**High-Hole-Mobility Metal-Organic Framework as Dopant-Free Hole Transport Layer for****Perovskite Solar Cells**

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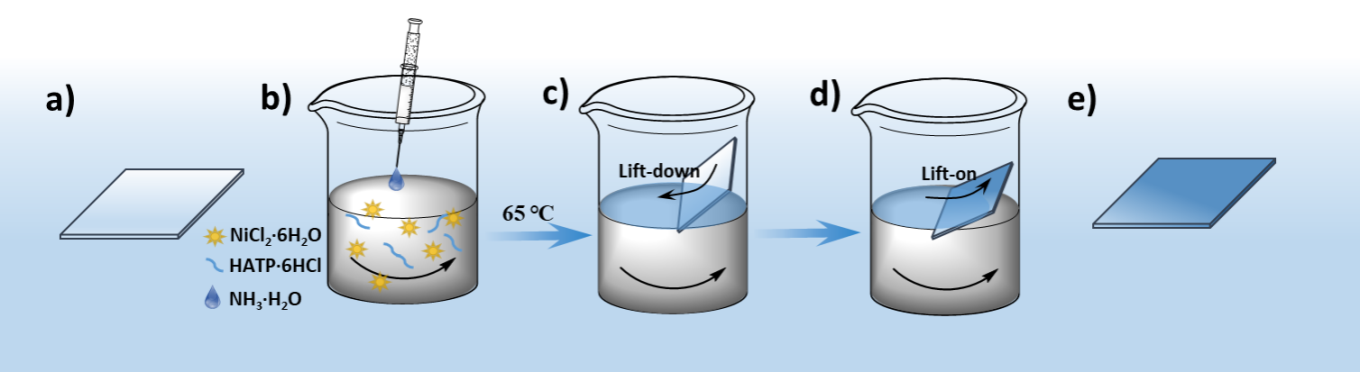
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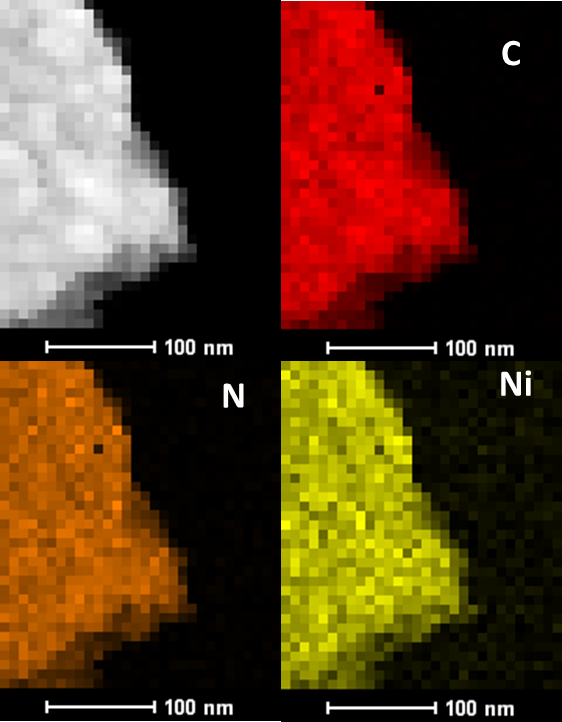
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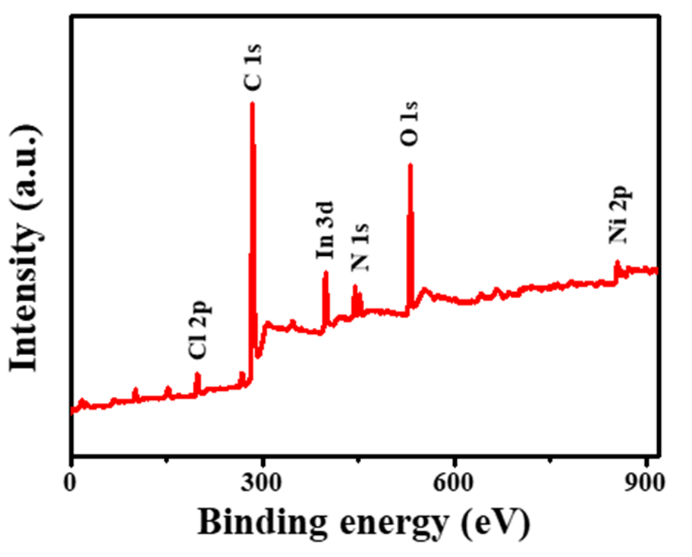
**Supporting figures**

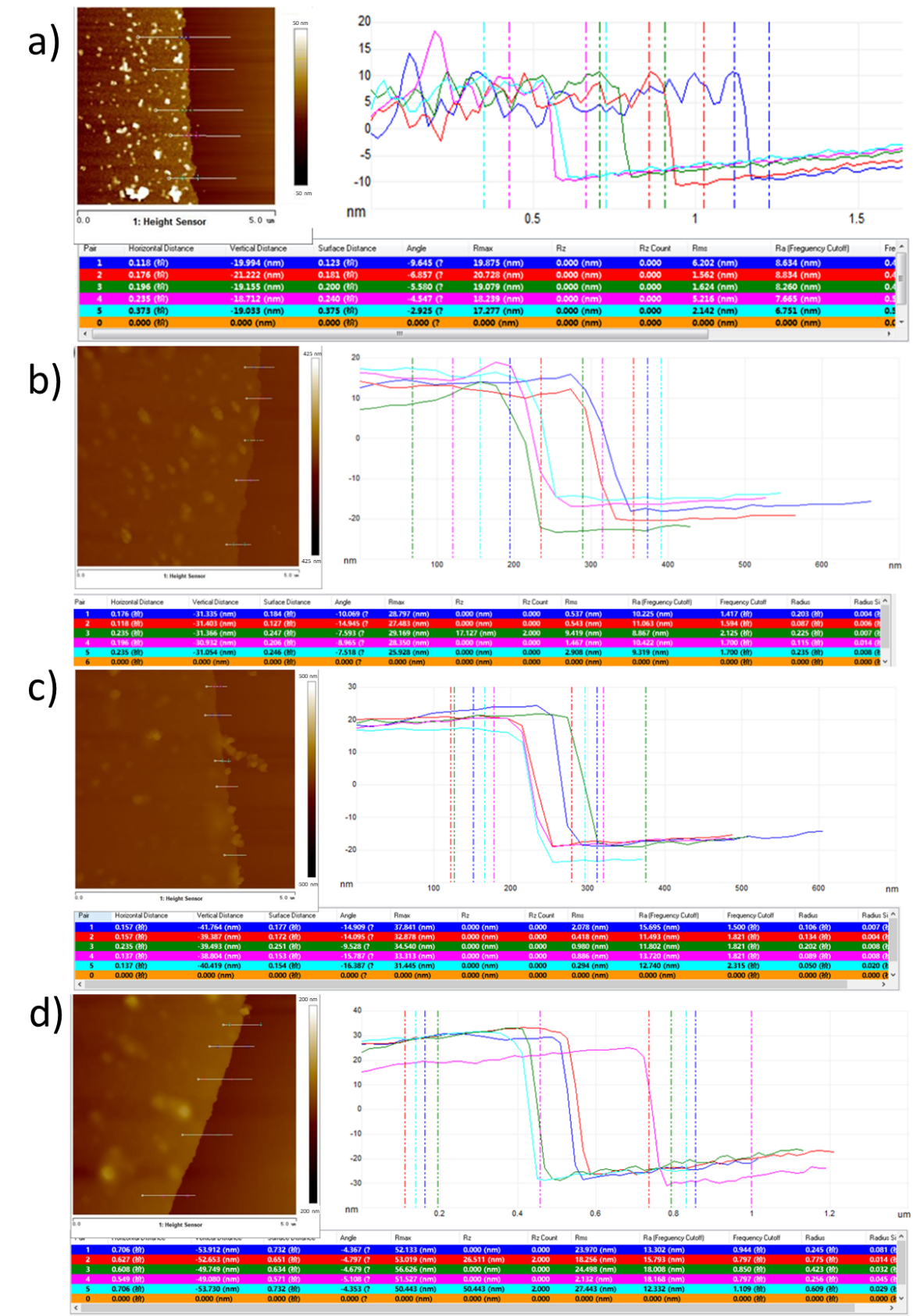
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**Figure S1.** Illustration of the assembly process for the Ni3(HITP)2 film. a) The cleaned ITO substrate; b) The precursor solution for synthesis of Ni3(HITP)2 film; c) After the Ni3(HITP)2 film is formed, lift-down ITO along of the beaker; d) Lift-on the membrane by slowly raising the ITO and e) ITO substrate coated with complete Ni3(HITP)2 film.

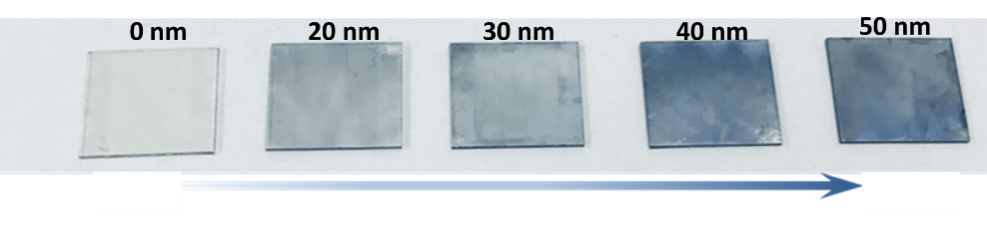
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**Figure S2.** EDX mapping of theNi3(HITP)2 membranes.

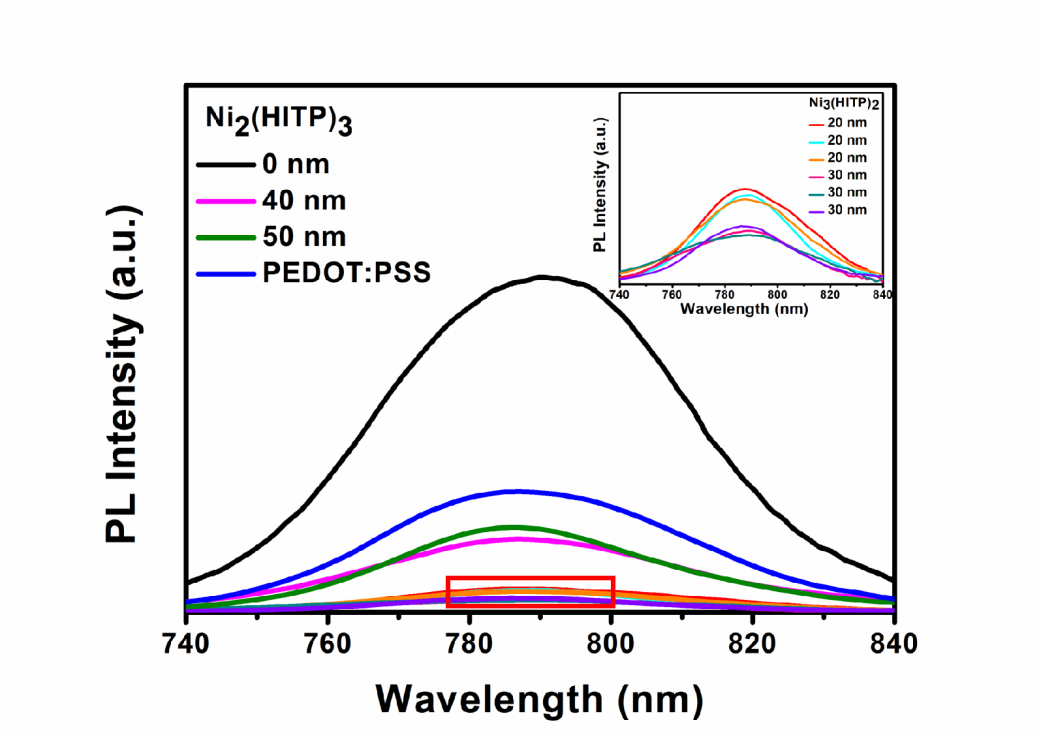
**Figure S3.** XPS spectra of theNi3(HITP)2 membranes.



**Figure S4.** AFM images of different thickness of Ni3(HITP)2 membranes.

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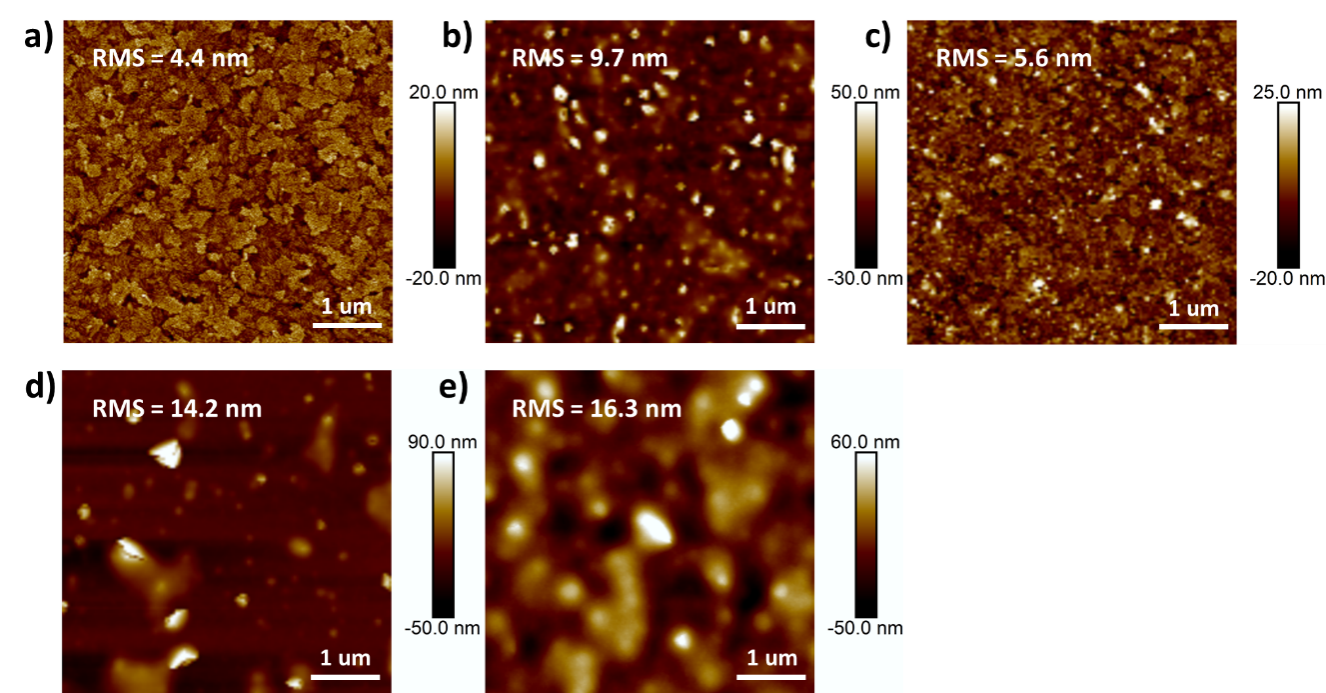
**Figure S5.** Photographs of the bare ITO glass and with different thickness of Ni3(HITP)2 membranes.

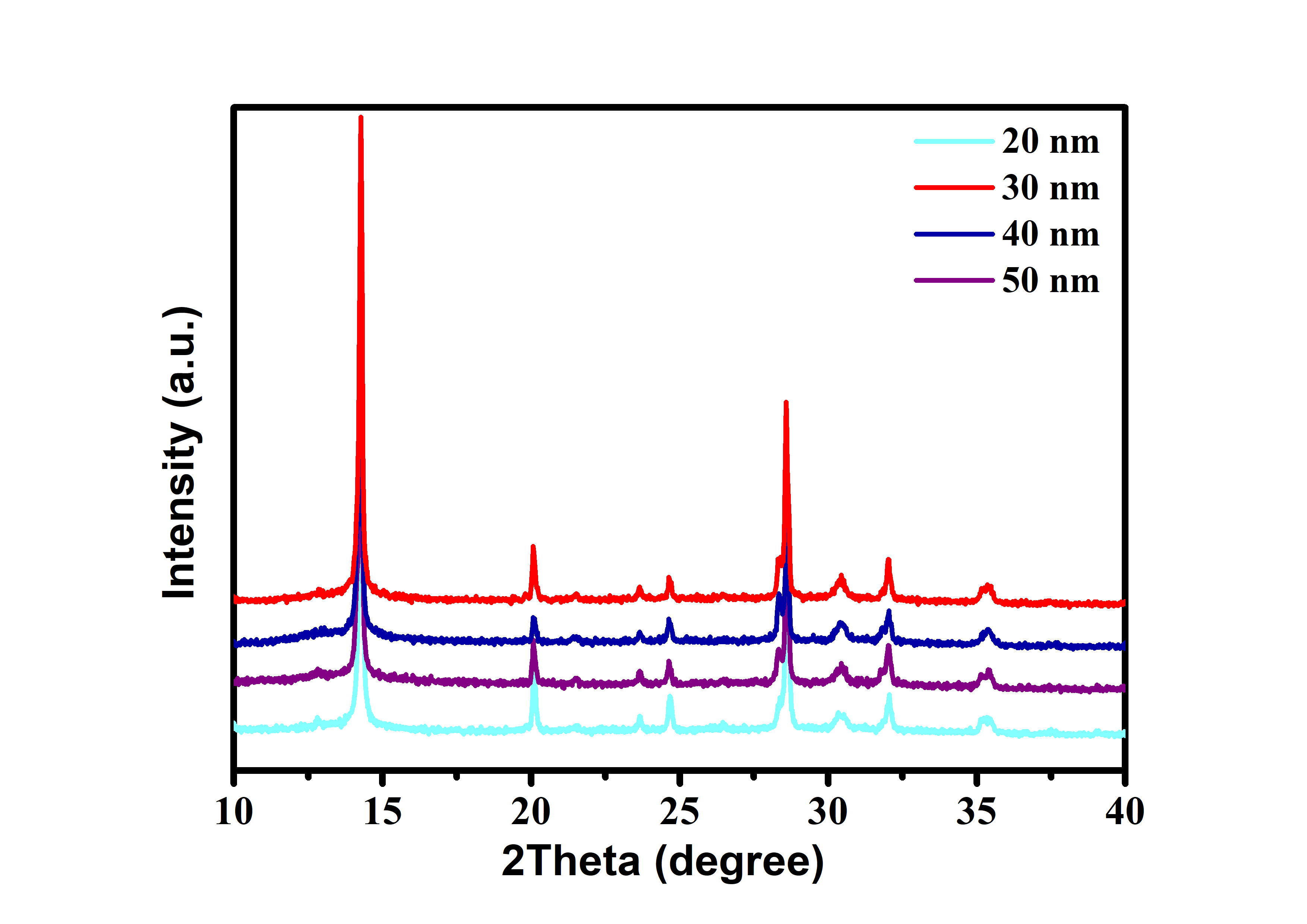


**Figure S6.** Steady-state PL spectra of perovskite/ITO based on 20nm, 30nm, 40nm, and 50nm Ni3(HITP)2 film and PEDOT/PSS; Inset: the enlarged steady-state PL spectra of perovskite on the 20nm and 30nm Ni3(HITP)2 film repeated by three times, respectively.

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**Figure S7.** Time-resolved PL spectra of perovskite on the ITO substrate with different thicknesses of Ni3(HITP)2 film and PEDOT/PSS.

**Figure S8.** AFM images of different thickness of Ni3(HITP)2 membranes. a) Bare ITO glass, b)-e) Corresponding to 20 nm, 30 nm, 40nm and 50 nm Ni3(HITP)2 membranes, respectively.



**Figure S9.** XRD pattern of perovskite films deposition on different thickness of Ni3(HITP)2 membranes.

**Table S1.** The average decay lifetimes of the perovskite/ITO with different hole transport layers.

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
|  | Without | PEDOT/PSS | Ni3(HITP)2 (20 nm) | Ni3(HITP)2 (30 nm) | Ni3(HITP)2 (40 nm) | Ni3(HITP)2 (50 nm) |
| Average τ(ns) | 18.14 | 6.17 | 1.89 | 1.18 | 2.82 | 3.02 |