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Abstract. Cerium doped Zinc Oxide (ZnO:Ce) nanocrystals were synthesized through precipitation of nitrate solution by employing ultrasound irradiation. The precipitate products were calcinated at various temperature. The calcination temperature is an important key on the formation and properties of nanocrystal. This paper studied influence of calcination temperature on the formation and optical properties of ZnO:Ce nanocrystal. ZnO:Ce nanocrystals were characterized by x-ray diffractometer and UV-Vis spectrophotometer. The x-ray diffraction patterns revealed the formation of hexagonal wurtzite crystal structure for ZnO:Ce nanocrystals. The increase in calcination temperature improved crystallinity and reduced the band gap energy of ZnO:Ce. The result showed that the calcination temperature strongly influenced ZnO:Ce nanocrystals formation. The optical properties of ZnO:Ce nanocrystal can be modified by varying calcination temperature.

1. Introduction

Zinc Oxide (ZnO) is a semiconductor with a direct and wide band gap energy of 3.37 eV that has variety applications. The hexagonal structure with lattice parameters of $a = b = 5{,}250$ Å and $c = 5.206$ Å forms ZnO with good chemical stability [1]. In the past few years, ZnO has been considered as a photocatalyst with better performance than TiO_2 and as an excellent antibacterial agent. In the field of photocatalyst application, the wide band gap of ZnO can absorb a wider ultraviolet spectrum and produce a higher hydroxyl oxidation potential than TiO_2 [2-5] In addition, the wide energy band gap of ZnO produces electron hole pairs with a relatively low energy level inducing reactive oxygen species (ROS) that inhibit bacterial growth [6, 7]. However, the wide band gap energy of ZnO possesses a rapid recombination of electron-hole pairs that reduced the performance of both photocatalytic and antibacterial properties of ZnO [2-5,7]

Rare earth metals doping such as Cerium (Ce), has been developed to improve the photocatalyst and antibacterial activity of ZnO. Ce doping can shift the band gap toward visible light and also encourage the formation of crystals to reduce particle size with large the surface area [4,7]. A number of methods have been reported for synthesizing Ce doped ZnO (ZnO:Ce) nanomaterials such as Sol gel [2,8], Hydrothermal [7, 9, 10] simple chemical precipitation [3,11,12], Sonochemical [4, 13, 14] and microwave irradiation [15]. The low temperature and fast reaction method is needed in term of low energy consumption, low production cost and large-scale production of nanomaterials. Hydrothermal, sonochemical and microwave irradiation methods have been successfully used to synthesize ZnO:Ce nanomaterials at low temperature, however those method require long reaction times and sometimes require post heat treatment. Sol gel is versatile method for large-scale production of nanomaterials

without post heat treatment due to high temperature reaction. The precipitation method has advantages over other methods, particularly due to simple apparatus and low production costs [1].

In the previous report, pure ZnO and ZnO:Ce nanocrystal have been synthesized at calcination temperature as low as 100 °C using precipitation method in combination with ultrasound irradiation [16,17]. The utilization of ultrasound irradiation to precipitation process resulted in high crystalline nature of ZnO:Ce nanocrystal. The formation of crystalline structure and modification of optical properties of ZnO:Ce can be achieved by selecting synthesis parameters. This paper aims to report the influence of calcination temperature on the formation of ZnO:Ce nanocrystals and their optical properties to obtain functional ZnO:Ce nanocrystal.

2. Experimental

2.1. Synthesis

The simple precipitation method was used to synthesize ZnO:Ce nanocrystals. Precipitation process was carried out by dissolving Zn $(NO_3)_2 \cdot 6H_2O$ and Ce $(NO_3)_3 \cdot 6H_2O$ (molar ratio Ce / Ce + Zn = 3%) into 100 mL demineralized water (DW). The precursor solution was stirred using a magnetic stirrer for 10 minutes at room temperature. During the stirring process, 3 M ammonium solution was dropped into the precursor solution until pH the solution reached to 10. The solution was then irradiated by ultrasound for 30 minutes. The precipitates was washed using distilled water and ethanol and calcined at temperature of 100 °C, 200 °C and 300 °C for 3 hours.

2.2. Characterization

The x-ray diffraction (XRD) pattern of obtained ZnO: Ce nanocrystal was recorded using x-ray diffractometer (Rigaku miniplex 600). UV-visible (UV-vis) spectra in the range 300-800 nm was measured using spectrophotometer (Shimadzu 1420).

3. Results and discussion

3.1. Crystal structure

Figure 1 shows the XRD patterns of the synthesized ZnO:Ce at calcination temperature of 100°C, 200°C and 300°C. Identification diffraction peaks of all ZnO:Ce revealed that the peaks correnponding to (100), (002), (101), (102), (110), (103), (200), (112), and (201) which good agreement with the diffraction peaks of hexagonal wurtzite ZnO (JCPDS No. 36-1431). The calcination at 100°C resulted in weak diffraction peaks. The strong diffraction peaks obtained for ZnO:Ce calcinated at 200°C and 300°C indicating well crystalline nature. In addition, there are not observed peaks related to other phase suggesting the formation a single phase hexagonal wurtzite structure that confirmed Ce incorporation into lattice site of Zn [3,4,18].

Incorporation of Ce into Zn lattice changes the diffraction plane distance. Based on the Bragg's law: $2\,d \sin\theta = n\,\lambda$, the relation between lattice parameter and diffraction plane distance of hexagonal crystal described by Equation (1) [1,16,18].

$$\frac{1}{d_{hkl^2}} = \frac{4}{3}\left[\frac{h^2 + hk + k^2}{a^2}\right] + \frac{l^2}{c^2} \quad (1)$$

where h, k and l are the Miller's index, d_{hkl} is the distance between the planes representing the miller's index, a and c are lattice parameters. Lattice parameters of ZnO:Ce were calculated using (100) and (002) diffraction peaks as follow [1,9, 16, 18]:

$$a = \frac{\lambda}{\sqrt{3}\sin\theta_{(100)}} \quad (2)$$

$$c = \frac{\lambda}{sin\ \theta_{(002)}} \quad (3)$$

where λ is the wavelength of x-rays (1.54056 Å).

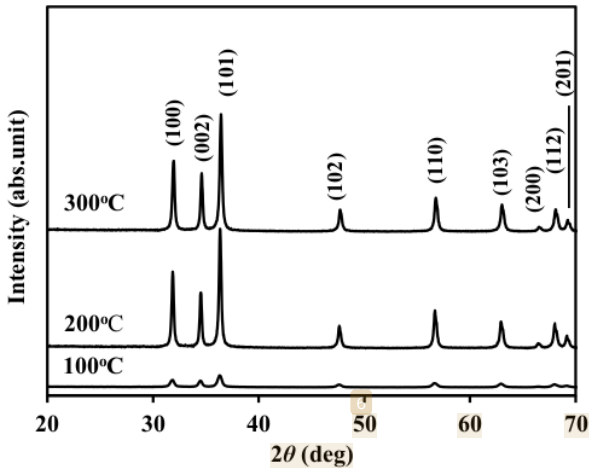

Figure 1. X-ray diffraction patterns of ZnO:Ce nanocrystals

The lattice parameter *a* dan *c* of ZnO:Ce nanocrystal are smaller than standard value of pure ZnO which confirmed the Ce incorporation into Zn lattice [3, 18]. Beside the same value of c/a ratio for all ZnO:Ce nanocrystal and close to standard value of 1.60 are also showed that the increase in calcination temperature does not change hexagonal structure. The shift in diffraction peaks toward the larger diffraction angle and the decrease in lattice parameters with improvement calcination temperature are attributed to the decrease in Zn–O bond length as shown in Table 1. The bond length was calculated by Equation (4) [18].

$$L = \left[\frac{a^2}{3} + \left(\frac{1}{2} - u\right)^2 c^2\right]^{1/2} \quad (4)$$

where $u = a^2/3c^2 + 0.25$ represents the potential parameter of hexagonal structure. The other structural parameters of potential parameter and unit cell volume of ZnO:Ce are also tabulated in Table 1. Both of potential parameter and unit cell volume are in good agreement with standard value in JCPDS. It indicates that calcination temperature in the range 100-300°C appropriate for hexagonal structure formation of ZnO:Ce nanocrystals.

Table 1. Structural parameters of ZnO:Ce nanocrystals

Calcination temperature (°C)	Lattice parameter			unit cell volume	Potential parameter	Zn–O bond length
	a (Å)	*c* (Å)	*c*/*a*	(Å^3)	*u*	*L* (nm)
100	3.2482	5.2020	1.60	47.51	0.3799	1.9764
200	3.2451	5.1938	1.60	47.36	0.3801	1.9742
300	3.2379	5.1813	1.60	47.04	0.3802	1.9697
JCPDS 36-1451	3.2490	5.2060	1.60	47.59	0.3799	1.9774

The peak broadening of XRD pattern is contribution of crystallite size and micro-strain. The crystallite size and lattice strain were estimated using the size strain plot (SSP) method as given in Equation (5) [16, 19]

$$d\beta_{hkl}\cos\theta = \frac{K}{D}(d^2\beta_{hkl}\cos\theta + \left(\frac{\varepsilon}{2}\right)^2 \quad (5)$$

where β_{hkl} is full width at half of maximum (FWHM), K is ¾ for spherical crystal, D is crystallite size and ε is the lattice strain. Figure 2 depicts SSP plot of ZnO:Ce calcined at temperature of 100°C, 200°C and 300°C. The SSP method shows high compatibility to experimental data with correlation coefficient $R^2 > 0.9$.

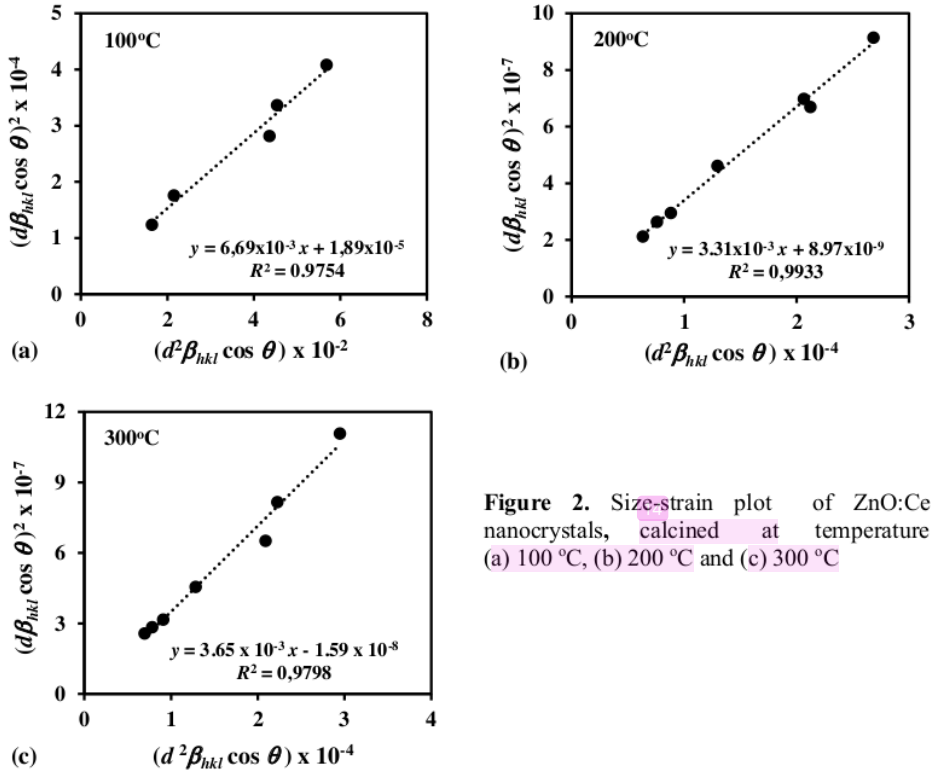

Figure 2. Size-strain plot of ZnO:Ce nanocrystals, calcined at temperature (a) 100 °C, (b) 200 °C and (c) 300 °C

The crystallite size is determined from the slope of SSP plot and the root mean square (RMS) strain is obtained from the root of y-intercept. The tabulation of crystallite size and strain for ZnO:Ce nanocrystal depicted in Table 2. The increase in calcination temperature from 100 °C to 200 °C lead to increase crystallite size over two times. As a consequent the strain value of ZnO:Ce nanocrystal decreased with increase in calcination temperature.

Table 2. Crystallite size and strain of ZnO:Ce nanocrystal

Calcination temperature (°C)	*D* (nm)	Strain
100	11.2	7.824×10^{-4}
200	22.6	1.600×10^{-4}
300	20.6	-2.027×10^{-4}

3.2 Optical properties

Optical properties of ZnO:Ce nanocryatals was analysed using UV-visible spectrophotometry. Figure 3(a) shows the UV-visible absorbance spectra of ZnO: Ce nanocrystals in the 300-800 nm range. The increase in calcination temperature lead the absorption peak shifted to the higher wavelength and improved the absorption properties of ZnO:Ce nanocrystals. The absorption peak shift can be related to formation localized state in the band gap due to Ce incorporation into Zn lattice [3], meanwhile the strong absorption properties can be related to the good crystallinity of ZnO:Ce as can be seen in the XRD pattern. The sharp peak in the absorbance at the 360 – 380 nm wavelength interval can be associated with the band gap energy of ZnO [3, 4].

The band gap energy of ZnO:Ce nanocrystals was estimated using Tauc's plot methods as depicted in Figure 3(b). The band gap energies of 3.08 eV, 2.98 eV and 2.67 eV were found for ZnO:Ce nanocrystal with increasing calcination temperature from 100 °C to 300 °C. The increase in calcination temperature reduced the band gap energy of ZnO:Ce nanocrystal. It can be associated to formation of bigger nanocrystal as found from XRD analysis. This finding shows that calcination temperature strongly influenced optical properties of ZnO:Ce nanocrystal. The band gap energy of ZnO:Ce can be modified by varying calcination temperature.

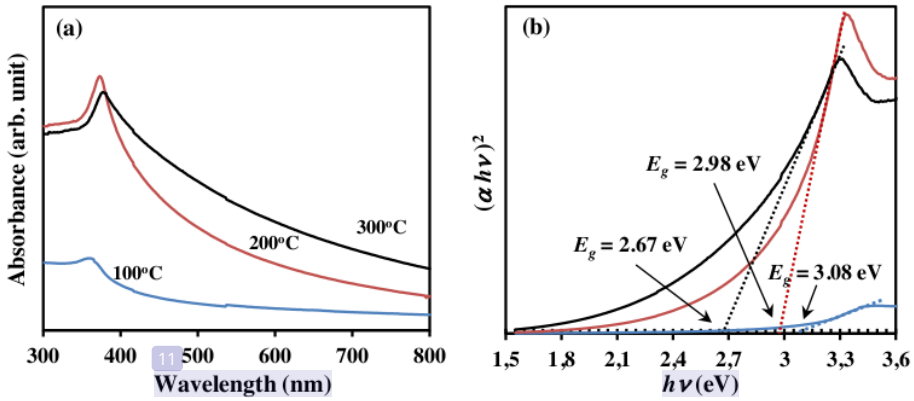

Figure 3. (a) UV-visible absorbance spectrum and (b) Tauc's plot of ZnO:Ce nanocrystal

4. Conclusion

The results show calcination temperature of 100 °C, 200 °C and 300 °C feasible for formation of hexagonal ZnO:Ce nanocrystal. The calcination temperature has a profound influence on crystallinity and optical properties of ZnO:Ce nanocrystals. High crystalline nature and strong absorption of ZnO:Ce obtained at calcination temperature of 200 °C. The band gap energy of ZnO:Ce nanocrystals narrowed with increase in calcination temperature due to formation larger nanocrystals.

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