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Characterization of ZnO:Al thin films obtained by spray pyrolysis technique

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Abstract

Films of zinc oxide doped with Al (ZnO:Al) are prepared using the spray pyrolysis technique. The effect of doping Al on the physical properties of ZnO:Al is studied. In this study the polycrystalline ZnO:Al films with the different Al concentration ([Al]/[Zn] in the starting solution was varied from 0 to 0.6 wt.%) were prepared. These films were confirmed to show the high crystallinity by X-ray diffraction technique. The smallest sheet resistance value, around 207 Ω/□, was obtained using a [Al]/[Zn] ratio of 0.125 wt.% in starting solution. The optical transmittance was about 75% in visible range for the optimum film.

Keywords: ZnO; ZnO:Al; Spray pyrolysis

1. Introduction

Thin films of non-stoichiometric and doped metallic oxides of ZnO, In_2O_3 ,SnO_2, CdO, In_2O_3:Sn (ITO),SnO_2:F, In_2O_3:F, ZnO:Al etc. have attracted a large interest because of their high electrical conductivity and optical transmittance in the visible region of solar spectrum making them suitable for their application in different kinds of opto-electronic devices [1–4]. Because of the fact that a high conductivity and a high transmittance in the visible region can be achieved simultaneously, they are used as transparent electrodes in thin film solar cells [5–7]. Among these materials, zinc oxide was felt important compared with tin oxide and indium oxide due to its stability in hydrogen plasma, which is of unique importance in amorphous and microcrystalline silicon areas [8]. ZnO thin films are also used as UV and ozone gas sensor [9].

The spray technique is one of the most commonly used technique for preparation of transparent and conducting oxides owing to its simplicity, safety, non-vacuum system of deposition and hence inexpensive method for large area coatings [10].By characterizing the deposition parameters especially the spraying temperature, flow rate and the amount of doping concentration, it is possible to deposit Al-doped ZnO (AZO) thin films having suitable values of sheet resistance and transmittance. ZnO films can be doped with Ga [11], In [12], Cu, Fe, Sn [13] and F [14]. In this paper, we present results of investigation on changes in physical properties of Al doped zinc oxide films deposited by the spray pyrolysis technique. The aim of the work is to show the effect of doping using a simple way of optimization of the samples with the help of two probes and XRD technique.

2. Experimental procedures

Al-doped ZnO films were deposited using spray pyrolysis technique on soda lime glass substrate. The spray

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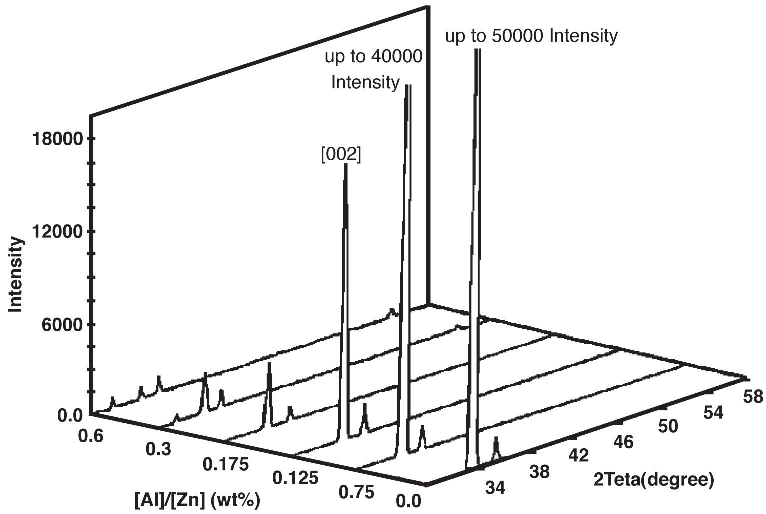

Fig. 1. X-ray spectra for films deposited at several Al concentration (substrate temperature 500 °C).

pyrolysis apparatus used in this work consists of a home made spraying unit, substrate holder with heater, and enclosure. The glass substrate was kept on a stainless steel (ss) plate that was heated by a 3 kW heater using canthal-heating coil. The heater is capable of heating the substrate up to a temperature of 700 °C. The temperature of the substrate is controlled by using temperature controller and Chromel–Alomel thermocouple kept at the center of a ss plate. The carrier gas used in all the experiments was air, which was supplied by an air compressor. The air produced by the compressor was first filtered and then connected to the glass spray-gun (atomizer) through a flow meter to control its flow.

The custom glass spray gun having a nozzle diameter of 0.2 mm was positioned at a distance of 30 cm above the substrate. The whole assembly was kept in an enclosure connected to an exhaust.

Zinc acetate dehydrate, aluminum chloride hexahydrate, deionized water and ethanol were used as a starting material. To stabilize the starting solution, a few drops of acetic acid were added. The ratio of [Al]/[Zn] in the starting solution was varied between 0 and 0.6 wt.% keeping all other parameters constant (i.e., substrate temperature 500 °C, air flow rate 6 l/min(l pm) , distance between substrate to nozzle $D_{sn}=30$ cm and solution composition). The thickness of the films can be calculated by using interference pattern observed in the visible region following the formula given by Swanepoel [15].The thickness of the films was found to lie in the range of 1200–1400 nm.

The crystalline structure was obtained by means of an X-ray diffractometer with Cu-Kα radiation (Philips-pw-1830). In addition, to evaluate the electrical and optical properties of films, a two-probe measurements and a UV–VIS spectroscopy (Cary 100 Scan Varian) were carried out respectively.

3. Results and discussion

Fig. 1 shows the X-ray diffraction patterns (XRD) of ZnO: Al films deposited at different quantity of aluminum chloride as a dopant. It is observed that in all cases, these films are polycrystalline showing a preferential orientation in the (002) direction to be the most predominant for films prepared with [Al]/[Zn] ratio of 0 to 0.3 wt.%. However, for Al concentration higher than 0.3 wt.%, the intensity of (101) orientation become comparable to (002)

Table 1
Variation of sheet resistance, visible transmission (at 550 nm) and XRD peak intensity of ZnO and ZnO:Al thin films with Al concentration

Al concentration (wt.%)	0	0.05	0.075	0.1	0.125	0.15	0.175	0.2	0.3	0.4	0.5	0.6
Sheet resistance Ω/□	750	550	370	285	207	230	280	560	1000	1200	1400	3000
Visible transmission % at 550 nm	75	72	72	72	75	68	69	68	68	68	69	69
(100) Peak intensity (×1000)	–	–	–	–	–	–	–	0.5	0.6	0.6	0.85	1
(002) Peak intensity (×1000)	50	41	40	22	16	11.2	3.75	3.5	2.5	1.05	0.95	1
(101) Peak intensity (×1000)	0.2	1.25	1.25	0.4	2	1.5	0.9	1.2	1.2	1.05	1	1.1
(102) Peak intensity (×1000)	–	–	–	–	–	–	–	–	–	–	–	0.17
(110) Peak intensity (×1000)	–	–	–	–	–	–	–	0.2	0.2	0.2	0.5	0.6