

## **SUPPLEMENTARY INFORMATION**

### **Micropatterned ZnO Semiconductors for High Performance Thin Film Transistors via Chemical Imprinting with PDMS Stamp**

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### **Experimental Section**

**Materials.** Poly(dimethyl siloxane) (PDMS; Dow Corning), Zinc oxide(99.999%, Aldrich), Lithium hydroxide monohydrate (99.95%, Aldrich), Ammonia water (25.0~28.0 wt%; Daejung, Korea) and methanol(99.5%, Daejung, Korea) were used without purification.

**PDMS stamps.** For preparation of the PDMS stamp, Si master was prepared through photolithography process on Si wafer. The aspect ratio of the PDMS stamp must be considered for the adequate patterning process to avoid problems such as sagging and pairing induced by intrinsic low young's modulus (2 MPa) of PDMS.<sup>[29]</sup> For the successful patterning of semiconductor films, we fabricated the semiconductor patterns (length of square: 500 μm, depth of patterns: 35 μm) on Si wafer via photolithography. PDMS base and curing agent

(10:1) were poured out on Si master, which has positive relief in the petri-dish. For curing, the mixed materials were annealed at 80°C for 4 hours, and then the PDMS stamp was peeled off from the Si master. To avoid uncured PDMS oligomers, the PDMS stamp was post-cured at 80°C for an additional 12 hours. After that, the PDMS stamp with negative relief structure was obtained. The prepared stamp was soaked in the binary solvent composed of ammonia water and methanol (volume ratio, 5:5) for one hour. After one hour, the soaked stamp was blown with nitrogen gas to remove solvent on its surface.

**Preparation of ZnO semiconductor films.** 0.001 mole of ZnO solution (ZnO powder 0.08139 g in 12 mL NH<sub>4</sub>OH) was refrigerated for 5 hours to increase the solubility of ZnO.<sup>[25]</sup> The prepared solution was spin-coated (3000 rpm, 30 s) on Si wafer with a thermally grown SiO<sub>2</sub> layer (thickness ~ 200 nm). The ZnO thin films were cured at 300 °C for 1hour. For fabrication high performance TFTs, 0.1mL of LiOH solution (LiOH powder 0.2395g in 10mL de-ionized water) was added to the zinc oxide solution (ZnO powder 0.08139 g in 12 mL NH<sub>4</sub>OH); this resulted in 10% Li doped ZnO solution. The 10% Li doped ZnO precursor solution was spin-coated (3000 rpm, 30 s) on SiO<sub>2</sub>/Si wafer (SiO<sub>2</sub> thickness ~ 200 nm).<sup>[1]</sup>

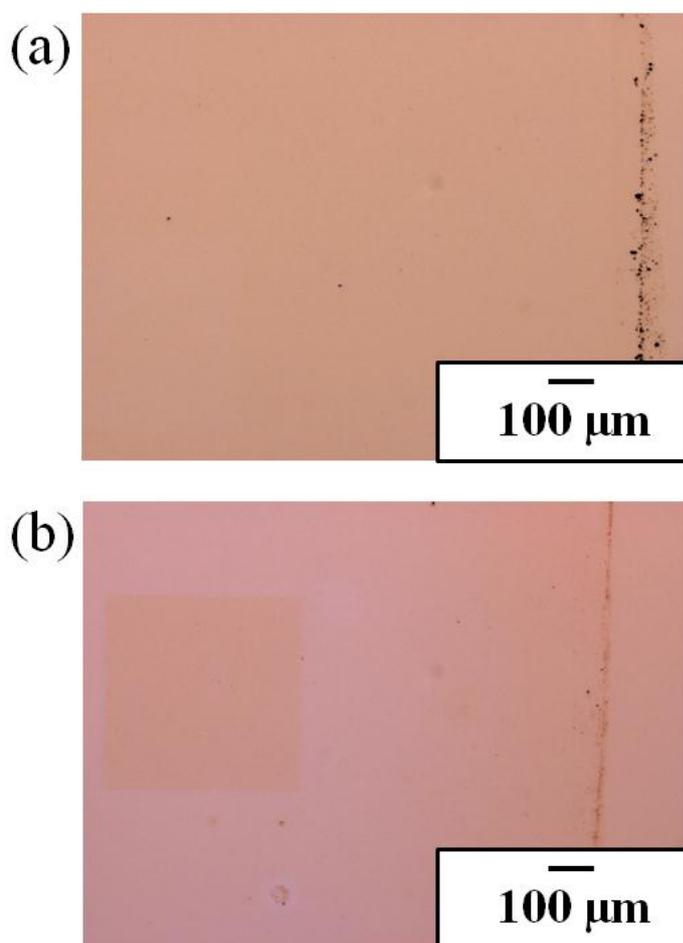
**Chemical imprinting.** The prepared stamp was contacted on ZnO thin films for 15 minutes to transform to the aqueous salt selectively in a refrigerator. After detaching the stamp from the ZnO thin films, the patterned ZnO thin films were washed with de-ionized water to dissolve the formed aqueous salt on the contact regions with PDMS stamp. To remove residual water and ammonia, the patterned ZnO thin films were post-annealed at 110 °C for 30 minutes to make only ZnO patterns. For fabricating high performance TFTs, this patterning method was applied to Li doped ZnO thin films with the same process.

**Fabrication of TFTs.** After the chemical imprinting, the TFTs were fabricated on patterned Li doped ZnO semiconductor films. The form of patterned Li doped ZnO semiconductor films were rectangular with a length of 500  $\mu\text{m}$ . As source/drain electrodes, Aluminum (Al, 100 nm) was evaporated thermally on Li doped ZnO thin films. For comparison of the patterning effect, as unpatterned devices, the TFTs were fabricated on the spin-coated Li doped ZnO thin films with the same process as patterned devices. As a result, the patterned devices' width was 500  $\mu\text{m}$  and length was 50  $\mu\text{m}$  (W/L=10), while the unpatterned devices' width was 1000  $\mu\text{m}$  and length was 50  $\mu\text{m}$  (W/L=20).

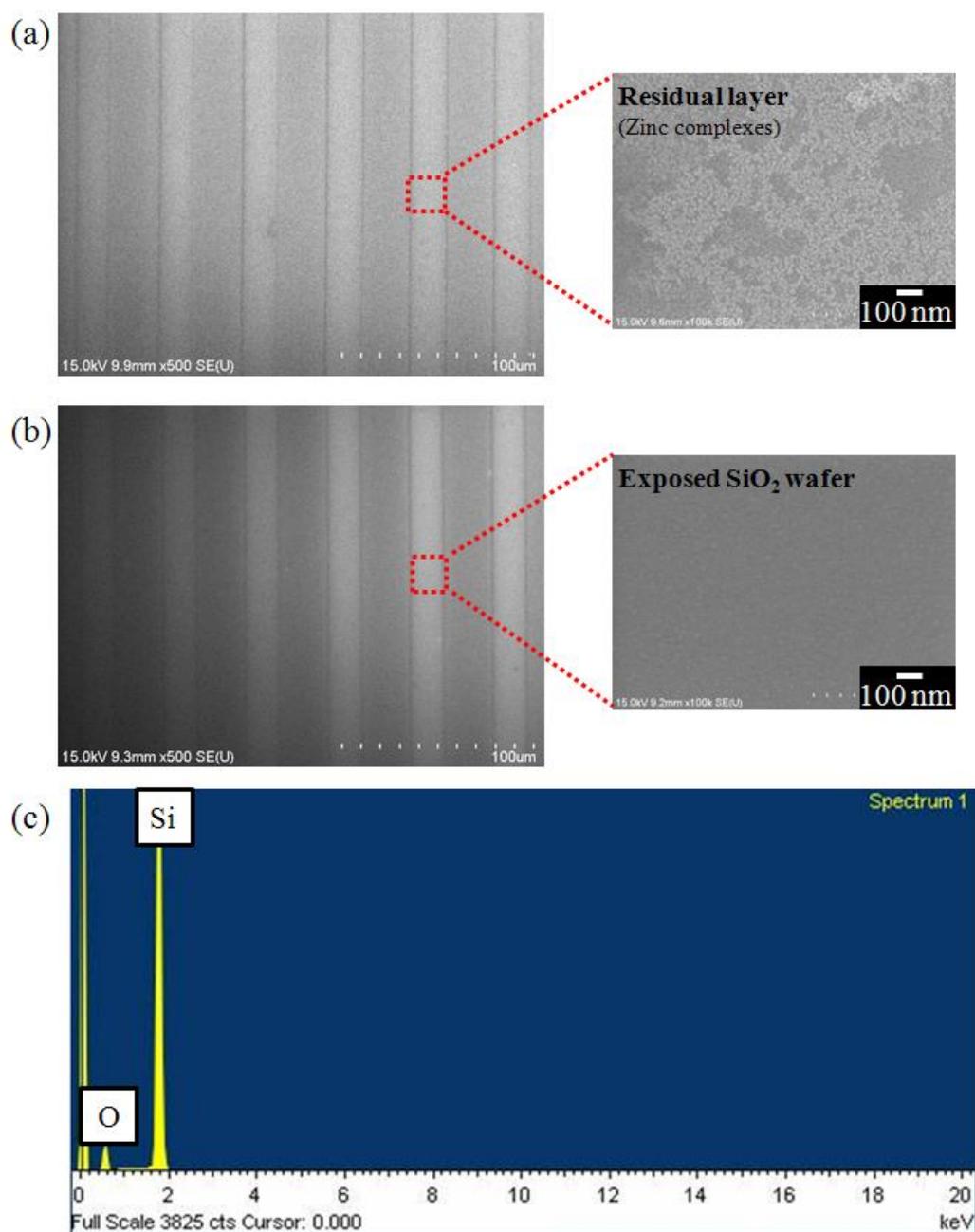
**Observation of the patterned ZnO thin films and TFTs.** The patterned ZnO thin films and the unpatterned ZnO thin films were characterized by a field emission scanning electron microscope (FE-SEM; S-4800: Hitachi), an atomic force microscope (AFM; XE100: PSIA) and a transmission electron microscope (TEM; JEM-2100F: JEOL) The current–voltage measurements were executed under ambient conditions by an Agilent 4155B semiconductor parameter analyzer.

[1] S. Y. Park, B. J. Kim, K. Kim, M. S. Kang, K. H. Lim, T. Il Lee, J. M. Myoung, H. K. Baik, J. H. Cho, Y. S. Kim, *Adv. Mater.* **2012**, *24*, 834.

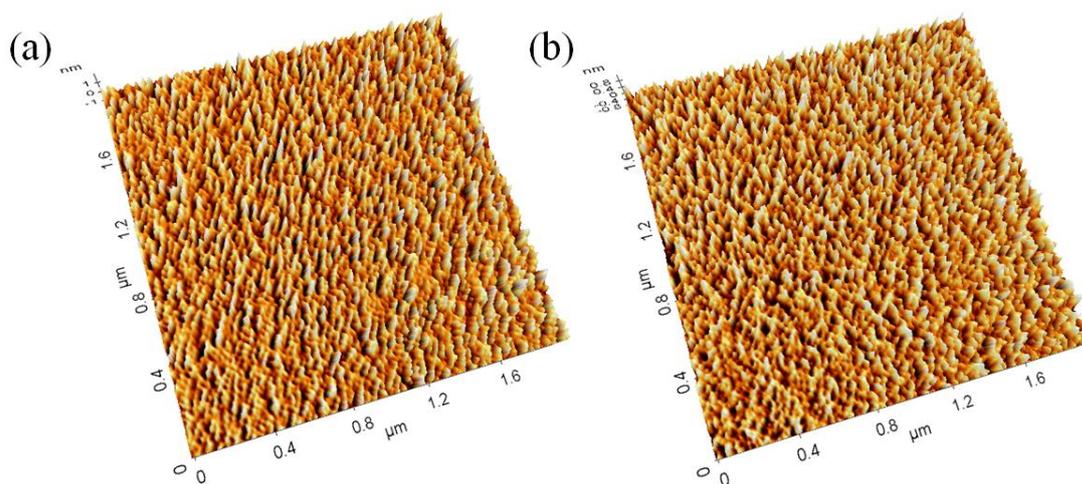
**Figure S1.** (a) ZnO thin films after the contact with the intrinsic PDMS stamps and washing with de-ionized water. (b) Patterned ZnO thin films after the contact with the soaked PDMS stamps in the binary solvent composed of ammonia water and methanol and washing with de-ionized water.



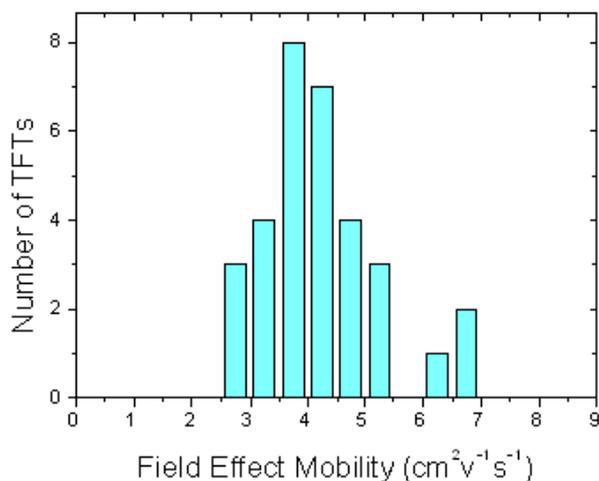
**Figure S2.** (a) ~ (b) SEM images of micro-patterned ZnO thin films. (a) the micro-patterned ZnO films after chemical imprinting. The SEM images showed the aqueous ammonia-ZnO salt forms on the region of chemical imprinting. (b) after rinsing with de-ionized water, no residual was shown in SEM image. (c) the EDS data of selectively exposed SiO<sub>2</sub> wafer indicated no residual layer of ZnO films.



**Figure S3.** Atomic force microscopy images. (a) spin-coated ZnO thin films. (b) micro-patterned ZnO thin films. Root mean square roughness of spin-coated ZnO thin films were 0.324nm; the patterned ZnO thin films were 0.222nm.



**Figure S4.** The field effect mobilities of micro-patterned Li doped ZnO TFTs array obtained as one-run process after sintering at 300°C.



The 36 TFTs were measured from the micro-patterned Li doped ZnO TFTs array fabricated by one run. Except 3 points, the electrical characteristics of TFTs were successfully obtained, and the average field effect mobility was  $4.16\text{cm}^2\cdot\text{v}^{-1}\cdot\text{s}^{-1}$  with a standard deviation of  $1.19\text{cm}^2\cdot\text{v}^{-1}\cdot\text{s}^{-1}$  on 33 points of TFTs. Although the TFTs were fabricated in the ambient condition, over 90% of the micro-patterned Li doped ZnO TFTs showed good electrical characteristics.

**Figure S5.** Hysteresis of the electrical characteristics on micro-patterned Li doped ZnO TFTs.

