Phys. Status Solidi A 208, No. 12, 2830–2832 (2011) / DOI 10.1002/pssa.201127365

physica status solidi a
www.pss-a.com
applications and materials science

Luminescence and scintillation of Ce^{3+}-doped oxide glass with high Gd_2O_3 concentration

W. Chewpraditkul*,[1], X. He[2,3], D. Chen**,[2], Y. Shen[2], Q. Sheng[2], B. Yu[3], M. Nikl[4], R. Kucerkova[4], A. Beitlerova[4], C. Wanarak[1], and A. Phunpueok[1]

[1] Department of Physics, King Mongkut's University of Technology Thonburi, Bangkok 10140, Thailand
[2] Shanghai Institute of Optics and Fine Mechanics, CAS, Shanghai 201800, P.R. China
[3] Department of Physics, Shanghai University, Shanghai 201800, P.R. China
[4] Institute of Physics, AS CR, Cukrovarnicka 10, 16253 Prague, Czech Republic

Received 22 June 2011, revised 29 July 2011, accepted 5 August 2011
Published online 1 September 2011

Keywords Ce^{3+}, light yield, luminescence, oxide glasses, scintillation, time-resolved luminescence

* Corresponding authors: e-mail weerapong.che@kmutt.ac.th, Phone: +66 2470 8944, Fax: +66 2470 8900
** e-mail d-chen@mail.siom.ac.cn, Fax: 86 2159911975

Ce^{3+}-doped oxide glass with high Gd_2O_3 (30 mol%) concentration was prepared as a scintillating glass under CO reducing atmosphere. A dominant emission band of the Ce^{3+} 5d → 4f transition peaking around 400 nm was observed in the photo- and radioluminescence (PL, RL) spectra. PL decay was governed by a few tens of nanoseconds decay time. The integral scintillation efficiency of about 30% of the $Bi_4Ge_3O_{12}$ scintillator was observed as well. It exhibits a light yield of about 910 photons/MeV with an energy resolution of 18.3% (FWHM) in the pulse height spectrum of 662 keV γ-rays from a ^{137}Cs source.

1 Introduction Scintillating glasses can be used for the detection of X-rays, γ-rays, and neutrons [1–3]. They are cheaper and easier to fabricate with respect to single crystal materials, but they suffer from inefficient energy transfer and concentration quenching at higher content of emission activator ions. In phosphate glasses the former problem was overcome by embedding the energy guiding sublattice made by high concentration of Gd^{3+} ions [2]. Nevertheless, the energy transfer was finally classified as phonon assisted [4] due to crystalline field fluctuation in glass matrix. Consequently, scintillation response contained a large amount of slow components. Scintillating glasses are required to have high light yield, high density, short decay time and good radiation hardness. To increase the density for effective detection of X-/γ-rays, Ce^{3+}-doped dense glasses with fast decay time were investigated using high concentration of Gd_2O_3 at levels up to 30 mol% in the glass matrices [2, 5–7].

This paper provides the photo- and radioluminescence (PL, RL) characteristics of Ce^{3+}-doped glass containing of 30 mol% Gd_2O_3 in the oxide glass matrix. The scintillation response under excitation with 662 keV γ-rays from a ^{137}Cs source is measured and compared to the response of the $Bi_4Ge_3O_{12}$ (BGO) single crystal scintillator.

2 Experimental The composition of the glass prepared in this work was $15SiO_2$–$30B_2O_3$–$25Al_2O_3$–$30Gd_2O_3$ (mol%), and doped with CeO_2 (1.0 wt%). The starting materials were reagent-grade SiO_2, H_3BO_3, $Al(OH)_3$, Gd_2O_3, and CeO_2. Powders of starting materials were mixed homogeneously in an agate mortar and melted in an alumina crucible under CO reducing atmosphere at 1580 °C for 1 h. The melt was poured onto a preheated stainless steel plate and then annealed at 600 °C for 4 h followed by cooling in the furnace at its natural cooling rate to ambient temperature. Polished plate of about $7 \times 7 \times 1\ mm^3$ was used for all measurements. The glass density was 4.70 g/cm^3 determined by Archimedes method.

The excitation and emission spectra were recorded by a Hitachi F-2500 fluorescence spectrophotometer equipped with a 150 W xenon lamp source. RL spectra and PL decays were obtained using the custom made 5000M fluorometer, Horiba Jobin Yvon, equipped with X-ray tube (40 kV, Mo anode) and pulsed (ns) hydrogen flashlamp as excitation sources, respectively, for details see Ref. [8].

Photoelectron yield, expressed as a number of photoelectrons per MeV (phe/MeV) of absorbed γ-photon, and energy resolution were measured by a Photonis XP5200B PMT under the excitation with 662 keV γ-rays from a ^{137}Cs source. The measurements were carried out with 4 μs shaping time constant in a spectroscopy amplifier. The PC-based multichannel analyzer (Tukan 8k MCA) was used to record pulse height spectra. The light yield, expressed

as a number of photons per MeV (ph/MeV) of absorbed γ-photon, was determined by relating the response of Ce^{3+}-doped Gd glass to 662 keV photopeak to the response of a reference BGO crystal (light yield of ~8 500 ph/MeV), and by taking into account the photocathode quantum efficiency of PMT for BGO crystal and Ce^{3+}-doped Gd glass emission wavelengths.

3 Results and discussion Absorption and excitation spectra for the Ce^{3+}-doped Gd glass at room temperature (RT) are given in Fig. 1. The broad bands (230–375 nm) are attributed to the 4f → 5d transitions of the Ce^{3+} ions admixed with $^8S_{7/2} \rightarrow {}^6I_J, {}^6P_J$ transitions of the Gd^{3+} ions. Energy transfer from higher lying states (^6I_J, $^6P_{7/2}$) of Gd^{3+} to lower lying 5d states of Ce^{3+} can further strengthen emission intensity of Ce^{3+}. In the absorption spectrum, the absorption edge located around 375 nm is due to the 4f → $5d_1$ transition of Ce^{3+}. The charge transfer absorption band of Ce^{4+} is located at longer wavelength with an absorption edge around 450 nm in case when the glass is made under an air atmosphere [7]. As it is absent in Fig. 1 it indicates that Ce^{4+} ions efficiently transform into Ce^{3+} ions when glass melts under the CO reducing atmosphere.

Characteristic broad emission band of Ce^{3+} is peaking around 400 nm under 300 nm excitation, but it is worth to note that the band is inhomogeneously broadened. This is rather typical situation of Ce^{3+} center in glassy environment. RL spectra of both Ce^{3+}-doped Gd glass and BGO standard sample at RT are given in Fig. 2. The samples have about the same shape and the measurements were performed in closely similar conditions, so that the spectra can be compared in an absolute way. The integral scintillation efficiency (integral of RL spectra) of the Ce^{3+}-doped Gd glass sample is ~30% of the BGO standard sample.

The PL decay at 386 nm of the Ce^{3+}-doped Gd glass sample under excitation with $\lambda_{ex} = 310$ nm is given in Fig. 3(a). Its double-exponential approximation, $I(t) = \Sigma A_i \exp(-t/\tau_i) + \text{background}$, $i = 1,2$, yields the decay times $\tau_1 = 28.3$ ns and $\tau_2 = 165$ ns with respective component intensities $I_i = (A_i\tau_i)/(\Sigma A_i\tau_i)$, namely, $I_1 = 92\%$ and

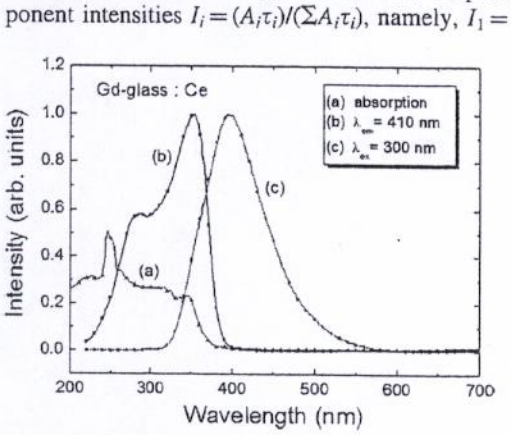


Figure 1 (online color at: www.pss-a.com) Absorption (a), excitation (b) and emission (c) spectra of Ce^{3+}-doped Gd glass sample as measured at RT.

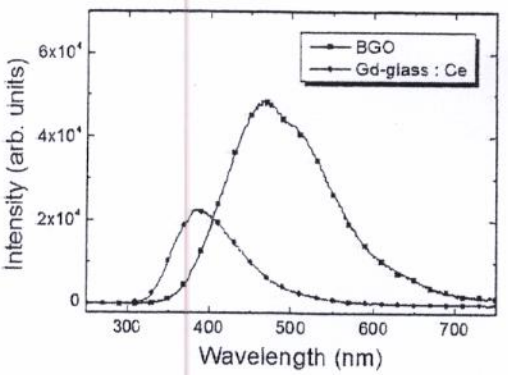


Figure 2 (online color at: www.pss-a.com) RL spectra (X-ray excitation: 40 kV, 15 mA) of Ce^{3+}-doped Gd glass and BGO crystal as measured at RT. The spectra are mutually comparable in an absolute way.

$I_2 = 8\%$, respectively. The PL decay at longer wavelength of 470 nm under excitation with $\lambda_{ex} = 360$ nm was also measured and is given in Fig. 3(b). Its double-exponential approximation, $I(t) = 1481\exp(-t/40.3\text{ ns}) + 49\ \exp(-t/136\text{ ns}) + 0.17$, yields the decay times $\tau_1 = 40.3$ ns and $\tau_2 = 136$ ns with respective component intensities $I_1 = 90\%$ and $I_2 = 10\%$, respectively. While τ_1 decay times are typical for the $5d_1 \rightarrow 4f$ radiative transition of Ce^{3+} [9] there is no immediate explanation of longer τ_2 ones though respective component amplitude is very small.

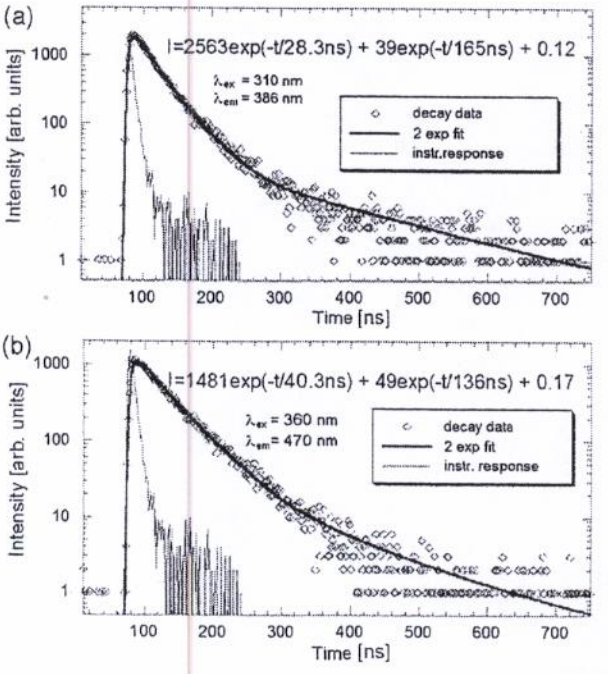


Figure 3 (online color at: www.pss-a.com) PL decay curves of the Ce^{3+}-doped Gd glass [(a) $\lambda_{ex} = 310$ nm, $\lambda_{em} = 386$ nm; (b) $\lambda_{ex} = 360$ nm, $\lambda_{em} = 470$ nm] at RT. The solid lines are the convolution of the instrumental response and the function $I(t)$, given in the figures.

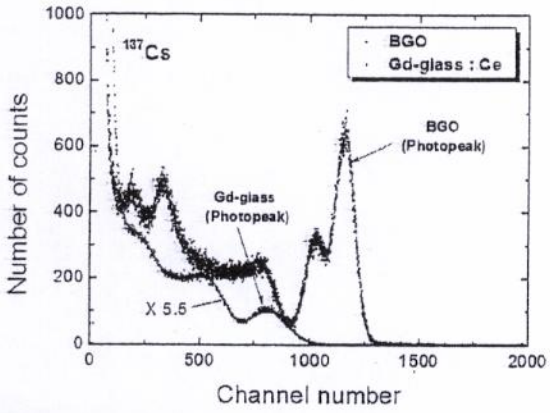


Figure 4 (online color at: www.pss-a.com) Pulse height spectra of 662 keV γ-rays from a ^{137}Cs source as measured with BGO crystal and Ce^{3+}-doped Gd glass (with a higher gain factor of 5.5).

Rather non-exponential decay of Ce^{3+} center in Fig. 3 and varying τ_1 values with emission wavelength are worth a comment. Similar phenomena can be noticed in the silica glasses doped with Ce^{3+} ions [10, 11] and Eu^{2+} ions [3]: the 5d excited state of both Ce^{3+} and Eu^{2+} is sensitive to symmetry, short-range order and crystal field strength at a given site of glass host. Due to glass matrix, the dopant is embedded at a number of slightly inequivalent sites, the emission band broadens inhomogeneously and the decay time value varies as well. Nevertheless, position and shape of PL and RL spectra in Figs. 1 and 2 nearly coincide and decay non-exponentiality is rather small (note quite low I_2 intensity of the decay component associated with longer τ_2 decay times) which means that the inhomogeneity of the Ce^{3+} sites is rather small.

Pulse height spectra of 662 keV γ-rays from a ^{137}Cs source are given in Fig. 4 for both BGO crystal and Ce^{3+}-doped Gd glass (with a higher amplification gain factor of 5.5). To our present knowledge, this is the first time to obtain the photopeak, which is well-separated from the Compton continuum in the pulse height spectrum of 662 keV γ-rays as measured with Ce^{3+}-doped Gd glass. Based on the recorded photopeak positions for Ce^{3+}-doped Gd glass and BGO crystal, and by taking into account the photocathode quantum efficiency of PMT for BGO crystal (~20% at 480 nm) and Ce^{3+}-doped Gd glass (~29% at 390 nm), we estimated the light yield of Ce^{3+}-doped Gd glass to be about 907 ph/MeV as measured at 4 μs shaping time constant. This light yield is about 11% of BGO. The photoelectron yield, light yield and energy resolution for both samples at 662 keV γ-rays are given in Table 1.

Table 1 Photoelectron yield, light yield, and energy resolution at 662 keV γ-rays for Ce^{3+}-doped Gd glass and BGO crystal coupled to the XP5200B PMT as measured at 4 μs shaping time constant.

sample	photoelectron yield (phe/MeV)	light yield (ph/MeV)	energy resolution (%)
Gd glass:Ce^{3+}	263	907	18.3
BGO	1700	8500	9.0

4 Conclusions The Ce^{3+}-doped Gd glass with small inhomogeneity of Ce^{3+} sites was successfully prepared under CO reducing atmosphere. It exhibited an intense violet-blue emission of Ce^{3+} ions under UV and X-ray excitations and an integral scintillation efficiency of about 30% of the BGO scintillator. Despite of low light yield (~11% of BGO), it shows photopeak in the pulse height spectrum of 662 keV γ-rays, which is well-separated from the Compton continuum. Based on good performance in γ-ray detection, short decay time, lower cost and easier fabrication, the Ce^{3+}-doped Gd glass is very attractive scintillator for practical applications *e.g.*, in high-energy physics. However, its scintillation light yield should be further enhanced *e.g.*, by optimizing the concentrations of $Gd^{3+}$and Ce^{3+} in the SiO_2–B_2O_3–Al_2O_3–Gd_2O_3 system.

Acknowledgements This work was supported by grants from the NRCT-NSFC project (No. 50911140475 and 60878043) and the National Research University Project of Thailand, Office of the Higher Education Commission. Partial support of Czech MSMT KONTAKT grant ME10084 is also gratefully acknowledged.

References

[1] G. Zanella, R. Zannoni, R. Dall'Igna, P. Polato, and M. Bettinelli, Nucl. Instrum. Methods Phys. Res. A **359**, 547 (1995).
[2] M. Nikl, K. Nitsch, E. Mihokova, N. Solovieva, J. A. Mares, P. Fabeni, G. P. Pazzi, M. Martini, A. Vedda, and S. Baccaro, Appl. Phys. Lett. **77**, 2159 (2000).
[3] W. Chewpraditkul, D. Chen, B. Yu, Q. Zhang, Y. Shen, M. Nikl, R. Kucerkova, A. Beitlerova, C. Wanarak, and A. Phunpueok, Phys. Status Solidi RRL **5**, 40 (2011).
[4] N. Solovieva, M. Nikl, and K. Nitsch, Opt. Mater. **30**, 113 (2007).
[5] S. Baccaro, R. Dall'Igna, P. Fabeni, M. Martini, J. A. Mares, F. Meinardi, M. Nikl, K. Nitsch, G. P. Pazzi, P. Polato, C. Susini, A. Vedda, G. Zanella, and R. Zannoni, J. Lumin. **87–89**, 673 (2000).
[6] M. Rodova, A. Cihlar, K. Knizek, K. Nitsch, and N. Solovieva, Radiat. Meas. **38**, 489 (2004).
[7] J. Fu, J. M. Parker, R. M. Brown, and P. S. Flower, J. Non-Cryst. Solids **326/327**, 335 (2003).
[8] H. Feng, V. Jary, E. Mihokova, D. Ding, M. Nikl, G. Ren, H. Li, S. Pan, A. Beitlerova, and R. Kucerkova, J. Appl. Phys. **108**, 033519 (2010).
[9] G. Blasse and B. C. Grabmeier, Luminescent Materials (Springer, Berlin, 1994).
[10] A. Vedda, N. Chiodini, D. Di Martino, M. Fasoli, S. Keffer, A. Lauria, M. Martini, F. Moretti, G. Spinolo, M. Nikl, N. Solovieva, and G. Brambilla, Appl. Phys. Lett. **85**, 6356 (2004).
[11] A. Vedda, N. Chiodini, D. Di Martino, M. Fasoli, F. Morazzoni, F. Moretti, R. Scotti, G. Spinolo, A. Baraldi, R. Capelletti, M. Mazzera, and M. Nikl, Chem. Mater. **18**, 6178 (2006).

SCImago Journal & Country Rank

EST MODUS IN REBUS
Horatio (Satire 1,1,106)

Home
Journal Rankings
Journal Search
Country Rankings
Country Search
Compare
Map Generator
Help
About Us

Show this information in your own website

Physica Status Solidi (A) Applications and Materials

Indicator	2004-2011	Value
SJR		0.18
Cites per doc		0.89
Total cites		1342

www.scimagojr.com

☑ Display journal title

Just copy the code below and paste within your html page:

How to cite this website?

SJR is developed by:

SCIMAGO LAB

Powered by SCOPUS

Journal Search

Search query

Exact phrase

Physica Status Solidi (A) Applications and Materials

Country: Germany

Subject Area: Materials Science | Physics and Astronomy

Subject Category: Condensed Matter Physics, Electronic, Optical and Magnetic Materials, Surfaces, Coatings and Films

Publisher: Wiley - V C H Verlag GmbbH & Co.. Publication type: Journals. ISSN: 18626300, 18626319

Coverage: 2005-2011

H Index: 60

Scope:

physica status solidi a - applications and materials science covers the preparation, structural analysis, and numerical simulation of advanced materials, [...]
Show full scope

Charts Data

SJR indicator vs. Cites per Doc (2y)

The SJR indicator measures the scientific influence of the average article in a journal, it expresses how central to the global scientific discussion an average article of the journal is. Cites per Doc. (2y) measures the scientific impact of an average article published in the journal, it is computed using the same formula that journal impact factor ™ (Thomson Reuters).

Citation vs. Self-Citation

WILEY-VCH

Browse our products:

Books

» Journals

Please specify

› Journals from A to Z

Online Submission of Manuscripts

New journals

Latest issues

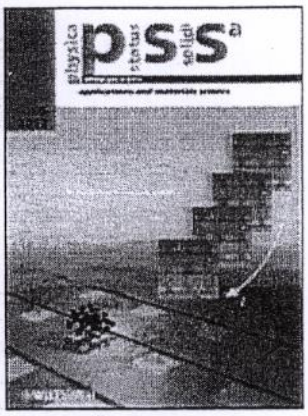
Featured sites

Choose your area of interest:

Please specify

Website Search

physica status solidi (a) - applications and materials science



Latest Issue

0 0

physica status solidi is devoted to the thorough peer review and the rapid publication of new and important results in all fields of solid state and materials physics, from basic science to applications and devices. Being among the largest and most important international publications, the pss journals publish review articles, letters and original work as well as special issues and conference contributions.

physica status solidi a - applications and materials science covers the preparation, structural analysis, and numerical simulation of advanced materials, nanostructures, surfaces and interfaces, as well as properties of such materials and structures relevant for device applications (magnetic, electromechanical, or electronic devices, photonics, spintronics, sensors...).

physica status solidi (a) has further raised its 2010 Impact Factor to **1.472 (+20%)**. Articles are thus cited more often and faster than ever, confirming pss as one of the most important publication brands in solid state physics.

Sign up for the free MaterialsViews.com newsletter and get weekly updates on the latest developments and exciting breakthroughs in the vast field of materials science.

Journal information

Subscription

pss(a) pss(b) pss(c)

Editorial Board

Contact

Download Flyer

Readership: Physicists, crystallographers, materials scientists, solid-state chemists, dealing with solid-state physics or materials science in basic and applied research and teaching

Print ISSN: 1862-6300
Online ISSN: 1862-6319
Volume 209.
12 issues in 2012.
Language of Publication: English

physica status solidi

pss

www.pss-journals.com

reprint

WILEY-VCH