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Defect modification in ZnInSnO transistor with solution-processed Al_2O_3 dielectric by annealing

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ABSTRACT

The properties of solution-processed Al_2O_3 thin films annealed at different temperatures were thoroughly studied through thermogravimetry–differential thermal analysis, UV–vis-NIR spectrophotometer measurements, scanning electron microscopy, X-ray diffraction, atomic force microscopy and a series of electrical measurements. The solution-processed ZnInSnO thin films transistors (TFTs) with the prepared Al_2O_3 dielectric were annealed at different temperatures. The TFTs annealed at 600 °C have displayed excellent electrical performance such as the field-effect mobility of 116.9 $cm^2 V^{-1} s^{-1}$ and a subthreshold slope of 93.3 mV/dec. The performance of TFT device could be controlled by adjusting the annealing temperature. The results of two-dimensional device simulations demonstrate that the improvement of device performance are closely related with the reduction of interface defects between channel and dielectric and subgap density of stats (DOS) in the channel layer.

1. Introduction

Printed electronics has drawn tremendous attention during the past few decades due to their potential of reducing manufacturing costs and environmentally-friendly products [1–4]. As one of the most important devices among various kinds of printed electronics, the amorphous oxide thin film transistors (TFTs) have been grabbing the researchers' eyes for their high mobility, optical transparency and solution processability [5,6]. Nowadays, the solution-processed methods have been successfully applied to fabricate the oxide TFTs, such as amorphous indium-gallium-zinc oxide (IGZO) [7], amorphous zinc-indium-tin oxide (ZITO) [8], Hafnium-indium-zinc oxide (HIZO) [9]. Among the amorphous oxide semiconductors, the ZITO material is promising to achieve high mobility because of its conductivity path and some studies have demonstrated the ZITO TFTs with the mobility higher than 30 cm^2/V s [10,11]. Besides, much efforts is also being put to study high-k gate dielectrics to improve the performance of TFTs [12,13]. Among high-k material dielectrics, Al_2O_3 is an excellent alternate for the gate dielectrics because of its abundance in nature, low cost, good chemical stability, high breakdown voltage and low interfacial trap density with oxide semiconductors [14].

Several groups have successfully fabricated the oxide TFTs using Al_2O_3 dielectric based on the vacuum technology or the solution method [15,16]. However, they just focused on the properties of Al_2O_3 dielectric with the annealing temperature and few of them studied the effects of interface between Al_2O_3 dielectric and semiconductors on the device performance. Although C.S. Fuh and S.W. Jeong et al. have studied the effects of interface trap sites between the dielectrics and semiconductors on the TFTs' electrical properties based on the vacuum technologies [17,18], very little is known about the effect of the post-annealing process on the interface trap density in Al_2O_3/ZITO-based semiconductor systems using the solution-processed methods, let alone the reasons for the threshold-voltage changes of ZITO-based oxide transistors in different annealing temperatures.

In this paper, the effects of annealing temperature on the solution-processed Al_2O_3 thin films have been studied in detail. The changes of interface trap charge between the Al_2O_3 dielectric and ZITO semiconductor controlled with the annealing temperature as well as the effects on the device performance have also been investigated. In addition, two-dimensional device simulations integrated with the modeling methodology based on the subgap density of states (DOS) and the interface charge for the solution-processed ZITO TFTs are presented. We combine experiments and simulation to study the effects of annealing temperatures on the electrical characteristics of the solution-processed ZITO TFTs with the Al_2O_3 dielectric.

2. Experimental

To begin with, the 0.3 mol/L Al_2O_3 solution is prepared by dissolving $Al(NO_3)_3 \cdot 9H_2O$ in 2-methoxyethanol (2-MOE). The

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0.3 mol/L ZITO solution is synthesized by dissolving zinc acetate dehydrate ($Zn(OC_2H_4)_2 \cdot 2H_2O$), indium chloride ($InCl_3$), and tin chloride pentahydrate ($SnCl_4 \cdot 5H_2O$) in 2-MOE solvent. Next, ethanolamine (99%) as a stabilizing agent is added into the mixed metal salt solutions. Both the ZITO and Al_2O_3 solutions are stirred vigorously in the water bath at 70 °C for 3 h, and then aged at least 1 d.

Three kinds of samples are fabricated to study the properties of Al_2O_3 dielectrics. The first one is the bare glass, which is used to study the thickness, crystallinity and roughness. The second one with ITO/Al_2O_3/Al metal–insulator–metal (MIM) capacitor structures is fabricated to characterize the electrical properties of Al_2O_3 films. The last one is TFTs with bottom gate top contact (BGTC) structure.

The BGTC TFTs were fabricated as the followings. Firstly, a 50-nm-thick ITO layer is sputter-deposited on a Corning EXG glass substrate (0.7 mm thick) for gate formation. Secondly, the synthesized Al_2O_3 gate dielectric is spin-coated at a speed of 3000 rpm for 30 s onto the sputter-deposited and patterned ITO gate substrate, subsequently annealed at 300 °C for 1 h. The aforementioned processes are repeated five times to achieve a thickness of approximately 110 nm. After the Al_2O_3 layer patterned via a wet-etch method with diluted hydrofluoric acid, ZITO solution is spin-coated (30 s at 3000 rpm) and annealed (300 °C for 1 h) at the same condition as the dielectric layer coated on the prepared gate insulator. The ZITO layers are repeated twice to achieve a prospective thickness about 50 nm. Next, active-island patterns are fabricated by a wet-etch method with oxalic acid. After the patterning process, the ZITO films are annealed at 350 °C for 1 h in air. Subsequentially, an ITO layer with a thickness of 50 nm is deposited on the ZITO thin-film by sputtering and patterned by a wet-etch method with PAN (a mixture of phosphoric acid, acetic acid, nitric acid, and water) to form source/drain electrodes, creating transistors with channel length (L) and width (W) of 6 and 30 μm, respectively. After all devices are completely fabricated, the TFTs are annealed with different temperatures of 400 °C, 500 °C and 600 °C for 2 h at air atmosphere in the tube furnace.

The thermal behavior of the Al_2O_3 precursor solution is analyzed by the thermogravimetry–differential thermal analysis (TG-DTA, Q600 analyzer) at a heating rate of 4 °C/min. The optical transmission of Al_2O_3 films is measured with the wavelength ranging from 300 to 800 nm by UV–vis-NIR spectrophotometer (H-3900). The thickness of Al_2O_3 film is characterized by using the scanning electron microscopy (SEM, Hitachi S-4800). The crystallization of Al_2O_3 thin films is detected by X-ray diffraction (XRD, D/MAX-2550) with the glancing incident angle of 1°. Surface analyses of the Al_2O_3 films are observed through atomic force microscopy (AFM, Nanonavi SPA-400). The capacitance–voltage (C–V) and current–voltage (I–V) curves of the ITO/Al_2O_3/Al capacitors and the transfer characteristics of the TFTs are measured by Agilent E4980A precision LCR meter and Agilent 4155C semiconductor parameter analyzer, respectively.

3. Results and discussion

TG-DTA is performed to monitor the effects of temperature on the thermal decomposition behavior of Al_2O_3 solution, as shown in Fig. 1. The significant weight loss of the solution accompanied with a distinct endothermic reaction happens in the range of 25–110 °C, which is attributed to the evaporation of both the solvent and organic molecules. The endothermic reaction near ~290 °C is attributed to the hydrolysis of the metal precursors [19]. With the alloy process at 290–400 °C, the organic molecules are decomposed gradually, and the organic-related residuals and defects in

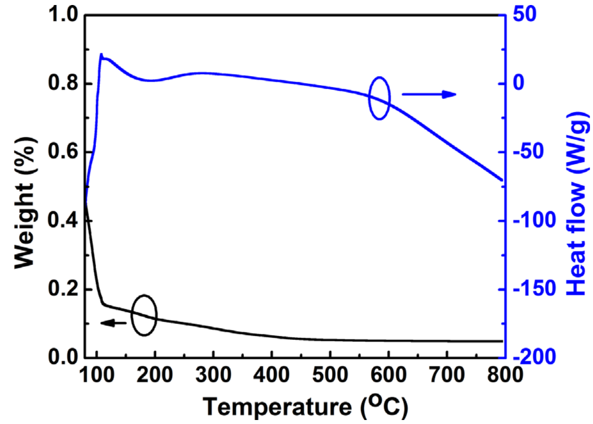

Fig. 1. TG-DTA curves of Al_2O_3 precursor solution, measured at a heating rate of 5 °C/min.

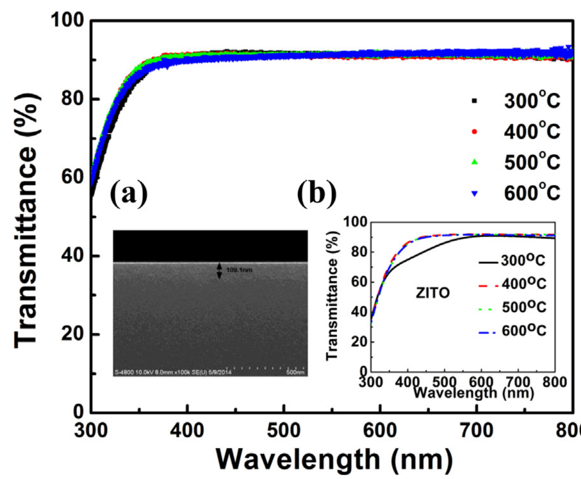

Fig. 2. Optical transmission of the Al_2O_3 films with various annealing temperatures. Inset (a) is a typical SEM section view image of the Al_2O_3 thin films, inset (b) is the transmission of the ZITO films with various annealing temperatures.

the film are further decreased. Above 400 °C, no significant weight and chemical reaction are observed, implying that the thermal reaction and alloy mechanism are almost completed.

Fig. 2 shows the optical transmission of the Al_2O_3 films on the glass substrate annealed at different temperatures. It can be seen that all the transmittance is about 90% (including the glass substrate) in the visible range of 400–800 nm. The result also suggests that the solution processed Al_2O_3 thin films have excellent optical properties and could be the promising insulators applied in the transparent TFTs. The inset (a) in Fig. 2 shows the typical image of the prepared Al_2O_3 thin films annealed at 500 °C, and the thickness of the films is ~109.1 nm. Inset (b) shows that all the films have high transmittance in the visible range of 400–800 nm except for the film annealed at 300 °C, which implies that there are little defects in the films [20]. In Fig. 3, the XRD patterns of Al_2O_3 films annealed at different temperatures shows that all the samples stayed amorphous phase. Previous researches have shown that the Al_2O_3 films showed high crystallization temperature up to 1000 °C [21]. The amorphous phase is preferred to the Al_2O_3 films for the absence of crystal boundaries contributing to a fewer electron diffusion path, which reduces leakage current and increases breakdown voltage [22–23].

It is well known that the good interface roughness of the gate insulators is one of the most important parameters to the device performance, which can reduce the carrier scattering and obtain high field effect mobility [24]. Fig. 4 shows the AFM images