Supplementary Information

A Flexible and Stable Interpenetrated Indium-Pyridylcarboxylate Framework with Breathing Behaviours and High Selective Adsorption of Cationic Dyes

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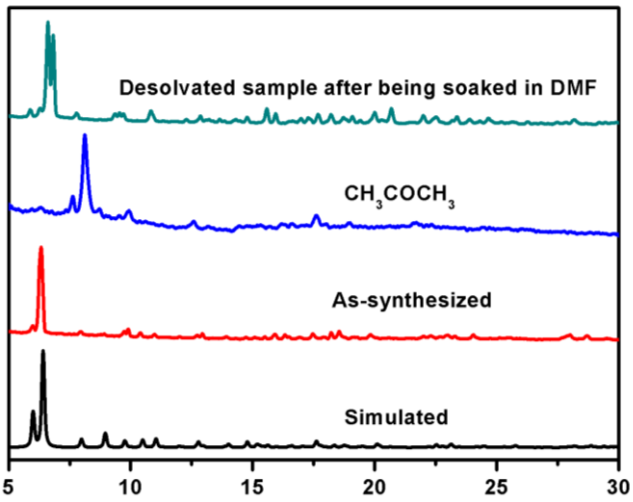


Figure S1. PXRD patterns of **1** simulated from the X-ray single-crystal structure, as-synthesized, desolvated sample and desolvated sample after being soaked in water of **1**.

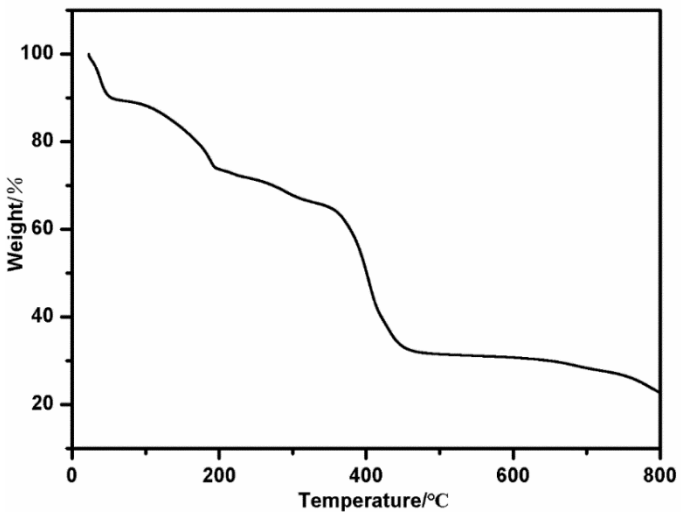


Figure S2. TGA plots of **1** under N_2 environment.

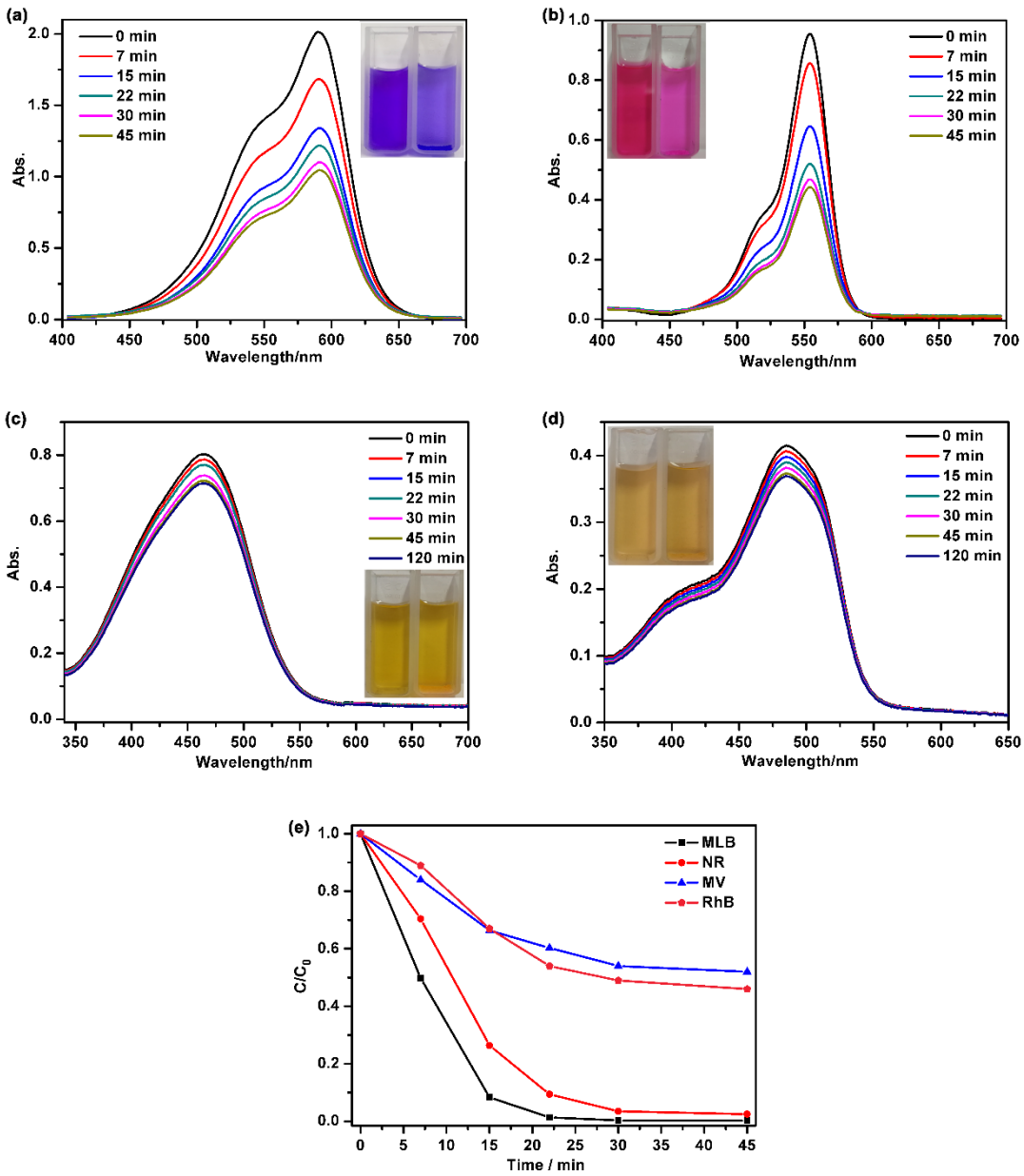


Figure S3. UV-vis spectra of MV (a), RhB (b), MO (c) and OrII (d) in aqueous solutions at different time; (e) the adsorption rate of MLB, NR, MV and RhB in **1**.

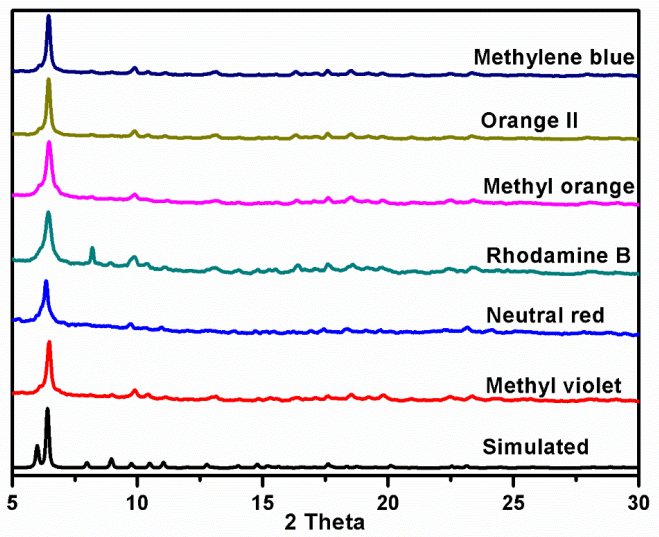


Figure S4. The PXRD patterns of **1** after immersing in different organic dyes.

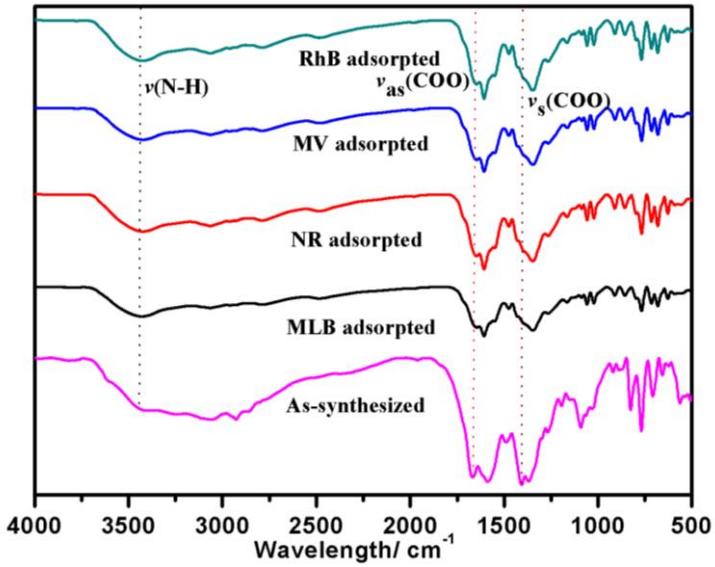


Figure S5. FT-IR spectra of **1** at the as-synthesized, MLB, NR, MV and RhB adsorbed states.

Computational methods

Based on the density functional theory (DFT), we chose the Vienna Ab-initio Simulation Package (VASP) package for first-principle calculations. The generalized gradient approximation (GGA) with the Perdew− Burke−Ernzerhof (PBE) functional were used to describe the electronic exchange and correlation effects. Uniform G-centered k-points meshes with a resolution of 2π*0.03 $Å^{-1}$ and Methfessel-Paxton electronic smearing were adopted for the integration in the Brillouin zone for geometric optimization. The simulation was run with a cutoff energy of 500 eV throughout the computations. These settings ensure convergence of the total energies to within 1 meV per atom. Structure relaxation proceeded until all forces on atoms were less than 1 meV $Å^{-1}$ and the total stress tensor was within 0.01 GPa of the target value. The adsorption energy of molecule on MOF surface ($\Delta E_{adsorption}$) was defined as

$$\Delta E_{adsorption} = E_{MOF+dye} - E_{MOF} - E_{dye}$$

where $E_{MOF+dye}$, E_{MOF} and E_{dye} stand for the ground-state energies of the substrate binding with molecules.

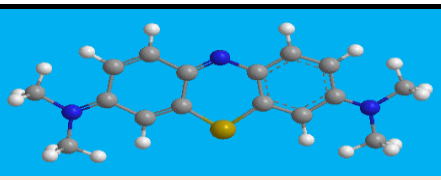
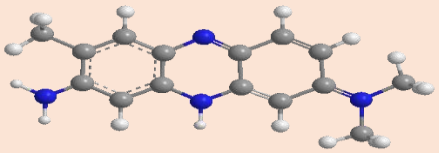
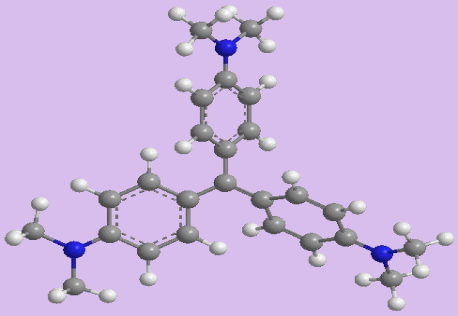
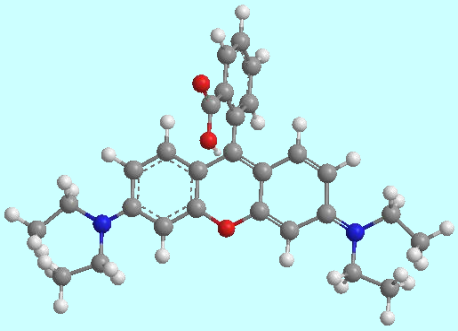
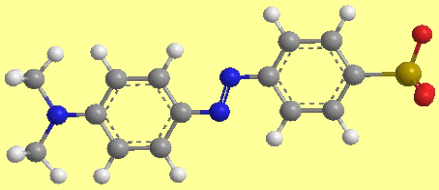
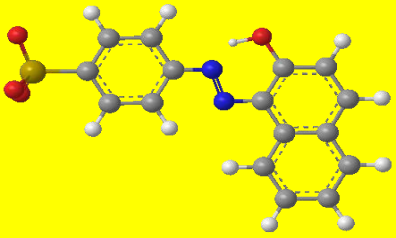
Table S1 The maximum adsorption amounts for MLB and NR on **1**, based on the data of different initial concentrations experiments.

Dyes	1	2	3	4	5	Average (mg/g)
MLB	415	412	408	400	418	410
NR	200	200	210	205	198	202

Table S2 Comparison of maximum adsorption amounts for MLB on various adsorbents.

Adsorbent	Q_e(mg/g)	Solvent	Ref.
CTS-g-PAA/10% VMT	1685.6	Water	1
MIL-100(Fe)	1105	Water	2
$[Ca(HDCPP)_2(H_2O)_2](DMF)_{1.5}$	952	Water	3
ZJU-24	902	Water	4
$H_3PW_{12}O_{40}$@ZIF-8	810	Water	5
MIL-100(Cr)	645	Water	6
PAC2	588	Water	7
1	410	Water	This work
Cd-MOF (2)	318	Water	8
JLU-Liu39	308	C_2H_5OH	9
Sr-BTTC	270	DMA	10
MOF-235	187	Water	11

Table S3 Molecular parameters of the dyes

Dyes	Charge	Dimensions (Å)	Molecular structure
Methylene blue	+1	1.8×5.5×14.2	
Neutral red	+1	2.3×6.4×12.6	
Methyl violet	+1	3.5×13.0×13.7	
Rhodamine B	+1	15.6×13.5×4.2	
Methyl orange	-1	4.5×6.0×14.8	
Orange II	-1	13.5×7.2×2.8	

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