## Supplementary Information

## Nondestructive Passivation of TiO<sub>2</sub> Electronic Transport Layer in Perovskite Solar Cells by PEIE-2D MOF Interfacial Modified Layer

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Figure S1. The molecular structure of polyethylenimine ethoxylated (PEIE).



Figure S2. Molecular structure diagram of the Cd<sub>3</sub>(C<sub>6</sub>H<sub>2</sub>TeO<sub>4</sub>)<sub>3</sub>·4DMF twodimensional metal organic framework.<sup>1</sup>



Figure S3. The contact angle and surface energy data of  $TiO_2$ ,  $PEIE/TiO_2$  and PEIE-2D MOF/TiO<sub>2</sub> film.



Figure S4. Dark saturation current of PSCs with/without modified.



Figure S5. Reverse (roundness) and Forward (rhombus) scanned J-V curves of the PSCs with and without modification layer. Inset: Device efficiency calculated from the curves.



Figure S6. AMF topography of  $TiO_2$  ETL modified by PEIE-2D MOF of different thickness. The weight concentration of the precursors is 0%, 0.05%, 0.1%, 0.2%, 0.4%, 0.6. The blue curves on the topography represent the roughness of the area corresponding to the line. The average roughness calculated from the curves are shown in the figure.



Figure S7. SEM images of perovskite film surface morphology. The perovskite films were deposited on  $TiO_2$  ETL with different PEIE-2D MOF modified layer. The inserted chart is the corresponding I-V curve and performance parameters of PSCs.



Figure S8. The decay curves of detailed performance parameters of perovskite solar cell.

## The detailed synthetic procedure of Cd<sub>3</sub>(C<sub>6</sub>H<sub>2</sub>TeO<sub>4</sub>)<sub>3</sub>·4DMF 2D MOF:

A mixture of tellerophene-2,5-dicarboxylic acid (120.0 mg, 0.45 mmol) and  $Cd(NO_3)_2.4H_2O$  (140.0mg, 0.90mmol) in a mixed solvent of DMF/ethyl alcohol (12 mL/1.5 mL) was placed in a sealed glass tube, which was heated at 110 °C for 24 h. After cooling slowly to room temperature, the crystalline solid was filtered, repeatedly washed with DMF (3 mL×3) and dried at room temperature to afford yellowish crystals. Yield: 0.60g. Calcd. for  $[Cd_3(C_6H_2TeO_4)_3](C_3H_7NO)_4$ :C, 25.25; H, 2.38; N, 3.92%. Found: C, 24.94; H, 2.46; N, 3.85%. An aqueous solution of PEIE (37 wt%) was diluted with 2-methoxyethanol to a weight concentration of 0.4%. MOF bulk (5mg) was added into PEIE aqueous solution (1mL), and the mixture was sonicated for 5 h in ice-water bath.