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Improvement of Scintillation Properties on Ce doped $Y_3Al_5O_{12}$ scintillator by divalent cations co-doping

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Abstract

AE co-doped Ce: YAG single crystal were grown by the micro-pulling down (μ-PD) method and characterized as for the structure and chemical composition. The Mg 500ppm co-doped sample showed a light output of around 150% compared to the non co-doped Ce:YAG sample. Details of results about crystal growth with other dopants and their luminescence and radiation response characteristics will be shown in our presentation. Band structure deference of Ce:YAG and AE co-doping effects on energy levels in the band will be discussed.

I. INTRODUCTION

The white light emitting diodes (LEDs) have a number of advantages over the conventional illumination devices including the incandescent and the fluorescent lamps in the viewpoints of low electric consumption, high brightness and long lifetime [1]. Most commonly used white LED is a combination of blue LED and yellow scintillator (Ce:$Y_3Al_5O_{12}$) [2,3]. The function of the Ce:YAG phosphor is to absorb the blue light emitted from the blue LED and converts it to yellow light. The phosphor has a broad emission band due to the 5d → 4f transition of Ce^{3+}. The combination of the transmitted blue light and the yellow fluorescent makes white light.

In the past decades, great effort was made to develop more efficient and fast scintillators to detect ionizing radiation. As a result, Ce-doped silicates, namely, the Gd_2SiO_5 (GSO), Lu_2SiO_5 (LSO), $(Lu_{1-x}Y_x)_2SiO_5$ (LYSO) and most recently $LaBr_3$ single crystal hosts were developed. At present, the best combination of stopping power, decay time, and light yield is achieved by Ce-activated materials, but there is still a continuous demand for new scintillator materials with even better properties. The most recent studies point to tunneling-driven radiative recombination between Ce emission centers and nearby lying deep electron traps which can in principle deteriorate scintillation performance. Y2SiO5:Ce (YSO:Ce) single crystals were studied comparatively less than LSO:Ce and LYSO:Ce [4]. Both LSO:Ce and YSO:Ce single crystals co-doped with Ca^{2+} have been recently investigated [5] and improvement in their scintillation characteristics, namely afterglow suppression and scintillation decay acceleration, were claimed. Most recently, divalent cation co-doping effects were investigated in LaBr3 :Ce,Sr [6] and LuAG:Ce,Mg [7] .

The aim of this work is to investigate AE(=Mg, Ca, Sr) co-doping effects on luminescence and scintillation properties of Ce doped YAG scintillator. In this report, AE co-doped Ce: YAG single crystal were grown by the micro-pulling down (m-PD) method and characterized as for the structure and chemical composition. Luminescence and radiation response characteristics were measured.

II. RESULTS

A. Crystal growth

A stoichiometric mixture of 4N CeO2, Y_2O_3 , Mg_2CO_3 , Ca_2CO , Sr_2CO and α-Al_2O_3 powders (High Purity Chemicals Co.) was used as starting material. Nominally, Lu^{3+} site were substituted by Ce^{3+} and AE^{2+} according to the formula of $AE_xCe_y,Y_{3-x-y}Al_5O_{12}$. Single crystals were grown by the micro-pulling-down (μ-PD) method with an RF heating system. Typical pulling rates were 0.05–0.07 mm/min and the diameter was around 3 mm. Crystals were grown from an Ir crucible under N_2 atmosphere to prevent evaporation of gallium oxide. The seed crystals were <111> oriented undoped YAG crystals. Plates of 3mmϕ x 1mm were cut and polished for the measurements of luminescence and scintillation properties. Example photos of grown crystals are shown in Figure 1.

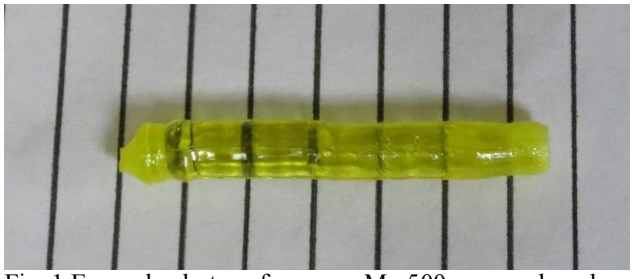

Fig. 1 Example photos of a grown Mg 500ppm co-doped Ce1%:YAG single crystal.

B. Luminescence and scintillation properties

Figure 2 shows absorbance spectra of Mg co-doped Ce1%:YAG samples. As increasing Mg concentration, Absorption in 200-350 range originated from Ce^{4+}. Radioluminescence spectra and photoluminescence decays at room temperature (RT) were measured at Spectrofluorometer 199S (Edinburgh Instrument) using an X-ray tube (operated at 35 kV and 16 mA, Mo cathode) and the pulsed nanosecond hydrogen-filled flashlamp for the excitation, respectively. Single grating excitation and emission monochromators and photon counting detection were employed. Fig.3 shows radioluminescence spectra of the Mg co-doped Ce:YAG samples. The expected 530 nm emission of Ce^{3+} 4f-5d and 330nm emission originated from anti-site defects in garnet structure have been observed. Intensity of 530nm emission was increased by Mg co-doping.

The Gamma-ray responses were measured using APD S8664-55(Hamamatsu). The signal was fed into a pre-amplifier (CP580K), a shaping amplifier (CP 4417), a pocket multichannel analyzer (MCA) (Amptec 8000A), and finally to a personal computer. The bias for the APDs was supplied by CP 6641. The bias voltage was 350V. The shaping time of the shaping amplifier was set at 2 μs. Figure 4 exemplifies the Mg co-doped Ce1%:YAG samples irradiated by ^{137}Cs, where 662 keV. The Mg 500ppm co-doped sample showed a light output of around 150% compared to the non co-doped Ce:YAG sample.

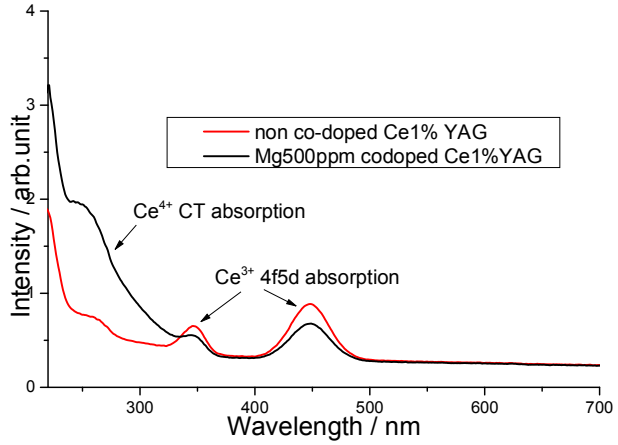

Fig. 2 absorption spectra of Mg co-doped Ce:YAG

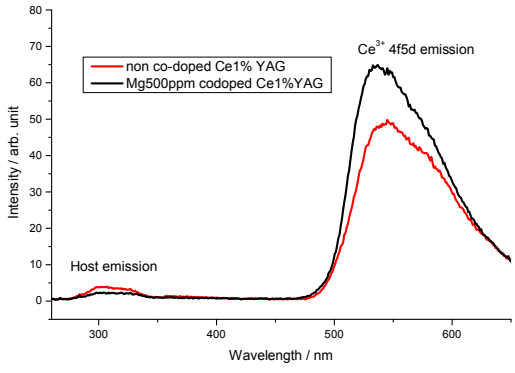

Fig. 3 Radioluminescence spectra of Mg co-doped Ce:YAG

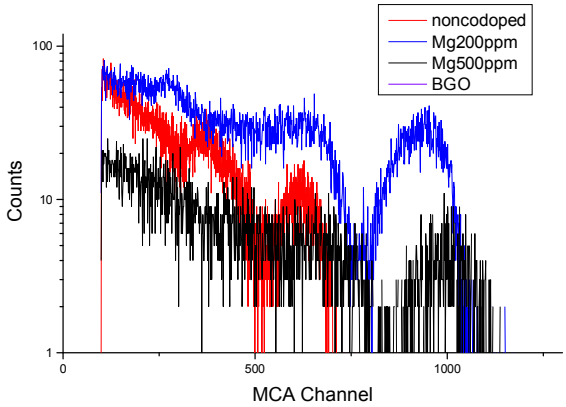

Fig. 4. Energy spectra of the grown Mg co-doped Ce:LuAG

III. CONCLUSION AND FUTURE PROSPECTS

In this presentation, AE(=Mg, Ca, Sr) co-doping effects on luminescence and scintillation properties of Ce: YAG will be reported. AE co-doped Ce: YAG single crystal were grown by the micro-pulling down (μ-PD) method and characterized as for the structure and chemical composition. The Mg 500ppm co-doped sample showed a light output of around 150% compared to the non co-doped Ce:YAG sample. Details of results about crystal growth with other dopants and their luminescence and radiation response characteristics will be shown in our presentation. Band structure deference of Ce:YAG and AE co-doping effects on energy levels in the band will be discussed.

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