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Impact of active layer thickness in thin-film transistors based on Zinc Oxide by ultrasonic spray pyrolysis

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ABSTRACT

In this work, the preparation of Zinc Oxide (ZnO) films by ultrasonic spray pyrolysis at low-temperature and its application in thin-film transistors (TFTs) are presented, as well, the impact of the active layer thickness and gate dielectric thickness in the electrical performance of the ZnO TFTs. A thinner active layer resulted in better transfer characteristics such as higher on/off-current ratio, while a thicker active layer resulted in better output characteristics. The ZnO films were deposited from 0.2 M precursor solution of Zinc acetate in methanol, using air as carrier gas on a hotplate at 200 °C. The ZnO films obtained at 200 °C were characterized by optical transmittance, Photoluminescence spectroscopy and X-ray diffraction.

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1. Introduction

Currently, Zinc Oxide (ZnO) films are attractive to be an alternative to amorphous silicon in thin-film transistors (TFTs) commercial applications [\[1\]](#page--1-0). ZnO films can be obtained by several techniques such as pulsed laser deposition, sputtering, and chemical vapor deposition $[2-5]$. Although some of these techniques allow the deposition of oxide films at room temperature, they present some technical problems such as low compatibility with large-area substrates and high-cost. On the other hand, spray pyrolysis offers low-cost, simplicity, compatibility with large-area substrates and no need of high vacuum [\[6,7\]](#page--1-0). These advantages make of great potential the use of oxide semiconductors by spray pyrolysis as active layer in TFTs for transparent, flexible and large-area applications. Although, oxide TFTs fabricated by spray pyrolysis have already been demonstrated $[8-12]$, the temperature of deposition to obtain high performance devices is still high to be compatible with most of the low-cost plastic substrates used in the aforementioned technologies. Then, it is necessary reduce the temperature of deposition at values about 200 °C or less in order to be a real alternative for large-area and flexible applications [\[1,13–15\].](#page--1-0)

There are different ways to spray the solution onto the substrates, Ortel et al. [\[8\]](#page--1-0) sprayed the solution with a perfume

⇑ Corresponding author. Tel./fax: +52 (222) 229 55 00x7876. E-mail address: madominguezj@gmail.com (M.A. Dominguez). atomizer, Adamopoulos et al. [\[9\]](#page--1-0) sprayed the solution with an airbrush. While in this work, we used an ultrasonic humidifier and air as carrier gas to spray the solution onto the samples. The application of ZnO films obtained by ultrasonic spray pyrolysis at lowtemperature (200 °C) as active layer in TFTs is presented, as well, the impact of the gate dielectric thickness and active layer thickness in the electrical performance of the ZnO TFTs.

2. Experiment

The ZnO films were deposited using a typical home-made ultrasonic spray pyrolysis deposition system adapted from an ultrasonic 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 on a hotplate at 200 \degree C. The orientation of the as-deposited films were obtained using a X-ray diffractometer (XRD) (Discover D8-Bruker axs) at 2 θ range between 20 $^{\circ}$ and 80 $^{\circ}$ and 0.002 $^{\circ}$ step. The optical transmittance was measured from 200 nm to 900 nm. For the Photoluminescence spectroscopy, it was used as exciting source a laser of He–Cd with 325 nm line. The photoluminescence measurement was performed using a silicon PIN Thorlabs (DET-210) detector with a spectral response of 200–1100 nm. It was employed the conventional lock-in technique seeking to minimize the effect of noise. The voltage signal is acquired by a data acquisition card brand Computer Boards CIO-DAS08 model, which operates in conjunction with a stepper motor controller to position the monochromator Sciencetech 9040 to the appropriate wavelength. The data are analyzed and plotted using software designed in lab-view. The 110 nm-thick ZnO samples for material characterization were prepared in corning glass 2947 by ultrasonic spray pyrolysis as mentioned above.

For the fabrication of the ZnO TFTs, inverted coplanar structure (bottom-contact bottom-gate) was used. ZnO active layer was deposited over 100 nm-thick aluminum electrodes (e-gun evaporated) patterned on thermally grown $SiO₂$ on highly doped Si wafers. The highly doped Si wafer was used as the gate electrode. Different sets of devices were fabricated, where 35 nm and 110 nm-thick ZnO active layer were used to compare the effects of the active layer thickness and 50 nm and 100 nm-thick thermally grown $SiO₂$ were used to compare the effects of the gate dielectric thickness. After the solution was sprayed onto the samples, these were kept at 200 °C on the hotplate for 3–5 min to allow the evaporation of the solvents. The electrical characteristics were measured using the Keithley-4200 Semiconductor Characterization System, under dark conditions, air ambient and room temperature. Commonly, in solution-processed TFTs fabricated at low-temperature there is present a moderate hysteresis, as in our case. The transfer and output characteristics were measured both forward and reverse biased. The average of the curves were obtained to extract the device parameters. The average values extracted were affected approximately ±15% of its value by the hysteresis. It is important to mention that the electrical performance of the devices is similar at different zones of the wafer. The electrical characteristics here reported are representative of 10 measured devices.

3. Results and discussion

Fig. 1 shows the XRD pattern of the as-deposited ZnO film at 200 °C. The ZnO film shows three weak peaks at $2\theta = 31.72^{\circ}$, 34.42 $^{\circ}$ and 56.64 $^{\circ}$ which are associated to the (100), (002) and (11 0) planes. From the overall XRD diffractogram the ZnO film tends to present some polycrystallinity and are in agreement with the JCPDS Card No. 36-1451. Further material characterization is needed to address this subject.

Fig. 2 shows the optical transmittance for ZnO films in the wavelength range of 200–900 nm. The film is highly transparent in the visible range. The optical gap energy was estimated by extrapolation of the linear region of the $(\alpha h v)^2$ \sim A($h v$ – Eg), where α is the absorption coefficient, hv is the photon energy, A is a constant and Eg is the optical gap energy. The value of 3.26 eV was obtained which is slightly lower than the typical reported [\[6\].](#page--1-0)

Fig. 1. XRD patterns of the as-deposited ZnO films by ultrasonic spray pyrolysis at 200 °C.

Fig. 2. Transmittance of the as-deposited ZnO films by ultrasonic spray pyrolysis at $200 °C$.

Fig. 3 shows the room temperature photoluminescence (PL) spectra of the ZnO film. It shows a normal PL spectrum with a high intensity peak centered at 390 nm and a broad band from 450 to 700 nm. The UV emission peak at 390 nm is associated to the near band-edge (NBE) emission of the wide gap attributed to the recombination of the free excitons [\[16–19\]](#page--1-0). The visible emission broad band from 450 to 700 nm includes the impurities and defects, which are not negligible in our case. The origin of this visible emission band can be possible due to oxygen vacancies, zinc vacancies, oxygen antisites, zinc antisites, oxygen interstitial and zinc interstitial [\[17–24\].](#page--1-0) Many authors have used photoluminescence spectroscopy to study the role of the impurities and defects in ZnO. However, there have been contradictory results reported even with the same experimental conditions $[24]$. Therefore, the role of the impurities and defects distribution in electronic properties of ZnO is still controversial, since they are highly dependent of the deposition technique and its conditions.

To demonstrate the application of the ZnO films obtained at 200 \degree C, inverted coplanar ZnO TFTs were fabricated as was indicated in the experimental section and [Fig. 4](#page--1-0). [Fig. 5](#page--1-0) shows the average of the forward and reverse measured transfer characteristics of the ZnO TFTs with 35 nm-thick active layer and gate dielectric of 50 nm and 100 nm-thick. For ZnO TFTs with 100 nm-thick gate dielectric, it can be observed an on/off-current ratio close to $10²$. While for ZnO TFTs with 50 nm-thick gate dielectric, the observed

Fig. 3. Photoluminescence of the as-deposited ZnO films by ultrasonic spray pyrolysis at 200 °C.

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