# Supplementary Information

# Direct patterning of sol-gel metal oxide semiconductor and dielectric films via selective surface wetting

Sujin Sung,<sup>*a*</sup> Sungjun Park,<sup>*a*</sup> Seungbok Cha,<sup>*a*</sup> Won-June Lee,<sup>*a*</sup> Chang-Hyun Kim<sup>*a,b*</sup> and Myung-Han Yoon<sup>\**a,b*</sup>

<sup>a</sup>School of Materials Science and Engineering, Gwangju Institute of Science and Technology, 123 Cheomdan-gwagiro, Buk-gu, Gwangju 500-712, Republic of Korea

<sup>b</sup>Research Institute for Solar and Sustainable Energies, Gwangju Institute of Science and Technology, 123 Cheomdan-gwagiro, Buk-gu, Gwangju 500-712, Republic of Korea

\* Corresponding author: Prof. Myung-Han Yoon, mhyoon@gist.ac.kr

#### EXPERIMENTAL

#### Preparation of sol-gel precursor solutions

All chemical reagents were purchased from Sigma-Aldrich and used without further purification. In preparation for the semiconductor layers, the IGZO precursors, i.e., indium nitrate, gallium nitrate, and zinc acetate were dissolved in 2-methoxyethanol (2-ME, anhydrous, 99.8%). The mixed solution was stirred for 12 hours at 75°C before use. For aluminum oxide as the dielectric layers, a 0.1 M precursor solution was prepared by dissolving aluminum nitrate in 2-ME, and the same thermal treatment/stirring process was applied before the dielectric film deposition.

#### Deposition of self-assembled monolayers (SAMs)

All substrates were cleaned by two successive sonication processes in acetone and isopropyl alcohol for 10 min. n-Octadecyltrichlorosilane (OTS, 95%, Gelest, Inc.) was deposited on p<sup>++</sup>-Si substrates with a 300-nm-thick thermally grown silicon dioxide (highly boron-doped silicon <100>, 0.001–0.003 Ohm·cm, Taewon Scientific Co., South Korea) and n<sup>++</sup>-Si substrates (highly arsenic-doped silicon <100>, < 0.0045 Ohm·cm, Taewon Scientific Co., South Korea) and n<sup>++</sup>-Si substrates (highly arsenic-doped silicon <100>, < 0.0045 Ohm·cm, Taewon Scientific Co., South Korea) for hydrophobic passivation. Briefly, silicon substrates were immersed in a 5 mM solution of OTS in anhydrous toluene for 30 min under an inert atmosphere of nitrogen, followed by rinsing several times with toluene, acetone, and isopropyl alcohol. In the case of SAM deposition on AlO<sub>x</sub>, metal oxide-coated substrates were immersed in a 5 mM solution of n-octadecylphosphonic acid (ODPA, 97%, Alfa Aesar) in isopropyl alcohol (Merck) for 20 min in ambient conditions.

#### Patterning of metal oxide films and fabrication of thin-film transistors (TFTs)

For selective surface activation, OTS- or OPDA-treated substrates were exposed to oxygen plasma, the plasma exposure time between 4 and 7 sec and plasma power 50 W, while an appropriate shadow mask was tightly pressed on top of the substrates. Subsequently, the prestirred solutions of sol-gel metal oxide precursors (i.e., IGZO and AIO<sub>x</sub>) were spin-coated on plasma-activated substrates at a spinning speed of 4000 rpm for 30s, followed by a post annealing at 350°C for 90 min at ambient conditions. All TFT devices were constructed on highly n-doped silicon substrates in a bottom-gate and top-contact configuration. For top source and drain electrodes, 40 nm-thick aluminum films were deposited by thermal evaporation through a shadow mask defining channel length (*L*) and width (*W*) of 50 and 500  $\mu$ m, respectively. For dielectric analysis, metal-insulator-semiconductor (MIS) devices were fabricated on directly patterned AIO<sub>x</sub>/n<sup>++</sup>-Si substrates by deposition of 40-nm thick gold top electrodes via thermal evaporation.

#### Characterizations

Electrical characteristics of TFT devices were evaluated using a semiconductor parameter analyzer (Keithley 4200, USA). Dielectric properties in MIS devices were measured using the same equipment coupled with a capacitance-voltage measurement module. Surface chemical functionality on SAM-treated and subsequent plasma-activated substrates was examined with attenuated total reflectance Fourier transform infrared spectroscopy (ATR-FTIR, Varian 660-IR, USA). Additionally, surface contact angle was measured for wettability analysis (Phoenix-300 Touch, SEO, South Korea). Thickness and surface roughness of sol-gel metal oxide films were determined by ellipsometry (Elli-SE, Ellipso Technology, South Korea) and tapping-mode atomic force microscopy (AFM, XE-100, Park Systems, USA), respectively.

### **OPTICAL IMAGES**



Fig. S1 Optical micrographs of directly-patterned IGZO island arrays with various plasma powers and exposure times.

## CONTACT ANGLE MEASUREMENTS



**Fig. S2** Variation of the water contact angle on aluminium oxide with increasing immersion time in an ODPA solution. The inset shows the chemical structure of ODPA.