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High Performance Amorphous ZnMgO/Carbon Nanotubes Composite Thin-Film Transistors with Tunable Threshold Voltage

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Abstract: Here we report the fabrication and characterizations of high mobility amorphous ZnMgO/single-wall carbon nanotubes composite thin film transistors (TFTs) with tunable threshold voltage. By properly controlling the ratio of MgO, ZnO and carbon nanotubes, high performance

¹⁰ composite TFTs can be obtained with the field-effect mobility up to 135 cm²/V·s, a low threshold voltage of 1 V and a subthreshold swing as small as 200 mV/decade, promising a new solution-processed material for high performance functional circuits. A low voltage inverter is demonstrated with functional frequency exceeding 5 kHz, which is only limited by parasitic capacitance rather than the intrinsic material speed. The overall device performance of the composite TFTs greatly surpasses not only that of

¹⁵ the solution-processed TFTs, but also that of the conventional amorphous or polycrystalline silicon TFTs. It therefore has the potential to open up a new avenue to high-performance, solution-processed flexible electronics to significantly impact the existing applications and enable a whole new generation of flexible, wearable, or disposable electronics.

1. Introduction

With exceptional optical transparency, decent mobility and excellent stability, amorphous metal oxide semiconductors have attracted considerable interest as a unique class of materials for thin film transistors (TFTs).¹⁻² Among various materials, ZnO,³⁻⁴ ZnMgO (ZMO),⁵ In2O3,⁶⁻⁷ InGaZnO,⁸⁻¹¹ InZnO (IZO),¹² and ²⁵ ZnInSnO,¹³⁻¹⁴ have been explored as the promising materials for high device performance. Together with a potentially low-temperature solution processibility, these materials could in principle enable practical and unconventional flexible electronic devices with optical transparency.¹⁵⁻¹⁸ However, the typical ³⁰ mobility of these metal oxide TFTs (1~10 cm²/V·s) is still lower than that of poly-silicon TFTs (10-200 cm²/V·s).¹⁹ The formation of hybrid composite materials could allow for the development of materials with multiple combined advantages over single

component building blocks.²⁰ By incorporating single-wall ³⁵ nanotubes (SWNTs) into the amorphous IZO thin films, we have recently demonstrated that oxide/SWNTs composite TFTs can be

created with greatly improved carrier mobility up to $140 \text{ cm}^2/\text{V} \cdot \text{s}$. Additionally, the composite TFTs also showed exceptional mechanical robustness for flexible electronics on plastics.²¹ This 40 significant improvements could be attributed to the excellent carrier transport and mechanical flexibility of SWNTs.²² The improved mobility was attributed to the reduction of the effective channel length by the embedded SWNTs network in the oxide matrix.²³ However, the electrostatic screening effect of the 45 SWNTs in the composite thin film makes the semiconductor oxide between SWNTs difficult to be pinched off, resulting in a depletion mode TFT with a relatively large negative threshold voltage (V_{th}) and large subthreshold swing (SS), unsuitable for applications in high-speed low power logic circuits.²⁴⁻²⁶ By ⁵⁰ incorporating high oxygen affinities cations (e.g. Mg²⁺) into oxide matrix, here we report the fabrication of high mobility oxide/SWNTs composite TFTs with tunable V_{th} for low voltage logic application.

It has been shown that V_{th} of oxide thin film transistors can ⁵⁵ be modulated by the incorporation high oxygen affinities cations into binary and ternary compound metal oxide thin films, such as Mg²⁷⁻²⁸ and Ga.^{18, 29} Oxygen affinities of these metal ions are higher than that of In, Sn and Zn, resulting in a suppressed carrier concentration to reduce the V_{th} , but also typically coming with a ⁶⁰ severe mobility degradation.^{13,27} Importantly, here we show that high performance TFTs with controllable threshold voltage can be achieved by incorporating SWNTs into an oxide matrix with controlled composition. Amorphous ZMO/SWNTs (a-ZMO/SWNTs) composite TFTs with low V_{th} around 1 V and ⁶⁵ small SS of 200 mV/decade have been fabricated through a solgel route. We further show that these high performance TFTs can

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be used to create logic inverters with voltage gain and excellent speed performance. Our study defines a clear pathway to high performance amorphous oxide/nanotube TFTs with controllable threshold voltage, and represents a critical step forward in 5 developing high speed oxide TFTs for diverse applications in large area flexible electronics.



Fig. 1 (a) Schematic of a TFT with the a-ZMO/SWNTs composite active channel. (b) The Raman spectra of the a-ZMO/SWNTs composite thin 10 film in a device channel; the inset shows an optical microscope image of a TFT. The scale bar is 20 μ m. (c) The transfer characteristics of the a-ZMO/SWNTs composite TFT with different Mg:Zn molar ratio concentrations. (d) The dependence of mobility and V_{th} on the molar ratio of Mg:Zn.

15 2. Results and Discussion

2.1. a-ZMO/SWNTs TFTs

A spin-coating process was employed to prepare the a-ZMO/SWCNTs composite thin films on 100 nm SiO₂ coated p^+ -Si substrate (Supporting Information S1). Two-step ²⁰ photolithography was employed to fabricate the TFTs. The first photolithography was carried out to define the as-prepared thin film into isolated pads, which can suppress the gate leakage current. The second photolithography was conducted to define source and drain electrodes (S/D). The Cr/Au (15 nm/30 nm) ²⁵ electrodes were completed by metal evaporation and lift-off processes. The transmission electron microscope image of the pure SWNTs used in this work is shown in the Fig. S1. The TFTs geometry employed in this work is shown in Fig. 1a. Fig. 1b is the Raman spectra of the composite thin film in a device channel, ³⁰ which is well consistent with that of SWNTs. The electrical measurements were performed with the Lake Shore TTPX Probe Station and Agilent 4155C Semiconductor Parameter Analyzer. The TFTs with Cr/Au contact are in the linear region operation (V_{ds} =1 V), the transfer characteristics of the fabricated TFTs show typical *n*-type characteristics expected for the oxide matrix (Fig. 1c). The field effect mobility is calculated by using the derivative of transfer characteristics:

$$\mu_{FE} = \frac{g_{\rm m}L}{V_{\rm de}C_{\rm i}W} \tag{1}$$

ķ

where the g_m can be obtained by deriving the I_{ds} - V_g curve. In ⁴⁰ general, high mobility values exceeding 100 cm²/V·s are observed in the composite TFTs with relatively low Mg concentration (<10%) (Fig. 1d). It is found that the mobility decreases with the increasing molar ratio of Mg:Zn (e.g. 60 cm²/Vs for Mg:Zn=1:5), which can be attributed to the higher 45 oxygen affinity of Mg than Zn as previously mentioned. 13, 18, 27-29 It is importantly to note that the V_{th} (determined from the horizontal intercept of a linear part in $I_{ds}^{1/2}$ versus V_g plot) can be readily modulated by adjusting the Mg concentration. The TFTs changed from the depletion mode $(V_{th}=-11.2 \text{ V})$ to the ⁵⁰ enhancement mode ($V_{th}=6$ V) when the Mg:Zn ratio is increased from 0 to 1:5 (averaged over dozens of devices). Together, for composite TFT with a Mg:Zn=1:8, a high mobility value of 130 cm^2/Vs and a small V_{th} of 0.8 V can be simultaneously achieved, promising an attractive TFT material for high performance low 55 power applications. These studies demonstrate the carrier mobility and threshold voltage of composite TFTs can be readily tuned to meet the requirement for specific applications by simply adjusting the molar ratio of Mg in the a-ZMO/SWNTs thin film.



 $_{60}$ Fig. 2 (a) The transfer characteristics of the a-ZMO/SWNTs composite TFTs with different SWNTs concentrations. The incorporation of SWNTs enhance the mobility, the molar ratio of Mg:Zn is fixed at 1:8. Due to the incorporated SWNTs, the V_{th} shift from the positive towards negative axis. (b) The output character of the composite thin film transistor with 1 wt% SWNTs. (c) The plot of the field effect mobility and threshold voltage as a function of SWNTs concentration.

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Fig. 3 The performance of a-ZMO/SWNTs composite thin film transistor with different channel length (*W* is fixed at 75 μ m), in which the composite thin film are composed of a-ZMO (Mg:Zn=1:8) and 1 wt% SWNTs. (a) The transfer character curves of the fabricated devices. (b) The plot of mobility and V_{th} versus channel length. (c) Intrinsic delay τ versus channel length. The solid linear is a fit to the data point.

Subsequently, to confirm the 1 wt% SWNTs concentration is also the optimum amount in a-ZMO/SWNTs composite thin film, the TFTs with a fixed 1:8 molar ratio of Mg:Zn with variable SWNTs concentrations from 0 to 2 wt% are fabricated to evaluate the performance of the devices. The transfer characteristics are 10 shown in Fig. 2a. The results are similar to those of the a-IZO/SWNTs composite thin films. Fig. 2b is the output characteristics of the TFT devices with 1 wt% SWNTs. The clear pinch-off and I_{ds} saturation indicate that the carriers transport in active channels is completely controlled by the gate bias.³² Fig. 2c 15 shows the mobility and V_{th} as a function of SWNTs concentrations, it clearly reveals that the mobility is significantly enhanced as the SWNTs concentration increases in the range of 0-1 wt%. Moreover, the V_{th} drifts to 0.8 V with 1 wt% SWNTs, which is much closer to 0 V. Due to the electrostatic screening 20 effect of SWNTs, high concentration SWNTs weakens the gate field, leading to a degraded mobility and requiring large bias to pinch off the channel, namely, the metallic SWNTs network dominates the carriers transport. These studies demonstrate that 1 wt% SWNTs concentration is also the best recipe for the a-25 ZMO/SWNT composite thin film.

2.2. a-ZMO/SWNTs TFTs Based Inverter

To probe the reliability and channel length scaling relations of the composite TFTs, we have prepared and studied a-ZMO/SWNT composite TFTs with the channel length *L* varying ³⁰ from 5 µm to 20 µm. Fig. 3a compares the transfer characteristics of TFTs with different channel length. Fig. 3b summarizes the mobility and V_{th} versus *L*. The field effect mobility values of the TFTs with different channel lengths are all as large as 130 cm²/V·s. An important benchmark of transistor performance is ³⁵ the intrinsic gate delay time, which represents the fundamental *RC* (where *R* is the device resistance and *C* is the capacitance) delay of the device. It provides a speed limitation for transistor operation, which is relatively insensitive to gate dielectrics and device width. Therefore, the intrinsic gate delay time is used as ⁴⁰ an important parameter for comparing different types of electronic materials,³³⁻³⁵ which is given by:

$$\tau = \frac{C_{\rm g}}{g_{\rm m}} = \frac{L^2}{\mu_{FE} V_{ds}} \qquad (2)$$

The intrinsic delay time is varied from 2.2 ns for the $L=5 \mu m$ devices to 31.8 ns for the $L=20 \mu m$ devices, which is observed at ⁴⁵ the largest gm. It is also possible to determine intrinsic cut-off frequency equation:

$$f_T = \frac{1}{2\pi\tau} \qquad (3)$$

where the projected f_T of the TFT devices could be up to 72 MHz for the TFT device with a channel length of 5 µm. A summary of τ from a-ZMO/SWNTs composite TFTs versus *L* is shown in Fig. 4c; the results formations of the devices are fit with L^2 . These advantages, together with the low V_{th} =1 V, suggest that a-ZMO/SWNTs composite TFTs can be applied to the fabrication of high-speed inverters with low power-consumption.



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Fig. 4 The DC and AC properties of the inverter. (a) The transfer characteristics of the TFT (with 50-nm SiO₂ insulator) adopted as the 'switch' of the inverter. (b) The voltage transfer characteristic (left vertical axis) and DC gain (right vertical axis) of an inverter, the inverter s is schematically depicted in the inset. (c) Output waveform by 0.1 kHz with a sinusoidal signal of V_{pp} =500 mV on the input terminal of the amplifier, (d) AC gain (dB) of the inverter at different frequencies. This inverter shows $f_T > 5$ kHz.

The ability to reliably fabricate high mobility composite TFTs 10 with controllable threshold voltage can readily allow us to construct the functional digital circuits. To this end, we have used two TFTs to configure an inverter. First, high field effect mobility is necessary to achieve large voltage gain inverters.³⁶⁻³⁷ Second, a low V_{th}, small SS and high ON/OFF ratio are necessary to 15 minimize power dissipation.^{2, 38-41} Third, a reduced channel length is desirable to minimize the intrinsic gate delay time in high frequency digital circuitry.⁴²⁻⁴⁴ Considering the above requirements, we fabricated a-ZMO/SWNTs TFTs (with 1 wt% SWNTs) on the 50 nm SiO₂ coated p^+ -Si, and employed a L/W = $_{20}$ 5 μ m/75 μ m TFTs for the inverter. The transfer characteristic is displayed in Fig. 4a. The field effect mobility is derived from gm with the value as large as 135 $\text{cm}^2/\text{V}\cdot\text{s}$. Obviously, the SS is decreased to the value of 200 mV/decade. In addition, the a-ZMO/SWNTs TFT is an enhancement-mode device with $V_{th}=0.8$ $_{25}$ V, which is off at $V_{o}=0$ V.

To evaluate both the static DC and AC performance of the inverter, we adopt another a-ZMO/SWNTs composite TFT with $L/W=500\mu$ m/75 μ m working at the saturation state as the "load". The "switch" TFT with $L/W=5\mu$ m/75 μ m is connected with the ³⁰ "load" in series, as shown in the inset of Fig. 4b. The gate of the "switch" TFT acts as the input terminal, whereas the connection between the "switch" and the "load" acts as the output terminal. The static DC performance of an inverter is evaluated by its voltage transfer characteristics and gain shown in Fig. 4b, with

- ³⁵ $V_{dd}=V_{ds}=10$ V. The slope of the steepest region of the $V_{out}-V_g$ curve represents the voltage gain G of the amplifier. In this case, the gain of our amplifier is |G|=3. It is important to note that a gain higher than 1 is desirable for practical applications. When a small AC signal V_{in-AC} is superimposed on the DC bias V_g at the 40 input, $V_{in}=V_g+V_{in-AC}$, then under the right circumstances the
- ⁴⁰ input, $v_{in}-v_g \tau v_{in-AC}$, then under the right circumstances the transistor circuit can act as a linear amplifier. The transistor is first biased at a certain DC gate voltage to establish a desired current in the circuit. A small sinusoidal AC signal V_{in-AC} is then superimpose on the gate bias on the input, causing the output ⁴⁵ voltage V_{out} to oscillate synchronously with a phase difference of

180°, with respect to V_{in-AC} . For maximum output voltage swing, we supply a gate bias of $V_g=1$ V to the 'switch' TFT and superimpose small V_{in-AC} signals of $V_{pp}=500$ mV with different frequencies on the gate bias as V_{dd} so $=V_{ds}=5$ V. The amplified output signals with 0.1 Hz is shown in Fig. 4c. For low frequencies (0.1 kHz), the gain |G| defined as $\Delta V_{out}/\Delta V_{in}$ is larger than 6. By measuring the dynamic response of the inverter with a harmonic-waveform input signal from 10 to 1 MHz as shown in Fig. 4d, it is found that the inverter is so characterized by a cut-off frequency above 5 kHz. Note that this value is obtained for the TFT with a common gate configuration where a huge parasitic capacitance exists between the Si substrate gate and the large size S/D metal pads. So further performance improvement at the inverter level should also be possible through of optimization of the TFT geometry with top-gated structure for small parasitic capacitance.⁴⁵

3. Conclusions

In summary, we have demonstrated high-performance a-ZMO/SWNTs composite TFTs with high field-effect mobility, ⁶⁵ high ON/OFF ratio and tunable threshold voltage, through the low-cost sol-gel approach. The fabricated a-ZMO/SWNTs TFTs outperform the previous reported amorphous metal oxide based TFTs. Furthermore, the inverter based on a-ZMO/SWNTs TFTs demonstrates high-speed operation. These advantages, together ⁷⁰ with the demonstrated control over threshold voltage, suggest substantial potential of a-ZMO/SWNTs TFTs for highperformance low cost electronic applications.

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Contents Entry:

High performance a-ZMO/SWNTs composite TFTs with tunable threshold voltage are fabricated by sol-gel method. A low voltage inverter is demonstrated with functional frequency exceeding 5 kHz

