Effect of Li^+ ions co-doping on luminescence, scintillation properties and defects characteristics of LuAG:Ce ceramics

Shuping Liu¹, Xiqi Feng¹, Jiri A. Mares², Vladimir Babin², Chen Hu¹, Huamin Kou^{*,1}, Carmelo D'Ambrosio³, Jiang Li¹, Yubai Pan⁴, Martin Nikl^{*,2}

Abstract:

Monovalent Li^+ codoped $\text{Lu}_3\text{Al}_5\text{O}_{12}:\text{Ce}$ (LuAG:Ce) optical ceramics were fabricated by solid state reaction method and further optimized by an air-annealing process. Optical absorption, radioluminescence spectra and scintillation properties such as light yield, scintillation decay times and afterglow were measured and compared with those of the Li^+ free LuAG:Ce ceramic and the commercial LuAG:Ce single crystal samples. Positive effect of Li^+ codopant consists mainly in the significant increase of scintillation light yield, acceleration of scintillation decay as well as the decrease of afterglow intensity. With 0.3% Li codoping, the obtained

LuAG:Ce,Li ceramic displays a light yield of ~29200 ph/MeV at 10 μ s shaping time, higher than that of the LuAG:Ce single crystal and optical ceramaic scintillators ever reported. The partial conversion of the stable Ce^{3+} to Ce^{4+} centers and the shallow and deep traps effect suppression by the Li^+ codoping are discussed.

Key words: LuAG:Ce,Li ceramics, scintillator, Li^+ codoping, Ce^{4+} and Ce^{3+} centers

1. Introduction

Scintillation materials convert the ionizing radiation into visible or ultraviolet light which can be easily detected by conventional photodetectors such as photomultiplier tubes (PMT) and CMOS camera [1, 2]. Within the last two decades, a number of excellent scintillators based on the Ce^{3+} and Pr^{3+} -doped materials, together with truly novel nanoscintillator, nanocomposite and phase separated material systems, were discovered and studied in depth [3-7]. Effective strategies like band-gap engineering, defect engineering and solid solution concept were also developed to improve the scintillation performance of advanced materials [8-10].

Recent studies have shown that the divalent ion Me^{2+} ($\text{Me} = \text{Ca}, \text{Sr}, \text{Mg}$) co-doping could improve both the light yield and scintillation decay of $\text{Lu}_2\text{SiO}_5\text{:Ce}$ (LSO:Ce,Ca) and $\text{LaBr}_3\text{:Ce}$ single crystals [11-13]. The presence of Ce^{4+} centers due to local charge compensation following Me^{2+} co-doping was clearly identified in the former compounds and considered responsible for the improved scintillation performance displayed by the materials. Similar and very encouraging results were also obtained in Mg^{2+} co-doped $\text{Lu}_3\text{Al}_5\text{O}_{12}\text{:Ce,Mg}$ (LuAG:Ce,Mg) optical ceramics. The scintillation

mechanism of stable Ce^{4+} center which provides an additional fast recombination pathway in $\text{LSO}:\text{Ce,Ca}$ and $\text{LuAG}:\text{Ce,Mg}$ scintillators were studied and proposed [14-16], consisting in the sequence of steps: (i) an immediate capture of electron from the conduction band by a stable Ce^{4+} center; (ii) radiative de-excitation of such a temporary Ce^{3+} and (iii) capture of a hole from the valence band or nearby defect to complete the cycle, returning cerium center into the stable $4+$ charge state. Furthermore, the occurrence of Me^{2+} -perturbed variants of O^- center was proposed and it was considered to play a key role in stabilizing and dynamically restoring the Ce^{4+} centers which contribute to fastest part of scintillation response working in parallel with the stable Ce^{3+} centers [17]. However, the attempt to improve the scintillation performance by Me^{2+} co-doping strategy was unsuccessful in $\text{Lu}_{0.8}\text{Sc}_{0.2}\text{BO}_3:\text{Ce}$ [18] and $\text{LuAlO}_3:\text{Ce}$ [19] single crystals. An acceleration of scintillation decay was accompanied, unfortunately, with a big decrease of light yield (LY). Some LY decrease was also observed in the $\text{Mg}^{2+}/\text{Ca}^{2+}$ co-doped $\text{Gd}_3\text{Al}_2\text{Ga}_3\text{O}_{12}:\text{Ce}$ (GAGG:Ce) garnet single crystals [20]. In Ref. [21, 22], a clear correlation between the scintillation performance and the changes of crystal field and defect structures were provided in Ca^{2+} , Ba^{2+} and B^{3+} codoped GAGG single crystals. It was found that codoping with Ca^{2+} ions suppress shallow trap centers but at the same time introduces deeper trap centers reflected in thermoluminescence peaks above room temperature.

In this paper, a monovalent Li^+ co-doping strategy was carried out in $\text{LuAG}:\text{Ce}$ optical ceramics. Its influence on the tuning the stable $\text{Ce}^{3+}/\text{Ce}^{4+}$ centers ratio and the

luminescence and scintillation properties were systematically clarified by employing optical absorption, radioluminescence (RL), light yield, scintillation decay and afterglow measurements. Besides, the characteristics of point defects in LuAG:Ce,Li ceramics were also investigated by thermoluminescence (TSL) technique. Discussion on the results of material optimization includes the comparison with Li⁺ free LuAG:Ce ceramic and the commercial LuAG:Ce single crystal samples.

2. Experimental Procedure

LuAG:0.3 at.% Ce, x at.% Li (x = 0.1, 0.15, 0.2 and 0.3, respectively) ceramics were prepared by solid-state reaction method and further optimized by an air-annealing process. The detailed technological route has been already described in previous works [23, 24]. In order to clarify the role of Li⁺ co-dopant, a 0% Li (Li free) LuAG:0.3 at.% Ce ceramic was also included (the one designated as *dI* in reference [25]). After being vacuum sintered at 1880 °C for 10-30 h and air-annealed at 1450 °C for 20 h, all the samples were double-polished to a thickness of 1-1.5 mm for further experiments. Commercial references, the LuAG:Ce single crystal grown by Czochralski method (by Crytur Ltd., Turnov, Czech Republic), as well as a Bridgman grown BGO single crystal provided by SICCAS Corp. (Shanghai, China) were also considered for comparison.

Absorption spectra were measured using the Shimadzu 3101 PC spectrometer in 200-800 nm range. RL spectra were measured under an X-ray tube excitation (operated at 40 kV, 15 mA) and recorded in reflection geometry at the custom-made

5000M model Horiba Jobin Yvon spectrofluorometer equipped with a TXB-04 photon counting detector. For afterglow measurement, samples were irradiated by X-rays for about 10 seconds with the same X-ray tube at room temperature, after which the beam was cut off and the residual luminescence emitted from samples was recorded as a function of time. Thermoluminescence (TSL) measurements were performed in the 77-500 K range in the cryostat Optistat, Oxford Instruments, after X-ray irradiation at 77 K (operated at 40 kV for 15 minutes). A 5 K /min heating rate was adopted.

Scintillation light yield was investigated using hybrid photodetectors (HPMT hybrid photomultipliers DEP PP0470 or PP0475B ones). Measured ceramics are characterized by wide emission bands peaking in the green spectral range where quantum efficiency of HPMT is around or below 10 % and the overall accuracy of light yield measurements is $\pm 5\%$. Scintillation decay curves were obtained by using the Hamamatsu PMT R375 and Tectronix TDS3052 digital oscilloscope under excitation by 662 keV γ -ray photons of ^{137}Cs source. To enhance collection of scintillation photons a Teflon reflection tape was used to cover the measured samples. Good optical coupling between samples and HPMT photocathode window was ensured by Q2-3067 optical grease.

3. Results and Discussion

Fig. 1 shows the absorption spectra of the air annealed LuAG:Ce,Li ceramics. The absorption bands at about 447 and 347 nm are related to the $\text{Ce}^{3+} 4f \rightarrow 5d_1$ and $5d_2$

transitions, respectively. They appear somewhat saturated due to the stray luminescence light of Ce^{3+} centers. The absorption pattern below 300 nm is composite [26, 27] due to the overlap of higher $4f \rightarrow 5d$ Ce^{3+} absorption bands and various intrinsic and trace impurity (Fe^{3+} , Yb^{2+} , or defects located at the grain boundaries of the ceramics) related absorptions, as well as the charge transfer absorption of Ce^{4+} centers [14-16] which was introduced by Li^+ codoping and air-annealing. A high level of absorbance of the samples in transparency region above 500 nm can be explained by higher light scattering effects. When compared to the Mg^{2+} co-doped LuAG:Ce,Mg ceramics in which the Ce^{4+} charge transfer absorption is increasing with Mg^{2+} content [28], the tuned cerium valence from Ce^{3+} to Ce^{4+} state in LuAG:Ce,Li ceramics cannot be observed very clearly by increasing the concentration of Li^+ co-dopant. This may be ascribed to poorer optical qualities of the obtained LuAG:Ce,Li ceramics and the thicker sample size used in the measurements.

In **Fig. 2**, the RL spectra of the LuAG:Ce,Li ceramics are displayed together with the LuAG:Ce reference single crystal. The $5d_1 - 4f$ ($^2F_{7/2, 5/2}$ doublet) transition of Ce^{3+} peaking at ~512 nm is observed in all the samples. The aliovalent impurity Li^+ codopant leads to a reduced RL intensity when compared to Li^+ free LuAG:Ce ceramic. Similar phenomenon was also observed in Mg^{2+} codoped LuAG:Ce,Mg ceramics [14, 28]. On the other hand, the RL intensities of Li^+ codoped LuAG:Ce,Li ceramics are higher than that of LuAG:Ce single crystal, and it tends to increase as the content of Li^+ co-dopants increases. This effect, however, can be also influenced by partial opacity of the Li-codoped samples. Besides, the Lu_{Al} antisite defect related

luminescence in the UV region [29] decreases dramatically after Li^+ codoping and it decreases with increasing of Li^+ co-dopant concentration, see the inset of Fig. 2. This improvement is considered due to the lower fabrication temperature of ceramics during which no liquid phase arises as well as the positive effect of Li^+ co-doping.

In order to verify the scintillation properties of Li^+ codoped LuAG:Ce,Li ceramics, the light yield, scintillation decay and afterglow time profiles are measured and compared with those of Li^+ free ceramic and the LuAG:Ce reference single crystal. The results are summarized in **Table 1**. Similar to the trend of RL intensity, light yield values of LuAG:Ce,Li ceramics increases with increasing of Li^+ concentration. It is worth noting that the light yield of 0.3% Li^+ codoped LuAG:Ce,Li ceramic reaches 17740 ± 890 ph/MeV when using a short shaping time of 1 μs , higher than that of the Li^+ free one (14760 ± 740 ph/MeV) and of the reference LuAG:Ce single crystal (16390 ± 820 ph/MeV). In **Fig. 3**, the light yield values of selected samples are reported as a function of shaping time from 0.5 μs to 10 μs . Apparently, the highest light yield value is observed at the longest shaping time of 10 μs and it reaches 29200 ± 1460 ph/MeV for 0.3% Li codoped LuAG:Ce,Li ceramic, the highest light yield value ever reported in LuAG:Ce single crystal and optical ceramic scintillators.

Table 1. Sample information, light yield (excitation ^{137}Cs , 662 keV, 1 μs shaping time) and scintillation decay of the LuAG:Ce,Li ceramics and LuAG:Ce reference single crystal. A double-exponential function $I(t) = \sum A_i \exp [-t/\tau_i]$ $i=1, 2$ was used to fit

the scintillation decay curves and the relative intensity of each component $I_i = A_i\tau_i / \sum A_j\tau_j \times 100$ ($i, j = 1, 2$) is reported as well.

Li concentration (at. %)	Size (mm)	LY (ph/MeV)	τ_1 (ns)/ I_1 (%)	τ_2 (ns)/ I_2 (%)	Afterglow at 4 ms (%) / 400 ms (%)
Li Free	Ø15×1	14760	69/41	1120/59	6.8/0.3
0.1	Ø15×1.5	13860	64/39	1150/61	6.1/0.4
0.15	Ø15×1.5	13870	63/42	1070/58	5.2/0.2
0.2	Ø15×1.5	15140	61/45	1160/55	6.3/0.3
0.3	Ø15×1.5	17740	61/43	1240/57	5.8/0.2
LuAG:Ce sc	Ø10×1	16390	70/34	1700/66	1.3/0.1

Fig. 4 shows the scintillation decay curves of selected samples. All the samples show typical two component scintillation decays based on the prompt and delayed radiative recombination at both the stable Ce^{3+} and temporary Ce^{3+} (created after electron capture by Ce^{4+}) centres. It is worth noting that the Li^+ codoped LuAG:Ce,Li ceramics have faster decays than that of the Li^+ free one and the LuAG:Ce single crystal. With increasing Li^+ content, the scintillation decay times of the LuAG:Ce,Li ceramics decrease and the relative weight of the fast decay component increases, see Table 1. Both the improvement of light yield and scintillation decay properties after Li^+ codoping can be explained by the creation of the stable Ce^{4+} center, which is considered to create an additional fast recombination pathway working in parallel with the classical mechanism based on the stable Ce^{3+} centers [14].

In **Fig. 5**, the afterglow characteristics are shown together with BGO single crystal which is known by its extremely low afterglow. Apparently, the afterglow intensities of Li^+ codoped LuAG:Ce,Li ceramics are weaker than that of the Li free one, which is

consistent with the observed higher light yield values and faster scintillation decay measured above after Li^+ codoping. However, the LuAG:Ce,Li ceramics still show worse afterglow characteristics when compared with the Mg^{2+} codoped ones [28] and LuAG:Ce single crystal as well. The relative values of afterglow intensity at 4 ms and at 400 ms after X-ray cut off (the signal level with X-rays being on is normalized to 1) are reported in Table 1.

Finally, TSL measurements were performed with the aim to evidence the influence of Li^+ codoping on the shallow and deep traps in LuAG:Ce ceramics. The spectrally unresolved TSL glow curves of Li free and 0.3% Li^+ codoped LuAG:Ce ceramics are reported in **Fig. 6**. It can be seen that a complex profile including broad structureless and overlapped TSL peaks was obtained in both samples. Their positions stay the same and match very well with the ones previously observed in LuAG:Ce single crystals [30]. So it seems that Li^+ codoping does not introduce additional electron traps in the forbidden band of LuAG:Ce ceramics. However, the TSL intensity of Li codoped LuAG:Ce,Li ceramic is clearly higher than that of Li free one below 340 K while the trend above 370 K is rather opposite. This means that a higher concentration of defects induced by various lattice deformation and charge mismatch may exist in LuAG:Ce,Li ceramics. Besides, by comparing the luminescence and scintillation properties measured above, one can see that the Li-free LuAG:Ce ceramic showed higher RL intensity, but lower light yield and TSL intensity than Li codoped ones. It means that some non-radiative recombination might occur in the Li-free LuAG:Ce ceramics. Further study is needed to explain these phenomena in more detail.

4. Conclusions

To summarize, LuAG:Ce,Li optical ceramics with Li^+ concentrations varying from 0.1 to 0.3 at% were designed and fabricated. Significant increase of scintillation light yield, acceleration of scintillation decay as well as a decrease of afterglow intensity were achieved, which demonstrates the positive effect of monovalent Li^+ codopant. With 0.3% Li^+ concentration, the obtained LuAG:Ce,Li ceramic displays a light yield of ~ 29200 ph/MeV at 10 μs shaping time, which is higher than that of the optimized LuAG:Ce,Mg scintillators. The partial conversion of stable Ce^{3+} to Ce^{4+} and the introduction of additional fast radiative recombination pathway through the stable Ce^{4+} centers were considered the most probable reason for the observed improvement.

However, high level of light scattering centers and significant intensity of “slow scintillation light” generated by the delayed radiative recombination are obvious disadvantages of LuAG:Ce,Li ceramics. In the light of our investigation, simultaneous codoping by both the divalent Me^{2+} and monovalent M^+ are proposed as an effective strategy to improve further the scintillation performance of LuAG:Ce scintillators. Li^+ seems more effective than the divalent ones in tuning the cerium from Ce^{3+} to Ce^{4+} state, while the Mg^{2+} codoping was shown to act also as an effective sintering aids improving considerably the ceramic transparency. Future research should be focused on the optimization of optical quality of LuAG:Ce ceramics by double codoping with

Mg²⁺ and Li⁺ ions and the detailed investigation of point defects and scintillation mechanism induced by such double codoping.

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Figure Captions

Fig. 1. Optical absorption spectra of LuAG:Ce,Li ceramics with different Li^+ concentrations.

Fig. 2. RL spectra of LuAG:Ce,Li ceramics with different Li^+ concentrations, and the LuAG:Ce and BGO reference single crystals. In the inset, the normalized RL spectra of Lu_{Al} antisite related defect between 250 and 420 nm is given.

Fig. 3. Light yield of LuAG:Ce reference single crystal and LuAG:Ce,Li ceramics with different Li^+ concentrations as a function of shaping time in the 0.5-10 μs range.

Fig. 4. Normalized scintillation decay curves of selected LuAG:Ce,Li ceramics and LuAG:Ce ceramic and single crystal reference samples.

Fig. 5. Spectrally unresolved afterglow time profile of LuAG:Ce,Li ceramics with different Li^+ concentrations, BGO and LuAG:Ce single crystals after the 40 kV X-ray excitation cut-off.

Figure 6. Spectrally unresolved TSL glow curves of the LuAG:Ce,Li ceramics after 15 min X-ray excitation at 77 K.

Figure 1-Abs

Figure 2-RL

Figure 3-LY

Figure 4-decay

Figure 5-afterglow

Figure 6-TSL