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Effects of annealing temperature on ZnO and AZO films prepared by sol–gel technique

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ABSTRACT

Zinc oxide (ZnO) films have the potential in the emerging thin-film technologies which can be employed in thin-film solar cells, transistors, sensors and other optoelectronic devices. In this work, low cost sol–gel spin-coating technique was used to synthesize the ZnO films. The influences of annealing temperature on the structural and optical properties of ZnO and aluminum doped ZnO (AZO) films were investigated. The structural properties of the ZnO films such as surface morphology and crystallinity were determined using atomic force microscopy (AFM) and X-ray diffractometry (XRD), respectively. The optical properties of the ZnO films were characterized by the ultraviolet–visible (UV–vis) spectroscopy and Tauc method was adopted to estimate the optical gap. The experimental results reveal that the thermal annealing treatment affects the properties of the ZnO films. The effects of the low range annealing temperature on the sol–gel ZnO films addressed in this investigation will be discussed in this paper.

1. Introduction

As a semiconducting material with wide band gap (3.37 eV) and large exciton binding energy (60 meV), zinc oxide (ZnO) films have gained the attention among researchers in the studies of their practical applications in thin-film solar cells, thin film transistors, surface acoustic wave devices and sensors [1–5]. Grown ZnO films usually exhibit n-type conductivity which is caused by intrinsic defects, interstitial zinc and oxygen vacancies [6,7]. The improvement of the electrical conductivity of ZnO films can be achieved by adding doping components such as aluminum (Al) and indium (In). Various growth techniques such as molecular beam epitaxy (MBE) [8], metal-organic chemical vapor deposition (MOCVD) [9,10], and sputtering technique [11,12] have been employed to produce ZnO films. These techniques can be used to produce good quality ZnO films, but the use of sophisticated and high cost equipment set-up raises the total manufacturing cost. On the other hand, sol–gel spin coating method offers simplicity, easy to control, and cost efficient way to fabricate ZnO films [13,14]. The characteristics of the films are typically influenced by sol–gel process parameters such as precursor concentration and aging time, as reported in literature [15–18]. There are studies on the effect of annealing temperature as well [19–22]. However, most of the works were focused on annealing temperature above 400 °C. For example in the work of Sengupta et al. [19], the effects of annealing temperature from 400 °C to 700 °C on the structural and optical properties of ZnO films were studied. Such temperature range may cause problem to the glass or plastic substrates since they can only sustain temperatures up to 500 °C. Therefore, the effects of lower temperature annealing treatment need to be investigated in order to determine the optimum annealing temperature in the fabrication of ZnO films on glass substrate.

In this work, ZnO and AZO films were deposited onto a glass substrate using sol–gel spin coating method. The main goals of this study are to investigate the influence of the low range annealing temperature (100–400 °C) on the quality of the ZnO and AZO films, and to find out the optimum temperature for annealing treatment. The effects on structural properties such as grain size, lateral size, surface roughness and crystallinity as well as optical properties of both ZnO and AZO films were investigated.

2. Experimental

Glass substrates were cleaned using isopropanol (IPA), rinsed in deionized (DI) water, and wiped dry with clean room grade

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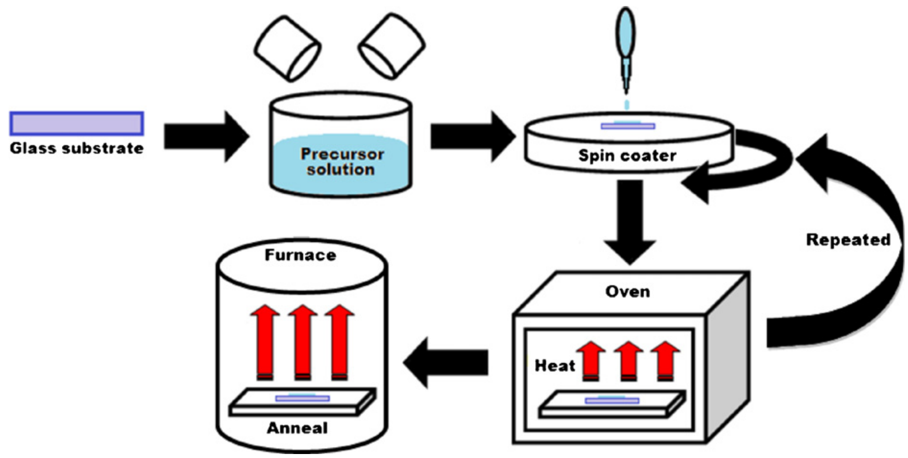

Fig. 1. Sol–gel spin coating process.

lint free wipers. The ZnO solution was prepared with the mixture of zinc acetate dihydrate [$Zn(CH_3COO)_2 \cdot 2H_2O$] as precursor, isopropanol (IPA) [C_3H_3OH] as solvent, and monoethanolamine as stabilizer. The precursor concentration was maintained at 0.5 mol. Meanwhile, the AZO solution was prepared separately by adding aluminum nitrate nonahydrate as dopant into the ZnO solution, while keeping the molar ratio of Al dopant at 1%. The resultant solutions were stirred with the speed of 200 revolutions per minute (rpm) at 60 °C for 2 h using a magnetic stirrer. It was then kept in a room temperature for aging process for another 22 h. The solutions were then dropped onto glass substrates, and spin coated at 3000 rpm for 20 s. After spin coating, the substrates were then pre-heated at 150 °C for 3 min. Finally, the substrates were annealed using a high temperature furnace. The process of sol–gel spin coating is as shown in Fig. 1. In order to study the effect of annealing treatment, the samples were annealed at temperatures of 100 °C, 200 °C, 300 °C, and 400 °C for 1 h.

The characterization of the ZnO films began with the optical transmittance of the ZnO films, which was determined using the

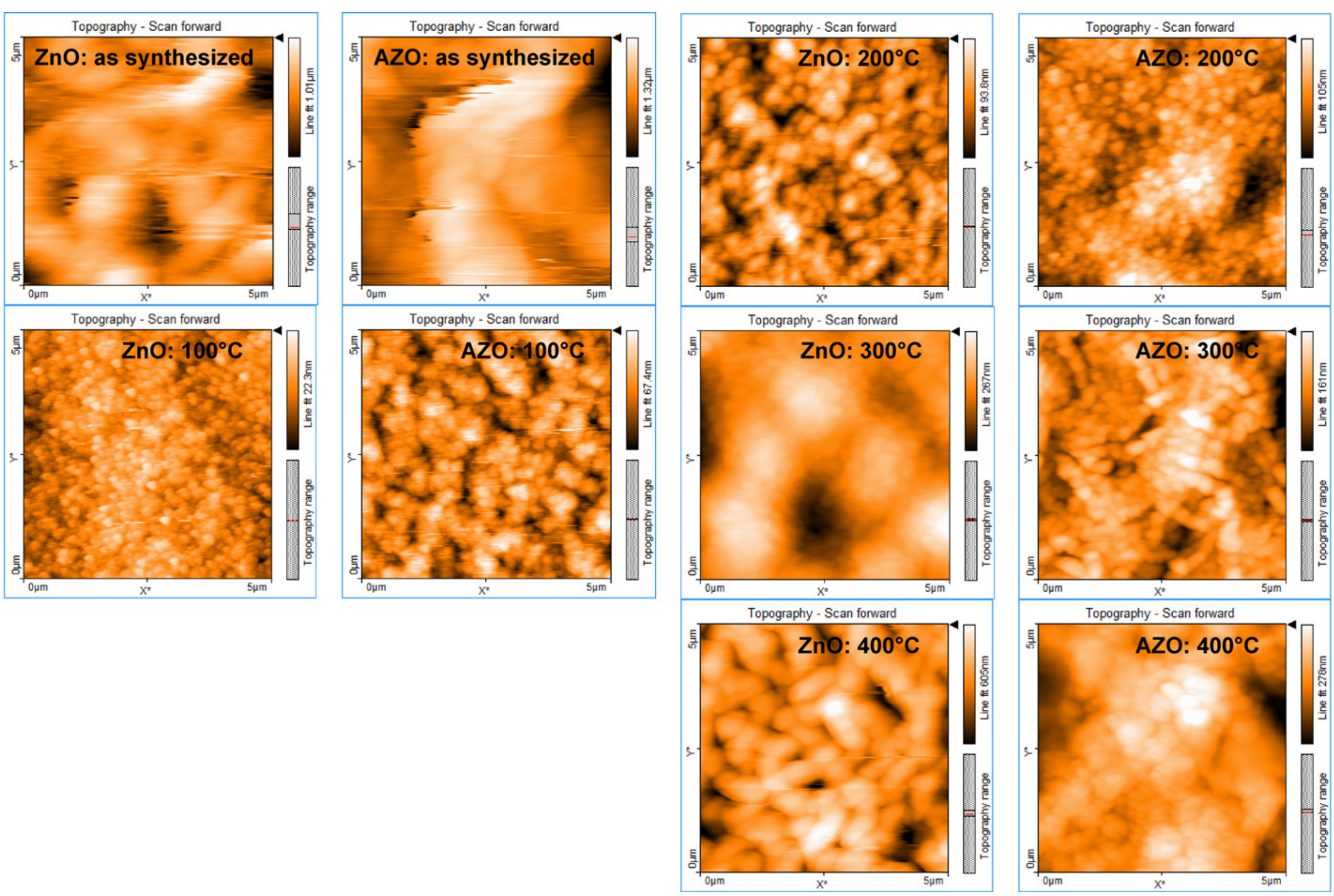

Fig. 2. AFM micrograph of ZnO and AZO films as synthesized and annealed at different temperatures.