Nuclear Instruments and Methods in Physics Research A 816 (2016) 176–183

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research A

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

Effect of Mg^{2+} ions co-doping on timing performance and radiation tolerance of Cerium doped $Gd_3Al_2Ga_3O_{12}$ crystals

M.T. Lucchini [a,*], V. Babin [c], P. Bohacek [c], S. Gundacker [a], K. Kamada [d,f], M. Nikl [c], A. Petrosyan [b], A. Yoshikawa [d,e,f], E. Auffray [a]

[a] CERN, 1211 Geneva 23, Switzerland
[b] Institute for Physical Research, Ashtarak, Armenia
[c] Institute of Physics AS CR, Prague, Czech Republic
[d] Tohoku University, New Industry Creation Hatchery Center, Sendai, Japan
[e] Tohoku University, Institute for Material Research, Sendai, Japan
[f] C&A Corporation, T-Biz, Sendai, Japan

ARTICLE INFO

Article history:
Received 21 December 2015
Received in revised form
29 January 2016
Accepted 1 February 2016
Available online 11 February 2016

Keywords:
Inorganic scintillators
Medical imaging
Calorimeters
Time-of-flight
Radiation tolerance

ABSTRACT

Inorganic scintillators with high density and high light yield are of major interest for applications in medical imaging and high energy physics detectors. In this work, the optical and scintillation properties of Mg co-doped Ce:$Gd_3Al_2Ga_3O_{12}$ crystals, grown using Czochralski technique, have been investigated and compared with Ce:$Gd_3Al_2Ga_3O_{12}$ ones prepared with identical technology. Improvements in the timing performance of the Mg co-doped samples with respect to Ce:$Gd_3Al_2Ga_3O_{12}$ ones have been measured, namely a substantial shortening of the rise time and scintillation decay components and lower afterglow were achieved. In particular, a significantly better coincidence time resolution of 233 ps FWHM, being a fundamental parameter for TOF-PET devices, has been observed in Mg co-doped crystals. The samples have also shown a good radiation tolerance under high doses of γ-rays, making them suitable candidates for applications in harsh radiation environments, such as detectors at future collider experiments.

1. Introduction

Inorganic scintillators combined with photodetectors are commonly used as efficient radiation detectors for applications in medical imaging and high energy physics [1–4]. In particular for time-of-flight positron emission tomography devices (TOF-PET) and light-based calorimeters at future colliders, a lot of efforts are being spent to improve the timing performance of such detectors. In the former case, the coincidence time resolution (CTR) represents a key parameter to improve noise suppression and to enhance signal to noise ratio in the reconstructed image [5–8]. In the latter, fast timing capabilities accompanied by a sufficient radiation tolerance of the scintillator would permit the detectors to operate in high rate conditions at future collider experiments [9,10].

In many inorganic scintillators such as Cerium-doped silicate and garnet single crystals the delayed radiative recombination at Ce emission centers due to electron traps can deteriorate scintillation performance leading to slow decay components and afterglow. In previous studies on Ce-doped orthosilicates (LSO, LYSO) [11,12], it was observed that Ca^{2+} co-doping can improve the scintillation characteristics due to the suppression of such undesired slow delayed recombination processes. Similar and very encouraging results were recently obtained with single crystal aluminum garnets (LuAG, YAG) by means of divalent ions co-doping such as Mg^{2+} and Ca^{2+} cations [13–15].

In recently discovered Ce:$Gd_3Al_2Ga_3O_{12}$ crystals (Ce: GAGG) [16,17] the defect-engineering of this kind to improve the scintillation mechanism has been applied as well. The detailed study of optical and scintillation characteristics of Ca co-doped GAGG:Ce single crystals and the Ca co-dopant effect on the stabilization of Ce^{4+} center in GAGG host has also been recently published [18]. Excellent scintillation properties of Ce:GAGG crystals and its relatively high density of 6.63 g/cm^3 make it a good candidate for radiation detectors.

Although previous publications have already started to study the properties of Mg- and Ca co-doped GAGG:Ce samples [17,18], the present study is focused on other crystal properties and their effects, which have not been investigated in the aforementioned

* Corresponding author.
E-mail address: Marco.Toliman.Lucchini@cern.ch (M.T. Lucchini).

http://dx.doi.org/10.1016/j.nima.2016.02.004

publications. In this paper we present the characterization of optical and scintillation properties of GAGG:Ce and GAGG:Ce:Mg samples grown by the Czochralski method. In particular, an improvement of the coincidence time resolution in GAGG:Ce:Mg crystals has been measured. Furthermore, an irradiation study using gamma-rays up to high doses ($\simeq 120$ kGy) has also been performed to investigate the radiation tolerance of such material.

2. Experimental methods and results

2.1. Crystal growth

Crystals of GGAG:Ce and GGAG:Ce:Mg were prepared by Czochralski method using an iridium crucible under N_2 atmosphere containing 2% O_2. The seed crystal of ⟨100⟩ orientation was purchased from C&A Corporation, Sendai, Japan. Mixtures of oxides of the purity 5N with compositions of $Gd_{2.985}Ce_{0.015}Ga_{2.7}Al_{2.3}O_{12}$ and $Gd_{2.982}Ce_{0.015}Mg_{0.003}Ga_{2.7}Al_{2.3}O_{12}$ were used as starting materials. A crystal growth velocity of 1.2 mm/hour was used and no excess of gallium oxide was added in the melt.

The set of samples used for the experiments consisted of a pair of GAGG:Ce and GAGG:Ce:Mg pixels ($2 \times 2 \times 10$ mm^3) and a pair of GAGG:Ce and GAGG:Ce:Mg cubes ($10 \times 10 \times 10$ mm^3). A BGO crystal in the form of $7 \times 7 \times 1$ mm^3 was also used for comparison in some of the measurements. The pixels were prepared from platelets which were cut perpendicularly to the crystal growth axis from the first third of the crystals whereas the cubes were obtained from the central part. All the faces of the samples were polished to optical quality.

Another couple of platelets, adjacent to those mentioned above, were analyzed using an electron micro-probe. The compositions of platelets from Mg-free and Mg co-doped crystals have been found to be $Gd_{3.09}Ce_{0.0027}Ga_{2.58}Al_{2.33}O_{12}$ and $Gd_{3.10}Ce_{0.0024}Mg_{0.0018}Ga_{2.54}Al_{2.36}O_{12}$ respectively. The distribution coefficients of Ce and Mg have been determined to be: $k_{Ce} = 0.14$ and $k_{Mg} = 0.45$.

The value of k_{Ce} we measured differs strongly from the one in [17], being 0.793. This is probably related to different growth methods (micro-pulling down in [17]) and different growth velocities, being 1.2 mm/h for our samples and 3.0 – 4.2 mm/h for the crystals in [17]. As the distribution coefficient approaches to 1 when the crystal growth velocity is increased [19], this could explain the larger value of k_{Ce} reported in [17].

2.2. Optical and scintillation properties

Absorption spectra, measured at Shimadzu 3101PC spectrometer across 2 mm thickness, are reported in Fig. 1 and show the $4f \rightarrow 5d^{1,2}$ transitions of Ce^{3+} center and the ^8S-^6P_x,^6I_x transitions of Gd^{3+}. In the Mg co-doped sample a clear fingerprint of Ce^{4+} charge transfer absorption is also observed, in agreement with previously published papers [12–14,18].

Transmission curves, measured at $10 \times 10 \times 10$ mm^3 cubes using a Perkin Elmer (Lambda 650 UV/VIS) spectrometer in the 200–800 nm range, are shown in Fig. 2. Both type of crystals have a good transmission of about 82% in the emission peak region (around 535 nm). The Mg co-doped samples show a stronger absorption in the UV region due to the presence of stable Ce^{4+} centers, see also Fig. 1 and related description.

Radio-luminescence spectra, in Fig. 3, were measured using a custom made 5000M Horiba Jobin Yvon spectrometer and an excitation X-ray source ISOVOLT 60 kV, Seifert Gmbh. A broad emission peak with the maximum around 530–535 nm was observed with no difference in the peak position between GAGG:Ce and GAGG:Ce:Mg samples. Absolute intensity of radio-luminescence is about 20% lower in the Mg co-doped samples, compared to Mg-free ones.

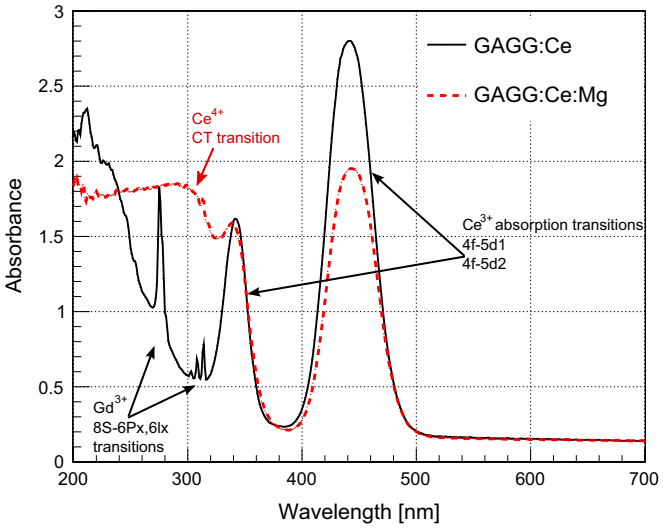

Fig. 1. Absorption spectra of GGAG:Ce and GGAG:Ce:Mg measured at $2 \times 2 \times 10$ mm^3 samples across 2 mm thickness.

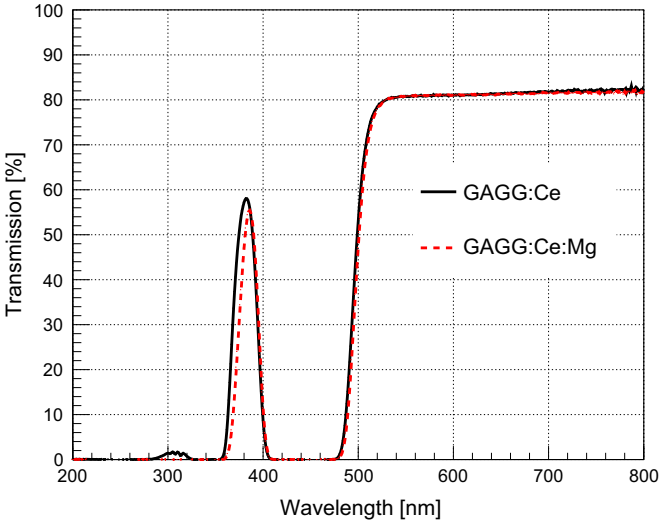

Fig. 2. Transmission curves for GAGG:Ce (black line) and GAGG:Ce:Mg (red dashed line) cubic samples. Lower transmission below 350 nm is observed for co-doped sample. (For interpretation of the references to color in this figure caption, the reader is referred to the web version of this paper.)

The light yield has been measured with a Photonics R2059 photomultiplier tube (PMT) under excitation by a ^{137}Cs source. Before measurement, the crystals were kept in the dark for one day to reduce the level of phosphorescence, see below Section 2.3. The samples were wrapped with Teflon tape and optically coupled with the PMT window using optical grease (refractive index $n = 1.41$). The quantum efficiency of the PMT weighed over the emission spectrum of GAGG:Ce was calculated to be $6.5 \pm 0.3\%$. The uncertainty on the light yield measurement, related to the error on the quantum efficiency calculation and to the calibration of the PMT, was estimated to be around 5%.

Light yield results are reported in Fig. 4 and summarized in Table 1. Using a 3 μs integration gate, a light yield of 35600 ± 1700 ph/MeV was measured for the GAGG:Ce cubic sample, whereas the Mg co-doped cubic sample shows a lower light yield of about 27800 ± 1400 ph/MeV. The energy resolution was found to be 7.1% and 7.6% respectively.

The light yield of the pixel samples was measured by collecting the light from the 2×2 mm^2 face. A light yield of 34700 ± 1700 ph/MeV and 26700 ± 1300 ph/MeV was obtained for the Mg-free