Supporting Information for:

Broadening the scope of high structural dimensionality nanomaterials using pyridine-based curcuminoids

Laura Rodríguez-Cid,[a]¥ Wenjie Qian,[a]¥ Joseline Iribarra-Araya,[b] Álvaro Etcheverry-Berríos,[b] Eulalia Martínez-Olmos,[a] Duane Choquesillo-Lazarte,[c] Eva Carolina Sañudo,[d,e] Olivier Roubeau,[f] Ana María López-Periago,[a] Arántzazu González-Campo,[a] José G. Planas,[a] Mònica Soler,*[b] Concepción Domingo*[a] and Núria Aliaga-Alcalde*[a,g]

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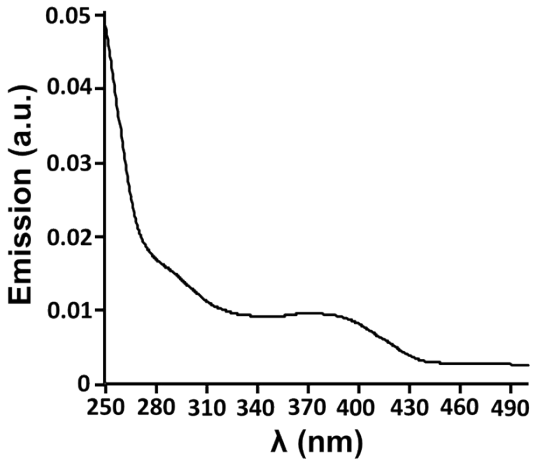


Figure S1. UV-Vis absorption spectrum between 250 and 500 nm of 3pyCCMoid in H_2O.

Table S1. Crystallographic data of the CPs.

Name	**3pyCCMoid**	**1**	**2**	**4**
Formula	$C_{17}H_{14}N_2O_2$	$C_{17}H_{13}N_2O_2Zn_{0.5}$	$C_{34}H_{26}N_6O_{10}Zn_2$	$C_{21}H_{25}N_3O_7Zn$
Formula weight	278.30	309.98	809.35	496.8
Crystal system	monoclinic	monoclinic	monoclinic	orthorhombic
Space group	$P2_1/c$	$C2/c$	$P2_1/n$	$P2_12_12_1$
a (Å)	17.531(8)	14.5539(12)	9.0611(3)	15.000
b(Å)	10.383(5)	20.3868(18)	14.0313(4)	15.360
c (Å)	7.561(4)	9.5213	28.1117(8)	9.980
α (◦)	90	90	90	90
β (◦)	102.080(6)	91.462(3)	95.0900(10)	90
γ (◦)	90	90	90	90
V (Å)^3	1345.8(11)	2824.1	3560.00	2299.39
Z	4	8	4	4
ρ (gcm^{-3})	1.374	1.458	1.510	1.429
Temperature (K)	100(2)	100(2)	293(2)	100(2)
Wavelength (Å)	0.7749	0.82654	0.71073	0.729
μ(mm^{-1})	0.110	1.372	1.412	1.192
F(000)	584	1280	1648	1024
Reflections collected		13947	34882	
Independent reflections	3014	2377	6303	6885
R_{int}	0.0576	0.049	0.0417	0.0365
Parameters	193	195	606	292
Restraints	1	0	183	0
Goodness-of-fit on F^2	1.052	1.185	1.070	1.180
R1 [I>2σ(I)]	0.0448	0.0810	0.0554	0.0295
wR2 [I>2σ(I)]	0.1170	0.1828	0.1415	0.0768
R1 (all data)	0.0611	0.0858	0.0790	0.0296
wR2 (all data)	0.1265	0.1859	0.1586	0.0769

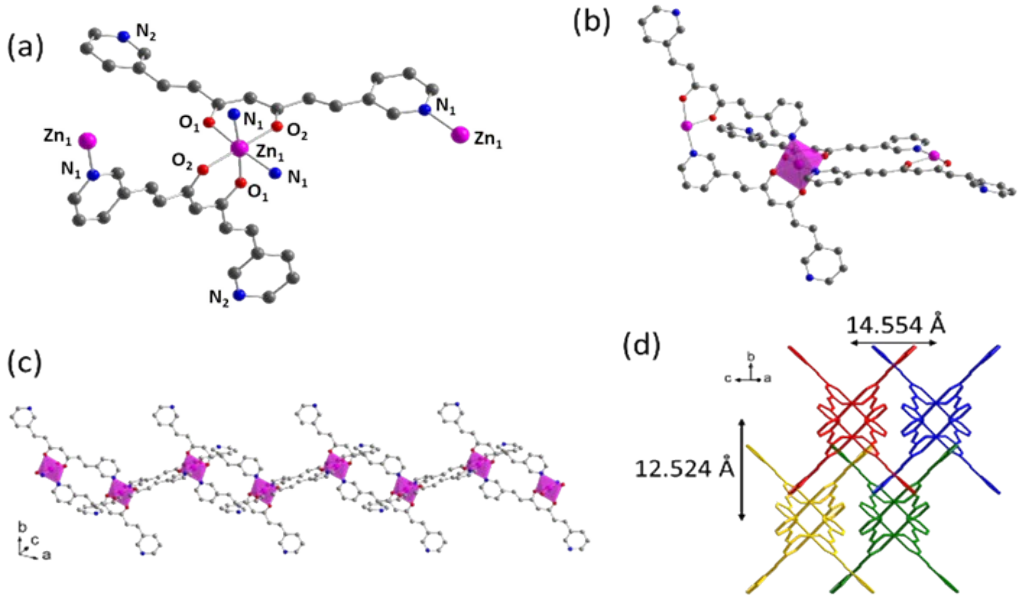


Figure S2. enlarged view of Figure 3. Crystal structure of **1**: (a) Representation with the coordination sphere of a Zn^{II} centre and the coordination modes of 3pyCCMoid molecules; (b) representation of two loops connected through the intermediate Zn(II) centre; (c) extended view of one chain; (d) arrangement of four chains in the crystal. Colour legend: Zn, pink; C, grey; O, red; N, blue; H are omitted for clarity.

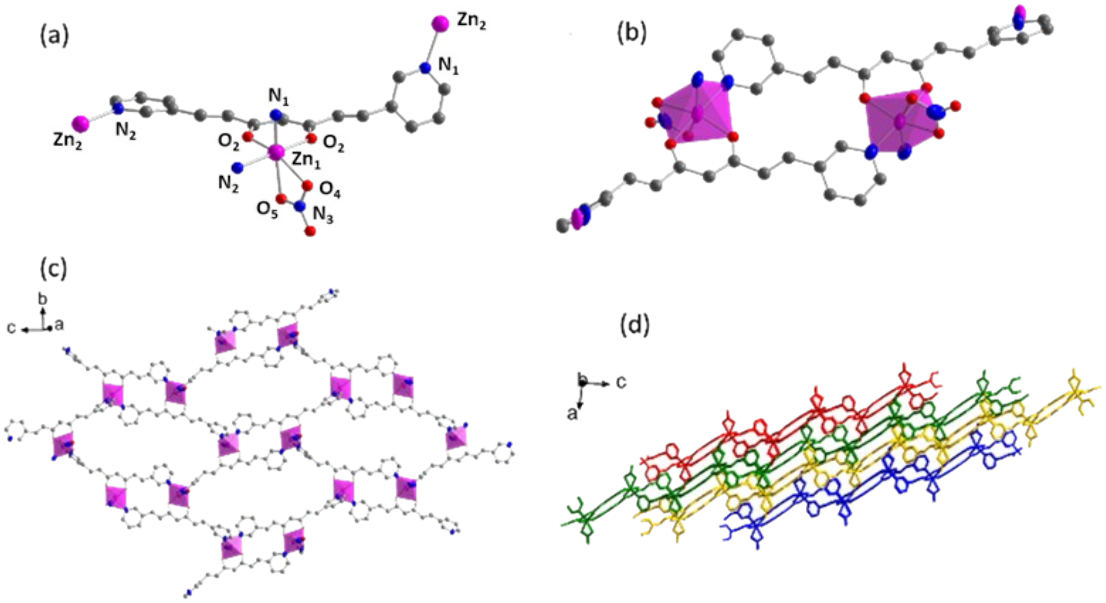


Figure S3. enlarged view of Figure 4. Crystal structure of **2**: (a) Representation with the coordination sphere of a Zn^{II} centre and the coordination mode of 3pyCCMoid; (b) picture of the smallest loop in the layer; (c) 2D Zn-CCMoid layer; (d) framework consisting on packed 2D layers (each layer represented by a different colour). Colour legend: Zn, pink; C, grey; O, red; N, blue; H are omitted for clarity.

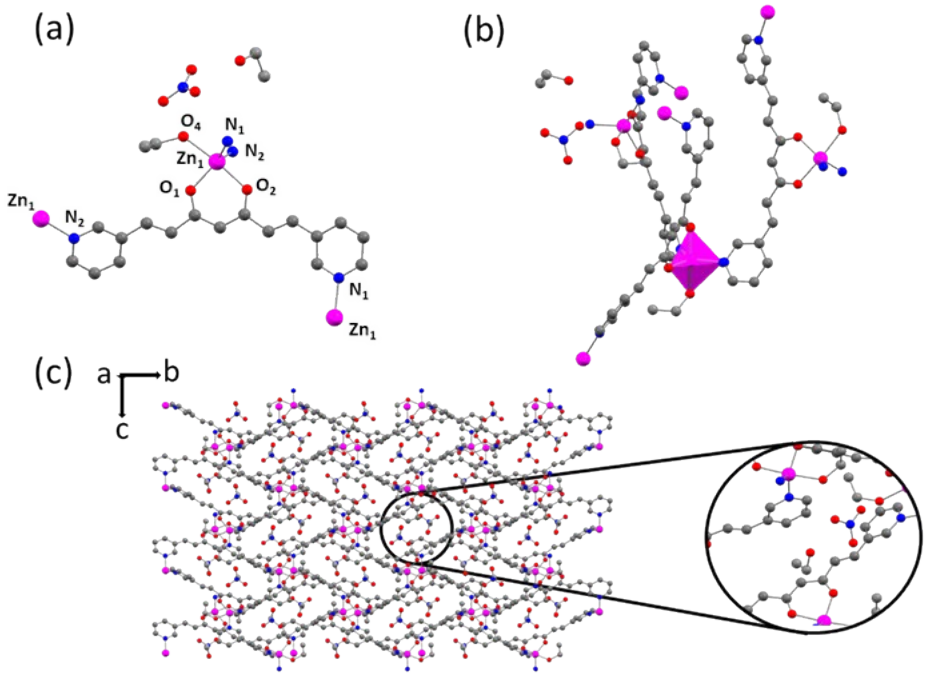
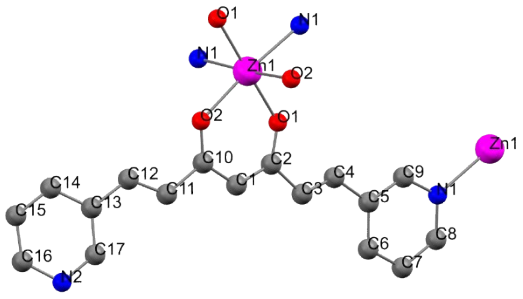


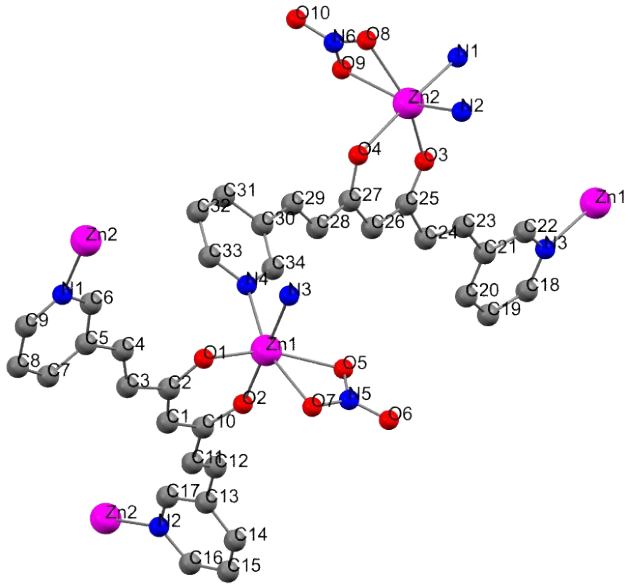
Figure S4. Enlarged view of Figure 5. Crystal structure of **4**: (a) Representation with the coordination sphere of a Zn^{II} centre and the coordination mode of 3pyCCMoid; (b) picture of the connection of two neighbour CCMoids; (c) 3D **4** structure with molecules of solvent and zoom of their disposition in the cavity. (Zn, pink; C, grey; O, red; N, blue; H are omitted for clarity).

Table S2. Repeated unit of CP **1** with label atoms and selected angles and distances.



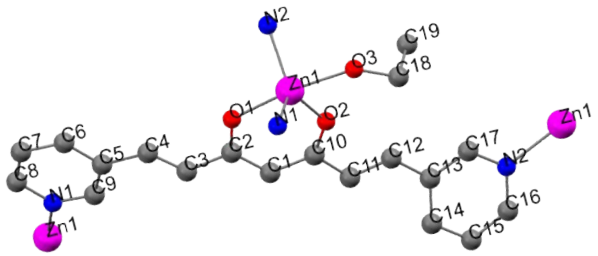
Atoms	Lengths [Å]	Atoms	Angles [°]
Zn1-N1	2.211	O1-Zn1-O2	87.53
Zn1-O1	2.026	O1-Zn1-O2	93.79
Zn1-O2	2.131	O1-Zn1-O1	178.11
C2-O1	1.241	O1-Zn1-N1	87.02
C10-O2	1.256	O1-Zn1-N1	91.68
C1-C2	1.394	O2-Zn1-N1	87.83
C1-C10	1.401	O2-Zn1-O2	91.55
N1-C8	1.321	O2-Zn1-O1	93.79
N1-C9	1.349	N1-Zn1-N1	93.31
		N1-Zn1-O1	87.02
		O1'-Zn1-O2	87.53
		O1-Zn1-N1	91.68
		O2-Zn1-N1	87.83
		Zn1-O1-C2	129.50
		Zn1-O2-C10	124.93
		O1-C2-C1	124.88
		O2-C10-C1	126.04
		C2-C1-C10	126.48
		C1-C5-N1	167.82
		C1-C13-N2	123.28

Table S3. Repeated unit of CP **2** with label atoms and selected angles and distances.



Atoms	Lengths [Å]	Atoms	Angles [°]	Atoms	Angles [°]
Zn1-N4	2.220	O1-Zn1-O2	88.76	O4-Zn2-O9	89.76
Zn1-N3	2.189	O1-Zn1-N3	88.79	O8-Zn2-O9	56.21
Zn1-O1	1.988	O1-Zn1-O7	98.21	O9-Zn2-N1	90.62
Zn1-O2	2.058	O1-Zn1-N4	99.78	O8-Zn2-N1	92.67
Zn1-O5	2.238	O2-Zn1-N3	176.79	O8-Zn2-N2	100.90
Zn1-O7	2.272	O2-Zn1-N4	98.29	O8-N6-O9	116.93
C2-O1	1.271	O2-Zn1-O5	87.89	N1-Zn2-N2	90.68
C10-O2	1.264	O2-Zn1-O7	89.92	Zn2-O3-C25	129.11
C1-C2	1.392	O7-Zn1-O5	57.25	Zn2-O4-C27	128.65
C1-C10	1.388	O7-Zn1-N3	88.39	O3-C25-C26	132.90
N1-C6	1.350	O5-Zn1-N3	93.49	O4-C27-C26	125.4
N1-C9	1.333	O5-Zn1-N4	105.00	C27-C26-C25	119.53
N1-C16	1.317	O5-N5-O7	116.03	C26-C30-N4	131.35
N1-C17	1.326	N3-Zn1-N4	84.15	C26-C21-N3	168.56
Zn2-N1	2.145	Zn1-O2-C10	126.35		
Zn2-N2	2.125	Zn1-O1-C2	128.27		
Zn2-O3	1.985	O1-C2-C1	124.90		
Zn2-O4	2.227	O2-C10-C1	125.63		
Zn2-O8	2.191	C2-C1-C10	125.59		
Zn2-O9	2.311	C1-C13-N2	134.88		
C25-O3	1.282	C1-C5-N1	167.41		
C27-O4	1.291	O3-Zn2-O4	84.27		
C26-C25	1.423	O3-Zn2-N1	91.64		
C26-C27	1.392	O3-Zn2-O9	102.56		
N3-C18	1.337	O3-Zn2-N2	100.25		
N3-C22	1.336	O4-Zn2-N1	175.88		
N4-C33	1.340	O4-Zn2-N2	90.56		
N4-C34	1.361	O4-Zn2-O8	90.95		

Table S4. Repeated unit of CP **4** with label atoms and selected angles and distances.



Atoms	Lengths [Å]	Atoms	Angles [°]
Zn1-N1	2.067	O1-Zn1-O2	88.80
Zn1-N2	2.054	O1-Zn1-N1	89.60
Zn1-O1	2.077	O1-Zn1-N2	93.68
Zn1-O2	1.966	O1-Zn1-O3	172.81
Zn1-O3	2.142	O2-Zn1-N1	117.53
C2-O1	1.271	O2-Zn1-N2	120.89
C10-O2	1.280	O2-Zn1-O3	85.25
C1-C2	1.410	O3-Zn1-N1	89.58
C1-C10	1.402	O3-Zn1-N2	92.84
		N1-Zn1-N2	121.53
		Zn1-O1-C2	125.03
		Zn1-O2-C10	127.48
		O1-C2-C1	125.23
		O2-C10-C1	125.58
		C2-C1-C10	124.32
		C1-C5-N1	124.06
		C1-C13-N2	169.85

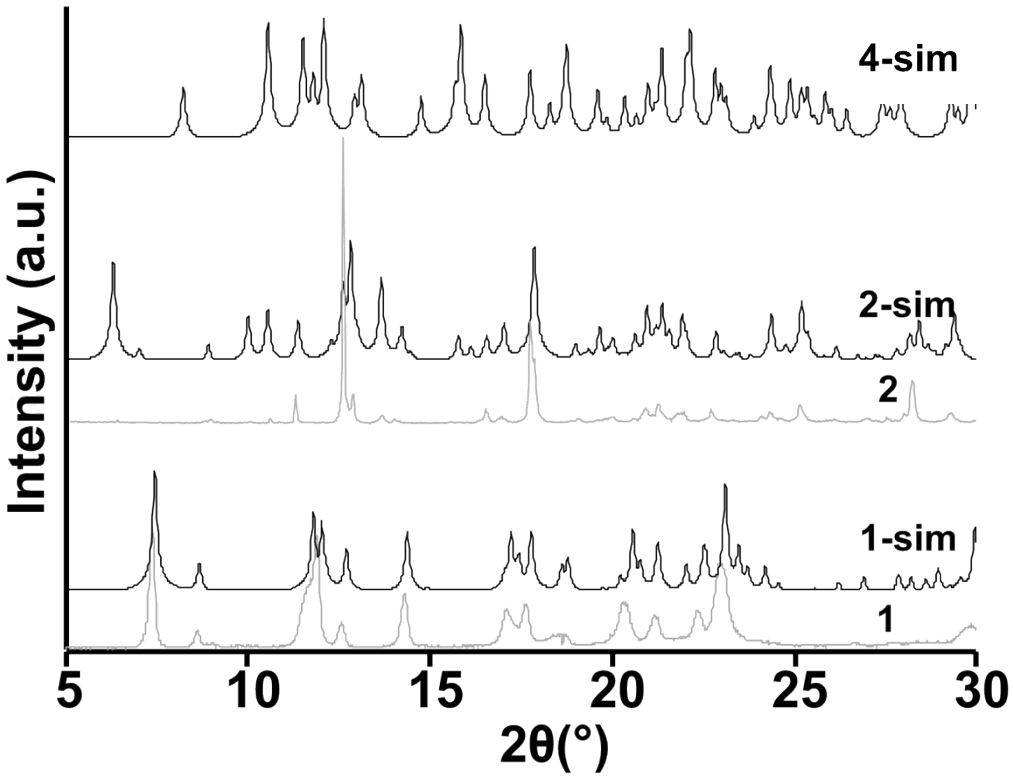


Figure S5. Comparison between the powder XDR of all the CPs (**1**,**2** and **4**), either simulated from the crystal structure with mercury (black) and/or obtained experimentally (grey).

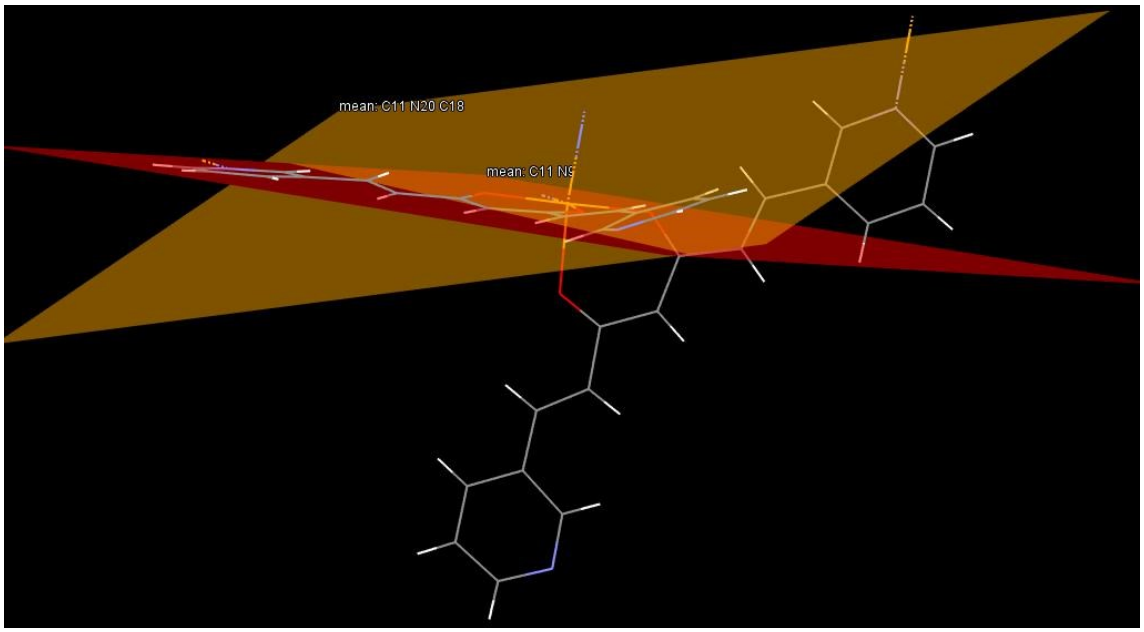
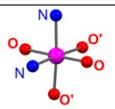
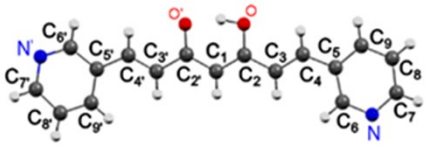
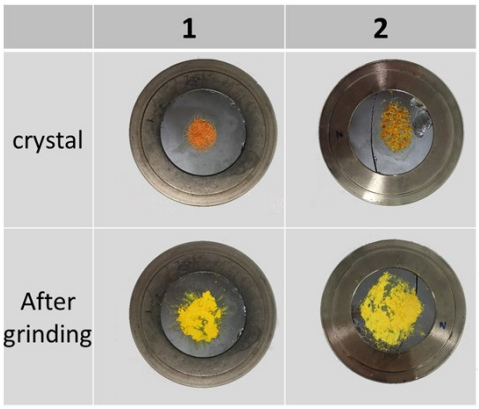


Figure S6. The two planes used in to measure planarity were made by using the *meta*- positions of the aromatic rings and the central carbon of the studied 3pyCCMoid, hence N'-C_8-C_1 and N-C_8-C_1 atoms from Figure 1 in the main manuscript.

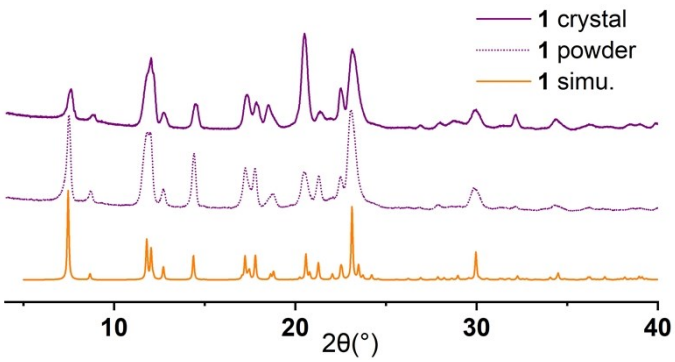
Table S5. Geometry, and coordination environment of the metal in the CP structures.

CPs	Metal coordination	Metal geometry	Metal coordination environment
1	Hexacoordinate	Pseudo-octahedral	
2	Hexacoordinate	Pseudo-octahedral	
4	Pentacoordinate	Pseudo-trigonal bipyramid	
			

(a)



(b)



(c)

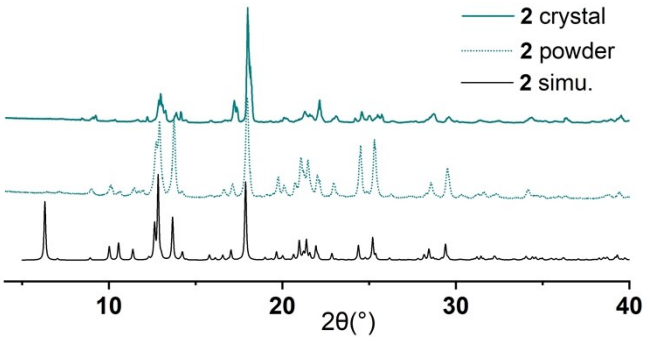


Figure S7. (a) Optical picture of samples **1** and **2** before and after grinding. Powder X-ray diffraction of **1** (a) and **2** (b). In both cases, the patterns are displaying the recorded experimental data from the single-crystal (top), the bulk crystalline sample, after grinding, (middle) and the simulated profile from the single-crystals structure (bottom**).**

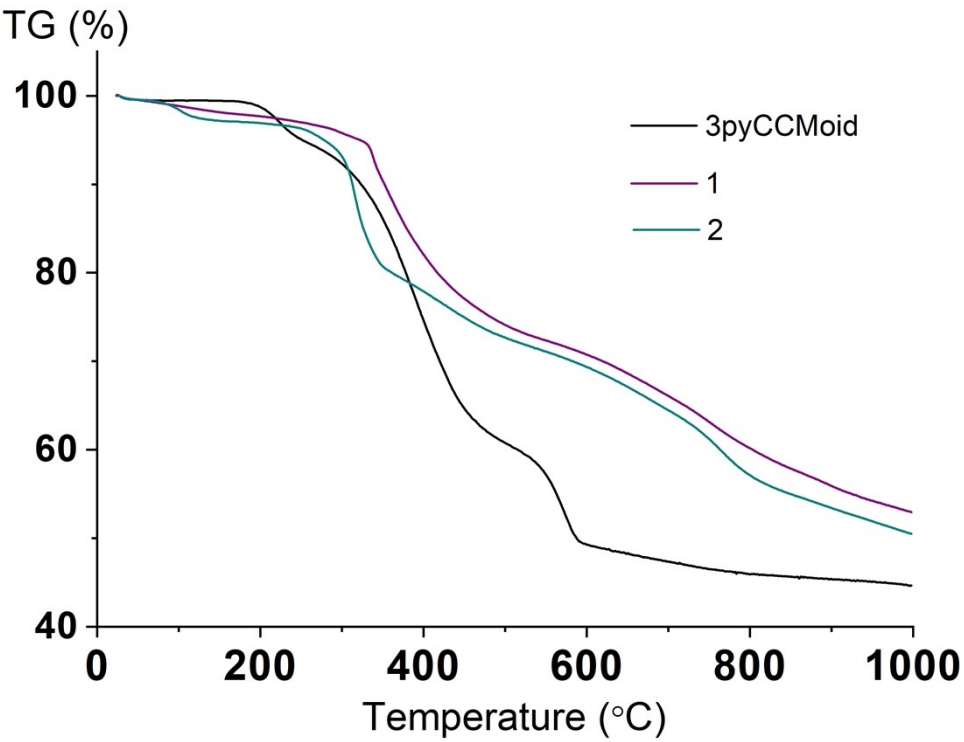
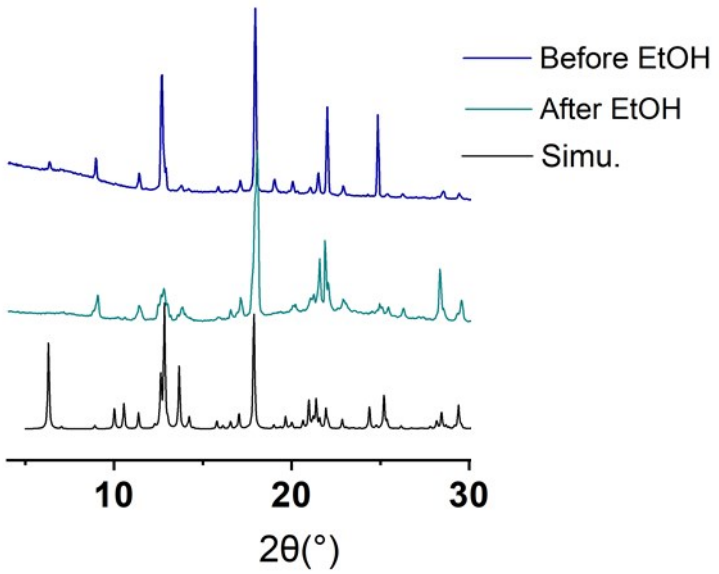


Figure S8. TGA analyses of 3pyCCMoid, and compounds **1** and **2**.

(a)



(b)

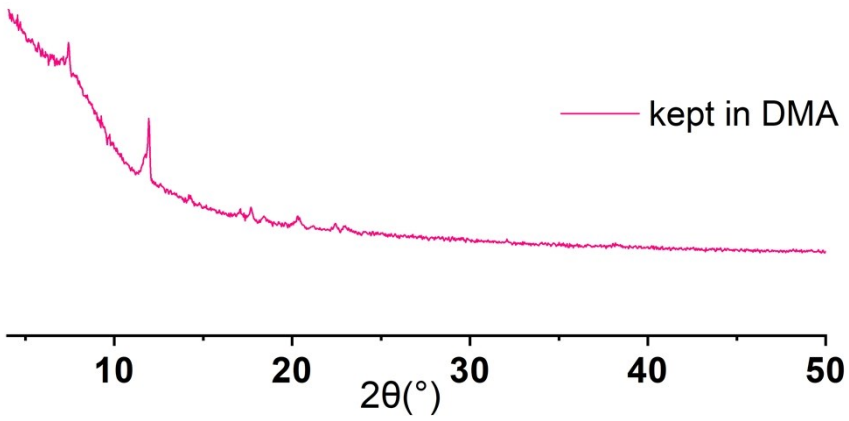


Figure S9. Powder XRD patterns of **2** (a) before and after washing with EtOH, together with the simulated profile from single-crystal structure, and (b) kept in DMA overnight.

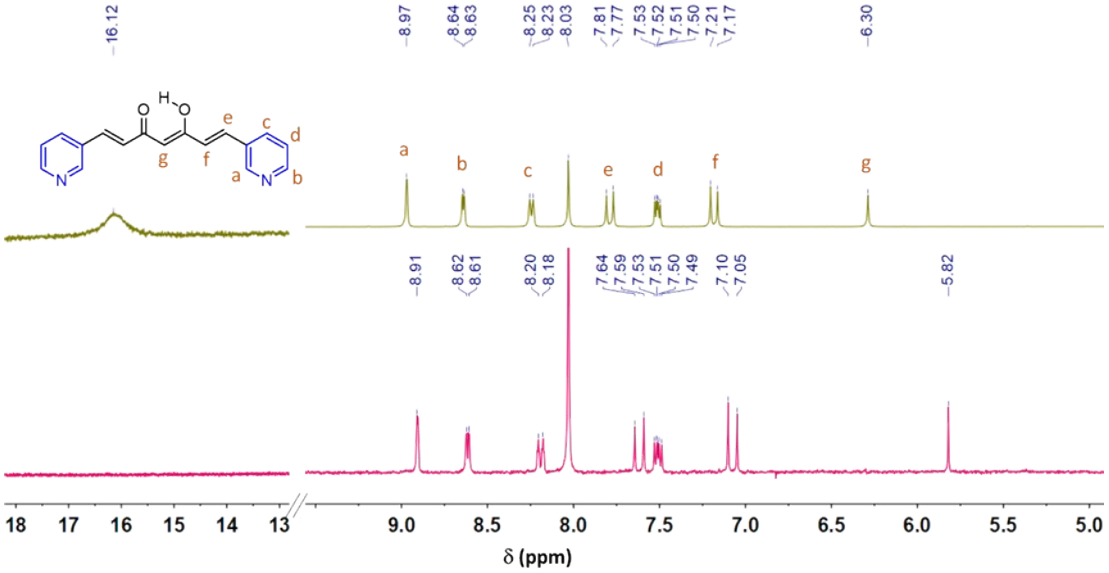
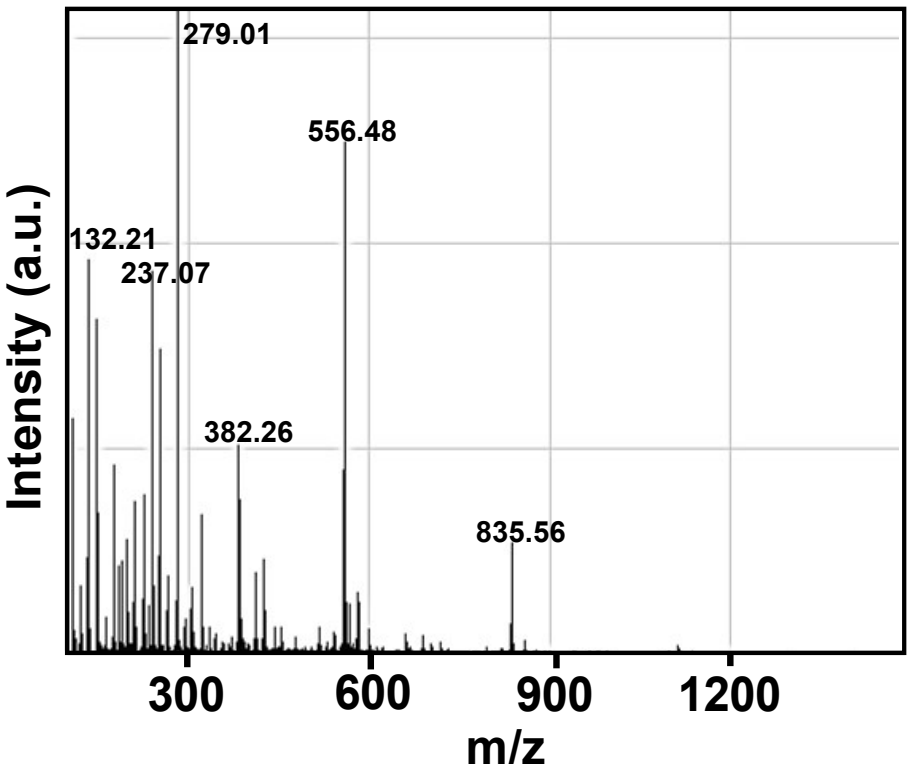


Figure S10. ^{1}H NMR in DMF-d_7 of 3pyCCMoid and a sample of **2**.



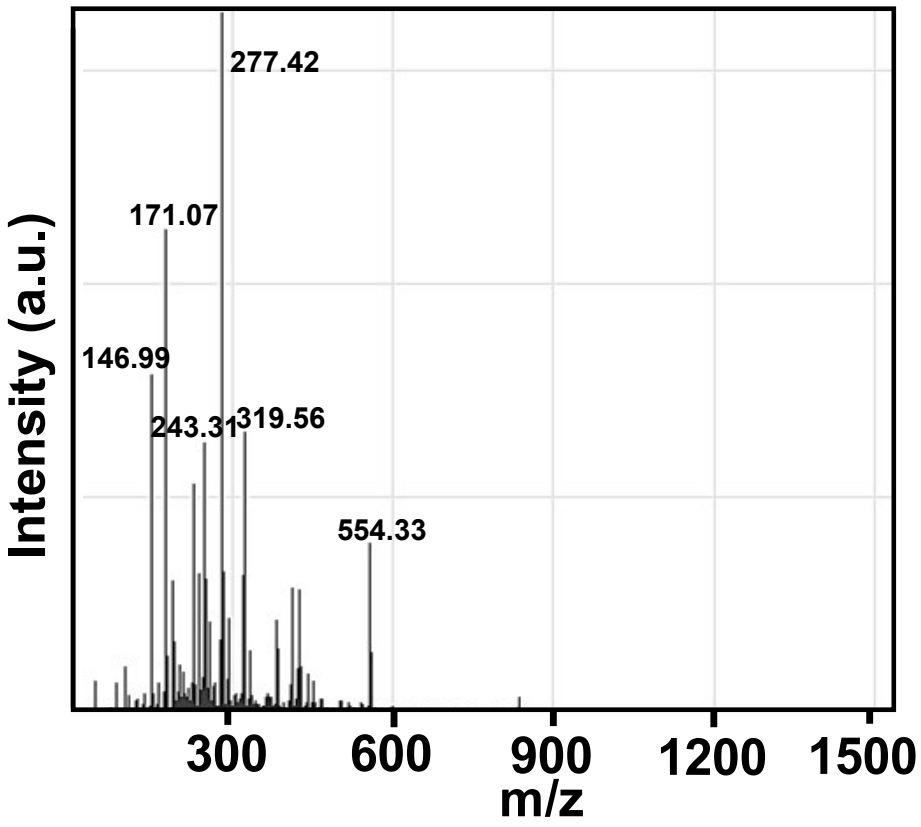


Figure S11. Mass spectrum of **2** after its partial re-dissolution in DMA, positive (top) and negative mode (down).

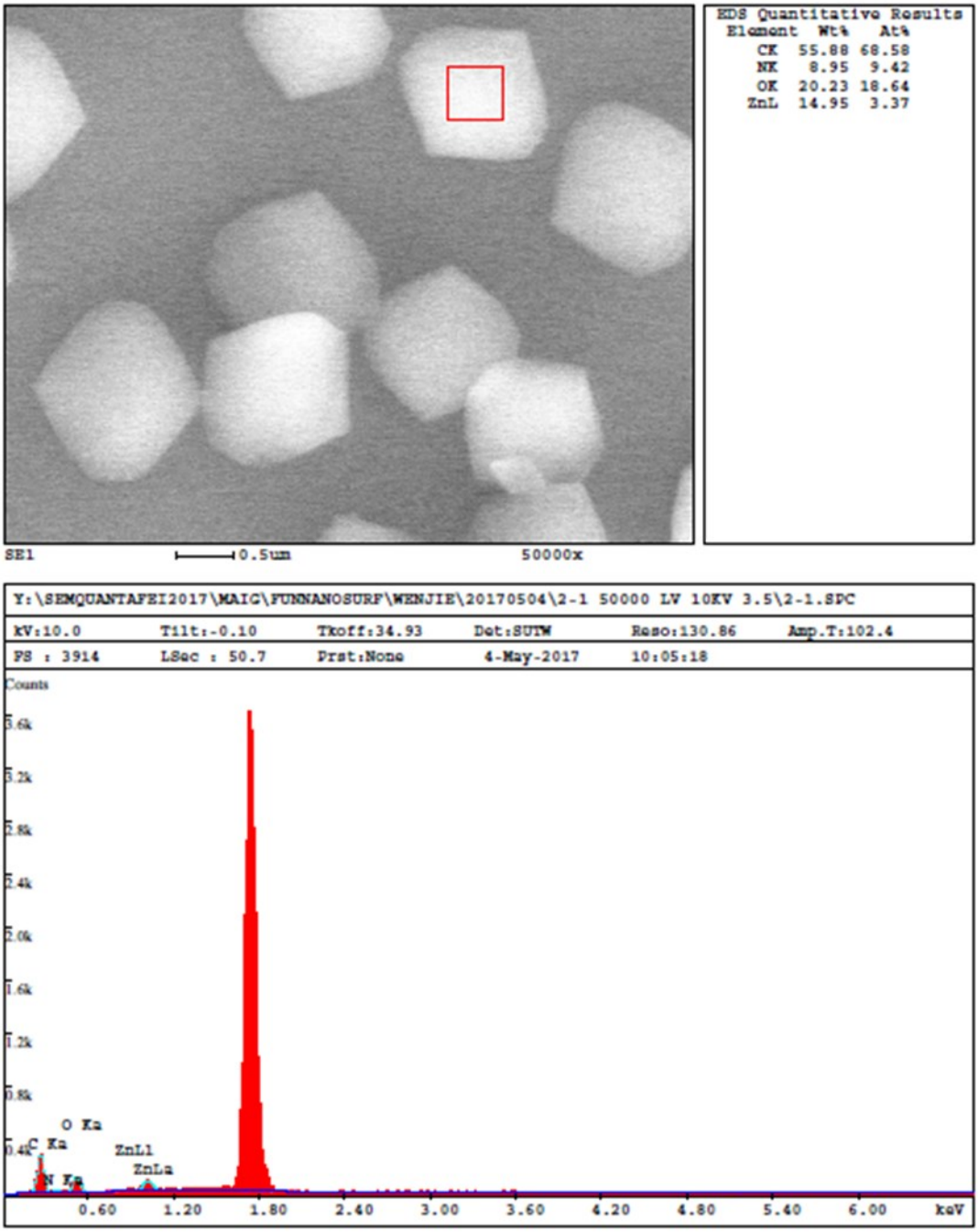


Figure S12. EDX spectrum of the cubes formed after re-dissolution in DMA of compound **2**, showing the same elements than the original crystal (C, O, N and Zn).

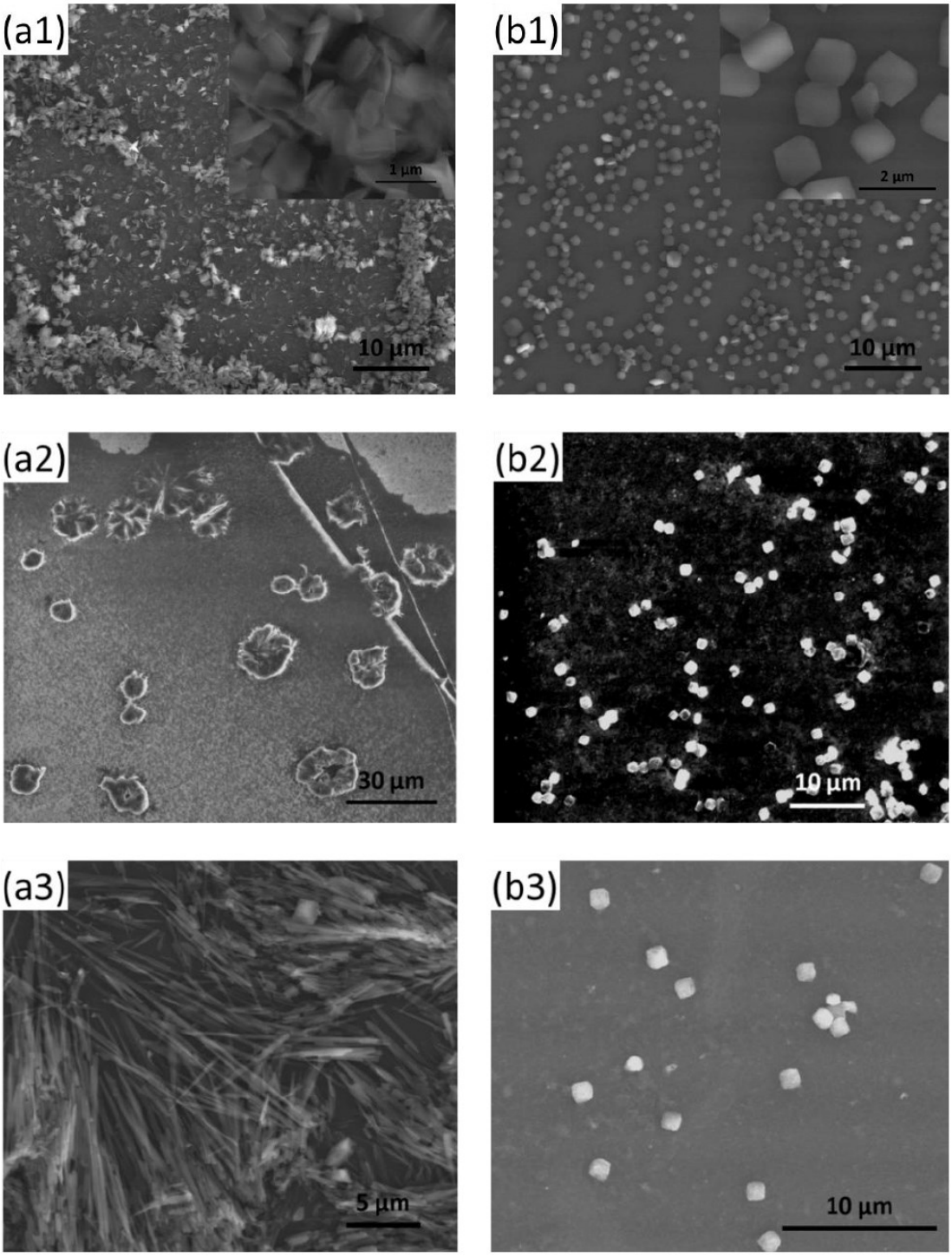


Figure S13. SEM images of 2 place onto different substrates of after adding DMA: (a1-3) aggregates from the unfiltered solution and (b1-3) solid part that remained after the addition of DMA. Numbers 1, 2 and 3 in (a) and (b) relate to SiO2/Si, mica and HOPG surfaces, respectively.

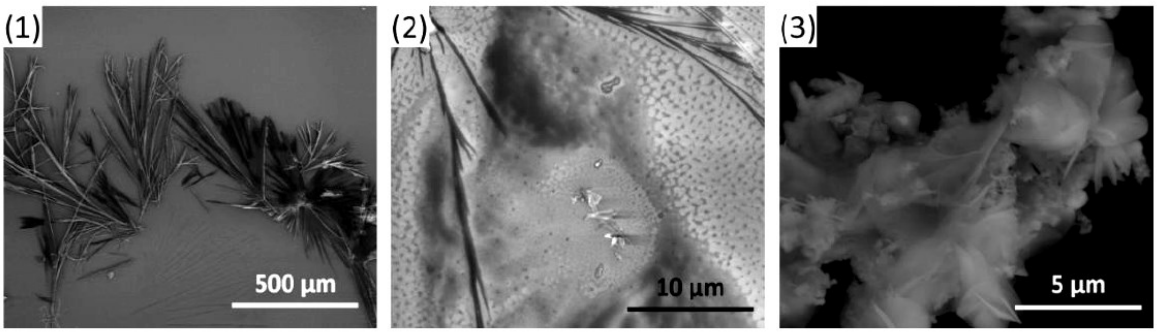


Figure S14. SEM images of 3pyCCMoid in different substrates: (1) SiO_2/Si wafer, (2) mica and (3) HOPG.

Figure S15. Appearance of a vial involving exfoliated 2 after 6h of resting. The largest nanosheets are precipitate at the bottom of the vial (left image) while smaller nanosheets remain dispersed in the solution, which is detected by Tyndall effect (right image).

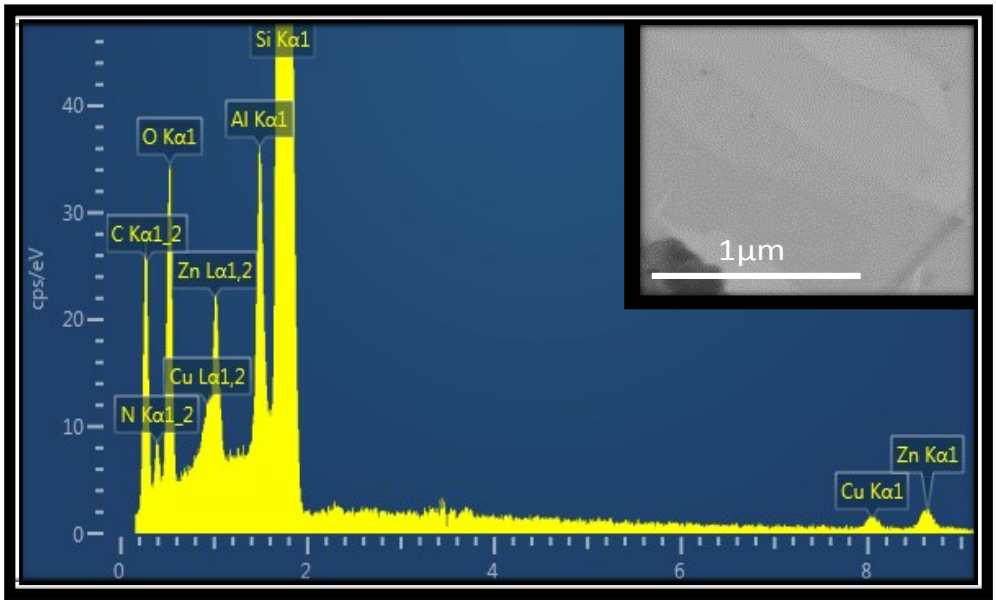


Figure S16. EDX spectrum of a single nanosheet of 2 displayed in the STEM picture, showing the same elements than the original crystal (C, O, N and Zn). Cu, Al and Si are related with the SEM components and the grid where the nanosheets are deposited.

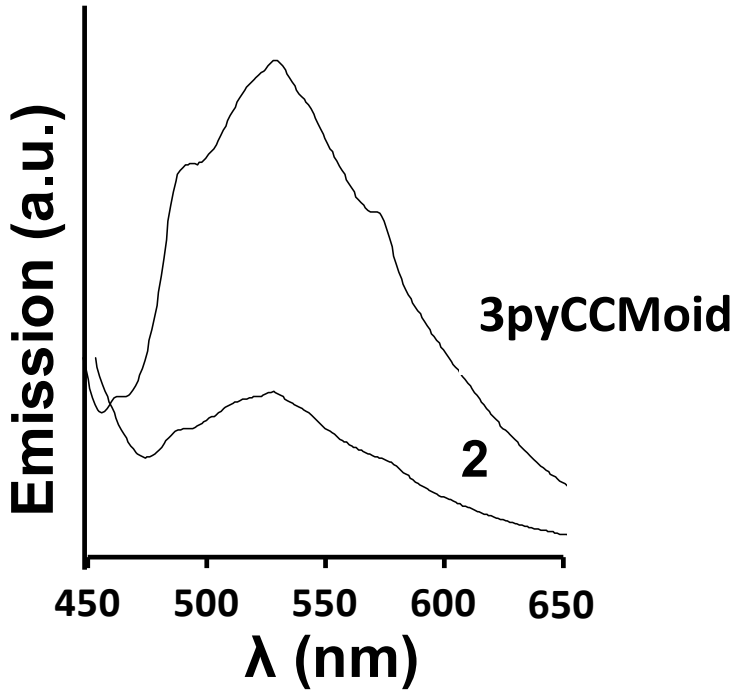


Figure S17. Fluorescence spectra of 3pyCCMoid (λmax=531 nm) and 2 (λmax=530nm). λ of excitation: 420 nm, excitation and emission slits 10 for 2, and 5 for 3pyCCMoid, in that order.