



Synthesis of crystalline Ce-activated garnet phosphor powders and technique to characterize their scintillation light yield

E. Gordienko^a, A. Fedorov^b, E. Radiuk^b, V. Mechinsky^c, G. Dosovitskiy^{a,*},
E. Vashchenkova^a, D. Kuznetsova^a, V. Retivov^a, A. Dosovitskiy^d, M. Korjik^c, R. Sandu^e

^a Institute of Chemical Reagents and High Purity Chemical Substances of National Research Center "Kurchatov Institute" (NRC "Kurchatov Institute" – IREA), 107076, Moscow, Bogorodskiy Val Str. 3, Russia

^b RIINC LLC, Minsk, Belarus

^c Institute for Nuclear Problems, Minsk, Belarus

^d NeoChem JSC, Moscow, Russia

^e National Research Center "Kurchatov Institute", 123098, Moscow, Ploshad Akad. Kurchatova 1, Russia

ARTICLE INFO

Article history:

Received 29 December 2017

Received in revised form

13 February 2018

Accepted 19 February 2018

Keywords:

Phosphor

Powder

Scintillator

Light yield

YAG:Ce

GGAG:Ce

ABSTRACT

This work reports on a process of preparation of garnet phosphor powders and a technique for light yield evaluation of strongly light scattering samples. Powders of scintillation compounds could be used as individual materials or as samples for express tests of scintillation properties. However, estimation of their light yield (LY) is complicated by strong light scattering of this kind of materials. Ce³⁺-activated yttrium-aluminum and gallium-gadolinium-aluminum garnet phosphor powders, Y₃Al₅O₁₂ (YAG:Ce) and Gd₃Ga₃Al₂O₁₂ (GGAG:Ce), were obtained using a modified coprecipitation technique. Ga tends to residue in mother liquor in ammonia media, but the modification allows to avoid the loss of components. We propose an approach for sample preparation and LY measurement setup with alpha particles excitation, allowing to decrease light scattering influence and to estimate a light yield of powder samples. This approach is used to evaluate the obtained powders.

© 2018 Published by Elsevier B.V.

1. Introduction

Devices for medical diagnostics, environmental monitoring, non-destructive testing, security systems etc. are often based on ionizing radiation detection methods, utilizing scintillation detectors [1]. Scintillation yield (light yield) is one of the most important characteristics of a scintillation detector. High light yield is a necessary condition for the effectiveness of the diagnostic equipment and reduction the dose received by the patient, image contrast, accuracy of all measurements involving ionizing radiation detection [2].

Ce-activated complex oxides with garnet structure, which could be described by a general formula (Gd,Y)₃(Ga,Al)₅O₁₂:Ce, are considered among the most promising scintillators, but their properties are strongly sensitive to composition [3–10]. A lot of

experimental work is required to find crucial tendencies and tune a composition to achieve highest possible characteristics. Preliminary experiments using powder samples could be useful for these purposes, as powder synthesis is much less laborious, than singly crystal growth. Moreover, some applications use phosphor powders in form of semi-transparent coatings or screens [11]. Translucent light scattering ceramics has also some existing applications, such as X-ray computed tomography (CT) [12], and some possible applications in high energy physics as cheap alternatives to single crystals [13] or as a scintillation material, which could be formed using 3D-printing [14]. Using untransparent ceramics as samples for preliminary studies in single crystal development is also discussed [15]. In all these examples the light yield of radio-luminescence is a key exploitative characteristic of a material. Therefore, precise, reliable and efficient method to estimate scintillation light yield of such objects is an important instrument in the process of development and production of scintillators and phosphors. However, powders were shown to be strongly light scattering, which could deteriorate measurements results [16].

Well-known method to estimate the light yield of scintillation

* Corresponding author.

E-mail addresses: gordienko@irea.org.ru (E. Gordienko), george.dos@gmail.com (G. Dosovitskiy), daria_kyznecova@inbox.ru (D. Kuznetsova).

detectors is based on comparison of pulse height spectra, measured with the evaluated and the reference detector successively placed on a photodetector, e.g. a vacuum photomultiplier tube (PMT). Ionizing radiation source is placed in close proximity to the detectors; pulse height spectra are accumulated in a multichannel pulse height analyzer. If it is possible to find well-defined lines in the spectra, such as the total absorption peak of 662 keV from Cs-137 gamma-source, then the ratio of the peak positions of the detectors is the ratio of their light yields [17].

This method works well with scintillation materials, having high optical transparency to its own emitted light wavelengths, in which interior scattering and absorption of the scintillation light are minimal and do not introduce additional errors in the measurement result. To implement the method, compared spectra must contain expressed peculiarities, such as lines or peaks of full absorption, the position of which in channels (or energies) can be reliably determined. In light scattering material, the light collection is non-uniform and strongly affected by geometry of scintillators samples being compared. As a result, pulse height spectra of light scattering materials contain no characteristic features which could be used to identify position on the energy scale.

There is also a method of comparison of brightness of scintillators, when measurements are carried out in the current mode, where no pulse shape spectra are recorded, but only mean photocurrent values are being compared [17]. However, the light signal in this case is influenced by the same factors, as in pulse mode measurements – by absorption length of an exciting radiation in a phosphor material and optical transparency and light scattering in the measured material layer. Thus, this approach does not have advantages over pulse height spectra approach in estimating light yield of untransparent or semi-transparent samples. Moreover, peak of total absorption (if could be measured) in pulse height spectrum gives a light yield mark free of electronic noise and radiation background, when registration in current measurement mode requires careful quantification and consideration of these factors.

In this work we propose a technique to evaluate scintillation light yield of powder samples or, more generally, light scattering samples. This technique is applied to characterize powdered yttrium aluminum garnet $Y_3Al_5O_{12}:Ce$, (YAG:Ce) and gadolinium gallium aluminum garnet $Gd_3Ga_3Al_5O_{12}:Ce$, (GGAG:Ce) phosphors obtained by co-precipitation and subsequent high temperature heat treatment calcination. An approach to co-precipitation of Ga-containing compounds is described as well as an important step to obtain powders with controlled composition.

2. Experiment

2.1. Powders preparation and characterization

Powders were obtained by several variations of co-precipitation technique. Basic approach included following sequence of steps: Nitrate solutions with concentrations of 1 mol/l were prepared from Gd oxide (5 N), Ga nitrate (4 N), Al nitrate (5 N), NH_4 -Ce nitrate (4 N), and then mixed in proportion to match desired composition of either $Y_{2.97}Ce_{0.03}Al_5O_{12}$ or $Gd_{2.97}Ce_{0.03}Ga_3Al_2O_{12}$. This joint solution was added to ammonia solution with concentration 2.5 mol/l at a rate 30 ml/min with constant stirring. Then the precipitate was filtered, washed with water, dried at 100 °C, crushed, sieved through 100 μ m polyamide net, calcined at 600 °C and then sintered at 900–1600 °C in air for 2 h in corundum crucibles.

Synthesis procedure variations were applied to ensure control over precipitate composition. Variation 1 consisted in additional stage of heating of the precipitate slurry after the precipitate formation, which was kept boiling under stirring on a hot plate for

30 min, and then processed according to the basic consequence (filtering, washing, drying, calcination, sintering). Variation 2 consisted in elimination of filtering and washing steps. Precipitate slurry was heated up to 100 °C on a hot plate in a molten silica beaker under stirring until the liquid was evaporated, then the temperature was increased up to 300 °C in 50 °C steps to form a dry precipitate and decompose nitrates formed, then the beaker was moved to a furnace and heated up to 600 °C in 100 °C steps. After that it was sieved, calcined and sintered. The discussion on synthesis results is given below in appropriate section.

Mother liquors after precipitation experiments were studied using either mass-spectrometry or gravimetry in order to study the precipitation habits of GGAG:Ce constituting elements. Element analysis was performed using ICP mass-spectrometer Elan DRC-e, PerkinElmer. Phase composition of the powders was studied using powder X-ray diffractometer eMMA, GBC Scientific, with Cu K α radiation. Powders microstructure was studied using Jeol JSM 7100 F scanning electron microscope. Optical microscopy was done using metallographic microscope MET5C, Altami, with $\times 100$ lens having numerical aperture 0.85.

2.2. Measurements procedures

Powder samples were prepared for measurements in a form of a compact cassette as follows: Sheet of polyethylene film with a thickness of 0.02–0.05 mm was stacked on a thin UV-transparent glass plate; aluminum ring with an inner diameter of 12 mm and walls 1 mm thick was placed on top of polyethylene film, and phosphor powder was poured inside the ring so that the powder layer located on a pat with the upper ring plane. A drop of optical UV-curing adhesive Norland 61 (0.10–0.12 cm³) was placed in the center of the powder layer with a syringe, and the blank was kept in the dark for at least 2 h. After the glue is completely soaked, second sheet of polyethylene film is placed on the top of the workpiece and covered with second glass plate. The preform then was cured under a luminescent UV light source (wavelength – 360 nm, total power – 30 wt) for 5 min from each side and left for 48 h at room temperature for hardening. Then the sample was removed from the polyethylene film and excess adhesive was removed with a scalpel. Mechanically bonded powder sample obtained following these procedures is shown on the inset of Fig. 1.

Scintillation light yield of the samples was performed using the “reflection” geometry, in which excitation source and photodetector were placed on one side of a sample (Fig. 1). ²⁴¹Am with alpha particles energy of ~5.5 MeV and activity of 10⁴ Bq was used for scintillations excitation. Thin wire frame was used for the source and the sample mounting to reduce the loss of scintillation light; the entire assembly was covered with Teflon[®] reflector cap. A typical scintillation spectrometer was used, consisting of: PHILIPS XP2020 photomultiplier with spectral sensitivity range of 290–650 nm and a photocathode diameter 44 mm; high-voltage power supply; spectrometric amplifier; a multi-channel pulse height analyzer. Alpha particles were chosen as a source of excitation due to two main reasons: short absorption lengths (tens of microns) and high energy, allowing to separate peak of full absorption from a background.

Pulse height spectra were recorded using the described spectrometer. ROOT v.5.26 software package [18] was used for pulse height spectra processing. Where it could be separated, peak of full absorption was approximated by Gauss function and peak maximum was located. The measured sample peak position was compared to a peak position of a reference sample of YAG:Ce single crystal. It had dimensions $\varnothing 12 \times 1$ mm, grinded front surface, and back surface covered with Teflon reflector. The reference sample pulse height spectra were measured at the same setup, using the

Download English Version:

<https://daneshyari.com/en/article/7907330>

Download Persian Version:

<https://daneshyari.com/article/7907330>

[Daneshyari.com](https://daneshyari.com)