Supplementary information for

Increased dimensionalities of zinc-diphenic acid metal-organic framework compounds by simultaneous or subsequent addition of neutral bridging ligands

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Experimental details for the interconversion of compound **1** into compounds **2** or **3**.

Figure S1. Powder diffraction patterns for reaction of compound 1 with dabco.

Figure S2. Powder diffraction patterns for reaction of compound 1 with 4,4'-bpy.

Figure S3. Proposed mechanism for the changes of lattice parameters with temperature in compound **3**.

Figure S4. Observed powder pattern for **3** at 357ºC and hypothetical patterns for the interpenetrated and non-interpenetrated structures.

Experimental details for the interconversion reactions:

1. Compound **1** (0.110 g, 0.34 mmol), triethylenediamine (0.038 g, 0.34 mmol), 15 mL of ethanol and 3 mL of water were put into a Teflon-lined autoclave and reacted for two days at 120°C. Filtration yielded an opaque substance with the original needle-shaped morphology of compound 1 that was identified as a mixture **1** and **2**.
2. Compound **1** (0.110 g, 0.34 mmol), triethylenediamine (0.038 g, 0.34 mmol), 15 mL of ethanol and 3 mL of water were put into a Teflon-lined autoclave and reacted for seven days at 120°C. Filtration yielded an opaque substance with the original needle-shaped morphology of compound 1 that was identified as a mixture **1** and **2**.
3. Compound **1** (0.045 g, 0.14 mmol), triethylenediamine (0.016 g, 0.14 mmol), and 15 mL of ethanol were put into a Teflon-lined autoclave and reacted for six days at 150°C. Filtration yielded a slightly grey-tinged substance which was identified as X-ray pure **2**.
4. Compound **1** (0.110 g, 0.34 mmol), 4,4'-bipyridyl (0.053 g, 0.34 mmol), 15 mL of ethanol and 3 mL of water were put into a Teflon-lined autoclave and reacted for two days at 120°C. Filtration yielded a mixture of colourless rectangular crystals of **3** and long colourless needles of **1**.
5. Compound **1** (0.110 g, 0.34 mmol), 4,4'-bipyridyl (0.053 g, 0.34 mmol), 15 mL of ethanol and 3 mL of water were put into a Teflon-lined autoclave and reacted for seven days at 120°C. Filtration yielded a mixture of colourless rectangular crystals of **3** and long colourless needles of **1**.
6. Compound **1** (0.115 g, 0.36 mmol), 4,4'-bipyridyl (0.111 g, 0.71 mmol), 15 mL of ethanol and 3 mL of water were put into a Teflon-lined autoclave and reacted for seven days at 120°C. Filtration yielded colourless rectangular crystals of **3** and very few remaining long colourless needles of **1**.

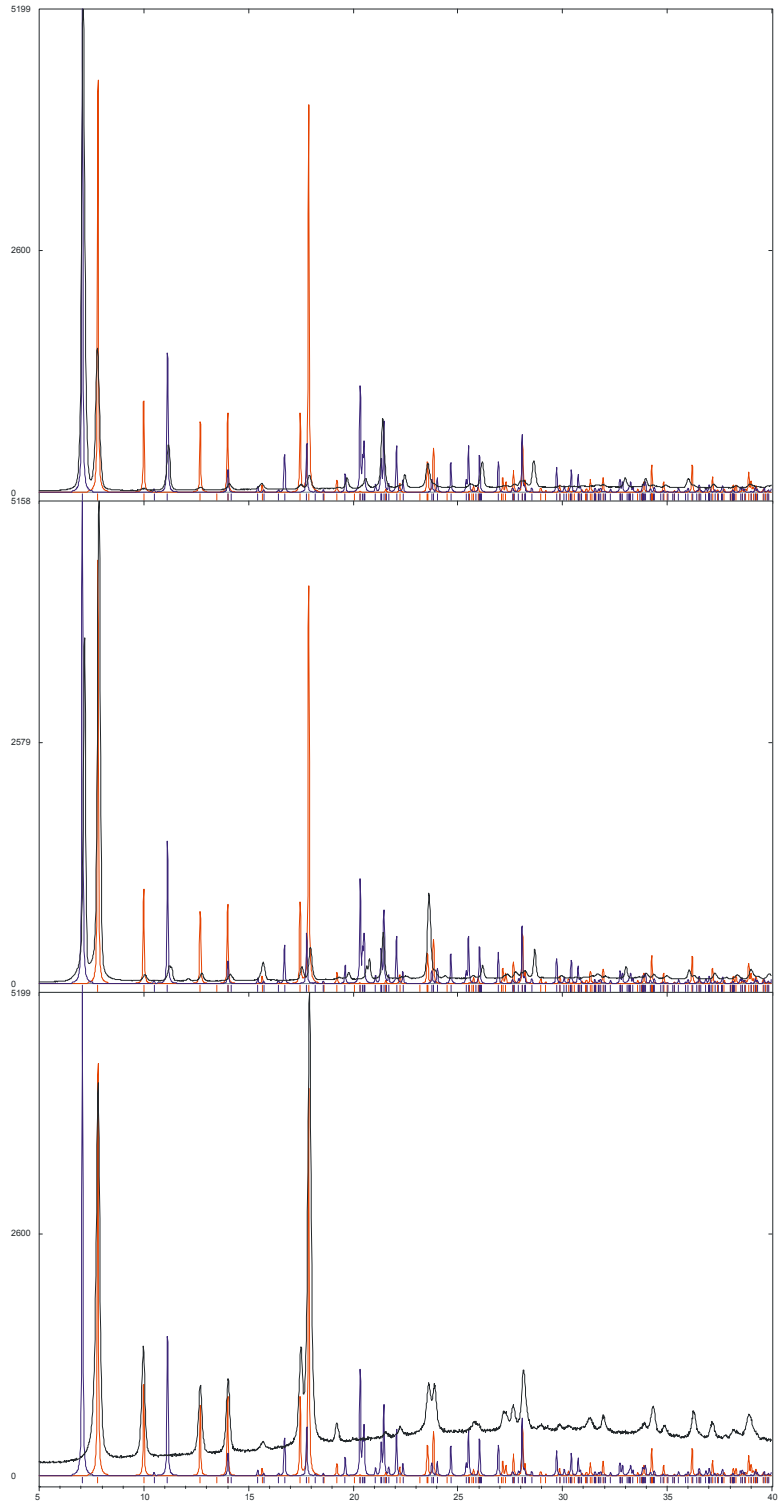

Figure S1. Powder diffraction patterns for reaction of compound **1** with dabco (top: after 3 d and 120°C, middle: after 7 d and 120°C, bottom: after 6 d and 150°C; blue: calculated pattern for compound **1**, red: calculated pattern for compound **2**).

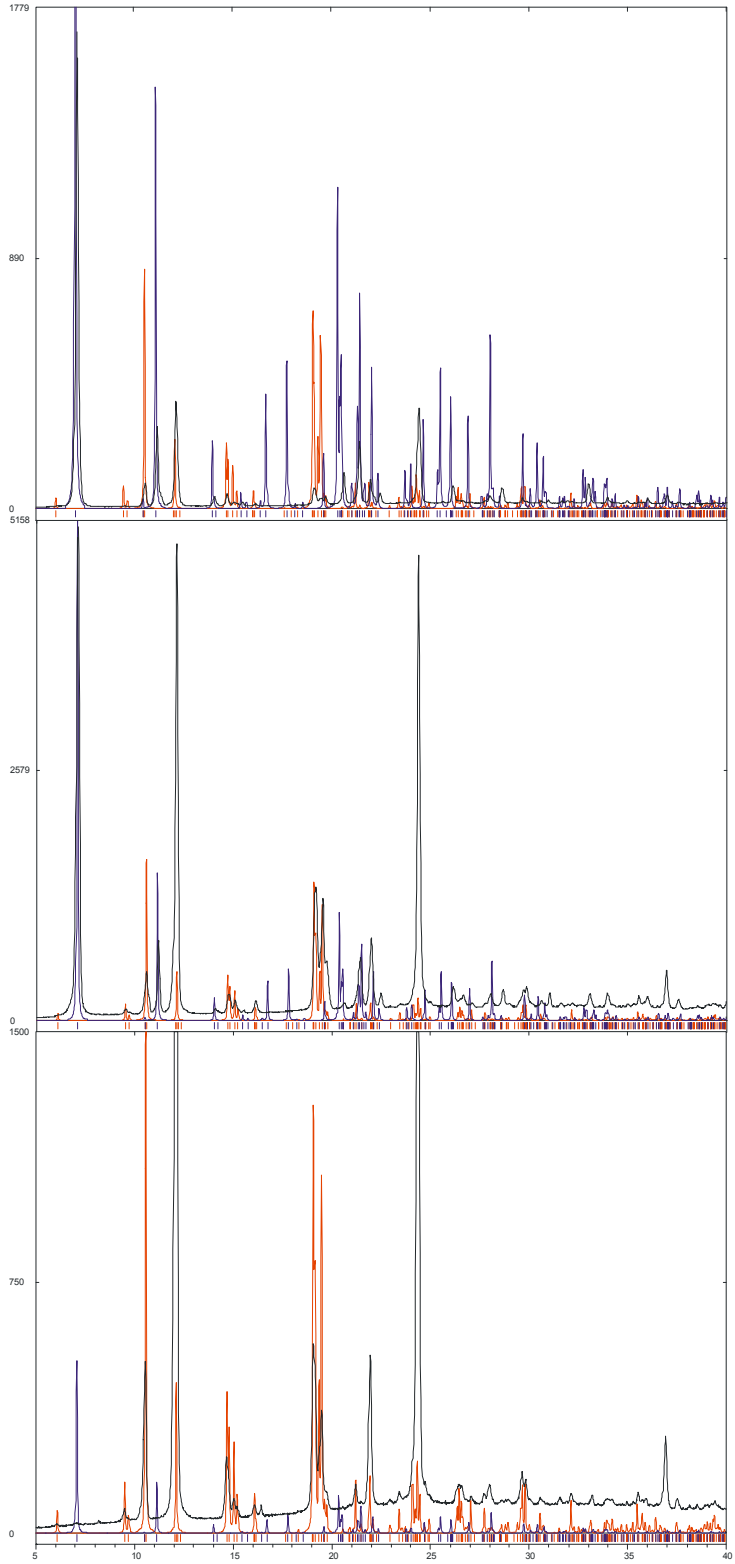

Figure S2. Powder diffraction patterns for reaction of compound **1** with 4,4'-bpy (top: after 2 d and 120°C, middle: after 7 d and 120°C, bottom: after 7 d and 120°C with a one-molar excess of 4,4-bpy; blue: calculated pattern for compound **1**, red: calculated pattern for compound **3**; differences between calculated and observed intensities are due to orientational effects).

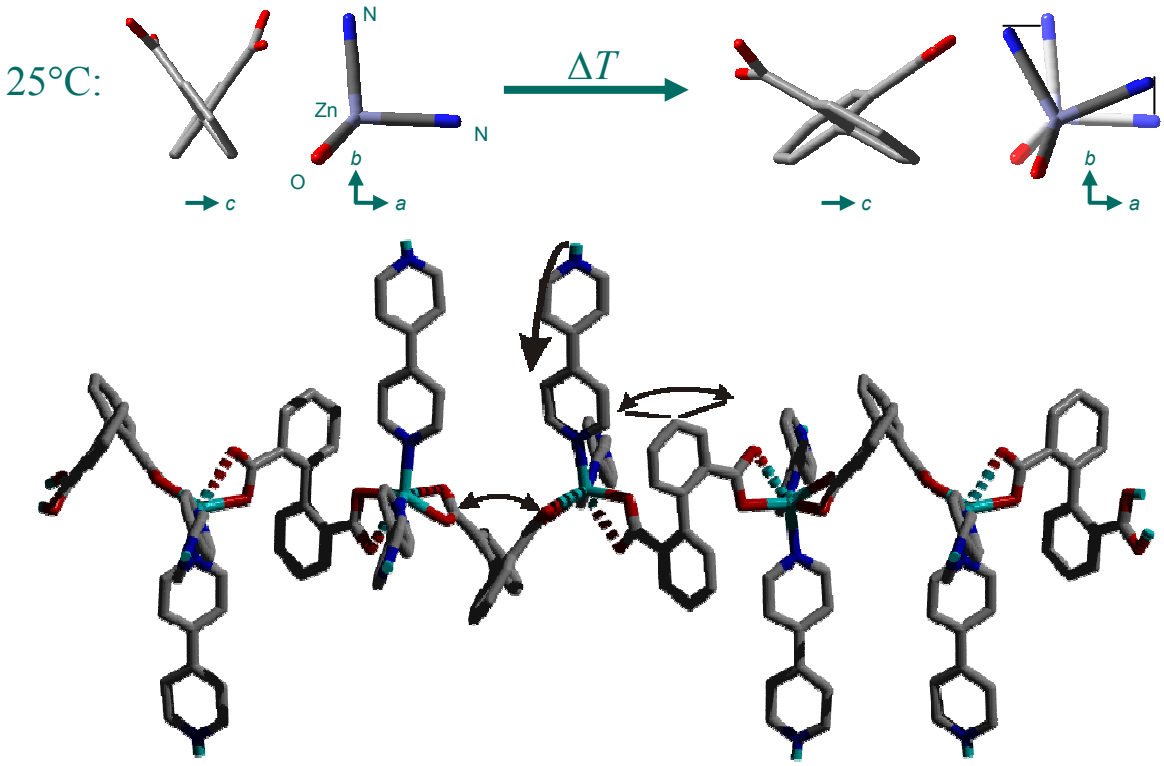

Figure S3. Proposed mechanism for how the widening of the angle between the planes of the aromatic rings in the 2,2'-dicarboxylatobiphenyl leads to a decrease of the *a* and *b* lattice parameters.

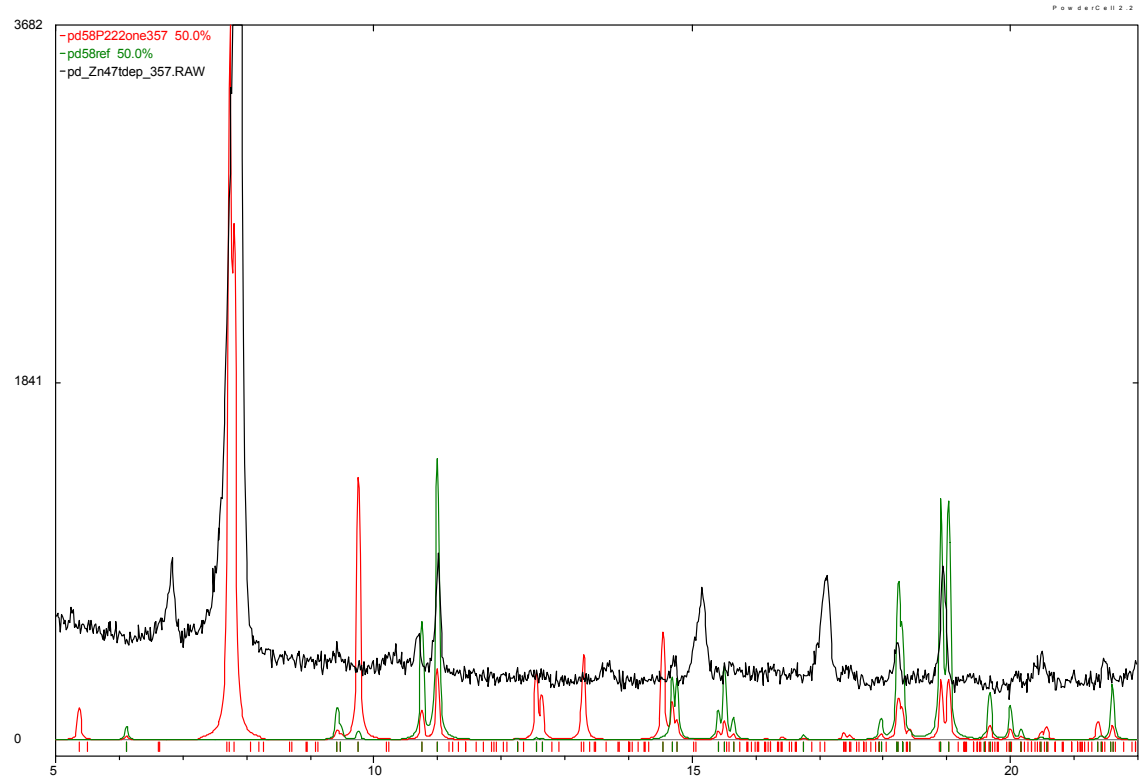

Figure S4. Observed powder patterns for **3** at 357ºC and hypothetical patterns for the interpenetrated and non-interpenetrated structures (green: calculated pattern for the interpenetrated structure with lattice parameters adjusted for the temperature; red: calculated pattern for non-interpenetrated single network structure with the same lattice parameters).