

Metal-organic Frameworks Based on Flexible Ligands (FL-MOFs): Structure and

Application

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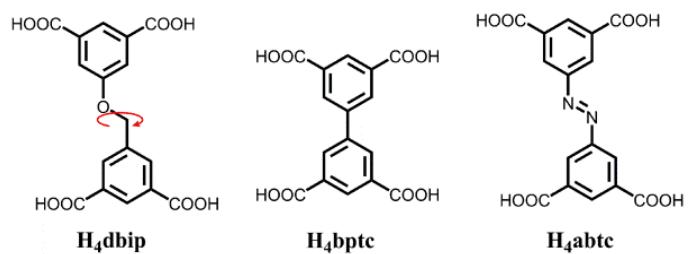


Fig. S1 The flexible and rigid tetracarboxylate ligands.

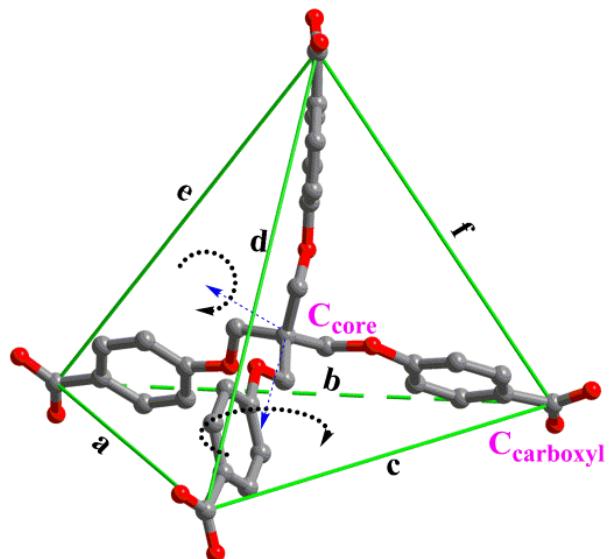


Fig. S2 The C_{carboxyl} tetrahedron in H₄tcm ligand, which can be uniquely determined by the lengths (denoted as *a-f*) of six edges (C_{carboxyl}-C_{carboxyl}). C_{carboxyl}-C_{carboxyl} edge scheme: *a*, the shortest edge; *b-e*: coplanar with *a* edge, *b* or *c* is the smallest value among *b-e*; view from C_{core}, both the edges *a-b-c* and *a-d-e* are clockwise arrangements.

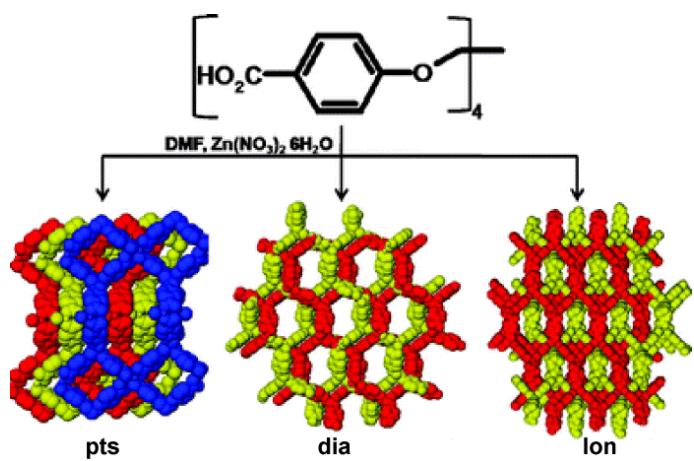


Fig. S3 Supramolecular isomers based on H₄tcm and zinc cations.

Table S1 The lengths of six edges in the C_{carboxyl} tetrahedron of the H₄tcm.

Formulae	a (Å)	b (Å)	c (Å)	d (Å)	e (Å)	f (Å)	comments	References
{[Zn ₂ (tcm)(H ₂ O)(DMF)]·3DMF} _n	11.983	13.025	13.566	13.071	13.639	12.186	isomers	¹
{[Zn ₂ (tcm)(DMF)]·x(H ₂ O,DMF)} _n	8.296	12.524	14.404	12.95	13.344	13.033		
{[Zn(H ₂ tcm)]·x(H ₂ O, DMF)} _n	10.172	13.995	12.999	13.558	14.328	11.347		
{[Zn ₂ (tcm)(H ₂ O)] ₂ ·H ₂ O} _n	9.319	9.896	14.945	13.105	15.031	11.419	isomers	²
{[Zn ₄ O(tcm) _{1.5}]·4DMA·10DEF·10H ₂ O} _n	10.68	11.256	14.883	13.658	14.883	10.68		
{[Cu ₂ (tcm)(H ₂ O) ₂]·7DMF·3(1,4-dioxane)·MeOH} _n (SNU-21)	10.805	11.507	15.341	11.507	15.341	12.318		
{[Cu ₂ (tcm)(H ₂ O) ₂]·4DMA·(H ₂ O) ₂ } _n	8.227	13.177	13.226	13.177	13.226	13.66	isomers	⁴
{Cd ₆ (tcm) ₃ (H ₂ O) ₆ ·Guest} _n	11.773	13.89	13.019	13.879	13.421	11.812		
{Cd ₂ (tcm)(H ₂ O) ₂ ·Guest} _n	11.509	14.012	12.947	13.455	13.811	11.272		
{Cd ₂ (tcm)(H ₂ O) ₃ ·Guest} _n	11.708	13.913	13.159	13.753	13.337	11.383	isomers	⁵
{[Co ₃ (Htcm) ₂]·(DMF) ₈ (H ₂ O) ₁₃ } _n	9.452	14.06	12.975	13.821	13.796	11.949		
{[Zn ₄ (tcm) ₂ (H ₂ O) ₃ (DMA)]·2H ₂ O} _n	10.734	13.378	11.197	14.934	12.707	13.031		
{[Co ₃ (tcm) ₂]·[NH ₂ (CH ₃) ₂] ₂ ·8DMA} _n	7.977	12.975	13.771	14.152	14.555	11.56	isomers	⁶
{[Cd ₂ (tcm)(DMA) ₂ (H ₂ O) ₂ } _n	9.274	14.98	10.287	15.104	13.137	11.755		
{[Mn ₃ (tcm) ₂]·[NH ₂ (CH ₃) ₂] ₂ ·8DMA} _n	9.028	14.882	10	15.152	13.146	11.458		
{[Cd ₃ (tcm) ₂]·[NH ₂ (CH ₃) ₂] ₂ ·8DMA} _n	10.998	13.753	13.753	13.753	13.753	10.998	isomorphous	⁷
{[Y ₃ (tcm) ₂ ·(NO ₃)·(DMA) ₂ ·(H ₂ O)]·5DMA·2H ₂ O} _n	10.02	14.018	14.018	14.018	14.018	10.02		
{[Dy ₃ (tcm) ₂ ·(NO ₃)·(DMA) ₂ ·(H ₂ O)]·5DMA·2H ₂ O} _n	7.546	12.338	13.123	14.089	14.849	12.308		
{[In ₂ (tcm) ₃ ·(OH) ₂]·3DMA·6H ₂ O} _n	8.516	13.446	15.076	13.712	15.115	8.635	isomorphous	⁸
{[In ₂ (tcm) ₃ ·(OH) ₂]·3DMA·6H ₂ O} _n	8.652	15.112	13.434	15.108	13.684	8.683		

{[Pb ₂ (tcm)·(DMA) ₂]·2DMA} _n	10.609	15.333	12.065	15.333	12.065	11.194			
{[Zn(tcm)(bipy)]·xsolvent} _n	10.448	13.536	13.245	13.792	14.57	10.495		⁹	
{[Zn ₂ (tcm)(CH ₃ CH ₂ OH)]·3H ₂ O} _n	11.136	11.997	13.516	12.761	14.01	13.057		¹⁰	
{[Mn ₃ (tcm) ₂] ²⁻ ·2[NH ₂ (CH ₃) ₂] ⁺ ·9DMF} _n	7.518	13.859	12.802	14.881	13.093	11.514		¹¹	
{Mn _{1.3} Ni _{2.7} (tcm) ₂ (H ₂ O) ₇ (DMF)·16DMF} _n	8.29	12.735	13.774	14.226	14.39	11.838			
{[Mn ₃ (tcm) ₂] ²⁻ ·2[NH ₂ (CH ₃) ₂] ⁺ ·9DMF} _n	8.927	13.546	12.408	14.608	13.686	12.261			
{MnCo ₃ (tcm) ₂ (H ₂ O) ₂ ·12DMF} _n	7.839	13.085	14	14.297	14.424	11.262			
{Mn _{1.3} Ni _{2.7} (tcm) ₂ (H ₂ O) ₇ (DMF)·16DMF} _n	8.535	12.968	14.153	13.523	14.392	11.903			
{[Y(tcm)[NH ₂ (CH ₃) ₂] ⁺ ·xsolent} _n	5.373	14.191	13.654	14.615	14.313	6.361	isomorphous	¹²	
{[La(tcm)[NH ₂ (CH ₃) ₂] ⁺ ·xsolent} _n	5.176	13.871	14.463	14.545	14.737	6.235			
{[Co ₂ (tcm)(dib)]·3DMF} _n	12.066	12.686	13.541	12.74	13.0731	12.233		¹³	
{[Co ₄ (tcm) ₂ (dib) ₃ (H ₂ O) ₄] ⁺ ·4H ₂ O} _n	10.785	13.601	13.339	14.114	13.461	11.476			
{[Co ₂ (tcm)(dibp)]·5DMF} _n	10.484	15.271	11.587	15.285	12.966	10.526			
{Co ₂ (tcm)(dibp) ₂ } _n	10.253	13.557	13.557	13.557	13.557	10.253			
{Co ₂ (tcm)(tt) ₂ } _n	11.817	12.892	13.954	13.155	13.954	11.817	isomorphous	¹⁴	
{Mn ₂ (tcm)(tt) ₂ } _n	11.87	12.903	14.096	13.339	14.096	11.87			
{Zn ₂ (tcm)(tt) ₂ } _n	11.955	13.91	12.701	13.91	13.113	11.955			
{Cd ₂ (tcm)(tt) ₂ } _n	12.104	13.926	12.191	13.926	12.859	12.104			
{[Co ₂ (tcm)(tim)]·4.25H ₂ O} _n	10.122	14.09	13.409	14.058	14.212	10.184			
{[Cd ₄ (tcm) ₂ (tim) ₂] ⁺ ·4H ₂ O·DMF} _n	10.36	13.922	13.991	13.922	13.991	10.454		¹⁵	
{[Co ₂ (tcm)(etbipy)]·2DMF·5H ₂ O} _n	12.26	12.429	12.707	14.295	12.789	12.283	isomorphous		
{[Zn ₂ (tcm)(etbipy)]·2.5DMF·2H ₂ O} _n	12.289	12.425	12.776	14.263	12.708	12.291			
{[Cd(H ₂ tcm)]·3DMF·3.5H ₂ O} _n	9.352	15.005	11.406	15.123	12.129	12.719			

H₄tcm = tetrakis[4-(carboxyphenyl)oxamethyl] methane acid; bipy = 4,4'-bipyridine; dibp = 4,4'-di(1H-imidazol-1-yl)-1,1'-biphenyl; dib = 1,4-di(1H-imidazol-1-yl)benzene; tt = 4-Tolyl-2,2':6',2''-terpyridine; tim = tetrakis(imidazol-1-ylmethyl)methane; etbipy = (E)-1,2-di(pyridin-4-yl)ethane.

Table S2 Selected gas adsorption and separation of FL-MOFs.

Compound	Formula	Pore Volum e	Surface area (m ² g ⁻¹) ^a		Adsorption Capacity (wt %)		Q_{st} (CO ₂) ^d	CO ₂ /N ₂ ^e	CO ₂ /CH ₄ ^e	Ref.
		Ratio	BET	Langmuir	H ₂ ^b	CO ₂ ^c				
	{[Zn ₂ (tcm)(CH ₃ CH ₂ OH)]·3H ₂ O} _n	34.4	367	479	0.32	—	—	—	—	10
	{[Zn ₄ O(tcm) _{1.5}]·4DMA·10DEF·10H ₂ O} _n	80	—	—	—	—	—	—	—	3
SNU-21S	{[Cu ₂ (tcm)(H ₂ O) ₂]·7DMF ·3(1,4-dioxane)·MeOH} _n	73.8	—	905	1.95	18.4	36.1	19.7 ^g	2.73 ^g	17
	{[Cu ₂ (tcm)(H ₂ O) ₂]·4DMA·(H ₂ O) ₂ } _n	48	382	507	1.22	—	—	—	—	5
	{Cu ₄ (ptmtip)(H ₂ O) ₄ ·(solvent)} _n	72	2896	—	2.37	16.7 ^h	35.2	—	—	18
	{Cu ₄ (H ₄ phmhip ₂)(H ₂ O) _x (DMF) _{4-x} ·(solvent)} _n	56	1490	—	1.80	18.4 ^h	29.7	—	—	
	{Cu ₄ (bmatip ₃)(H ₂ O) ₃ (DMF) ₃ ·(solvent)} _n	76	—	—	—	—	—	—	—	
	{Cu(ppip)·xsolvent} _n	58.6	—	—	—	—	—	—	—	19
	{Cu(mpip)·xsolvent} _n	53.6	—	—	—	—	—	—	—	
	{Cu(mpip)·xsolvent} _n	58.6	—	—	—	—	—	—	—	
	{Zn ₂₄ (bcbd) ₈ (H ₂ O) ₂₄ } _n	71	74.47	—	—	—	—	—	—	20
rht-MOF-7	{Cu ₃ (tdpat)(H ₂ O) ₃ ·xsolvent} _n	70	—	2170 ⁱ	—	28.7	44.7	—	—	21

	{[Cu ₄ (tdm)(H ₂ O) ₄]·xsolvent} _n	64.0	—	—	—	—	—	—	—	—	33
			1115	1722	2.12	27.5	—	—	—	—	34
PCN-26			1854	2545	2.57	35.6	—	49	8.4	—	35
	{In ₂ (tdm)(NH ₂ CH ₃) ₂ ·xsolvent} _n	65.1	1555	1707	1.49	19.6	21.14	—	—	—	36
			752	991	1.12	12.1	—	15.5 ^j	—	—	37
	{[In(dibp)·(Me ₂ NH ₂)]·xsolvent} _n	70.3	8.12	19.45	—	—	—	—	—	—	38
	{[In(dibp)·tma)]·xsolvent} _n	—	13.67	21.35	—	—	—	—	—	—	
	{[In(dibp)·tea)]·xsolvent} _n	—	5.81	13.63	—	—	—	—	—	—	
	{[In(dibp)·tpa)]·xsolvent} _n	—	37.41	57.05	—	—	—	—	—	—	
	{[In(dibp)·(tba)]·xsolvent} _n	—	325.65	477.77	—	—	—	—	—	—	
	{[In ₂ (tdm)·(Me ₂ NH ₂)]·xsolvent} _n	65.7	83.39	—	—	—	—	—	—	—	
	{[In ₂ (tdm)·(tma)]·xsolvent} _n	—	13.40	-22.55	—	—	—	—	—	—	
	{[In ₂ (tdm)·(tea)]·xsolvent} _n	—	754.63	1099.46	—	—	—	—	—	—	
	{[In ₂ (tdm)·(tpa)]·xsolvent} _n	—	36.26	54.17	—	—	—	—	—	—	
	{[In ₂ (tdm)·(tba)]·xsolvent} _n	—	120.76	181.45	—	—	—	—	—	—	
	{[Ln(cpi) ₂ (H ₂ O) ₂]·4H ₂ O} _n	32.1	—	147	—	4.3	—	—	—	—	39
	{Cd ₆ (tcm) ₃ ·(H ₂ O) ₆ } _n	42.3	231	249	—	5.6 ^h	—	—	—	—	1
	{In ₂ (OH) ₂ (obb) ₂ } _n	47.7	354.1	518	—	7.2	—	—	—	—	40

MSF-2	MSF-2	39.8	622	—	1.03	—	—	—	—	—	41
MSF-4	MSF-4	39.3	767	—	1.58	—	—	—	—	—	
	{Zn ₂ (tcm)(bipy)} _n	—	1150	—	—	5 ^h	—	—	—	—	42
	{[M ₂ (tpom)(D-cam) ₂]·2H ₂ O} _n	26.4%	111	380	—	—	—	—	—	—	43
	{[Zn ₃ (tcpt) ₂ (H ₂ O) ₂]·3H ₂ O·TEA·2DMF} _n	41.4	—	535	1.02	8.1	—	—	—	—	44
	{[Cd ₃ (tcpt) ₂ (H ₂ O) ₆]·1.5H ₂ O·2EtOH·DMF} _n	41.5	—	417	1.17	11.0	—	—	—	—	
NJU-bai3	{[Cu ₃ (H ₃ cip) ₂ (H ₂ O) ₅]·xGuest} _n	76.9	2690	3100	—	27.3	36.5	—	—	—	45
HNUST-1	{Cu ₂ (bdpt)} _n	71.0	1400	1620	2.14	30.7	~30.5	39.8	7.2	—	46
HNUST-3	{[Cu ₂ bdpo]·xsolvent} _n	73.2	2412	2784	2.2	16.6 ^h	24.8	26.1 ^g	7.9 ^g	—	47
	{[Cu ₂₄ (bcbd) ₈ (H ₂ O) ₂₄]·xsolvent} _n	74	3160	3570	—	42.54	26.3	22 ^k	—	—	48
	{Cu ₃ (btbip)} _n	81.1	3288	—	—	~24	34.3	8.6	—	49	
	{Cu ₃ (tatbip)} _n		3360	—	—						
Cu-tdpat	{[Cu ₃ (tdpta)(H ₂ O) ₃]·10H ₂ O·5DMA} _n	70.2	1938	2608	2.65	44.5	42.2	16 ^l	—	—	50
	{[Zn(btz)]·DMF·0.5H ₂ O} _n	45.6	1151	1222	2.28	35.6	31.2	35.6	21.1	—	51
SNU-100	{[Zn ₃ (TCPT) ₂ (HCOO)][NH ₂ (CH ₃) ₂ }] _n	38.1	814	—	1.81	14.1 ^h	—	25.5 ^f	—	52	
SNU-Li	—	—	924	—	1.93	15.3 ^h	—	37.8 ^f	—		
SNU-Mg	—	—	959	—	1.99	15.1 ^h	—	32.6 ^f	—		
SNU-Ca	—	—	935	—	1.88	15.1 ^h	—	40.4 ^f	—		

SNU-Co	—	—	1000	—	2.16	16.8 ^h	—	31.0 ^f	—	
SNU-Ni	—	—	982	—	1.98	16.6 ^h	—	37.4 ^f	—	

^a Derived from N₂ adsorption at 77 K. ^b Measured at 77 K and 1 atm. ^c Measured at 273 K and 1 atm. ^d The zero coverage isosteric heats of CO₂ adsorption (Q_{st}) calculated by using Clausius–Clapeyron equation. ^e Selectivities were calculated from the ratios of Henry constants for gas adsorption isotherms at 273 K. ^f Selectivities were calculated from the ratios of Henry constants for gas adsorption isotherms at 298 K. ^g The selectivities calculated from the uptake ratio by weight at 298 K. ^h Measured at 298 K and 1 atm. ⁱ Derived from Ar isotherm at 87 K. ^j The selectivities calculated from the uptake ratio by molar ratios at 273 K. ^k The IAST-predicted selectivity for equimolar CO₂/N₂ mixtures at 298 K. ^lThe selectivities calculated from the uptake ratio by molar ratios at 298 K.

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