



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]. 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]. 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].

There are different ways to spray the solution onto the substrates, Ortel et al. [8] sprayed the solution with a perfume

atomizer, Adamopoulos et al. [9] sprayed the solution with an air-brush. 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 low-temperature (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 °C. The orientation of the as-deposited films were obtained using a X-ray diffractometer (XRD) (Discover D8-Bruker axs) at 2θ range between 20° and 80° and 0.002° 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

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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 $\pm 15\%$ 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° and 56.64° which are associated to the (100), (002) and (110) 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 $(\alpha h\nu)^2 \sim A(h\nu - E_g)$, where α is the absorption coefficient, $h\nu$ is the photon energy, A is a constant and E_g is the optical gap energy. The value of 3.26 eV was obtained which is slightly lower than the typical reported [6].

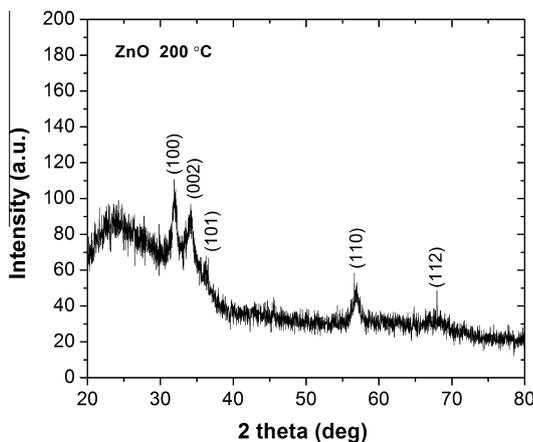


Fig. 1. XRD patterns of the as-deposited ZnO films by ultrasonic spray pyrolysis at 200 °C.

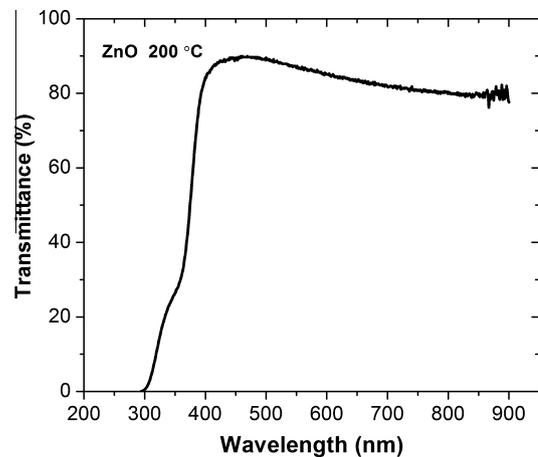


Fig. 2. Transmittance of the as-deposited ZnO films by ultrasonic spray pyrolysis at 200 °C.

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]. 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]. 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 °C, inverted coplanar ZnO TFTs were fabricated as was indicated in the experimental section and Fig. 4. Fig. 5 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 10^2 . While for ZnO TFTs with 50 nm-thick gate dielectric, the observed

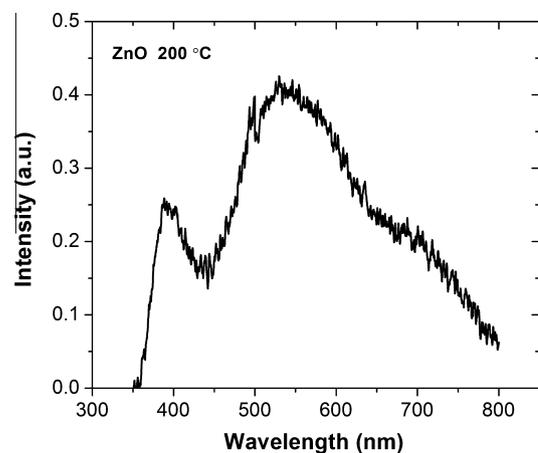


Fig. 3. Photoluminescence of the as-deposited ZnO films by ultrasonic spray pyrolysis at 200 °C.

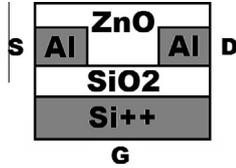


Fig. 4. Cross section of the fabricated bottom-gate ZnO TFT (not to scale).

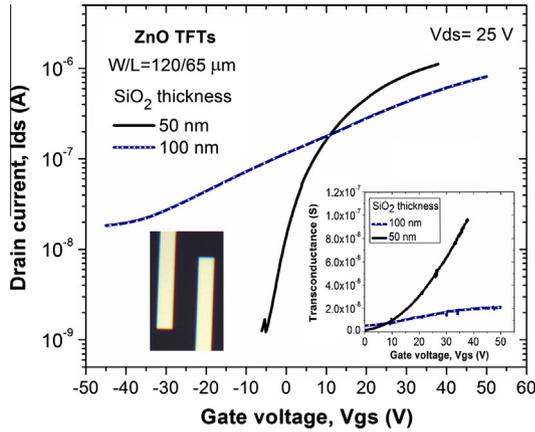


Fig. 5. Transfer characteristics of the low-temperature ZnO thin-film transistors with different gate dielectric thickness. Inset: top-view of the fabricated ZnO TFTs and comparison of the transconductance of the ZnO thin-film transistors.

on/off-current ratio is 10^3 which is similar to the values reported for solution-processed ZnO TFTs at 200°C [9,25–28].

On the other hand, the electron mobility and threshold voltage V_T were extracted from the square root of I_{ds} vs. V_{gs} in the saturation regime ($V_{ds} = 25\text{ V}$) [29], where the values obtained were $0.01\text{ cm}^2/\text{Vs}$ and 2.8 V , and $0.002\text{ cm}^2/\text{Vs}$ and -20 V for ZnO TFTs with 50 nm and 100 nm -thick gate dielectric, respectively.

The obtained results for ZnO TFTs with 50 nm -thick gate dielectric are better than those reported by Adamopoulos et al. [9]. They reported carrier mobilities from 0.003 to $0.001\text{ cm}^2/\text{Vs}$ and on/off-current ratios from 10^2 to 10^1 at deposition temperature of 200°C . Although Bashir et al. [12] reported carrier mobilities from 0.13 to $0.11\text{ cm}^2/\text{Vs}$ and on/off-current ratios from 10^5 to 10^4 , they obtained a much higher threshold voltage (from 16 to 13.8 V), and also, they used different S/D electrodes and top-contact structure.

The improvement in the electrical performance of the ZnO TFTs with 50 nm -thick gate dielectric may be due to the increase in the transconductance of the devices, since the transconductance g_m is proportional to the oxide capacitance C_{ox} of the device as shows Eq. (1). However, at the same time, the gate leakage current increases as the thickness of the gate dielectric is decreased and may affect the electrical performance of the device. From the transfer characteristics, there is no appreciation of high leakage current effects. Inset in Fig. 5 shows a comparison of the transconductance of both ZnO TFTs.

$$g_m = (\mu C_{ox} W/L)(V_{gs} - V_T) \quad (1)$$

where μ is the field-effect mobility, C_{ox} is the capacitance per unit area of the gate dielectric, W and L are the channel width and the length, respectively, and V_T is the threshold voltage. To compare the effects of the active layer thickness, ZnO TFTs with 110 nm -thick active layer and gate dielectric of 50 nm -thick were fabricated. The same solution and procedure depicted in the experimental section were used. Fig. 6 shows the average of the forward and reverse measured transfer characteristics of the ZnO TFTs with 35 nm and

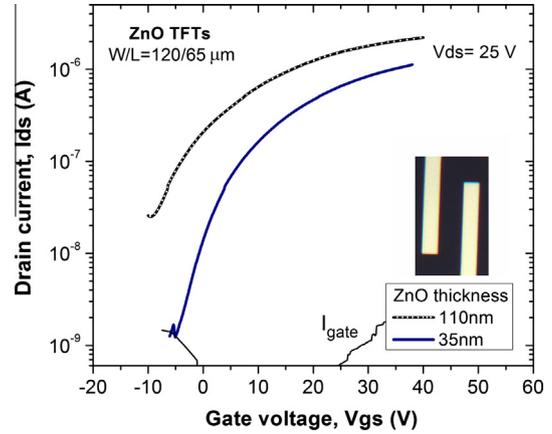


Fig. 6. Transfer characteristics of the low-temperature ZnO thin-film transistors with different active layer thickness.

110 nm -thick active layer and gate dielectric of 50 nm -thick. For ZnO TFTs with 110 nm -thick active layer, it can be observed an on/off-current ratio of 10^2 . While for ZnO TFTs with 35 nm -thick active layer, the observed on/off-current ratio is 10^3 . The electron mobility and threshold voltage V_T were $0.01\text{ cm}^2/\text{Vs}$ and 2.8 V , and $0.015\text{ cm}^2/\text{Vs}$ and -1.5 V for ZnO TFTs with 35 nm and 110 nm -thick active layer, respectively. As expected, the off-current and on-current are higher for ZnO TFTs with 110 nm -thick active layer. Typically, the effects of the active layer thickness are attributed to the trap states density and active layer conductance [30–32]. Our results are similar to those reported by Nomura et al. [30], where the on/off-current ratio is improved with thinner active layer.

On the other hand, the average of the forward and reverse measured output characteristics are shown in Fig. 7. It can be seen effects of current crowding at low values of V_{ds} , due to a high contact resistance resulted by an Al_2O_3 layer formed in the metal–semiconductor interface during the fabrication of the devices. This high contact resistance reduces the on/off-current ratio and masks the real value of the electron mobility. Also, the output characteristics show a decrease in the drain current at high drain voltage, this may be related to the hysteresis presented in the device. However, it can be noted that the output characteristics of the ZnO TFTs with 110 nm -thick active layer exhibit a better performance than the ZnO TFTs with 35 nm -thick active layer, such as, lower contact resistance effects at low drain voltage and less drain current drop at high drain voltage. Contrary to these results, Oh et al. [32] reported that thicker active layer increases the off-current resulting in poor output characteristics. Our results can be explained by the thickness of the ZnO active layer. For the case of the 110 nm -thick ZnO TFT, since in the metal–semiconductor interface the active layer is thicker than the aluminum electrodes, covering well the bottom-contacts and increasing the area of contact resulting in better quality contact. While in the case of the 35 nm -thick ZnO TFT, the active layer is very thin compared with the 100 nm -thick aluminum electrodes, thus, the area of contact is much smaller resulting in a poor quality contact. These agree with the small difference in the values of electron mobility extracted from the aforementioned ZnO TFTs.

It is difficult to determine the optimum active layer thickness for these inverted coplanar ZnO TFTs, because 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 dependency of the electrical performance on the structure of the TFTs has been widely studied in a-Si TFTs and can be useful for the improvement of Oxide TFTs [29,33].

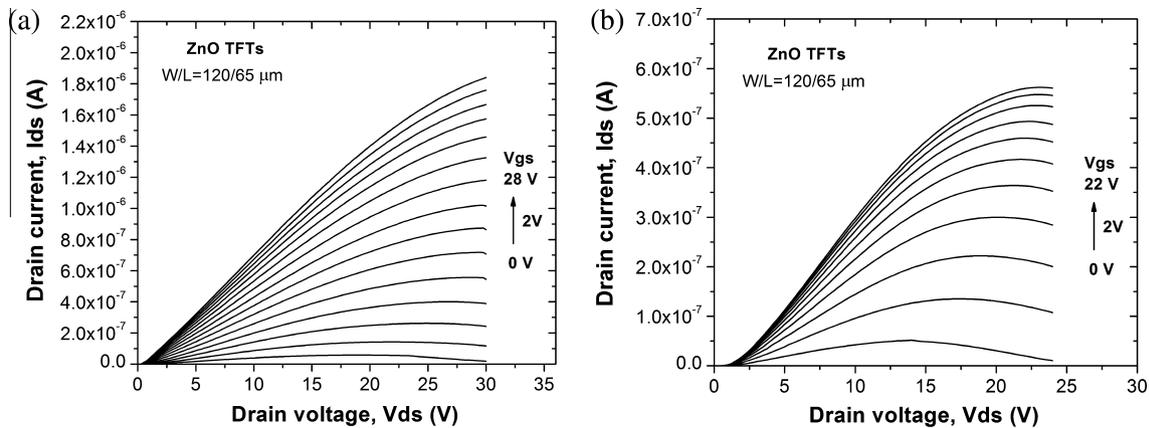


Fig. 7. Output characteristics of the low-temperature ZnO thin-film transistors (a) 110 nm-thick active layer and (b) 35 nm-thick active layer.

4. Conclusions

The characterization of ZnO films by ultrasonic spray pyrolysis at 200 °C and its application in inverted coplanar ZnO TFTs are demonstrated. The improvement in the electrical performance of the ZnO TFTs with thinner gate dielectric is due to the increase of the transconductance of the device. There is no observation of high leakage current effects by the thinner gate dielectric in the transfer characteristics. Thicker active layer results in better output characteristics (I_{ds} vs. V_{ds}), while thinner active layer results in better on/off-current ratio. The improvement in the output characteristics is associated to a larger area of contact with the thicker active layer, this agrees with the relatively higher electron mobility extracted in the 110 nm-thick ZnO TFTs.

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References

- [1] Street R. Thin-film transistors. *Adv Mater* 2009;21:2007.
- [2] Fay S, Kroll U, Bucher C, Vallat-Sauvain E, Shah A. Low pressure chemical vapour deposition of ZnO layers for thin-film solar cells: temperature-induced morphological changes. *Sol Energy Mater Sol Cells* 2005;86:385.
- [3] Mohamed SH, Drese R. Structural and optical properties of direct current sputtered zinc aluminum oxides with a high Al concentration. *Thin Solid Films* 2006;513:64.
- [4] Bouhssira N, Abed S, Tomasella E, Cellier J, Mosbah A, Aida M, et al. Influence of annealing temperature on the properties of ZnO thin films deposited by thermal evaporation. *Appl Surf Sci* 2006;252:5594.
- [5] Nishii J, Hossain FM, Takagi A, Aita T, Saikusa K, Ohmaki Y, et al. High mobility thin film transistors with transparent ZnO channels. *Jpn J Appl Phys* 2003;42:L347.
- [6] Olvera M, Gomez H, Maldonado A. Doping, vacuum annealing, and thickness effect on the physical properties of zinc oxide films deposited by spray pyrolysis. *Sol Energy Mater Sol Cells* 2007;91:1449.
- [7] Nunes P, Fernandes B, Fortunato E, Vilarinho P, Martins R. Performances presented by zinc oxide thin films deposited by spray pyrolysis. *Thin Solid Films* 1999;337:176.
- [8] Ortel M, Trostyanskaya Y, Wagner V. Spray pyrolysis of ZnO-TFTs utilizing a perfume atomizer. *Solid State Electron* 2013;86:22.
- [9] Adamopoulos G, Bashir A, Gillin W, Georgakopoulos S, Shkunov M, Baklar M, et al. Structural and electrical characterization of ZnO films grown by spray pyrolysis and their application in thin-film transistors. *Adv Funct Mater* 2011;21:525.
- [10] Oertel S, Jank M, Teuber E, Bauer A, Frey L. High-mobility metal-oxide thin-film transistors by spray deposition of environmentally friendly precursors. *Thin Solid Films* 2014;553:114.
- [11] Ming Y, Ling X, Yu L, Yan D, Jing H. Electrical characteristics of high-performance ZnO field-effect transistors prepared by ultrasonic spray pyrolysis technique. *Chin Phys Lett* 2011;28:017302.
- [12] Bashir A, Wobkenberg P, Smith J, Ball J, Adamopoulos G, Bradley D, et al. Structural and electrical characterization of ZnO films grown by spray pyrolysis and their application in thin-film transistors. *Adv Mater* 2009;21:2226.
- [13] Hwang J, Park J, Ki H. UV-assisted rapid thermal annealing for solution-processed zinc oxide thin-film transistors. *Semicond Sci Technol* 2014;29:095019.
- [14] Jeong Y, Pearson C, Lee Y, Winchester L, Hwang J, Kim H, et al. Zinc oxide thin-film transistors fabricated at low temperature by chemical spray pyrolysis. *J Electron Mater* 2014;43:4241.
- [15] Jeong Y, Pearson C, Lee Y, Ahn K, Cho C, Hwang J, et al. Effects of hydrogen plasma treatment on the electrical behavior of solution-processed ZnO transistors. *J Appl Phys* 2014;116:074509.
- [16] Xiao S, Zhao L, Liu Y, Lian J. Nanocrystalline ZnO films prepared by pulsed laser deposition and their abnormal optical properties. *Appl Surf Sci* 2013;283:781.
- [17] Santra S, Guha P, Ali S, Hiralal P, Unalan H, Covington J, et al. ZnO nanowires grown on SOI CMOS substrate for ethanol sensing. *Sens Actuators B Chem* 2010;146:559.
- [18] Padilla D, Vadillo J, Laserna J. Room temperature pulsed laser deposited ZnO thin films as photoluminescence gas sensors. *Appl Surf Sci* 2012;259:806.
- [19] Sepulveda S, Rejea B, De la Rosa E, Ortiz U, Reyes C, Cruz R, et al. Room-temperature deposition of crystalline patterned ZnO films by confined dewetting lithography. *Appl Surf Sci* 2010;256:3386.
- [20] Lin B, Fu Z, Jia Y. Green luminescence center in undoped zinc oxide films deposited on silicon substrates. *Appl Phys Lett* 2001;79:943.
- [21] Janotti A, Van de Walle C. Native point defects in ZnO. *Phys Rev B* 2007;76:165202.
- [22] Bera A, Basak D. Correlation between the microstructure and the origin of the green luminescence in ZnO: a case study on the thin films and nanowires. *Chem Phys Lett* 2009;476:262.
- [23] Tong Y, Liu Y, Shao C, Liu Y, Xu C, Zhang J, et al. Growth and optical properties of faceted hexagonal ZnO nanotubes. *J Phys Chem B* 2006;110:14714.
- [24] Wang D, Seo H, Tin C, Bozack M, Williams J, Park M, et al. Effects of postgrowth annealing treatment on the photoluminescence of zinc oxide nanorods. *J Appl Phys* 2006;99:113509.
- [25] Fortunato E, Barquinha P, Martins R. Oxide semiconductor thin-film transistors: a review of recent advances. *Adv Mater* 2012;24:2945.
- [26] Yang C, Hong K, Jang J, Chung D, An T, Choi W, et al. Solution-processed flexible ZnO transparent thin-film transistors with a polymer gate dielectric fabricated by microwave heating. *Nanotechnology* 2009;20:465201.
- [27] Jun J, Park B, Cho K, Kim S. Flexible TFTs based on solution-processed ZnO nanoparticles. *Nanotechnology* 2009;20:505201.
- [28] Choi S, Park B, Jung H. Luminescence and electrical properties of solution-processed ZnO thin films by adding fluorides and annealing atmosphere. *Mater Res Bull* 2011;46:810.
- [29] Dominguez M, Rosales P, Torres A. Performance improvement of low-temperature a-SiGe: H thin-film transistors. *Solid State Electron* 2012;76:44.
- [30] Nomura K, Kamiya T, Hosono H. Ambipolar oxide thin-film transistor. *Adv Mater* 2011;23:3431.
- [31] Chung J, Lee J, Kim H, Jang N, Kim J. Effect of thickness of ZnO active layer on ZnO-TFT's characteristics. *Thin Solid Films* 2008;516:5597.
- [32] Oh B, Jeong M, Ham M, Myoung J. Effects of the channel thickness on the structural and electrical characteristics of room-temperature fabricated ZnO thin-film transistors. *Semicond Sci Technol* 2007;22:608.
- [33] Dominguez M, Rosales P, Torres A, Flores F, Molina J, Moreno M, et al. Planarized ambipolar a-SiGe: H thin-film transistors: influence of the sequence of fabrication process. *Solid State Electron* 2014;99:45.