

## Supporting Information

### A Solid Solution Approach to 2D Coordination Polymers for CH<sub>4</sub>/CO<sub>2</sub> and CH<sub>4</sub>/C<sub>2</sub>H<sub>6</sub> Gas Separation: Equilibrium and Kinetic Studies

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#### Experimental

[Zn(5NO<sub>2</sub>-ip)(bpy)]<sub>n</sub> (CID-5, 5NO<sub>2</sub>-ip = 5-nitroisophthalate), [Zn(5MeO-ip)(bpy)]<sub>n</sub> (CID-6, 5MeO-ip = 5-methoxyisophthalate), and their ligand-base solid solution compounds [Zn(5NO<sub>2</sub>-ip)<sub>1-x</sub>(5MeO-ip)<sub>x</sub>(bpy)]<sub>n</sub> (CID-5/6,  $x = 0.1, 0.2, 0.4$ ) were synthesized according to reported procedures.<sup>1</sup> Characterization of these compounds was performed by powder X-ray diffraction data collected on a Rigaku RINT-2200HF (Ultima) diffractometer with CuK $\alpha$  radiation and <sup>1</sup>H NMR measured with a JEOL JNM-A 500 FT NMR system. Equilibrium gas adsorption and desorption isotherms were measured by a BEL HP instrument. All measurements were executed at 273 K, at a pressure range of 0.0~1.0 MPa and an equilibrium time for each pressure point was set for 500 seconds.

Breakthrough curve measurements were performed using a hand-made gas flowing system. Sample cell was filled with sample powders and temperatures of the cell were controlled by a refrigerant circulating system. For CH<sub>4</sub>/CO<sub>2</sub> mixture gas, the gas fraction was CH<sub>4</sub>:CO<sub>2</sub> = 60:40 (*vol*) and the measurements were executed at 0.80 MPa of total pressure at 273 K with a space velocity of 6 min<sup>-1</sup>. The relative pressure of CO<sub>2</sub> was 0.32 MPa. For CH<sub>4</sub>/C<sub>2</sub>H<sub>6</sub>, the gas fraction was CH<sub>4</sub>:C<sub>2</sub>H<sub>6</sub> = 90:10 (*vol*) and the measurements were executed at 0.80 MPa of total pressure at 273 K with a space velocity of 6 min<sup>-1</sup>. The relative pressure of C<sub>2</sub>H<sub>6</sub> was 0.08 MPa.

1. T. Fukushima, S. Horike, Y. Inubushi, K. Nakagawa, Y. Kubota, M. Takata and S. Kitagawa, *Angew. Chem. Int. Ed.*, 2010, **49**, 4820-4824.