Supporting Information

Aqueous Zinc Ammine Complex For Solution-processed ZnO Semiconductors in Thin Film Transistors

Si Yun Park\(^{a\dagger}\), Sunyoung Kim\(^{a\dagger}\), Jeeyoung Yoo\(^a\), Keon-Hee Lim\(^a\), Eungkyu Lee\(^a\), Kyongjun Kim\(^a\), Joohee Kim\(^a\), and Youn Sang Kim\(^{a,b*}\)

\(^a\)Program in Nano Science and Technology, Graduate School of Convergence Science and Technology, Seoul National University, Seoul 151-744, Republic of Korea. Tel: +82-31-888-9143; E-mail: younskim@snu.ac.kr.

\(^b\)Advanced Institutes of Convergence Technology, 864-1 Iui-dong, Yeongtong-gu, Suwon-si, Gyeonggi-do 443-270, Republic of Korea.

\(^\dagger\) Equally contributed to this work

Experimental Section

Precipitated Zn(OH)\(_2\). The synthesis of zinc ammine complex using precipitated Zn(OH)\(_2\) is as follows; Zn(NO\(_3\))\(_2\)·6H\(_2\)O (Sigma Aldrich, 98%) in distilled H\(_2\)O to a total concentration of 0.5 M Zn. Ten milliliters of 2.5 M NaOH (Duksan Pure Chemicals, 95%) was added dropwise to the zinc nitrate solution while stirring at 500 rpm for 5 min. To minimize residual Na\(^+\) and NO\(_3^-\), the precipitated zinc hydroxide was centrifuged four times with DI water. Following final centrifugation, the supernatant was removed and dried at vacuum oven at 50 °C to get the Zn(OH)\(_2\) powder from synthesis.
**Fabrication and observation of TFTs.** All prepared solutions were filtered through a hydrophilic 0.45 μm PTFE syringe filter and spin-coated with 3000 rpm for 30 s on SiO$_2$/Si substrates for TFTs fabrication. A thermally grown 200 nm SiO$_2$ (capacitance ~17 nF/cm$^2$) and heavily doped Si substrate were used as a gate insulator and gate electrode, respectively. After deposition, the ZnO film was annealed immediately on preheated hot plate at 300 °C for 1 h. The 100 nm of Al source and drain electrodes are thermally evaporated with mask (Width/Length = 1000 μm / 50 μm). The current-voltage measurements were executed under dark ambient conditions (Agilent 4155B) and the surface morphologies were characterized by an Atomic Force Microscope (AFM; XE100: PSIA). HRTEM (JEM-2100F) was used for the investigation of the ZnO crystal structure. To observe the chemical composition in the ZnO semiconductor films, XPS (SIGMA PROBE: ThermoVG) and H-NMR (600MHz, VNS, VARIAN).
Figure S1. Photographs of zinc ammine complexes of different zinc oxide sources. When the as-prepared solution was refrigerated for 1 day, the zinc ammine complex (90 mM) in 10ml ammonia water was dissolved well. (a) device 1 (ZnO powder), (b) device 2 (intrinsic Zn(OH)$_2$), (c) device 3 (precipitated Zn(OH)$_2$).
Figure S2. $^1$H-NMR spectra of zinc ammine complex with various zinc oxide sources. The $^1$H NMR data were obtained in ppm (δ) from the internal standard and chemical shift. (a) ZnO powder, (b) intrinsic Zn(OH)$_2$, (c) precipitated Zn(OH)$_2$. 
Figure S3. Output characteristic of various ZnO TFTs. (a) device 1 (ZnO powder), (b) device 2 (intrinsic Zn(OH)$_2$), (c) device 3 (precipitated Zn(OH)$_2$).
Figure S4. The histogram of field effect mobility of various ZnO TFTs as one run at 300 °C.

We fabricated 4 samples of each device. One sample had 15 TFTs (60 TFTs). 75 % of ZnO TFTs (45 TFTs) worked normally.
Figure S5. The hysteresis behavior of various ZnO TFTs with SiO_2 gate dielectric. (a) device 1 (ZnO powder), (b) device 2 (intrinsic Zn(OH)_2), (c) device 3 (precipitated Zn(OH)_2).
Figure S6. (a), (b) and (c) Evolution of the linear transfer curves of various ZnO TFTs, as a function of negative bias stress time (40 V, 0-6000 s), (d) Relative threshold voltage shift ($\Delta V_{th}$) of various ZnO TFTs as a function of stress time.
Figure S7. Images of FFT-SAED patterns of various ZnO films made from zinc ammine complex prepared with various zinc oxide sources (ZnO powder, intrinsic Zn(OH)$_2$ and precipitated Zn(OH)$_2$) by HR-TEM. (a) ZnO powder, (b) intrinsic Zn(OH)$_2$, (c) precipitated Zn(OH)$_2$. 