Supplementary Information:

Flexible Transparent Layered Metal Oxides for Organic Devices

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Supplementary Text

Theoretical optical modeling calculations

Two types of transfer matrix are used in calculation: Transmission matrix *L*, which indicates the phase change in the same layer and is related to the thickness of material and to the angle between the surface normal and the wave vector; Interface matrix *I*, which indicates the reflection and refraction at interface of two media and is obtained by Fresnel reflection and transmission coefficients. Optical electric field *E* coming out of the OLED can be calculated by the methods descripted in reference.[1]

To investigate the weak microcavity effect in OLED devices, microcavity effect factor is introduced[2] in calculation. The output optical electric field E_{out} can be described by the following equation:

$$E_{out} = \sqrt{\frac{[1 + R_t + 2\sqrt{R_t}cos(- \varphi_t + \frac{4\pi n_{source} d cos(\theta)}{\lambda}}{(1 - \sqrt{R_t R_b})^2 + 4\sqrt{R_t R_b} sin^2(\frac{\Delta\varphi}{2})}} E \qquad (1)$$

R_t and R_b are the reflectivity of cathode and anode respectively, n_{source} is the refractive index of emitting layer, and *d* is the distance between dipole source and cathode. Δφ is the total phase shift in the cavity, φ_t is the phase shift at cathode. So, the relative spectral power distribution $P(\lambda)$ is calculated by:

$$P(\lambda) = |E_{out}|^2 \qquad (2)$$

There is a little deviation between the calculated and experimental result, this may be due to the measurement error of complex refractive index.

Discussion on Gibbs free energy for chemical reaction between Al and MoO_3

The XPS data in the main text provide a strong evidence that the redox reaction occurs at the Al-MoO_3 interface. To study the thermodynamics involved in this reaction, we explored the Gibbs free energy change of the reaction below:

$$2Al(s) + 3MoO_3(s) \rightarrow Al_2O_3(s) + 3MoO_2(s) \qquad (3)$$

The standard Gibbs free energies (or formation energies) of involved oxides are taken from literature[3] and shown as below:

$$2Al(s) + \frac{3}{2}O_2(g) \rightarrow Al_2O_3(s) \qquad \Delta G^0 = -378.2\ kcal/mol \qquad (4)$$

$$Mo(s) + O_2(g) \rightarrow MoO_2(s) \qquad \Delta G^0 = -127.39\ kcal/mol \qquad (5)$$

$$Mo(s) + \frac{3}{2}O_2(g) \rightarrow MoO_3(s) \qquad \Delta G^0 = -159.66\ kcal/mol \qquad (6)$$

The standard Gibbs free energy change of the reaction (3) can be calculated according to the reaction (4) (5) and (6):

$$2Al(s) + 3MoO_3(s) \rightarrow Al_2O_3(s) + 3MoO_2(s) \qquad \Delta G = -281.39\ kcal/mol \qquad (7)$$

Thus, the redox reaction is energetically favorable and occurs spontaneously at the interface.

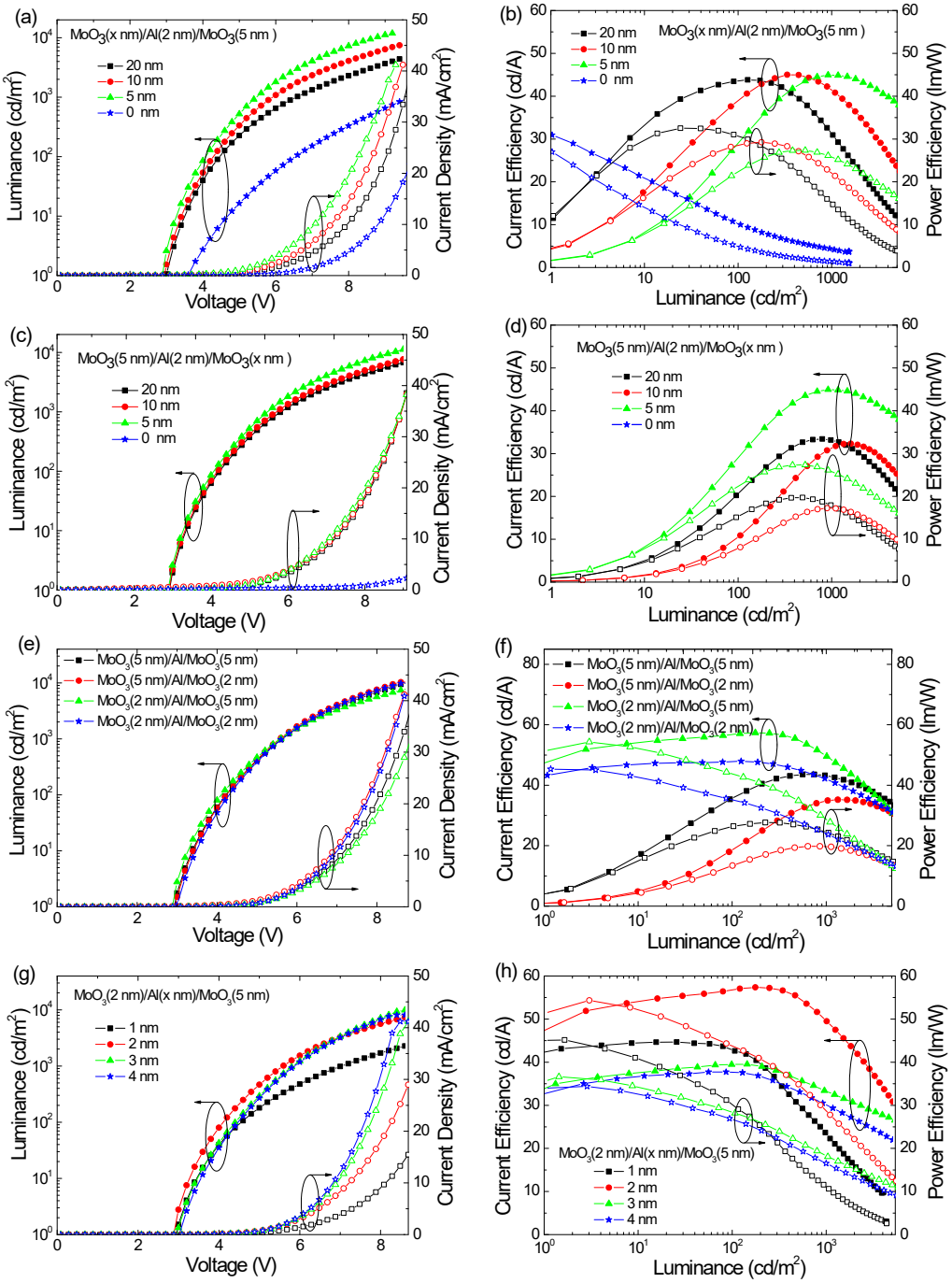

Fig. S1. The MoO_3/Al/MoO_3 based OLED devices characteristics of 2×12 mm^2 active area. (a) Luminance-current -voltage (L-I-V), (b) current and power efficiency-luminance characteristics with various thicknesses of outer MoO_3 layer. (c) L-I-V, (d) current and power efficiency- luminance characteristics with various thicknesses of inner MoO_3 layer. (e) L-I-V, (f) current and power efficiency- luminance characteristics with various thicknesses of outer and inner MoO_3 layer. (g) L-I-V, (h) current and power efficiency-luminance characteristics with various thicknesses of Al layer.

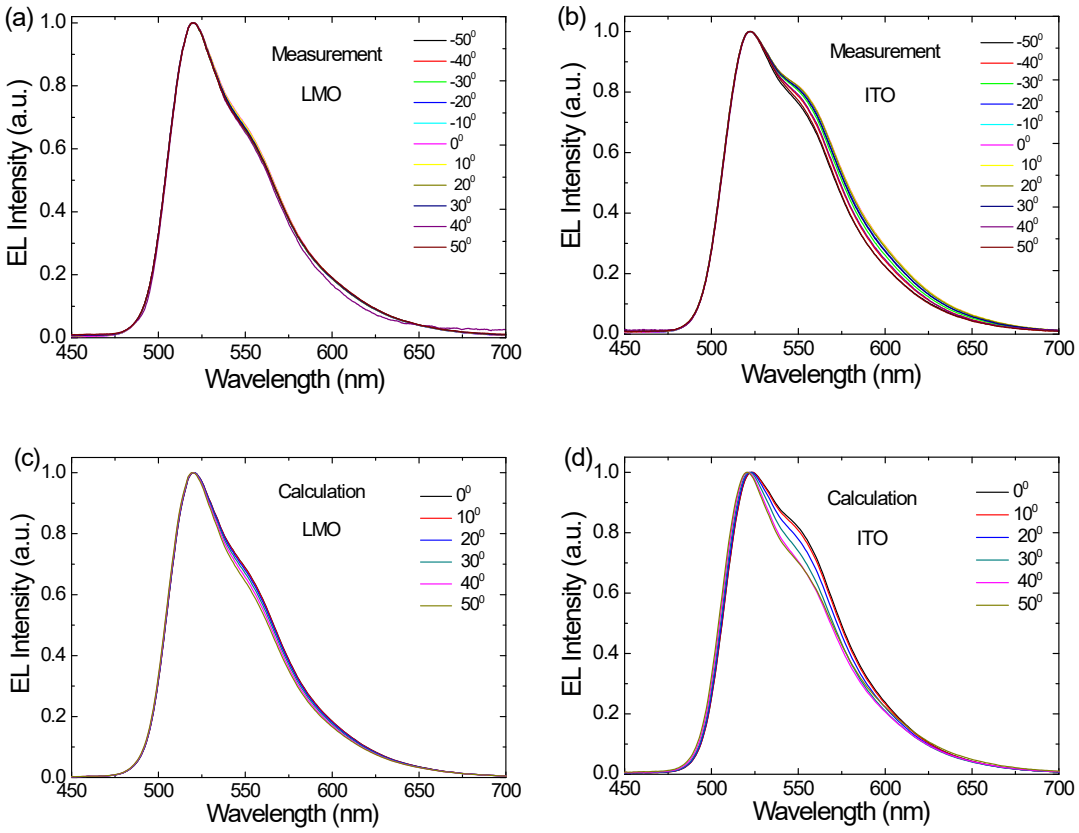

Fig. S2. Measurement emission spectra at various angles from an OLED with (a) LMO electrode, (b) ITO electrode. Calculated spectral radiance for OLED with (c) LMO electrode, (d) ITO electrode.

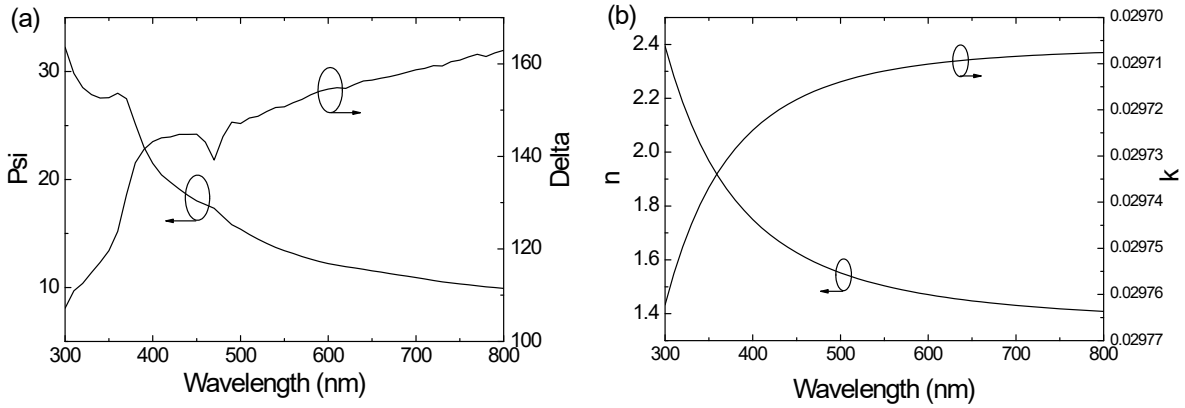

Fig. S3. The optical constants (a) Psi, Delta (b) n, k of LMO in optical model. LMO on Si substrate at the incident angle of 70° measured by a spectroscopic ellipsometer.

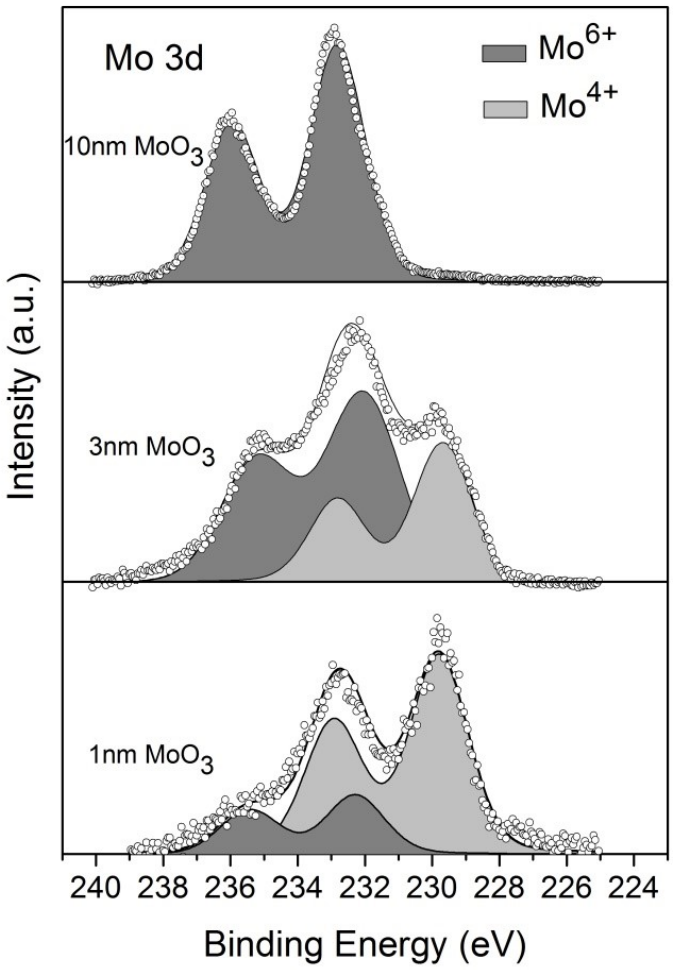

Fig. S4. Mo 3d XPS spectra for MoO_3 films of various thicknesses deposited onto Al substrate.

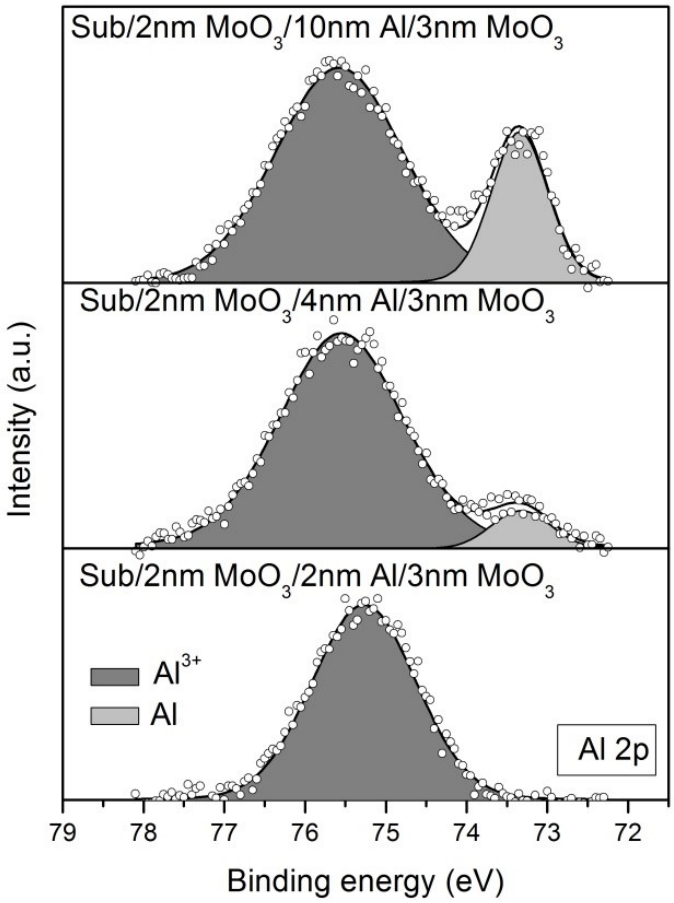

Fig. S5. Al 2p XPS spectra for 2, 4, and 10 nm Al. The peak centered ~ 73.3 eV and ~ 75.5 eV represent metallic Al and Al 3+ (Al_2O_3), respectively.

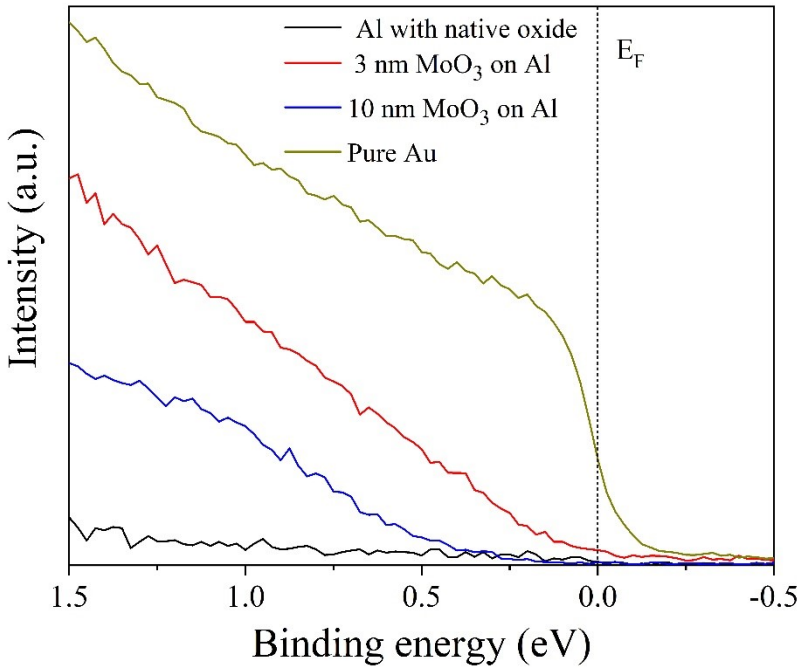

Fig. S6. UPS valence band spectra of MoO_3 films on Al substrates. The band edge is indicated by a dashed line. A gold UPS spectra showing a clear Fermi edge is also plotted as a reference.

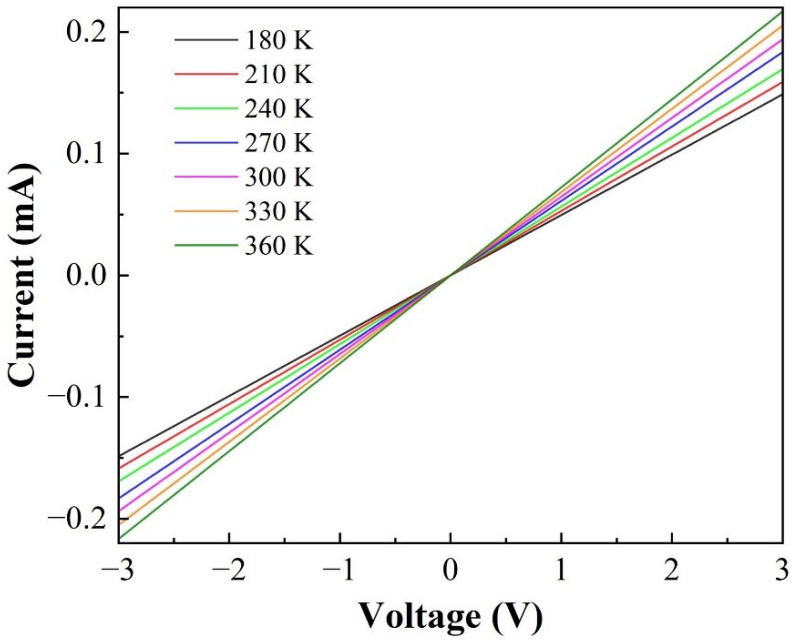

Fig. S7. Current–Voltage characteristics range from 180 K to 360 K of Glass/ Al finger electrodes (100 nm)/MoO_3 (2 nm)/Al (2 nm)/MoO_3 (5 nm)/NPB (100 nm).

References

1. Z. B. Wang, M. G. Helander, X. F. Xu, D. P. Puzzo, J. Qiu, M. T. Greiner, and Z. H. Lu, *Optical design of organic light emitting diodes*, J. Appl. Phys., 2011, **109**, 053107.

2. M. Thomschke, R. Nitsche, M. Furno, and K. Leo, *Optimized efficiency and angular emission characteristics of white top-emitting organic electroluminescent diodes*, Appl. Phys. Lett., 2009, **94**, 083303.

3. P. Patnaik, *Handbook of Inorganic Chemicals*, 2002, McGraw-Hill, New York.