

THE EFFECT OF CeO₂ ON Gd₂Lu_(1-x)Ce_xAl₄ScO₁₂ PROPERTIES FOR X-RAY DETECTING PHOSPHOR

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Abstract

The phosphors with Gd₂Lu_(1-x)Ce_xAl₄ScO₁₂ compound were studied for radiation detection material in this work. The effect of Ce content on structural, optical and luminescence properties of phosphor was investigated. The XRD pattern presented the cubic and orthorhombic crystalline structure with some derivative structures and defects. The optical spectra indicated the photon absorptions in ultraviolet - visible light region by host components and Ce³⁺ ion. The photoluminescence spectra performed the strong ultraviolet and visible light emission by 4f - 5d transition of Ce³⁺ and other discrete energy in structure. The decay time of phosphors was in a range of 65 to 73 ns. The radioluminescence excited by X-ray revealed the various and complicated emission peaks of ultraviolet, visible light, and infrared, originating from Gd³⁺, Ce³⁺, and defects in phosphors. The developed Gd₂Lu_{0.95}Ce_{0.05}Al₄ScO₁₂ phosphors showed high potential for using in X-ray detecting screen and detector device.

Keywords: Aluminium Garnet Phosphor; Cerium; Luminescence

Introduction

The phosphor material known as luminescence powder has been widely used in the light source or photon generating devices such as display, fluorescence lamp, LED, watch and medical- / flaw detection- / security inspection- imaging system. The radiation detecting phosphor requires the high radiation absorption coefficient, appropriate band gap, efficient energy transfer to luminescence center, and strong light emitting. The multi-components (Gd,Y)₃(Al,Ga)₅O₁₂ garnets have been researched and developed as radiation detection

materials in form of single-crystal (Ashurov *et al.*, 1977; Kamada *et al.*, 2011; Chewpraditkul *et al.*, 2017), ceramic (Chen *et al.*, 2020) and phosphor (Asami *et al.*, 2018; Kaura *et al.*, 2020; Ou *et al.*, 2020). The optimal ratio of Gd to Ga content enhances the host band gap by suppressing the shallow traps and reducing the thermal ionization of luminescence Ce³⁺ ion. The efficient photon interaction in this garnet is facilitated by high effective atomic number of Gd element, and the effective energy transfer from Gd³⁺ ions to the

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luminescence dopant. There were several works tried to change some elements in (Gd,Y)₃(Al,Ga)₅O₁₂ garnets to optimize their properties, particularly luminescence and radiation detecting potential (Ueda and Tanabe, 2019). It is very attractive to replace Y³⁺ (0.90 Å) by Lu³⁺ (0.98 Å) in the garnet's dodecahedral site because both belong to the rare-earth element group and possess similar ionic radii (Ziolkowski, 1985). Furthermore, Sc³⁺ located at the octahedral site as same as Ga³⁺ in this garnet has reported (Ueda and Tanabe, 2019). The luminescence Ce³⁺ ion exhibits characteristic signature 4f-5d transitions, resulting in strong emission in the ultraviolet (UV) - visible light (VIS) with a short decay time. That emission region nicely responds to the photo-multiplier tube (PMT) commonly used in the commercial radiation detector or imaging system. The 5d state of Ce³⁺ can be perturbed by the crystal field and coordination with surrounding atoms/ions because its 5d orbital is not covered by 5s / 5p unlike 4f one. It results to the 5d state distribution and the spectroscopic shift based on the Ce³⁺ lattice site (Ueda and Tanabe, 2019). Consequently, there were several researches about the Ce³⁺-doped phosphors such as BaF₂:Ce³⁺ (Gavhane *et al.*, 2019), CaSc₂O₄:Ce³⁺ (Kang *et al.*, 2019), BaB₄O₇:Ce³⁺ (Gavhane *et al.*, 2020), Li₂Ca₂Si₂O₇:Ce³⁺ (Wu *et al.*, 2020), and Gd₂YAl₂Ga₃O₁₂:Ce³⁺ (Asami *et al.*, 2018) for the radiation detection applications.

This work investigated the structural, optical, photoluminescence (PL) and radioluminescence (RL) properties of Gd₂Lu_(1-x)Ce_xAl₄ScO₁₂ phosphors. The influence of Ce content on these properties was evaluated to determine the optimal phosphor composition for X-ray detecting applications.

Experiment and Method

The Gd₂Lu_(1-x)Ce_xAl₄ScO₁₂ phosphors were synthesized by a solid state reaction in ambient atmosphere. The Ce amount (x) in each phosphor sample was different as 0.00, 0.025, 0.050, 0.075 and 0.100 substituted on the Lu content (1-x). The samples were labeled as Ce0, Ce025, Ce050, Ce075 and Ce100, respectively. The raw chemicals such as Gd₂O₃, Lu₂O₃, CeO₂, Al₂O₃, and Sc₂O₃ were mixed in stoichiometry and merged with acetone before they were taken for ball-milling at 200 rpm for 2 h. The mixtures were compressed into tablets using a hydraulic press at 50 MPa. At first thermal process, all samples were calcined at 1000°C for 60 h in a furnace before being ground into powder. After that, the powders were pressed by the same hydraulic system to be tablets again and were sintered at

1600°C for 10 h as a second thermal process. Once sintered, the phosphors cooled down on their own to room temperature.

The structural properties of phosphors were studied using an X-ray diffractometer (Shimadzu, XRD-6100) with the CuK_α radiation source (wavelength ~1.54 Å). A UV - VIS - near infrared (NIR) spectrophotometer (Shimadzu, UV-3600) was used to measure the absorption spectra of samples. The phosphor's PL emission and excitation spectra were measured using a fluorescence spectrophotometer (Hitachi, F-4600). A fluorescence lifetime measuring device (Horiba Scientific, DD-290) with a 286 nm diode laser source was operated to study the PL decay curves of the phosphors. The RL spectra in process of X-ray induced optical luminescence were measured using a setup comprising a Cu-target X-ray generator operated at 50 kV and 30 mA, an optical fiber - spectrometer (Ocean Optics, QE65 Pro), and a cooling system.

Result

Phosphor sample

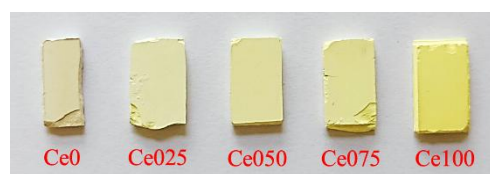


Figure 1. Photograph of Gd₂Lu_(1-x)Ce_xAl₄ScO₁₂ phosphor samples.

In Figure 1, the Gd₂Lu_(1-x)Ce_xAl₄ScO₁₂ phosphors are presented as yellow rectangular samples. The yellow color of phosphor possibly related to the VIS absorption in blue region. The heat treatment and tablet press during the synthesis made the powders combine as bulk materials which were convenient for measuring their spectroscopic properties.

Structure

The X-ray diffraction (XRD) patterns of phosphors are exhibited in Figure 2(a). All samples possessed two-phases of the crystalline structure consistent with the patterns of Gd₃Al₅O₁₂ (ICDD 00-032-0383; red square) and GdAlO₃ (ICDD 00-046-0395; blue circle) (Wang, 1994) data base. A Gd₃Al₅O₁₂ lattice is a cubic crystalline structure in *Ia* $\bar{3}$ *d* space group that is good agreement with the typical {A}₃[B]₂(C)₃O₁₂ garnet structure (Ueda and Tanabe, 2019). The dodecahedral {A} site is occupied by Gd³⁺ either Lu³⁺ ions. The octahedral

[B] site could be occupied by both Al^{3+} or Sc^{3+} ions, while tetrahedral (C) site is exclusively occupied by Al^{3+} ion. However, the ICDD 00-032-0383 database reports that the $\text{Gd}_3\text{Al}_5\text{O}_{12}$ can be decomposed to the GdAlO_3 at 1500°C . This is the reason that the GdAlO_3 phase was observed in the phosphor. The crystalline structure of GdAlO_3 was orthorhombic and explained as the perovskite $\{\text{X}\}[\text{Y}]\text{O}_3$ formation within $Pnma$ space group (Kumar *et al.*, 2023). The distorted square-antiprismatic $\{\text{X}\}$ site and octahedral $[\text{Y}]$ site was likely occupied by $\text{Gd}^{3+}/\text{Lu}^{3+}$ and $\text{Al}^{3+}/\text{Sc}^{3+}$, respectively. Due to similar ionic radii, Ce^{3+} dopant could replace $\text{Gd}^{3+}/\text{Lu}^{3+}$ ion at the $\{\text{A}\}$ site of the cubic and the $\{\text{X}\}$ site of the orthorhombic lattice. However, the diffraction patterns had some noises and there were also the unidentified diffraction (U) peaks, as observed in Figure 2(b). Those U peaks likely associated with the derivative structures or defects originated from the mixing of two distinct lattices in the phosphor. The (2 1 1) diffraction peak of the cubic and (0 0 2) of the orthorhombic lattice, which were strong and looked cleaner from U peak than others, were chosen for analysis of diffraction and the crystallite size.

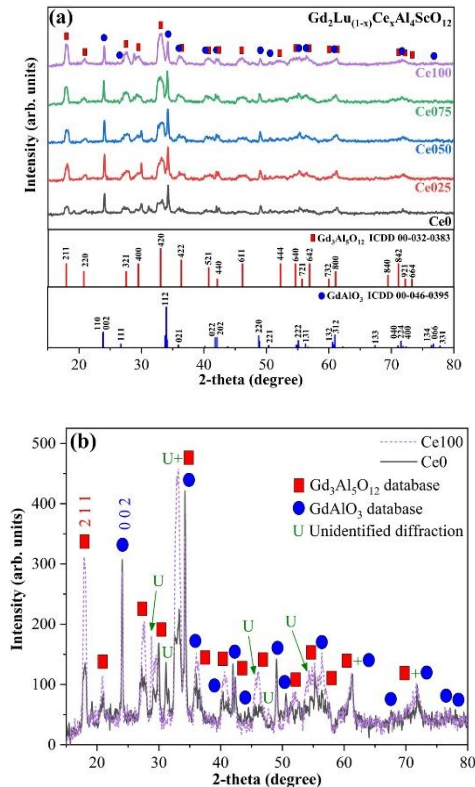


Figure 2. (a) XRD patterns of $\text{Gd}_2\text{Lu}_{(1-x)}\text{Ce}_x\text{Al}_4\text{ScO}_{12}$ phosphors compared with the JCPDS data base of $\text{Gd}_3\text{Al}_5\text{O}_{12}$ (red) and GdAlO_3 (blue) (b) comparative XRD pattern of Ce_0 and Ce_{100} phosphors

The XRD intensities of the (2 1 1) - cubic (I_{Cubic}) and the (0 0 2) - orthorhombic ($I_{\text{Orthorhombic}}$) atomic planes, exhibiting the degree of atomic ordering in each plane, were represented in Figure 3(a). In phosphors with $0.000 \leq x \leq 0.075$, I_{Cubic} was lower than $I_{\text{Orthorhombic}}$, indicating the dominance of orthorhombic plane. However, I_{Cubic} turned to rise more than $I_{\text{Orthorhombic}}$ in the Ce_{100} phosphor. The crystallite size (ε) of the atomic plane in lattice can be evaluated from the full width at half maximum (β) and diffraction angle (θ) of XRD peak by Scherrer's equation (Langford and Wilson, 1978; Kumar *et al.*, 2023);

$$\varepsilon = \frac{k\lambda}{\beta \cos \theta} \quad (1)$$

where k and λ is the Scherrer constant and X-ray wavelength, respectively.

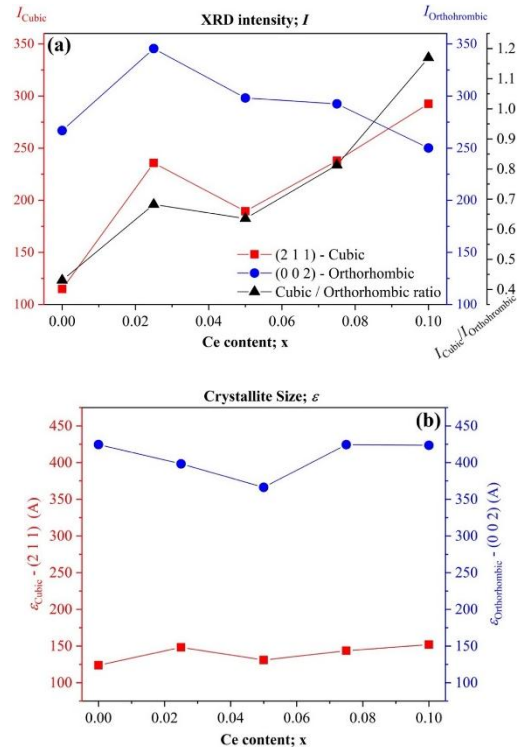


Figure 3. (a) The XRD intensity of (2 1 1) - cubic (I_{Cubic}) and (0 0 2) - orthorhombic ($I_{\text{Orthorhombic}}$). (b) The crystallite size of (2 1 1) - cubic ($\varepsilon_{\text{Cubic}}$) and (0 0 2) - orthorhombic ($\varepsilon_{\text{Orthorhombic}}$) atomic plane of $\text{Gd}_2\text{Lu}_{(1-x)}\text{Ce}_x\text{Al}_4\text{ScO}_{12}$ phosphors.

The crystallite sizes of the (2 1 1) - cubic ($\varepsilon_{\text{Cubic}}$) and (0 0 2) - orthorhombic ($\varepsilon_{\text{Orthorhombic}}$) atomic planes are shown in Figure 3(b). No evident trend was seen in the variation of $\varepsilon_{\text{Cubic}}$ and $\varepsilon_{\text{Orthorhombic}}$ values with varying Ce content. The $\varepsilon_{\text{Cubic}}$

value was ranged from 124 (Ce0) to 152 (Ce100) Å, while the $\varepsilon_{\text{Orthorhombic}}$ was ranged from 366 (Ce005) to 414 (Ce0) Å. The $\varepsilon_{\text{Orthorhombic}}$ were larger than $\varepsilon_{\text{Cubic}}$ in all samples. The crystallite size relates inversely to the crystallite boundary. The orthorhombic lattice in this phosphor compared to the cubic lattice exhibits larger crystallites, resulting in less crystallite boundary. Furthermore, Ce050 sample has the degraded quality in crystalline structure, observed from the drop of XRD intensity and crystallite size in Figure 3(a and b).

Optical absorption

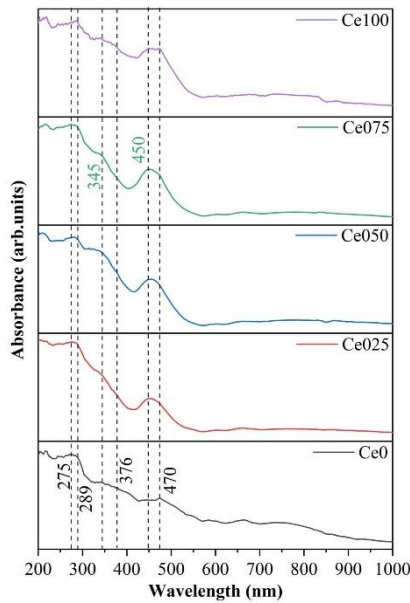


Figure 4. The absorption spectra of Gd₂Lu_(1-x)Ce_xAl₄ScO₁₂ phosphors.

The absorption spectra of the phosphors, measured in the wavelength range of 200 - 1000 nm, are presented in Figure 4. There was the strong absorption in the UV - VIS region for Ce0 sample, particularly at peaks / kinks around 275, 289, 376 and 470 nm. The absorption peak at 275 nm corresponds to the $^8S_{7/2} \rightarrow ^6I_{17/2}$ transition of the Gd³⁺ ion (Carnall *et al.*, 1968), while the kink at 289 nm likely originates from either the band gap transition or exciton transition. Photons of 376 and 470 nm may be absorbed by the ambiguous discrete energy (ADE) of derivative structures or defects. For phosphors doped with Ce, additional absorption peaks at approximately 345 and 450 nm correspond to the $4f \rightarrow 5d_2$ and $4f \rightarrow 5d_1$ transitions (Asami *et al.*, 2018) respectively. These peaks overlap with the host absorption region. The change of absorption strength by varying Ce content is not in any trend. The strong VIS absorption in the blue region by the phosphors is responsible for their yellow color.

Photoluminescence

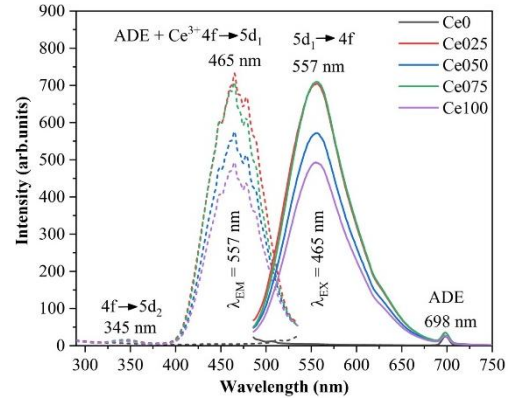


Figure 5. The PL emission spectra with $\lambda_{\text{EX}} = 465$ nm (solid line) and excitation spectra with $\lambda_{\text{EM}} = 557$ nm (dash line) of Gd₂Lu_(1-x)Ce_xAl₄ScO₁₂ phosphors.

To study the $4f - 5d_1$ transition of Ce³⁺, the PL spectra with emission wavelength (λ_{EM}) at 557 nm and excitation wavelength (λ_{EX}) at 465 nm were measured and shown in Figure 5. The emission spectra under $\lambda_{\text{EX}} = 465$ nm consisted of a broad and smooth peak at 557 nm in Ce-doped samples, while all samples showed a small peak at 698 nm. The peak at 557 nm corresponds to the $5d_1 \rightarrow 4f$ transition of Ce³⁺ (Asami *et al.*, 2018; Tiana *et al.*, 2022), while the peak at 698 nm is possibly due to a transition of ADE in phosphor structure. For excitation spectra with $\lambda_{\text{EM}} = 557$ nm, there were a broad peak at 465 nm with spikes attributed to the $4f \rightarrow 5d_1$ transition of Ce³⁺ ions with additional contributions from ADE, and a tiny 345 nm corresponding to the $4f \rightarrow 5d_2$ transition of Ce³⁺ ions (Ueda and Tanabe, 2019; Asami *et al.*, 2018; Tiana *et al.*, 2022). For the mechanism of these PL spectra, the VIS photon with 465 nm could excited Ce³⁺ ion and ADE in structure. The electrons in Ce³⁺ ions were excited from the ground $4f$ state to the $5d_1$ orbital and then non-radiatively relaxed to the lowest vibrational state available within the $5d_1$. Finally, the $5d_1 \rightarrow 4f$ transition of Ce³⁺ generated the broad emission centered around 557 nm. The ADE could also be excited by such VIS photon and this excitation energy transferred to Ce³⁺ ion for emission respectively. Under PL with $\lambda_{\text{EX}} = 465$ nm and $\lambda_{\text{EM}} = 557$ nm, the Ce025 and Ce075 samples exhibited higher PL intensity compared to other samples. The decreased PL intensity for the Ce050 phosphor could be attributed to its degraded crystalline quality previously mentioned. Additionally, concentration quenching caused the lowest PL intensity in the Ce100 sample.

For the investigation of PL through Gd³⁺ excitation, the PL emission spectra under $\lambda_{\text{EX}} = 275$

nm are presented in Figure 6(a). The PL spectra exhibited three distinct groups: a sharp peak at 312 nm, a board peak centered around 362 nm, and the spike peaks with a maximum intensity at 469 nm. The UV with 275 nm provided the $^8S_{7/2} \rightarrow ^6I_{17/2}$ excitation of Gd^{3+} before this ion non-radiatively relaxed to $^6P_{7/2}$ state. At this point, some Gd^{3+} emitted a sharp peak at 312 nm through the $^6P_{7/2} \rightarrow ^8S_{7/2}$ transition, some could transfer their energy to Ce^{3+} (Kaewnuam *et al.*, 2022) and some probably transferred their energy to the ADE. The Ce^{3+} acceptor might emit photons through the $5d_2 \rightarrow 4f$ transition at 362 nm (Ueda and Tanabe, 2019; Kaewnuam *et al.*, 2022). However, the Ce0 sample also exhibited this peak. The board emission peak at 362 nm was likely a superposition of the $5d_2 \rightarrow 4f$ from Ce^{3+} and the ADE emission from derivative structures or defects. Additionally, these ADE acceptors also generated the emissions as spike peaks with the highest one at 469 nm. The strongest emission at 312 (Ce^{3+}), 362 (ADE+ Ce^{3+}), and 469 (ADE) nm belonged to Ce0, Ce075 and Ce025 sample, respectively.

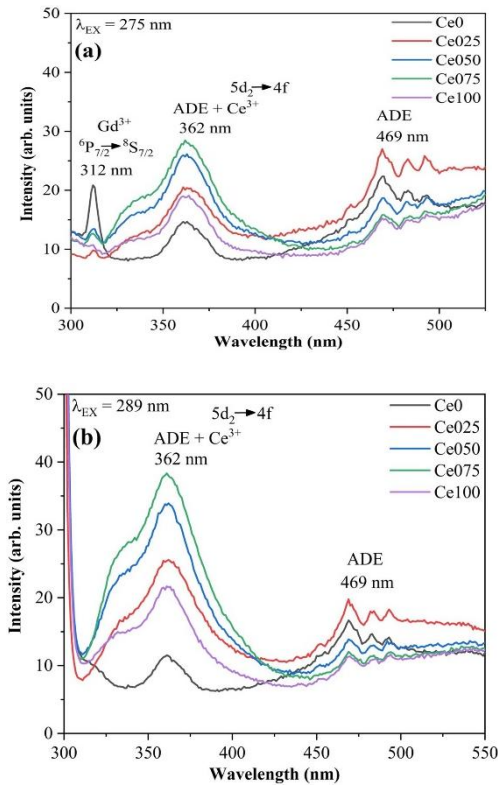


Figure 6. (a) The PL emission spectra with $\lambda_{EX} = 275$ nm and (b) $\lambda_{EX} = 289$ nm of $Gd_2Lu_{(1-x)}Ce_xAl_4ScO_{12}$ phosphors.

Since UV with 289 nm could generated the band gap transition or exciton of phosphor, it was also used as λ_{EX} to study the PL emission spectra, as

shown in Figure 6(b). Their patterns were like PL spectra with $\lambda_{EX} = 275$ nm, but lacked the Gd^{3+} emission peak. After electron-hole or exciton generation, the charge carrier migrated to the discrete-energy center such as Ce^{3+} ion and derivative structures / defects. Then, the $5d_2 \rightarrow 4f$ and ADE transitions generated the PL peaks observed at 362 and 469 nm. The PL intensity under $\lambda_{EX} = 289$ nm was higher than that of $\lambda_{EX} = 275$ nm because generation of electron-hole pairs or excitons was more efficient compared to Gd^{3+} excitation.

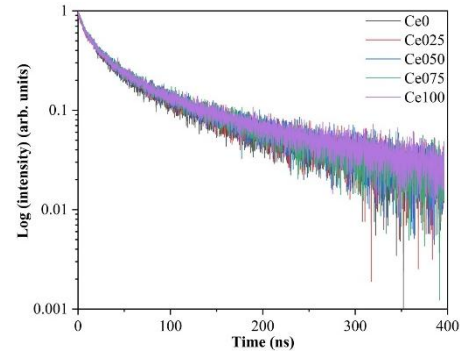


Figure 7. PL decay curves for $Gd_2Lu_{(1-x)}Ce_xAl_4ScO_{12}$ phosphors excited at $\lambda_{EX} = 286$ nm.

Table 1. Fitted parameters from PL decay curves

Sample	1 st -decay component		2 nd -decay component		τ_{ave}
	A_1	τ_1	A_2	τ_2	
Ce0	0.58	12.15	0.35	78.24	64.69
Ce025	0.53	13.09	0.35	86.27	72.72
Ce050	0.55	12.44	0.36	84.67	71.62
Ce075	0.56	13.01	0.36	86.79	72.85
Ce100	0.54	12.15	0.36	82.61	69.91

In Figure 7, the PL decay profiles of all phosphors excited at $\lambda_{EX} = 286$ nm are presented and exhibit features of a double-components exponential decay, which was therefore used for fitting with the following expression (Lu *et al.*, 2023);

$$I = A_0 + A_1 e^{(-t/\tau_1)} + A_2 e^{(-t/\tau_2)} \quad (2)$$

where A_0 , A_1 and A_2 are the arbitrary constant, 1st-decay constant, and 2nd-decay constant, respectively. Additionally, τ_1 and τ_2 denote the 1st-decay time and 2nd-decay time, respectively. The average decay time (τ_{ave}) was evaluated by using the following relation (Lu *et al.*, 2023);

$$\tau_{ave} = \frac{A_1 \tau_1^2 + A_2 \tau_2^2}{A_1 \tau_1 + A_2 \tau_2} \quad (3)$$

The parameter values obtained from fitting are listed in Table 1. The values of 1st component-decay time were more similar and showed little change with increasing Ce content, which suggested their association with the luminescence time of ADE within structure. For the 2nd component, the slow τ_2 values in Ce-doped phosphors were longer compared to the undoped CeO sample. The 2nd decay in undoped CeO sample likely originates from another ADE emission, while it was superposed with 4f-5d emission of Ce³⁺ in Ce-doped phosphors. The existence of only ADE in 1st component-decay and mixed ADE+Ce³⁺ on 2nd component-decay profile corresponds to the PL spectra with $\lambda_{EX} = 289$ nm in Figure 6(b). Such PL peaks consist of two groups, only ADE centered around 469 nm and mixed ADE+Ce³⁺ centered around 362 nm. The average decay times of phosphors were ranged from 65 to 73 ns, that the Ce-doped samples had higher value than the undoped one. This relatively short decay time makes these phosphors suitable for radiation detection material.

Radioluminescence

To determine the potential of these phosphors as X-ray detection material, their RL spectra were consequently measured and shown in Figure 8. The peaks of the RL emission spectra can be grouped into four: The first one at 313 nm is under $^6P_{7/2} \rightarrow ^8S_{7/2}$ transition of Gd³⁺; The second peak around 368 nm possibly corresponds to the 5d₂→4f transition of Ce³⁺ dopant; The third one at 575 nm is dominated by the 5d₁→4f transition of Ce³⁺, with less superposed from ADE on the longer wavelength; The fourth group, complicated and mixed emission peaks in a broad range of 530 - 990 nm, is likely dominated by ADE and the part on the left side overlapped with the third emission group. The undoped CeO sample owned the first (Gd³⁺) and fourth (only ADE) groups of RL peak. The fourth group resembles the RL peak observed in undoped GdYA₁₂GaO₁₂ phosphors in our previous work (Kaewnuam *et al.*, 2023). All four emission groups were observed in Ce-doped samples. For RL mechanism, the induction by X-ray generated the electron - hole pairs through photoelectric effect and Compton scattering in phosphors. The electron and hole are separated to occupy in the conduction and valence band, respectively. Those electron and hole can transfer to combine at the discrete energy levels of Ce³⁺ ion, Gd³⁺ ion, or the derivative structure / defects (ADE). Such electron-hole recombination finally causes the luminescence (Nikl, 2006). Some electron - hole pairs recombined at the inter 5d-4f level of Ce³⁺ ion, some at intra 4f-4f orbital of Gd³⁺ ion, and others at ADE of derivative structures / defects. After that, the Ce³⁺ generated emissions at

362 and 575 nm by itself and by absorbing the energy transferred from the Gd³⁺. The electron - hole recombination at Gd³⁺ generated the emission at 312 nm and possibly transferred energy to Ce³⁺ for luminescence. Since this phosphor structure was complicated by the mixed lattices of cubic, and orthorhombic, and the possible existence of derivative structures / defects, the ADE were consequently fluctuated and superposed. The electron - hole recombination at the ADE produced multiple non-Gaussian emission peaks between 530 - 990 nm. The highest intensity for the first RL peak was observed at 313 nm in the Ce025 sample. Similarly, the Ce025, Ce050, and Ce100 samples exhibited the most intense peaks at 368 nm, 575 nm, and 727 nm, respectively. The samples with highest RL intensity were differed from PL spectra, because X-ray likely induced damage or disturbance on the phosphor structure that impacted emission transitions. The Ce050 phosphor exhibited highly promising characteristics for X-ray detecting applications due to its strong RL intensity arising from both 4f-5d transitions of Ce³⁺ and its significant spectral overlap with the standard BGO crystal scintillator.

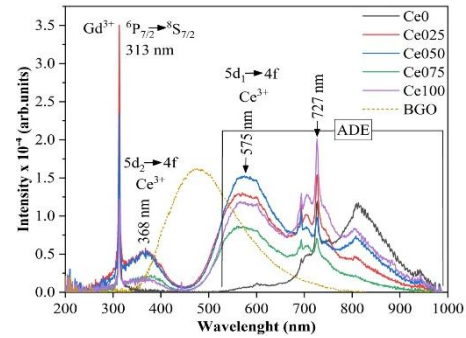


Figure 8. The RL spectra under X-ray induction of Gd₂Lu_(1-x)Ce_xAl₄ScO₁₂ phosphors

Conclusion

The Gd₂Lu_(1-x)Ce_xAl₄ScO₁₂ phosphors were synthesized by solid state reaction, resulting in two crystalline structures: a cubic lattice with *Ia3d* space group and an orthorhombic lattice with *Pnma* space group. The derivative structures and defects were also observed in all phosphor samples. The CeO₂ addition enhanced the construction of (2 1 1) - cubic atomic plane compared to (0 0 2) - orthorhombic atomic plane. The Ce³⁺, Gd³⁺ and other components in phosphors absorbed the photons in UV and VIS regions. Under 465 nm excitation, a strong photoluminescence (PL) emission peak at 557 nm was observed due to the 4f-5d₂ transition of Ce³⁺ ions, particularly in the Ce025 and Ce075

phosphors. Upon Gd^{3+} excitation at 275 nm, energy transfer occurred from Gd^{3+} to Ce^{3+} and derivative structures / defects. This resulted in the strongest emission peaks at 362 nm and 469 nm in the Ce075 phosphor, attributed to the 4f-5d₁ and ADE transitions. The expected band gap or exciton excitation by UV with 289 nm led to the emission similar to that observed with Gd^{3+} excitation. The decay curves of phosphors under 286 nm excitation exhibited double exponential behavior. Their average decay times were around 65 – 73 ns, likely due to a superposition of the ADE and 4f-5d transition. The RL spectra under X-ray induction compose of multiple emission peaks from four groups, such as 312 nm ($^6P_{7/2} \rightarrow ^8S_{7/2}$ of Gd^{3+}), 368 nm ($5d_2 \rightarrow 4f$ of Ce^{3+}), 575 nm ($5d_1 \rightarrow 4f$ of Ce^{3+}) and 530 - 990 nm (ADE). The highest RL intensities were different from those in the PL, likely due to the structural disturbances caused by X-ray. Since strong RL emission intercepted in RL range of BGO crystal and its short decay time, $Gd_2Lu_{0.95}Ce_{0.05}Al_4ScO_{12}$ phosphor demonstrates potential as an X-ray detecting material for the X-ray imaging screen and detector.

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References

- Asami, K., Ueda, J., Kitaura, M., and Tanabe, S. (2018). Investigation of luminescence quenching and persistent luminescence in Ce^{3+} doped $(Gd,Y)_3(Al,Ga)_5O_{12}$ garnet using vacuum referred binding energy diagram. *Journal of Luminescence*, 198:418-426. <https://doi.org/10.1016/j.jlumin.2018.01.041>
- Ashurov, M.K., Voronko, Y., Osiko, V.V., Sobol, A.A., and Timoshechkin, M.I. (1977). Spectroscopic study of stoichiometry deviation in crystals with garnet structure. *Physica Status Solidi (a)*, 42(1):101-110. <https://doi.org/10.1002/pssa.2210420108>
- Carnall, W.T., Fields, P.R., and Rajnak, K. (1968). Electronic energy levels of the trivalent lanthanide aquo ions. II. Gd^{3+} . *Journal of Chemical Physics*, 49(10):4443-4446. <https://doi.org/10.1063/1.1669894>
- Chen, X., Liu, X., Feng, Y., Li, X., Chen, H., Xie, T., Kou, H., Kucerkova, R., Beitlerova, A., Mihokova, E., Nikl, M., and Li, J. (2020). Microstructure evolution in two-step-sintering process toward transparent $Ce:(Y,Gd)_3(Ga,Al)_5O_{12}$ scintillation ceramics. *Journal of Alloys and Compounds*, 846:156377. <https://doi.org/10.1016/j.jallcom.2020.156377>
- Chewpraditkul, W., Sakthong, O., Pattanaboonmee, N., Chewpraditkul, W., Szczesniak, T., Swiderski, L., Moszynski, M., Kamada, K., Yoshikawa, A., and Nikl, M. (2017). Optical and scintillation characteristics of $Gd_2YAl_2Ga_3O_{12}:Ce$ and $Lu_2YAl_2Ga_3O_{12}:Ce$ single crystals. *Journal of Crystal Growth*, 468:395-398. <https://doi.org/10.1016/j.jcrysgro.2016.10.086>
- Gavhane, K.H., Bhadane, M.S., Bhoir, A.S., Kulkarni, P.P., Dahiwal, S.S., Patil, B.J., Bhorkar, V.N., Dhole, S.D., and Dahiwal, S.S. (2020). T_m - T_{stop} analysis and dosimetric properties of Ce doped $BaBaO_7$ phosphor. *Journal of Alloys and Compounds*, 817:152805. <https://doi.org/10.1016/j.jallcom.2019.152805>
- Gavhane, K.H., Bhadane, M.S., Jadhav, A.P., Patil, B.J., Bhorkar, V.N., Dahiwal, S.S., and Dhole, S.D. (2019). TL properties of $BaF_2:Ce$ phosphor for high gamma ray dosimetry. *Journal of Luminescence*, 209:316-320. <https://doi.org/10.1016/j.jlumin.2019.02.011>
- Kaewnuam, E., Angnanon, A., Wantana, N., and Kaewkhao, J. (2023). The study and development on $Gd_2Y_{(1-x)}Dy_xAl_2Ga_3O_{12}$ phosphor for x-ray detection material. *Radiation Physics and Chemistry*, 202:110508. <https://doi.org/10.1016/j.radphyschem.2022.110508>
- Kaewnuam, E., Wantana, N., Ruangtaweep, Y., Cadatal-Raduban, M., Yamanoi, K., Kim, H.J., Kidkhunthod, P., and Kaewkhao, J. (2022). The influence of CeF_3 on radiation hardness and luminescence properties of $Gd_2O_3-B_2O_3$ glass scintillator. *Scientific Reports*, 12:11059. <https://doi.org/10.1038/s41598-022-14833-3>
- Kamada, K., Yanagida, T., Pejchal, J., Nikl, M., Endo, T., Tsutsumi, K., Fujimoto, Y., Fukabori, A., and Yoshikawa, A. (2011). Scintillator-oriented combinatorial search in Ce-doped $(Y,Gd)_3(Ga,Al)_5O_{12}$ multicomponent garnet compounds. *Journal of Physics D: Applied Physics*, 44(50):505104. <https://doi.org/10.1088/0022-3727/44/50/505104>
- Kang, T., Lim, S., Lee, S., Kang, H., Yu, Y., and Kim, J. (2019). Luminescent properties of $CaSc_2O_4:Ce^{3+}$ green phosphor for white LED and its optical simulation. *Optical Materials*, 98:109501. <https://doi.org/10.1016/j.optmat.2019.109501>
- Kaura, J., Rania, S., and Lal, B. (2020). Luminescence properties of Eu^{3+} doped gadolinium aluminum garnet phosphors. *Optik - International Journal for Light and Electron Optics*, 212:164745. <https://doi.org/10.1016/j.ijleo.2020.164745>
- Kumar, P., Singh, D., Gupta, I., Singh, S., and Kumar, V. (2023). Structural and luminescent characteristics of orthorhombic $GdAlO_3:Sm^{3+}$ nanocrystalline materials for solid state lighting. *Chemical Physics Letters*, 812:140277. <https://doi.org/10.1016/j.cplett.2022.140277>
- Langford, J.I., and Wilson, A.J.C. (1978). Scherrer after sixty years: A survey and some new results in the determination of crystallite size. *Journal of Applied Crystallography*, 11(2):102-113. <https://doi.org/10.1107/S0021889878012844>
- Lu, S., Hu, S., Hong, Y., Wang, B., Li, W., He, Q., and Wang, Y. (2023). Blue luminescent properties of Ce^{3+}/Gd^{3+} co-doped $NaBaBO_3$ phosphors. *Chemical Physics*, 566:111789. <https://doi.org/10.1016/j.chemphys.2022.111789>
- Nikl, M. (2006). Scintillation detectors for x-rays. *Measurement Science and Technology*, 17(4):R37-R54. <https://doi.org/10.1088/0957-0233/17/4/R01>
- Ou, Y., Zhou, W., Ma, F., Liu, C., Zhou, R., Su, F., Huang, Y., Dorenbos, P., and Liang, H. (2020). Luminescence tuning of Ce^{3+} , Pr^{3+} activated $(Y,Gd)AGG$ system by band gap engineering and energy transfer. *Journal of Rare Earths*, 38(5):514-522. <https://doi.org/10.1016/j.jre.2020.01.001>

- Tian, Y., Chen, J., Yi, X., Jiang, R., Lin, H., Chen, L., and Zhou, S. (2022). Unravel the effect of lattice distortion on the 4f-5d excitation of Ce³⁺ in garnet phosphors. *Journal of Alloys and Compounds*, 907:164412. <https://doi.org/10.1016/j.jallcom.2022.164412>
- Ueda, J., and Tanabe, S. (2019). Review of luminescent properties of Ce³⁺-doped garnet phosphors: New insight into the effect of crystal and electronic structure. *Optical Materials*, 90:100018. <https://doi.org/10.1016/j.omx.2019.100018>
- Wang, P. (1994). Shanghai Institute of Ceramics, Chinese Academy of Sciences, Shanghai, China. ICDD Grant-in-Aid.
- Wu, Q., He, Z., Huang, Q., Zhao, Q., Zheng, P., Xiang, D., Ding, J., and Zhou, J. (2020). The electronic structure, site occupancy and luminescent properties of Ce³⁺-activated Li₂Ca₂Si₂O₇ blue phosphor. *Ceramics International*, 46(4):4511-4518. <https://doi.org/10.1016/j.ceramint.2019.10.178>
- Ziolkowski, J. (1985). New relation between ionic radii, bond length, and bond strength. *Journal of Solid State Chemistry*, 57(3):269-290. [https://doi.org/10.1016/0022-4596\(85\)90152-5](https://doi.org/10.1016/0022-4596(85)90152-5)