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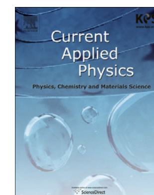

Highly stable hafnium–tin–zinc oxide thin film transistors with stacked bilayer active layers

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ABSTRACT

Hf–Sn–Zn–O (HTZO) thin films were prepared on SiO_2/SiN_x substrates at room temperature by the direct current (DC) magnetron sputtering of Hf-doped Sn–Zn–O targets. The characteristics of films with different amounts of Hf were analyzed. Amorphous HTZO films were obtained by increasing the Hf content, while polycrystalline films have not shown with Hf doping. With the proper Hf concentration in the HTZO films (~2.0 atomic % Hf/(Hf + Sn + Zn + O)), HTZO films demonstrated good performance as an oxide semiconductor channel material in thin film transistors (TFTs) with a field effect mobility (μ_{FE}) of 10.9 cm^2V^{-1} s^{-1}, an on/off current ratio of 10^9, and a subthreshold voltage swing of 0.71 V/decade.
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1. Introduction

Recently, ZnO-based amorphous oxide semiconductor (AOS) transparent thin film transistors (TTFTs) have been received much attention as an alternative candidate to amorphous-Si (*a*-Si) and/or polycrystalline-Si (poly-Si) TFTs in applications such as active-matrix liquid-crystal displays (AMLCDs), organic light-emitting diodes (OLEDs), and flexible electronics [1]. Very recently, LG Display released a 55 in, full high-definition (FHD) OLED television which utilizes an InGaZnO (IGZO) TFT active matrix [2]. AOS TFTs exhibit a high μ_{FE} compared to conventional a-Si:H TFTs (<1 cm^2/Vs). A number of groups have developed AOS TFTs using IGZO [3–8], In–Zn–O (IZO) [9] and Si, Hf-doped IZO [10,11], and Zn–Sn–O (ZTO) [12] and oxygen vacancy suppressor (Zr, Si, Hf, Al, and Ta) doped ZTO [13–17]. Although In- and Ga-based AOSs have shown excellent performance, they have disadvantages including toxicity, element scarcity, and In extraction in hydrogen plasma [18]. For these reasons, there is a need for In- and Ga-free oxide semiconductors that are inexpensive and nontoxic. As a result, Sn-doped AOSs, such as ZTO [12] and Hf-Zn-Sn-O (HZTO) [15], have been studied. These materials are appealing because tin ions (Sn^{4+}:$4d^{10}5s^0$) have orbitals similar to those of indium ions (In^{3+}:$4d^{10}5s^0$) and a high mobility expected [19] (see Table 3).

As mentioned earlier, many researchers have studied the zinc oxide-based amorphous oxide semiconductor thin film transistors by various channel materials and deposition method as summarized in Table 1. Additionally, solution-based methods such as spin-coated and printed AOSs are actively being studied as low-cost processes for the formation of oxide semiconductors [20]. For stable TFT performance, the qualities of the active channel materials must be excellent. In particular, the number of n-type carriers (mainly electrons) and trap sites must be carefully controlled. These properties are normally influenced by controlling processing conditions such as the O_2 partial pressure during deposition, the post annealing treatment [21,22], and by varying the film stoichiometry [23]. In the present work, we report hafnium–tin–zinc oxide (HTZO) semiconductor layers and demonstrated their application for active channel layers in order to resolve negative bias temperature instability (NBTI) issues associated with zinc oxide-based TFTs. Devices were formed with HTZO films deposited in a double-layer structure to investigate the origin of the negative bias stability improvement. To make these, an HTZO film with a thickness of 10 nm was first deposited onto the gate insulator (GI). Next, a 30 nm thick ZTO film was grown on the HTZO. For comparison, a second structure was made by performing the depositions in the reverse order (HTZO 30 nm/ZTO 10 nm/GI). The Hf in the HTZO

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Table 1
Atomic compositions of HTZO thin films determined by XPS analysis.

O_2 in sputtering atmosphere	Elements			
	Hf	Sn	Zn	O
0% (pure Ar)	2.1	11.9	38.3	47.7
5%	2.0	12.1	36.5	49.4
10%	2.2	12.4	35.7	49.7

films helps control the carrier concentration by curbing carrier generation via oxygen vacancy formation.

2. Experimental

2.1. Film preparation and characterization

Thin films of HTZO (40 nm thick) were deposited using a direct current (DC) magnetron sputtering system. ZTO and HTZO (2.0 atomic % Hf) targets were prepared and deposited on the substrates. The $ZnO:SnO_2$ molecular ratio in the ZTO and HTZO targets was maintained at 3:1. The deposition temperature was about 25 °C (room temperature), and sputtering was conducted in an Ar atmosphere with an O_2 partial pressure ranging between 0% and 10%. A 4 in diameter target was placed 4 cm from the substrate, and a plasma glow discharge power density of 0.49 W/cm^2 was applied.

Using X-ray diffraction (XRD, D-2500, Rigaku), the crystal structure of the film was examined in the conventional θ-2θ mode. The microstructural properties of the film were analyzed with high-resolution transmission electron microscopy (HR-TEM, JEOL FB-2100F). The composition and chemical bonding states of the HTZO films were analyzed by X-ray photoelectron spectroscopy (XPS, ESCA lab 220, VG Microtech). This system uses Mg-Kα radiation ($h\upsilon = 1253.6$ eV) as the excitation source. The X-ray beam energy and filament current were 12 kV and 10 mA, respectively. The atomic ratio of Hf in the HTZO film, as determined by inductively coupled plasma Auger electron spectroscopy (ICP-AES), was 2.0 atomic %.

2.2. Device fabrication

Bottom-gate TFTs (Fig. 1) were fabricated using HTZO channels deposited on heavily doped n-type silicon substrates which had a 100 nm thick SiN_x layer deposited by low-pressure chemical vapor deposition (LPCVD) and a buffered gate insulator consisting of a 10 nm thick SiO_2 layer (plasma enhanced chemical vapor deposition (PECVD))/100 nm thick SiN_x layer (LPCVD) structure. The TFT channel (W/L = 1000 μm/100 μm) and source/drain electrodes were patterned using shadow masks. Molybdenum (300 nm) deposited by DC magnetron sputtering, was used as the source/drain electrode. Finally, the TFT devices were subjected to annealing at 400 °C for 1 h in ambient air. All current–voltage measurements of the TFTs were performed using a semiconductor parameter analyzer (Agilent HP 4145B) at room temperature in the dark. A series of measurements were examined as the gate-source voltage was swept from −40 V to 40 V with drain-source bias (V_{DS}) fixed at 1 V. In this study, I_{ON}/I_{OFF} was obtained by measuring the drain current as a function of V_{GS} at fixed V_{DS} of 1 V.

3. Results and discussion

The atomic concentration of the HTZO films formed under the different deposition gas conditions are listed in Table 1. Although there is no noticeable variation in the Hf and O percentages in the films, the Zn and Sn contents did vary depending on the gas

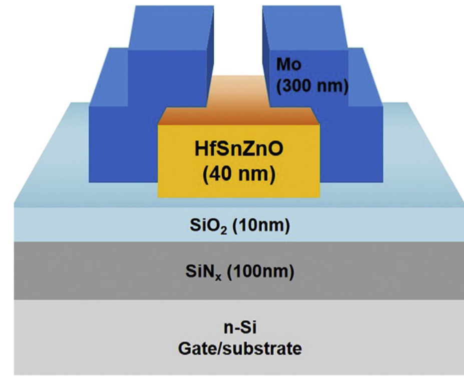
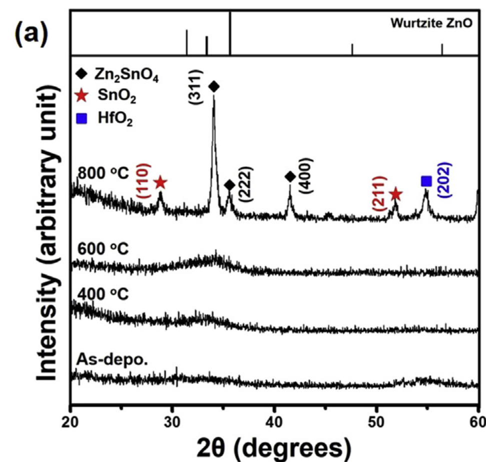


Fig. 1. Cross-sectional schematic of a bottom-gate-type HTZO TFT with a staggered structure.

conditions. This can be explained by the fact that the sputtering yield of the metal oxide ceramic target varied depending on the conditions of the sputtering chamber. XPS analyses show that the HTZO films are Zn-rich and oxygen-deficient, creating n-type carriers. These values are not noticeably affected by the sputtering conditions.

Fig. 2(a) shows the X-ray diffraction patterns of the HTZO films deposited with an Ar/O_2 ratio of 9:1 as a function of different post-annealing conditions (room temperature, 400, 600, or 800 °C for 1 h). The amorphous structure of the films was found to be stable up to a post-annealing temperature of 600 °C. The HTZO films annealed at 800 °C exhibited a crystalline phase. As the heat



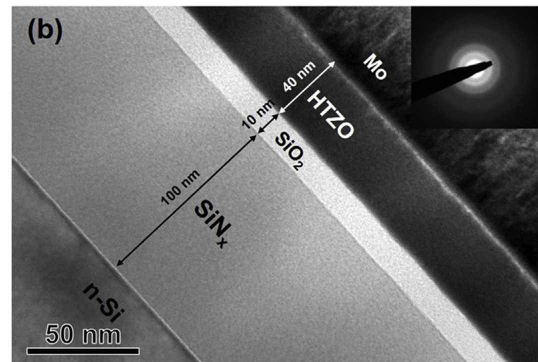


Fig. 2. (a) XRD patterns of HTZO films post-annealed at 400, 600, and 800 °C compared to the as-deposited film. (b) Cross-sectional HR-TEM images of the bottom-gate-type HTZO TFT with a staggered structure.

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