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Effects of low-temperature annealing on electrical properties of Thin-film Transistors based on Zinc Oxide films deposited by ultrasonic spray pyrolysis: Impact of annealing time

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ABSTRACT

In this work, the study of annealing effects on electrical properties of Zinc Oxide Thin-film Transistors is presented. The samples were annealed at 180 °C under Nitrogen ambient. The conductivity and contact resistance of ZnO films with Aluminum electrodes are studied by Transmission Line Method. Also, the Zinc Oxide films obtained by ultrasonic spray pyrolysis at 200 °C are studied by X-ray diffraction and Fourier transform infrared spectroscopy. A comparison of the electrical properties as a function of annealing time is presented. The results show an optimal annealing time and after this time, the metal-ZnO interface deteriorates.

1. Introduction

Currently, Zinc Oxide films (ZnO) have a great application in potential low-cost devices, such as solar cells and Thin-film Transistors (TFTs) [1–6]. In these devices are desirable to have a low contact resistance for an optimum electrical performance. In an ideal metal-semiconductor contact, there are no barriers for the carrier flow in either positive or negative polarization. Ideally, this occurs when metal and semiconductor work functions are of similar value and there are no interface states. However, usually, having a metal-ZnO contact without interface states is difficult. Also, matching the semiconductor and metal work functions is nearly impossible [7]. It is important to say that contact metallization technology in ZnO has not been explored extensively [7]. Some authors have attempted to reduce the contact resistance in metal-ZnO interfaces [8–16]. Moreover, different thermal treatments have been reported for ZnO alloys and TFTs [17–31]. However, it is well-known, that ZnO films are highly dependent on the deposition and post-treatments conditions, resulting in different surface conditions and defect density distribution. For this reason, different results have been reported. In [8], contacts annealed at 300 °C resulted in a reduction by two orders of magnitude in contact resistance; however, annealing at 500 °C resulted in a reduction of one order of magnitude only. Also, in other works, have been reported that metal-ZnO contact deteriorates after annealing at higher temperature, where the contact resistance is increased [9,16]. Contrary to these results, in [15] reported Ru-ZnO contacts annealed at 700 °C without any electrical degradation. Therefore, a quantitative comparison of the contact resistance would be unfeasible, because it depends on the quality of the ZnO film and metal contact used.

On the other hand, although the improvement of electrical characteristics of ZnO TFTs by post-deposition thermal annealing has already been demonstrated, the temperature of post-deposition treatments to improve performance is too high [23–31]. Since the aim of Oxide TFTs development is to be an alternative to the typical a-Si TFTs, which are fabricated at temperatures close to 200 °C [32–35], then it is necessary to keep the complete fabrication temperature at low values, less than 200 °C, in order to be a real alternative to a-Si for large-area and flexible applications [35].

In this work, the study of annealing effects on electrical properties of ZnO Thin-film Transistors is presented. The ZnO films were deposited by ultrasonic spray pyrolysis at 200 °C. The samples were annealed at 180 °C under Nitrogen ambient. The conductivity and contact resistance of the ZnO films with Aluminum (Al) electrodes are studied by Transmission Line Method (TLM), a comparison of the electrical properties as a function of annealing time is presented. Also, the ZnO films are studied by X-ray diffraction (XRD) and Fourier transform infrared spectroscopy (FTIR). It is important to note, as far as the authors know, low-temperature annealing effects on ZnO deposited by ultrasonic spray pyrolysis have not been published yet.

2. Experiment

The ZnO thin films were deposited using a home-made ultrasonic spray pyrolysis deposition system adapted from an ultrasonic

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humidifier (Heaven Fresh), from 0.2 M precursor solution of Zinc acetate in methanol, using air as carrier gas at flow rate of 467 sccm. The substrate was placed on a hot-plate at 200 °C during deposition. The nozzle is located at 8 cm-height to cover a substrate area of 5 cm × 5 cm. The post-deposition annealing was conducted in a furnace with Nitrogen atmosphere at 180 °C for different annealing times: 15, 30, 45 and 60 min.

The Al-ZnO contacts were characterized by TLM, which consists in current–voltage measurements at different contact separations [36]. The contact separation was 5, 10, 25, 30 and 35 μm, and the contact width was 120 μm. The resistance measured between the contacts is described by:

$$R_T = \rho sd/Z + 2R_C \quad (1)$$

where ρs is the resistivity, d is the contact separation, Z is the contact width and R_C is the contact resistance. To calculate the contact resistance, the interception at d = 0 in the plot of resistance as a function of d gives the value of $2R_C$.

The orientation of the film was investigated using X-ray diffractometer (XRD) (Discover D8-Bruker axs) at 2θ range between 10° and 90° and 0.002° step. The IR absorption spectra was measured with a "BRUCKER" FTIR spectrometer, Model Vertex-70.

The inverted coplanar ZnO TFTs (bottom-contact bottom-gate) were made of 35 nm-thick ZnO deposited over 100 nm-thick Aluminum electrodes patterned on 50 nm-thick thermally grown SiO_2 on heavily-doped Si wafers [4,5]. Better electrical performance in TFTs using thinner active layer has been reported, for this reason, active layer thickness of 35 nm was used [37,38]. The thickness was measured by ellipsometry and confirmed by measuring an etched step. The electrical characteristics were measured using the Keithley-4200 Semiconductor Characterization System at room temperature and under dark conditions.

3. Results and discussion

Fig. 1 shows the contact resistance and conductivity of Al-ZnO contacts annealed at 180 °C as a function of annealing time. The figure shows a reduction of two orders of magnitude in contact resistance for samples annealed for 30 min. This improvement is related to a higher carrier injection through the Al-ZnO interface. P. Nunes et al. [39–41] found that this is due to desorption of oxygen present at the grain boundaries of the ZnO film. This results in an increase of the effective carrier concentration by the oxygen loss near to the Al-ZnO interface [7,10–12]. The improvement of two orders of magnitude in contact resistance is similar than that previously reported by other authors [8, 13–16]. In [8], the annealing at 300 °C for 1 min under Nitrogen ambient of as-deposited contacts resulted in a reduction of two orders of magnitude in contact resistance; however, the annealing at 500 °C resulted in a reduction of one order of magnitude only.

On the other hand, to improve metal-ZnO contacts, a specific annealing time is used without any explanation [8,9,15,16,18–21,25–29]. Only few authors reported the use of different annealing times [30,31]. For this reason, in order to study the impact of annealing time, a longer annealing time is explored. Longer annealing than 30 min results in an increase of the contact resistance, as shown in Fig. 1. This is due to a change of the quality in the ZnO film, as can be interpreted with the reduction of conductivity. These results show that there is an optimal annealing time and after this time, the metal-ZnO interface deteriorates. In order to address this assumption, X-ray diffraction and FTIR spectroscopy were used to study the effects of annealing time on ZnO films.

Fig. 2 shows the FTIR spectra of ZnO films at different annealing times. It can be seen the peak at 415 cm^{-1} related to Zn—O stretching modes [42,43]. The characteristic Zn—O band indicates the ZnO formation. The figure clearly shows a reduction in Zn—O bonds as the annealing time is increased. This can be correlated with the results of conductivity and contact resistance, where the oxygen vacancies increase the carrier concentration. However, at 60 min of annealing, the reduction in Zn—O bonds does not completely explain the increase in contact resistance, although, a change in the structural properties can explain it. Fig. 3 shows the XRD patterns of as-deposited and annealed ZnO films. In the as-deposited films, the peaks related to the planes (100), (002), (101) and (110) can be identified, where the strongest peak is associated to the (100) plane. At 30 min of annealing, the peaks related to the planes (100), (002), (101), (102), (103), (112) and (110) can be identified, where the strongest peak is now associated to the (002) plane. The XRD diffractogram confirms a better polycrystalline nature of ZnO films for 30 min of annealing. The preferential orientation in (002) plane (in agreement with the JCPDS Card No. 36-1451) has been reported in high-performance ZnO films [42,44,45]. On the other hand, at 60 min of annealing, the preferential orientation in (002) plane is lost, moreover, the peaks related to the planes (102) and (103) are disappeared, indicating the loss of the polycrystalline nature of the film. In addition, a reduction in the peaks related to the planes (100) and (101) is appreciated for ZnO films annealed for 60 min compared to as-deposited films. These results confirm the degradation of the 60 min annealed ZnO film and are in agreement with the FTIR spectroscopy, conductivity and contact resistance results.

In order to demonstrate the above conclusions in the improvement of electrical characteristics of ZnO TFTs with Al electrodes, Fig. 4 shows

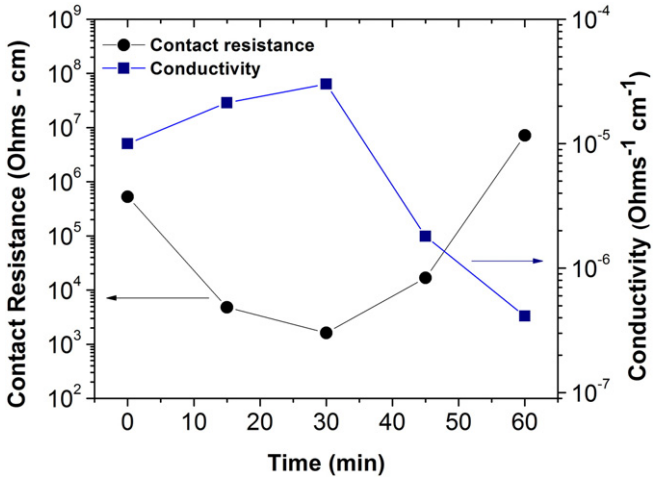

Fig. 1. Al-ZnO contact resistance and conductivity extracted by TLM as a function of annealing time.

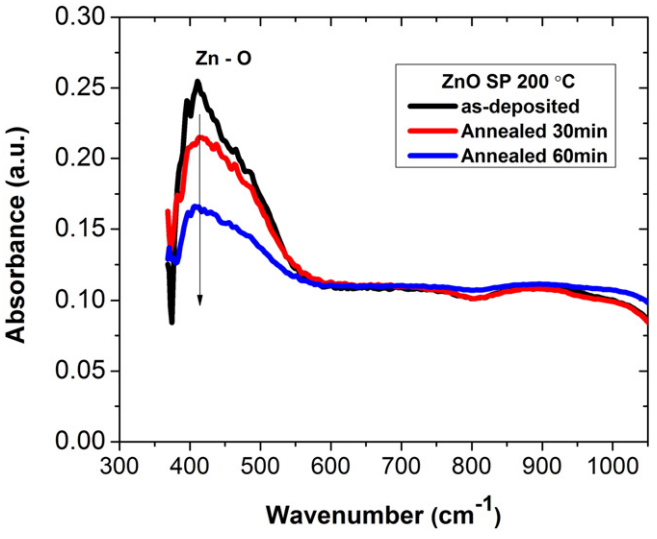

Fig. 2. FTIR spectra of as-deposited and annealed ZnO films.