

Supporting Information for

Correlation of Mesh-Size of Metal–Carboxylate Layer with Degree of Interpenetration in Pillared-Layer Frameworks

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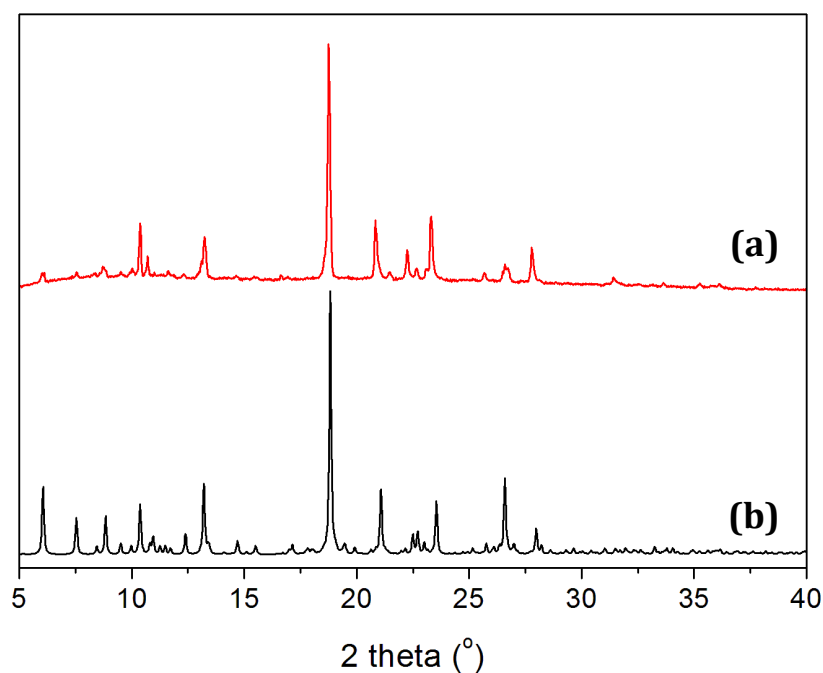


Figure S1. Powder X-Ray diffraction (PXRD) patterns of **1**: (a) a freshly grounded sample at room temperature; (b) simulated from the single-crystal data.

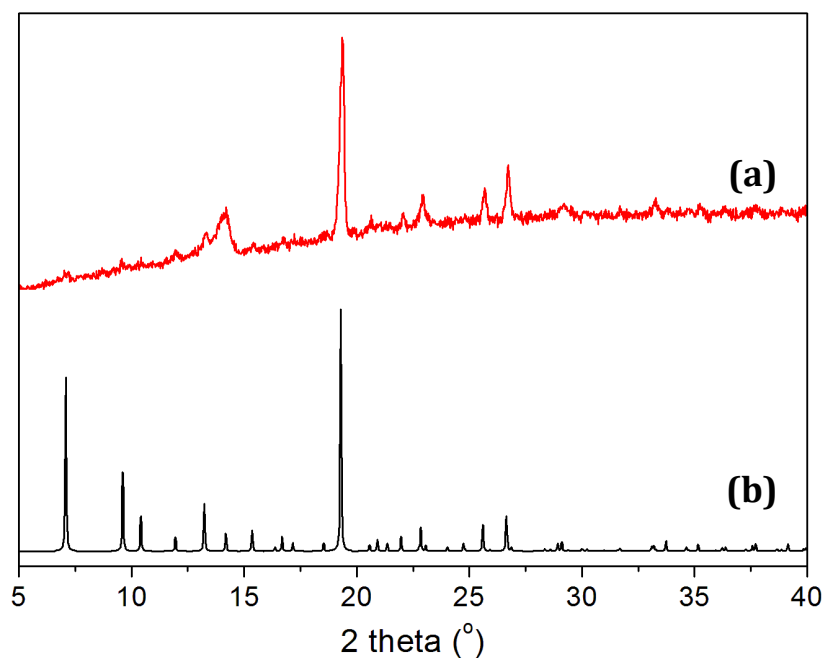


Figure S2. Powder X-ray diffraction (PXRD) patterns of **2**: (a) a freshly grounded sample at room temperature; (b) simulated from the single-crystal data. For **2**, it was a challenge to obtain a well-resolved PXRD pattern that is well-matched with the simulated PXRD patterns since the lattice solvent molecules might have escaped the crystal lattice on loading the sample for PXRD measurement.

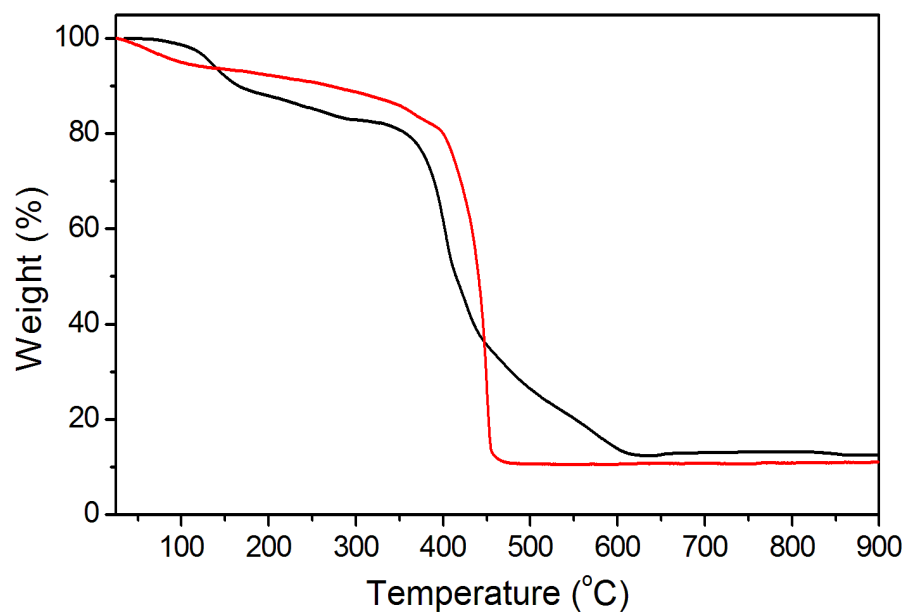


Figure S3. Thermogravimetric (TG) curves of compounds **1** (black line) and **2** (red line).

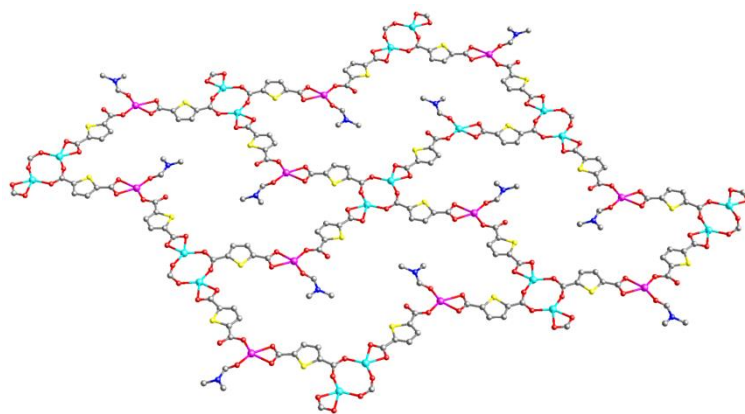


Figure S4. View of the 2D Co-thdc layer showing a rhomboid grid of 4⁴-**sql** topology in **1**.

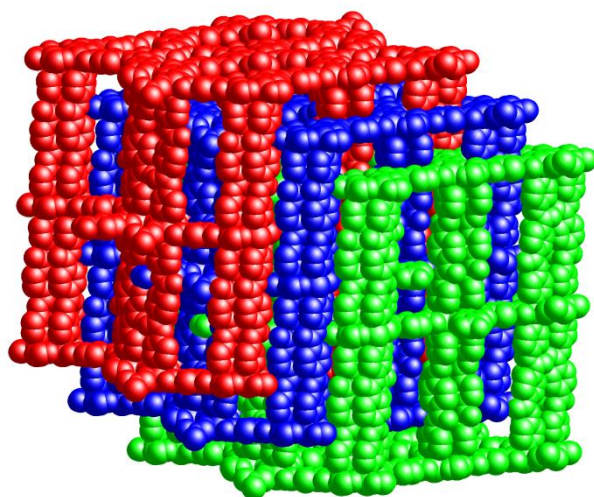


Figure S5. Space-filling representation of the 3-fold interpenetration of **pcu** net.

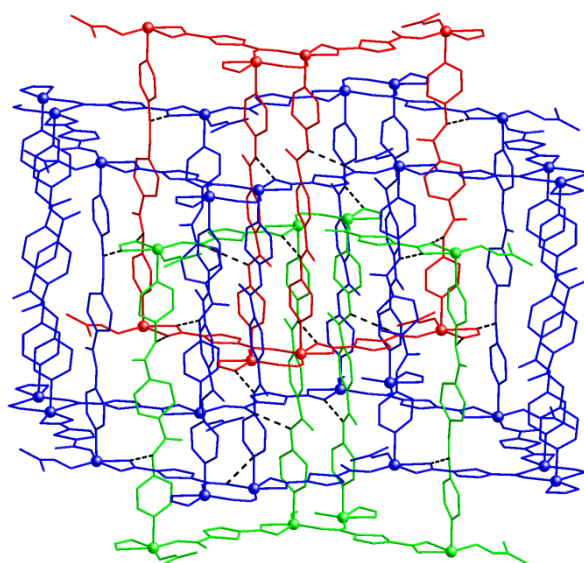


Figure S6. Side-view of the net-to-net N-H...O hydrogen-bonding interactions in **1**.

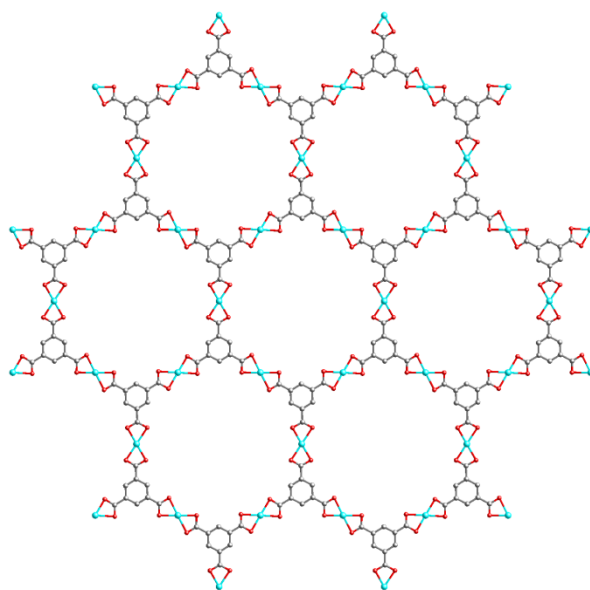


Figure S7. View of the 2D Co–btc layer showing a honeycomb-like 6³-topology in **2**.

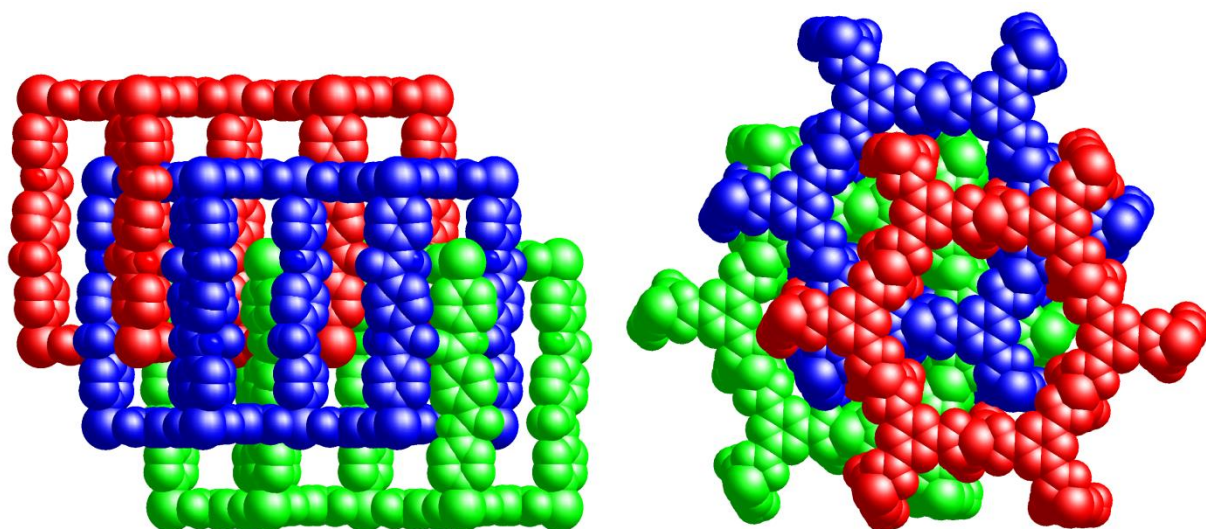


Figure S8. Space-filling representation of the 3-fold interpenetration of net in **2**, viewed along the crystallographic *b* (left) and *c* (right) axes.

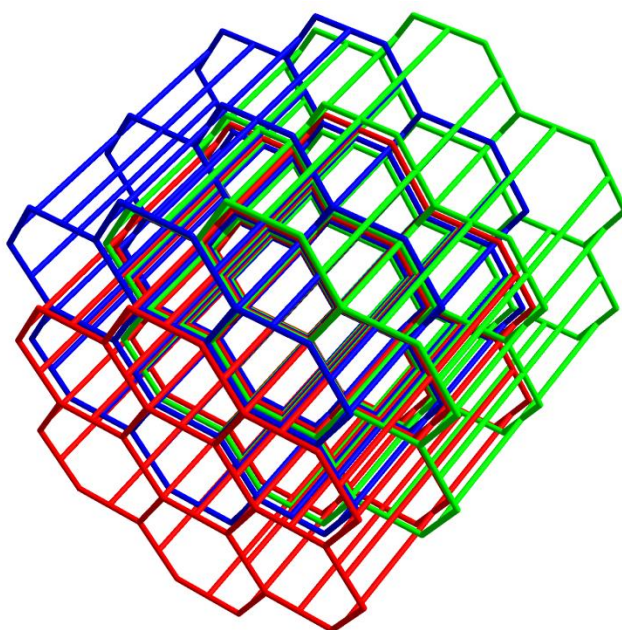


Figure S9. View of the 3-fold interpenetration of net in **2** along the crystallographic $[2,1,1]$ direction, showing that they are related by a single translation vector.

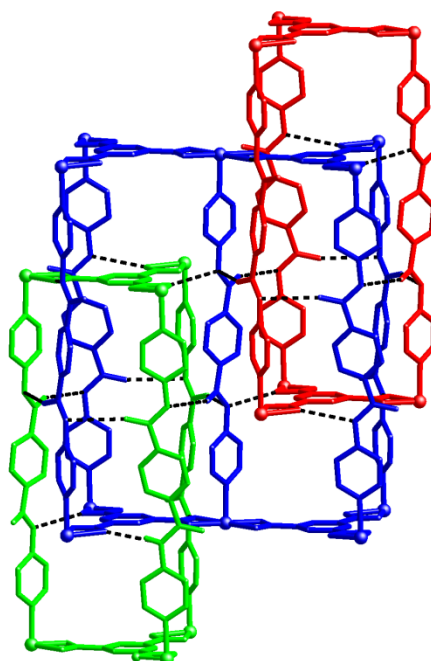


Figure S10. Side-view of the net-to-net N-H...O hydrogen-bonding interactions in **2**.