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Luminescence characteristics of $Lu_2SiO_5:Ce^{3+}$ (LSO:Ce) ceramic scintillators under VUV–UV excitation

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ABSTRACT

The luminescent properties of LSO:Ce (1 mol%) scintillators were characterized systematically under vacuum ultraviolet (VUV)–UV excitation. Under 185 nm VUV excitation, a broad emission band was detected in the wavelength range from 350 nm to 600 nm at different temperatures, which is ascribed to the 5d→4f electron transitions of Ce^{3+}. Excitation wavelength and temperature effects on luminescent properties of LSO:Ce ceramic scintillators were investigated in this paper. At 50 K, the emission intensity of Ce1 (Ce^{3+} locating in seven-oxygen-coordinated site) was much stronger than that of Ce2 (Ce^{3+} locating in six-oxygen-coordinated site), and the total emission intensity excited by 355 nm (Ce1) was higher than that excited by 185 nm (host). The total luminescence intensity increases as the rising temperature in the temperature range of 150–300 K, which is originated from thermal enhancement of transportation of free carriers dominating Ce1 emission.

1. Introduction

The good combination characteristics of Ce^{3+}-doped lutetium oxyorthosilicate (Lu_2SiO_5:Ce, LSO:Ce), such as high density (7.4 g/cm^3), high light yield (~27,300 photons/MeV), good energy resolution(7–13%) and short decay time (~40 ns), make it one of the most suitable scintillator in high-energy radiation detection such as Computed Tomography (CT) and Positron Emission Tomography (PET) in the last two decades [1–3]. However, as the doping Ce concentration increases from top to bottom in the crystal boule owing to the low segregation coefficient (~0.22) of Ce in LSO single crystal, the light output typically fluctuates 30–40% from the bottom to the top of the boule [4]. Such change results in variation of light output of final LSO:Ce single crystal. Moreover, the high melting point (2100 °C) of LSO is very close to the breakdown temperature of iridium crucibles [5], all of these disadvantages restrict its applications to a great extent. Compared to single crystals, polycrystalline ceramics possess the advantages of relative lower fabrication temperature, uniform doping concentration and easiness of mass production. Therefore, polycrystalline LSO:Ce ceramics are considered to be a candidate for corresponding LSO:Ce single crystals once high density and satisfactory optical transparency are achieved to meet the needs of sophisticated medical imaging and radiation detection by advanced ceramics processing techniques.

Various ceramics processing techniques including hot pressing (HP) [6], hot isostatic pressing (HIP) [7,8], spark plasma sintering (SPS) [9] and pressureless sintering [10] have been successfully developed to fabricate LSO ceramic scintillators in recent years. Scintillation properties (light yield and decay time) of the fabricated LSO ceramic scintallitors are comparable to that of LSO single crystal. However, photoluminescence (PL) and photoluminescence excitation (PLE) characteristics of LSO ceramic scintillators have not been investigated systematically up to now.

In present investigation, a translucent LSO:Ce ceramic scintillator was fabricated by pressureless sintering in a flowing H_2 atmosphere under 1700 °C for 6 h starting from synthetic submicron polycrystalline LSO:Ce powders. Luminescence properties of the LSO:Ce ceramic scintillators were systematically investigated under synchrotron radiation (SR) excitation. The luminescence characteristics of doped Ce ions with different local environment were discussed.

2. Experimental

2.1. Fabrication of LSO:Ce ceramic scintillators

The LSO:Ce (1 mol%) polycrystalline powder was obtained from a synthetic precursor after being calcined at 1000 °C for 2 h in air [11]. The ceramics powder was first ground in an agate mortar, sieved by a griddle of 200 mesh and then brought to a high packing density by uniaxial pressing of 25 MPa followed by cold

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isostatic pressing of 200 MPa. The as-prepared green pellet (19 mm in diameter and 2.5 mm in thickness) was pressureless sintered at 1700 °C for 6 h in a flowing H_2 atmosphere to achieve high density ceramics [10]. Subsequently the sintered LSO:Ce ceramics was subjected to a post-annealing treatment at 1250 °C for 6 h in air to eliminate the oxygen vacancy. A translucent LSO:Ce ceramic was finally obtained after lapping and mirror polishing.

2.2. Characterization of LSO:Ce powder and as-fabricated ceramic scintillators

The phase compositions of both LSO:Ce powder and ceramics specimen were determined by using an X-ray diffractometer (XRD, D\max-2550, Rigaku) with Cu Kα X-ray ($\lambda = 1.5406$ Å). The morphology and microstructure of LSO powder and ceramics were observed by field emission scanning electron microscopy (FE-SEM, JSM-6700F, JEOL). The particle size distribution of the LSO:Ce powder was measured by particle size analyzer (Zetasizer 3000HS, Malvern). The bulk density of the as-sintered ceramics in the form of a Φ15 mm × 1 mm disc was measured by the Archimedes method and expressed as a percentage of the theoretical density (7.4 g/cm^3) of LSO single crystal.

The luminescent properties LSO:Ce ceramics was thoroughly evaluated in terms of its VUV spectroscopic properties. Synchrotron radiation excitation and emission spectra were taken at the U24 VUV station of National Synchrotron Radiation Laboratory (NSRL, Hefei, China). The integration time of 1 s and wavelength step of 1 nm were applied for all VUV spectral measurements. Intensities of all spectra in each figure were divided by average current of the electron storage rings during the corresponding measurements for normalizing. All measurements were carried out at room temperature except for the temperature dependence test at VUV beam-line station using a closed loop liquid-helium apparatus. Decay profiles of the sintered LSO:Ce ceramics were recorded by a spectrofluorometer (FLSP920, Edinburgh Instruments) at room temperature. Pulse height spectra of LSO:Ce ceramics, bismuth germinate ($Bi_4Si_3O_{12}$, BGO) single crystal (15 mm × 15 mm × 1 mm) and LSO:Ce single crystal (15 mm × 15 mm × 1 mm) were recorded by a Hamamatsu R2059 photomultiplier (PMT) under excitation of 662 keV γ-ray (^{137}Cs source).

3. Results and discussion

3.1. Phase composition and microstructure of LSO:Ce powder and ceramic scintillators

The powder with high purity, uniform grain size and low agglomeration state is desirable for fabrication of ceramics with low porosity and good optical transparency. Fig. 1(a) shows the morphology of the synthetic LSO:Ce powder after calcining at 1000 °C for 2 h in air. SEM image of the as-calcined powder indicates that it is well dispersed with the average grain size of 70 nm. The inset in Fig. 1(a) indicates that average particle size of the LSO:Ce powder is 277 nm, and such kind of powder is beneficial to promote the densification of LSO:Ce ceramic scintillators. The fractured surface of the as-sintered LSO:Ce ceramics is given in Fig. 1(b), which shows that the ceramics with an average grain size of 5 μm is nearly pore-free and no secondary phases was observed within grains or at grain boundaries. The XRD patterns of LSO:Ce powder and ceramics shown in Fig. 1(c) demonstrate that all diffraction peaks of LSO:Ce powder and ceramics are well accord with the reported data (JCPDS 41-0239) of monoclinic LSO phase, no other secondary phases are detected. The relative density of the obtained translucent LSO ceramics reaches 99.6%. Fig. 1(d) displays a typical photograph of a LSO ceramics specimen with 15 mm in diameter and 1 mm in thickness in contact with characters. The sample exhibits optical translucence, the characters under the ceramics can be read. Considering large optical anisotropy (maximum difference of refractive index of LSO is 0.028, which is about 4 times of that of α-Al_2O_3) of LSO crystal structure, it is quite satisfied to attain such a translucent LSO ceramics.

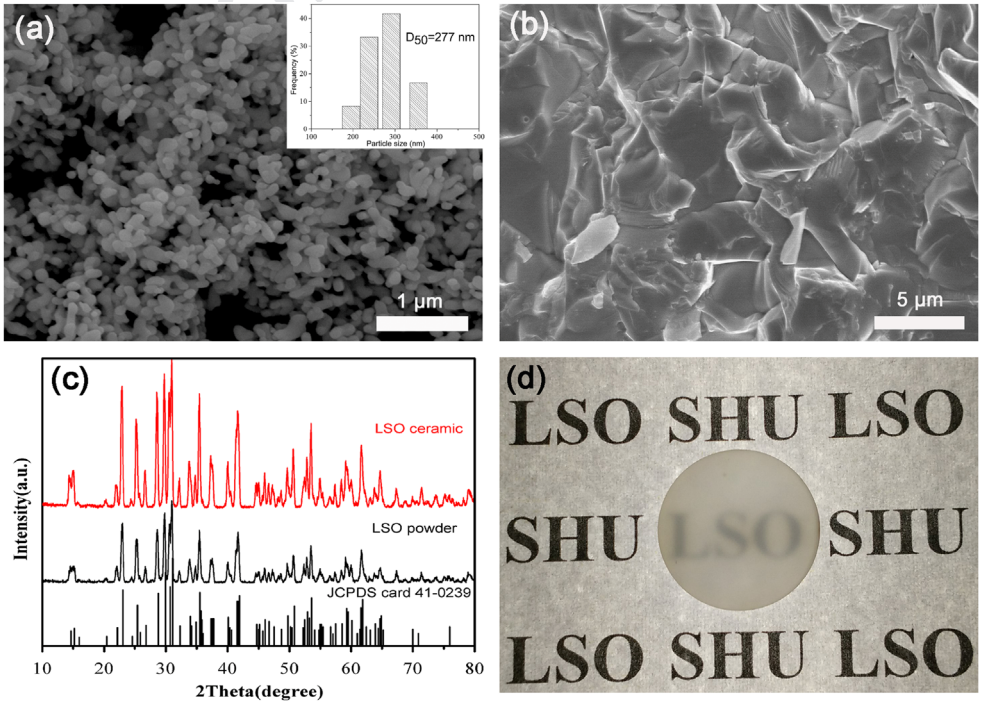

Fig. 1. SEM micrographs of (a) the as-prepared LSO:Ce powder calcined at 1000 °C for 2 h in air and (b) fractured surface of the LSO:Ce ceramics sintered at 1700 °C for 6 h, (c) XRD patterns of the as-prepared powders and as-sintered LSO:Ce ceramics, (d) photograph of a typical LSO:Ce ceramics specimen (Φ15 mm × 1 mm), inset is particle size distribution of the LSO:Ce powder.