

Highly transparent and lower resistivity of yttrium doped ZnO thin films grown on quartz glass by sol–gel method

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ABSTRACT

We prepared highly transparent yttrium-doped ZnO (YZO) thin films on quartz glass by a sol–gel method, and then annealed them at 600 °C in vacuum. All samples showed hexagonal wurtzite structure with a preferential orientation along the (002) direction. We observed the average grain size of Y: 2 at% thin film to be in the range of 15–20 nm. We observed blue shift in the optical bandgap (3.29 eV → 3.32 eV) by increasing the Y concentration (0–2 at%), due to increasing the number of electrons, and replacing the di-valent (Zn^{2+}) with tri-valent (Y^{3+}) dopants. Replacing the higher ionic radii (Y^{3+}) with smaller ionic radii (Zn^{2+}) expanded the local volume of the lattice, which reduced the lattice defects, and increased the intensity ratio of NBE/DLE emission ($I_{\text{NBE}}/I_{\text{DLE}}$). We also observed the lowest (172 meV) Urbach energy of Y: 2 at% thin film, and confirmed the high structural quality. Incorporation of the appropriate Y concentration (2 at%) improved the crystallinity of YZO thin films, which led to less carrier scattering and lower resistivity.

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1. Introduction

Zinc oxide (ZnO) thin films have aroused great interest as transparent conductive oxides (TCO), due to their wide bandgap (3.37 eV), chemical and thermal stability, and large exciton binding energy (60 meV) [1–5]. Due to the intrinsic properties of ZnO, thin films have attracted considerable interest for application in LEDs [6], diodes [7], detectors [8], UV lasers [9], solar cells [10], gas sensors [11], nanogenerators [12], flat-panel displays [13], magneto-optical switches [14], thin film transistors [15], and so on. Extensive research has been conducted on doped ZnO thin films (ZnO: Al, In, Ga, Y and so on), and has improved their optical and electrical properties [16–18]. In the last decade, rare earth dopant materials like scandium (Sc), lanthanum (La), and yttrium (Y) in ZnO thin films have attracted much attention, due to their non-toxicity and abundant availability in the Earth's crust [19–23]. The incorporation of rare earth atom/ions into suitable matrixes produces possible intra-4f optical transitions, which change the optical properties of ZnO thin films. Generally, the crystal orientation and the surface morphology of ZnO thin films depend upon the growth parameters, like dopant concentration and post-annealing. Therefore, we can alter the structural, physical, and electrical

properties of ZnO–TCOs by varying the dopant concentration and subsequent annealing.

Compared to dopant concentration, Y-doped ZnO (YZO) thin films have great importance because of the stronger bonding between oxygen and yttrium. Due to the high bonding energy of Y–O ($\sim 719.6 \text{ kJ mol}^{-1}$), there is less tendency to form oxygen vacancy defects, which improves the device stability. Jun et al. [24] found that YZO-based TFTs improved electrical performance due to lower defect density. Han et al. [19] determined that the incorporation of Y concentration reduced the resistivity of YZO thin films.

Taking into consideration the fabrication and processing scale of YZO thin films, the sol–gel method has been preferred. Yu et al. [25] observed a shift in the UV emission of YZO thin films prepared from sol–gel and subsequently annealed at 550 °C. Sharma et al. [26] found that the diameter and density of YZO nanorods could be controlled by modulating the grains of seed layers. Kaur et al. [27] demonstrated the relationship between stress relaxation and optical bandgap of YZO thin films. Heo et al. [28] used the Y concentration in ZnO thin films to tailor the physical and electrical properties grown on Si substrates by a sol–gel method. Earlier reports observed the resistivity of YZO (Y: 2 at%) thin films to be $\sim 10^{-2} \Omega\text{-cm}$, which is insufficient for device applications. In order to meet the requirement for the devices, it is necessary to reduce the resistivity, and show high transmittance in the visible region.

The main objective of our paper is to achieve highly

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transparent and conductive YZO thin films grown on quartz glass, which can be done by the appropriate addition of Y concentration and subsequent annealing. So we report the structural, optical and electrical properties of YZO thin films with respect to dopant concentration (Y: 0, 1, 2, 3, 4 and 5 at%) and subsequent annealing at 600 °C prepared on quartz glass substrates by a sol-gel method.

2. Experimental

We prepared Y-doped ZnO (YZO) thin films on quartz glass substrates by a sol-gel method at room temperature. Fig. 1 provides a flow chart and brief description for the synthesis of highly transparent YZO thin films. We prepared 0.5 M sol by dissolving the zinc acetate dihydrate ($\text{Zn}(\text{CH}_3\text{COO})_2 \cdot 2\text{H}_2\text{O}$), and yttrium acetate hydrate ($\text{Y}(\text{CH}_3\text{COO})_3 \cdot \text{H}_2\text{O}$) in 2-methoxyethanol ($\text{C}_3\text{H}_8\text{O}_2$) at room temperature. The Y concentration was varied between 1–5 at%. An equimolar amount of monoethanolamine (MEA) was added drop-by-drop to eliminate the turbidity and precipitates from the solution, which was then aged for 24 h to obtain optimal viscosity. The prepared solution was spin-coated on quartz glass substrates at 3000 rpm for 20 s, and dried in electric oven at 300 °C for 10 min. Before deposition of YZO thin films, the quartz glass was ultrasonically cleaned in 2-propanol, acetone, methanol, and deionized water for 15 min each step. We then annealed the deposited YZO thin films at 600 °C for 3 min in vacuum by using rapid thermal annealing (RTA).

We obtained the crystallographic characteristics from X-ray diffraction (XRD) (Rigaku-Ultima-IV), and monitored the surface morphology of the thin films by field emission-scanning electron microscopy (FE-SEM) (Hitachi-S-4700). The composition of films was confirmed from energy dispersive X-rays spectroscopy (EDX) (EX-220). The transmission and absorption spectra of YZO thin films were taken by UV-visible spectrophotometer (US-S-4100). The bandgap of YZO films was evaluated from Tauc's plot. Photoluminescence (PL) measurements were carried out by using excitation of He-Cd laser at the wavelength of 325 nm. We studied the electrical properties of YZO thin films by Van der Pauw Hall measurement system (Ecopia-HMS-3000) at room temperature.

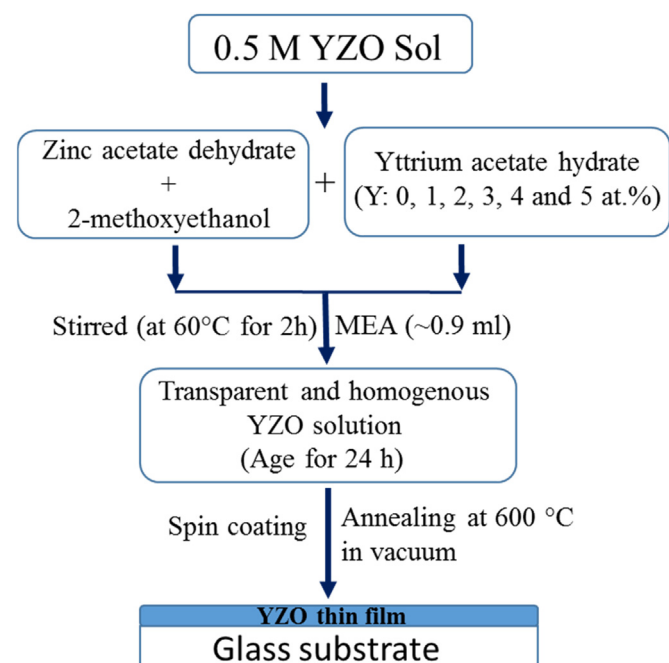


Fig. 1. Flow chart for the synthesis of YZO thin films.

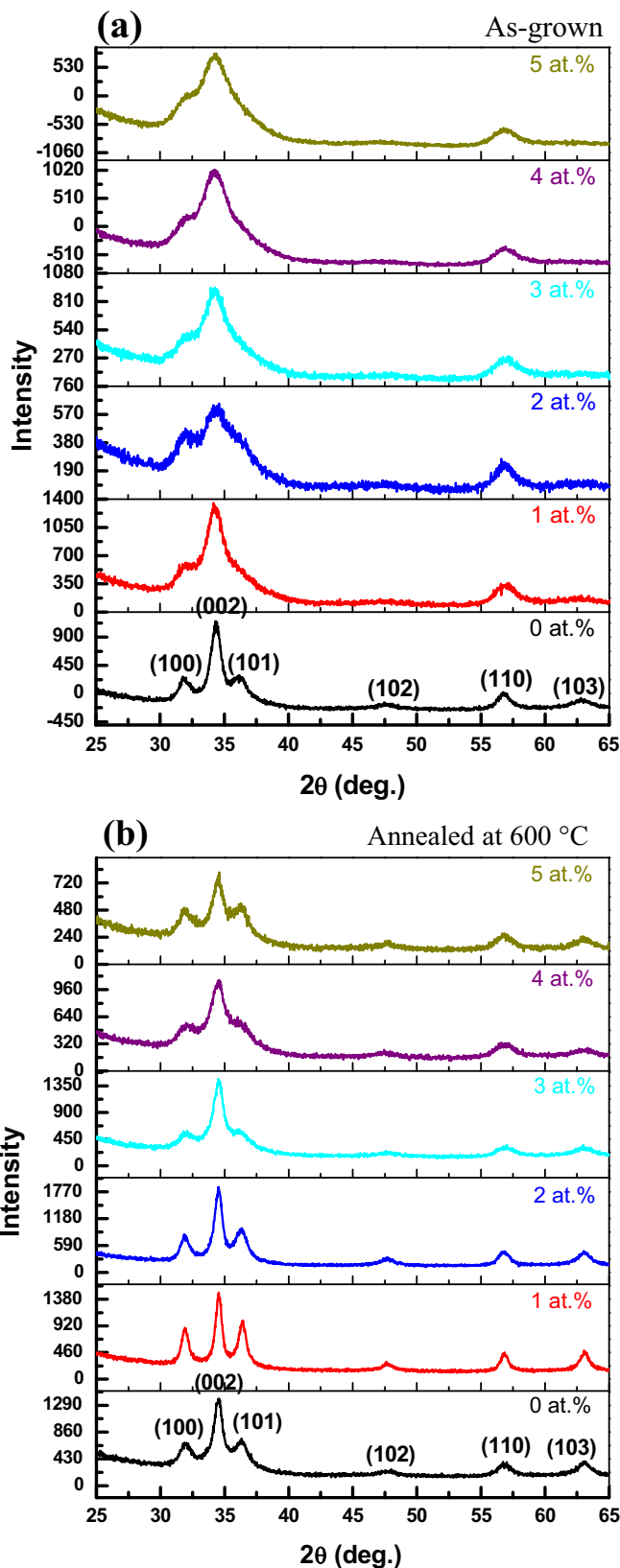


Fig. 2. X-ray diffraction pattern of YZO thin films with respect to Y concentration (Y: 0, 1, 2, 3, 4, and 5 at%), (a) as-grown films, and (b) annealed films at 600 °C in vacuum.

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