

Supporting Information

Study on the influence of local doping in ALD Al:ZnO thin film transistors

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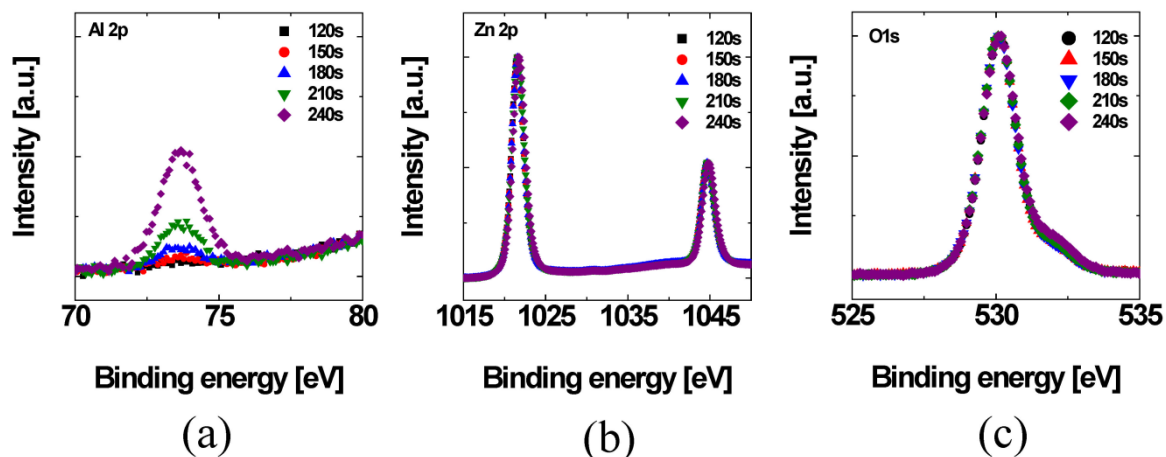
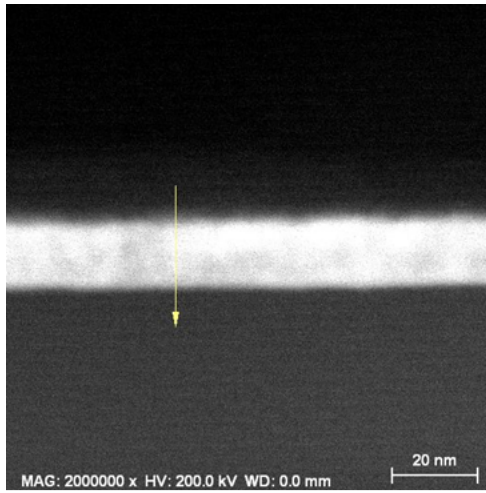
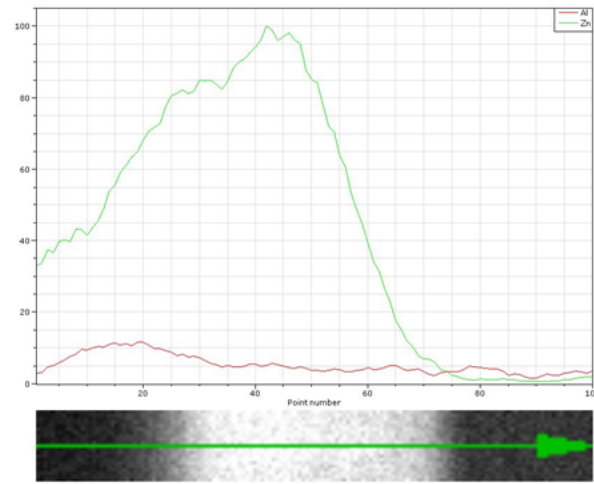


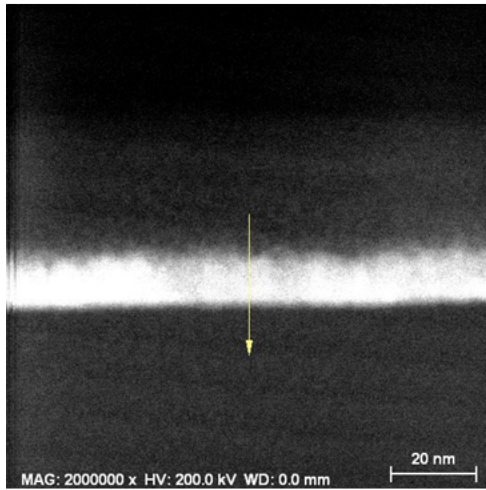
Figure S1. High resolution XPS profiles of the spectra given in Figure 1. (a), (b), and (c) show the profiles for Al 2p, Zn 2p, and O 1s peaks, respectively. As can be seen from the figures, the change in the profiles of Zn and O are relatively small compared to the evident appearance of Al. Careful examination shows that the Zn 2p 3/2 peak shows a shift from 1021.64 eV to 1021.72 eV, at the etch time of 210s and 240s. Peak binding energy values show no change at etch time shorter than 210s, implying that a significant amount of Al is required in the matrix for a notable shift to occur in the Zn 2p peak. A similar trend is observed in the O 1s peak, which only shows the precursor of a shoulder at higher binding energies when a significant amount of Al is present in the vicinity (etch time 240s).



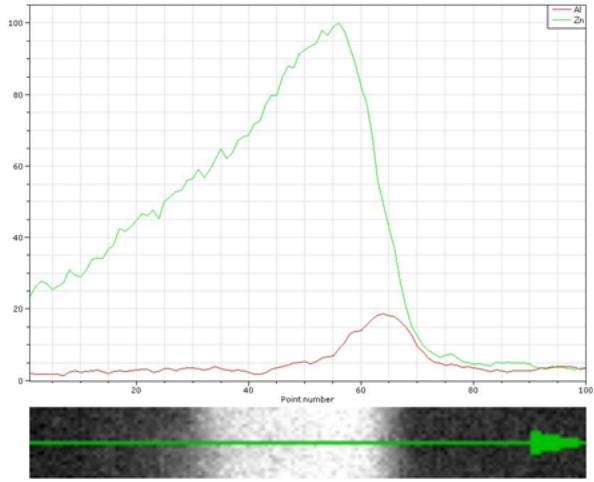
(a)



(b)



(c)



(d)

Figure S2. TEM and corresponding EDX line profiles of cases where doping was performed at the (a) surface and (b) gate dielectric/semiconductor interface regions of the film. As can be seen in the figures, it is clear from the EDX line profiles that doping can be achieved in any position with modest controllability. The TEM images could not be achieved as clear as in the case where doping was performed in the middle of the film mostly due to ion milling proficiencies in the top case and contrast was not as strong in the bottom case due to the SiO₂ layer lying directly beneath the ZnO layer.

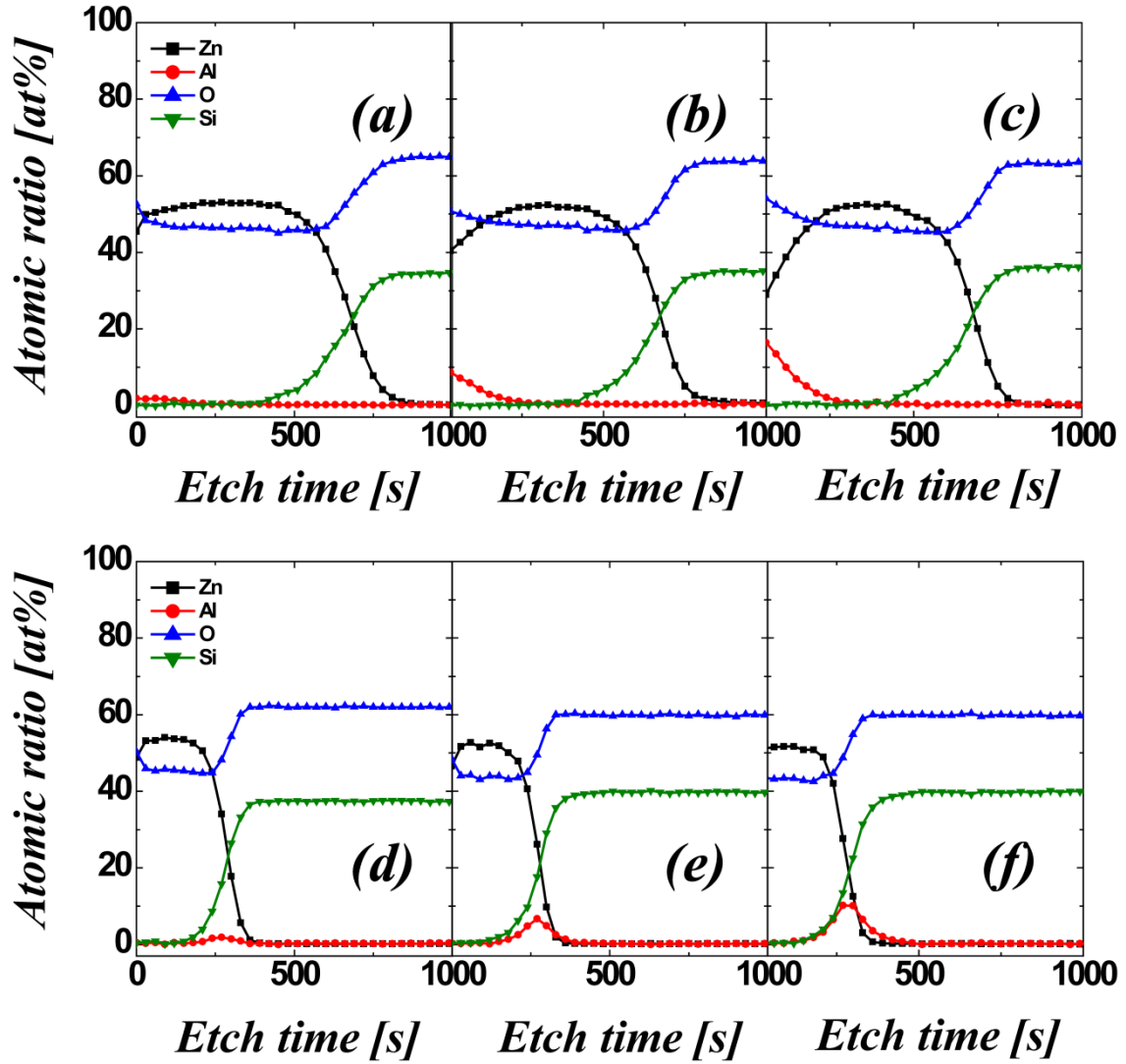


Figure S3. XPS depth profiles of locally doped films with various doping concentrations in (a)-(c) the bulk depletion regions and (d)-(f) the accumulation regions for 3, 7, and 17 at% Al cases, respectively. As can be seen from the figures, stringent concentration control can be achieved in each region of the film when experimental parameters are varied. The discrepancy found in the plotted chemical composition comes from the presence of a significant amount of Si detected when Al is near the gate dielectric interface. However, when comparing only the cation ratio between Al and Zn the two cases are in good agreement with each other, with ratios of $\sim 9:1$, $4:1$, and $2:1$ for the cases of 3, 7, and 17 at% Al doping in the doped regions.

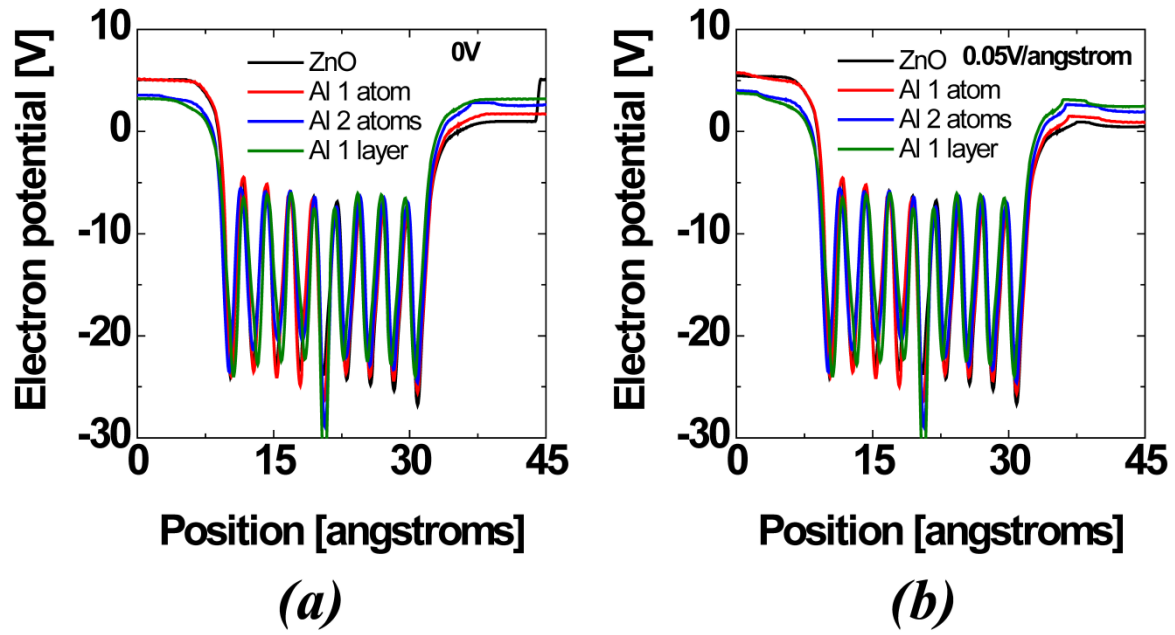


Figure S4. Electron potential profiles of the slabs generated by first principle calculations according to relative position within the slab. (a) shows the case for when no external field is applied to the slabs and (b) shows the case where an external field of $0.05\text{V}/\text{\AA}$ is applied to the structures. As can be seen in the figures the potential the electron experiences shows a sharp decline when Al atoms substitute Zn in the matrix. In both (a) and (b), the overall potential profile of the slabs with less Al content within them show a gradual potential change due to the polar character of ZnO, which is almost non-existent when 1 full Al layer is implemented.

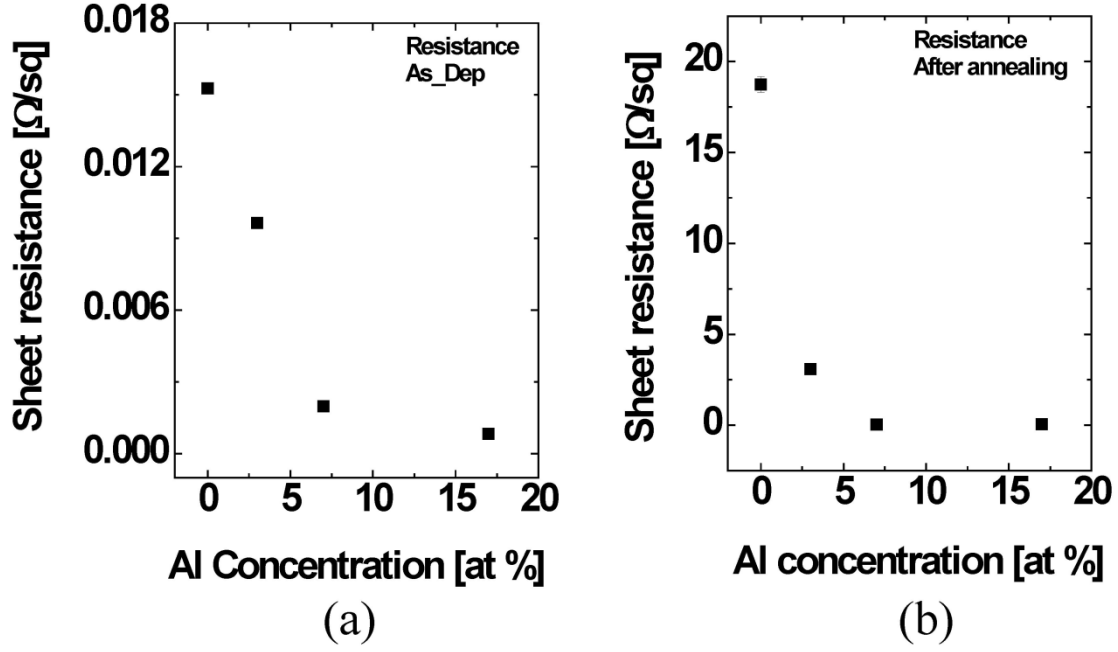


Figure S5. Sheet resistance values of bulk ZnO and Al:ZnO films with various Al content (a) before and (b) after thermal annealing. In both figures, a clear trend can be observed as the Al content increases. This type of behavior is widely accepted to come from the combination of many defect chemistry reactions that provide excess electrons, including but not limited to those such as $2\text{Al} + 3\text{ZnO} \rightarrow \text{Al}_2\text{O}_3 + 3\text{Zn} + 3\text{V}_\text{O}$ or $\text{Al}_2\text{O}_3 + \text{V}_\text{O} \rightarrow 2\text{Al}_\text{Zn} + 3\text{O}_\text{O}$. This coincides well with the first principle calculation results provided in the main article, which clearly show that excess electrons are generated when Al is substituted into the position of Zn in the ZnO matrix. As can be seen in (b) even when the films are subjected to post deposition annealing, the films with large Al content have low resistance values, which imply difficulty in depletion or modulation in a TFT structure. This agrees well with the profiles observed in figures 3 and 4 in the main article.