





## Superlattices and Microstructures

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# Influence of annealing temperature on structural and optical properties of ZnO: In thin films prepared by ultrasonic spray technique

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## Abstract

Transparent conducting indium doped zinc oxide was deposited on glass substrate by ultrasonic spray method. The In doped ZnO samples with indium concentration of 3 wt.% were deposited at 300, 350 and 400 °C with 2 min of deposition time. The effects of substrate temperature and annealing temperature on the structural, electrical and optical properties were examined. The DRX analyses indicated that In doped ZnO films have polycrystalline nature and hexagonal wurtzite structure with (002) preferential orientation and the maximum average crystallite size of ZnO: In before and annealed at 500 °C were 45.78 and 55.47 nm at a substrate temperature of 350 °C. The crystallinity of the thin films increased by increasing the substrate temperature up 350 °C, the crystallinity improved after annealing temperature at 500 °C. The film annealed at 500 °C and deposited at 350 °C show lower absorption within the visible wavelength region. The band gap energy increased from  $E_g=3.25$  to 3.36 eV for without annealing and annealed films at 500 °C, respectively, indicating that the increase in the transition tail width. This is due to the increase in the electrical conductivity of the films after annealing temperature.

## Highlights

- ▶ Indium doped ZnO films were prepared on glass substrates by ultrasonic spray.
- ▶ The films deposited at three different substrate temperature was 300, 350 and 350 °C.
- ▶ The peak corresponding to (002) said the direction along the *c*-axis orientation.
- ▶ The average crystallite sizes of the films are increases after annealing temperature.
- ▶ The band gap and electrical conductivity increases after annealing temperature.

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## Introduction

Zinc oxide (ZnO) which is one of the most important binary II–VI semiconductor compounds has a hexagonal wurtzite structure and a natural n-type electrical conductivity with a direct energy wide band gap of 3.37 eV at room temperature, a large exciton binding energy (~60 meV) [1]. The resistivity values of ZnO films may be adjusted between 10 and 10<sup>-4</sup> Ωcm by changing the annealing conditions and doping [2].

Transparent conducting oxides (TCO) films were used for applications in microelectronic devices, light emitting diodes, thin film, antireflection coatings transparent electrodes in solar cells [3], [4], gas sensors surface acoustic wave devices [5], varistors, spintronic devices, and lasers [6]. The conventional TCO films included tin-doped indium oxide (ITO). However, the cost of ITO films is expensive. In recent years, ZnO:Co films have been extensively studied because they present exhibit high mobility, high optical transparency, high electrical conductivity and have a lower material cost. Currently, many methods are being used to prepare TCO films, such as molecular beam epitaxy (MBE), chemical vapor deposition, electrochemical deposition [5], pulsed laser deposition (PLD), sol-gel process [6], reactive evaporation, magnetron sputtering technique and spray pyrolysis [7]. The conductivity property of the ZnO:Co films can be improved after doping and low temperature.

Although the ultrasonic spray method is especially efficient in producing thin films, transparent, multi-component oxide layers of many compositions on various substrates, including glass, there are still many factors affecting the physical properties of ZnO thin films. These factors include ZnO solution concentration, substrate temperature, post-annealing temperature, annealing atmosphere and film thickness [8], [9], [10], [11], [12]. Among these factors, the influence of substrate temperature and annealing temperature on structural and optical properties of ZnO thin films (especially indium doped ZnO thin films) derived from ultrasonic spray method was less studied. The chosen of temperature in range between 300 and 400°C for substrate temperature is important study with deposition the thin films on glass substrate and the chosen annealing temperature at 500°C for improvement the physical properties such a crystalline structure, optical gap energy and electrical conductivity of the doping thin films. The crystallization of the films at a temperature pass than 500°C the glass substrate is fragile at this temperature.

In this report, we have deposited In doped ZnO samples on glass substrates by ultrasonic spray technique, the indium concentration was used to prepare ZnO: In films is 3wt.%. The thin films were annealed at 500°C for improvement the physical properties. We have studied the effect of the substrate temperatures before and after annealing temperature on the crystallinity, band gap energy and electrical conductivity of the semiconductors.

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## Section snippets

### Preparation of spray solution

ZnO:In solution were prepared by dissolving 0.1 M  $\text{Zn}(\text{CH}_3\text{COO})_2 \cdot 2\text{H}_2\text{O}$  in the solvent containing equal volumes absolute ethanol solution (99.995%) purity, then have added a drops of HCl solution as a stabilized, the mixture solution was stirred at 50°C for 120min to yield a clear and transparency solution. then have added to the precedent solution a 0.03M solution of indium chloride, such that the ratio of In/Zn=0.03. This In content can also be expressed as 3wt.%. The solution became clear and...

### Crystalline structure

Fig. 1 shows the XRD patterns of ZnO:In thin films deposited on glass substrate at three substrate temperatures, the films were realized without annealing. Four diffraction peaks were observed at  $2\theta=31.74^\circ$ ,  $34.52^\circ$ ,  $36.40^\circ$  and  $47.50^\circ$  which can be attributed respectively to (100), (002), (101) and (102) planes of ZnO phase. This result showed the thin films were polycrystalline and had a hexagonal wurtzite structure. As discussed in our published paper [8], where only a (002) diffraction peak is...

## Conclusion

In conclusion, highly transparent conductive Indium doped ZnO thin films have been deposited on glass substrate by ultrasonic spray technique. The influence of substrate temperature before and after annealing temperature on structural, optical and electrical properties was investigated. The whole obtained films have a polycrystalline wurtzite structure and are mainly (002) oriented. We have observed an improvement in the films crystallinity with increasing substrate temperature up to 350 °C in...

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...Thus, for Ni (1455 °C) and Pd (1555 °C) [84], 500 °C falls into the temperature range at which the highest atomic mixing through diffusion takes place, resulting in the largest degree of surface alloying. Similarly, the crystallite size increase with increasing annealing temperature might be the reason behind the activity decay after 500 °C [85–87], as well as

grain agglomeration, which is not favorable for electrochemical reactions on bifunctional surfaces [85–88]. Thus, from the results presented above it is reasonable to infer that the rise and decay of the HOR geometric activity with increasing temperature demonstrates the relationship between Pd and Ni surface atomic distribution and the high HOR activity...

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