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Fabrication and Scintillation Performance of Nonstoichiometric LuAG:Ce Ceramics

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Nonstoichiometric LuAG:Ce Ceramics ([Lu$_{(1-x)}$Ce$_x$]$_3$Al$_5$O$_{12}$, $x = 0.005$) with different excess of Lu^{3+} were designed on the basis of Lu$_2$O$_3$-Al$_2$O$_3$ phase diagram and fabricated by a solid-state reaction method. Without using any traditional sintering aids, pure phase and good optical performance were obtained in such a Lu-rich LuAG:Ce ceramics. In addition, scintillation efficiency and light yield of 1% excess of Lu^{3+} ceramic sample were found 16 times and 1.82 times higher than that of commercial Bi$_4$Ge$_3$O$_{12}$ (BGO) single crystals, respectively. Such values are comparable or even better than those in most of LuAG:Ce single crystals. However, antisite defects were also induced by excess of Lu doping, whose luminescence was found at 350–410 nm in Lu-rich LuAG:Ce ceramics. The relationship of excess content of Lu and the microstructure, optical quality, and scintillation performance were clarified and discussed. Furthermore, by utilizing X-ray absorption near edge spectroscopy technique, the charge state stability of cerium in Lu-rich LuAG:Ce ceramics was examined. It appears that the excess of isovalence Lu^{3+} doping has a negligible effect on the cerium valence instability and creation of stable Ce^{4+} center.

I. Introduction

THE development of inorganic scintillators for the detection of X-ray and γ-ray photons, accelerated charged particles or even neutrons has a very long history.[1,2] Within the last two decades, a number of excellent scintillators based on Ce^{3+} and Pr^{3+}-doped materials were discovered and systematically studied, triggered mainly by industrial interest, such as the X-ray computed tomography (X-CT) and positron emission tomography (PET) in modern medical imaging and calorimetry in high-energy physics (HEP).[3–5] Among them, the Ce-doped Lu$_3$Al$_5$O$_{12}$ (LuAG:Ce) aluminum garnet attracted high attention for its great scintillation performance and was found as a promising candidate for the scintillation applications.[6] The high density and effective atomic number (ρ = 6.7 g/cm^3, Z$_{eff}$=63) of LuAG:Ce are advantageous in the case of hard X- and γ-ray detection. Fast and efficient luminescence is assured by the allowed $5d^1 \rightarrow 4f$ transition of Ce^{3+} ion the dominant emission of which located at around 520 nm matches well the Si photodiode spectral sensitivity.

However, the scintillation performance of LuAG:Ce single crystals was limited by the presence of Lu$_{Al}$ antisite defect and low Ce concentration in LuAG lattice. The latest Lu$_2$O$_3$–Al$_2$O$_3$ binary phase diagram showed that there existed a nonstoichiometric phase in LuAG garnet lattice and the concentration homogeneity range was about 4 mass % of Lu$_2$O$_3$.[7] Such a phenomenon could be ascribed to the presence of antisite defect (AD, part of Lu ions substituted the octahedral-coordinated sites of Al ions in the garnet lattice) in garnet single crystals grown from melt. With the help of atomistic calculation, RE$_{Al}$ antisite defect was found as the lowest energy intrinsic defect in both YAG and all other REAGs, (RE ranging from Lu to Gd).[8,9] Experimental studies have shown that Lu$_{Al}$ AD tended to occur more easily than Y$_{Al}$ due to higher growth temperature of LuAG single crystals and smaller misfit between the Al^{3+} and Lu^{3+} ionic radii. Unfortunately, numerous studies showed that migrating electrons could be retrapped at shallow traps induced by such AD, before their radiative recombination with holes at the Ce^{3+} centers. As a result, the light yield and scintillation response of LuAG:Ce single crystals were degraded seriously.[10,11] It seems that AD is an inevitable consequence of the growth from high-temperature melt. It can be fully eliminated in LuAG:Ce single crystalline films grown by liquid phase epitaxy due to much lower preparation temperature.[12] In addition, low segregation coefficient of Ce^{3+} ions in LuAG lattice is detrimental to the growth of high Ce^{3+} concentration and homogenously doped LuAG single crystals. It was shown that the light yield of LuAG:Ce single crystals can reach 26000 ph/MeV when 0.55 at.% Ce^{3+} were incorporated in high optical quality crystals, which is much higher than that achieved in most of LuAG:Ce single crystals doped only with ~0.1 at.% Ce^{3+} (10000–12500 ph/MeV).[13,14] In this study, we also show that high Ce^{3+} concentration could be accommodated in LuAG lattice due to excess occupancy by Lu^{3+} at octahedral Al^{3+} sites.

Due to the lower preparation temperature, AD was not detected in LuAG:Pr optical ceramics.[15] Indeed, scintillator ceramics were characterized not only by lower preparation temperature but also by uniform high concentration doping and simple preparation process. However, LuAG:Ce optical ceramics were first reported and fabricated by means of technologies providing the light yield of only 50% of that of BGO reference single crystal, even though their radioluminescence (RL) intensity reflecting intrinsic scintillation efficiency was about 10 times of BGO.[16–19] Compared with the detrimental effect of worse optical transmittance and grain boundaries, sintering aids were reported as the key factor to deteriorate the scintillation performance of ceramics, although they are essential to obtain highly transparent ceramics in most cases. The simultaneous use of traditional

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aliovalent sintering aids, such as $(C_2H_5)_4Si$ (tetraethyl ortho-silicate, TEOS) and MgO, caused serious point defects due to the charge compensation effects. Even the light yield of LuAG:Pr ceramics manufactured with isovalent sintering aids (La_2O_3, Sc_2O_3) was still lower than that of BGO reference single crystals.[20] It seems that sintering aids either entering the crystal lattice or segregating at grain boundaries could give rise to different kinds of charge carrier traps in the forbidden gap, thus effectively slowing the energy transfer to luminescence centers.

In this study, we reported another efficient way to fabricate highly transparent LuAG:Ce ceramics with good scintillation performance as well: inducing excess of Lu to the lattice. By adjusting the content of excess of Lu, nonstoichiometric LuAG:Ce ceramics were successfully fabricated without adding any other sintering aids. The LuAG:Ce ceramics with excess of Lu show significantly improved both optical quality and scintillation performance. We then discussed the current understanding of the formation of AD in LuAG ceramics and the role of excess of Lu in the fabrication and scintillation process of LuAG:Ce optical ceramics. Furthermore, the valence stability of cerium in Lu-rich LuAG:Ce ceramics was also examined by an advanced XANES technique.

II. Experimental Procedure

Commercial powders of CeO_2, Lu_2O_3, and α-Al_2O_3 (>99.99% purity, Alfa Aesar, Ward Hill, MA) were used as starting materials. In all experiments, CeO_2 and α-Al_2O_3 were weighted according to the chemical stoichiometry of $(Ce_xLu_{1-x})_3$ Al_5O_{12}, where $x = 0.005$. On the contrary, Lu_2O_3 powders were weighted with 1%, 2%, 3%, and 4 wt% in excess. The starting materials were mixed and ball-milled by highly pure Al_2O_3 ceramic balls with anhydrous alcohol for 12 h. After stove-dried and mesh-sieved, the powder mixtures were dry-pressed to form a disk with a diameter of 18 mm and subsequently isostatically cold-pressed at 250 MPa. The powder compacts were vacuum sintered in a vacuum tungsten wire furnace at 1850°C for 20 h and then air-annealed at 1450°C for 20 h. Finally, the resultant ceramics were double polished to a thickness of 1 mm for optical experiments.

The phase composition of the obtained sample was identified by X-ray diffraction (XRD, Model D/MAX-2550V, Rigaku Co., Tokyo, Japan). The microstructure of the ceramic was observed with a scanning electron microscopy (SEM, JSM-6390, JEOL, Tokyo, Japan). Mirror-polished samples on both surfaces with the thickness of 1.0 mm were used to measure the optical in-line transmittance (Cary 5000, UV-VIS-NIR spectrophotometer, Varian Inc., Melbourne, Australia). The X-ray excited luminescence spectra were conducted on an X-ray excited luminescence spectrometer, assembled at Shanghai Institute of Ceramics, Chinese Academy of Sciences. To determine the scintillation light yield (LY), the energy spectra were collected under the 662 keV gamma-ray excitation (^{137}Cs source) and detected by a photomultiplier (R878, Hamamatsu, Hamamatsu City, Japan) supplied by -1200 V bias. The shaping time was 1 and 0.75 μs. X-ray absorption near edge structure (XANES) spectra was recorded at Beijing Synchrotron Radiation Facility (BSRF). Measurements were recorded at room temperature, at the Ce L_{III}-edge (5723 eV).

III. Results and Discussion

As shown in Fig. 1, Lu-rich LuAG:Ce transparent ceramics were fabricated successfully without the use of any other traditional sintering aids. All the samples displayed high and uniform optical quality. The transmission spectra of as-sintered samples are given in Fig. 2. We can see that the transmittance increased with increasing Lu content. It is well-known that the beneficial role of sintering aids exploited in optical ceramics is due to their ability to reduce scattering
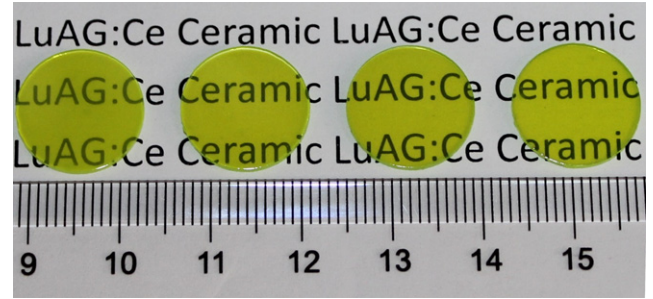

Fig. 1. Photograph of LuAG:Ce ceramics with various excess of Lu^{3+} (from left to right, 1%, 2%, 3%, and 4 at.% excess of Lu^{3+} samples).
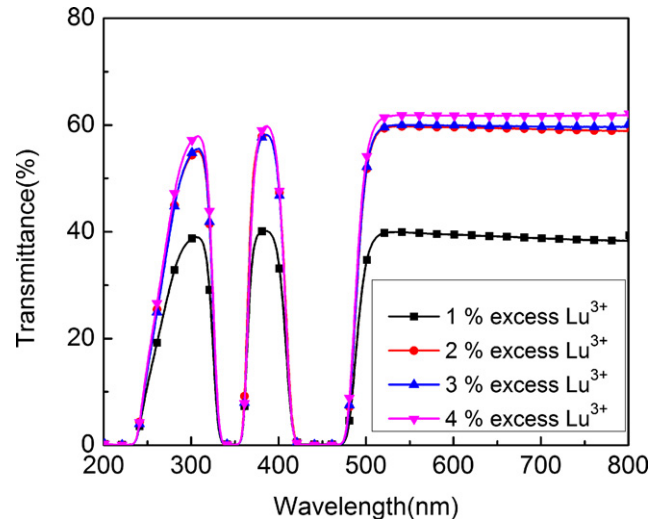

Fig. 2. The in-line transmittance of Ce:LuAG ceramics with various excess of Lu^{3+}.

centers related to the amount of residual pores, therefore improving transparency. As the excess of Lu_2O_3 in this study played the same role as traditional sintering aids, we call this "self-sintering aid" phenomenon. According to the Lu_2O_3–Al_2O_3 phase diagram,[7] the liquid phase should not be formed in the range of sintering temperatures of LuAG:Ce ceramic when excess of Lu was added to the mixture. Since LuAG garnet is a defect sensitive material, further lattice distortion and structural disorder might be the possible factors which enhance the sintering process of LuAG:Ce ceramics. However, further studies especially on the sintering kinetics is still needed to understand nature of the effect of Lu excess in the sintering process.

In the research work of Kanai *et al.* which related to the nonstoichiometric $Gd_{3+\delta}(Al,Ga)_{5-\delta}O_{12}$:Ce garnet scintillators, $GdAlO_3$ and β-Ga_2O_3 were observed in the XRD patterns when the composition has slight deviation from stoichiometry.[21] The XRD patterns of nonstoichiometric LuAG:Ce ceramics with various excess of Lu are shown in Fig. 3(a). They agree well with the standard card of $Lu_3Al_5O_{12}$ compound (JCPDS 73-1368), showing the single cubic garnet phase. This reveals that 4 at.% excess of Lu^{3+} can be soluble in LuAG lattice without any new obvious phase induced. In addition, based on the difference of the main diffraction peak of these samples, which is shown in Fig. 3(b), it is observed that the diffraction peak clearly shifts toward lower angle with increasing of Lu content. According to the Bragg equation, the shift of diffraction peaks toward lower angle reveals that the cell parameters of samples could continuously increase with the increase of Lu content. According to the Lu_2O_3–Al_2O_3 binary phase diagram,[7] it is discussed that excess of Lu_2O_3 (maximum 4 mass%) could be accommodated in the LuAG lattice
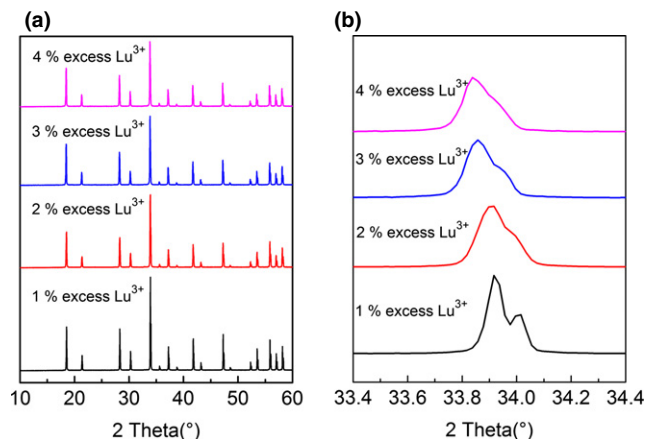
Fig. 3. (a) XRD patterns of LuAG:Ce ceramics with various excess of Lu^{3+}; (b) Expanded view of the 2θ diffraction peak between 33.4° and 34.4°.

Fig. 5. X-ray excited luminescence (XEL) spectra of Ce:LuAG ceramics with various Lu content. BGO single crystal was used for quantitative comparison; In the inset, the expanded view of the XEL spectra between 350 and 410 nm is given.

homogenously without an obvious second phase creation, which was ascribed to the substitution of Lu^{3+} (0.86 Å) to the octahedral site of Al^{3+} (0.54 Å) due to their similar ionic radius in octahedral coordination. Thus, in our case, the introduced excess of Lu^{3+} (from 1% to 4 at.%) can occupy octahedral site of Al^{3+}, and the larger radius of Lu^{3+} undoubtedly leads to a lattice constant increase, as observed in Fig. 3(b).

To investigate the microstructure of LuAG:Ce ceramics with an excess of Lu, SEM were performed on the samples with surface thermally etched at 1500°C for 3 h in air, see Fig. 4. As expected, very dense, almost pore-free microstructures are observed in the ceramic samples. There are no secondary phases observed both at the grain boundaries and inside the grains even for the 4% excess Lu LuAG:Ce ceramics. This interesting phenomenon seems to confirm again the good solubility of Ce^{3+} and Lu^{3+} in the LuAG host lattice.

In Fig. 5, the X-ray excited luminescence spectra of LuAG:Ce optical ceramics with various excess of Lu are shown and compared with a standard BGO scintillation single crystal with the same size. There is an evident trend in decreasing XEL intensity with increasing Lu content when the samples are compared: the integrals of XEL spectra are in the ratio 16:14:12.7:11.5:1 for the 1% excess, 2% excess, 3% excess, 4% excess of Lu samples and BGO single crystal, respectively. This result points to very high scintillation efficiency of Lu-rich samples compared with BGO single crystal. We note that the scintillation efficiency is by factor 1.5–2
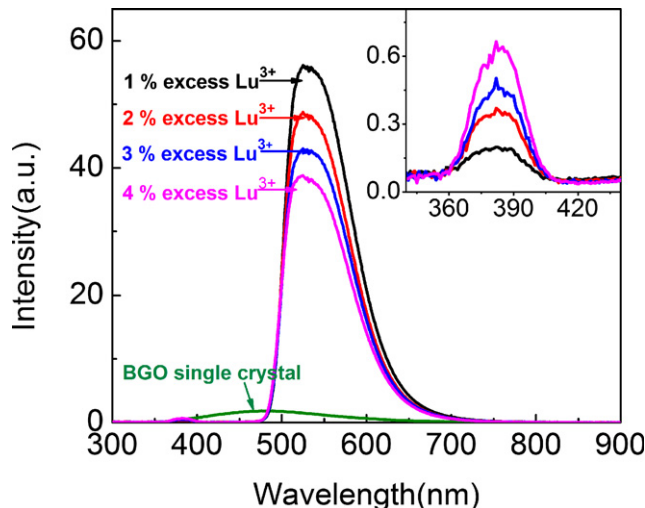
higher than that found in a typical commercial LuAG:Ce single crystal.[11] At the same time, we can see that the XEL intensities of four LuAG:Ce ceramics decrease gradually with the increase of Lu content. An additional luminescence peak located at 350–410 nm was also observed in Lu-rich LuAG:Ce ceramic samples, see the inset of Fig. 5. This result was in agreement with the known "Lu_{Al} antisite" defect-related emission of LuAG:Ce single crystals.[22] Moreover, the intensity of this additional peak increased with increasing content of excess of Lu in ceramic samples, which means that the higher excess Lu content, the stronger AD related luminescence intensity. Therefore, the decrease of scintillation efficiency seems to be related to the increased concentration of Lu_{Al} AD in these nonstoichiometric LuAG:Ce ceramics. However, it is also reported that the decay curves corresponding to AD emission at 225K contained both the fast (150 ns) and slow (1160 ns) components of emission.[12] Further investigations including the decay kinetic measurement and low-temperature thermoluminescence glow curves measurement are still needed to obtain more detailed information of AD effect in these samples.
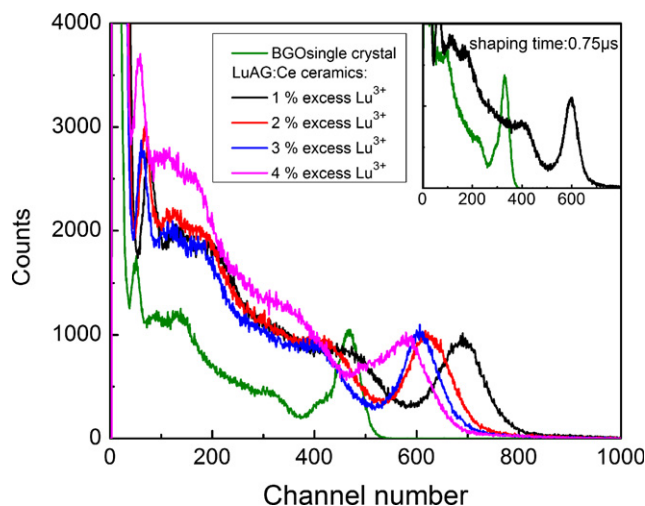

Fig. 6. Comparison of the pulse-height spectra of Lu-rich Ce:LuAG ceramics with BGO single crystal of the same size. The position of the photopeak determines the light yield value. In the inset, there is the light yield of 1 at.% Lu^{3+}-rich LuAG:Ce ceramic measured with 0.75 μs shaping time.
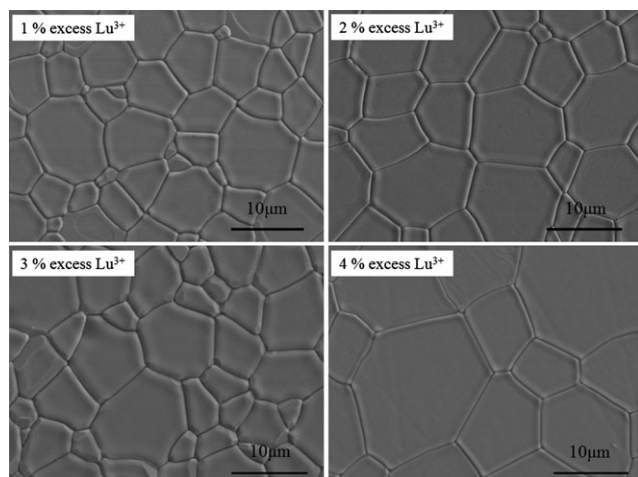

Fig. 4. The thermally etched surfaces of LuAG:Ce ceramics with various Lu content.

In Fig. 6, we show the pulse-height spectra, in which the light yield is given by the position of the photopeak in the figure (measured with 1 μs shaping time). For LuAG:Ce ceramics with increasing Lu content, a gradually decreasing light yield (light released within 1 μs after 662 keV photoabsorption) is observed. This trend is consistent with the observed X-ray luminescence intensities in Fig. 5. The mentioned retrapping of migrating electrons in shallow traps associated with Lu_{Al} AD during the transport stage of the scintillation mechanisms can indeed decrease the value of LY, but the decrease of radioluminescence intensity needs further investigation. To describe the absolute value of the light yield of Lu-rich LuAG:Ce ceramics, the relative comparison with the BGO standard sample is of significance in this case as both the samples emit in a similar spectral range, and therefore a spectral sensitivity of the photocathode of PMT is not much important. It is worth mentioning that all the Lu-rich LuAG:Ce ceramics display a higher light yield than that of BGO single crystal. The highest one within 0.75 μs shaping time in nonstoichiometric Lu 1% excess LuAG:Ce ceramics is about 1.82 times of that of BGO single crystal, see the inset of the Fig. 6. This result, however, is comparable to most of the LuAG:Ce single crystals, in which the concentration of Ce is limited at 0.01–0.1at.%. In addition, in the previous research work, we have reported that the light yield of stoichiometric LuAG:Ce ceramics containing Si and Mg sintering aids was only 60% of BGO single crystal, even though their transmittances at 550 nm is close to 80%.[23] Therefore, present results clearly demonstrate that the noticeable improvement of light yield can be obtained by inducing certain excess of Lu in the LuAG ceramics. Moreover, the deterioration effect on the scintillation characteristics of Lu_{Al} antisite defect is far smaller than in the case of the traditional sintering aids used.

Explanations about the improvement of light yield can be found in the work of Shen *et al.*[20] They have shown that both alio- and isovalent ion sintering aids may introduce structure disorder and impurities because of ionic radius mismatch between them and the matrix ions. In addition, the charge compensation effect may also give rise to the formation of defect complexes and color centers. All these phenomena were blamed for the formation of charge carrier traps in forbidden band, which are responsible for considerable intensity of scintillation light generated in the delayed recombination processes. However, as compared with alio- and isovalent sintering aids, neither charge compensation

effect nor impurities could be introduced to LuAG lattice in the case of excess Lu doping. As a consequence, high scintillation efficiency and light yield can be achieved in the case of Lu-rich LuAG:Ce ceramics.

Finally, the charge stability of Ce $^{3+/4+}$ is becoming a big issue in recent years. It has been reported that the ratio of Ce^{3+}/Ce^{4+} can be tuned both by the divalent ion Mg^{2+} codoping and isovalent ion Ga^{3+} admixture,[24,25] and the positive effect of Ce^{4+} ions in scintillation mechanism was proposed and evidenced in silicate and garnet materials. To investigate whether the valence state of cerium can be affected by excess of Lu^{3+} doping, X-ray absorption near edge structure (XANES) spectra of Lu-rich LuAG:Ce ceramics are measured. Fig. 7(a) shows the Ce L_{III}-edge XANES spectra of Lu-rich LuAG:Ce ceramics with various Lu content. CeF_3 and CeO_2 are the standard compounds with trivalent and tetravalent cerium, respectively, whose XANES spectra are shown in Fig. 7(b). By comparing the peak shape and the edge position with reference samples, the valence state of cerium in Lu excess LuAG ceramics could be qualitatively determined. However, as for different Lu excess LuAG:Ce ceramics, it appears that the cerium ion keeps almost stable the 3+ charge state, and the Ce^{4+} content remains almost at the same level, if at all present. This means that the excess of isovalence Lu^{3+} doping has a negligible effect on the cerium valence instability and creation of stable Ce^{4+} center.

IV. Conclusions

In this study, the influence of excess Lu admixture on the microstructure, optical and scintillation characteristics was studied in LuAG:Ce ceramics. Pure LuAG phase and good transmittance has been achieved up to 4% Lu_2O_3 excess, which demonstrates that excess of Lu could play the positive role of "self-sintering aid" in the ceramic sintering process. However, antisite defects were evidenced in Lu-rich LuAG:Ce ceramics. This result illustrates that the formation of Lu_{Al} antisite defects is not only related to the fabrication temperature but also related to the sample stoichiometry in agreement with theoretical calculations.[26] In addition, the scintillation efficiency of these Lu-rich LuAG:Ce ceramics reaches more than 10 times of that of BGO single crystal, which evidences no harmful role of such sintering aids in fabrication process. Furthermore, the highest light yield of Lu-rich LuAG:Ce ceramics was 1.82 times of that of BGO single crystal, and the charge state of cerium in Lu-rich LuAG:Ce was predominantly 3 +. Since Ce^{4+} ions were reported as fast and effective luminescence centers in the scintillation process, by their stabilization and an optimized Ce^{3+}/Ce^{4+} ratio further performance improvement seems very feasible.

Fig. 7. (a)The XANES spectra of the Lu-rich LuAG ceramics recorded near cerium's L_{III}-edge (5723 eV) at RT; (b)The XANES spectra of the Ce^{3+} and Ce^{4+} reference samples recorded at Ce L_{III}-edge at RT.

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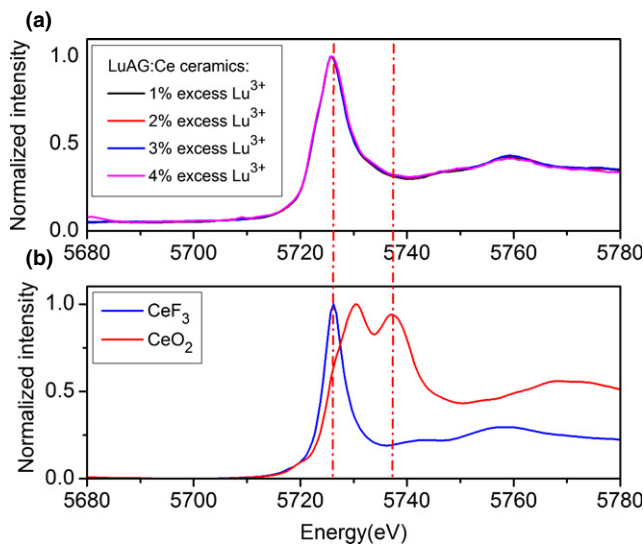
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