Journal of Nano- and Electronic Physics
Vol. **11** No 1, 01028(3pp) (2019)

Журнал нано- та електронної фізики
Том **11** № 1, 01028(3cc) (2019)

Short Communication

Sol-gel Synthesis and Structural Properties of Cu Doped ZnO Nanoparticles

P.K. Samanta[1,*], M. Das[2], N.K. Rana[3]

[1] *Department of Physics (PG & UG), Prabhat Kumar College, Contai-721404, West Bengal, India*
[2] *Department of Physics (PG & UG), Panskura Banamali College, Panskura-721152, West Bengal, India*
[3] *Department of Physics, Ghatal R. S. Mahavidyalaya, Ghatal-721212, West Bengal, India*

(Received 18 November 2018; revised manuscript received 02 February 2019; published online 25 February 2019)

We report here a sol-gel method to synthesize undoped and Cu doped ZnO nanoparticles. The nanoparticles were characterized using X-ray diffraction technique to understand the crystallographic properties. The results indicate the formation of pure and Cu doped ZnO nanoparticles. The growth was found to be anisotropic. The low concentration of Cu doping does not influence the particles size of ZnO. We have successfully synthesized undoped and Cu doped ZnO nanoparticles using a simple and cost effective sol-gel method. The synthesized nanoparticles exhibit high crystallinity and Cu doping was further confirmed from X-ray diffraction. The diffraction pattern also suggests that the growth of hexagonal ZnO is anisotropic. However due to this low percentage of Cu doping, no significant change in particle size was observed. In this paper, we report a very simple and cost effective sol-gel method to synthesize ZnO nanoparticles followed by typical structural characterization.

Keywords: ZnO, Doping, X-ray diffraction, Crystal, Anisotropy.

DOI: 10.21272/jnep.11(1).01028 PACS numbers: 78.67.Bf, 78.67.Rb, 87.64.-t

1. INTRODUCTION

Zinc oxide (ZnO) is a potential material in the present material research because of its unique size dependent tunable properties [1]. It is II-VI semiconductor with high band gap ~ 3.4 eV and large binding energy (~ 60 meV) for exciton at room temperature [2]. This has made ZnO a potential material for stable and short wavelength UV lasing. Huang *et al.* have successfully demonstrated UV lasing from ZnO nanorods grown on GaAs substrate by vapor-liquid-solid method [3]. ZnO usually crystallizes in wurtzite structure with alternatively stacked Zn and O ions along the c-axis of the hexagon [4]. But the structure is non-centrosymmetric, due to which ZnO exhibits piezoelectric property. Wang *et al.* have already demonstrated the piezoelectric property of ZnO nanorods and its use in pollution free energy generation from mechanical vibration [5]. ZnO exhibits high optical transparency in visible wavelength of the electromagnetic spectrum. It also has good electrical conductivity. Possession of this visible light transparence and high conductivity makes it a potential material to be used as transparent electrode in solar cell and other photonic/optoelectronic devices [6] ZnO is a very popular material for its excellent and efficient photoluminescence property. Due to its high band gap, the photoluminescence from ZnO is usually observed in the UV region. However, during the crystal growth, several defects like vacancies, interstitials and anti-sites appear [7-11]. Energy states are created in between the conduction band and valence band of ZnO. Thus these defect states with energy lower than the band gap give rise to the visible photoluminescence from ZnO nanostructures. Researchers have reported visible emission (violet, blue, green) from varieties of ZnO nanostructures. Several methodologies have already appeared in the literature to synthesize ZnO nanostructures. These includes: chemical and sol-gel method, sputtering (dc and ac), vapor-liquid-solid, electrochemical, physical and chemical evaporation and spray pyrolysis method [12-13]. Sol-gel method is a very popular and simple technique to synthesize ZnO nanoparticles. It does not require the maintenance of sophisticated experimental parameters like low pressure, high temperature, flow of carrier gas and many more. Here, in this paper, we report a very simple and cost effective sol-gel method to synthesize ZnO nanoparticles followed by typical structural characterization.

2. MATERIALS AND METHOD

All chemicals used in this experiment were used as supplied by Merck (99.99 % pure). To synthesize pure ZnO, aquatic solution of zinc acetate dehydrate was prepared by dissolving 2.195 g in predetermined amount of water. 1.048 g of LiOH was dissolved in predetermined amount of water to prepare 1 M solution. These two solutions were mixed and magnetically stirred for 2 hours at room temperature. At the end of the reaction the white precipitate collected, washed with distilled water and dried at 100 °C for characterization.

To synthesize Cu doped ZnO, we followed the same experimental process as described in the above section. We additionally used $CuCl_2$ solution of predetermined concentration. By varying the concentration of $CuCl_2$, the % of Cu doping was varied.

The X-ray diffraction (XRD) experiment was carried out in a Rigaku X-ray diffractometer and the data were collected over the angular range $30° < 2\theta < 70°$ with step size of 0.02°. Cu-$K\alpha$ radiation (wavelength ~1.5418 Å) was used as the source of X-ray. The detector was an ionization chamber type to count for the intensity of the diffracted beam.

*pijush.samanta@gmail.com

3. RESULTS AND DISCUSSION

Typical XRD patterns of undoped and Cu doped ZnO nanoparticles are shown in Fig. 1. Sharp diffraction peaks were observed indicating that the synthesized material is highly crystalline.

The pattern was indexed with hexagonal unit cell structure using the standard JCPDs Card No. 36-1451 with the presence of the diffraction peaks in the order (100), (002), (101), (102) , and (110).

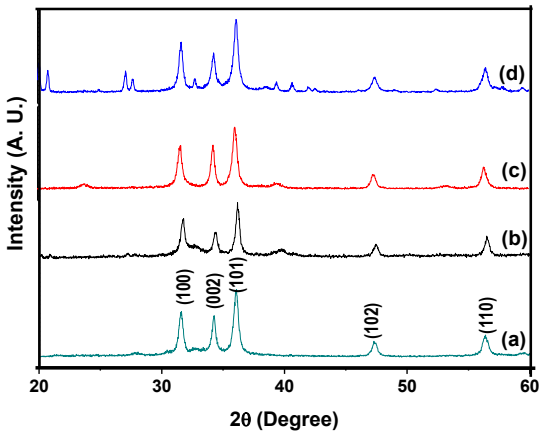

Fig. 1 – XRD pattern of undoped and Cu doped ZnO [(a) undoped ZnO, (b) 2 %, (c) 4 % and (d) 6 % Cu doped ZnO]

The data are very consistent with the JCPDS data. Some additional peaks appeared at 39.32° and 40.68°, respectively, for Cu-doped ZnO samples which are the diffraction peaks of Cu. This indicates that Cu is successfully doped in ZnO. No peak of CuO was observed; this indicates that there is no CuO phase mixed with the ZnO nanoparticles.

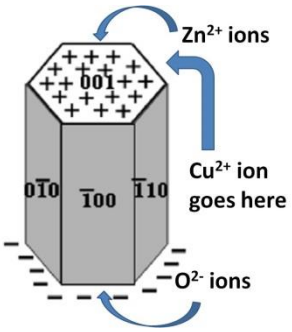

Fig. 2 – Schematic of the Cu doping in ZnO crystal

The crystal structure of ZnO is hexagonal/wurtzite. The Zn^{2+} and O^{2-} ions are alternatively stacked along the c-axis of the hexagon. The top face is terminated with the Zn^{2+} ion, while the bottom face is terminated by O^{2-} ion. Thus there exists a dipole moment along the *c*-axis. These two faces are polar in nature. The other faces are non-polar. Thus the energies of different facets are different. This indicates that growth rate of different planes will be different. This growth rate plays an important role in determining the morphology of the nanostructures. In our case we also observe the variation of intensity of different diffraction peaks that indicates the anisotropic growth of undoped and Cu doped ZnO nanoparticles. The growth rate of different diffraction peak follows the following order: (102) > (110) > (002) > (100) > (101). In this context it is worthy to mention that the higher the growth rate, the lower multiplicity of the plane is. We also observed that the intensity of (002) plane has been changed due to Cu doping. This is because, during chemical reaction Cu^{2+} ions are released in the solution and preferentially attached to the (001) plane towards the Zn^{2+} ions site. Thus the growth rate increases resulting in the decrease of the multiplicity of (001) plane. A schematic of Cu^{2+} ion incorporation in ZnO crystal is shown in Fig. 2.

Scherrer formula was employed to estimate the crystallite size as given by

$$R = 0.89\ \lambda / \beta \cos\theta,$$

here β is the full width at half maxima and λ is the wavelength of the X-ray used in the diffraction experiment.

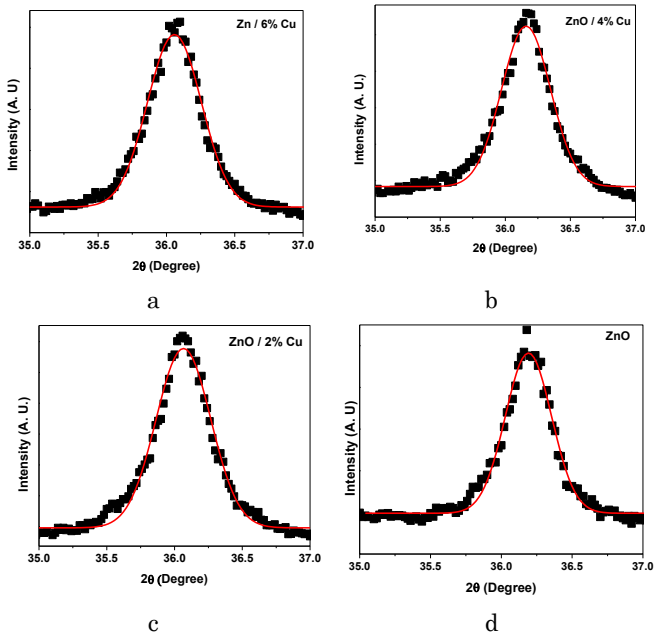

Fig. 3 – Gaussian fitting of (101) peak of undoped and Cu doped ZnO

For this calculation we choose the highest intensity peak (101) and used Origin 6.1 software to fit it as Gaussian type (see Fig. 3). No significant change in particle size was observed due to Cu doping and average particle size was calculated to be 24 nm.

4. CONCLUSIONS

In conclusion, we have successfully synthesized undoped and Cu doped ZnO nanoparticles using a simple and cost effective sol-gel method. The synthesized nanoparticles exhibit high crystallinity, and Cu doping was further confirmed from X-ray diffraction. The diffraction pattern also suggests that the growth of hexagonal ZnO is anisotropic. However due to this low percentage of Cu doping, no significant change in particle size was observed.

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Золь-гель синтез та структурні властивості наночастинок ZnO легованих Cu

P.K. Samanta[1], M. Das[2], N.K. Rana[3]

[1] *Department of Physics (PG & UG), Prabhat Kumar College, Contai-721404, West Bengal, India*
[2] *Department of Physics (PG & UG), Panskura Banamali College, Panskura-721152, West Bengal, India*
[3] *Department of Physics, Ghatal R. S. Mahavidyalaya, Ghatal-721212, West Bengal, India*

В роботі був використаний метод золь-гель синтезу наночастинок ZnO, нелегованих і легованих Cu. Кристалічна структура наночастинок ZnO була досліджена методом рентгенографії. Отримані результати свідчать про утворення чистих і легованих Cu наночастинок ZnO. Виявлено, що ріст наночастинок є анізотропним. Низька концентрація легування Cu не впливає на розмір частинок ZnO. Ми успішно синтезували наночастинки ZnO, нелеговані і леговані Cu, використовуючи простий і економічно ефективний золь-гель метод. Синтезовані наночастинки ZnO демонструють високу кристалічність, а легування Cu підтверджується рентгенівською дифракцією. Дифрактограма також свідчить про те, що зростання гексагонального ZnO є анізотропним. Проте, внаслідок низького відсотка легування Cu, суттєвих змін у розмірі частинок не спостерігалося. У даній роботі наведено дуже простий і економічно ефективний метод золь-гель синтезу наночастинок ZnO з типовими структурними характеристиками.

Ключові слова: ZnO, Легування, Рентгенівська дифракція, Кристал, Анізотропія.