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Effect of Lu-to-Y ratio and Mo coactivation on scintillation properties of LuYAG:Pr and LuAG:Pr,Mo crystals

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ABSTRACT

In this paper the basic scintillation and thermoluminescence properties of LuYAG:Pr and LuAG:Pr,Mo crystals are reported. It is shown that, primarily, both materials display significantly higher scintillation yields than their prototype LuAG:Pr. The results of radioluminescence, low and high temperature thermoluminescence, and scintillation time profile measurements are analyzed quantitatively to allow us a better understanding of the scintillation process in LuYAG:Pr and LuAG:Pr,Mo, as well as to let us propose or verify possible explanations of the observed yield enhancement.

1. Introduction

Rare-earth activated garnet compounds are one of the most promising groups of scintillators introduced to the market in the recent years [1]. Fitting into the actual research trends related to this kind of materials, $(Lu_xY_{1-x})_3Al_5O_{12}$:Pr (LuYAG:Pr) and $Lu_3Al_5O_{12}$:Pr,Mo (LuAG:Pr,Mo) crystals have been grown under the project entitled "Study of the Influence of Electron Trap Distribution on the Efficiency of Host-to-Ion Energy Transfer in (Lu,Y)AG:Pr(,Mo) Scintillator Crystals", aimed at improving their scintillation properties with an anticipated measurable effect of light yield increase. It has already been reported that a 1 mm high sample of $(Lu_{0.75}Y_{0.25})_3Al_5O_{12}$:Pr displays a reasonably high yield of 33,000 ph/MeV [2], which denotes a significant step forward compared to $Lu_3Al_5O_{12}$:Pr (~20,000 ph/MeV). Recent investigations on LuAG:Pr,Mo samples have also demonstrated their higher yields against LuAG:Pr [3]. The achieved enhancement has been attributed to some specific changes in distributions of electron traps occurring in particular hosts, depending on the lutetium-to-yttrium ratio and molybdenum coactivation. In this paper the results of pulse height (PH), radioluminescence (RL), low and high temperature thermoluminescence (ltTL and htTL), and scintillation time profile (STP) measurements performed on LuYAG:Pr and LuAG:Pr,Mo crystals are presented and discussed in order to broaden the knowledge of the mechanisms responsible for the scintillation yield increase in these materials.

2. Materials and experiment

The boules (Fig. 1) of $(Lu_xY_{1-x})_3Al_5O_{12}$:Pr ($x$ = 0.00, 0.25, 0.50, 0.75, 1.00) and $Lu_3Al_5O_{12}$:Pr,Mo have been grown at ITME, Warsaw, by the Czochralski method as characterized in [2,4]. Cube $5 \times 5 \times 5$ mm^3 (for PH and STP measurements) and plate $5 \times 5 \times 1$ mm^3 (for RL, ltTL, and htTL measurements) samples have been cut from these boules and thoroughly polished. The two generations of $Lu_3Al_5O_{12}$:Pr reference crystals come from Furukawa Co. Ltd. (see [5] for technological details) and have been investigated previously ("old" in [2,6], "new" in [7–9]). The basic properties of all the samples used in the current research are specified in Table 1.

The PH, RL, ltTL, and STP experiments have been carried out using the same setups, methods and settings as reported in [2,10]. Details of the supplementary ltTL studies with the "$T_{max} - T_{stop}$" method can be found in [11]. Each htTL glow curve has been recorded with a Mikrolab RA'04 reader at a heating rate of 1 K/s, following a 100 s X-ray irradiation of the studied specimen.

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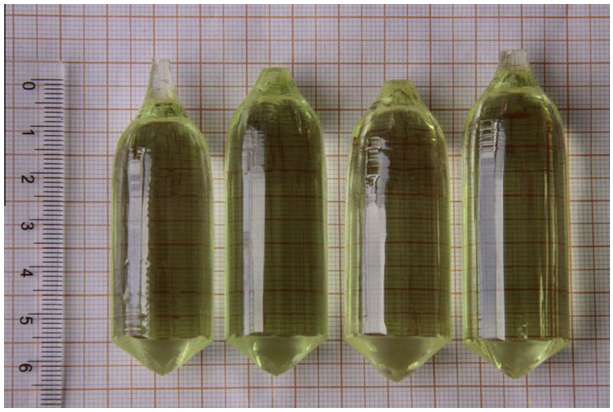

Fig. 1. "As grown" LuYAG:Pr crystals.

3. Results and discussion

3.1. Pulse height spectra

Representative PH spectra of LuAG:Pr, $Lu_{0.75}Y_{0.25}AG$:Pr and LuAG:Pr,Mo are shown in Fig. 2. It is evident that upon either partial replacement of lutetium with yttrium or molybdenum coactivation an increase of light output compared to LuAG:Pr, proved by photopeak shifts to higher channel numbers, takes place. The values of light yield (LY) and energy resolution (ER) of all the investigated samples, derived from the respective spectra, are listed in Table 1.

We note that PH spectra of each sample have been taken 3 times (i.e. using 3 different sides of the cube to stick it to the PMT window). Having at our disposal 4 cubes of each composition itemized in Table 1 we have obtained 12 values of LY and ER per material in this way. Since the spreads within the collected "twelves" are lower than an arbitrary level of 5% mostly approved as uncertainty of yield determination, for clarity of the paper in Table 1 we place the highest observed yields and lowest resolutions. We also mark that double Gaussian functions have been used to fit the shapes of the PH spectra in the photopeak region, taking into consideration the presence of the so-called escape peak, which is a small satellite peak on the left side of the photopeak. Since the *K*-shell electron binding energy for lutetium and yttrium is 63.3 keV and 17.0 keV [12], respectively, in case of ^{137}Cs PH spectra (661.6 keV) the escape peak should appear around 598.3 keV for LuAG:Pr and 644.6 keV for YAG:Pr, which agrees with our experimental results.

According to Table 1, $Lu_3Al_5O_{12}$:Pr from ITME is somewhat worse than the new generation of $Lu_3Al_5O_{12}$:Pr from Furukawa Co Ltd., particularly with respect to ER. To some extent it may be associated with the difference in praseodymium concentration, which is much closer to the yield-optimized one [5] in the latter crystal (0.23 at% vs. 0.12 at%). On the other hand, the samples from ITME coactivated with molybdenum have the same lower Pr content (0.12 at%), which does not prevent them from displaying high LYs (123–137% compared to LuAG:Pr from Furukawa Co. Ltd.). Interestingly, the magnitude of LY improvement decreases with increasing concentration of molybdenum, which is a similar effect to the one recognized for $LuAlO_3$:Ce,Mo [13]. It is the LuAG:Pr,Mo crystal with the least Mo content (0.0005 at%) that almost overtakes the best material of the $(Lu_xY_{1-x})_3Al_5O_{12}$:Pr family, i.e. $Lu_{0.75}Y_{0.25}AG$:Pr. It cannot be excluded that after refining all the compositions (the Lu-to-Y ratio and the concentrations of Pr and Mo) LuAG:Pr,Mo would be a much better scintillator than

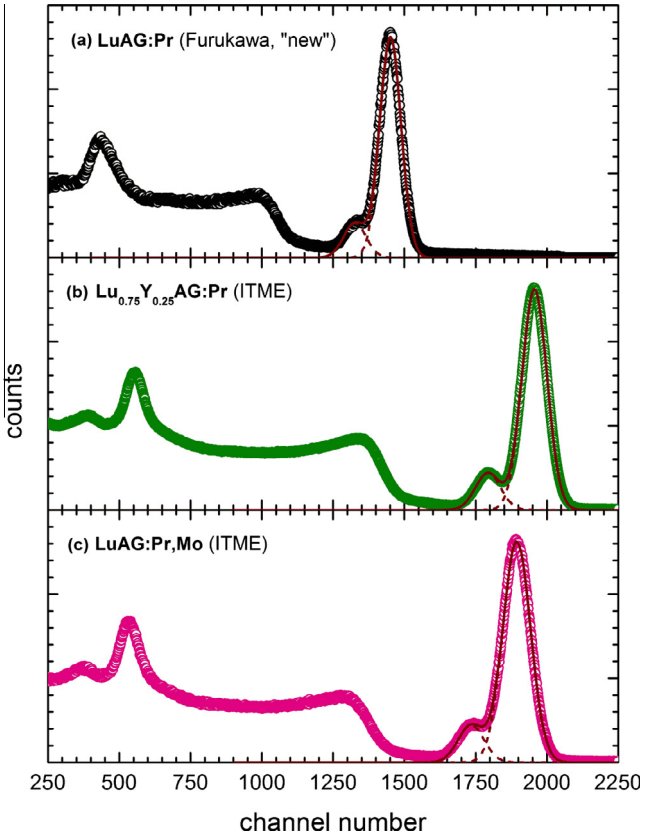

Fig. 2. 662 keV pulse height spectra recorded with (a) LuAG:0.23%Pr, (b) $Lu_{0.75}Y_{0.25}AG$:0.16%Pr, and (c) LuAG:0.12%Pr,0.0005%Mo.

Table 1
The basic properties of the studied crystals.

Host	Activator and concentration[a]	Density (g/cm^3)[a]	Company	Light yield (ph/MeV)[b]	Relative yield[b]	Resolution at 662 keV (%)[b]
$Lu_3Al_5O_{12}$ ("old")	Pr (0.23 at%)	6.7	Furukawa	16,700	0.874	6.1
$Lu_3Al_5O_{12}$ ("new")	Pr (0.23 at%)	6.7	Furukawa	19,100	1.00	6.1
$Lu_3Al_5O_{12}$	Pr (0.12 at%)	6.7	ITME	16,900	0.885	10.0
$(Lu_{0.75}Y_{0.25})_3Al_5O_{12}$	Pr (0.16 at%)	6.2	ITME	27,000	1.41	5.3
$(Lu_{0.5}Y_{0.5})_3Al_5O_{12}$	Pr (0.17 at%)	5.7	ITME	21,800	1.14	5.9
$(Lu_{0.25}Y_{0.75})_3Al_5O_{12}$	Pr (0.23 at%)	5.2	ITME	20,600	1.07	6.1
$Y_3Al_5O_{12}$	Pr (0.17 at%)	4.6	ITME	21,200	1.11	6.1
$Lu_3Al_5O_{12}$	Pr (0.12 at%), Mo (0.0005 at%)	6.7	ITME	26,200	1.37	6.1
$Lu_3Al_5O_{12}$	Pr (0.12 at%), Mo (0.0009 at%)	6.7	ITME	24,800	1.30	6.0
$Lu_3Al_5O_{12}$	Pr (0.12 at%), Mo (0.005 at%)	6.7	ITME	23,500	1.23	6.1

[a] The values of Pr and Mo concentration in the crystals from ITME have been determined by the ICP–OES, while the values of density by the pycnometer method.
[b] The scintillation parameters correspond to the cube $6 \times 6 \times 6$ mm^3 or $5 \times 5 \times 5$ mm^3 samples from Furukawa or ITME, respectively, and have been derived from PH spectra recorded with a shaping time of 2 μs.