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Chemical Synthesis of Non-hazardous ZnO and Potassium Doped ZnONanoparticles for Optoelectronic Devices.

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Abstract:

Zinc Oxide nanoparticles with energy efficient, cost effective and nontoxic material has opened the avenues of nanotechnology. Zinc oxide nanoparticles have advantages because of the extra ordinary physical and chemical properties. Chemical synthesis method has been carried out for the deposition of pure ZnO and K doped ZnO nano films. The structural, optical and electrical properties of the films have been investigated using X-ray diffraction (XRD), UV-Vis spectroscopy, I-V characteristics, Photoluminescence (PL), and Field Effect Scanning Electron Microscopy (FESEM).

Crystal size of undoped and K doped ZnO nano films were deduced to be 20.71nm and 15.62nm respectively. Films exhibits average transmittance more than 80%. I-V characteristics show increase in conductivity with doping of potassium. FESEM reveals the uniform chromosome type structure.

Keywords: Chemical synthesis; Nano films; K doped ZnO; sol-gel

Introduction:

Bulk Zinc oxide is cost-effective, nontoxic and centre symmetric structure material. Because of these advantages it is the highest tensor among all semiconductors for fabrication of optoelectronic devices. ZnO nano particles are II-VI group semiconductor with wide band gap of 3.3ev and high excitation energy of 60ev. It can be made more conductive by doping. It is having unique structural and photoluminescence properties [1]. Indium tin oxide (ITO) is most investigated region by the researchers, but due to the cost of fabrication and its toxic nature is hazardous to human and environment; there is necessity to attempt new combination of TCO. ZnO is a best alternative to tin oxide and Indium oxide due to its nontoxicity and low cost.

The study of nanomaterials belongs to morphological features of materials on the nanoscale. The properties of nanomaterials are a subject of recent nano technological research. In nanotechnology, reduction of size of particle exhibits great change in electrical optical properties. When the dimension of material is continuously reduced below 100nm, its properties show drastic changes. The geometrical structure, chemical bonds, ionization potential, mechanical strength, thermal and optical properties get affected by reduced particle size.

The electronic and optical properties of zinc oxide nanomaterial are attractive for optoelectronic application such as electronic material in light emitting diode, solar cells, flat panel display and transparent filed effect transistor [2, 3]. It is possible to evaluate optical constants by analyzing transmittance spectrum [4]. ZnO is semiconductor material having n-type electrical conductivity, due to intrinsic defects such as oxygen vacancies and zinc interstitials. Doping of different elements from group I (Li, Na & K) for Zn site and group V elements (antimony, bismuth) for oxygen sites forms P-type ZnO. Group I elements are better dopants materials than group V elements in terms of the shallow acceptor levels [5]. Comparative study of the effects of group I elements on the structural and optical properties of the ZnO nanoparticle is reported [6].

ZnO thin films have been deposited using different techniques, such as spray pyrolysis, molecular beam epitaxy (MBE), pulsed laser deposition (PLD) and sol-gel [7-11]. In the present investigation, chemical synthesis method such as sol-gel technique is used to prepare thin films of ZnO because the technique is simple and low cost. Moreover, the deposition rate, thickness of the films can be easily controlled over a wide range by changing deposition parameters sol-gel technique is also having advantages like homogeneity, simplicity and large area films can be prepared at lower temperature.

Present work focused on effect of K-doping ZnO thin films prepared by sol-get chemical synthesis method. By keeping post annealing temperature constant effect of K doping on the electrical and optical properties of deposited films is investigated.

Experimental:

ZnO and K doped ZnO nano films were deposited on glass substrates using chemical synthesis sol-gel method. Zinc acetate dihydrate ($Zn(CH_3COO)_2 .2H2O$), 2methoxy ethanol ($CH_3O(CH_2)OH$), potassium acetate, were used as a starting precursor. All the materials were purchased from sigma-Aldrich. 9% K ZnO

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solution was prepared for 0.4 moles. The mixture of the solution was vigorously stirred on hot magnetic plate at 60 □ c for 1 hour. This prepared solution was used for spin coating after the aging of the solution for 24hours.

The glass substrates were cleaned sequentially by chromic acid, labolean and acetone. Deposition of the films was carried out on spin coater maintained at constant rotation. The samples were coated repeatedly for ten times to get desired thickness. Preheating treatment is given to samples after each coating and cooled down at room temperature. Preheating treatment is given to samples for evaporation of the organic contents in the film. The deposited samples were post annealed at 350 □ c temperature in open air for one hour. Annealed samples were characterized for structural,optical and electrical properties. The crystal structure of ZnO nano films were investigated by Rigaku X-ray. Diffractometer (XRD) with cu-kα radiation source having wavelength of 1.5405 A □, and the scanning range of 2θ was between 20 □ to 60 □.

The optical transmittance of the samples was investigated by using double beam Shimadzu UV-2600 spectrophotometer in the UV visible range. UV-spectra of samples were recorded in the range 400 to 700 nm. The optical absorption of the films was carried out by the same instrument. The room temperature photoluminescence spectrum was measured by spectrofluorometer in the range with excitation wavelength of 330 nm. Samples have been characterized by current verses voltage curve. Surface morphologyof the deposited films were examined with field emission scanning electron microscope (FESEM)

Results and discussion:

ZnO and K doped ZnO nano film shows the significant results, which has been reported in the present study to investigate effect of doping on structural, electrical and optical properties.

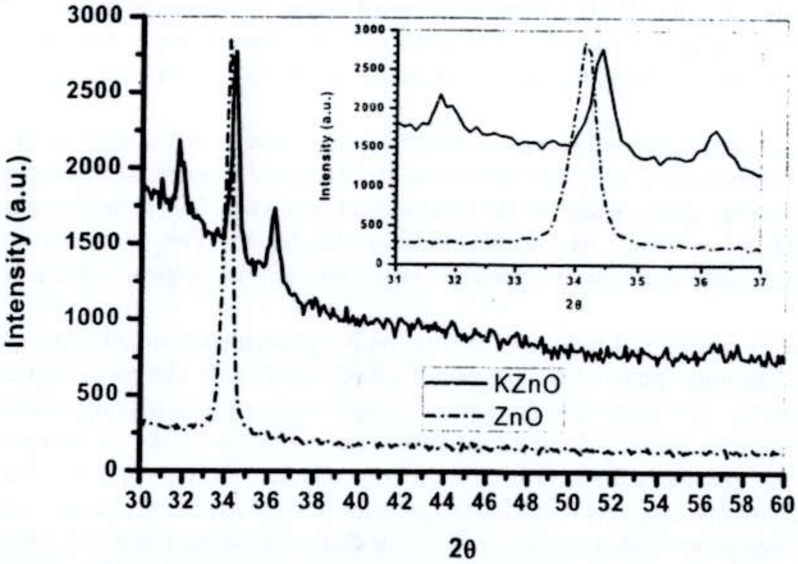

Fig.1: XRD pattern of undoped ZnO and K doped ZnO nano films. (Inset: Shifting of peak)

Fig. 1 represent the XRD pattern of undoped ZnO and K doped ZnO nano films.According to JCPDS card no 36-1451 [12]diffraction peak (002)of the hexagonal wurtzite ZnO at 34.08 □ indicating that undoped and K doped ZnO nano films were grown along preferential c-axis orientation (002). Potassium ions incorporated in ZnO improve the crystallinestructure[13]. The doping of Potassium does not deteriorate the crystal structure. However, the potassium heterogeneous nucleation is facilitated due to ions in the ZnO Structure[14].Crystallite size and lattice strain affect the Bragg peak by increasing peak width,intensity and shifting of peak position accordingly[15].The average crystalline size of undoped and K doped ZnO has been estimated from Debye Scherrer equation[16].

$$D = \frac{0.9\lambda}{\beta\cos\theta} \qquad (1)$$

Where, D is the average crystalline size of the film. λis the wave length of X-ray source(1.54059 Å),θ is the Braggdiffraction angle andβ is the full width at half maximum intensity (FWHM)in radians.

Average crystalline size estimated from XRD pattern and found to be 20.71 nm and 15.62 nm for undoped and K doped ZnO respectively.Grain size decreased with K doping. The diffraction angle of peak (002) is

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34.12 and 34.429 for undoped and K doped ZnO nanofilms respectively. Inset of fig. 1 shows the shifting of diffraction angle. Diffraction angle of K doped ZnO nanofilms shifted towards higher side which suggests that the lattice constant in c-axis of ZnO crystal decrease with substitution of K ions into Zn sites.
The strain of the films can be obtained using Williamson-Hall (W-H) method. W-H equation can be expressed in the form [17].

$$\varepsilon = \frac{\beta os\theta}{4Sin\,\theta} \qquad (2)$$

Where,ε is the micro strain associated with the nanoparticles. The macrostrain of undoped and K doped ZnO nanofilms was found to be 1.84e-3 and 7.4e-3 respectively. The structural parameters of undoped and K doped ZnO nanofilms were summarized in the table 1.

	Compound	2 θ ( )	FWHM (°)	Max. Intensity	Grain size D(nm)	Micro strain ε
Deposited samples	Undoped ZnO	34.12	0.24	2845	20.71	$1.84*10^{-3}$
	K doped ZnO	34.429	0.255	2773	15.62	$7.4*10^{-3}$

Table 1: Structural parameters of undoped and K doped ZnO nanofilms

Undoped and K doped ZnO nano films were studied by UV-Vis spectrophctometer for the optical properties. As ZnO is a transparent conducting oxide, it is essential to study effect of K doping on transmittance. Fig. 3 explores the optical transmittance of Undoped and K doped ZnO nano films. The films exhibit good transparency. The transmittance value of undoped and K doped ZnO nano films was found 83.41% and 86.33% respectively. The potassium doped ZnO film shows a higher transmittance than the undoped ZnO film due to smoother and more uniform surface.
The surface morphology of undoped and K doped ZnO nano films using FESEM were explored in fig. 2. Images of the nanofilms were taken at the scale of 5μm with magnification of 10,000 for undoped and K doped ZnO nano films. Uniform chromosome type structure is retained for both the films with change in crystal size.

Fig.2 FESEM images of undoped and K doped ZnO nano films

The optical band gap (E_g) of undoped and K doped ZnO nano films was determined by using the absorption coefficient and extrapolation method. Fig. 4 showed plot of photon energy verses absorption coefficient for undoped and K doped ZnO nano films. The graph showed the optical band gap energy value 3.25ev and 3.277ev for undoped and K doped ZnO nano films respectively. Doping of potassium showed broadening in the band gap.

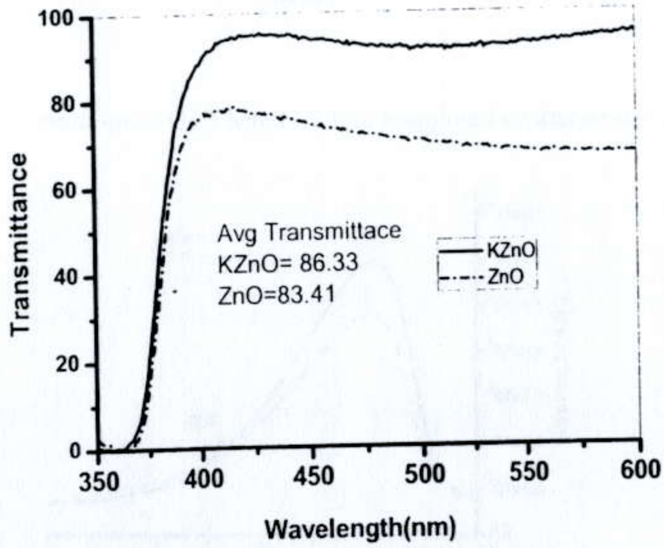

Fig. 3: Transmittance of undoped and K doped ZnO thin films

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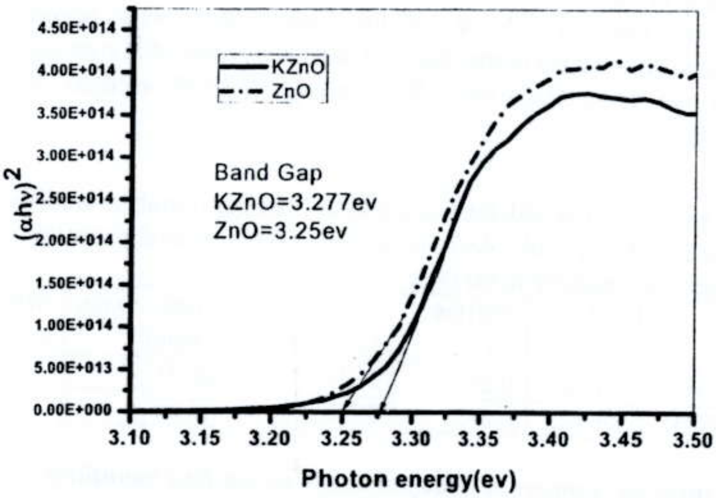

Fig. 4: Plot of absorption coefficient Vs photon energy.

The electrical behaviour of undoped and K doped ZnO nano films was investigated by current-voltage (I-V) measurements, which is realized in fig. 5. Forward bias and reverse bias study are carried out and it was found to exhibit ohmic nature. The linear, ohmic nature showed the good quality of deposited films. Maximum current of 0.201 μA was obtained for K doped ZnO nano film. The potassium concentration was an important factor to influence the current density with respect to bias voltage. The results revealed that conductivity of K doped ZnO nano films is higher than undoped ZnO nano films, due to decreasing grain size and increasing number of grain boundaries.

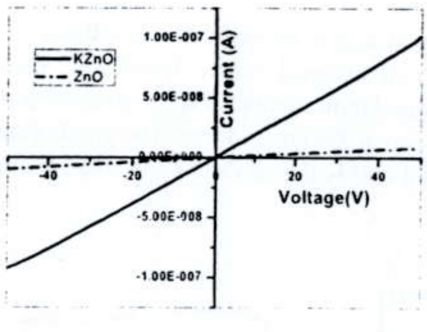

Fig. 5: I-V characteristics of undoped and K doped ZnO nano films

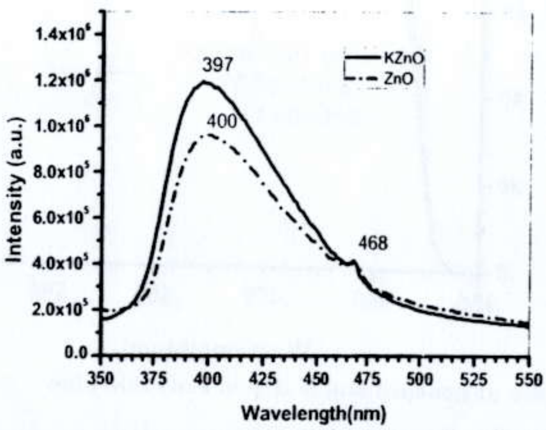

Fig. 6: PL Spectra of undoped and K doped ZnO nano films

Photoluminescence spectra is studied for investigation of the effects of potassium doping on the optical properties of ZnO nano materials. It is expected that the doped ZnO materials have different optical properties in comparison with undoped ZnO. Figure 6 shows the room temperature PL spectra of the undoped and K doped ZnO nano films.PL spectra of undoped and K doped ZnO nano film shows the strong peaks at 400nm and 397nm respectively and blue emission low intensity peak at 468nm. Potassium doped ZnO nano film shows a stronger peak than undoped, indicates that the doped ZnO films have higher optical quality than undoped ZnO nanoparticles. These results are in good agreement with those obtained from the UV Vis spectrophotometer. Since K (1.38Å) has bigger ionic radius than Zn (0.74Å) causes lattice distortion [18]. This effect influences the energy band structure of the ZnO nanoparticles. Oxygen vacancies can be introduced by the new band structure deformation.

Conclusions:

In summary, the Undoped and K doped ZnO nano film were chemically synthesized by simple and inexpensive sol-gel spin coating technique on glass substrate. The XRD spectra reveals the crystalline quality of K doped ZnO nano films without any degradation of the wurtzite structure of the zinc oxide. The size of grains was found to decrease with K incorporation in the films. The incorporation of K in ZnO and decrease in size was confirmed through FESEM, which reflects uniform chromosome type structure. Film showed more than 80% optical transparency in the visible range. Little increase in the band gap was observed with respect to doping of K in ZnO nanoparticles.I-V characteristics analysis reveals increase in currant with doping of potassium in ZnO nanoparticles. The PL spectra of the Undoped and K doped ZnO nano film contains two emission peaks dominated by a strong luminescence peak at 397nm followed by blue visible emission peak at 468nm. The analysis and investigation lead successful incorporation of K dopant in ZnO for transparent electrodes in optoelectronic devices.

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