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ABSTRACT

Ceramic and single crystal Lutetium Aluminum Garnet scintillators exhibit energy resolution with bialkali photomultiplier tube detection as good as 8.6% at 662 keV. Ceramic fabrication allows production of garnets that cannot easily be grown as single crystals, such as Gadolinium Aluminum Garnet and Terbium Aluminum Garnet. Measured scintillation light yields of Cerium-doped ceramic garnets indicate prospects for high energy resolution.

Keywords: Scintillators, garnets, gamma ray detectors, transparent ceramics

1. INTRODUCTION

Inorganic scintillators are widely employed in radiation detectors for high energy physics, medical diagnostic imaging, radiation monitoring, and environmental applications. At present, $LaBr_3$:Ce is the scintillator providing the highest energy resolution, ~2.6% at 662 keV, but it is highly hygroscopic and its growth is still challenging. For homeland security applications, detection of weak gamma ray sources and rapid unambiguous isotope identification are required. High sensitivity and effective isotope identification can be attained with scintillator detectors offering high energy resolution, large size, high Z, and fast decay time. Additionally, the possibility to grow very large-sized scintillators at low production costs can facilitate detector deployment.

Lanthanide Aluminum Garnets doped with Cerium, such as Lutetium Aluminum Garnet (LuAG:Ce), Gadolinium Aluminum Garnet (GdAG:Ce) and Terbium Aluminum Garnet (TbAG:Ce) are scintillators featuring high density and effective atomic number (e.g. ρ=6.7 g/cm^3 and Z_{eff}=58.9 for LuAG), which result in high sensitivity. Furthermore, Lanthanide Orthoaluminate and Garnet Oxides exhibit good chemical robustness, and tend to exhibit proportional light yield as a function of electron energy.

Garnet single crystals are typically grown by the Czochralski method, and production of large-sized optics is difficult and expensive. LuAG:Ce scintillators have been described [1],[2], but the presence of intrinsic radioactivity arising from the beta and gamma emission from Lu-176 (2.59% natural abundance) is an undesirable interference [3]. One paper on single crystal Ce-doped Gadolinium Scandium Aluminum Garnet indicated promising optical and scintillation properties for this material, which showed an energy resolution of 12.5% at 662 keV [4]. Transparent polycrystalline ceramics not only allow production costs to be substantially reduced, but the activator concentration and uniformity can be considerably increased. In addition, there is potential to fabricate garnet phases that cannot be grown from melt due to incongruent melting, such as GdAG and TbAG.

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2. EQUIPMENT AND METHODS

Beta radioluminescence employed a ^{90}Sr source (beta endpoint energy 0.546 MeV). Radioluminescence spectra were collected with a Princeton Instruments/Acton Spec 10 spectrograph coupled to a thermoelectrically cooled CCD camera. A flashlamp pumped Nd:YAG laser at 10 Hz was operated at the 4th harmonic of 266 nm, producing 20 ns FWHM pulses to excite the sample. Luminescence is collected with a monochromator coupled to an R928 Hamamatsu PMT and read out by an oscilloscope.

The single crystal LuAG:Ce was grown by Czochralski at Crystal Photonics. The garnet ceramics were formed using stoichiometric mixed metal oxide particles synthesized via flame spray pyrolysis. Fully dense, transparent polycrystalline ceramics were formed by cold pressing green bodies that were subsequently vacuum sintered, and residual porosity removed by hot isostatic pressing. LuAG ceramics prepared by this method approach the level of transparency of a single crystal, and exhibit average grain size of ~10 microns. More details about the ceramic formation process are published elsewhere [5].

The scintillation light produced by the samples was detected by a commercially available Hamamatsu R329EGP PMT, which possesses a quantum efficiency at 550 nm, of 15% [6]. The scintillators were optically coupled to the PMT by means of BC 630 Saint Gobain silicone grease (index of refraction n = 1.465) and wrapped with several layers of Teflon tape. For all measurements, the scintillator was placed in the center of the entrance window of the PMT because in this region the photocathode has better uniformity and photoelectron collection at the first dynode is optimized. The signals from the PMT anode were collected on a 500 Ω load resistor, shaped with a Tennelec TC 244 spectroscopy amplifier (set with a shaping time equal to 8 μs for all the tested crystals) and then recorded with the Amptek MCA8000-A multi-channel analyzer for offline analysis.

3. RESULTS AND DISCUSSION

We characterized a LuAG single crystal (nominal doping of 1% Ce), and compared to results from a LuAG ceramic (0.02 atomic fraction Ce, substituting for Lu). Additional results are reported for a GdYAG:Ce ceramic ($Gd_{1.5-x}Y_{1.5}Al_5O_{12}:Ce_x$, x = 0.02) and a TbAG:Ce ceramic ($Tb_{3-x}Al_5O_{12}:Ce_x$, x = 0.08). Photographs of two of the ceramics characterized in this report are shown in Figure 1.

3.1 Radioluminescence Spectra and Decay Times

Under steady-state beta excitation, the emission spectra recorded represent the integral luminescence over all timescales. In Figure 2, the beta excited luminescence of the garnet ceramics are shown, in comparison to a CsI:Tl standard crystal. While the measured integral light yields are very high for GdYAG:Ce and TbAG:Ce (105,000 and 84,000 Photons/MeV, respectively), some long decay components (see Figure 3) will not contribute to the scintillation light pulse measured in the pulse height spectra. The amplitudes of the slower decay components may possibly be decreased by further increasing the Ce doping level or co-doping with ions that fill intra-bandgap traps. The decay of TbAG:Ce at Ce atomic fraction 0.08 (major component ~1.5 μs), while considerably slower than that of LuAG:Ce and GdYAG:Ce (71 and 106 ns, respectively), is still fast enough to permit use in scintillator detectors. Some slower decay components with small amplitudes are present, and may be more prominent upon excitation with gamma rays than observed here with direct excitation of the Ce^{3+} ion.

3.2 Pulse Height Spectra

The gamma ray pulse-height spectra of the 662 keV line of the Cs-137 radioactive source have been acquired with the LuAG:Ce single crystal and ceramics and with a commercial CsI:Tl for comparison. The total absorption peaks have been processed with a Gaussian fit procedure to evaluate the peak position and the peak full width at half maximum, in order to estimate the scintillation light yield and the energy resolution, respectively. In Figure 4.the pulse-height spectra are reported; The measured light yields of LuAG:Ce single

crystal and ceramic are ~25% and ~16% of CsI(Tl), respectively, and the energy resolutions at 662 keV are 8.6% and 12%, very good considering the transparency of the ceramic can be improved.. The TbAG:Ce sample indicated very promising gamma ray detection features showing a light yield of ~21% compared to a commercial canned CsI(Tl) crystal and a FWHM energy resolution of 11%.

3.3 Conclusions

The high light yields observed under beta excitation suggest that with improved transparency and uniformity, garnet ceramics could offer high energy resolution gamma ray spectroscopy. The overall light yield measured with the beta excitation technique includes slow components, some of which may not be measurable with the 8 μs shaping time. However, improved energy resolution is expected if ceramics fabrication of Gadolinium and Terbium Aluminum Garnet is improved to near-single crystal optical transparency, and if high QE, low noise photodetectors in green-red region are used. Silicon photodiodes or PMTs with unconventional photocathode materials should allow realization of energy resolution in the 2-5% range at 662 keV with garnet scintillators.

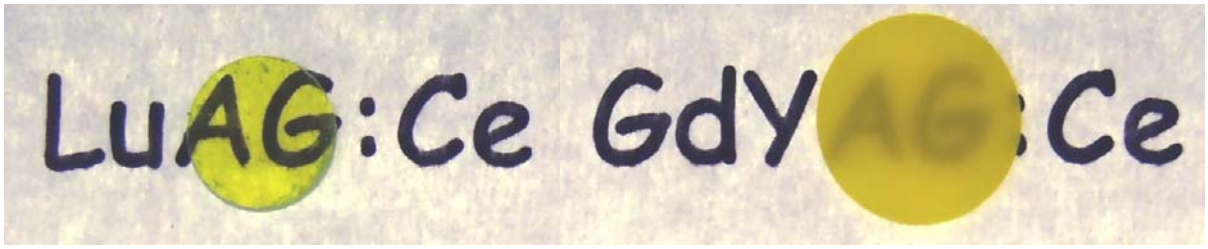

Fig. 1. (left) Photograph of a Cerium-doped Lutetium Aluminum Garnet ceramic, (right) Photograph of a Cerium-doped Gadolinium Yttrium Aluminum Garnet ceramic. Lower transparency is obtained for the GdYAG and TbAG ceramics so far.

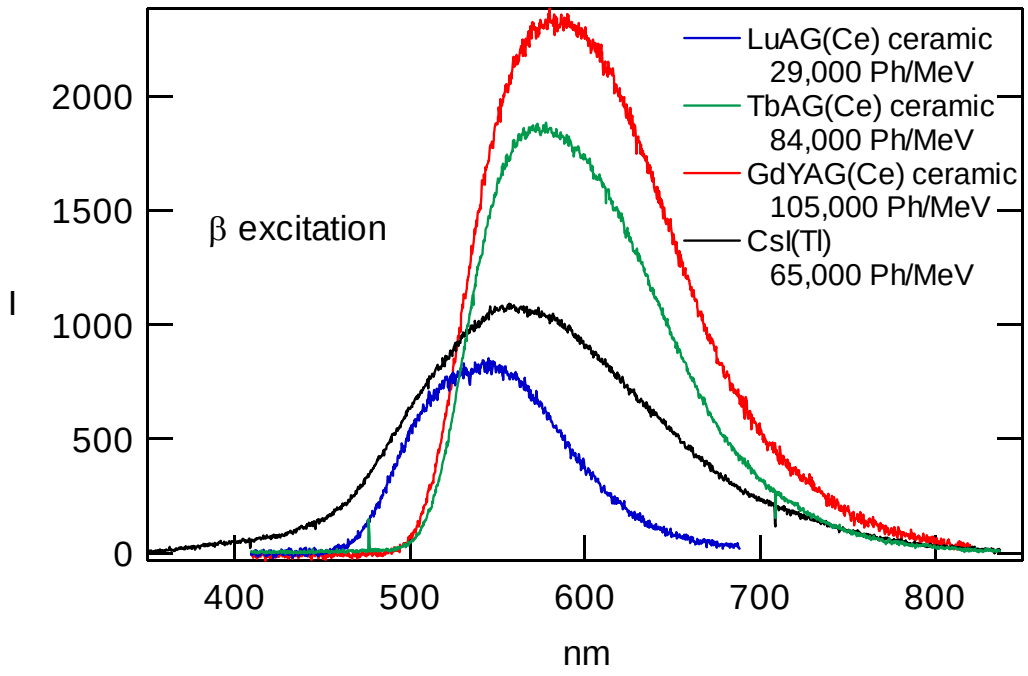

Fig. 2. Beta-excited radioluminescence spectra acquired of a Ce-doped Lutetium Aluminum Garnet ceramic (blue), a Ce-doped Gadolinium Aluminum Garnet ceramic (red), and a Ce-doped Terbium Aluminum Garnet ceramic (green), compared to Tl-doped Cesium Iodide (black).

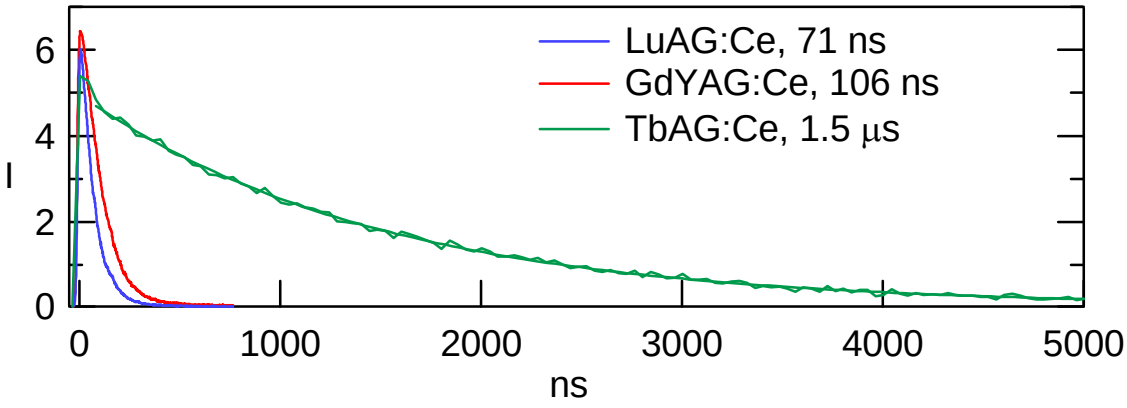

Fig. 3. Time-resolved luminescence decays acquired by excitation with 30 ns laser pulses at 266 nm. All fit to single exponential functions, with additional small amplitude long-lived components.

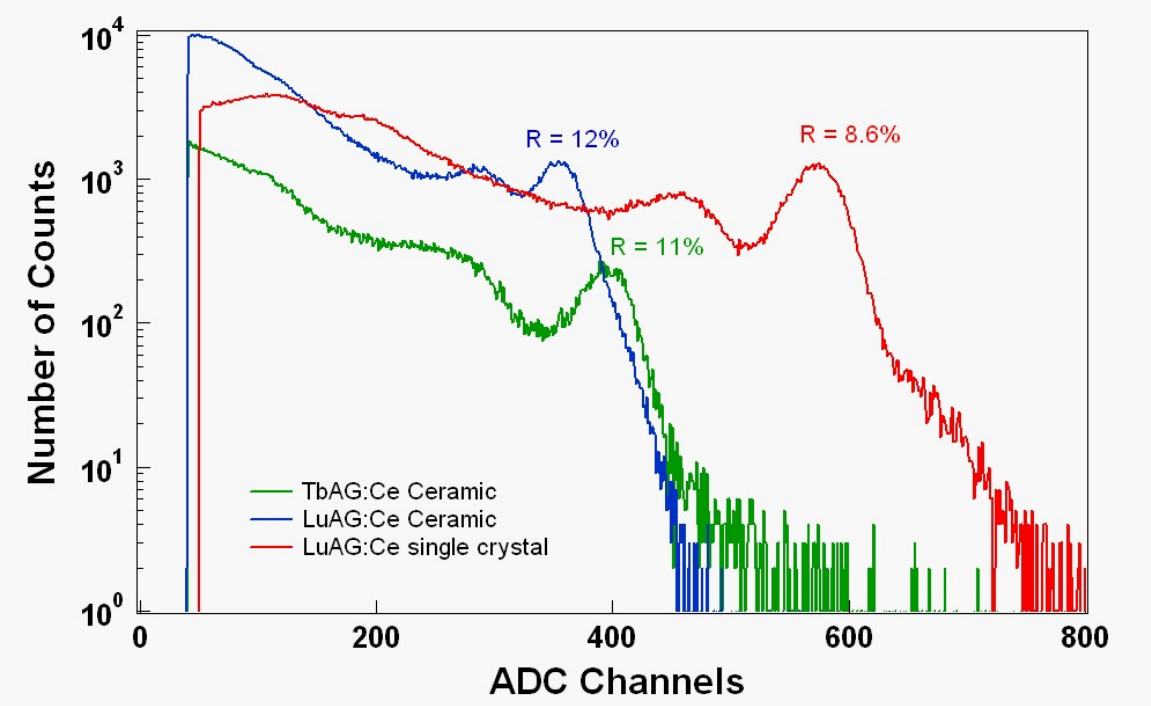

Fig. 4. Pulse height spectra acquired of a Ce-doped Lutetium Aluminum Garnet single crystal (red), a Ce-doped Lutetium Aluminum Garnet ceramic (blue), and a Ce-doped Terbium Aluminum Garnet ceramic (green).

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