Electrical Properties of the Thin-Film Transistor with an Indium Gallium Zinc Oxide Channel and Aluminium Oxide Gate Dielectric Stack Formed by Solution-based Atmospheric Pressure Deposition

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***Abstract*—We developed a thin-film transistor (TFT) with an amorphous InGaZnOx (IGZO) channel and aluminium oxide (AlOx) gate dielectric stack that was formed using a solution-based atmospheric pressure chemical vapour deposition. A breakdown electric field of 5.9 MV/cm and a dielectric constant of 6.8 were achieved for the AlOx gate dielectric. The non-vacuum processed IGZO TFT gave a field effect mobility of 4.2 $cm^2 \cdot V^{-1} \cdot s^{-1}$ and an on/off current ratio of over 10^8. Moreover, the proposed deposition method is a powerful tool for material research to explore multi-component oxide insulators and semiconductors.**

***Index Terms*—indium gallium zinc oxide (IGZO), aluminum oxide, chemical vapour deposition, thin-film transistor (TFT)**

I. Introduction

High mobility (μ) oxide thin-film transistors (TFTs) have received considerable attention regarding their possible use in next generation flat panel displays [1–5]. High μ amorphous-oxide TFTs have been demonstrated by using vacuum depositions. Very recently, several research groups have exhibited ZnSnO (ZTO), $InGaZnO_4$ (IGZO), and InZnO (IZO) TFTs through solution-based processes such as sol-gel and spin coating [6–8]. The non-vacuum process is a simple and powerful tool for reducing production cost and equipment investment in the flat-panel display (FPD) industry; however, most of the current research still utilises vacuum deposited films as a gate dielectric. The atmospheric pressure deposition of both oxide semiconductor and gate dielectric is a challenging and key development milestone in the achievement of simple and cost-effective TFT processes. Avis *et al.* [9] demonstrated high μ ZTO TFT with a solution processed ZTO/AlOx gate dielectric stack. However, the spin coating and subsequent hot plate annealing were repeated a number of times in order to have a suitable film thickness for the TFT.

In this letter, we report a TFT with an IGZO channel and AlOx gate dielectric stack that was deposited by a solution-based atmospheric pressure chemical vapour deposition (AP-CVD). The proposed solution-based AP-CVD process is safe, has low energy consumption, and has low cost without vacuum equipment [10]. Moreover, film thickness can be controlled by adjusting the deposition time.

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II. Experimental Procedure

A schematic illustration of the solution-based AP-CVD (known as a “mist-CVD”) system is shown in Fig. 1. The mist-CVD apparatus consists of a source solution supply unit and a deposition unit. The precursor solution is atomised using ultrasonic transducers in the source solution supply unit. The atomised precursor solution is transferred to the deposition unit with carrier gas (air), and the films are deposited by thermal decomposition and chemical reactions.

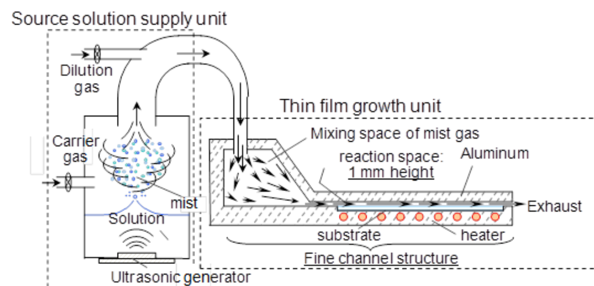

Fig. 1. Schematic illustration of the “mist-CVD” system.

To apply this method to oxide TFTs, we investigated the structural and electrical properties of AlOx film for a gate dielectric and IGZO film for an active channel. The precursor solute and deposition conditions of the AlOx and IGZO films are summarised in Table. 1.

III. Results and Discussion

The dielectric constant value (k) and the breakdown field (E_{BD}) of the 50-nm thick AlOx films deposited on p^+-Si wafers were evaluated with a Mercury probe capacitance vs. voltage (C-V) and a current vs. voltage (I-V) systems. Fig. 2(a) shows the dependence of E_{BD} on deposition temperature (T_S). The E_{BD} dramatically improves from 0.4 to over 5 MV/cm when T_S rises from 350 to 400 °C.

TABLE I DEPOSITION CONDITIONS OF ALOX AND INGAZNO

For AlOx deposition	
Solute	0.020 mol/L Aluminum acetylacetonate
Solvent	Distilled water : Methanol (10 : 90 in vol.%)
Carrier gas	Air, 5.0 L/min
Dilution gas	Air, 20.0 L/min
Deposition temp.	300–430 °C
Deposition rate	5.3-17.8 nm/min at 300-430 °C
For IGZO deposition	
Solute	0.03 mol/L Indium acetylacetonate, Gallium acetylacetonate, and Zinc acetylacetonate (Molar ratio of In:Ga:Zn is 1:1:1)
Solvent	Distilled water : Methanol (10 : 90 in vol.%)
Carrier gas	Air, 5.0 L/min
Dilution gas	Air, 20.0 L/min
Deposition temp.	300 and 350 °C
Deposition rate	19.0 and 27.8 nm/min at 300 and 350 °C

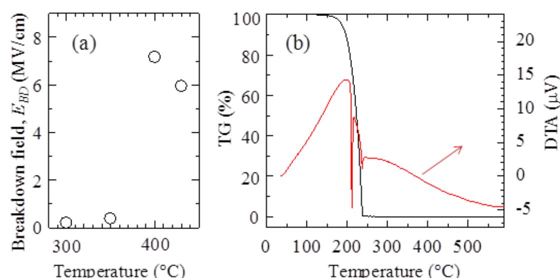

Fig. 2. (a) Deposition temperature dependence of the breakdown field (E_{BD}) of AlOx film, and (b) TG-DTA results of the aluminum acetylacetonate [$Al(C_5H_7O_2)_3$].

To determine the possible chemical reactions of the AlOx deposition, thermogravimetry and differential thermal analysis (TG-DTA) was performed in an air atmosphere [11]. Fig. 2(b) shows the TG-DTA results of the powder $Al(C_5H_7O_2)_3$ used as the source solute for AlOx deposition. The sharp endothermic peaks at 211.9 and 237.8 °C accompanied by a large weight loss represents the $Al(C_5H_7O_2)_3$ decomposition to alumina and aluminium-oxyhydroxide (AlOOH). From a Fourier-transform infrared spectroscopy analysis [not shown], the stretching vibrations of the -OH groups in the AlOx declined as the T_S rose to more than 400 °C. The DTA result also indicated a broad exothermal process up to 600 °C. These results suggest that the E_{BD} of AlOx is influenced by the hydrogen concentration in the film; thus, the appropriate deposition temperature is mainly determined by the decomposition and the reaction temperature of the source solute. A dielectric constant (k) of 6.8 and a breakdown field (E_{BD}) of 5.9 MV/cm were obtained at the T_S of 430 °C. The stoichiometry of the AlOx was evaluated using Auger electron spectroscopy (AES). The AlOx film was found to have an Al:O ratio of 36.5:63.5%, which is oxygen rich (O/Al = 1.74) compared to the stoichiometry of Al_2O_3.

For the IGZO deposition, the molar ratio of In:Ga:Zn in the mixed solution was set at 1:1:1. Figs. 3(a) and 3(b) show atomic force microscope (AFM) images of the IGZO films deposited at 300 and 350 °C, respectively. Although the IGZO film deposited at 300 °C shows a porous microstructure with a rough surface, the film became a dense structure with a smooth surface as the T_S increased to 350 °C. Fig. 3(c) shows the X-ray diffraction pattern of the 200-nm thick IGZO film. The IGZO film does not have any sharp diffraction peaks, indicating an amorphous nature of the film. The In:Ga:Zn ratio in the IGZO film was evaluated by X-ray photoelectron spectroscopy (XPS). A sputter-deposited IGZO film from the IGZO (In:Ga:Zn = 1:1:1) target was used for comparative analysis. The In:Ga:Zn ratios in sputter-deposited and mist-CVD deposited IGZO films are 1:1.3:1.5 and 1:5.0:3.4, respectively. This result suggests that Ga and Zn ions possess stronger chemical bonds with oxygen than In ions during the CVD process. Further studies are needed to clarify the growth mechanism and to control the material composition of multi-component oxide films.

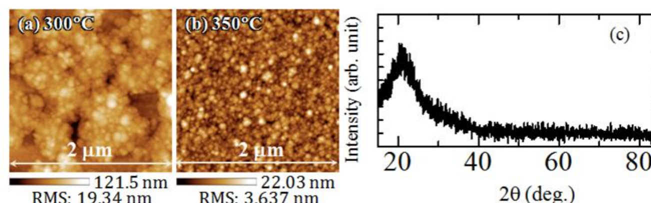

Fig. 3. AFM images of the 200-nm thick IGZO films deposited at (a) 300 and (b) 350 °C on quartz substrates, respectively. (c) XRD spectra of 200-nm thick IGZO film deposited at 350 °C.

Next, bottom-gate TFT was fabricated. Fig. 4 shows a schematic cross sectional view of the IGZO TFT. Firstly, a chromium (Cr) gate electrode was formed on a glass substrate. Then, a 116-nm thick AlOx gate dielectric and a 47-nm thick IGZO channel were deposited at atmospheric pressure by the mist-CVD. The T_S of the AlOx and IGZO deposition were chosen to be 430 and 350 °C, respectively. We waited several hours between the AlOx and the IGZO depositions in order to change the deposition temperature. After patterning the IGZO channel (photolithography and dry-etching), indium-tin-oxide (ITO) source/drain electrodes were formed by lift-off. Finally, the TFTs were annealed at 350 °C for 1 h in forming gas (5 % H_2 in N_2) to improve the electrical properties of the TFT. The channel length (L) and width (W) of the TFT were 30 and 45 μm, respectively.

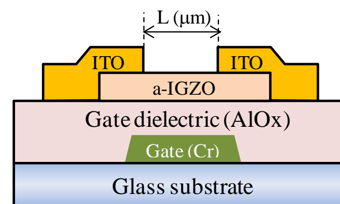

Fig. 4 Schematic cross sectional view of the IGZO TFT

Output characteristics of the IGZO TFT are shown in Fig. 5(a). The IGZO TFT exhibited clear pinch-off and good saturation behaviour. No current crowding was observed, indicating good ohmic properties between the IZGO channel and the source/drain electrodes. The transfer characteristics are shown in Fig. 5(b). Two sets of drain current versus gate voltage (I_D-V_G) curves were measured at drain voltages (V_D) of 0.1 (linear region) and 20.1 V (saturation region) with the double sweep gate voltage mode. The leakage current of the

AlOx gate dielectric [not shown] was suppressed below 1 pA.

The field effect motilities in linear (μ_{lin}) and saturation (μ_{sat}) regions were 4.2 and 4.0 $cm^2 \cdot V^{-1} \cdot s^{-1}$, respectively. In the saturation region, the threshold voltage (V_{th}), which was extracted from the square root plot of the drain current, was estimated to be 3.6 V. An on/off current ratio of over 10^8 was also obtained.

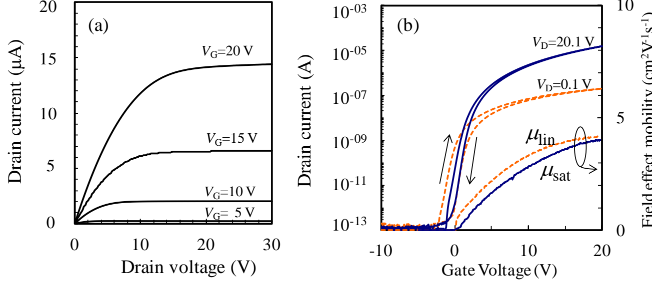

Fig. 5. (a) Output and (b) transfer characteristics of the IGZO TFT.

The subthreshold swing (S), which is defined as the V_G required to increase the I_D by one decade (from 10 to 100 pA), was 0.55 V/dec. The S value is influenced by the maximum area density of states (N_S^{max}), including the interfacial and semiconductor bulk traps. The N_S^{max} can be calculated using following equation:

$$N_S^{max} = \left(\frac{S \times \log e}{kT/q} - 1\right)\frac{C_i}{q} \qquad (1)$$

where q is the unit charge, k is the Boltzmann constant, C_i is the gate capacitance per unit area, and T is the temperature. The N_S^{max} is 2.7×10^{12} $cm^{-2}eV^{-1}$, which is over ten times higher than that of sputter-deposited IGZO TFT [4]. Moreover, the clockwise hysteresis (ΔV_H), which is defined by the difference in the V_G at the I_D of 1 nA between the V_G sweeps of off-to-on and on-to-off, was 0.68 V. The clockwise hysteresis is most likely due to negative charge trapping in an oxide semiconductor or in an oxide/date dielectric interface.

The experimental N_S^{max} and ΔV_H suggests that the trap formation occurs at the oxide/gate dielectric interface or in an early stage of the IGZO deposition. For the AP-CVD of the IGZO/AlOx stack, the adsorption of carbon-related species on the gate dielectric surface should be considered a contamination. It is noted that we waited several hours between the AlOx and the IGZO depositions due to an experimental limitation. In addition, the surface treatment was not applied to the gate dielectric surface prior to the IGZO deposition. Thus, surface treatment of the gate dielectric would further improve electrical performance.

In addition, uniformity and reliability are important issues for TFTs with CVD deposited multi-component oxide semiconductors. We previously reported that the surface treatment of a gate dielectric affects the uniformity of the oxide TFTs [12]. Furthermore, passivation of the TFT is necessary for reliability measurements in order to avoid environmental effects [13]. Future research should thoroughly investigate the uniformity and reliability of the TFTs.

IV. CONCLUSION

In summary, we have developed an IGZO TFT with a non-vacuum processed IGZO/AlOx gate dielectric stack. The IGZO TFT gave a μ of 4.2 $cm^2 \cdot V^{-1} \cdot s^{-1}$and an on/off current ratio of over 10^8. Non-vacuum CVD processes can deliver both energy savings and fast returns on investments in the FPD industry. Moreover, the solution-based CVD is a powerful tool for material research to explore multi-component oxide insulators and semiconductors.

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