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Muscovite mica as a flexible substrate for transparent conductive AZO thin films deposited by spray pyrolysis

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ABSTRACT

In this study, Aluminum doped and undoped ZnO transparent conductive oxide thin films on a specific substrate, i.e., the muscovite mica sheet, were prepared by the spray pyrolysis technique. The substrate was used in this study for its unique properties, namely simultaneous flexibility and high thermal stability. The effects of doping on structural, morphological, optical and electrical characteristics of the samples were investigated and discussed. The obtained thin films were mostly polycrystalline with a hexagonal structure. UV–Visible spectroscopy analysis showed high average transparency (more than 80%) in visible and near-infrared region. Field emission scanning electron microscopy analysis proved the homogenous morphology of the films and showed their resistivity to be in the range of 10^{-3} Ω cm.

1. Introduction

Zinc oxide (ZnO), an n-type semiconductor oxide, has gained much interest for its broad and various applications in optoelectronic, microelectronic and spin transport electronic devices [1–4]. ZnO is a wide direct band gap oxide (3.34 eV) and has found many applications in solar cells [5], diodes [6], heat mirrors [7,8], piezoelectric transducers [9,10], gas, chemical and biological sensors [11,12]. The photocatalytic properties of metal oxides such as ZnO are also well known [13]. Recently, some efforts have been done to utilize the unique properties of graphene to improve efficiency of photocatalysis [14–17], solar cells [18,19] and batteries [20,21]. Although, many of interior mechanism and details are still not completely clear, but results show that the incorporation of ZnO/graphene improve the performance of the mentioned devices. Moreover, ZnO high conductivity and nontoxicity, attract most researcher's attention to this transparent conductive oxide. Additionally, abundance in nature and high stability in hydrogen plasma [22] make this oxide economical and a suitable replacement for indium tin oxide thin films.

Many deposition techniques have been used to form ZnO thin films on different substrates, including pulsed laser [23], sol-gel [24], sputtering [25], chemical bath deposition [26], arc discharge [27] and spray pyrolysis [28]. The spray pyrolysis is an attractive and practical technique for thin film deposition in large area and good quality. This method is cheap and simple to use due to being independent of vacuum condition for deposition [29].

Replacement of the substrate with flexible substrates is obtaining a great interest due to lightness, flexibility and transparency. Several flexible substrate such as polyimide [30,31], polycarbonate [32], polyethylene terephthalate [33,34] and polyethylene naphtahlate [35,36] have been used for ZnO and AZO thin films deposition by different techniques. However, there are some restrictions for polymer substrates. They have low processing temperature. Moreover, thermal stresses between the substrate and the thin films are significant. Although, the glass substrates and metal foils can overcome the mentioned limitations, but they are rigid and can't applied for flexible devices [37,38].

The muscovite mica sheet with $KAl_3Si_3O_{10}(OH)_2$ formula is a well-known substrate for growth of semiconductors, metal thin films and well-oriented nanowire arrays [39,40]. It is a perfect electrical-thermal insulator, which is stable in water, inert to most acids, alkalis, solvent and oil and also, is highly transparent and flexible in ultra-thin form. Some information on the muscovite mica has been reported [41,42]. The mica substrate is easily cleaved and obtains a highly smooth and flat surface area that makes it an ideal substrate for AFM imaging [43–45] and thin film applications [46–48]. These sheets are stable up to 600 °C [49]. It can be expected that mica muscovite will be widely used as a suitable substrate for optoelectronic devices in the future. A few investigations have been carried out about thin film preparation on the muscovite mica sheet. These works often belong to the formation mechanism and physical characterization of Au and Ag films [50–54] and biological studying [55,56]. The prepared samples were deposited on freshly cleaved mica substrates. The surface contaminations were removed by washing in distilled water and heating [52–54,57].

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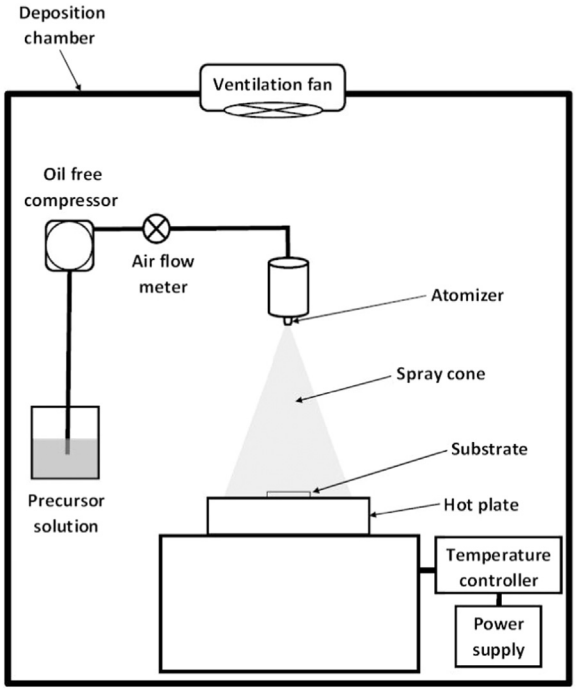

Fig. 1. The schematic diagram of the spray pyrolysis system.

2. Experimental details

The ZnO films were prepared from the zinc acetate-dehydrated solution in a blend of deionized water, acetic acid (CH_3COOH) and absolute ethanol (8:3:1 vol ratio). Addition of some acetic acid was required to stabilize the pH and avoid precipitation of zinc hydroxide in the precursor solution.

The resulting solution was sprayed on the heated cleaved mica sheets ($35 \times 40 \times 3\ mm^3$) by using compressed air as a carrier gas. The substrates were placed on the metal plate. The plate temperature was controlled through heating element inside the metal plate. Doping of the films was achieved by adding weight percent (wt%) $AlCl_3$ to the precursor solution. The nozzle to substrate distance and deposition flow-rate during the deposition process was kept ≈ 30 cm and 12 lit/min, respectively. The scheme of our system is displayed in Fig. 1.

Philips (PW 1800 model) X-ray diffraction system was used for studying the structural properties of the films. The samples were placed under CuK_α radiation (1.5406 Å) as the X-ray source at 40 KeV and 30 mA over a range from 0 to 60 degrees and a rate of 2 degrees per minute. The morphology of the obtained thin films was observed by TESCAN MIRA3 XMU VP-FESEM system at 15 kV. Electrical analysis was performed using the van der Pauw and Hall Effect measurement system. The optical transmittance of the ZnO and AZO samples was investigated by the UV–visible spectrophotometer (Cary 100 Scan Varian) in the range from 200 to 800 nm.

3. Results and discussion

3.1. Structural characteristics

The mica sheets had crystalline structure so that the other peaks (*), were related to the sheet substrate (Fig. 2) [58]. Certainly, some of the ZnO peaks overlapped with the mica sheet substrate peaks. This occurred for (101), which was placed at ≈ 36.3°.

The XRD pattern of ZnO doped with different concentrations of Al is shown in Fig. 2. All the samples were prepared and optimized at 550 °C. Diffraction peaks indicated that the films were polycrystalline with a hexagonal wurtzite crystal structure. These measurements revealed that

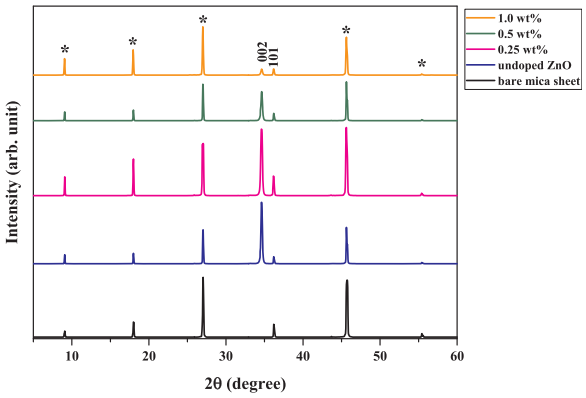

Fig. 2. XRD patterns for bare mica sheet, ZnO and different Al doped ZnO (AZO).

the sprayed films preferred growth orientation along c-axis, (002) plane, perpendicular to the substrate. Similarly, this preferred growth orientation was reported by others investigators [59,60] and no changing in orientation occurred for the doped films.

This reveals the presence of a single-phase ZnO. It was further found that increasing in the doping level led to an initial increase in the intensity of the (002) peak [61]. For the low doped films (0.25 and 0.5 wt %), AZO crystallites were strongly oriented on (002) direction and analogical with another mica sheet peak. Furthermore, increasing Al doping to high level (1.0 wt%) led to lower orientation and poor crystallinity. This deterioration can be attributed to Al interstitial inclusion in the ZnO lattice [61]. In higher doping level, aluminum atoms tend to create more nucleation sites in the ZnO lattice and this behavior prohibit the growth of grains. Prajapati et al. have reported that the cristallinity of the sprayed AZO thin films increases with the increasing aluminum concentration up to 2 at% and on further increasing of the doping level, the crystallinity has observed to deteriorate [62]. Whereas, some other researchers have found the reduced cristallinity of the spray pyrolysis deposited Al-doped ZnO thin films observed after increasing the aluminum doping amount up to 3.0 at% [42,63]. The (002) orientation had the lowest surface energy density (SED) in the ZnO crystal. Thus, when the films grew with the (002) orientation, the grains became larger and growth orientation progressed in a crystallographic direction with the lowest SED. Meanwhile, the Al doping level affected the SED and growth process [7,64].

The lattice constants for the hexagonal structure of the ZnO films, 'a' and 'c', were calculated by the following equation [65]:

$$\frac{1}{d^2(hkl)} = \frac{4}{3}\left(\frac{h^2 + hk + k^2}{a^2}\right) + \frac{l^2}{c^2} \quad (1)$$

In which $d_{(hkl)}$ is the spacing between lattice planes of the given miller indices h, k and l.

For calculation of the crystallite size (D), the Scherrer's formula was used [66]:

$$D = \frac{C\lambda}{\beta cos\theta} \quad (2)$$

Where C is constant and nearly equal to 0.9, λ is the X-ray wavelength ($k_{\alpha 1,2}$ of copper = 1.5406 Å), θ is the Bragg's angle and β is the full width at half maximum of (002) diffraction peak. The obtained lattice constants were in well agreement with the standard value from the JCPDS card no. 036–1451 (Table 1). The following table presents the calculated crystallite sizes and lattice constants of the different samples.

In order to investigate the effect of the doping level on the films morphology, Field emission scanning electron microscopy analysis was used (Fig. 3). All samples deposited at 550 °C were quite uniform with no crack. Observation showed that surface morphology was strongly affected by the doping level. With increasing doping concentration, the size of the particles decreased from 42.3 nm to nearly 35.2 nm