Light yield measurement of the Ce3+ activated LuAG nano-powder garnet scintillating detector

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Abstract− This paper focuses on the characterization of nano-powder (NP) scintillator, specifically presenting preliminary results of the measurement of the light yield (LY) of a newly synthesized Lutetium Aluminum (LuAG) garnet phosphor NP scintillator, doped with 0.1% cerium. The scintillation light yield was measured using the comparative method. Considering the light-scattering and self-absorbing properties of powder scintillators, alpha particles from ^{241}Am source were used for excitation instead of Gamma/X rays, while a Ce^{3+} activated LuAG single crystal (SC) was employed as the reference detector due to its well-established light yield. A photomultiplier tube (PMT) served as the photodetector, and signals were read out by a nuclear instrumentation chain. The results demonstrate that the newly prepared LuAG:Ce NP exhibits a light output that is approximately 90% of that of the single crystal reference. These findings highlight the promising potential of NP scintillators as a viable alternative to single crystals, especially in applications where the preference for powder scintillators is evident.

***Keywords*− LuAG scintillation detector, light yield measurement, alpha particles, nano-powder.**

Nomenclature

Am	Americium
HV	High Voltage
LY	Light Yield
MCA	Mulichannel Analyzer
NP	Nano-powder
PL	Photoluminescence
PMT	Photomultiplier Tube
SC	Single crystal

I. Introduction

Several applications [1], [2] such as homeland security, medical imaging diagnosis, high energy physics (HEP), industrial nondestructive control, environmental monitoring rely on radiation detection using scintillating materials as detectors. In these applications, certain scintillating detector characteristics are crucial for their effectiveness. For example, medical imaging diagnosis applications like positron emission tomography (PET) require high scintillation light yield (LY) and fast time resolution to improve signal-to-noise ratio, image quality, accuracy, and reduce patient dose[1], [3]–[5]. In certain applications, nano-sized scintillator, such as rare earth activated garnet phosphor NPs, can serve as an alternative to single crystal scintillators, as they are easier to synthesize compared to growing single crystals. This makes them a more cost-effective option for applications like high energy physics (HEP) and X-ray computed tomography (CT) [6].

Among the inorganic scintillators, Ce^{3+}-activated lutetium aluminum garnet with the general formula Lu3Al5O12 (LuAG:Ce^{3+}) in powder form shows promise due to its excellent properties, including chemical and mechanical stability, good optical properties, and high density (6.73 g/cm3) [7]. However, determining the optimal composition for achieving the best scintillation performances requires both theoretical and experimental investigations. Scintillators in powder form are known for light scattering and self-absorption [8]. Powder samples with a thickness of (1-3) mm are not transparent, resulting in a significant fraction of generated photons being scattered and reabsorbed by the material itself. As result, only a limited number of photons are able to reach the photodetector, Making it challenging to measure scintillation characteristics in the absence of certain elements, such as the lack of a photo-peak in the energy spectrum. This lack of efficiency, especially for gamma and X-rays is due to the requirement for significant absorption depth, which is not achievable with small powder samples. On the other hand, thick powder samples exhibit strong light scattering and absorption. Consequently, powder materials are not efficient in terms of gamma and X-ray absorption [9]. Therefore, careful attention should be given for the choice of excitation radiation when measuring the scintillation properties of powder materials[1][9]. In characterization tasks, achieving a high scintillation light yield (LY) is crucial for obtaining accurate measurement signals. However, obtaining a high LY with gamma and X-ray radiation is a challenge. On the other hand, it has been found that alpha particles are more efficient when interacting with powder scintillators. This is because alpha particles are absorbed in the surface layers of the sample (within a few microns) [1], Additionally, alpha particles have high energy, leading to a greater production of photons and thus better measurement signals. In such circumstances, the excitation geometry also plays a role in ensuring a good collection of the scintillation photons. Considering the excitation geometry is important for efficiency of the measurement setup[1][6]. In addition, and for the excitation with alpha particles, The investigation demonstrated that the

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reflection geometry is more suitable for evaluating the scintillation properties of powder scintillators, rather than transmission geometry [1][9].

The objective of this work is to measure the scintillation light yield of a newly prepared Ce^{3+} doped lutetium aluminum garnet $Lu_3Al_5O_{12}$ (LuAG) NP scintillator. The scintillator was synthesized using the Sol-Gel method, and a previous study investigated the effect of Ce^{3+} concentration on the photoluminescence (PL) properties, determining that the optimal Ce^{3+} dopant concentration for the highest PL performance is 0.1% [7].

Various methods are commonly used to measure the scintillation light yield of scintillator detectors, which are detailed in [10]. In this work, the comparison method was employed, and the light yield was estimated using a Ce-doped LuAG single crystal as a reference detector, as its light yield is known (12000-15000 ph/MeV) [11]–[13]. Alpha particles with an energy of 5.48 MeV from ^{241}Am were used for excitation, a photomultiplier tube (PMT) was served as a photodetector and the reflection geometry was applied.

II. Method and experimental details

A. Samples preparation

Ce^{3+} doped lutetium aluminum garnet NPs (LuAg:Ce^{3+}) were synthesized using the sol-gel method, as described in[7]. The NP scintillator was synthesized with a Ce^{3+} concentration of 0.1%. The resulting NPs were then pressed and compacted to form a pellet measuring approximately 1 cm in diameter and 3 mm in thickness, as illustrated in **Fig. 1**.

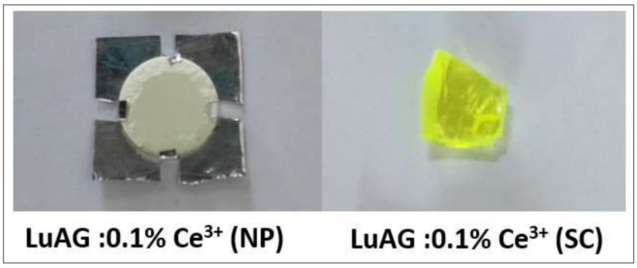

Fig. 1: Photography of the prepared LuAg:0.1%Ce^{3+} (NP) and the LuAg:0.1%Ce^{3+} (SC), the single crystal was used as a reference detector.

The LuAG:0.1%Ce^{3+} SC was developed using the Czochralski growth method. A small portion of comparable size with the NP sample was used as a reference for measuring the light yield. The single crystal sample is shown in Fig. 1. Further details regarding the development of the single crystal can be found in[14], [15].

B. Light yield measurement method

The scintillation light yield (LY), measured in Photons/MeV, is a key characteristic of scintillation detectors. Several methods, including the method of comparison, are commonly employed to determine the LY [16]. The comparison method, as described in the literature[1][10], involves determining the relative light yield (RLY) by comparing the signal amplitudes of the detector under characterization with those of a reference detector with a known LY. Both detectors should be measured in the same setup and under room temperature. The RLY can be calculated using Equation (1):

$$RLY_D(\%) = \frac{PP_{-D}}{PP_{-RD}} * \frac{G_{RD}}{G_D} \quad (1)$$

Where: RLY is the relative light yield (in % of the reference detector), PP_{-D} is the signal amplitude from the detector under test, PP_{-RD} is the signal amplitude from the reference detector, G_{RD} is the amplifier gain applied to reference detector and G_D is the amplifier gain applied to the detector under characterization. The absolute light yield (in ph/Mev) of the detector under characterization can be estimated by multiplying the RLY by the known LY of the reference detector [1] as given in the equation (2).

$$ALY_D = RLY_D . LY_{RD} \quad (2)$$

Where ALY_D is the absolute light yield (in ph/Mev) of the detector under characterization , LY_{RD} is the absolute light yield of the reference detector (in ph/Mev), the absolute light yield of the yield is reported in [11] .

For the measurement of the light yield (LY) of the prepared Ce-doped LuAG NP sample, the LuAg:0.1%Ce^{3+} single crystal was employed as a reference detector. Both of LuAg:Ce SC and NP are depicted in Fig. 1. A calibrated detection chain was utilized to acquire measurement signals and to collect energy spectrums from both the NP and single crystal detectors, in the same configuration and under the reflection geometry.

Alpha particles were used for excitation for two reasons. Firstly, their high energy enables effective discrimination of noise and the production of a well-defined full absorption peak [1]. Secondly, alpha particles interact effectively with scintillator materials over short distances (tens of microns). To determine the light yield (LY), alpha particles with an energy of 5.48 MeV from ^{241}Am (with an 85% yield) were employed as the excitation source. In order to prevent any external light from penetrating, a tightly closed box was used to house the excitation source, samples, and PMT.

The experimental setup used for measuring the light yield of the prepared sample is presented in Fig. 2. The light emitted by the scintillators under excitation by alpha particles was detected by a photomultiplier tube (PMT). The PMT model used was the 6292-DUMONT, featuring a multi-alkali S-9 photocathode. The spectral response of the PMT is well-matched to the spectral emission of the Ce-doped LuAG, with a maximum response in the range of 400-550 nm [17], which exactly aligns with the peak emission wavelength of LuAg:Ce at 500 nm [18]. Both the spectral emission of LuAg:Ce and the spectral response of the PMT fall within the visible light range.

The output of the PMT was connected to the input of the ORTEC 572 amplifier, which served to amplify and shape signals. The amplification gain was fixed at 50 for both prepared and reference detectors. Energy spectra were acquired using the EASY-MCA-8K-CH multichannel analyzer (MCA) from ORTEC and collected using Maestro Software, also from ORTEC.

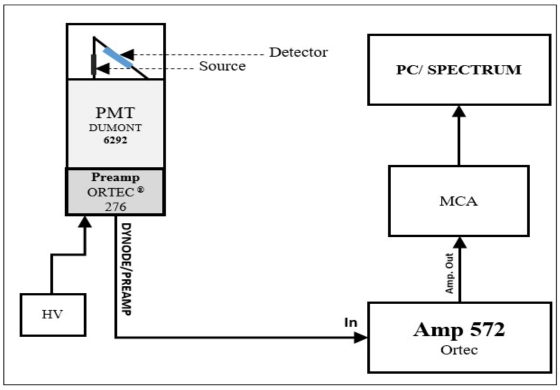

Fig.2: Schematic view of the light measurement setup.

A pulse height spectrum was initially recorded using the LuAg:0.1%Ce^{3+} single crystal reference detector. This process was then repeated with the LuAg:0.1%Ce^{3+} NP detector under characterization. By comparing the positions of the 5.48 MeV photo-peak obtained with the LuAg:Ce single crystal and the

NP, an estimation of the light output of LuAG:0.1%Ce NP can be determined. Fig. 3 illustrates the pulse height spectra recorded for both the LuAg:Ce NP and the single crystal under 5.48 MeV alpha particles excitation, using a shaping time of 6 µs for the amplifier. The selected shaping time allows for the collection of all charges produced in the PMT, ensuring the maximum collection of photons from the scintillator detectors[19]. The PMT was biased with a 1400V high voltage (HV) power supply, and the PMT bias, shaping time, and amplifier gain were the same for both spectrums. To accurately determine the photo-peak center positions, Gaussian fitting was performed on the obtained photo-peaks.

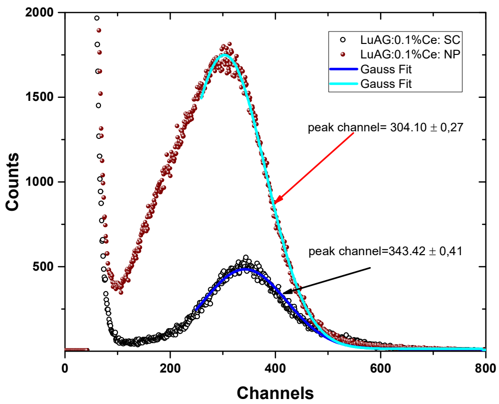

Fig.3: Pulse height spectra of 5.48 Mev alpha particles recorded with LuAg: Ce NP and single crystal (HV=1400V,Shaping time = 6µs, amplification gain = 50, spectra were collected for a time of 1000 s)

III. RESULTS AND DISCUSSION

As shown in the Fig.3, both spectrums show different photo-peak positions. By analyzing the obtained photo-peak for both the LuAg: 0.1%Ce^{3+} NP (detector under test) and the LuAg:0.1%Ce^{3+} SC (reference detector), it was determined that the light output of the prepared sample of the LuAg:0.1%Ce^{3+} NP is approximately 89% of the light output of the LuAg:0.1%Ce^{3+} SC used as reference. Results of the light yield (LY) measurement are presented in both relative and absolute values in Table 1. The absolute LY value of the reference detector used for comparison is that reported in [11] which is of 15000 Ph/MeV.

Table. I
LIGHT YIELD MEASUREMENT RESULTS

Scintillators	PEAK POSITIONS	RELATIVE LIGHT YIELD (%)	ABSOLUTE LIGHT YIELD (PH/MEV)
LuAG SC	343.42±0.4	100	15000
LuAG NP	304.10±0.3	88.55±0.13	13395±25

IV. CONCLUSION

This study is a part of an ongoing endeavor to develop and enhance NP scintillating detectors. The main focus of this research was to characterize the scintillation light yield. By utilizing a reference detector and alpha particles, the light output of a newly developed Ce-activated LuAG NP scintillator was successfully measured. The results indicated that the NP scintillator demonstrates a relatively high light output, comparable to that of the single crystal reference. These findings suggest that NP scintillators have the potential to serve as an alternative to single crystals in certain applications, owing to their ease of development. However, it is important to acknowledge that characterizing powder scintillators presents additional challenges compared to single crystals due to their tendency for light scattering, which can reduce efficiency when detecting certain types of radiation, such as Gamma and X-rays. Further advancements in characterization methods are necessary in this regard. Future work will involve investigating other scintillation properties of NPs, such as timing characteristics, as well as exploring the impact of dopant concentrations on scintillation characteristics.

REFERENCES

[1] E. Gordienko et al., “Synthesis of crystalline Ce-activated garnet phosphor powders and technique to characterize their scintillation light yield,” Opt. Mater. (Amst)., vol. 78, pp. 312–318, 2018, doi: 10.1016/j.optmat.2018.02.045.

[2] C. Dujardin et al., “Needs, trends, and advances in inorganic scintillators,” IEEE Trans. Nucl. Sci., vol. 65, no. 8, pp. 1977–1997, 2018, doi: 10.1109/TNS.2018.2840160.

[3] E. Auffray et al., “A comprehensive and systematic study of coincidence time resolution and light yield using scintillators of different size and wrapping,” IEEE Trans. Nucl. Sci., vol. 60, no. 5, pp. 3163–3171, 2013, doi: 10.1109/TNS.2013.2270089.

[4] M. Salomoni, R. Pots, E. Auffray, and P. Lecoq, “Enhancing light extraction of inorganic scintillators using photonic crystals,” Crystals, vol. 8, no. 2, pp. 1–35, 2018, doi: 10.3390/cryst8020078.

[5] A. Knapitsch and P. Lecoq, “Review on photonic crystal coatings for scintillators,” Int. J. Mod. Phys. A, vol. 29, no. 30, 2015, doi: 10.1142/S0217751X14300701.

[6] G. Dosovitskiy et al., “Polycrystalline scintillators for large area detectors in HEP experiments,” J. Instrum., vol. 12, no. 6, pp. 1–23, 2017, doi: 10.1088/1748-0221/12/06/C06045.

[7] A. Boukerika and L. Guerbous, “Investigation of the structural and photoluminescence properties of Ce3+-doped LuAG nanopowders prepared via sol-gel method,” Opt. Mater. (Amst)., vol. 40, pp. 14–19, 2015, doi: 10.1016/j.optmat.2014.11.039.

[8] S. Sinha, J. Kwong, W. G. J. Langeveld, and P. Ryge, “Characterization of ZnO, BaF2 and PbWO4 scintillator detectors for cargo inspection using transmitted X-ray spectroscopy,” IEEE Trans. Nucl. Sci., vol. 60, no. 2, pp. 1016–1023, 2013, doi: 10.1109/TNS.2013.2241451.

[9] H. Mekki, L. Guerbous, A. Boukerika, and K. Lebbou, “Scintillation properties of (Lu1−x Yx)3Al5O12:Ce3+ nanoscintillator solid solution garnet materials,” J. Instrum., vol. 18, 2023, doi: https://doi.org/10.1088/1748-0221/18/02/P02007.

[10] E. Sysoeva, V. Tarasov, and O. Zelenskaya, “Comparison of the methods for determination of scintillation light yield,” Nucl. Instruments Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip., vol. 486, no. 1–2, pp. 67–73, 2002, doi: 10.1016/S0168-9002(02)00676-9.

[11] “High Precision Scintilation Crystals Phosphor Ce:LuAG Crystal High Light Yield,” 2021. http://www.crystro.com/sale-11745046-high-precision-scintilation-crystals-phosphor-ce-luag-crystal-high-light-yield.html

[12] S. Liu, X. Feng, Z. Zhou, M. Nikl, Y. Shi, and Y. Pan, “Effect of Mg2+ co-doping on the scintillation performance of LuAG: Ce ceramics,” Phys. Status Solidi - Rapid Res. Lett., vol. 8, no. 1, pp. 105–109, 2014, doi: 10.1002/pssr.201308199.

[13] T. Yanagida et al., “Scintillation properties of LuAG (Ce) ceramic and single crystalline scintillator,” IEEE Nucl. Sci. Symp. Conf. Rec., pp. 1612–1614, 2010, doi: 10.1109/NSSMIC.2010.5874048.

[14] K. L. A. Nehari, G. Alombert-Goget, O. Benamara, H.Cabane, M. Dumortier, P. Jeandel, I. Lasloudji, F.Mokhtari, T.Baron, G.Wong, M. Allani, J. Boy, S.Alzuaga, L. Arapan, F.Gegot, T.Dufar, “Czochralski crystal growth and characterization of large Langatate (La3Ga5.5Ta0.5O14, LGT) crystals for SAW application,” R. Soc. Chem., vol. 21, no. 11, pp. 1764–1771, 2019, doi: 10.1039/C8CE02157K.

[15] H. Li, E. A. Ghezal, A. Nehari, A. Brenier, and K. Lebbou, “Bubbles defects distribution in sapphire bulk crystals grown by Czochralski technique,” Opt. Mater. (Amst)., vol. 35, pp. 1071–1076, 2013, doi: 10.1016/j.optmat.2012.12.022.

[16] K. S. Shah et al., “CeBr/sub 3/ scintillators for gamma-ray spectroscopy,” in IEEE Symposium Conference Record Nuclear Science 2004., 2004, vol. 7, pp. 4278–4281. doi: 10.1109/NSSMIC.2004.1466835.

[17] DUMONT, “DUMONT_PMT_6292.” [Online]. Available: https://frank.pocnet.net/sheets/085/6/6292.pdf02/Dumont_PMT_6292.pdf

[18] A. Boukerika and L. Guerbous, “Investigation of the structural and photoluminescence properties of Ce3+-doped LuAG nanopowders prepared via sol-gel method,” Opt. Mater. (Amst)., vol. 40, pp. 14–19, 2015, doi: 10.1016/j.optmat.2014.11.039.

[19] K. S. Shah et al., “CeBr 3 scintillators for gamma-ray spectroscopy,” IEEE Trans. Nucl. Sci., vol. 52, no. 6, pp. 3157–3159, 2005, doi: 10.1109/TNS.2005.860155.

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