(Supplementary Information)

Solution-Processed Amorphous Hafnium-Lanthanum Oxide Gate Insulator for Oxide Thin-Film Transistors

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Fig. S1 ATR-FT IR spectroscopy of the HfLaO_x films at different temperatures.



Fig. S2 The ln(J) versus $E^{1/2}$ plots for HfLaO_x films were measured at various temperature ranges.

The solid line shows the linearly fitted graphs by thermionic emission model. To figure out the conduction mechanism of $HfLaO_x$ film, additional experiments were carried out. We

checked the temperature dependency of $HfLaO_x$ films' leakage current density and matched it with various conduction mechanisms in MIS capacitors. As a result, thermionic emission model could explain the conduction mechanism of the $HfLaO_x$ films as shown in the above figure.

Thermionic emission model;

$$J = A^{**}T^2 \exp\left[\frac{-q(\emptyset_B - \sqrt{\frac{q\epsilon_i}{4\pi\epsilon_i}})}{kT}\right]$$

 A^{**} = effective Richardson constant. ϕ_B = barrier height. ϵ_i = electric field in insulator. ε_i = insulator permittivity. $\sqrt{\frac{q}{4\pi\varepsilon_i}}$ = constants. ¹)

Thermionic emission is responsible for carrier transport between the metal-insulator barrier or the insulator-semiconductor barrier. In the increment of temperature in metal, the electrons would be moving faster and some could escape from their bulk. We don't fully understand why these films show the thermionic emission but we carefully suggest that the existence of native oxide between silicon substrate and $HfLaO_x$ dielectric film made the oxide barrier and led thermionic emission.

In the other previous report (*C. An, M. S. Lee, J. Choi, and H. Kim, Appl. Phys., Lett. 2009, 94, 262901*), the ALD deposited HfLaO_x films were shown to follow the Poole-Frenkel emission model. It is interesting that two results were shown different conduction mechanism. However, in this study, we just showed that the thermionic emission model could explain the electrical conduction of solution-processed HfLaO_x dielectric films. We remain the detail study of conduction mechanism as a further study.



Fig. S3 ZnO TFT device structure with $HfLaO_x$ as a dielectric layer.



Fig. S4 Hysteresis of Li-ZnO TFTs on (a) 200 nm SiO_2 and (b) HfLaO_x gate insulator.



Fig. S5 Output and transfer characteristics of ZnO TFTs with 200 nm SiO₂ gate insulator. (*p*-type Si / 200 nm SiO₂ / 5 nm ZnO / 100 nm Al) ((a), (b): V_{DS} , $V_{GS} = 60$ V, (c), (d): V_{DS} , $V_{GS} = 5$ V, (e),(f): V_{DS} , $V_{GS} = 1$ V)



Fig. S6 Leakage current density versus voltage of metal oxide films measured with metalinsulator-metal structure. (*p*-type Si substrate as bottom electrode / metal oxide dielectric layer / aluminium top electrode).



Fig. S7 Energy-dispersive X-ray spectroscopy (EDX) results of a SiO₂ interlayer (SiO₂ layer in Figure 5(a))