Electronic Supplementary Information

Reliability enhancement in thin film transistors using Hf and Al co-incorporated ZnO active channels deposited by atomic-layer-deposition

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S1. Device characteristics of HAZO TFTs depending on Al concentration

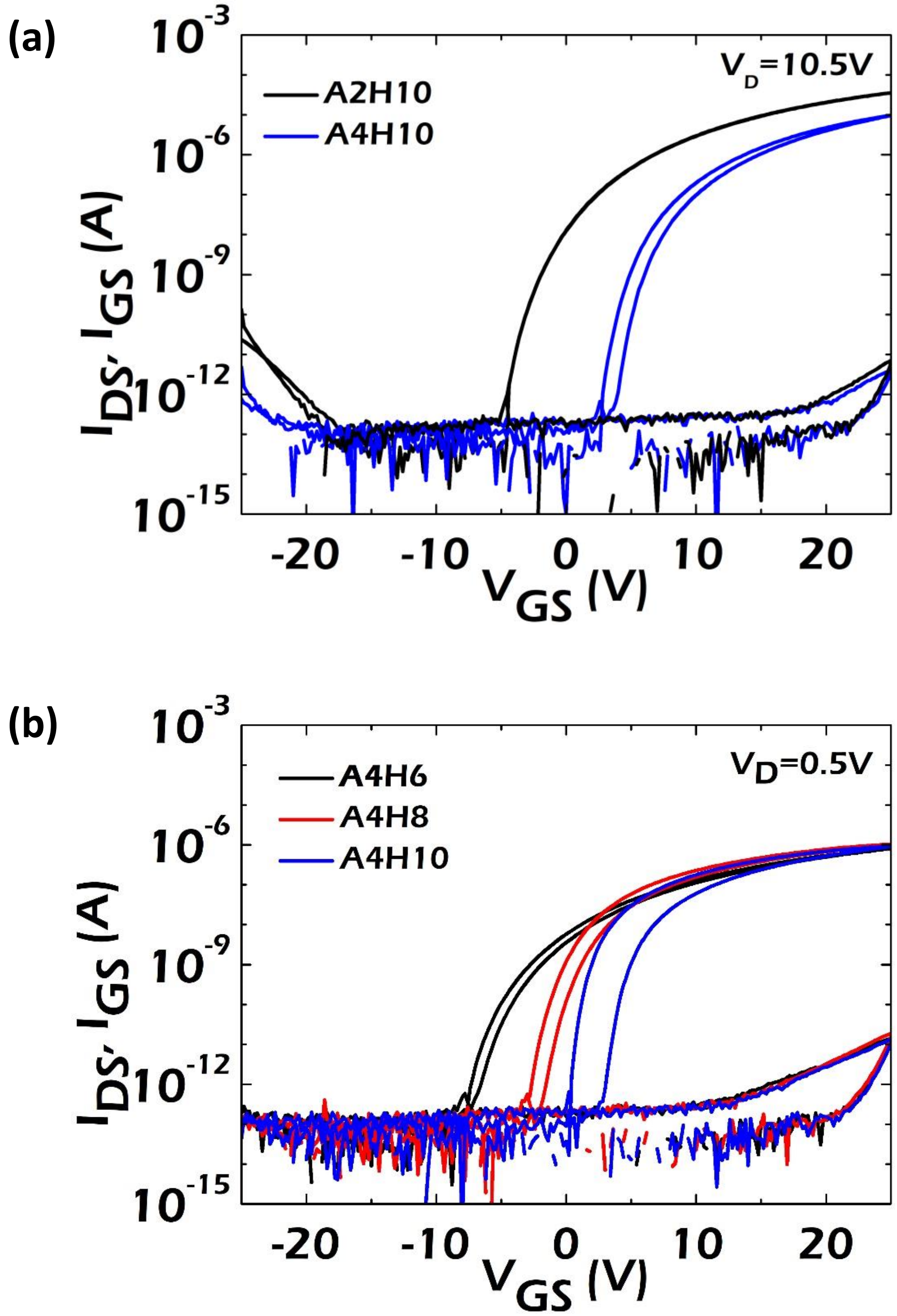

Figure S1. Comparisons of I_{DS}-V_{GS} characteristics and I_{GS} leakage components of the fabricated devices with (a) different Al concentrations when the incorporated Hf cycles were fixed as 10, and with (b) different Hf concentrations when the incorporated Al cycles were fixed as 4.

To investigate the effects of Al doping contents on the device characteristics, the incorporated Al cycles were varied from 2 to 4. Figure S1(a) represents the I_{DS}-V_{GS} curves of the HAZO TFTs when the ALD cycle numbers for Al incorporation were varied to 2 (Device A2H10) and 4 (Device A4H10) with keeping the ALD cycle numbers of Hf incorporation at 10. The A4H10 showed relatively high V_{TH} value compared with that of the A2H10. As mentioned in the manuscript, the electrical conductivity of the HAZO film can be effectively modulated by controlling the incorporated Al contents. However, when the ALD cycle numbers for Al incorporation increased to 4, undesirable large hysteresis was observed in the transfer characteristics when the V_{GS} was swept from −25 to 25 V in forward and reverse directions, as shown in the Figure S1(b), in which the I_{DS}-V_{GS} curves of the HAZO TFTs when the ALD cycle numbers for Hf incorporation were varied to 6 (Device A4H6), 8 (Device A4H8), and 10 (Device A4H10) with keeping the ALD cycle numbers of Al incorporation at 4. These characteristics were supposed to be ascribed from the incomplete dissociation of TMA source during the ALD process. In other words, the ALD process window for the TMA precursor used as Al sources is known to be in the temperature range from 200 to 250 ^{o}C. Therefore, the thermal energy was not sufficient for completely decomposing the TMA sources at an ALD temperature of 100 ^{o}C for the HAZO depositions. The byproducts such as $CH_3CH_2^-$ anions, which are generated by incomplete dissociation of precursors during the ALD process for the HAZO thin films, can induce considerable amounts of charge-trap centers and hence the hysteretic behaviors happen in I_{DS}-V_{GS} curves of the fabricated devices. Thus, in this manuscript, we focused on the effects of Hf incorporation into the Al-doped ZnO channel on the TFTs operations when the ALD cycle numbers for Al incorporation was 2.

S2. Surface morphology analysis of HAZO thin films by AFM images

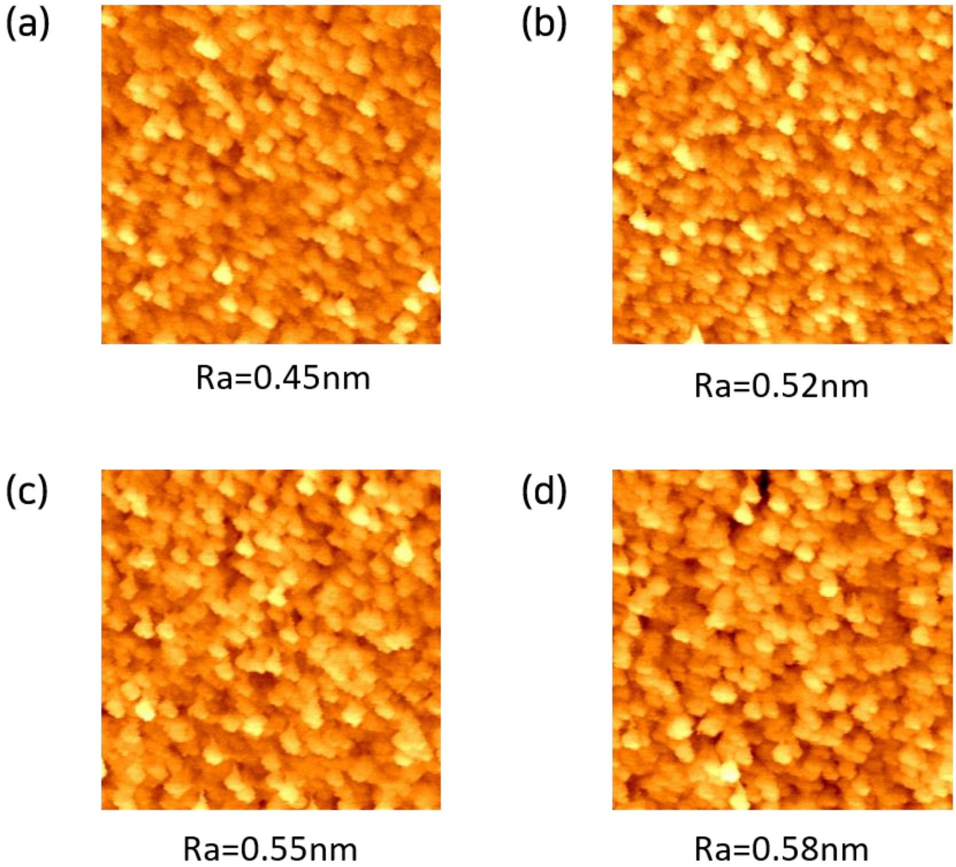

Figure S2. AFM images and R_a values of the HAZO thin films prepared with the number of inserted Hf cycles were (a) 4, (b) 6, (c) 8, and (d) 10 during the ALD process, respectively. The scan size was 2 x 2 μm^2.

Figure S2 (a)-(d) show the AFM images of the HAZO thin films when the ALD cycle numbers for Hf incorporation were varied to 4, 6, 8, and 10, respectively. The R_a values were 0.45, 0.52, 0.55, and 0.58 nm as the number of Hf cycle increased from 4 to 10. The HAZO films had uniform and smooth surface showing the R_a values of around 0.5 nm regardless of the film composition, although the R_a values showed a slight increasing trend with increasing the Hf concentration. These results originated from the amorphous natures of the HAZO thin films irrespective of the film compositions, as confirmed in XRD analysis discussed in Supplemental Information 3. Thanks to good surface morphologies, the ALD HAZO films could be concluded to be suitably applied to active channels in TFTs.

S3. Crystallinity of HAZO thin films by XRD analysis

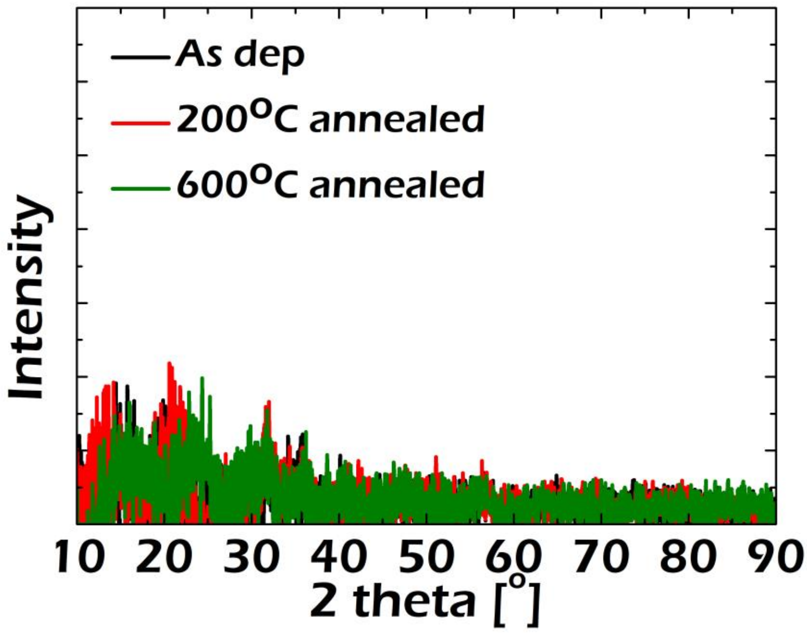

Figure S3. XRD spectra of the ALD HAZO thin films depending on the annealing temperature.

The XRD analysis was performed to examine the crystallinity of the HAZO thin films prepared by ALD at 100 ^oC. Figure S3 shows the XRD patterns for the HAZO films, which were identical to the active channels of the device H10 after the thermal treatments performed at 200 and 600 ^oC. Generally, the ZnO thin films were known to show a c-axis (002)-preferred crystalline orientation due to thermodynamically favor in the wurtzite structure under the equilibrium state. On the contrary, in the HAZO thin films, there were no diffraction peaks even after 600 ^oC annealing for 1 h. From these results, it can be suggested that the HAZO thin films remain as amorphous phases even when the TFT devices undergo thermal effects during the fabrication and/or post-annealing processes. Consequently, the amorphous nature of the HAZO thin films will be more desirable for the use of active channels in TFTs unlike the ZnO channel.

S4. Reliability properties under negative bias temperature stress condition of HAZO TFTs

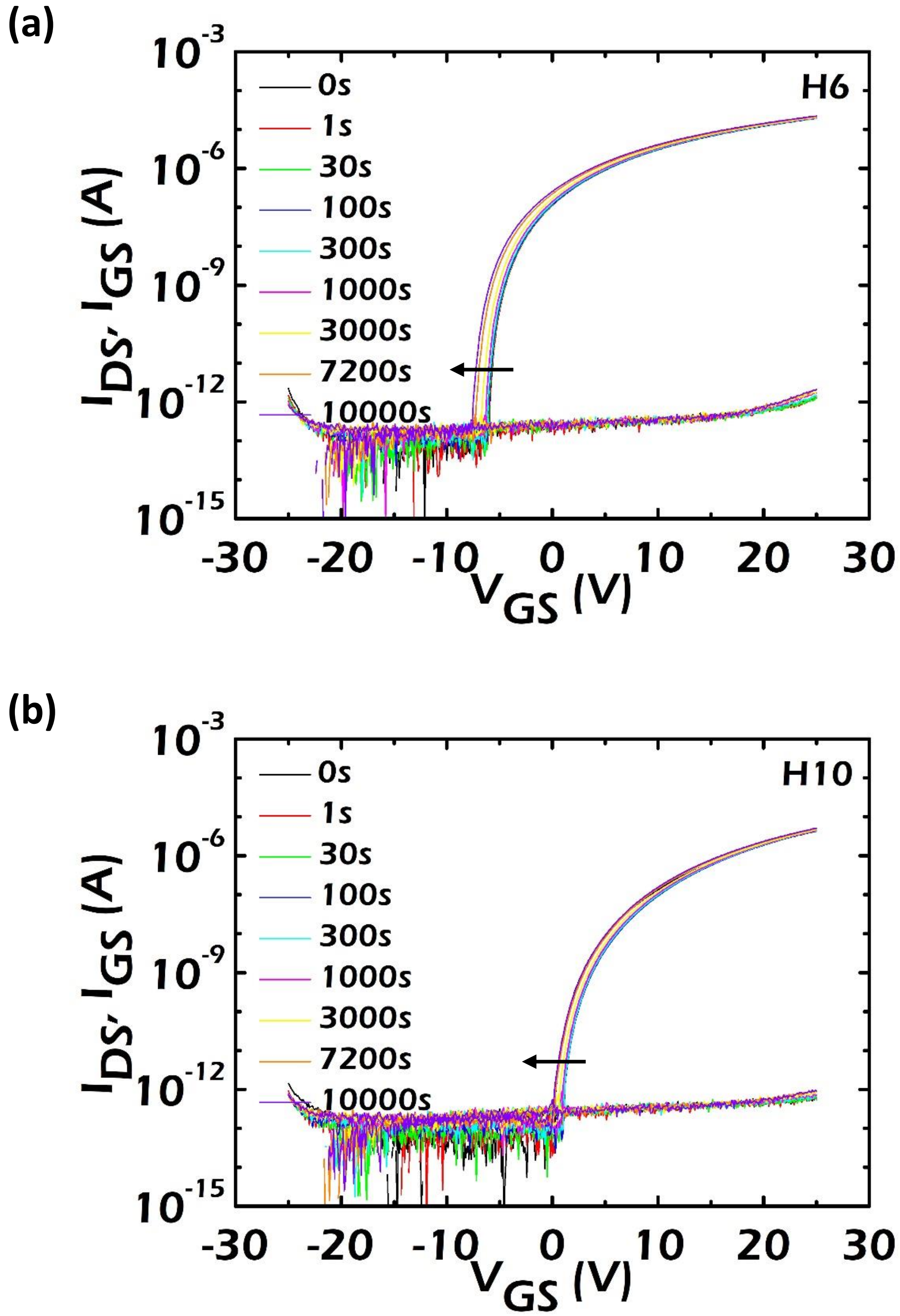

Figure S4. Variations in the I_{DS}-V_{GS} curves under the NBTS conditions of the HAZO TFTs at a V_{GS} of -20 V with a substrate temperature of 80 ^{o}C when the Hf cycles were varied to (a) 6, (b) 10, respectively.

The negative bias temperature stress (NBTS) stabilities were also examined. Figures S4a–b show the variations in transfer characteristics under the NBTS for the H6, and H10, respectively. The devices were stressed under a V_{GS} of -20 V and V_{DS} was fixed at 10.5 V while keeping the substrate temperature at 80 ^{o}C for 10,000 s. The ΔV_{TH}'s were -1.5, and -0.9 V for the H6 and H10, respectively, showing no marked degradation in carrier mobility and SS. Also, the ΔV_{TH} showed decreasing trend with increasing the incorporated Hf contents. Furthermore, the device H10 exhibited the ΔV_{TH}'s smaller than -1 V even at a high temperature of 80 ^{o}C. In conclusion, our fabricated HAZO TFTs exhibited stable operations even under the NBTS conditions showing small ΔV_{TH}'s without SS degradation.