Ultrasmooth, High Electron Mobility Amorphous In–Zn–O Films Grown by Atomic Layer Deposition

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Supporting Information

ABSTRACT: Ultrasmooth and highly conductive amorphous In–Zn–O (a-IZO) films are grown by atomic layer deposition (ALD). This opens a new pathway to highly transparent and conductive oxides with an extreme conformality. In this process, a-IZO films of various compositions are deposited by alternate stacking of ZnO and In2O3 atomic-layers at a temperature of 200 °C. The IZO films have an amorphous phase over a wide composition range, 43.2–91.5 at %, of In cation ratio. The In-rich a-IZO film (83.2 at % In) exhibits a very low resistivity of 3.9 × 10−4 Ω cm and extremely high electron mobility in excess of 50 cm2 V−1 s−1, one of the highest among the reported ALD-grown transparent conducting oxides. Moreover, it exhibits an ultrasmooth surface (∼0.2 nm in root-mean-square roughness), and can be conformally coated onto nanotrench structures (inlet size: 25 nm) with excellent step coverage of 96%.

1. INTRODUCTION

Amorphous transparent conductive oxides (a-TCOs) have attracted great interest for applications such as transparent electrodes and semiconducting channel layers in photovoltaics, flat panel displays, light emitting diodes (LEDs), and transparent electronics. Amorphous ZnO (a-ZnO), SnO2 (a-SnO2), In–Ga–Zn–O, In–Zn–Sn–O, and Zn–Cd–O, consist of binary or ternary TCOs that have different crystal structures and/or different coordination number of metal cations to oxygen. These materials have unusual high electron mobility over 10 cm2 V−1 s−1, despite the absence of a long-range order, owing to the overlap of large and spherical s-orbitals of heavy (n − 1)d10ns0 metal cations, such as In 5s1. At the same time, a-TCOs are optically transparent (bandgap larger than 3 eV) and can have a smooth surface attributable to their amorphous nature. In addition, a-TCOs hold additional advantages of a low-temperature deposition down to room temperature (contrary to >300 °C for conventional polycrystalline TCOs) and flexibility. Of the various a-TCOs, In–Zn–O (IZO) has been one of the most widely studied materials due to its extremely high electron mobility (20–70 cm2 V−1 s−1) and excellent thermal stability up to 600 °C. In addition, electrical conductivity of IZO can be widely tunable, from metallic to semiconducting (104–106 S cm−1), via adjusting film composition, or simple postannealing, or doping/alloying with other elements (Ga, Hf, Zr, etc.).

These remarkable advantages have led IZO and its derivatives to the industrial standard of both transparent electrodes and semiconductors in various applications. While conventional TCOs are mostly deposited by sputtering, TCOs grown by atomic layer deposition (ALD) have been recently investigated as transparent electrodes of emerging applications, such as nanocrystallized solar cells, photoelectrochemical energy-harvesting devices, and organic LEDs. These approaches are attributed to the ALD’s excellent film conformality onto complex substrates at a low temperature. Most studies have focused on ALD-grown polycrystalline TCOs, such as indium–tin–oxide (ITO), Al-doped ZnO (AZO), and Nb-doped SnO2. There have been relatively few ALD growth studies of a-TCOs, despite its additional merits of the extreme conformality and the ultrasmooth surface. Recently, Heo et al. and Lee et al. proposed an ALD process of amorphous Zn–Sn–O (ZTO) and successfully integrated that into a semiconducting channel layer of thin film transistors and into an electron-blocking layer of thin film solar cells. These examples highlight the advantages of ALD-grown a-TCOs. Still, there are many other applications to benefit from the excellent electrical conductivity, mobility, and wide tunability of a-IZO films grown by ALD. The development of ultrathin a-IZO, in particular, will shed...
light into a high performance three-dimensional transparent electrode. The ultrasmooth surface and the absence of grain boundaries would minimize scattering of transporting electrons. Efforts on the ALD growth of In₂O₃ films have been made, and some of those successfully resulted in the high-quality films.²²−²⁴ Nevertheless, to the best of the authors’ knowledge, there have been no reports on the ALD-growth of α-IZO films.

In this article, we report on structural and electrical properties of ALD-grown IZO films with outstanding electrical conductivity and extremely smooth surface. Our previous efforts on the ALD growth of ZnO-based TCOs and In₂O₃ films paved the way to this IZO growth.²⁵−²⁸ The successful development of the ALD-In₂O₃ process using trimethylindium (TMI) and water vapor, in particular, motivated us to explore ALD-grown α-IZO films. IZO films of various cation ratios, ranging over all compositions, were deposited by alternate stacking of ZnO and In₂O₃ atomic-layers. Film growth behavior and structural and electrical properties of ALD-IZO films were studied using various tools, including inductively coupled plasma-atomic emission spectroscopy (ICP-AES), transmission electron microscopy (TEM), atomic force microscopy (AFM), and Hall measurements. The IZO films were amorphous over a wide composition range of 47.2−91.5 at % In cation ratios. At 83.2 at % In, the film exhibited the lowest resistivity of 3.9 × 10⁻⁴ Ω cm, an extraordinary high electron mobility in excess of 50 cm² V⁻¹ s⁻¹, and an ultrasmooth surface (∼0.2 nm in root-mean-square roughness). In addition, when an ultrathin α-IZO film was conformally coated onto nanotrench structures (inlet size: 25 nm), it showed excellent step coverage of 96%.

### 2. EXPERIMENTAL DETAILS

IZO films were deposited in a traveling-wave type ALD apparatus (LUCIDA D-100, NCD Co., Ltd, Korea) at a deposition temperature of 200 °C, as calibrated by a thermocouple inside the chamber. Thermally grown SiO₂ (100 nm)/Si wafers were used as a substrate. Diethylzinc (DEZ, EG Chem Co., Ltd., Korea) and TMI (EG Chem Co., Ltd., Korea) were used as precursors for the ZnO and In₂O₃ deposition, respectively. DEZ canister was cooled to 10 °C and TMI canister was heated to 50 °C of which both precursors allowed high working pressure increases above 50 mTorr during the exposure. Water vapor, held at room temperature, was commonly used as a reactant. A working pressure increase during the water vapor exposure was kept high as 600−700 mTorr to provide sufficient Langmuir exposure (∼2 Torr s) for the deposition of In₂O₃.²⁸ N₂ gas with a flow rate of 50 sccm (standard cubic centimeter per minute) was introduced in the chamber during the entire process. The optimized ALD cycle consists of precursor delivery (1 s pulsing accompanied by 5 s extended exposure without evacuation) − N₂ purge 20 s − H₂O pulsing (0.5 s for ZnO and 3 s for In₂O₃) − N₂ purge 20 s (Figure S1 in SI). Herein, the extended precursor exposure was applied to provide sufficient adsorption time to overcome the adsorption delay. The IZO films with various cation ratios were deposited by varying an ALD cycle ratio between ZnO and In₂O₃. The number of total ALD cycles was adjusted between 300 and 2000 as the films have similar thicknesses of 40−50 nm. For instance, the film with In cation ratio of 83.2 at % was deposited by repeating 200 supercycles consisting of nine cycles of In₂O₃ and one cycle of ZnO.

ALD-grown IZO films were characterized by various tools. Film thicknesses were measured by ellipsometry (Gaertner Scientific Corp., L2W155830) at a wavelength of 632.8 nm. The cation ratios were determined by ICP-AES (Perkin-Elmer, Optima-4300 DV), and compositional depth profiles and impurity levels were confirmed by AES (Perkin-Elmer, PHI 660). Structural characterizations were performed using XRD diffractometry (New D8 Advance, Bruker), TEM (JEOL, JEM-3000F operated with a 300 kV gun), and AFM (Surface Imaging Systems, NANOStation II and Scanning Probe Image Processor Ver. 4.2). Electrical properties were studied by Hall measurements (BIO-RAD, HLS500PC) using the Van der Pauw configuration. A UV−vis−NIR spectrophotometer (Carry 5000, Varian) was used to obtain optical transparency of the films that were deposited on a glass substrate (Eagle XG, Samsung Corning Co., Ltd., Korea).

### 3. RESULTS AND DISCUSSION

For the growth of ALD-IZO films, deposition temperature was set to 200 °C since it is the lower temperature boundary of the ALD-In₂O₃ growth window.²⁸ In this study, we introduced a modified precursor exposure recipe to overcome adsorption delay of the precursors during the ALD-growth of IZO films. Specifically, the precursor was pulsed into the chamber for a short time (1 s) and exposed for prolonged duration (5 s) without evacuation (Figure S1 in Supporting Information [SI]). First, individual ZnO and In₂O₃ films were deposited using the extended exposure recipe, and properties of those films were characterized. As summarized in Table 1, the ZnO and In₂O₃ films showed the growth rates of 1.51 Å cycle⁻¹ and 0.48 Å cycle⁻¹, respectively. X-ray diffraction (XRD) analysis confirmed that the ZnO and In₂O₃ films have Wurtzite and Bixbyite structures, respectively. The resistivity of In₂O₃ film (3.1 × 10⁻³ Ω cm) was significantly lower than that of ZnO (1.9 × 10⁻² Ω cm), attributable to a much higher electron mobility (62.2 cm² V⁻¹ s⁻¹ for In₂O₃ and 17.9 cm² V⁻¹ s⁻¹ for ZnO). The carrier concentrations of both films were laid in the low 10¹⁹ cm⁻³ levels. These electrical properties are well matched to other reports.²⁸−³⁰

Once the individual films were characterized, IZO films of various compositions were deposited by alternately stacking ZnO and In₂O₃ atomic layers with various ALD cycle ratios. The total numbers of the ALD cycles were adjusted for the films to have thicknesses of 40−50 nm. Figure 1a shows the growth rates of ALD-grown IZO films as a function of In₂O₃ ALD cycle ratio. Herein, In₂O₃ ALD cycle ratio, the x-axis of the plot, indicates a ratio of the number of In₂O₃ cycles to the total ALD cycles. For instance, 10% cycle ratio means a film is deposited by repeating nine cycles of ZnO and one cycle of In₂O₃. The estimated film growth rates (Goverall) were determined from the growth rates of individual ZnO (GZnO) and In₂O₃ (GInO) as

\[
G_{\text{overall}} = R_{\text{ALD}} G_{\text{InO}} + (1 - R_{\text{ALD}}) G_{\text{ZnO}}
\]

<table>
<thead>
<tr>
<th>Film</th>
<th>Crystal structure</th>
<th>Resistivity (Ω cm)</th>
<th>Density (g cm⁻³)</th>
<th>Growth rate (Å cycle⁻¹)</th>
<th>AREAL DENSITY OF ATOM DEPOSITED (1 ALD CYCLE) (cm⁻²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>ZnO</td>
<td>Wurtzite</td>
<td>5.56</td>
<td>1.51</td>
<td>6.22 × 10¹⁴</td>
<td></td>
</tr>
<tr>
<td>In₂O₃</td>
<td>Bixbyite</td>
<td>7.19</td>
<td>0.48</td>
<td>1.50 × 10¹⁴</td>
<td></td>
</tr>
</tbody>
</table>

where $R_{\text{ALD}}$ is the In$_2$O$_3$ ALD cycle ratio in fraction. In Figure 1a, it is obvious that the measured growth rates (symbols) are much lower than the estimated values (rule of mixture). This indicates that the adsorption of precursors was retarded during the growth. The film growth rate decreased with increasing the In$_2$O$_3$ ALD cycle ratio and exhibited the lowest values of 0.19–0.24 Å cycle$^{-1}$ at 80–93.3 ALD cycle %. Once the growth rate showed the lowest value, it increased with increasing In$_2$O$_3$ ALD cycle ratio and reached that of undoped In$_2$O$_3$.

Film growth behavior was further studied by measuring cation compositions of IZO films. Figure 1b shows the cation ratio of In [In/(Zn + In)], measured by ICP-AES (symbols) and estimated from the rule of mixture (solid line), as a function of the In$_2$O$_3$ ALD cycle ratio. The estimated In cation ratios were determined as,

$$\frac{\text{In}}{(\text{Zn} + \text{In})} = \frac{R_{\text{ALD}}N_{\text{In}}}{R_{\text{ALD}}N_{\text{In}} + (1 - R_{\text{ALD}})N_{\text{Zn}}}$$

(2)

where $N_{\text{In}}$ ($1.50 \times 10^{14}$ cm$^{-2}$) and $N_{\text{Zn}}$ ($6.22 \times 10^{14}$ cm$^{-2}$) are the areal densities of In and Zn atoms deposited for 1 ALD cycle of pristine films. In the overall ALD cycle ratios, the measured In cation ratios were higher than the estimated values from eq 2. For instance, when the In$_2$O$_3$ cycle ratio was 50% (ZnO:1:In$_2$O$_3$:1), the film had the In cation ratio of 47.2 at % and this is more than 2-fold higher than that expected (19.4 at %). These results suggest that the retarded adsorption of DEZ would be mainly responsible for the much lower film growth rates than expected in Figure 1a. Details on the growth kinetics of ALD-IZO films will be reported elsewhere.

For the ALD-IZO films grown above, the structural properties were characterized by AES, XRD, and TEM. Figure 2 shows the XRD patterns of IZO films with various In cation ratios that are measured by ICP-AES. For the low In cation ratios (4.7 and 8.2 at %), the films had the Wurtzite structure, the same as the undoped ZnO film in this study and the reported ALD-AZO and TZO films.25,27,28 This indicates that ZnO forms the matrix and the In$_2$O$_3$ atomic-doping-layers are spatially distributed, corresponding to the depth proﬁling patterns of the undoped ZnO, IZO with various In cation ratios, and undoped In$_2$O$_3$ films. The cation ratios are measured by ICP-AES.

of either crystalline ZnO or In$_2$O$_3$. Rather, a very broad peak is observed between In$_2$O$_3$ (222) peak and ZnO (0002) peak. These XRD peaks are well matched to those of amorphous IZO films.9,30 In these films, Zn and In compositions were quite uniform throughout the film growth direction (Figure S2c in SI), unlike the films with spatial In$_2$O$_3$ or ZnO atomic-doping layers. All films studied by AES depth profiling had a low carbon impurity level below 1 at %.

Even the XRD analysis provided a rough estimation of the crystal structures of ALD-grown IZO films; some ultrathin (<50 nm) films resulted in low XRD intensities. This hindered the clear interpretation of the crystal structure. Therefore, the microstructures of ALD-IZO films were further investigated by TEM. Figure 3 shows cross-sectional views of bright field (BF, a–d) and high-resolution (HR, e) TEM micrographs of (a) undoped ZnO, IZO with the measured In cation ratios of (b) 4.7 at %, (c) 47.2 at %, and (d,e) 83.2 at %, and (f) undoped In$_2$O$_3$ films. Insets in (a–d,f) indicate the local area electron diffraction (SAED) pattern. As shown in Figure 3a, the undoped ZnO film had the Wurtzite structure with polycrystalline, columnar grains. The same features were also found in the low In cation ratio IZO film (4.7 at %). In Figure 3b, spatial In$_2$O$_3$ atomic-doping layers are clearly observed within the ZnO matrix, as in the case of ALD$_3$O$_3$ atomic-layers doping.33,34 Consecutive TEM studies revealed that the Wurtzite structure was conserved until the In cation ratio increased up to 27.3 at %. Further increase in the In cation ratio led to transformation to an amorphous phase over a wide range of In cation ratio of 47.2–91.5 at %. These amorphous ranges correspond well to the reported IZO films deposited by other methods.9,30 One interesting feature is that the Zn-rich a-IZO film (In 47.2 at %, Figure 3c) consists of an amorphous matrix at the base and 10s of nanometer-sized crystallites above that (Figure S3 in SI). From the SAED patterns, these crystallites were indexed as (1 0 6), (0 0 14), and (0 0 16) peaks of a homologous (ZnO)$_x$(In$_2$O$_3$)$_{1-x}$ phase.31 Such a bilayered structure is unusual in the layer-by-layer grown ALD films. Rather, it looks like the film initially grew with the amorphous phase, and it partially crystallized once the film became thicker and provided a kinetic energy for the crystallization. The coexistence of amorphous and partially crystallized phases has also been observed for PLD-grown film (In 40 at %), but the crystallites were columnar shaped.32 Further study needs to be conducted to more clearly understand this unusual crystal-
zation behavior. For the higher In cation ratios, these crystallites were not observed, and the films were fully amorphous with an extremely smooth surface. Images d and e of Figure 3 show the representative TEM micrographs of α-IZO film (In 83.2 at %). The HRTEM and its fast Fourier transform images clearly reveal the amorphous phase. Once the In cation ratio increases above 91.5 at %, the films have the Bixbyite structure with large, polycrystalline grains like the undoped In2O3 film (Figure 3f), identical to the XRD results in this study. The structural properties studied here clearly demonstrate the feasibility of growing smooth, compositionally uniform, and low-impurity level α-IZO films by the ALD process.

For all the composition ranges of the ALD-IZO films, the electrical properties were investigated using Hall measurements. Figure 4 shows the electrical resistivities of IZO films as a function of the measured In cation ratios. The crystal structures of IZO films are shaded into three regimes. For the small amount of In doping into the ZnO matrix (Wurtzite structure regime), the electrical resistivity decreased about one order of magnitude compared to the undoped ZnO film. As the In doping concentration increased up to 8.2 at %, the resistivity decreased down to 2.6 × 10⁻³ Ω cm. This result indicates that the incorporated In atoms worked as dopants either by substituting into Zn sites (In2⁺zns)³³ or by taking oxygen from the ZnO matrix and forming oxygen vacancies (Vₐ²⁺)/zinc interstitials (Zn⁺⁺).²² Above those cation ratios, the resistivity slightly increased and fluctuated. Once the films were amorphized, by adding more In cations (47.2–91.5 at %), there was a drastic decrease of electrical resistivities to below 10⁻³ Ω cm. Increasing the In cation ratio to 83.2 at %, the α-IZO film exhibited a lowest resistivity of 3.9 × 10⁻⁴ Ω cm. This value is much lower than those of ALD-grown doped ZnO films [(1.7–4.4) × 10⁻³ Ω cm for Al²⁵,²⁷,³⁴,³⁵ (8.0 × 10⁻⁴)–(1.4 × 10⁻³) Ω cm for Ga²⁶,³⁶ 1.0 × 10⁻³ Ω cm for Ti²⁷ and 1.3 × 10⁻³ Ω cm for Zr dopings]³⁷ and is comparable to that of ALD-ITO films [(2.4–3.9) × 10⁻⁴ Ω cm]¹⁷,³⁸ and that of high-quality sputtered IZO films [(2.6–8.4) × 10⁻⁴ Ω cm].⁹,³⁹,⁴¹ The In cation ratio of the lowest resistivity ALD-IZO film (83.2 at %) is well matched to that of an industrial α-IZO TCOs (In 83 at %, as 10 wt % ZnO incorporated).³⁹ Once the crystal structure is transformed to the Bixbyite for higher In cation ratios (≥93.0 at %), that low electrical resistivity has disappeared. Instead, by inserting the ZnO atomic-doping layers into the In₂O₃ matrix, the electrical resistivity drastically increased to even above 10⁻¹ Ω cm.

Figure 5 shows the (a) Hall mobilities and (b) carrier concentrations of IZO films as a function of the measured In cation ratio. For the low In cation ratios (Wurtzite regime), the small amount of In doping resulted in a drastic increase of the carrier concentration. The film doped with 8.2 at % In exhibited the highest carrier concentration of 7.1 × 10²⁰ cm⁻³·³. This value is more than twice of the ALD-grown AZO and TZO films [(2–3) × 10²⁰ cm⁻³].²⁵,²⁷,³⁴,³⁵ However, the Hall mobility decreased down to 3.5 cm² V⁻¹ s⁻¹ with increasing In cation ratio to 8.2 at % and canceled the benefit of higher electron concentration to electrical conductivity. That mobility is much lower than that of the ALD-grown AZO (14.6 cm² V⁻¹ s⁻¹) and TZO films (20.4 cm² V⁻¹ s⁻¹).²⁷ The In doping also resulted in much lower mobility in sputtered films, attributed to a larger scattering cross-section of In ions with larger ionic radius (In³⁺: 0.81 Å, Al³⁺: 0.57 Å, Ti⁴⁺: 0.64 Å).⁴² The decrease of mobility upon an increase of carrier concentration in this regime can be explained by a typical ionized impurity scattering.⁴³

Once the films are amorphized for the higher In cation ratios, the Hall mobility gradually increased with increasing In cation ratio. Specifically, the In-rich α-IZO film (83.2 at %) exhibited
an extraordinary high electron mobility of 52.1 cm² V⁻¹ s⁻¹. This value is even slightly higher than the ALD-grown polycrystalline ITO films (∼48 cm² V⁻¹ s⁻¹) of the optimal conductivities. The extremely high electron mobility of α-IZO would be attributed to the less surface scattering originating from the ultrasmooth surface and/or the absence of grain boundaries as scattering centers. In Figure 5a, the mobility increase with increasing In cation ratio is probably due to the enhanced overlap of large, spherical In 5s orbitals. At the same time, surprisingly, the carrier concentration also increased slightly. This tendency, the concurrent increase of both mobility and carrier concentration, suggests that the electron transport through α-IZO films is not limited by the ionized impurity scattering but is dominated by other mechanisms such as lattice (or phonon) scattering. Indeed, Leenheer et al. found that the mobility of sputtered α-IZO films decreased gradually with increasing measurement temperature, without a change of carrier concentration, indicative of lattice scattering. By adding more In cations, the high mobility disappeared in the Bixbyite structure regime. Rather, both the mobility and carrier concentration decreased simultaneously with insertion of the ZnO atomic-doping layers into the In₂O₃ matrix. This indicates that the incorporated Zn atoms might result in the formation of killer acceptors, such as substitutional Zn (Zn_{In}) or In vacancies (V_{In}), and those impurities might work as scattering centers for the electron transport. Indeed, the drastic drop of the electron density down to 10¹⁹ cm⁻³ may lead to the decrease of mobility since the depletion of electrons at/near the grain boundaries would bring up an energy barrier for the electron transport and result in the increased grain boundary scattering at that low carrier concentration regime (below 10²⁰ cm⁻³). Our results clearly demonstrate that, by careful optimization of the composition, the ALD-grown In-rich α-IZO film (In 83.2 at %) can excel in electrical properties such as very low resistivity down to 3.9 × 10⁻⁴ Ω cm and extreme electron mobility in excess of 50 cm² V⁻¹ s⁻¹.

Focusing on the optimally conductive In-rich α-IZO film (In 83.2 at %), we investigated optical transparency, surface roughness, and conformality to high-aspect-ratio nanostructures. Figure S4 in SI shows the transmittance curves of undoped ZnO, α-IZO, and undoped In₂O₃ films, as referenced to bare glass. The α-IZO film exhibited high optical transmittance over 80% for most of the visible spectrum. At 550 nm, the α-IZO film had a transmittance of 83%, and this value is slightly lower than those of undoped ZnO and In₂O₃ films (≥86%). Figure 6 shows the surface topologies of undoped ZnO, α-IZO, and undoped In₂O₃ films scanned by AFM. The polycrystalline ZnO and In₂O₃ films exhibited a rough surface with root-mean-square roughness (Rₚ) of 1.1 and 2.0 nm, respectively. In contrast, the ALD-grown, 49-nm-thick α-IZO film exhibited an ultrasmooth surface with a very low Rₛ of 0.2 nm, a replication of the underlying SiO₂ surface. The extremely low roughness observed in this study is comparable to those of sputtered IZO films (<0.4 nm) and might be attributed to its amorphous nature, as demonstrated for other ALD-grown amorphous dielectrics. It should be noted that the roughness of ALD-grown α-IZO film is considerably lower than those of polycrystalline ALD-TCOs, 8.9 nm for ITO (89-nm-thick) and 0.8 nm for AZO (98.4-nm-thick) films.

Finally, the conformality of ALD-grown α-IZO film was investigated. Figure 7 shows the cross-sectional view TEM micrographs of α-IZO films coated onto the nanotrench structures. The nanotrenches have an aspect ratio of 4:5:1 with very narrow inlets of 25 nm. In Figure 7a, the ultrathin α-IZO film (∼10 nm) was successfully coated on the multiple trenches. In the magnified image (Figure 7b), it is clearly observed that the α-IZO film was coated conformally, from the top of the trenches to a narrow bottom side, and the step coverage was as good as 96%. Our findings of the ultrasmooth surface and excellent conformality, and a comparable or even better electrical conductivity and mobility, highlight the advantages of ALD-grown α-IZO films over the conventional polycrystalline ALD-TCOs.
4. CONCLUSIONS

Highly conductive, transparent, and ultrasmooth α-IZO films were grown by ALD via alternate stacking of ZnO and In$_2$O$_3$ atomic-layers. The growth behaviors and structural and electrical properties of ALD-IZO films with various compositions were investigated. The crystal structures and microstructures were studied by XRD, AES, and TEM, and the IZO films were found to be amorphous over wide In cation ratios of 43.2–91.5 at %. The α-IZO films exhibited a very low electrical resistivity down to $3.9 \times 10^{-4}$ Ω cm and extremely high mobility in excess of 50 cm$^2$ V$^{-1}$ s$^{-1}$, surpassing the electrical properties of polycrystalline ALD-TCOs. In accordance with the excellent electrical properties and high optical transparency, the ultrasmooth surface with a near-zero roughness (0.2 nm) and a perfect conformity onto the nanostructures (step coverage of 96%) highlight the merits of ALD-grown α-IZO films over those of the conventional polycrystalline ALD-TCOs. The development of ALD-grown ultrathin, ultrasmooth, and ultraconformal α-TCOs of extraordinarily high conductivity and transparency in this study could enable new advances or breakthroughs in such applications as nanostructured photovoltaics, photoelectrochemical energy-harvesting devices, and transparent flexible electronics.