## **Supplementary Information**

Bottom-up approach for the preparation of hybrid nanosheets based on coordination polymers made of metal-diethyloxaloacetates complexes linked by 4,4'-bipyridine

Núria Portolés-Gil, Ricard Parra-Aliana, Ángel Álvarez-Larena, Concepción Domingo, José A. Ayllón, Ana López-Periago.

Cu(1)-O(19)	1.919(2)
Cu(1)-O(12)	1.930(2)
Cu(1)-O(14A)	1.9425(19)
Cu(1)-O(24A)	1.954(2)
Cu(1)-O(1)	2.298(2)
O(19)-Cu(1)-O(12)	88.12(8)
O(19)-Cu(1)-O(14A)	168.65(9)
O(12)-Cu(1)-O(14A)	92.87(8)
O(19)-Cu(1)-O(24A)	92.45(8)
O(12)-Cu(1)-O(24A)	172.72(9)
O(14A)-Cu(1)-O(24A)	85.16(8)
O(19)-Cu(1)-O(1)	91.90(9)
O(12)-Cu(1)-O(1)	97.40(9)
O(14A)-Cu(1)-O(1)	99.17(9)
O(24A)-Cu(1)-O(1)	89.83(9)

Table S1. Selected bond lengths [Å] and angles  $[\circ]$  for  $[Cu(deox)_2(H_2O)]$ .

Co(1)-O(1)	2.026(2)
Co(1)-O(1)#1	2.026(2)
Co(1)-O(4)	2.086(2)
Co(1)-O(4)#1	2.086(2)
Co(1)-N(1)	2.183(3)
Co(1)-N(1)#1	2.183(3)
O(1)-Co(1)-O(1)#1	94.61(13)
O(1)-Co(1)-O(4)	89.74(9)
O(1)#1-Co(1)-O(4)	175.23(9)
O(1)-Co(1)-O(4)#1	175.23(9)
O(1)#1-Co(1)-O(4)#1	89.74(9)
O(4)-Co(1)-O(4)#1	85.97(12)
O(1)-Co(1)-N(1)	90.11(10)
O(1)#1-Co(1)-N(1)	89.58(10)
O(4)-Co(1)-N(1)	88.47(10)
O(4)#1-Co(1)-N(1)	91.87(10)
O(1)-Co(1)-N(1)#1	89.58(10)
O(1)#1-Co(1)-N(1)#1	90.11(10)
O(4)-Co(1)-N(1)#1	91.87(10)
O(4)#1-Co(1)-N(1)#1	88.47(10)
N(1)-Co(1)-N(1)#1	179.53(15

**Table S2.** Selected bond lengths [Å] and angles  $[\circ]$  for  $[Co(deox)_2(tbpy)_2] \cdot H_2O$ .

Symmetry transformations used to generate equivalent atoms: #1 -x,-y+1,z



**Fig. S1**. ATR-FTIR recorded spectra of  $[M(deox)_2(H_2O)_n]$  (M = Cu, n=1; M = Co, Mn, Ni, Zn; n=2). From bottom to top, M= Co, Cu, Mn, Ni and Zn respectively.



Fig. S2. <sup>1</sup>H- NMR of [Zn(deox)<sub>2</sub>(H<sub>2</sub>O)<sub>2</sub>] measured in CDCl<sub>3</sub>.



Fig. S3. <sup>13</sup>C- NMR of [Zn(deox)<sub>2</sub>(H<sub>2</sub>O)<sub>2</sub>] measured in CDCl<sub>3</sub>.



Fig. S4. X-ray diffractograms of [Cu(deox)<sub>2</sub>(H<sub>2</sub>O)]·(up) measured at room temperature. Calculated pattern from resolved crystal structure is also included (down) as a reference.



**Fig. S5**. ATR-FTIR recorded spectra of  $[M(deox)_2(tbpy)_2] \cdot nH_2O$  (M = Cu, n=1; M = Co, Mn, Ni, Zn; n=2). From bottom to top, M= Co, Cu, Mn, Ni and Zn respectively.



Fig. S6. <sup>1</sup>H-NMR of [Zn(deox)<sub>2</sub>(tbpy)<sub>2</sub>] 2H<sub>2</sub>O measured in CDCl<sub>3</sub>.



Fig. S7. <sup>13</sup>C-NMR of [Zn(deox)<sub>2</sub>(tbpy)<sub>2</sub>] 2H<sub>2</sub>O measured in CDCl<sub>3</sub>.



**Fig. S8.** X-ray diffractograms of  $[Co(deox)_2(tbpy)_2] \cdot H_2O$  (up) measured at room temperature. Calculated pattern from resolved crystal structure is also included (down) as a reference.



**Fig. S9**. ATR-FTIR recorded spectra of  $[M(deox)_2(bipy)]_{\infty}$ . From bottom to top, M= Co, Cu, Mn, Ni and Zn respectively.



**Fig. S10.** ATR-FTIR spectra (up) and powder XRD pattern (down) of  $[Co(deox)_2(bipy)]_{\infty}$  samples precipitated in conventional solvent (methanol, bottom), in scCO2 with 2% ethanol (middle) and neat scCO<sub>2</sub> from  $[Co(deox)_2(tbpy)_2] H_2O$ .



**Fig. S11.** ATR-FTIR spectra (up) and powder XRD pattern (down) of  $[Cu(deox)_2(bipy)]_{\infty}$  samples precipitated in conventional solvent (methanol, bottom), in scCO2 with 2% ethanol (middle) and neat scCO<sub>2</sub> from  $[Cu(deox)_2(tbpy)_2] \cdot 2H_2O$ .



**Fig. S12.** ATR-FTIR spectra (up) and powder XRD pattern (down) of  $[Mn(deox)_2(bipy)]_{\infty}$  samples precipitated in conventional solvent (ethanol, bottom), in scCO2 with 2% ethanol (middle) and neat scCO<sub>2</sub> from  $[Mn(deox)_2(tbpy)] \cdot 1.5H_2O$ .



**Fig. S13.** ATR-FTIR spectra (up) and powder XRD pattern (down) of  $[Ni(deox)_2(bipy)]_{\infty}$  samples precipitated in conventional solvent (methanol, bottom), in scCO2 with 2% ethanol (middle) and neat scCO<sub>2</sub> from  $[Ni(deox)_2(tbpy)_2]$ .



**Fig. S14.** ATR-FTIR spectra (up) and powder XRD pattern (down) of  $[Zn(deox)_2(bipy)]_{\infty}$  samples precipitated in conventional solvent (methanol, bottom), in scCO2 with 2% ethanol (middle) and neat scCO<sub>2</sub> from  $[Zn(deox)_2(tbpy)_2] 2H_2O$ .