Fully Solution-Processed Sodium Doped ZnO Thin-Film Transistors via a Low-Temperature Aqueous Route

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***Abstract*—In this study, we develop a facile and eco-friendly aqueous route for the fabrication of sodium (Na) doped ZnO/AlO_x thin-film transistors (TFTs). To prepare Na doped ZnO and AlO_x thin films, ammonia water and deionized water were used as the precursor solvents. The AlO_x thin film annealed at 300 °C showed an areal-capacitance of 129 nF/cm^2 at 1 kHz. On the basis of its implementation as the gate oxide, fully solution-processed Na doped ZnO TFTs were fabricated and the electrical characteristics were systematically studied. The fully solution processed Na doped ZnO/AlO_x TFTs exhibited a high field effect mobility of 20.73 cm^{2}V^{-1}s^{-1}, a subthreshold swing off 0.58 V/decade, a threshold voltage of 0.8 V, and an on/off current ratio of 2×10^4.**

Keywords-solution-processed; metal oxide thin-film transistors; low-temperature; aqueous route

I. INTRODUCTION

Metal oxide thin-film transistors (MOTFTs) have been extensively investigated for next-generation displays and sensors due to their high mobility, excellent stability, and large area uniformity [1]. The solution processed MOTFTs have been developed due to the possibility of low-cost and large-area fabrication without high-vacuum deposition techniques. Various metal-alloy zinc oxides, such as indium gallium zinc oxide (IGZO), indium zinc oxide (IZO), and indium titanium zinc oxide (ITZO) TFT have been introduced in order to compensate for the poor electrical performance of pure ZnO processed at relatively low temperature [2-4]. However, the cost of indium (In) has increased rapidly due to supply shortage. To find an alternative route to improve the carrier mobility of solution-processed ZnO, various dopants have been introduced to form indium-free zinc oxide semiconductors. Unfortunately, toxic organic solvents such as 2-methoxyethanol (2-ME) have been used [5] Recently, a facile and environmental friendly 'aqueous route' was proposed for the fabrication of MOTFTs at low temperature. In the present work, we report on the fabrication of Na doped ZnO (Na-ZnO) and AlO_x thin films using water as precursor solvent. The electrical characteristics of solution-processed AlO_x high-*k* dielectrics and fully solution-processed Na-ZnO TFTs were studied in detail. In particular, The Na-ZnO/AlO_x TFTs annealed at 300 °C showed good mobility of 20.73 cm^{2}V^{-1}s^{-1} at low operation voltage of 3 V.

II. EXPERIMENTAL

The AlO_x precursor solution was prepared by dissolving 2.5 M aluminum nitrate nonahydrate ($Al(NO_3)_3{\cdot}9H_2O$) into deionized (DI) water. For the Na-ZnO precursor solution, 81.39 mg zinc oxide and sodium hydroxide were dissolved into 10 ml of ammonia water (28%). The molar ratio between sodium hydroxide and ZnO was 15 mol%. All the precursors were stirred

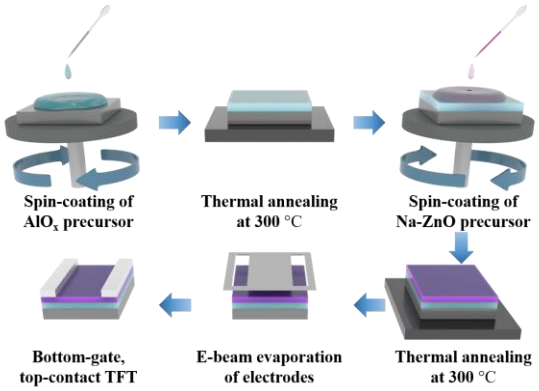

Figure 1. A schematic process flow of the fabrication of fully solution-processed Na-ZnO/AlO_x TFTs.

vigorously for 30 min. and then filtered through 0.45 μm polyethersulfone (PES) syringe filters before spin coating. The fabrication procedure for the Na-ZnO/AlO_x TFTs is illustrated in Fig. 1. Heavily doped n-type Si substrates, acting as the common gate electrodes, were exposed to air plasma for 30 min. to enhance the hydrophilicity before spin coating. The AlO_x precursor solution was spun onto the substrate at 4500 rpm for 40 s and then annealed at 300 °C under ambient atmosphere for 1 h. For the metal-insulator-metal (MIM) devices, Al electrodes (300 nm) were deposited on the AlO_x layer by electron beam evaporation. To fabricate the TFTs with a bottom-gate top-contact structure, the Na-ZnO precursor solution was spun onto the AlO_x gate dielectric at 3000 rmp for 30 s and annealed at 300 °C under ambient atmosphere for 1 h to achieve thickness of about 20 nm. Finally, fabrication of the Na-ZnO/AlO_x TFTs was completed with the electron beam evaporation of 300 nm-thick Al top source/drain and bottom gate electrodes through shadow masks. The channel width and length were 1500 and 100 μm, respectively.

III. Results and Discussion

Fig. 2 shows the capacitance-frequency curve for the AlO_x thin film. The film annealed at 300 °C had areal capacitance of 129 nF/cm^2 at 1000 Hz, corresponding to dielectric constant of 8.7. In addition, the AlO_x thin film showed weak frequency dependence of capacitance, consistent with low leakage current level, indicating dense metal-oxide bonding formation and low defect density. The surface morphology of the solution-processed AlO_x film was observed by the atomic force microscope (AFM). The root-mean-square (rms) roughness of AlO_x was 0.41 nm, indicating an ultra-smooth surface. The transfer and output characteristics of Na-ZnO/AlO_x TFTs annealed at 300 °C are presented in Fig. 3. The device showed an average mobility of 20.73 cm^{2}V^{-1}s^{-1}, a subthreshold swing off 0.58 V/decade, a threshold voltage of 0.8 V, and an on/off current ratio of 2×10^4. For Na-ZnO/AlO_x TFTs, the mobility was improved by a factor of 2 compared to Na-ZnO/SiO_x TFT reported previously [6]. This is due to the combination of high-*k* AlO_x dielectric bringing an excellent heterogeneous interfacial layer formed with

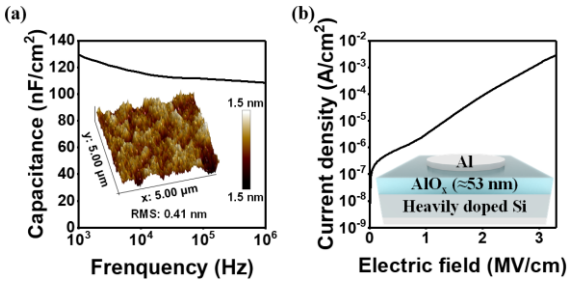

Figure 2. (a) Capacitance versus frequency of AlO_x dielectrics and AFM image of the AlO_x film (inset). (b) Current density versus electric field of AlO_x dielectrics and the schemic of the device configuration (inset).

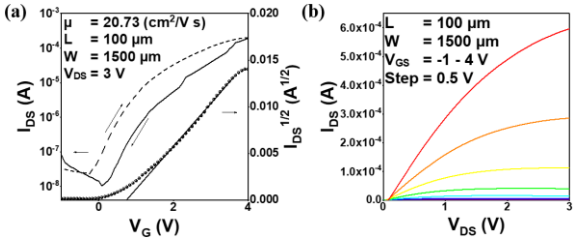

Figure 3. (a) Transfer and (b) output characteristics of a bottom-gate, top-contact Na-ZnO/AlO_x TFTs.

metal oxide semiconductor. Additionally, a counterclockwise hysteresis was observed in the transfer curve. Donor-like electron traps in the AlO_x might be responsible for this hysteresis as well as the mobility enhancement by improving the steady-state electron density in the semiconductor layer [7]. There might be donor-like electron traps injecting electrons into the Na-ZnO through a thermally activated emissive process. The electron injection results in an increase of the steady-state electron concentration in the Na-ZnO, which leads to an increase in electron mobility as transport is trap limited [7].

IV. Conclusion

In conclusion, we report on fully solution-processed Na doped ZnO/AlO_x thin-film transistors via an aqueous route. The electrical properties of the aqueous solution-processed high-*k* AlO_x dielectric and the MOTFTs were intensively investigated. The water-induced AlO_x thin film exhibited ultra-smooth morphology and high areal capacitance of 129 nF/cm^2. The fully solution-processed 15% Na-doped/AlO_x TFT showed a mobility of 20.73 cm^{2}V^{-1}s^{-1}, a subthreshold swing off 0.58 V/decade, a threshold voltage of 0.8 V, and an on/off current ratio of 2×10^4. This low-temperature aqueous route is applicable to a broad range of ZnO based TFTs, it should provide an eco-friendly and facile approach towards the development of indium-free ZnO-based metal oxide TFTs.

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