## **Supporting Information**

to the paper

## Catalytic properties of a series of coordination networks: cyanosilylation of aldehydes catalyzed by Zn(II)-4,4' -bpy-carboxylato complexes

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Three porous crystalline coordination polymers,  $\{[Zn_3(4,4'-bpy)_{3.5}(\mu-O_2CH)_4 (H_2O)_2](ClO_4)_2(H_2O)_2\}_n$ (1),  $\{[Zn_3(4,4'-bpy)_3(\mu-O_2CCH_3)_4(H_2O)_2](PF_6)_2(H_2O)_2\}_n$  (2), and  $\{[Zn_3(4,4'-bpy)_4(\mu-O_2CCH_2CH_3)_4]$ (ClO<sub>4</sub>)<sub>2</sub>(4,4'-bpy)<sub>2</sub> (H<sub>2</sub>O)<sub>4</sub> $\}_n$  (3), exhibit voids that contain the counter-ions and guest molecules. The removal and reintroduction of guest water molecules for compounds 2 and 3 have been explored for their dynamic structural transformation and all Zn(II) compounds are active heterogeneous catalysts for the high-yield cyanosilylation of acetaldehyde in dichloromethane.



Figure S1.1 (a) The polycrystalline EPR spectrum of 1 doped with Cu(II) at 70 K (b) The polycrystalline EPR spectrum of 1 doped with Mn(II) at 70 K



Figure S1.2 (a) The polycrystalline EPR spectrum of **3** doped with Cu(II) at 70 K (b) The polycrystalline EPR spectrum of **3** doped with Mn(II) at 70 K



Figure S2.1 XRPD patterns of 1 (a) as-synthesized, (b) doped with Cu(II) and (c) doped with Mn(II)



Figure S2.2 XRPD patterns of 2 (a) as-synthesized, (b) doped with Cu(II) and (c) doped with Mn(II)



Figure S2.3 XRPD patterns of 3 (a) as-synthesized, (b) doped with Cu(II) and (c) doped with Mn(II)



Figure S3.1 IR spectra of 1 (a) doped with Cu(II) and (b) doped with Mn(II)



Figure S3.2 IR spectra of 2 (a) doped with Cu(II) and (b) doped with Mn(II)



Figure S3.3 IR spectra of 3 (a) doped with Cu(II) and (b) doped with Mn(II)