



# Article Influence of Active Channel Layer Thickness on SnO<sub>2</sub> Thin-Film Transistor Performance

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**Abstract:** Sol-gel processed SnO<sub>2</sub> thin-film transistors (TFTs) were fabricated on SiO<sub>2</sub>/p<sub>+</sub> Si substrates. The SnO<sub>2</sub> active channel layer was deposited by the sol-gel spin coating method. Precursor concentration influenced the film thickness and surface roughness. As the concentration of the precursor was increased, the deposited films were thicker and smoother. The device performance was influenced by the thickness and roughness of the SnO<sub>2</sub> active channel layer. Decreased precursor concentration resulted in a fabricated device with lower field-effect mobility, larger subthreshold swing (*SS*), and increased threshold voltage (*V*<sub>th</sub>), originating from the lower free carrier concentration and increase in trap sites. The fabricated SnO<sub>2</sub> TFTs, with an optimized 0.030 M precursor, had a field-effect mobility of 9.38 cm<sup>2</sup>/Vs, an *SS* of 1.99, an *I*<sub>on</sub>/*I*<sub>off</sub> value of ~4.0 × 10<sup>7</sup>, and showed enhancement mode operation and positive *V*<sub>th</sub>, equal to 9.83 V.

Keywords: sol-gel; thickness; precursor concentration; SnO<sub>2</sub>; thin-film transistor



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# 1. Introduction

Metal-oxide semiconductors are promising channel layer materials for thin-film transistors (TFTs). Their field-effect mobility is higher than that of amorphous Si, and it is well known that amorphous-phase metal-oxide semiconductors have the advantage of uniform electrical properties compared to poly-Si semiconductors [1-6]. Currently, vacuum-based fabrication processes are typically used to fabricate devices. However, vacuum-based fabrication processes are cost-intensive. They require complex equipment and are not suitable processes for large-area applications. However, to deposit metal-oxide semiconductors, there are many useful processes for large-area applications, such as spin casting, printing, and the sol-gel process [7-13]. There are three representative n-type semiconductors fabricated by these processes: ZnO, In<sub>2</sub>O<sub>3</sub>, and SnO<sub>2</sub>. In particular, indium-based metal oxide has received much attention due to its excellent electrical properties, which originate from its electronic configuration (1s<sup>2</sup>2s<sup>2</sup>p<sup>6</sup>3s<sup>2</sup>p<sup>6</sup>d<sup>10</sup>4s<sup>2</sup>p<sup>6</sup>d<sup>10</sup>5s<sup>2</sup>p<sup>1</sup>). Unfortunately, indium is a rare metal and mined in only a few locations worldwide. Therefore,  $SnO_2$  is an attractive choice as an indium-free, abundant, metal-oxide semiconductor. SnO<sub>2</sub> shows the highest mobility, a large optical bandgap—which is required for highly transparent applications—and a relatively lower melting point compared to ZnO and In<sub>2</sub>O<sub>3</sub>, making crystallization easier and related to high conductivity [14]. These advantages are useful for realizing high-performance transparent electronic devices [15–20]. In this work, solgel processed SnO<sub>2</sub> TFTs were fabricated. Precursor concentration was shown to impact SnO<sub>2</sub> film thickness and film uniformity. The changes in film thickness and uniformity affected device performance, such as field effect mobility in the saturation regime and the subthreshold swing (SS). A thick  $SnO_2$  active channel layer was formed with a high

precursor concentration. The fabricated SnO<sub>2</sub> thin-film transistor, consisting of thick films, showed high performance, including a field-effect mobility of 10.83 cm<sup>2</sup>/Vs, on/off current ratio of ~ $6.7 \times 10^7$ , and SS of 1.04. In addition, the optimized precursor concentration could successfully control the film thickness and carrier concentration. SnO<sub>2</sub> TFTs, made of 0.030 M precursor, showed enhancement mode operation and good electrical properties, including a field-effect mobility of 9.38 cm<sup>2</sup>/Vs, an on/off current ratio of ~ $4.0 \times 10^7$ , and an SS of 1.99, at the same time.

### 2. Materials and Methods

In this experiment, tin (II) chloride dihydrate (SnCl<sub>2</sub>·2H<sub>2</sub>O) was purchased from Sigma Aldrich. To make the precursor solution, we dissolved tin (II) chloride dihydrate into ethanol in ambient air at room temperature. Precursor solutions were made in three different concentrations: 0.020 M, 0.030 M, and 0.033 M. To fabricate bottom-gate coplanar type structures, a heavily doped p-type silicon wafer, which had a thermally grown 100-nm-thick  $SiO_2$  dielectric layer, was used. An e-beam evaporator was used to deposit a 50-nm-thick Au source/drain. The width and height of the electrodes were 1000  $\mu$ m and 100 µm, respectively. After dicing the substrate into small pieces, the pieces were blown by N<sub>2</sub> and cleaned by a UV/ozone cleaner (SENLights SSP16-110) for 3 h to eliminate organic impurities and to improve adhesion. The solutions, prepared with different concentrations, were spin-coated at 3000 rpm for 50 sec. The spin-coated devices were soft-baked on a hotplate (CORNING PC-420D) at 150 °C for 10 min to dry out the ethanol. Then, the devices were annealed at 500 °C for 2 h in a furnace (U1Tech PTF-1203). After cooling down, the  $SnO_2$  film active channel layer was patterned by mechanical removal to avoid the fringing effect and gate leakage current. Transmission electron microscopy (TEM: Titan G2 ChemiSTEM Cs Probe) was used to confirm the structural properties of the device, such as the interface roughness, surface roughness, and film thickness. The cross-sectional TEM specimens were fabricated by focused ion beam. The optical properties needed for the calculation of the optical band gap were obtained through ultraviolet-visible spectroscopy (UV-Vis; LAMBDA 265) measurements. To compare the chemical composition of the SnO<sub>2</sub> films, X-ray photoelectron spectroscopy (XPS: ULVAC-PHI) was used. The electrical characteristics of the devices were investigated using a probe station (MST T-4000A) in ambient air with a KEITHLEY 2636B Source Meter.

#### 3. Results and Discussion

Figure 1a–c show cross-sectional TEM images of SnO<sub>2</sub> films with a carbon protective layer as a function of different precursor concentrations. The estimated film thickness was below 6 nm for all precursor concentrations. All deposited films showed clearly regular atomic spacing, which indicated that the deposited films were in the polycrystalline phase [21]. Figure 1d shows a schematic diagram illustrating the definition of parameters to estimate the film thickness uniformity. The horizontal line through the profile represents the mean height.  $\Delta T$  was defined as the maximum height of the profile above the mean height line. The estimated *T* and  $\Delta T$  values are plotted in Figure 1e,*f*, respectively. Figure 1e reveals that film thickness increased with the increasing concentration of the precursor, from 0.020 M to 0.033 M. These results are well-matched with the previous report of W. W. Flack et al., who demonstrated that an increase in the concentration of the precursor results in thicker films. Increased concentration leads to an increase in viscosity and convective radical flow [22]. However, in terms of  $\Delta T$ , thicker films, with higher precursor concentrations, show lower  $\Delta T$  values, which indicates smooth and uniform film formation.



Figure 1. Cont.



**Figure 1.** (**a**–**c**) Cross-sectional TEM images of 0.020, 0.030, and 0.033 M SnO<sub>2</sub> films, respectively. (**d**) Schematic diagram illustrating the definition of parameters and estimated (**e**) average thickness and (**f**)  $\Delta T$  parameters.

The optical properties of  $SnO_2$  films, as a function of precursor concentration, were investigated by UV-VIS spectroscopy. Figure 2a,b give the transmittance and absorbance spectra, respectively, of  $SnO_2$  thin films as a function of precursor concentration. The obtained transmission results for all  $SnO_2$  films, ranging from 250 to 900 nm, revealed higher transmittance in the visible range. All  $SnO_2$  films showed high transmittance, over 97.0%, in the visible range. These values are adequate for the realization of a solar cell module, which requires over 95% transmittance. The dramatic drop in the ultraviolet regime, around 350 nm, originates from bandgap absorption, which has a value close to the intrinsic bandgap of  $SnO_2$  (3.60 eV). The energy band gap values were obtained by extrapolating the line segment in Figure 2c. To extrapolate the line segment, the following equation is used:

$$(\alpha h\nu)^{1/n} = A(h\nu - E_g), \tag{1}$$

where  $\alpha$  is the absorption coefficient, *A* is a constant, and  $E_g$  is the bandgap of SnO<sub>2</sub> films. The value 0.5 (direct transition) is used for the power factor (*n*). The obtained bandgap values are 4.24 eV, 3.86 eV, and 3.82 eV for 0.020 M, 0.030 M, and 0.033 M, respectively. The value 2 (indirect transition) is used for the power factor (*n*). The obtained bandgap values are 3.63 eV, 3.28 eV, and 3.15 eV for 0.020 M, 0.030 M, and 0.033 M, respectively. These values are larger than that of bulk SnO<sub>2</sub>. The increased bandgap with decreasing SnO<sub>2</sub> film thickness originates from the quantum confinement effect since the thickness is close to the Bohr radius. It is well known that the Bohr radius of SnO<sub>2</sub> is 2.7 nm. The change in energy bandgap can be expected by using the following equation [23–25]:

$$\Delta E_g = \frac{h^2}{8t^2} \left( \frac{1}{m_e^*} + \frac{1}{m_h^*} \right), \tag{2}$$

where *h* is Plank's constant, *t* is the film thickness,  $m_e^*$  is the effective mass of electrons, and  $m_h^*$  is the effective mass of holes. The calculated bandgap values are 4.23 eV, 3.93 eV, and 3.9 eV, showing a similar trend to the experimentally obtained values of energy quantization. The reduced film thickness affects the physical properties due to the energy quantization phenomena.

XPS analysis of SnO<sub>2</sub> thin films as a function of precursor concentrations was conducted to determine the chemical state of the elements. Figure 3a–c show the O1s spectra. All SnO<sub>2</sub> films show three components. Three peaks, occurring at 529.8 eV, 530.7 eV, and 531.9 eV corresponded to the oxygen ions ( $O^{2-}$ ) ( $O_{Lattice}$ ) combined with metal cations, the oxygen vacancy ( $O_{Vacancy}$ ) in the structure, and the hydroxyl groups (-OH), respectively. Figure 3d shows the relative contents of the  $O_{Lattice}$ ,  $O_{Vacancy}$ , and –OH of SnO<sub>2</sub> thin films after the annealing process. As the film thickness is decreased, it is noted that the amount of the oxygen vacancy is increased. Based on the cross-sectional TEM images, the thinnest SnO<sub>2</sub> film, made of a low precursor concentration, showed rougher surface characteristics and an increase in air-exposed surface area during the annealing process. It is well known that the increase of the exposure area accelerates the increase of oxygen vacancy formation [26].

Figure 4 shows the representative output curve and transfer curve of the fabricated  $SnO_2$  TFTs as a function of precursor concentration. All the fabricated  $SnO_2$  TFTs show conventional n-type semiconductor properties. All output curves show a similar tendency, except for the value of drain current. All four graphs exhibit non-linear properties at low drain voltage, indicating that a Schottky contact is formed between the source/drain electrode and the Au electrode. This originates from the high work function of Au [9]. The electrical characteristics of the fabricated  $SnO_2$  TFTs were estimated by extracting the representative four parameters, such as field-effect mobility in the saturation regime, subthreshold swing (*SS*), on/off current ratio, and threshold voltage ( $V_{th}$ ). The formulas used for analysis are

$$I_D = \mu C_i \frac{W}{2L} (V_G - V_{th})^2$$
(3)

and

$$SS = \left(\frac{\partial log I_D}{\partial V_G}\right)^{-1} \tag{4}$$

where  $C_i$  (insulator capacitance) =  $3.45 \times 10^{-8}$  F/cm<sup>2</sup>, W (channel width) = 1000 µm, and L (channel length) = 100 µm. For precursor concentrations of 0.020 M, 0.030 M, and 0.033 M, the field-effect mobilities were 1.01 cm<sup>2</sup>/Vs, 9.38 cm<sup>2</sup>/Vs, and 10.83 cm<sup>2</sup>/Vs, *SS* values were 2.51 V/decade, 1.99 V/decade, and 1.04 V/decade, and on/off ratios were  $1.5 \times 10^6$ ,  $4.0 \times 10^7$ , and  $6.7 \times 10^7$ , respectively. The extracted  $V_{th}$  was 14.9 V, 9.83 V, and -6.77 V for precursor concentrations of 0.020 M, 0.030 M, and 0.033 M, respectively. In order to check the statistical distribution of performance parameters, we compared the extracted parameters of each TFTs (10 devices each). Extracted performance parameters are plotted in Figure 5.



**Figure 2.** (a) Transmittance of the prepared SnO<sub>2</sub> films; the inset shows absorbance spectra. (b)  $(ahv)^2$  vs. photon energy and (c)  $(ahv)^{0.5}$  vs. photon energy for prepared SnO<sub>2</sub> films. (d) Energy bandgap as a function of the precursor concentrations.



Figure 3. (a–c) O1s XPS spectra of  $SnO_2$  thin films and (d) O1s composition as a function of precursor concentrations.



**Figure 4.** (**a**–**c**)  $I_D$ - $V_D$  curves and (**d**) transfer curves of SnO<sub>2</sub> thin-film transistors as a function of precursor concentrations.



**Figure 5.** Extracted thin-film transistor performance parameters: (**a**) field-effect mobility in saturation regime, (**b**) subthreshold swing (*SS*), (**c**)  $V_{th}$ , and (**d**)  $I_{on}$  and  $I_{off}$ .

The thicker films showed better performance with respect to field-effect mobility in the saturation regime, on/off current ratio, and SS. The field-effect mobility in the saturation regime increased with increasing precursor concentration. The increased precursor concentration leads to an increase in channel layer thickness. This phenomenon, the relationship between field-effect mobility and active channel thickness, is similar to the results for other metal-oxide and amorphous-silicon-based TFTs. The free carrier concentration per unit accumulation area is proportional to the active channel layer thickness. Thicker films made with higher precursor concentrations have higher free carrier concentrations and show increased film conductivity [27,28]. This results in strong n-type semiconductor properties with high electron carrier concentrations. The increased induced carrier concentration shifts  $V_{th}$  to negative values, causing the transistors to turn on early. In addition, fabricated SnO<sub>2</sub> TFTs consisting of thicker channel layers exhibit a lower value of SS. It is well known that the SS values are strongly affected by the trap density ( $N_t$ ) of the interfaces between the semiconductor and dielectric layers [29] by

$$N_t = \left[\frac{SSlog_{10}^{(e)}}{\frac{kT}{q}} - 1\right] \left(\frac{C_i}{q}\right),\tag{5}$$

where *SS* is the subthreshold swing, *k* is Boltzmann's constant, *T* is the temperature, *q* is the electron charge, and  $C_i$  is the capacitance of the dielectric layer. The SnO<sub>2</sub> TFTs consisting of a thinner channel layer show higher and poorer *SS* values, indicating the formation of a larger number of trap sites between the semiconductor and dielectric layers. The conductivity and the concentration of trap sites at the surface of the channel layer can be changed by the chemisorption process. It is well known that the adsorption of O<sub>2</sub> or H<sub>2</sub>O molecules on the back-channel of the metal oxide can change device performance. O<sub>2</sub> and H<sub>2</sub>O molecules can capture the electrons from the conduction band and form a depletion region in the back-channel, resulting in a positive  $V_{th}$  shift [30]. The following formulas illustrate the electron capture:

$$O_2 + e^- \to O_2^- \tag{6}$$

and

$$H_2O + O_2 \to H_2O^+ + O_2^-,$$
 (7)

which work as the surface trap center in the back-channel. Even though the chemisorption can be constant, regardless of film thickness, this phenomenon can more dominantly affect thinner films. In addition, the rougher surface of thinner films is more sensitive to this chemisorption, resulting in the degradation of device performance.

## 4. Conclusions

Sol-gel processed SnO<sub>2</sub> TFTs were fabricated on SiO<sub>2</sub>/p<sub>+</sub> Si substrates. The SnO<sub>2</sub> active channel layer was deposited by the sol-gel spin coating method. The precursor concentration influenced the film thickness and surface roughness. As the concentration of the precursor was increased, the deposited films were thicker and smoother. It was found that the device performance was influenced by the film thickness and roughness of the SnO<sub>2</sub> active channel layer. As the concentration of the precursor was decreased, the fabricated device showed lower field-effect mobility, larger subthreshold swing (*SS*), and increased threshold voltage ( $V_{th}$ ), originating from the lower free carrier concentration and increased trap sites. The fabricated SnO<sub>2</sub> TFTs with the optimum precursor concentration of 0.030 M had a field-effect mobility of 9.38 cm<sup>2</sup>/Vs, a subthreshold swing of 1.99, an  $I_{ont}/I_{off}$  value of ~4.0 × 10<sup>7</sup>, and showed enhancement mode operation, with positive  $V_{th}$  equal to 9.83 V.

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