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Oxide-Based Electric-Double-Layer Thin-Film Transistors on a Flexible Substrate

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Abstract— Flexible electric-double-layer (EDL) InGaZnO thin-film transistors (TFTs) were fabricated on a plastic substrate at room temperature. A large EDL gate capacitance, $0.22 \mu\text{F}/\text{cm}^2$, at 20 Hz was achieved using 200-nm-thick radio-frequency magnetron sputtered porous SiO_2 as the dielectric layer, which is equivalent to ~ 15.7 nm thermally grown SiO_2 . The devices therefore show a low operating voltage of 1 V, a high current on-off ratio $> 10^5$ and a low subthreshold swing < 0.12 V/dec. These properties were maintained even after bending, suggesting that the TFTs are suitable for applications in portable sensors and rollable displays.

Index Terms— Electric-double-layer (EDL), flexible thin-film transistors (TFTs), radio-frequency magnetron sputtered SiO_2 , InGaZnO.

I. INTRODUCTION

Oxide-based thin film transistors (TFTs) have gained significant attention especially in recent years due to their large field-effect mobility, low process temperature and large-area uniformity [1-4]. Electronic devices fabricated on flexible substrates provide technological solutions to many applications that cannot be achieved with conventional Si-based ones, such as mechanical flexibility and optical transparency [5]. However, most flexible TFTs require a large operating voltage, leading to high power consumption – a critical barrier to the realization of battery-powered portable electronics [6].

A common method employed to reduce the operating voltage is to reduce the thickness of dielectric layer, e.g. using ultra-thin cross-linked films or self-assembled monolayers [7, 8]. However, such transistors normally suffer large gate leakages due to the difficulty of maintaining good uniformity

over large area. An alternative is to use high relative permittivity (high- κ) dielectric materials as they can increase the capacitive coupling between the gate electrode and channel layer [9, 10]. However, such high- κ dielectric materials usually have problems with threshold voltage control and therefore serious issues with long-term reliability [11].

Recently, some electronically insulating but ion-conducting electrolytes have been used to solve the problem. The most commonly used electrolytes are ionic liquids (ion gels) and polymer electrolytes (polyelectrolytes) [12, 13]. However, it is hard to control the shape, thickness and uniformity of ionic liquids and ion gels. Polymer electrolytes also have limitations in stability, especially at high temperatures. As a result of these limitations, solid-state insulators with porous structures have been developed [14, 15]. Such porous structures allow ions to diffuse, leading to high ionic conductivities. Under a positive gate voltage, the electric field drives the positive mobile ions, such as protons, towards the interface between the electrolyte and the semiconductor channel, and thereby induces electrons within the channel layer. The formation of an electric-double-layer at the interface gives a relatively high specific capacitance and ensures the TFT has a low operating voltage [16]. Plasma-enhanced chemical vapor deposition (PECVD)-deposited porous SiO_2 has been demonstrated as a suitable solid-state electrolyte candidate [17, 18]. Hydrogen molecules from SiH_4 in the plasma are believed able to enter the porous SiO_2 layer and induce mobile protons inside the SiO_2 layer to form a three-coordinate oxygen center, $\text{Si-OH}^+-\text{Si}$ [18]. Since the bonds are not stable, protons hop between two neighboring hydroxyl groups and move to the dielectric/gate interface or dielectric/channel interface according to the polarity of the external electric field [19, 20]. However, PECVD might have toxic, explosive and flammable processing gases and by-products. We recently used radio-frequency (RF) magnetron sputtering to develop a cost-effective, industry compatible way to deposit a porous SiO_2 electrolyte that uses absorbed water molecules to generate mobile protons [20, 21].

In this letter, we report electric-double-layer transistors on polyethylene naphthalate (PEN) substrates fabricated at room temperature using RF magnetron sputtering. The devices have an operating voltage of 1 V and show excellent electrical properties with a current on-off ratio $> 10^5$, a subthreshold swing < 0.12 V/dec before and after bending.

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II. EXPERIMENTAL PROCEDURE

Flexible InGaZnO (IGZO)-based TFTs with a bottom-gate, top-contact structure were fabricated on PEN substrates at room temperature. Firstly, a 200-nm-thick polymethyl methacrylate (PMMA) layer was spin-coated on to the PEN to improve the smoothness of the substrate. 100-nm-thick Al was thermally evaporated to form the gate electrode. Using a SiO₂ target, a 200-nm-thick SiO₂ gate dielectric layer was deposited at 85 W by RF magnetron sputtering. A 70-nm-thick IGZO (In:Ga:Zn = 1:1:1) channel layer was deposited at 40 W by RF magnetron sputtering. Finally, 100-nm-thick Al source and drain were deposited by thermal evaporation. All the sputtering processes were kept at a pressure of 5×10^{-3} mbar in pure Ar ambient and all layers were patterned using shadow masks. Following the deposition of each layer, the samples were brought to atmospheric pressure and removed from the chamber. The channel length and width are 60 μm and 2 mm, respectively.

A schematic diagram of the devices is shown in Fig. 1(a). Fig. 1(b) shows the PEN TFT arrays on the probe station for electrical measurement on a curved surface, indicating the flexibility. The devices were measured using an Agilent E5270B semiconductor analyser and an Agilent E4980A LCR meter in air ambient. The cross-sectional views of SiO₂ and IGZO were investigated using a FEI Nova NanoSEM 450 scanning electron microscope (SEM).

Fig. 1. (a) Schematic diagram of flexible EDL TFTs. (b) Optical image of the electrical measurement of PEN TFT arrays bent on a curved surface with a curvature radius of 3.5 cm.

III. RESULTS AND ANALYSIS

Fig. 2. (a) Cross-sectional SEM image of SiO₂ and IGZO layers. (b) Specific capacitance of SiO₂ gate dielectric as a function of frequency (20 Hz to 100 kHz) with inset showing the capacitance test structure.

Fig. 2(a) shows the SEM of the SiO₂ gate dielectric layer and the IGZO channel layer. The thicknesses of the SiO₂ and the IGZO are approximately 200 nm and 70 nm, respectively. In Fig. 2(a), the IGZO has a similar microstructure to the SiO₂, due to sequential deposition of IGZO on top of SiO₂. As previously reported [21], despite the polycrystalline-like microstructure shown in the SEM image, the SiO₂ layer is amorphous with an inhomogeneous density distribution due to the porous microstructure. Similarly, the IGZO layer should also be amorphous, which is in agreement with the microstructure of most reported IGZO thin films that are deposited at room temperature [4, 5, 22].

Fig. 2(b) shows the specific capacitance of the SiO₂ gate dielectric using an Al/SiO₂/Al sandwich test structure. For conventional, thermally grown SiO₂, a 200-nm-thick SiO₂ layer would have a capacitance of 17.3 nF/cm². In Fig. 2(b), the capacitance is 0.22 $\mu\text{F}/\text{cm}^2$ at 20 Hz and 0.031 $\mu\text{F}/\text{cm}^2$ at 100 kHz. A possible reason for this frequency dependent capacitance is the low ionic mobility of the mobile protons in the SiO₂ dielectric. As indicated in our previous study [21], some water molecules might be absorbed from the ambient air, forming three coordinate oxygen centers, Si-OH⁺-Si, in the SiO₂ electrolyte [18, 23]. The external electric field will force the protons to jump from one hydroxyl group to the next due to the non-stable bonding between hydrogen and oxygen. When a positive bias is applied to the gate electrode, the protons accumulate at the dielectric/channel interface and induce electrons inside the IGZO channel layer to accumulate at the channel/dielectric interface. At low frequencies, the mobile protons inside the SiO₂ layer have enough time to accumulate at SiO₂/Al interface in response to the external electrical field. However, at high frequencies, protons are less able to respond and accumulate at the interface before the electric field switches orientation [24].

Fig. 3. (a) Output characteristics with V_D swept from 0 V to 1 V and V_G swept from 0 V to 1 V with a step of 0.2 V. (b) I_D , $I_D^{1/2}$ and I_G as a function of V_G from -0.5 V to 1 V at $V_D = 1$ V.

Fig. 3(a) shows the output characteristics (I_D - V_D) of the flexible TFTs, indicating the device operates in n -type enhancement mode. Drain current, I_D , is found to increase linearly with the drain voltage, V_D , at low gate voltage, V_G . This demonstrates that a good ohmic contact is formed between the Al source and drain contacts and the IGZO. The estimated contact resistance of the device is less than 10% of the total channel resistance, even at $V_G = 1$ V. It is also found that I_D exhibits a clear pinch off and saturates at higher V_D . A slight decrease of I_D is observed in the saturation regime, which might be caused by a hot carrier effect and/or self-heating effects [25-28]. Under such effects, charge trapping occurs, resulting in a positive threshold voltage shift and a slight decrease in I_D . Fig. 3(b) shows the corresponding transfer characteristics. The devices show an ultralow hysteresis with a current on-off ratio larger than 10^5 . The subthreshold swing, SS , is found to be 79 mV/dec for the forward sweep and 84 mV/dec for the backward sweep, both of which are very close to the theoretical minimum [29]. The gate leakage current, I_G , is less than 0.4 nA, which is smaller than the previously reported EDL transistors gated with polymer electrolytes or ionic liquids [12, 30]. Fig. 3(b) also displays the transfer characteristics as $I_D^{1/2}$ - V_G , by which a threshold voltage, V_{TH} , of 0.22 V is extracted.

In the saturation region ($V_D > V_G - V_{TH}$),

$$I_D = \frac{1}{2} \frac{W}{L} \mu C (V_G - V_{TH})^2 \quad (1)$$

where, W/L is the ratio of channel width and length, C is the capacitance per unit area. By using the capacitance at 20 Hz ($\sim 0.22 \mu\text{F}/\text{cm}^2$), the mobility, μ , is estimated to be $\sim 5.6 \text{ cm}^2/\text{Vs}$, which is reasonable and can be further improved by reducing the channel thickness or by thermal annealing [31, 32].

Fig. 4. (a) Switching stability driven by a 0.1 Hz square wave pulse between -0.5 V and 1 V at $V_{DD} = 1$ V. Inset shows the test circuit and a zoom in part of the input voltage and output voltage against time. (b) Transfer characteristics at a constant V_D of 1 V before and after bias stress. (c) Transfer characteristics with V_G swept from -0.5 V to 1 V at a constant V_D of 1 V before and after bending

In order to study the switching stability, the device was tested by connecting it with a $1 \text{ M}\Omega$ resistor in an inverter setup, as shown in the left inset of Fig. 4(a). The supply voltage, V_{DD} , was fixed at 1 V and the input signal was a 0.1 Hz square wave between -0.5 V and 1 V. The right inset of Fig. 4(a) clearly shows an on to off switch in the output voltage, suggesting the device works as an inverter. The output voltage is 0.5 V in the on state because the internal resistance of the oscilloscope is $1 \text{ M}\Omega$ (R_{OSC} in Fig. 4(a)). The whole circuit was tested for 1000 s and no obvious output voltage drop was observed, indicating that the EDL TFTs have good switching stability. The device was also tested by continuously biasing it at $V_G = -1$ V and $V_D = 1$ V under illumination (2000 lx white LED, 3 cm away from the device), and only a +0.06 V threshold voltage shift was observed after 30 minutes, as shown in Fig. 4(b). It is possible that the stability of the devices can be further improved by adding a capping layer to passivate the top surface of the devices [33].

Table I

C/A (NF/CM ²) AT 100 HZ BEFORE AND AFTER BENDING					
Sample number	1	2	3	4	5
Before bending	179	185	159	165	155
After bending	179	183	160	164	159

The electrical properties of the flexible TFTs were also measured on a curved surface with a 3.5 cm radius of curvature, as shown in Fig. 1(b). Several samples were measured and the gate capacitance was found to be almost the

same before and after bending, as summarized in Table I. Also, the devices showed no breakdown even after a gate bias up to 5 V before and after bending. Fig. 4(c) shows the transfer characteristics of the device at 1 V drain voltage before and after bending. The green curve (before bending) and red curve (during bending) show the current on-off ratio decreases from about 10^6 to 10^5 due to increase of the off current. It is possible that during bending, the distance between atoms in every layer of the transistor increased, resulting in an effective decrease in the energy level splitting of the bonding and antibonding orbitals between the atoms in the IGZO layer [34]. Therefore, some donor electrons might be able to reach the antibonding state after bending, resulting in the increase of off current and a threshold voltage left shift of about 0.07 V [35]. The device was tested again 20 minutes after being removed from the bending holder. The black curve (20 minutes after removal from the holder) shows a decrease in off current and a slight right shift of the threshold voltage when comparing with the red one (before removing from the holder). The comparison of the black curve and green curve indicates the change induced by bending is reversible. Throughout the whole test the current on-off ratio remained $> 10^5$ and the subthreshold swing was $< 0.12 \text{ V}/\text{dec}$. These results are maintained even after several bending events.

Some groups have reported flexible IGZO TFTs before [36-39]. Hsu *et al.* reported an IGZO TFT on a polycarbonate substrate that could achieve a SS of $\sim 0.13 \text{ V}/\text{dec}$ after being bent to a curve with 3.5 cm curvature radius [36]. Lee and co-workers reported an IGZO TFT on a thin glass substrate and the obtained results show a threshold voltage change of 0.25 V and a SS of $\sim 0.28 \text{ V}/\text{dec}$ after being bent to a curve with 4 cm curvature radius [37]. Thus, the electrical properties of our devices match, and even surpass, those of other IGZO TFTs under similar bending conditions. Moreover, thanks to the formation of the EDL, an operating voltage of 1 V is achieved in our devices.

In conclusion, we reported flexible EDL transistors that are fabricated using an industry-compatible sputtering process at room temperature. The work might have important implications in future large-scale, low-cost manufacturing of wearable electronics where a low operating voltage and mechanical flexibility are necessary.

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