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Supplementary Information

Chemically Improved High Performance Printed Indium Gallium Zinc Oxide Thin-Film Transistors

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Experimental Deatails

Precursor Solution Synthesis. All reagents, zinc acetate dihydrate ($Zn(C_2H_3O_2)_2 \cdot 2H_2O$, 98%), indium nitrate hydrate ($In(NO_3)_3 \cdot H_2O$, 99.9%), and gallium nitrate hydrate ($Ga(NO_3)_3 \cdot H_2O$, 99.9%), ethylene glycol ($HOCH_2CH_2OH$, anhydrous 99.8%), monoethanolamine ($NH_2CH_2CH_2OH$, 99%), and 2-methoxyethanol ($CH_3OCH_2CH_2OH$, anhydrous 99.8%) were purchased from Aldrich and used without additional purification. 0.375 M metal precursor solutions were prepared in 2-methoxyethanol including the different amount of ethylene glycol. The composition of ethylene glycol was varied from 0 to 30 vol%, and chemical composition ratio of IGZO precursor solution was In:Ga:Zn = 63:10:27. Monoethanolamine and DI-water were additionally incorporated as a stabilizer and a reactant for hydrolysis reaction, respectively. The prepared clear solutions were stirred for 3 hr at room temperature prior to spin coating.

Transistor Fabrication and Electrical Performance Characterization. Doped silicon substrate with a 100 nm-thick thermal SiO_2 layer, used as a gate dielectric, was sonicated with absolute ethanol and dried with an N_2 stream, followed by UV/O_3 treatment for 5 min. For fabricating the transistors with top contact device architecture, each precursor solution was spin coated at 4000 rpm for 35 sec and 20 vol% EG added precursor solution was ink-jet

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printed on gate dielectric layer. The printer setup is composed of a drop-on-demand piezoelectric inkjet nozzle manufactured by Microfab Technologies, Inc. (Plano, TX), and the nozzle with an orifice diameter of 30 μm was used. The resulting IGZO semiconducting layers were annealed at 400 ^{o}C for 30 min, and then Al source and drain electrodes with 50 nm thickness were deposited via thermal evaporation (pressure ~10^{-6} Torr) through shadow masks. The channel length and width are 100 and 100 μm, respectively. For transistors with bottom contact device architecture, ITO source and drain electrodes were patterned using a photolithography and wet etching method on the top of 100 nm-thick SiO_2 gate dielectric. The channel length and width are 100 and 100 μm, respectively. 20 vol% EG added precursor solution was ink-jet printed and then annealed at 400 ^{o}C for 30 min. In order to analyze the contact resistance for both printed devices, a transmission line method (TLM) analysis was carried out. For both printed TFTs with either Al or ITO electrode, the source and drain electrodes had a fixed channel width of 100 μm and varying channel lengths between 20 and 100 μm. The electrical performance of the transistors was analyzed in ambient condition using an Agilent E5270B source-measure unit. Saturation mobilities were extracted from the slope of (drain current)$^{1/2}$ versus gate voltage derived from the device transfer plot. All device fabrication and measurement were carried out in ambient atmosphere.

Film Characterization. The chemical structures of oxide semiconductors were examined by X-ray photoelectron spectroscopy (XPS, Omicron, ESCA Probe). The surface XPS data were collected using monochromatic AlKα radiation (1486.6 eV) in an ultrahigh vacuum system with a base pressure of ~10^{-10} Torr. The crystal structures were analyzed with a grazing angle X-ray diffraction (XRD) using CuKα radiation on a Rigaku ATX-G thin-film diffraction workstation. The grain size and surface morphology were observed by atomic force microscope (Digital Instruments, Nano Scope IV), and the thermal behaviors of metal

salt precursors were monitored using thermal gravimetric analyses (SDT2960, TA Instruments).

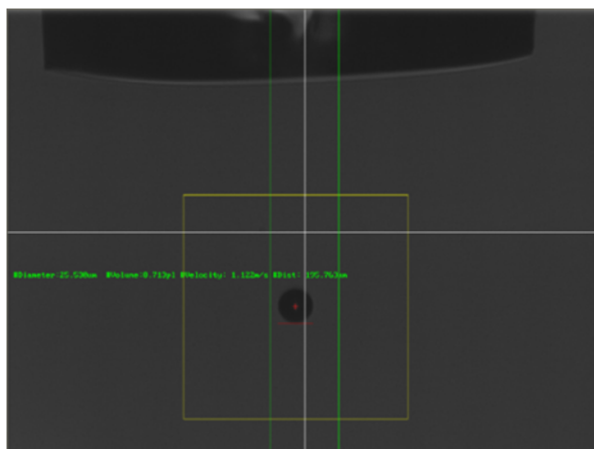

Figure S1. The image showing the jetting behavior of IGZO precursor droplet ejected through the ink-jet printing nozzle.

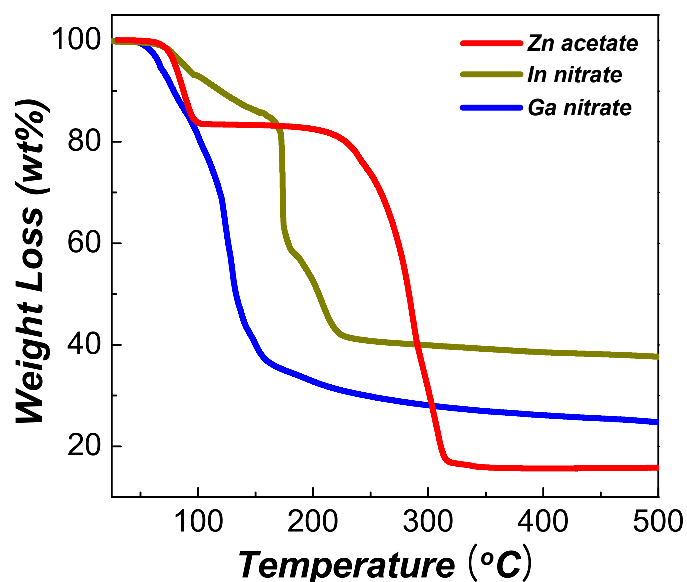

Figure S2. Thermal gravimetric analysis curves of indium nitrate hydrate, zinc acetate dihydrate, and gallium nitrate hydrate. The measurement was carried out in air.

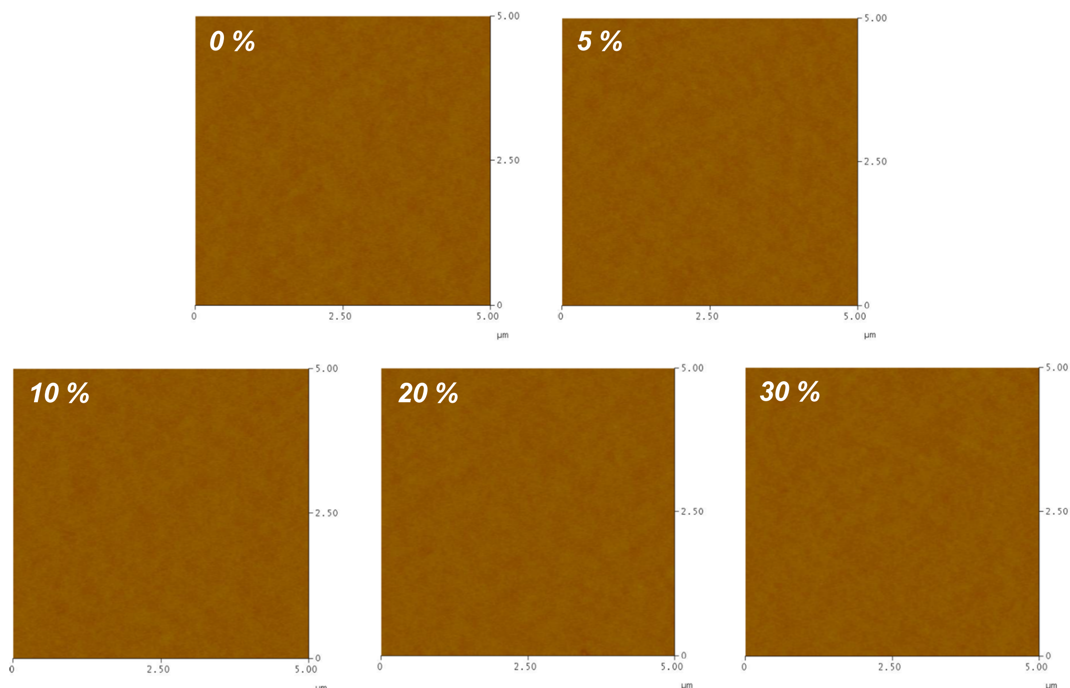

Figure S3. (a) AFM images for IGZO semiconductors prepared using precursor solution containing the different amount of EG between 0 and 20 vol%. All IGZO layers were annealed at 400 °C in air.

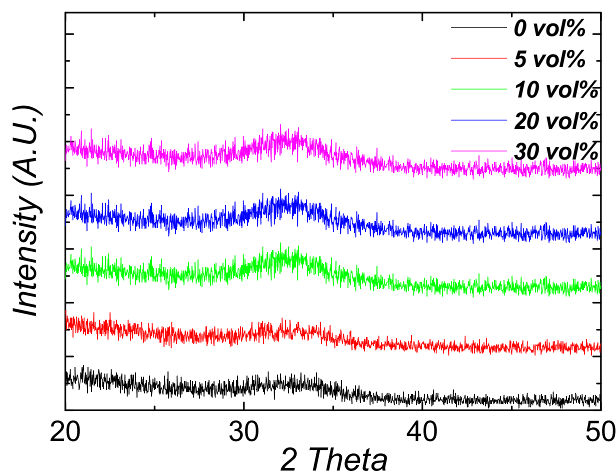

Figure S4. XRD results for IGZO semiconductors prepared using precursor solution containing the different amount of EG between 0 and 20 vol%. All IGZO layers were annealed at 400 °C in air.

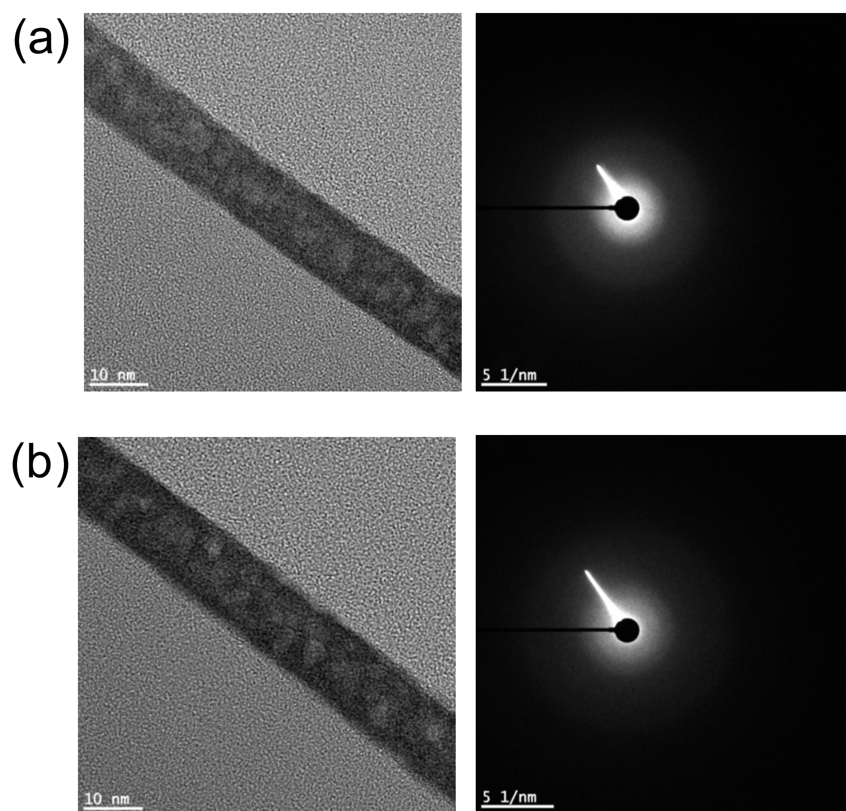

Figure S5. TEM image and selective area diffraction pattern for IGZO semiconductor layer prepared from IGZO precursor solution containing EG of (a) 20 vol% and (b) 30 vol%.

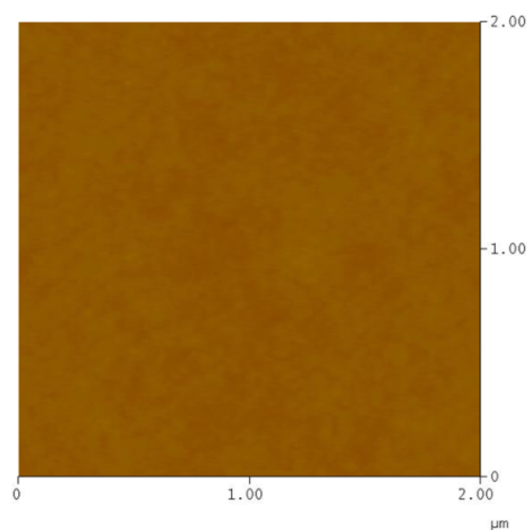

Figure S6. AFM image for the IGZO semiconductor printed layer using a 20 % EG added precursor solution.

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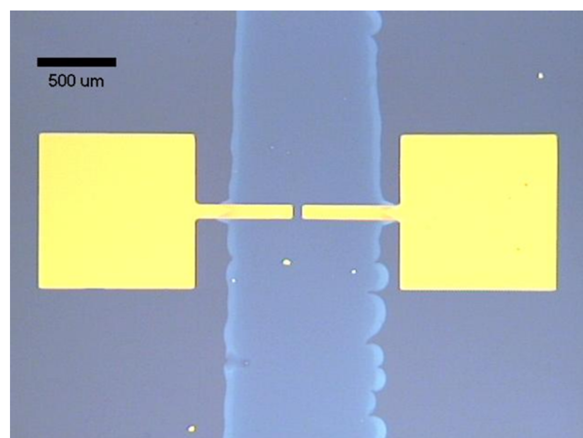

Figure S7. Optical microscope image for the printed IGZO TFT with ITO source/drain electrode.