Supplementary Information

Exploration of Structural Transition Phenomenon in Flexible Metal-Organic Framework Formed on Polymer Substrate

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1. Formation of different types of MOFs with structural transition

Figure S1. SEM images of samples prepared using the ion-doped doped substrate (scale bars: 1 μ m); XRD patterns of as-prepared sample before and after heat treatment for solvent desorption and after re-adsorption of guest molecules.





Figure S2. Optical images of water drop on samples prepared via conventional method and present two-step and one-pot approaches.

3. Influence of polyimide on the structural transition



Figure S3. XRD patterns of samples prepared via conventional method in the presence of different amount (a: 0 wt%, b: 1 wt%, c: 3 wt%, d: 5 wt%) of poly(amic acid) before (A) and after heat treatment for solvent desorption (B) and after re-adsorption of the guest molecules (C).



Figure S4. TEM images of samples prepared via conventional method in the presence of different amount (a: 0 wt%, b: 1 wt%, c: 3 wt%, d: 5 wt%) of poly(amic acid).



Figure S5. FT-IR spectra of samples prepared via conventional method in the presence of different amount (a: 0 wt%, b: 1 wt%, c: 3 wt%, d: 5 wt%) of poly(amic acid).

4. Heat and water resistances of obtained crystals prepared by two-step approach



Figure S6. SEM image and XRD patterns of samples prepared by using only layer ligand (scale bar: $1 \mu m$).



Figure S7. (a) TG analysis of samples prepared via present two-step approach. (b) XRD patterns and (c) FT-IR spectra of samples prepared via present two-step approach during water resistance test.

5. Effect of KOH treatment time and temperature of polyimide films on the number of adsorbed metal ions and the thickness of hydrolyzed layer



Figure S8. Effect of KOH treatment condition of polyimide film on the amount of potassium ions and cross-sectional SEM and EDX mapping images of polyimide at various hydrolysis temperature. Scale bars are 1 μ m.

6. Reproducibility of the structural transition



Figure S9. XRD patterns of reversible structural transition of samples prepared via twostep approach using polyimide films hydrolyzed at 20 °C.