High performance In2O3 thin film transistors using chemically derived aluminum oxide dielectric
Pradipta K. Nayak, M. N. Hedhili, Dongkyu Cha, and H. N. Alshareef

Citation: Applied Physics Letters 103, 033518 (2013); doi: 10.1063/1.4816060
View online: http://dx.doi.org/10.1063/1.4816060
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/103/3?ver=pdfcov
Published by the AIP Publishing

Articles you may be interested in
Oxygen plasma assisted high performance solution-processed Al2Ox gate insulator for combustion-processed InGaZnOx thin film transistors

The effects of buffer layers on the performance and stability of flexible InGaZnO thin film transistors on polyimide substrates

Low-voltage InGaZnO thin-film transistors with Al 2 O 3 gate insulator grown by atomic layer deposition

High-performance InGaZnO thin-film transistors with high- k amorphous Ba 0.5 Sr 0.5 Ti O 3 gate insulator

GaN metal-oxide-semiconductor high-electron-mobility-transistor with atomic layer deposited Al 2 O 3 as gate dielectric
Appl. Phys. Lett. 86, 063501 (2005); 10.1063/1.1861122
High performance In$_2$O$_3$ thin film transistors using chemically derived aluminum oxide dielectric

Pradipta K. Nayak,¹ M. N. Hedhili,² Dongkyu Cha,² and H. N. Alshareef¹,a)

¹Materials Science and Engineering, King Abdullah University of Science and Technology (KAUST), Thuwal 23955-6900, Saudi Arabia
²Imaging and Characterization Laboratory, King Abdullah University of Science and Technology (KAUST), Thuwal 23955-6900, Saudi Arabia

(Received 8 June 2013; accepted 1 July 2013; published online 18 July 2013)

We report high performance solution-deposited indium oxide thin film transistors with field-effect mobility of 127 cm$^2$/Vs and an $I_{on}/I_{off}$ ratio of $10^6$. This excellent performance is achieved by controlling the hydroxyl group content in chemically derived aluminum oxide (AlO$_x$) thin-film dielectrics. The AlO$_x$ films annealed in the temperature range of 250–350°C showed higher amount of Al-OH groups compared to the films annealed at 500°C, and correspondingly higher mobility. It is proposed that the presence of Al-OH groups at the AlO$_x$ surface facilitates unintentional Al-doping and efficient oxidation of the indium oxide channel layer, leading to improved device performance. © 2013 AIP Publishing LLC.

Over the past 10 years, transparent oxide thin film transistors (TFTs) have been extensively studied due to their potential use in large area flat-panel display (FPD) applications. Oxide semiconductor based TFTs have been proven to be the alternative candidates to the low mobility silicon-based TFTs, which are currently being used for display applications.⁴ High performance oxide TFTs have been reported using vacuum deposition techniques²–⁵ and have also been used for the demonstration of display devices.⁶,⁷ Solution deposition techniques have also been employed for the fabrication of oxide TFTs. During the last few years, zinc oxide,⁸,⁹ indium oxide,¹⁰,¹¹ zinc tin oxide,¹²,¹³ indium zinc oxide,¹⁴,¹⁵ and indium gallium zinc oxide¹⁶–¹⁸ based thin film transistors with reasonable performance have been reported using spin coating technique. It is noted that, in most of the previously reports on solution processed TFTs, high annealing temperatures have been used to achieve better device performances and the TFTs are operated in very high operating voltage ranges. In practical, however, TFTs prepared at low processing temperatures with high field-effect mobility, high $I_{on}/I_{off}$ ratio, low turn-on voltage, and small sub-threshold swing values are needed for low-voltage operation and faster switching applications. The use of appropriate metal precursors, solvents, and appropriate gate dielectric are very crucial parameters to enable low process temperatures and good device performances. Low voltage operated TFTs with good performance have been reported using solution-processed gate dielectrics such as zirconium oxide¹³ and sodium beta-alumina.¹⁹ However, the processing temperature of typical dielectric layers is very high in these cases (>500°C), which is not suitable to be used for practical display applications.

In the present work, we demonstrated the fabrication of high performance and low-voltage operated TFTs using solution processed In$_2$O$_3$ as the semiconducting layer and aluminium oxide (AlO$_x$) as the gate dielectrics at significantly lower temperature. We found that the aluminium hydroxide content present in spin coated aluminium oxide dielectrics plays an important role to obtain high performance In$_2$O$_3$ TFTs.

The precursor solution for the preparation of AlO$_x$ films was prepared by dissolving aluminum chloride (0.5 M) in a mixture of acetonitrile (AN) and ethylene glycol (EG) with AN to EG ratio of 3:7. The mixed solutions were stirred at room temperature in air for 4 h. Heavily doped p-type silicon wafers were used as the gate electrode and substrate. The Si substrates were cleaned ultrasonically by ethanol, acetone, and isopropanol and then exposed to oxygen plasma for 2 min to enhance the hydrophilicity. The AlO$_x$ precursor solution was spun on the cleaned Si surfaces at a rotation speed of 3000 rpm in air. After spin coating, the wet films were placed on a hot plate at 90°C for 5 min and then subjected to rapid annealing at the temperature range of 250-500°C in air for 10 min. The spin coating and annealing process was repeated for three times. Finally, the AlO$_x$ films were annealed in the temperature range of 250-500°C for 2 h in air. The AlO$_x$ films prepared at 250°C, 350°C, and 500°C, hereafter will be called as AlO$_x$-250°C, AlO$_x$-350°C, and AlO$_x$-500°C, respectively. The average thickness of the AlO$_x$-250°C, AlO$_x$-350°C, and AlO$_x$-500°C were found to be ~120 nm, ~115 nm, and ~95 nm, respectively. Indium chloride (0.05 M) dissolved in acetonitrile and ethylene glycol (9:1 v/v) was used as the precursor solution for the preparation of In$_2$O$_3$ semiconductor channel layer. The mixed solution was stirred at ~50°C for 1 h. One layer of the indium oxide precursor solution was spun on AlO$_x$ films at a rotation speed of 4000 rpm in air. The wet films were kept on a hot plate at 80°C for 5 min and then subjected to rapid annealing at 250°C for 1 h in air using a tube furnace. Circular shaped aluminum contacts (thickness ~80 nm) with diameter of 100 μm were used for the capacitance and leakage current measurements of the AlO$_x$ films. Aluminum source and drain electrodes with channel width to length ratio (W/L) of 10 (W = 1000 μm and L = 100 μm) were used for the transistor characteristic measurements. The aluminum contacts were deposited using shadow mask and e-beam evaporation. Crystallinity of the AlO$_x$ and In$_2$O$_3$ films were studied by x-ray diffraction (XRD) diffractometer (Bruker D8...
AlOx films prepared at different annealing temperatures are gated by XPS measurements. The XPS O1s peaks of the films prepared at different annealing temperatures are shown in Fig. 1(a). No peak corresponding to the crystalline Al2O3 was observed, suggesting amorphous AlOx films were obtained over the entire range of annealing temperatures. The AFM images of the corresponding AlOx films are shown in Figs. 1(b)–1(d). The interface roughness of the gate dielectric is one of the crucial parameter to reduce carrier scattering and obtain high field-effect mobility. The root mean square surface roughness of the AlOx-250°C, AlOx-350°C, and AlOx-500°C was measured to be 0.3 nm, 0.2 nm, and 0.2 nm, respectively, and the obtained low surface roughness values are ideal for obtaining good device performances.

The chemical composition of the AlOx films was investigated by XPS measurements. The XPS O1s peaks of the AlOx films prepared at different annealing temperatures are shown in Fig. 2(a). The open circles show the measured data and the solid lines represents the Gaussian peak fitting results. The deconvoluted results of O1s peaks exhibited a peak at 531.0 eV, attributed to the O 2- in Al2O3 and another peak at 532.3 eV, attributed to oxygen associated to the OH- in aluminum hydroxide. The intensity of the two peaks at 531.0 eV and 532.3 eV were comparable in case of AlOx-250°C. Interestingly, the intensity of the peak at 532.3 eV decreased and subsequently, the intensity of the peak at 531.0 eV increased with increasing annealing temperature above 250°C. The decreased intensity of the peak at 532.3 eV may be attributed to the conversion of more aluminum hydroxides to form Al2O3 at higher temperatures. It may be noted here that the associated OH groups are mainly due to the water absorbed from the environment in the AlOx films after the final annealing. At low annealing temperatures, all the aluminum atoms are not completely oxidized and hence they absorb water molecules from the environment to form aluminum hydroxides. Fig. 2(b) shows the Al2p peaks of the AlOx films prepared at different annealing temperatures. The XPS spectra show a singlet Al2p peak at ~74.3 eV in case of AlOx-250°C and AlOx-350°C, whereas, it was shifted to 74.2 eV in case of AlOx-500°C. The shifting of the Al2p peak towards lower binding energy in case of AlOx-500°C may be attributed to the decrease in the concentration of OH- ions or coordination number of Al3+ ions in the film.

The schematic of the TFT structure used in this work is shown in Fig. 3(a). Figs. 3(c)–3(e) show the out-put characteristics of the fabricated In2O3 TFTs using AlOx-250°C, AlOx-350°C, and AlOx-500°C as the gate dielectrics. The capacitance of the aforementioned p+-Si/AlOx/Al structures were measured in the frequency range of 1 kHz to 1 MHz and the obtained curves are shown in the inset of Fig. 2(c). The measured capacitance at 1 MHz for AlOx-250°C, AlOx-350°C, and AlOx-500°C were found to be 66 nF/cm², 75 nF/cm², and 58 nF/cm², respectively. The high value of capacitance in case of the low temperature annealed AlOx films may attribute to the presence of OH-groups.

The schematic of the TFT structure used in this work is shown in Fig. 3(a). Figs. 3(c)–3(e) show the out-put characteristics of the fabricated In2O3 TFTs using AlOx-250°C, AlOx-350°C, and AlOx-500°C as the gate dielectrics. The capacitance of the aforementioned p+-Si/AlOx/Al structures were measured in the frequency range of 1 kHz to 1 MHz and the obtained curves are shown in the inset of Fig. 2(c). The measured capacitance at 1 MHz for AlOx-250°C, AlOx-350°C, and AlOx-500°C were found to be 66 nF/cm², 75 nF/cm², and 58 nF/cm², respectively. The high value of capacitance in case of the low temperature annealed AlOx films may attribute to the presence of OH-groups.
AlOx-250°C as evident from the I-V curve shown in Fig. 2(c). As seen from Table I, the TFTs fabricated using AlOx-500°C as the gate dielectric showed very inferior performance compared to the TFTs fabricated on AlOx-250°C and AlOx-350°C.

Material characterizations of the In2O3 films were performed to understand the effect of annealing temperature of AlOx thin film dielectrics on the performance of In2O3 TFTs. The crystallinity of the In2O3 films was investigated by XRD using grazing incidence geometry with an incident angle of 1°. The grazing incidence XRD patterns (Fig. 4(a)) of the In2O3 films deposited on different AlOx films showed a broad peak at ~30.5° corresponding to the (222) plane of In2O3. The Gaussian peak fitting of the observed peaks revealed that the full width at half maximum (FWHM) for the In2O3 films deposited on AlOx-250°C (FWHM = 2.5°) and AlOx-350°C (FWHM = 2.8°) are higher than that of the In2O3 film deposited on AlOx-500°C (FWHM = 2.1°). Furthermore, Fig. 4(b) shows the XPS O1s spectra of the In2O3 films deposited on AlOx films annealed at different temperatures. The Gaussian fitting (solid lines) of all the O1s peaks exhibited a strong peak at 529.8 eV along with two shoulder peaks at 531.1 eV and 532.3 eV. The peak at 529.8 eV is attributed to the oxygen in In2O3 lattice without oxygen vacancy (Ov).23 The peaks at 531.1 eV and 532.3 eV are attributed to the oxygen in In2O3 lattice with oxygen vacancy (OV) and the OH groups formed to understand the effect of annealing temperature of AlOx thin film dielectrics.

### Table I. Electrical parameters of In2O3 TFTs fabricated using AlOx films annealed at different temperatures as the gate dielectrics.

<table>
<thead>
<tr>
<th>AlOx annealing temperature</th>
<th>OV (%)</th>
<th>$\mu_{sat}$ (cm²/Vs)</th>
<th>$I_{on}/I_{off}$</th>
<th>$V_{on}$ (V)</th>
<th>SS (V/dec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>250°C</td>
<td>24%</td>
<td>82</td>
<td>$3 \times 10^{4}$</td>
<td>-0.1</td>
<td>0.271</td>
</tr>
<tr>
<td>350°C</td>
<td>28%</td>
<td>127</td>
<td>$1 \times 10^{6}$</td>
<td>0.1</td>
<td>0.137</td>
</tr>
<tr>
<td>500°C</td>
<td>31%</td>
<td>5</td>
<td>$2 \times 10^{4}$</td>
<td>-2.6</td>
<td>0.951</td>
</tr>
</tbody>
</table>

FIG. 3. (a) Schematic of the TFT structure, (b) transfer characteristic curves of the In2O3 TFTs fabricated using AlOx films prepared at different temperatures as the gate dielectrics. Out-put characteristic curves of the In2O3 TFTs fabricated using (c) AlOx-250°C, (d) AlOx-350°C, and (e) AlOx-500°C as the gate dielectrics. The inset in (e) shows the $I_D$ vs $V_D$ curves measured at zero gate voltage for the TFTs fabricated using AlOx dielectrics annealed at different temperatures.

FIG. 4. (a) XRD patterns and (b) O1s XPS spectra of the In2O3 films deposited on different AlOx films annealed at different temperatures.

TABLE I. Electrical parameters of In2O3 TFTs fabricated using AlOx films annealed at different temperatures as the gate dielectrics.

<table>
<thead>
<tr>
<th>AlOx annealing temperature</th>
<th>OV (%)</th>
<th>$\mu_{sat}$ (cm²/Vs)</th>
<th>$I_{on}/I_{off}$</th>
<th>$V_{on}$ (V)</th>
<th>SS (V/dec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>250°C</td>
<td>24%</td>
<td>82</td>
<td>$3 \times 10^{4}$</td>
<td>-0.1</td>
<td>0.271</td>
</tr>
<tr>
<td>350°C</td>
<td>28%</td>
<td>127</td>
<td>$1 \times 10^{6}$</td>
<td>0.1</td>
<td>0.137</td>
</tr>
<tr>
<td>500°C</td>
<td>31%</td>
<td>5</td>
<td>$2 \times 10^{4}$</td>
<td>-2.6</td>
<td>0.951</td>
</tr>
</tbody>
</table>

where $I_D$ is the drain to source current, $V_G$ is the gate to source voltage, $V_{TH}$ is the threshold voltage, $W$ and $L$ are the channel width and length, respectively, and $C_ox$ is the capacitance per unit area of the gate dielectric. The values of $\mu_{sat}$ were found to be 82 cm²/V·s⁻¹ and 127 cm²/V·s⁻¹ for the In2O3 TFTs fabricated on AlOx-250°C and AlOx-350°C, respectively. The values of $V_{on}$ for the TFTs fabricated on AlOx-250°C and AlOx-350°C were found to be -0.1 V and 0.1 V, respectively. The drain off-current to on-current ratio ($I_{off}/I_{sat}$) for the TFTs fabricated on AlOx-250°C and AlOx-350°C were $3 \times 10^{4}$ and $1 \times 10^{6}$, respectively. The low $I_{off}/I_{sat}$ ratio in case of the TFTs fabricated on AlOx-250°C can be attributed to high leakage of AlOx-250°C as evident from the I-V curve shown in Fig. 2(c). As seen from Table I, the TFTs fabricated using AlOx-500°C as the gate dielectric showed very inferior performance compared to the TFTs fabricated on AlOx-250°C and AlOx-350°C.

Material characterizations of the In2O3 films were performed to understand the effect of annealing temperature of AlOx thin film dielectrics on the performance of In2O3 TFTs. The crystallinity of the In2O3 films was investigated by XRD using grazing incidence geometry with an incident angle of 1°. The grazing incidence XRD patterns (Fig. 4(a)) of the In2O3 films deposited on different AlOx films showed a broad peak at ~30.5° corresponding to the (222) plane of In2O3. The Gaussian peak fitting of the observed peaks revealed that the full width at half maximum (FWHM) for the In2O3 films deposited on AlOx-250°C (FWHM = 2.5°) and AlOx-350°C (FWHM = 2.8°) are higher than that of the In2O3 film deposited on AlOx-500°C (FWHM = 2.1°). Furthermore, Fig. 4(b) shows the XPS O1s spectra of the In2O3 films deposited on AlOx films annealed at different temperatures. The Gaussian fitting (solid lines) of all the O1s peaks exhibited a strong peak at 529.8 eV along with two shoulder peaks at 531.1 eV and 532.3 eV. The peak at 529.8 eV is attributed to the oxygen in In2O3 lattice without oxygen vacancy (Ov).23 The peaks at 531.1 eV and 532.3 eV are attributed to the oxygen in In2O3 lattice with oxygen vacancy (OV) and the OH groups formed to understand the effect of annealing temperature of AlOx thin film dielectrics.

### Table I. Electrical parameters of In2O3 TFTs fabricated using AlOx films annealed at different temperatures as the gate dielectrics.

<table>
<thead>
<tr>
<th>AlOx annealing temperature</th>
<th>OV (%)</th>
<th>$\mu_{sat}$ (cm²/Vs)</th>
<th>$I_{on}/I_{off}$</th>
<th>$V_{on}$ (V)</th>
<th>SS (V/dec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>250°C</td>
<td>24%</td>
<td>82</td>
<td>$3 \times 10^{4}$</td>
<td>-0.1</td>
<td>0.271</td>
</tr>
<tr>
<td>350°C</td>
<td>28%</td>
<td>127</td>
<td>$1 \times 10^{6}$</td>
<td>0.1</td>
<td>0.137</td>
</tr>
<tr>
<td>500°C</td>
<td>31%</td>
<td>5</td>
<td>$2 \times 10^{4}$</td>
<td>-2.6</td>
<td>0.951</td>
</tr>
</tbody>
</table>
In$_2$O$_3$ films deposited on AlO$_x$-250$^\circ$C and AlO$_x$-350$^\circ$C, indicating the presence of smaller concentration of oxygen vacancies in the latter two cases. It is interesting to discuss the origin of the observed broadness of the XRD peaks and lower oxygen vacancy concentration in In$_2$O$_3$ films deposited on the AlO$_x$-250$^\circ$C and AlO$_x$-350$^\circ$C dielectrics. First, the XRD peak broadening most likely comes from the development of a more amorphous-like structure, a trend which has been previously reported for Al-doped In$_2$O$_3$ films,$^{11}$ possibly owing to the big difference between the atomic radii of Al (1.43 Å) and In (1.66 Å).$^{24}$ Thus, we suggest that the XRD peak broadening in case of In$_2$O$_3$ films deposited on AlO$_x$-250$^\circ$C and AlO$_x$-350$^\circ$C films is likely due to Al diffusion into the In$_2$O$_3$ channel from the underlying AlO$_x$ films that are rich in Al-OH groups. Second, the OH-groups attached to the Al at the interface of AlO$_x$ as seen from the XPS O1s spectra (Fig. 2(a)) can act as oxygen source and induce a more efficient oxidation of In$_2$O$_3$ after annealing at 250 $^\circ$C. Both Al-doping and OH-assisted oxidation of In$_2$O$_3$ are expected to reduce the oxygen vacancy concentration in In$_2$O$_3$ films deposited on AlO$_x$-250$^\circ$C and AlO$_x$-350$^\circ$C films.$^{11,25}$ In contrast, higher oxygen vacancy concentration is expected in the In$_2$O$_3$ deposited on AlO$_x$-500$^\circ$C due to the reduced OH group concentration in AlO$_x$-500$^\circ$C (see XPS in Fig. 2(a)). This is also expected to increase the carrier concentration and conductivity of In$_2$O$_3$, which is believed to be controlled by oxygen vacancies.$^{26}$ The increased carrier concentration of In$_2$O$_3$ TFTs made on AlOxC-500$^\circ$C is indeed observed as can be seen from the plot of drain current vs drain voltage measured at zero gate bias, as shown in the inset of Fig. 3(e). Fig. 3(e) shows significantly higher drain voltage measured at zero gate bias, as shown in the inset of Fig. 3(e). The high value of “SS” in case of the TFTs made on AlOxC-500$^\circ$C, further confirms the increased carrier concentration in In$_2$O$_3$ deposited on AlOxC-500$^\circ$C. Thus, the inferior performance in case of the In$_2$O$_3$ TFT fabricated using AlOxC-500$^\circ$C as the gate dielectric can be attributed to the scattering of carriers due to the high carrier concentration in the corresponding In$_2$O$_3$ semiconductor layer. Hence, we find that the presence of Al-OH groups in low temperature annealed AlO$_x$ film plays a crucial role in In$_2$O$_3$ TFT performance. It is noted that the performance of our devices are superior to that of previously reported results for the solution processed In$_2$O$_3$ TFTs.$^{10,11,27}$

In summary, thin film transistors were fabricated using solution-processed In$_2$O$_3$ semiconductor as the channel layer and AlO$_x$ films annealed at different temperatures as the gate dielectric. The TFTs were operated at very low voltage range and exhibited better performance than previously reported In$_2$O$_3$ TFTs processed at these temperatures. High performance In$_2$O$_3$ TFTs could be obtained at relatively low processing temperatures by controlling the Al-OH content in solution processed AlO$_x$ gate dielectrics.

The authors acknowledge the generous support of the KAUST baseline fund.