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Description	

Combustion synthesized Indium-Tin-Oxide (ITO) thin film for Source/Drain electrodes in all solution-processed oxide thin film transistors

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We report combustion solution synthesized (SCS) indium-tin-oxide (ITO) thin film, which is a well-known transparent conductive oxide, for source/drain (S/D) electrodes in solution-processed amorphous zirconium-indium-zinc-oxide (ZIZO) TFT. A redox-based combustion synthetic approach is applied to ITO thin film using acetylacetone as a fuel and metal nitrate as oxidizer. The structural and electrical properties of SCS-ITO precursor solution and thin films were systematically investigated with changes of tin concentration, indium metal precursors, and annealing conditions such as temperature, time, and ambient. It was found that at optimal conditions the SCS-ITO thin film exhibited high crystalline quality, atomically smooth surface (RMS $\sim 4.1 \text{ \AA}$), and low electrical resistivity ($4.2 \times 10^{-4} \text{ \Omega cm}$). The TFT using SCS-ITO film as the S/D electrodes showed excellent electrical properties with negligible hysteresis. The obtained “on/off” current ratio, sub-threshold swing factor, sub-threshold voltage, and field-effect mobility were 5×10^7 , 0.43 V/decade, 0.7 V, and $2.1 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, respectively. The performance and stability of the SCS-ITO TFT are comparable to those of the sputtered-ITO TFT emphasizing that the SCS-ITO film is a promising candidate for totally solution-processed oxide TFTs.

1. Introduction

Metal oxide semiconductor thin film transistors (TFTs) have drawn much attention for use in switching devices for large-sized electronics due to their good uniformity, transparency and better device performance compared with conventional amorphous Si-based and organic TFTs [1-4]. However, high-cost vacuum-based manufacturing processes are the main obstacle for modern, mass-produced, and large electronics. In contrast, solution-based deposition techniques such as spin-coating and ink-jet printing offer many advantages such as simplicity, low-cost, high throughput, and direct pattern-ability, enabling the fabrication of low-cost printed electronics [5-21]. So far, researchers have mainly focused on solution processing for only the oxide semiconductor [5-19]. Nevertheless, in order to realize ultimate reduction of fabrication cost it is obviously that solution processing for the other components of a TFT such as gate insulator and source/drain (S/D) electrodes is indispensable. In particular, as for the S/D electrodes, conductive indium-tin-oxide (ITO) has been mainly used because of its excellent transparency, high electrical conductivity, and ohmic contact with oxide semiconducting channel layer [1, 22, 23]. Though a number of researches on solution-processed ITO films have ever tried, most of the films exhibit poor quality as compared to those prepared from vacuum-based techniques. Consequently, various attempts have been done to enhance the structural and electrical properties of ITO thin films fabricated by solution-based processes. For example, highly conductive ITO film ($313 \Omega^{-1}\text{cm}^{-1}$) was developed using a nanoparticle-based solution process through the control of oxygen partial pressure during annealing [24]. Addition of ITO crystalline particles to alkoxide sol resulted in a lower crystallization temperature and higher electrical conductivity [25]. ITO thin films prepared from alcoholic solutions of indium chloride and stannic chloride showed significant decrease in the resistance values after annealed in N_2/H_2 (96/4%) atmosphere ($\sim 11 \Omega/\square$) [26]. A sol-gel route assisted with an ultraviolet laser beam annealing was proposed to lower resistivity and annealing temperature ($6 \times 10^{-1} \Omega\text{cm}$ at $300 \text{ }^\circ\text{C}$) [27]. Annealing in reducing atmosphere such as Ar/H_2 (95/5%) and $\text{N}_2+\text{H}_2\text{O}$ vapor was also considered to improve electrical conductivity [28, 29]. Recently, solution combustion synthesis (SCS) was reported as a novel method for producing device-quality oxide semiconducting thin film at temperature as low as $200 \text{ }^\circ\text{C}$ [30]. The self-generated heat of combustion synthesis provides a localized energy supply, eliminating the need for high,

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externally applied processing temperatures. In addition, the atomically local oxidizer supply can efficiently remove organic impurities without coke formation. In this work, a redox-based combustion synthetic approach is applied to ITO thin film using acetylacetone as a fuel and metal nitrate as oxidizer. The structural and electrical properties of SCS-ITO precursor solution and thin films were systematically investigated with changes of tin concentration, indium metal precursors, and annealing conditions such as temperature, time, and ambient. After that, the optimized SCS-ITO thin film was applied for source/drain (S/D) electrodes in a total solution-processed amorphous oxide TFT. The performance and stability of the SCS-ITO TFT were evaluated and compared to those of the TFT with sputtered-ITO S/D electrodes.

2. Experimental methods

Synthesis of functional precursor solutions

A 0.2 mol/kg Zr-In-Zn-O (ZIZO) solution was synthesized by using the sol-gel method with indium (III) acetylacetonate [$\text{In}(\text{OCCH}_3\text{CHOCCCH}_3)_3$, Sigma-Aldrich], zinc chloride [ZnCl_2 , Kanto Chemical], and zirconium (IV) butoxide [$\text{Zr}(\text{OC}_4\text{H}_9)_4$, Sigma-Aldrich] dissolved in propionic acid solvent. The mole ratio of Zr/In/Zn was fixed at 0.05/2/1 based on previous study. A 0.4 mol/kg La-Zr-O (LZO) solution with the La/Zr mole ratio of 1/1 was prepared in propionic acid solvent using a mixture of lanthanum (III) acetate hydrate [$\text{La}(\text{O}_2\text{C}_2\text{H}_3)_3 \times 1.5 \text{ H}_2\text{O}$, Kanto Chemical, 98% purity] and zirconium (IV) butoxide [$\text{Zr}(\text{OC}_4\text{H}_9)_4$]. All the solutions were stirred at 1000 rpm at 110 °C for 30 minutes to form transparent and homogeneous solutions, and were then filtered through a 0.2 μm syringe filter [poly(tetrafluoroethylene), GE]. A commercially available polysilazane-based solution (NP110-20, AZ Electronic Materials) consisting of a basic unit of $(\text{H}_2\text{Si-NH})_n$ was used as the precursor material for an etch stopper layer. A 0.6 mol/kg In_2O_3 solution was synthesized by dissolving indium nitrate trihydrate [$\text{In}(\text{NO}_3)_3 \cdot 3\text{H}_2\text{O}$, Kanto Chemical] in 2-methoxyethanol (2ME). Then, acetylacetone and ammonium nitrate are added to the solution to serve as fuel and oxidizer, respectively. A 0.6 mol/kg SnCl_2 solution was prepared by dissolving tin chloride in 2ME. The solution combustion synthesized (SCS)-ITO solutions were prepared by mixing In_2O_3 and SnCl_2 solutions with different ratios such as 90/10, 92/8, 95/5, and 97/3 wt%. As a control experiment, two conventional indium precursor solutions,

which were synthesized by mixing indium (III) acetylacetonate [In-acac/PrA] and indium (III) nitrate trihydrate [In-NO/PrA] with propionic acid in the same manner as the SCS-ITO solution, were also prepared. SnCl₂ (5wt%) were added to these indium precursors to form ITO solutions.

Oxide TFT fabrication

The bottom-gate, top-contact structure was selected for fabrication of the TFTs. First, 100-nm-thick Pt/Ti was deposited on a glass substrate using RF-sputtering technique. Next, a 250-nm-thick LZO layer was spin-coated on the Pt/Ti substrate. Then, it was annealed at 550 °C for 10 min in oxygen ambient using rapid thermal annealing (RTA). Next, 20-nm-thick ZIZO active layer was deposited on the LZO layer by spin-coating, and annealed at 500 °C for 10 min in oxygen using RTA. The ZIZO layer was patterned by physical dry-etching technique using argon gas. As a next step, 100-nm-thick SiO₂ etch-stopper layer was deposited by spin-coating, followed by a wet-oxygen annealing at 500 °C for 30 min. The etch-stopper layer was patterned by reactive ion etching (RIE) technique using CF₄ gas. As S/D electrodes, 120-nm-thick ITO layer was deposited by spin-coating and annealed at 250 °C under low-pressure condition (14 Pa) for 30 min. The ITO S/D patterns were isolated by the physical dry-etching. Finally, 400-nm-thick SiO₂ film was spin-coated and annealed at 250 °C using silsesquioxane precursor solution for passivation layer.

Characterization

Thermal behaviors of ITO solutions were analyzed by thermal gravity-differential thermal analysis (TG-DTA) using a TG/DTA 6200 SII NanoTechnology Inc. system. The crystalline phases of the ITO thin films were analyzed using X-ray diffraction (XRD), with a Philips X'Pert system using Cu K α radiation. The sheet resistance values of the samples were measured at room temperature using the four-point probe (SDY-4) system. Surface morphology of the ITO film was characterized by atomic force microscope (AFM) using SPA-400 system. Device operation characterization was carried out at room temperature by the Agilent 4155C Semiconductor Parameter Analyzer.

3. Results and discussion

3.1 Investigation on SCS-ITO precursor solutions and thin film formation

3.1.1. ITO precursor solutions

Figure 1 shows thermal gravity (TG) and differential thermal analysis (DTA) of ITO

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solutions with different precursors. The DTA spectra of conventional ITO solution systems based on nitrate and acetylacetonate exhibited broad exothermic peaks at 316 and 344 °C, respectively, which can be attributed to dehydroxylation/crystallization process. On the other hand, the DTA spectrum of the SCS-ITO solution showed a single and intense exothermic peak at 188 °C corresponding to an abrupt weight loss in the TG spectrum. This could be attributed to a redox reaction between the fuel (acetylacetonate) and oxidizer (nitrate) in the precursor solution [30]. This reaction generates internal heat facilitating conversion from metal precursor to metal oxide. The combustion process eliminated the need for high external heat, which might lower process temperature below 200 °C.

3.1.2. The crystalline phases of the ITO thin films

The XRD patterns of ITO films (8wt% Sn) prepared at 500 °C for 30 min in vacuum using different precursor solutions including SCS-ITO, nitrate (In-NO/PrA), chloride (In-Cl/PrA), and acetylacetonate (In-acac/PrA)-based are shown in Fig. 2(a). Compared to the other films, the XRD pattern of the SCS-ITO film exhibited sharpest and highest peak intensity indicating that high crystalline quality was obtained. This could be attributed to effective conversion of the precursor to oxide film due to the internal combustion effect. Fig. 2(b) shows the XRD patterns of SCS-ITO films annealed at 250-500 °C for 30 min in vacuum as a function of Sn content (3, 5, 8, and 10wt%). We found that the intensity of the main (400222) peak initially increased as the Sn content rose up, and then decreased at the Sn content of 10wt%. This results from the fact that Sn⁴⁺ ions substitute for In³⁺ ones in crystal lattice providing more free-electron carriers which is a source of electrical conductivity [31]. The decrease of crystalline quality at 10wt% Sn is probably caused by an aggregation of excess Sn element [32].

The XRD patterns of SCS-ITO films (8wt% Sn) prepared at 500 °C as a function of annealing time (10-120 min) are shown in Fig. 2(c). We found that optimal annealing time was approximately 30 minutes. Longer annealing time might result in out-diffusion of Sn atoms from ITO lattice leading to decrease of crystallinity.

The XRD patterns of SCS-ITO films (8wt% Sn) prepared at different annealing temperatures (200-600 °C) for 30 min are summarized in Fig. 2(d). The characteristics 2θ lines at 30.8, 35.7, and 51.2° appeared as reflections from the (222), (400), and (440) planes, respectively. This matched well with the card PDF#65-3170 [33]. The XRD patterns show that the film

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annealed at 250-600 °C were polycrystalline with a cubic In₂O₃ structure. All of the patterns preferentially centered at 2θ at about 30.8°, which corresponded to the (222) plane of In₂O₃. No phases corresponding to tin oxide or to other tin compounds were detected in the XRD patterns. This can be attributed to the incorporation of Sn into the In₂O₃ lattice. Consequently, the ITO films with single- phase formed. With the increase of annealing temperature the crystalline structures of ITO films did not change. However, the crystallinity was improved as the annealing temperature increased and the size of In₂O₃ grains grew [34]. The 250 °C-annealed ITO film showed sufficient crystallinity for application to electrodes.

3.1.3. Morphology of the ITO thin films

Figure 3 shows the AFM images of SCS-ITO films as a function of annealing temperatures (200-600 °C). Atomically flat and uniform surface morphologies were observed in all cases. Elevation of the annealing temperature resulted in increases of grain size as well as surface roughness, indicating an improvement of crystallinity [33, 35]. However, the roughness mean square (RMS) value was less than 8.5 Å in all cases (particularly, being 4.1 Å for 250 °C-annealed ITO film). This tendency is consistent to what observed from the XRD patterns, and could be explained by the aggregation of native grains into larger clusters upon annealing. There was no significant difference observed in surface morphologies of the SCS-ITO films with respect to either the Sn content or the annealing time.

3.1.4. Electrical properties of the ITO thin films

Figure 4 (a) shows sheet resistance of the 70-nm-thick ITO films annealed in vacuum or nitrogen atmosphere as a function of annealing temperature (250-550 °C) for 10 minutes. Generally, the sheet resistance decreased as the annealing temperature increased in both cases of vacuum or nitrogen annealing due to an improvement of film crystallinity. Annealing under vacuum condition resulted in lower sheet resistance, which is originated from the fact that number of oxygen vacancies increased as annealed in vacuum [31, 36]. The lowest sheet resistance ($\sim 24 \Omega/\square$) could be obtained at the annealing temperature of 450 °C. Reduction of conductivity at higher annealing temperatures is because of the crystallinity degradation as observed from XRD. At above 450 °C, the conductivity were fluctuated which is inconsistent with observed XRD results. Probably, the actual content of Sn in the ITO films might be fluctuated due to the in/out diffusion of Sn element at the high annealing temperature, which led to the conductivity inconstancy of the resultant ITO films.

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To demonstrate merits of SCS process for low temperature processing, the dependence of 250 °C-annealed ITO film's sheet resistance on the annealing time is shown in Fig. 4 (b). The sheet resistance initially decreased as the time increased, reached minimum at 30-min-annealing, then increased with longer annealing time, which is also consistent to the XRD result. The lowest sheet resistance ($\sim 81 \Omega/\square$) corresponding to a resistivity of $4.2 \times 10^{-4} \Omega\text{cm}$ was obtained under vacuum condition. This condition would be eligibly applied for S/D electrodes of TFTs.

3.2 Operation of oxide TFTs

In the next step, we adopted the SCS-ITO film for source/drain electrodes in a bottom-gate top-contact TFT structure. A schematic structure of the fabricated TFT is shown in Fig. 5 (a). Figure 5 shows the transfer and output characteristics of the fabricated oxide TFTs using SCS-ITO as the S/D electrodes. As for the transfer curve (Fig. 5 (b)), gate voltage was scanning from -15 V to 20 V while a constant source-drain voltage of 15 V was applied. A typical n-type transistor characteristic was obtained. The TFT showed excellent electrical properties with negligible hysteresis. The obtained “on/off” current ratio, sub-threshold swing factor, sub-threshold voltage, and field-effect mobility were 5×10^7 , 0.43 V/decade, 0.7 V, and $2.1 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$, respectively. The output curve exhibited a good drain current saturation (Fig. 5 (c)). A linear increase of the drain current at small drain voltage indicated that ~~contact resistance~~ an ohmic contact between the SCS-ITO S/D electrodes and the ZIZO channel was ~~negligible~~ achieved. In other words, an ohmic contact was practically realized even though the S/D and channel layers were formed by the solution process. The TFT using sputtered-ITO for S/D electrodes was also fabricated for comparison. It was found that the performance of the SCS-ITO TFT is comparable to that of the sputtered-ITO TFT.

The effect of applying a gate-bias stress of 15 V to the gate contact for 10^4 s is shown in Fig. 6 (a). There was a positive displacement (0.42 V) of the transfer characteristic along the voltage axis. On the other hand, when a bias of -15 V was applied to the gate, the characteristics shift in the negative direction (5.45 V) (Fig. 6 (b)). However, once the stress bias was removed, the devices under test recover their original performance after approximately 60 min at room temperature. These device characteristics indicate that, during the periods of stress, both electrons and holes, depending on the polarity of the applied bias, are temporarily trapped in the gate insulator or at the channel/insulator interface. Upon the

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removal of the stress, these carriers are then released gradually from the trap states, resulting in the fresh characteristics that are again being obtained [37]. Furthermore, a logarithmic time-dependent threshold-voltage shift was observed. Based on these observations, it suggests that temporary charge trapping at fast interface states or in shallow traps in the oxide may be the cause of the device instability [38-41]. For comparison, the same experiments were done with the sputtered-ITO TFT. The threshold-voltage shifts were observed to be 2.37 V and 7.86 V for the positive and negative biases, respectively. These results indicate that the use of the SCS-ITO for S/D electrodes would not degrade the device performance stability.

4. Conclusions

A redox-based combustion synthetic approach is applied to ITO thin film using acetylacetone as a fuel and metal nitrate as oxidizer. The structural and electrical properties of SCS-ITO precursor solution and thin films were systematically investigated with changes of tin concentration, indium metal precursors, and annealing conditions such as temperature, time, and ambient. It was found that at optimal conditions the SCS-ITO thin film exhibited high crystalline quality, atomically smooth surface (RMS $\sim 4.1\text{\AA}$), and low electrical resistivity ($4.2 \times 10^{-4} \text{ }\Omega\text{cm}$). The TFT using SCS-ITO film as the S/D electrodes showed excellent electrical properties with negligible hysteresis. The obtained “on/off” current ratio, sub-threshold swing factor, sub-threshold voltage, and field-effect mobility were 5×10^7 , 0.43 V/decade, 0.7 V, and $2.1 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$, respectively. The performance and stability of the SCS-ITO TFT are comparable to those of the sputtered-ITO TFT emphasizing that the SCS-ITO film is a promising candidate for totally solution-processed oxide TFTs.

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Figure Captions

Fig. 1. (Color online) (a) Thermal Gravity (TG) and (b) Differential Thermal Analysis (DTA) of ITO solutions with different precursors.

Fig. 2. (Color online) XRD patterns of the SCS-ITO films as a function of: (a) ITO precursor solutions, (b) Sn content, (c) annealing time, and (d) annealing temperature.

Fig. 3. (Color online) AFM images of the SCS-ITO films as a function of annealing temperatures (200-600 °C). The films were annealed in vacuum for 30 min.

Fig. 4. (Color online) Sheet resistance of the SCS-ITO films as a function of: (a) annealing temperature and (b) annealing time.

Fig. 5. (a) Cross-sectional structure, (b) transfer characteristics, and (c) output characteristics of the fabricated oxide TFTs.

Fig. 6. (a) Positive bias stress (PBS) and (b) Negative bias stress of the fabricated SCS-ITO TFTs.