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Low-temperature ultrasonic spray deposited aluminum doped zinc oxide film and its application in flexible Metal-Insulator-Semiconductor diodes



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ABSTRACT

In this work, the fabrication and characterization of fully solution-processed flexible Metal-Insulator-Semiconductor (MIS) diodes are presented. The MIS structure was fabricated using aluminum doped zinc oxide and spin-on glass as semiconductor and insulator, respectively. The maximum temperature used was 200 °C. The electrical characteristics of the flexible devices show a good agreement with the typical characteristics of a semiconductor diode even while bent to 5 mm tensile radius.

1. Introduction

Currently, several techniques allow the deposition of thin films at room temperature; however, some present technical problems such as low compatibility with large-area substrates, high vacuum need and high-cost. On the other hand, deposition of thin films using solution processes offers low-cost, simplicity, compatibility with large-area substrates and no need of high vacuum [1-4]. Ultrasonic spray pyrolysis allows the deposition of denser films compared with conventional spin-coating technique. Despite on this, usually, the use of high deposition temperature is still required to obtain high performance devices [5-7]. Deposition or annealing temperatures higher than 200 °C are not compatible with most of the low-cost plastic substrates used in the emerging technology of flexible electronics. Therefore, the device fabrication at low temperature is one of the most demanding research issues in terms of device performance in this technology [8,9].

Semiconductor diodes are extensively used in electronics as rectifiers, mixing, isolating and detection of signals. Recently, Son et al. [10], reported Schottky diodes using solution-processed zinc tin oxide on corning glass. The Metal-Insulator-Semiconductor (MIS) diodes, also called MIS Tunnel diodes, use a very thin insulator film to allow the tunneling of carriers in one bias condition [11-15]. In [16], it was reported the application of MIS diodes fabricated on Silicon substrates as temperature sensors.

In this work, the fabrication and characterization of fully solutionprocessed flexible MIS diodes is presented. The MIS structure was fabricated using aluminum doped zinc oxide (AZO) and spin-on glass (SOG) as semiconductor and insulator, respectively. The AZO thin film was deposited by ultrasonic spray pyrolysis. The maximum temperature

used was 200 °C.

2. Experiment

The AZO precursor solution was prepared from 0.2 M of zinc nitrate (Sigma-Aldrich) dissolved in distilled water. The aluminum doping of the film was facilitated by adding aluminum nitrate (Sigma-Aldrich) to the precursor solution at different molar concentrations respect to zinc nitrate (1, 3, 5 and 10 wt%). The flexible MIS diodes were fabricated on indium tin oxide (ITO) coated polyethylene terephthalate (PET) substrates (Sigma-Aldrich). First, the SOG solution (SOG700B from Filmtronics) was diluted 2:1 with deionized water (DI). Then, the SOG/ DI solution was spin-coated at 5000 RPM for 30 s and cured at 200 °C for 1 h. After that, the AZO precursor solution was ultrasonically sprayed to the samples using air as carrier gas at a 0.5 ml/min flow rate. The AZO films were deposited at 200 °C using a home-made ultrasonic spray pyrolysis deposition system adapted from an ultrasonic nebulizer (CITIZEN CUN-60 @2.5 MHz). The nozzle is 10 cm height from the substrate. Finally, as top electrodes, Silver ink (AgIC Inks) was patterned. The contact area was 0.012 cm². Fig. 1 shows the fabricated structure. The average thickness of the SOG/DI and AZO films were 15 nm and 70 nm, respectively.

The optical transmittance was measured using a Thermo Scientific evolution 600 equipment. The resistivity of the AZO films above corning glass 2947 was measured by four-point probe. The orientation of the as-deposited films was obtained using a X-ray diffractometer (XRD) (Discover D8-Bruker axs).

The electrical characteristics of the flexible MIS diodes were measured using the Keithley-4200 Semiconductor Characterization System

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Fig. 1. Schematic structure of the flexible MIS diodes.

at room temperature and under dark conditions.

3. Results and discussion





Fig. 2. Optical transmittance of the solution-processed thin films. a) SOG/DI film and b) AZO films at different aluminum doping concentration.



Fig. 3. XRD patterns of the AZO films at different aluminum doping concentrations.

films. Fig. 2a shows the transmittance of the SOG/DI films. The films are highly transparent in the visible range, which makes feasible their use in transparent electronics. In previous work [17], it was demonstrated that dilution of SOG is necessary to obtain good quality films at 200 °C, where the dilution makes easier the evaporation of the organic material present in the SOG solution. The refractive index of the film was calculated close to 1.45 along with a dielectric constant very close to that of thermally grown silicon oxide. These make the SOG/DI film feasible to be used as insulator in flexible electronic devices. On the other hand, Fig. 2b shows the optical transmittance of the AZO films at different Aluminum doping concentration. The films are also highly transparent in the visible range. The increase in doping concentration has no considerable effect in the optical transmittance. This agrees with that reported by [4].

Fig. 3 shows the XRD pattern of the AZO films at different Aluminum doping concentrations. The AZO films exhibit an amorphous structure regardless of the aluminum doping concentration. Also, the undoped zinc oxide film does not reveal a polycrystalline structure, therefore the amorphous structure cannot be attributed to the aluminum doping. This can be expected due to the low temperature of deposition [6].

Fig. 4 shows the resistivity of the AZO films at different doping concentrations. As the doping concentration increases, the resistivity decreases to find the lowest value at 3 wt%. This decrease is attributed



Fig. 4. Resistivity of the AZO films at different aluminum doping concentrations.

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