

Studies on in-Doped ZnO Transparent Conducting thin Films

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ABSTRACT: In this manuscript we have investigated the influences of indium dopants on zinc oxide (ZnO) thin films regarding physico-chemical properties for application in modern conducting devices. As a starting material, Indium (III) chloride, and $Zn(CH_3COO)_2 \cdot 2H_2O$ were used. The complex TSDC spectrum was obtained by submitting the sample to a constant electrical field $E_p = 10M$ V/m during 2 min at a varying polarization temperature of $T_{max} = 150^{\circ}C$. A minimal sheet resistance with electrical resistivity as low in the range of $10^{-3} \Omega \cdot cm$ was found for this thin film.

Keywords: ZnO, Transparent conducting oxide, Thin films, Spin-coating

I. INTRODUCTION

Modern optical and electrical applications are two core areas which have been intensively investigated for conducting oxide (TCO) films. The examples are liquid crystals, organic light-emitting diodes, thin-film transistors, and thin-film solar cells [1-5]. The ideal situations for these thin films are having low resistivity, high transmittance in the visible region with appropriate stability in terms of thermal /chemical [6-7]. Indium oxide based ITO's have been considered superb because of good electrical and optical properties [8-9]. Recently, zinc oxide (ZnO) has come as a replacement because of wide band gap (3.37 eV at room temperature) with high excitation binding energy (60 meV) and multiple functionalities [10-11]. III group elements from the periodic table have been used for improving the electrical conductivity and optical transmittance of ZnO films [12]. In this study, In-doped ZnO (hereafter IZO) thin films have been prepared by sol-gel spin-coating method. We study the effect of these dopants on the microstructure, electrical, and optical properties as a function of doping concentration.

Experimental details

Thin films were prepared by doping of Indium (III) chloride, and different $[In]/[In+Zn]$ atomic per cent ratios, *i.e.*, 1.0, 2.0 and 2.5 at% were tested. A molten tin bath was used for substrate heating, and the temperature of the bath was controlled and monitored by a thermocouple. Substrate temperatures selected were 400, 425 and 450 $^{\circ}C$, within ± 1 $^{\circ}C$ variation. The deposited thin films were presented an apparent homogeneous surface and very good adhesion with the substrate. The sheet resistance, of indium thin film were measured by the four-point probe technique, taking into account the adequate geometrical corrections. The vacuum aluminized film was sandwiched between two electrodes. The sample was thermally polarized at different temperatures and electric fields. The sample holder was placed in a digitally controlled oven heated up to poling temperature. The sample was then allowed to remain at that temperature for about 30 min. Then desired strength of electric field was applied for 1 h at poling temperature. The sample was allowed to cool down at room temperature in the presence of applied field.

The complex TSDC spectrum was obtained by submitting the sample to a constant electrical field $E_p = 10M$ V/m during 2 min at a varying polarization temperature of $T_{max} = 150^{\circ}C$. The sample is then cooled, with a ramp of 20 C/min, to liquid nitrogen temperature in the presence of the electrical field. The field was then suppressed at around 25 $^{\circ}C$ and the electrodes were short-circuited for 15 min. All measurements were repeated to verify the reproducibility and the accuracy of the results. The current was measured with an electrometer and recorded by using X-Y plotter. The electrometer was also coupled to a PC for data collection. A platinum temperature sensor Pt100, mounted in the sample holder and adjacent to the film, allowed the temperature measurement with a precision of 0.05 $^{\circ}C$. The heating rate of 5 $^{\circ}C$ /min was used and controlled by a temperature regulator. The DSC measurements were performed in a Perkin-Elmer with a controlled cooling accessory.

II. RESULT & DISCUSSION

Figure 1 showed the depolarizing current vs. Temperature at constant polarising temperature T_p (*i.e.* 40, 60, 80 and 90 $^{\circ}C$). Figures 2 showed the TC curve for charging mode obtained for polarising temperature T_p (*i.e.* 40, 60, 80 and 90 $^{\circ}C$) with polarizing field E_p (05MV/m) for Al-Al electrode system respectively. These characteristics are showing two to three peaks at each temperature, one at low temperature (β peak), which is

associated with dipolar relaxation, and other at higher temperature (α peak) that appears due to space charge relaxation. The analysis of the shape of TSDC peak makes it possible to obtain the activation energy, relaxation time and charge released. When polarizing field was increased, the magnitudes of both peaks increased.

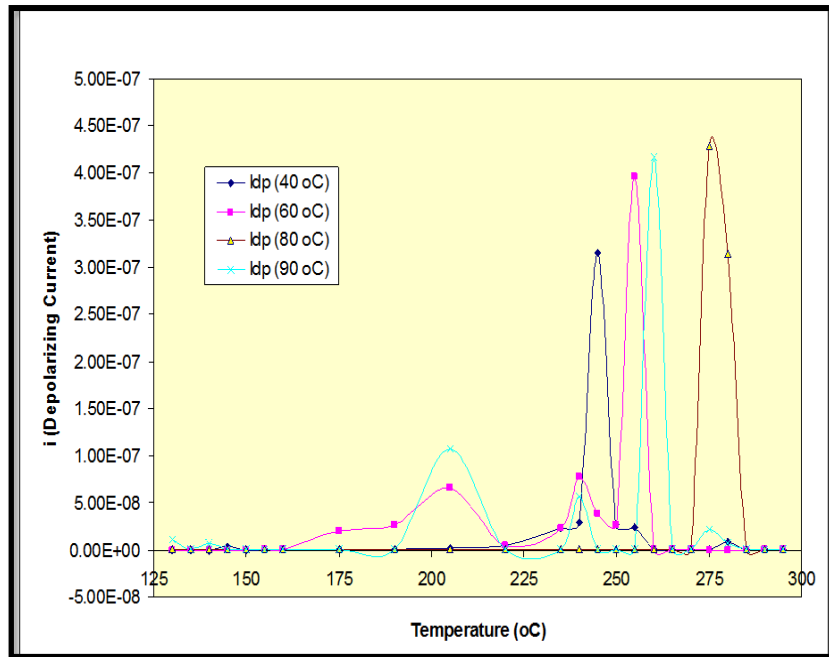


Figure 1- Depolarizing current of doped system

The present study on polarizing field and temperature dependence of electrical conduction in undoped and doped films has been carried out to understand the role of dopant when added in varying concentrations.

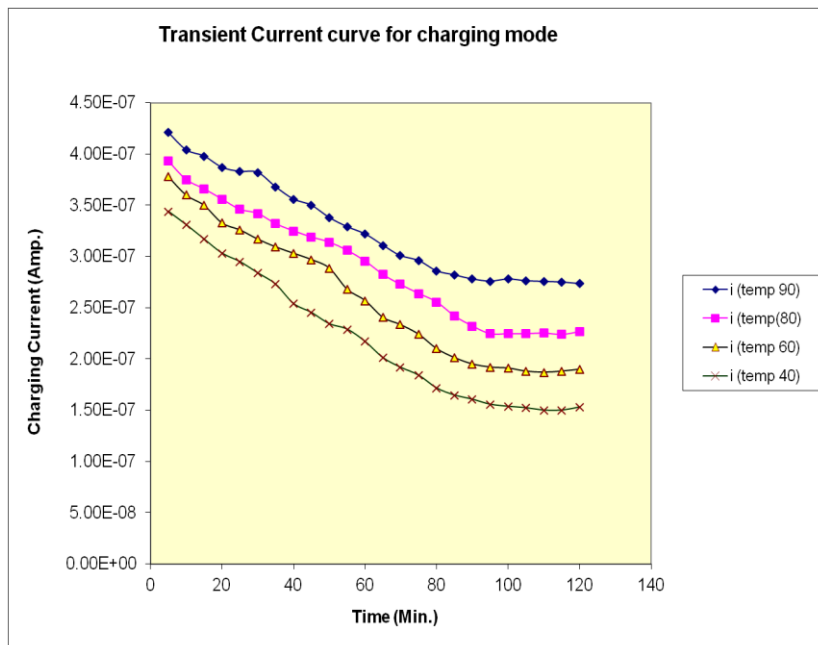


Figure 2- TC curve for charging mode

The conductivity also depends on temperature. As the temperature increases film becomes soft and mobility of the main chain segments as well as the rotation of side groups become easier. Thus, at higher temperature more and more dipoles are oriented resulting in the higher equivalent surface charge density i.e. as temperature increases conductivity increases as per Arrhenius equation

$$\sigma = \sigma_0 \exp(-E_a/KT),$$

where σ_0 is the pre-exponential factor, E_a the activation energy of conduction and K the Boltzmann's constant. The activation energy for these low and high temperature regions are also different. Electrical conductivity of all the samples were measured by the four-point probe method by considering the geometry of electrical contacts located over them. It was observed that, with the increase in the substrate temperature, the resistivity of the ZnO:In thin films decreases, reaching a minimum value, for a fixed [In]/[In+Zn] ratio in the starting solution. From the measured electrical conductivity of the samples presented in Table 1. Low resistivity of the ZnO:In films containing 2.5 at% nominal indium contents fabricated at $T_s = 450$ °C might be associated with their compact surface morphology and bigger crystallite sizes.

[In]/[In+Zn]	Electrical Resistivity ($\Omega \cdot \text{cm}$)		
	400 °C	425 °C	450 °C
1.0	1.1×10^{-2}	4.4×10^{-3}	4.1×10^{-3}
2	8.6×10^{-3}	3.9×10^{-3}	3.3×10^{-3}
2.5	3.5×10^{-3}	2.2×10^{-2}	5.5×10^{-3}

III. CONCLUSION

Indium-doped ZnO thin films were prepared by sol-gel spin-coating method for TCO applications. All films had a crystal structure, and a minimum sheet resistance of $10^3 \Omega \cdot \text{cm}$ for doped ZnO thin film. In conclusion, the structural, morphological, electrical, and optical characteristics of IZO thin films were observed, and In doping seems to be more effective.

Competing interests

The authors declare that they have no competing interests.

REFERENCES

- [1]. Oztas M, Bedir M: Thickness dependence of structural, electrical and optical properties of sprayed ZnO:Cu films. *Thin Solid Films* 2008, 516: 1703.
- [2]. Chen M, Pei ZL, Sun C, Gong J, Huang RF, Wen LS: ZAO: an attractive potential substitute for ITO in flat display panels. *Mater Sci and Eng B* 2001, 85: 212.
- [3]. Ohyama M, Kozuka H, Yoko T: Sol-gel preparation of transparent and conductive aluminum-doped zinc oxide films with highly preferential crystal orientation. *J Am Ceram Soc* 1998, 81: 1622.
- [4]. Cheong KY, Muti N, Ramanan SR: Electrical and optical studies of ZnO:Ga thin films fabricated via the sol-gel technique. *Thin Solid Films* 2002, 410: 142.
- [5]. Rech B, Wagner H: Potential of amorphous silicon for solar cells. *Appl. Phys. A. Mater Sci Process* 1999, 69: 155.
- [6]. Fu Z, Lin B, Zu J: Photoluminescence and structure of ZnO films deposited on Si substrates by metal-organic chemical vapor deposition. *Thin Solid Films* 2002, 402: 302.
- [7]. Nunes P, Fernandes B, Fortunato E, Vilarinho P, Martins R: Performances presented by zinc oxide thin films deposited by spray pyrolysis. *Thin Solid Films* 1999, 337: 176.
- [8]. Asmar RA, Juillaguet S, Ramonda M, Giani A, Combette P, Khoury A, Foucaran A: Fabrication and characterization of high quality undoped and Ga₂O₃-doped ZnO thin films by reactive electron beam co-evaporation technique. *J Cryst Growth* 2005, 275: 512.
- [9]. Cooray NF, Kushiya K, Fujimaki A, Sugiyama I, Miura T, Okumura D, Sato M, Ooshita M, Yamase O: Large area ZnO films optimized for graded band-gap Cu(InGa) Se₂-based thin-film mini-modules. *Sol Energy Mater and Sol Cells*. 1997, 49: 291.
- [10]. Kluth O, Schöpe G, Rech B, Menner R, Oertel M, Orgassa K, Schock HW: Comparative material study on rf and dc magnetron sputtered ZnO:Al films. *Thin Solid Films* 2006, 502: 311.
- [11]. Sub ES, Kang HS, Kang JS, Kim JH, Lee SY: Effect of the variation of film thickness on the structural and optical properties of ZnO thin films deposited on sapphire substrate using PLD. *Appl Surf Sci* 2002, 186: 474.
- [12]. Fang GJ, Li DJ, Yao BL: Effect of vacuum annealing on the properties of transparent conductive AZO thin films prepared by dc magnetron sputtering. *Phys Status Solidi A* 2002, 193: 139.
- [13]. Sanon G, Rup R, Mansingh A: Growth and characterization of tin oxide films prepared by chemical vapour deposition. *Thin Solid Films* 1990, 190: 287.