

The influence of substrate temperature and deposition pressure on pulsed laser deposited thin films of CaS:Eu^{2+} phosphors



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ABSTRACT

The aim of this study was to investigate the influence of substrate temperature and argon deposition pressure on the structure, morphology and photoluminescence emission (PL) properties of pulsed laser deposited thin films of CaS:Eu^{2+} . The PL intensity improved significantly upon reaching substrate temperature of 650 °C. The (200) peak gradually became the preferred orientation. The increase in PL intensity as well as surface roughness is attributed to improved crystallinity and higher growth rates, respectively. The best PL intensity as a function of deposition pressure was obtained at an argon pressure of 80 mTorr. The initial increase and eventual drop in PL intensity as deposition pressure increases is ascribed to the changes in growth rates.

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1. Introduction

There is a growing interest in rare-earth-doped alkaline earth sulfides such as CaS:Eu^{2+} (red-emitting) and CaS:Ce^{3+} (green-emitting) as candidates for down conversion of blue light for phosphor-converted white LEDs (pc-WLEDs). This is so because both of these phosphors strongly absorb the blue light emitted by a blue LED [1] to emit red and green, respectively. Europium doped calcium sulfide has been studied by a substantial number of workers for its application in cathode ray tubes [2,3] and pc LEDs [4,5]. The development of a WLED as a source of light is important because LEDs have a high durability and low energy consumption when compared to other light sources, such as incandescent and fluorescent lamps. Thus, the LED as a source of light promises to overtake, if not displace and make obsolete our current sources of light energy. As opposed to the traditional powder phosphors, thin films offer the benefits of a reduction of material wastage, and better adhesion to solid substrates [6,7]. A variety of thin film growth techniques have been developed, including, spray pyrolysis, sputtering, metal organic chemical vapor deposition (MOCVD), pulsed laser deposition (PLD) [18], and Dey et al. [19] has grown CaS thin films using the atomic layer deposition (ALD) technique. Among these methods, PLD has emerged as one of the most popular techniques, offering a number of advantages over other techniques, which include, but not limited to,

congruent transfer of target materials, versatility, flexibility, and relatively low substrate temperature [8]. Nsimama and co-workers [9] showed that the structure and other properties of $\text{SrAl}_2\text{O}_4\text{:Eu}^{2+}, \text{Dy}^{3+}$ thin films during a PLD process are affected by such parameters as number of laser pulses, while [11] showed that wavelength of the laser radiation is important for the growth of CaS:Eu^{2+} . Substrate temperature and deposition pressure, which have a strong bearing on the kinetics of particles on the surface of the growing film and thus have an influence on the quality of the thin film.

In this paper, we report the growth, and characterization of CaS:Eu^{2+} thin films deposited on Si (111) substrates in an argon atmosphere for deposition pressure in the range 40–350 mTorr, and the substrate temperature varying from 200 °C to 650 °C. The subsequent influence of substrate temperature and deposition pressure on the growth rate is evaluated. Photoluminescence properties were investigated and evaluated for possible applications in pc-white LED technologies.

2. Experimental

CaS:Eu^{2+} , a commercial powder, obtained from Phosphor Technology (UK) was used in this study. To make the target for ablation, the powder was cold-pressed without binders using a custom made target holder [10]. The target was annealed in air at 250 °C for a 3 h period to harden and remove adventitious water-containing species and other volatile adsorbed substances. Thin

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films of CaS:Eu^{2+} were grown on Si (111) substrates using the Spectra Physics Quanta-Ray Pro-270 Nd: YAG laser operated in the 266 nm wavelength mode. Subsequently, the substrates were prepared and cleaned using the procedure outlined elsewhere [11] in greater detail. The deposition chamber had been evacuated to base pressure of 5.7×10^{-5} mbar before backfilling with the Ar gas at a pressure of 5.33×10^{-2} mbar (40 mTorr). The laser energy, number of pulses and pulse repetition rate were fixed at 40 mJ (a fluence of 5.09 J/cm^2), 18,000 and 10 Hz, respectively. The substrate temperature was in the range 200–650 °C, argon gas deposition pressure was varied from 40 mTorr to 350 mTorr, while the target was kept at 4.5 cm from the non-rotating substrate holder.

3. Characterization

The crystalline nature and phase composition of the thin films was investigated using a Bruker AXS D8 ADVANCE X-ray diffractometer (XRD), operated at 40 kV and 40 mA, with a $\text{CuK}\alpha$ radiation of wavelength 1.5406 Å. Diffraction patterns were recorded in the 2θ range of 15–50°, with a counting time of 1 s for each step size of 0.0037°. The Shimadzu SPM-9600 atomic force microscopy (AFM) operated in the contact mode was used to observe the surface morphology and roughness of the thin films. Photoluminescent (PL) spectra measurements of the thin films were carried out using a Varian Carry eclipse (model LS 55) spectrophotometer at room temperature with a monochromatized 150 W xenon flash lamp as the source of excitation. The PMT voltage was set to 600 V, while the emission and excitation slit widths were both 10 nm.

4. Results and discussion

4.1. XRD results

Fig. 1(i) shows the XRD patterns of the films deposited on Si (111) at substrate temperature ranging from 200 °C to 650 °C. The peaks marked S are from the substrate, while those marked with an asterisk* are suspected to come from CaSO_4 , as we showed in an earlier study [12]. For all deposition temperatures, the films exhibited a (200) texture, while the (111) texture increased as the substrate temperature was increased from 200 °C to 400 °C, then gradually decreased as the substrate temperature rose to 550 °C, and eventually disappeared at substrate temperature of 650 °C.

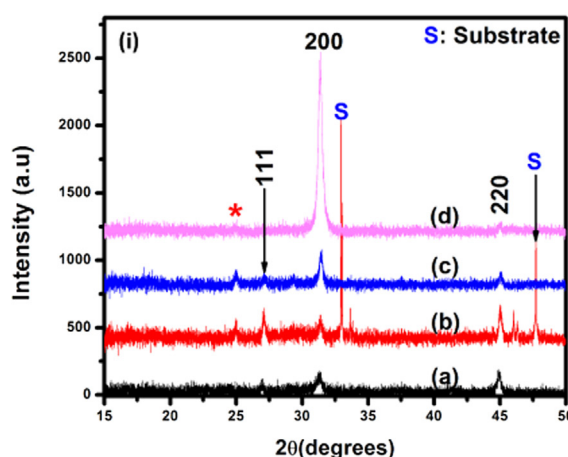


Fig. 1. (i) X-ray diffraction patterns for CaS:Eu^{2+} films deposited at various temperatures: (a) 200 °C, (b) 400 °C, (c) 550 °C, (d) 650 °C. (ii) Variation of FWHM of (200) peak with substrate temperature.

Table 1
Variation of crystallite size with substrate temperature.

Sub Temp.	2θ (°)	FWHM (°)	D (nm)
200	31.321	0.3149	25.92
400	31.382	0.2519	32.41
550	31.394	0.2204	37.04
650	31.373	0.2047	39.88

Also, as the (200) preferred orientation became more pronounced, the (220) orientation became more diminished. From these observations, it implies that as the substrate temperature increased, and crystallinity improved, the preferred orientation of the films became (200). The (200) peak (Fig. 1(i)), was used for the estimation of the crystallite size (D), using Scherer's formula [13,14]:

$$D = \frac{\kappa \lambda}{\beta \cos \theta} \quad (1)$$

where β is the FWHM of the peak in radians, and κ is a factor which is usually set as 0.89, and λ is the X-ray radiation wavelength (0.154056 nm) and θ is Bragg's angle. The estimated sizes are shown in Table 1.

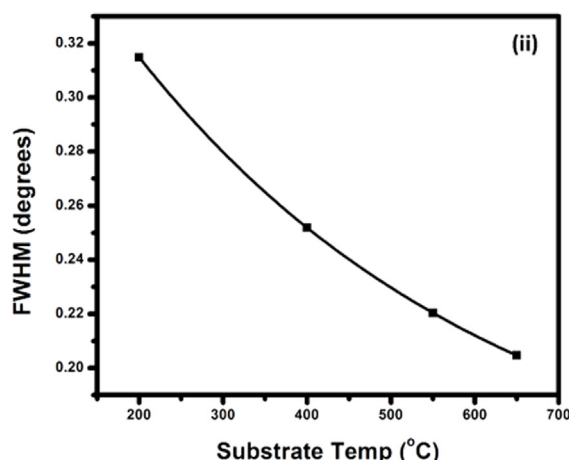
As can be seen from Table 1, and Fig. 1(ii), the full width at half maximum (FWHM) of the (200) peak decreased as the substrate temperature was increased, resulting to a corresponding increase of the crystallite size.

The X-ray diffraction patterns of the films deposited on Si (111) at deposition pressure ranging from 40 mTorr to 350 mTorr are shown in Fig. 2. Preferential growth of films on the (111) plane is more pronounced than on the (220) plane. The (200) plane is clearly visible for films deposited at pressures of 80 and 160 mTorr.

4.2. AFM results

Fig. 3 presents 3-dimensional AFM images for films deposited on the silicon substrate for different substrate temperatures. The images show well defined spherically shaped grains that are uniformly distributed over the surface with root mean square (RMS) roughness of 12 nm, 21 nm, 22 nm, and 24 nm, for films deposited at substrate temperatures of 200 °C, 400 °C, 550 °C, and 650 °C, respectively. The increased roughness with temperature rise is attributed to a higher rate of particle growth at higher temperatures [15].

Fig. 4 shows AFM images with root mean square (RMS) roughness of 21 nm, 26 nm, 12 nm, and 10 nm, for deposition pressures of 40 mTorr, 80 mTorr, 160 mTorr and 350 mTorr,



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