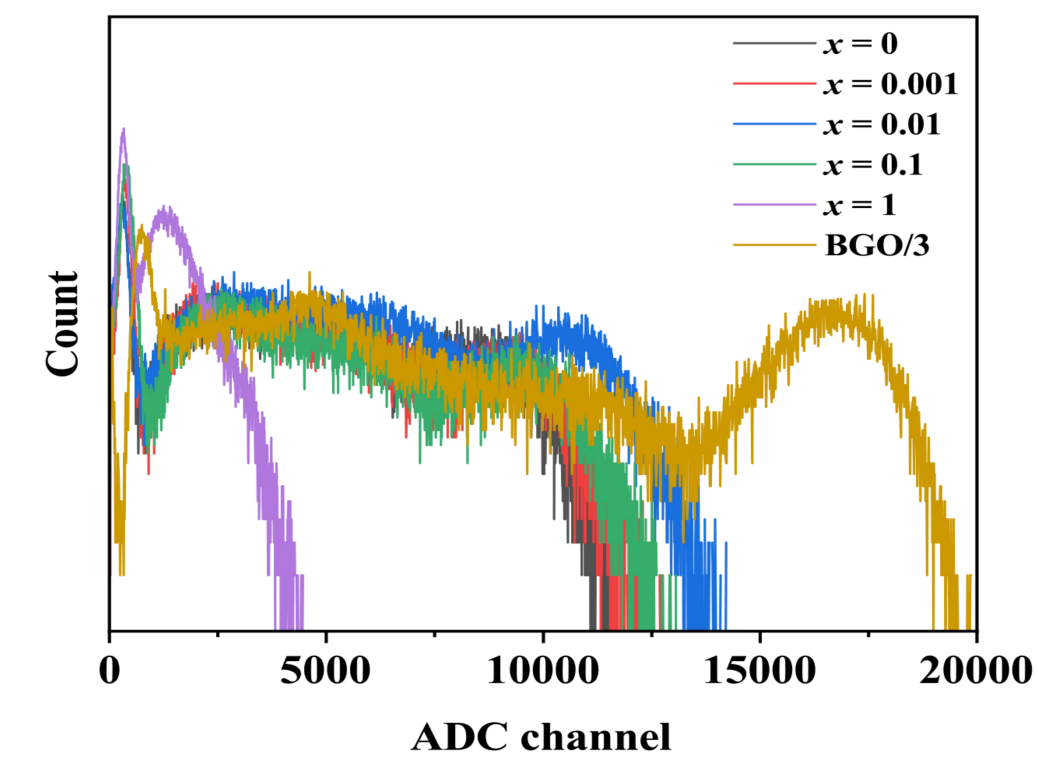
https://doi.org/10.1016/j.nima.2020.164963
Received 24 August 2020; Received in revised form 9 December 2020; Accepted 11 December 2020
Available online 15 December 2020
0168-9002/

- LY=98200 photons/MeV
- E.R=3.3%@662KeV
- Decay=840 ns

3.4 GS—co-doped with Ce and Y (Unpublished)



Doping Y into GFO Glass

	LY (ph/MeV)	ER (%@662keV)
1Ce+0Y	1185	40.6
1Ce+0.001Y	1301	33.8
1Ce+0.01Y	1485	31.5
1Ce+0.1Y	1350	34.8
1Ce+1Y	331	75.2

- Incorporating additional luminescent centers can reduce the non-radiative recombination between self-trapped excitons and glass defects, offering a potential approach to enhance the light yield of the GS.
- By adding a trace amount of Y^{+}, the light yield increased from 1200 to ~1500 ph/MeV.

Summary

- 1. High Light Yield Enhancement is possible!
- 2. We try our best to Enhance the Light Yield;
  - Density: **We really need the density of GFO larger than 6g/cc?**
  - Decay Time: **We really need the decay time of GFO less than 500ns?**
  - Wavelength Shift:
  - Co-doping: We need time and funding to find the new materials;
- 3. How to conrol the cost of GFO Glass.


N2+H2→NH3
element
COOH
CH3
e=mc²
See the unseen
change the unchanged
The Innovation

THANKS