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Letter

Flexible a-IZO thin film transistors fabricated by solution processes

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ABSTRACT

A thin film transistor (TFT) device, made using a flexible polyimide (PI) substrate contained twenty-seven top gate TFTs. The transistors contained both organic and inorganic components. The active channel layers used amorphous indium zinc oxide (a-IZO). The gate insulators were of cross-linked poly-4-vinylphenol (PVP). These components were prepared by solution processes and microwave heating under ambient air conditions. Microwave heating achieves the thin film annealing activation energy at lower bulk temperatures than conventional heating. The source, drain, and gate conducting electrodes were of aluminum metal. When operated in depletion mode, the device showed field-effect mobility levels as high as 6.9 $cm^2/V\,s$, a threshold voltage of 2 V, and an on/off current ratio greater than 10^6.

1. Introduction

Lightweight and mechanical flexibility are important to many new optoelectronic applications, such as flexible displays, flexible printed circuits, and flexible chemical sensors [1]. Organic polymers offer the desired mechanical flexibility, however channel mobility tends to diminish the performance levels of organic thin film transistors [2]. The situation demands hybrid transistors, with high mobility active channels and suitable gate insulators on flexible plastic substrates. Poly-4-vinylphenol (PVP) has acceptable dielectric properties when used as an organic gate insulator, making it potentially useful for OTFTs [3] and flexible electronics applications [4]. Existing organic and inorganic semi-conductive materials have been evaluated for use in flexible thin film transistors (TFTs) [5–8]. Various limitations, including sensitivity to light, and relatively low field-effect mobility (<1 $cm^2/V\,s$) have made most of these materials unsuitable for high performance TFT applications. However, there have been recent developments in field-effect transistors (FETs), focusing on the use of oxide semiconductors.

Multi-component amorphous oxide semiconductors (AOSs), such as indium–zinc oxide (IZO), zinc–tin oxide (ZTO), and indium–gallium–zinc oxide (IGZO), are emerging for utilizing use as the active channel layer in TFTs because of their high field-effect mobility, excellent thermal stability, film smoothness and low compressive stress. Among these AOSs, the amorphous indium zinc oxide (a-IZO) is a good candidate for flexible TFTs and flexible electronic circuit applications. A-IZO exhibits a wide optical band gap (3.7 eV), high mobility (<60 $cm^2/V\,s$), and resistivity between 10^{-4} and 10^1 $\Omega\,cm$ [9,10]. Several reports demonstrated that a-IZO films permit controllable conductivity and are appropriate for use as the active layers and source/drain electrodes in transparent TFTs [9–12].

Most a-IZO films and a-IZO TFTs are prepared on glass substrates [9–12] or silicon wafer [13,14] using physical vapor deposition (PVD) techniques such as RF magnetron sputtering and pulsed laser deposition. These require expensive equipment and result in high manufacturing costs. On the other hand, oxide semiconductor films fabricated using solution-based processes could eliminate the need for vacuum, and enable the fabrication of low cost and high performance electronics. A recent attempt to fabricate a-IZO TFTs utilized a chemical solution method which required a high temperature in excess of 400 °C [15–17]; thus that method is not feasible for TFTs producing on plastic substrates. Microwave heating is a useful method for the annealing of materials due to its selective heating resulting from a difference in the dielectric loss factors of the heating objects. Thus, microwave heating is often more convenient than conventional heating methods for the fabrication of a-IZO TFTs on plastic substrates. The method requires a lower bulk temperature than other processes, since materials absorb microwave energy at different rates [18].

The purposes of this study are: (1) to demonstrate a hybrid structure field-effect thin film transistor with high electrical performance; (2) to use solution processes to produce a gate insulator and an active channel on a flexible polyimide (PI) substrate; and (3) to deposit a-IZO film using a sol–gel method and microwave heating under ambient air conditions.

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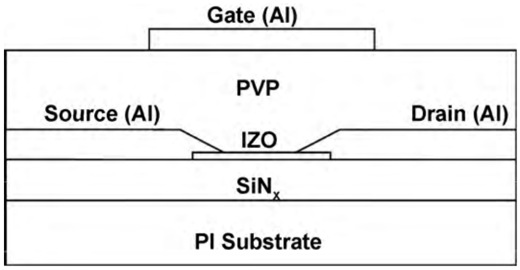

Fig. 1. Schematic cross-sectional view of the a-IZO TFT.

2. Experimental procedures

A transparent PI substrate (125 μm) was passivated by deposition of a 100 nm thickness of silicon nitride (SiN_x) as a barrier against thermal convection and material diffusion. An a-IZO film was made from a sol–gel solution and fabricated on the substrate by spin-coating method. The sol–gel solution contained 60 vol.% of 0.1 M zinc acetate ($Zn(CH_3COO)_2 \cdot 2H_2O$) and 40 vol.% of 0.1 M indium acetate ($In(CH_3COO)_3$) in 2-methoxyethanol; it was stabilized in advance by 0.4 M diethanolamine (DEA) and acetylacetone [16]. The device was baked twice on a hotplate to evaporate solvent and stabilizer, at 150 °C and 230 °C, respectively. To obtain an IZO film with high electron mobility, the organics and residues from the sol–gel film must be fully decomposed. Optimized microwave heating in a Spectra-650W microwave oven (SM-1210) with a 2.45 GHz working frequency decomposed the organics and residues. Then the IZO film was patterned with standard lithography and a wet etching process. Atomic force microscopy (AFM, Digital Instrument NS4/D3100CL) and high-resolution transmission electron microscopy (HRTEM, FEI/Philip Tecnai F20) were used to characterize the morphologies and structures of the prepared films. Cross-sectional views of the a-IZO TFT device were observed using a scanning electron microscope (SEM; Hitachi 4700).

The hybrid-structure a-IZO TFT was fabricated onto PI substrate as shown in Fig. 1. Optimized a-IZO semiconducting film was fabricated; the sources and drains were deposited on this film by sputtering a 70-nm-thick layer of Al; this Al film was patterned by the lift-off technique. Each source–drain pair had a channel width W = 500 μm and channel length L = 10 μm. A 400-nm-thick film of PVP was used for the gate dielectric layer; the film was spin-coated with a solution of poly-4-vinylphenol (4-PVP) and a cross-linking agent (melamine-co-formaldehyde; PMF) in propylene glycol monomethyl ether acetate (PGMEA) [19], cross-linked through UV, and then de-solvated and fully cured in an oven for 30 min at 200 °C. Finally, a 70-nm-thickness of Al was deposited by sputtering and patterned into gate electrodes by a shadow mask. The transistor devices were analyzed with an Agilent 4156C precision semiconductor parameter analyzer at room temperature and were measured in a dark box.

3. Results and discussion

Fig. 2 shows the surface morphology and structure of the IZO film on a PI substrate after microwave heating. The IZO thin film appears very smooth and uniform; it had a low root-mean-square (rms) roughness of 2 nm (Fig. 2(a)). Furthermore, the TEM image shows that the IZO thin film on the PI substrate had a thickness of approximately 15 nm (Fig. 2(b)). The electron diffraction pattern of the IZO thin film showed no any characteristic patterns (bottom right inset of Fig. 2(b)), thus therefore the prepared film was amorphous. Additionally, top inset of Fig. 2(b) shows that only the elements indium, zinc, and oxygen appear in the energy dispersive X-ray spectroscopy (EDX) pattern; this indicates that the residues in the a-IZO film were fully decomposed after the microwave heating. We reasoned that the most important cause of this decomposition was fast internal heating of the a-IZO film due to the dielectric loss factor.

Fig. 3 shows all layers of the a-IZO TFT device on the unbroken PI substrate. The a-IZO films heated by conventional heating methods required large activation energies. Conventional heating methods often result in degrades components during the time-consuming fabrication process. This device assembly remained undamaged during the fabrication process, because the a-IZO film activation energy achieved using microwave energy, occurs at a lower bulk temperature than from conventional heating. The processing temperature and time were significantly lower than would be required for conventional heating methods. Moreover, the SiN_x film between the a-IZO layer and the PI substrate has a low thermal conductance coefficient; therefore, it insulates the heat transfer from the a-IZO film to the PI substrate thus preventing damage. This process can yield semiconductors with precisely defined attributes. The oxidation states and distribution of Zn ions will dominate the electrical characteristics of the a-IZO channel. Due to indiums greater

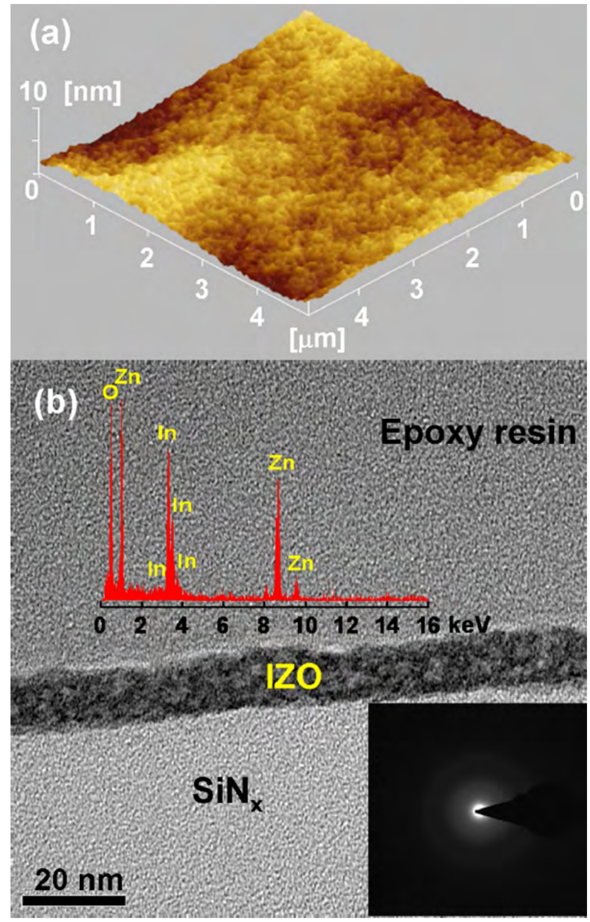

Fig. 2. (a) AFM image of the spin-coated IZO film on the PI substrate. (b) HRTEM cross-sectional image of the IZO film on the PI substrate; the two insets are the EDX pattern (top inset) and the electron diffraction pattern (bottom right inset) of the IZO film.

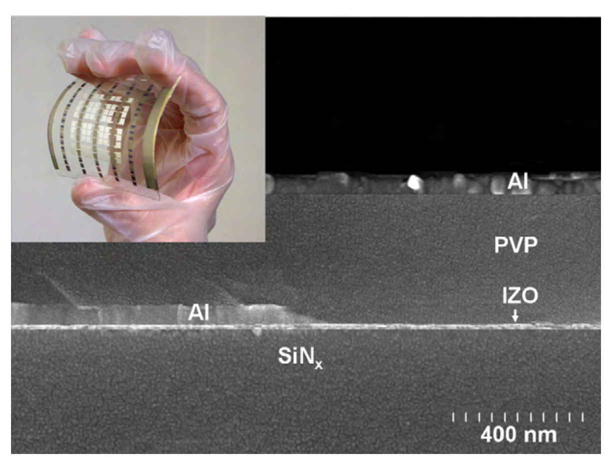

Fig. 3. SEM local cross-sectional image of the a-IZO TFT device on the PI substrate; the inset shows a flexed 5 cm × 5 cm PI substrate with 27 a-IZO TFT devices.

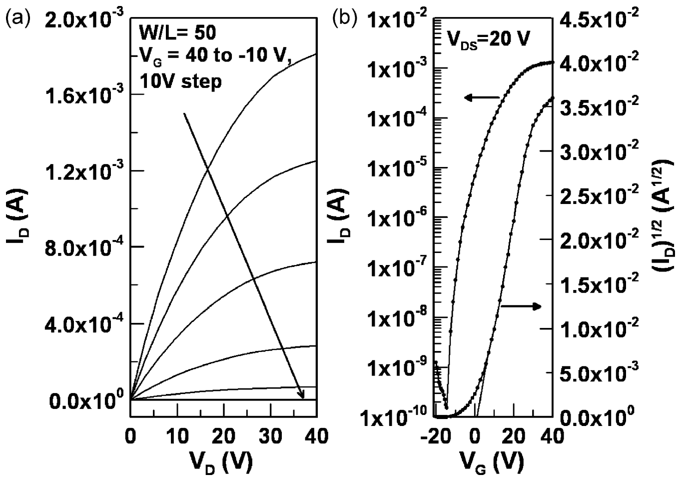

Fig. 4. (a) Drain current–drain voltage (I_D–V_D) curves at gate voltage (V_G) between −10 V and 40 V for an a-IZO TFT with a channel width-to-length ratio (W/L) of 50. (b) Transfer characteristics: I_D versus V_G at V_{DS} = 20 V for the same a-IZO TFT. The second curve traces the square root of drain current versus the gate voltage; this determines the threshold voltage.

ionic radius, zinc ions forms stronger chemical bonds with oxygen than indium ions do [12]. Microwave heating during fabrication enhances electronic mobility in the finished product. This process has a low oxygen diffusion time, and forms more Zn–O bonds than In–O bonds. The preponderance of Zn–O bonds causes formation of oxygen vacancy sites, which, in the finished product, suppresses carrier transmission, and improves electronic mobility. The device's top gate configuration also improves electronic mobility. The completed device has 27 a-IZO TFTs on a single, bendable 5 cm × 5 cm PI substrate (shown as an inset in Fig. 3).

The I_D–V_D curve and I_D–V_G curve of a typical a-IZO TFT are shown in Fig. 4. A positive gate voltage is required to induce a conducting channel in this device, and the current can still be measured at 0 V of gate voltage. From these observations, and from the current–voltage properties measured through the gate, one can infer that the a-IZO channel is an n-type depletion mode device. Fig. 4(a) shows the drain currents (I_D) as functions of source–drain voltage (V_D) for gate voltages (V_G) between −10 V and 40 V. The slope of each I_D curve is flat for large V_D, which means that there are many mobile charge carriers in the a-IZO thin films. Fig. 4(b) shows the corresponding transfer characteristic of I_D versus V_G at a fixed V_{DS} of 20 V for the same TFT device, such that gate leakage determines the transistor 'off' current, which is about 10^{-10} A for the PVP gate dielectric employed here. This reveals a drain current with an on/off ratio of more than 10^6. The threshold voltage (V_{th}) and saturation mobility were defined by fitting a straight line to the plot of the square root of I_D versus V_G, calculated by the formula of saturated regions:

$$I_{DS} = \mu_{FEsat}\left(\frac{W}{2L}\right)C_I(V_{GS} - V_{th})^2$$

where W and L are the channel width and length respectively, C_I is the capacitance per unit area of gate insulator. The V_{th} is 2 ± 0.1 V and the saturated region mobility (μ_{FEsat}) was calculated as 6.9 cm^2/V s.

4. Conclusions

We have demonstrated a solution process for the fabrication of hybrid-structure a-IZO TFTs on flexible PI substrates using microwave heating. Microwave annealing the a-IZO film effectively obtains desired electrical performance without damage to the flexible PI substrate. The a-IZO is one of a group of metal oxide semiconductors used as transparent active channel layers in an amorphous structure. It is more suited to application in flexible electronics than in crystalline films. This result extends the possibilities for new approaches using a-IZO for flexible electronics applications.

References

[1] M.C. McAlpine, H. Ahmad, D. Wang, J.R. Heath, Nat. Mater. 6 (2007) 379.
[2] H. Rost, J. Ficker, J.S. Alonso, L. Leenders, I. McCulloch, Synth. Met. 145 (2004) 83.
[3] J. Kim, J. Jeong, H.D. Cho, C. Lee, S.O. Kim, S.K. Kwon, Y. Hong, J. Phys. D: Appl. Phys. 42 (2009) 115107.
[4] H.Y. Choi, S.H. Kim, J. Jang, Adv. Mater. 16 (2004) 732.
[5] S.C. Wang, C.F. Yeh, C.K. Huang, Y.T. Dai, Jpn. J. Appl. Phys. 42 (2003) 1044.
[6] C.S. Yang, L.L. Smith, C.B. Arthur, G.N. Parsons, J. Vac. Sci. Technol. B 18 (2000) 683.
[7] H. Klauk, M. Halik, U. Zschieschang, G. Schmid, W. Radlik, W.J. Weber, J. Appl. Phys. 92 (2002) 5259.
[8] S.H. Kim, S.Y. Yang, K. Shin, H. Jeon, J.W. Lee, K.P. Hong, C.E. Park, Appl. Phys. Lett. 89 (2006) 183516.
[9] P. Barquinha, A. Pimentel, A. Marques, L. Pereira, R. Martins, E. Fortunato, J. Non-Cryst. Solids 352 (2006) 1794.
[10] J.I. Song, J.S. Park, H. Kim, Y.W. Heo, J.H. Lee, J.J. Kim, G.M. Kim, B.D. Chio, Appl. Phys. Lett. 90 (2007) 022106.
[11] P. Barquinha, G. Gonçalves, L. Pereira, R. Martins, E. Fortunato, Thin Solid Films 516 (2007) 8450.
[12] D.C. Paine, B. Yaglioglu, Z. Beiley, S. Lee, Thin Solid Films 516 (2008) 5894.
[13] M.G. McDowell, I.G. Hill, IEEE Trans. Electr. Dev. 56 (2009) 343.
[14] J.H. Choi, U.B. Han, K.C. Lee, J.H. Lee, J.J. Kim, I.T. Cho, J.H. Lee, Y.W. Heo, J. Vac. Sci. Technol. B 27 (2009) 622.
[15] S. Jeong, Y.G. Ha, J. Moon, A. Facchetti, T.J. Marks, Adv. Mater. 22 (2010) 1346.
[16] C.G. Choi, S.J. Seo, B.S. Bae, Electrochem. Solid-State Lett. 11 (2008) H7.
[17] D.H. Lee, Y.J. Chang, G.S. Herman, C.H. Chang, Adv. Mater. 22 (2007) 843.
[18] H.Y. Chang, K.S. Liu, I.N. Lin, J. Mater. Res. 10 (2006) 2052.
[19] D.K. Hwang, J.H. Park, J. Lee, J.M. Choi, J.H. Kim, E. Kim, S. Im, Electrochem. Solid-State Lett. 8 (2005) G140.