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Table S1 † Selected bond lengths (Å) and angles (°) for **1-5**

Complex **1**			
Cd1-O10^{i}	2.118 (2)	Co1-N1^{i}	2.159 (3)
Co1-O10	2.118 (2)	Co1-N1	2.159 (3)
Co1-N2^{i}	2.110 (3)	Co2-O1	1.941 (2)
Co2-O5^{ii}	1.962 (2)	Co2-N7	2.012 (3)
Co2-N8	2.041 (3)	Co1-N2	2.110 (3)
O10^{i}-Co1-O10	180.00 (13)	O10-Co1-N1	90.31 (9)
O10^{i}-Co1-N1^{i}	90.31 (9)	O10-Co1-N1^{i}	89.69 (9)
O10^{i}-Co1-N1	89.69 (9)	N1^{i}-Co1-N1	180.0
N2^{i}-Co1-O10	90.80 (10)	N2^{i}-Co1-O10^{i}	89.20 (10)
N2-Co1-O10	89.20 (10)	N2-Co1-O10^{i}	90.80 (10)
N2^{i}-Co1-N1	94.84 (10)	N2-Co1-N1	85.16 (10)
N2^{i}-Co1-N1^{i}	85.16 (10)	N2-Co1-N1^{i}	94.84 (10)
N2-Co1-N2^{i}	180.0	O1-Co2-O5^{ii}	108.33 (10)
O1-Co2-N7	112.52 (10)	O1-Co2-N8	111.96 (11)
O5^{ii}-Co2-N7	113.88 (10)	O5^{ii}-Co2-N8	100.21 (10)
N7-Co2-N8	109.35 (11)		
Symmetry codes: i) −x+2, −y+2, −z; ii) x, y−1, z			
Complex **2**			
Co1-O1	2.0499 (12)	Co1-O7^{ii}	2.0970 (14)
Co1-O3^{i}	2.0542 (12)	Co1-O8	2.1996 (13)
Co1-N1	2.1046 (16)	Co1-N5	2.1087 (16)
Co2-O4	2.0685 (13)	Co2-O4^{iii}	2.0684 (13)
Co2-O8^{iv}	2.1951 (12)	Co2-O8^{v}	2.1951 (12)
Co2-N4^{vi}	2.0986 (16)	Co2-N4^{vii}	2.0986 (16)
O3-Co1^{iv}	2.0542 (12)	O7-Co1^{ii}	2.0970 (14)
O8-Co2^{i}	2.1951 (12)	N4-Co2^{viii}	2.0987 (16)
O1-Co1-O3^{i}	173.04 (6)	O1-Co1-O7^{ii}	90.04 (6)
O1-Co1-O8	87.06 (5)	O1-Co1-N1	93.03 (6)
O1-Co1-N5	94.04 (6)	O3^{i}-Co1-O7^{ii}	83.09 (6)
O3^{i}-Co1-O8	93.72 (5)	O3^{i}-Co1-N1	93.81 (6)
O3^{i}-Co1-N5	84.68 (6)	O7^{ii}-Co1-O8	87.43 (5)
O7^{ii}-Co1-N1	176.64 (6)	O7^{ii}-Co1-N5	88.34 (6)
N1-Co1-O8	94.09 (6)	N1-Co1-N5	90.08 (6)
N5-Co1-O8	175.62 (6)	O4^{iii}-Co2-O4	180.0
O4-Co2-O8^{v}	82.14 (5)	O4^{iii}-Co2-O8^{iv}	82.14 (5)
O4-Co2-O8^{iv}	97.86 (5)	O4^{iii}-Co2-O8^{v}	97.86 (5)
O4^{iii}-Co2-N4^{vii}	90.29 (6)	O4^{iii}-Co2-N4^{vi}	89.71 (6)
O4-Co2-N4^{vi}	90.29 (6)	O4-Co2-N4^{vii}	89.71 (6)
O8^{iv}-Co2-O8^{v}	180.0	N4^{vii}-Co2-O8^{iv}	91.64 (6)
N4^{vii}-Co2-O8^{v}	88.36 (6)	N4^{vi}-Co2-O8^{iv}	88.36 (6)
N4^{vi}-Co2-O8^{v}	91.64 (6)	N4^{vii}-Co2-N4^{vi}	180.0
Co2^{i}-O8-Co1	117.46 (6)		

Symmetry codes: i) x+1, y, z; ii) −x+1, −y, −z+2; iii) −x, −y, −z+1; iv) x−1, y, z; v) −x+1, −y, −z+1; vi) x, y−1, z+1; vii) −x, −y+1, −z; viii) x, y+1, z−1;			
Complex **3**			
Co1-O2	1.9643 (14)	Co1-O4^{i}	1.9642 (14)
Co1-N1	2.0210 (17)	Co1-N4^{ii}	2.0412 (16)
O4-Co1^{iii}	1.9643 (14)	N4-Co1^{ii}	2.0413 (15)
O2-Co1-N1	111.84 (6)	O2-Co1-N4^{ii}	94.32 (6)
O4^{i}-Co1-O2	110.99 (6)	O4^{i}-Co1-N1	114.67 (6)
O4^{i}-Co1-N4^{ii}	112.51 (6)	N1-Co1-N4^{ii}	110.78 (6)
Symmetry codes: i) x, −y+1/2, z+1/2; ii) −x+2, −y+1, −z+2; iii) x, −y+1/2, z−1/2			
Complex **4**			
Co1-O2	2.054 (3)	Co1-O3^{i}	2.202 (3)
Co1-O4^{i}	2.191 (3)	Co1-N1	2.086 (4)
Co1-N5^{ii}	2.092 (4)	Co1-N6	2.223 (3)
Co2-O8	2.016 (3)	Co2-O10^{iii}	2.387 (4)
Co2-O11^{iii}	2.118 (3)	Co2-N3^{iv}	2.262 (3)
Co2-N8^{v}	2.104 (4)	Co2-N10	2.079 (3)
O4-Co1^{iii}	2.191 (3)	O11-Co2^{i}	2.118 (3)
O10-Co2^{i}	2.387 (4)	N3-Co2^{vi}	2.262 (3)
N5-Co1^{ii}	2.092 (4)	N8-Co2^{v}	2.104 (4)
O2-Co1-O3^{i}	147.29 (13)	O2-Co1-O4^{i}	88.07 (12)
O2-Co1-N1	87.19 (14)	O2-Co1-N5^{ii}	126.77 (13)
O2-Co1-N6	81.50 (13)	O3^{i}-Co1-N6	93.88 (14)
O4^{i}-Co1-O3^{i}	59.59 (12)	O4^{i}-Co1-N6	91.85 (13)
N1-Co1-O3^{i}	99.24 (15)	N1-Co1-O4^{i}	94.42 (13)
N1-Co1-N5^{ii}	96.23 (14)	N1-Co1-N6	166.87 (15)
N5^{ii}-Co1-O3^{i}	84.66 (13)	N5^{ii}-Co1-O4^{i}	143.91 (13)
N5^{ii}-Co1-N6	85.35 (14)	O8-Co2-O10^{iii}	155.01 (12)
O8-Co2-O11^{iii}	97.28 (13)	O8-Co2-N3^{iv}	83.13 (13)
O8-Co2-N8^{v}	122.86 (13)	O8-Co2-N10	93.57 (14)
O11^{iii}-Co2-O10^{iii}	57.74 (12)	O11^{iii}-Co2-N3^{iv}	85.75 (13)
N3^{iv}-Co2-O10^{iii}	93.59 (13)	N8^{v}-Co2-O10^{iii}	81.01 (13)
N8^{v}-Co2-O11^{iii}	136.37 (13)	N8^{v}-Co2-N3^{iv}	83.12 (13)
N10-Co2-O10^{iii}	91.76 (13)	N10-Co2-O11^{iii}	100.28 (13)
N10-Co2-N3^{iv}	173.48 (14)	N10-Co2-N8^{v}	94.04 (14)
Symmetry codes: i) x+1, y, z; ii) −x+1, −y+1, −z+1; iii) x−1, y, z; iv) x, y, z−1; v) −x+1, −y+2, −z; vi) x, y, z+1			
Complex **5**			
Co1-O1	2.0686 (16)	Co1-O3^{i}	2.0917 (17)
Co1-N6^{ii}	2.1127 (18)	Co1-N1	2.1185 (17)
Co1-N4^{iii}	2.1207 (18)	O3-Co1^{iv}	2.0917 (17)
N6-Co1^{ii}	2.1127 (18)	N4-Co1^{v}	2.1206 (18)
O1-Co1-O3^{i}	133.89 (7)	O1-Co1-N6^{ii}	135.80 (7)
O1-Co1-N1	84.82 (7)	O1-Co1-N4^{iii}	88.52 (7)

$O3^{i}$-Co1-$N6^{ii}$	90.31 (7)	$O3^{i}$-Co1-N1	92.23 (7)
$O3^{i}$-Co1-$N4^{iii}$	92.83 (7)	$N6^{ii}$-Co1-N1	94.93 (7)
$N6^{ii}$-Co1-$N4^{iii}$	89.41 (7)	N1-Co1-$N4^{iii}$	173.31 (7)

Symmetry codes: i) x+1, y, z; ii) −x+1, −y+1, −z+2; iii) x, y−1, z; iv) x−1, y, z; v) x, y+1, z

Table S2 † Comparison of the dye adsorption capacities

Adsorbent	Adsorption Capacity (MO)	Reference
$\{[Co_3(L)_2(4,4'\text{-bibp})_3(\mu_2\text{-}O)_2]\cdot 2H_2O\}_n$	97.1%	This work
$\{[Co_2(CHOO)_3(bibp)_2]\cdot NO_3\cdot H_2O\}_n$	95.1%	J. Solid State Chem., 2017, 248, 109
$\{[Ni_2(CHOO)_3(bibp)_2]\cdot NO_3\cdot H_2O\}_n$	95.1%	
$\{[Cu_2(CHOO)_3(bibp)_2]\cdot CHOO\}_n$	21.1%	
$\{[(CH_3)_2NH_2][Co_2NaL_2(CH_3COO)_2]\cdot xS\}_n$	No adsorption	J. Mater. Chem. A, 2015, 3, 12804
$[Co(L)]_n$	No adsorption	Inorg. Chem., 2016, 55, 8816
$\{[Co(L)(BIBP)]\cdot H_2O\}_n$	91.7%	
$\{[Co_3(L)(BPY)_{1.5}]\cdot H_2O\}_n$	No adsorption	
amino-MIL-101(Al)	99.3%	J. Mater. Chem. A, 2014, 2, 193
$(Me_2NH_2)_2[Zn_2L_{1.5}bpy]\cdot 2DMF$	No adsorption	J. Solid State Chem., 2016, 233, 143
Adsorbent	Adsorption Capacity (AH)	Reference
$\{[Co_{1.5}(HL)(4,4'\text{-bidpe})_2(H_2O)]\cdot 3H_2O\}_n$	92.8%	This work
$\{[Co_3(L)_2(4,4'\text{-bibp})_3(\mu_2\text{-}O)_2]\cdot 2H_2O\}_n$	91.3%	
$\{[Co(HL)(1,3\text{-bitl})]\cdot(1,4\text{-Diox})\}_n$	92.1%	
$[Co_2(HL)_2(3,5\text{-bipd})_2]_n$	90.0%	
$\{[Co(HL)(tib)]\cdot 0.5H_2O\cdot NMP\}_n$	90.6%	
$\{[(CH_3)_2NH_2][Co_2NaL_2(CH_3COO)_2]\cdot xS\}_n$	85.0%	J. Mater. Chem. A, 2015, 3, 12804
$(Me_2NH_2)_2[Zn_2L_{1.5}bpy]\cdot 2DMF$	95.0%	J. Solid State Chem., 2016, 233, 143
Adsorbent	Adsorption Capacity (GR)	Reference
$\{[Co_{1.5}(HL)(4,4'\text{-bidpe})_2(H_2O)]\cdot 3H_2O\}_n$	91.8%	This work
$\{[Co_3(L)_2(4,4'\text{-bibp})_3(\mu_2\text{-}O)_2]\cdot 2H_2O\}_n$	91.0%	
$\{[Co(HL)(1,3\text{-bitl})]\cdot(1,4\text{-Diox})\}_n$	91.8%	
$[Co_2(HL)_2(3,5\text{-bipd})_2]_n$	78.5%	
$\{[Co(HL)(tib)]\cdot 0.5H_2O\cdot NMP\}_n$	70.8%	
$[Co(L)]_n$	83.3%	Inorg. Chem., 2016, 55, 8816
$\{[Co(L)(BIBP)]\cdot H_2O\}_n$	87.5%	
$\{[Co_3(L)(BPY)_{1.5}]\cdot H_2O\}_n$	No adsorption	
$[Cu(bipy)(SO_4)]_n$	98.5%	Chem. Eng. J., 2015, 265, 157
$[Co(L1)(tp)]_n$	92.8%	Inorg. Chem. Commun., 2013, 37, 54
$[Co(L_2)(Htp)(tp)_{0.5}]_n$	62.9%	
$[Zn_2(L)_2(bpe)_2(H_2O)_2]_n$	92.0%	Dalton Trans., 2015, 44, 18795
$[Zn(L)(rctt\text{-tpcb})_{0.5}(H_2O)]_n$	86.0%	

Fig. S1

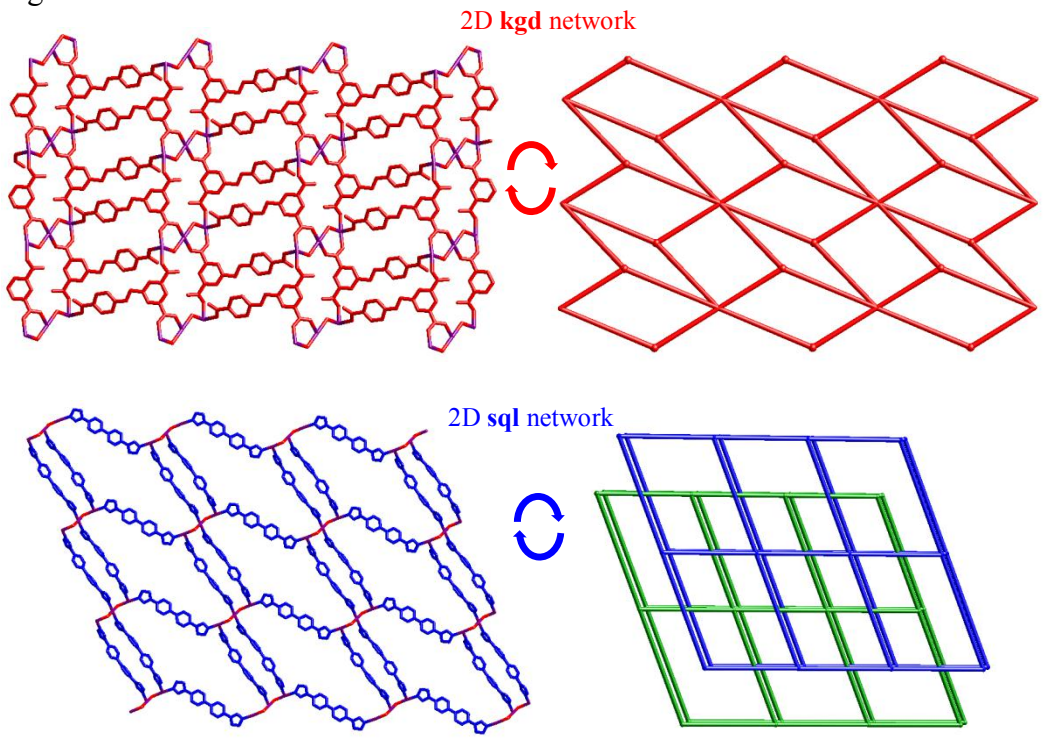


Fig. S1 View of the 2D **kgd** $[Co_3(L^{3-})_2]_n$ network and 2D **sql** $[Co_3(4,4'\text{-bibp})_2]_n^{6n+}$ network. (blue spheres: 4,4'-bibp ligands; red spheres: L^{3-} ligands)

Fig. S2

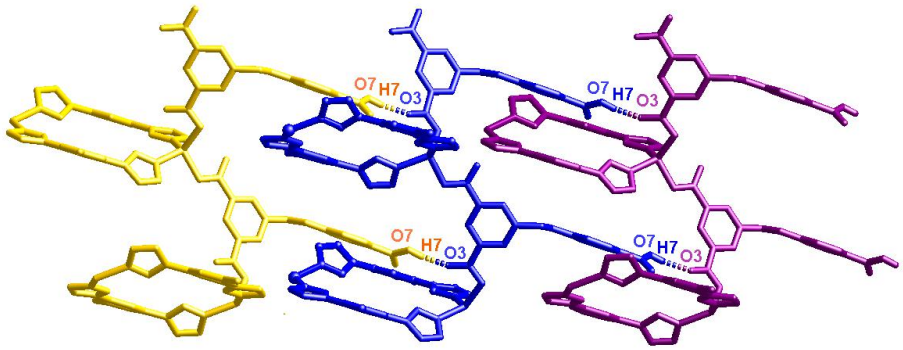


Fig. S2 The O-H···O bonds in complex **3**.

Fig. S3

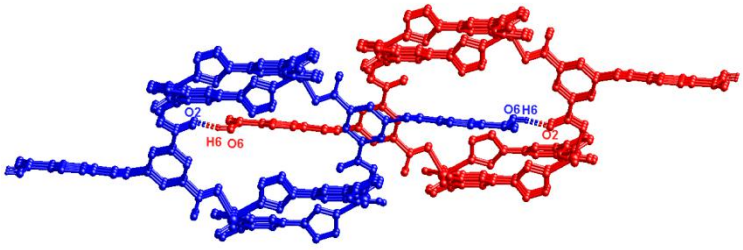


Fig. S3 The O-H···O bonds in complex **5**.

Fig. S4

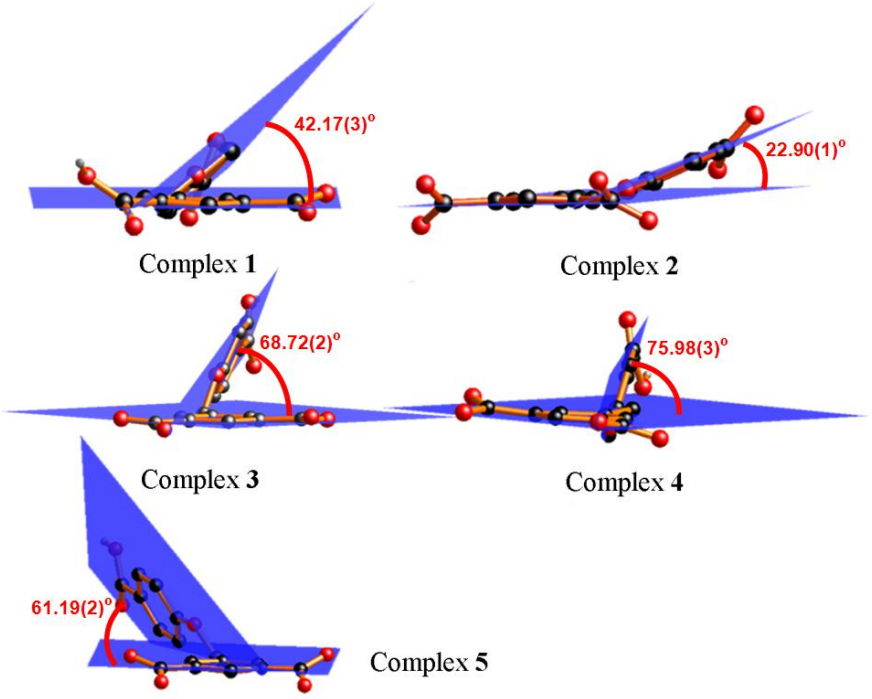


Fig. S4 The dihedral angles between the two phenyl rings of H_3L ligands in complexes **1**-**5**.

Fig. S5

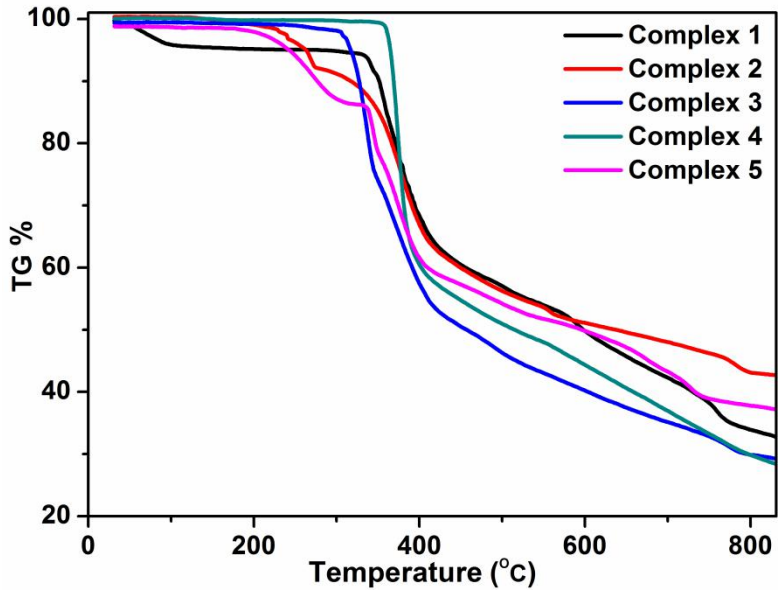


Fig. S5 The TGA curves for complexes **1**-**5**.

Fig. S6

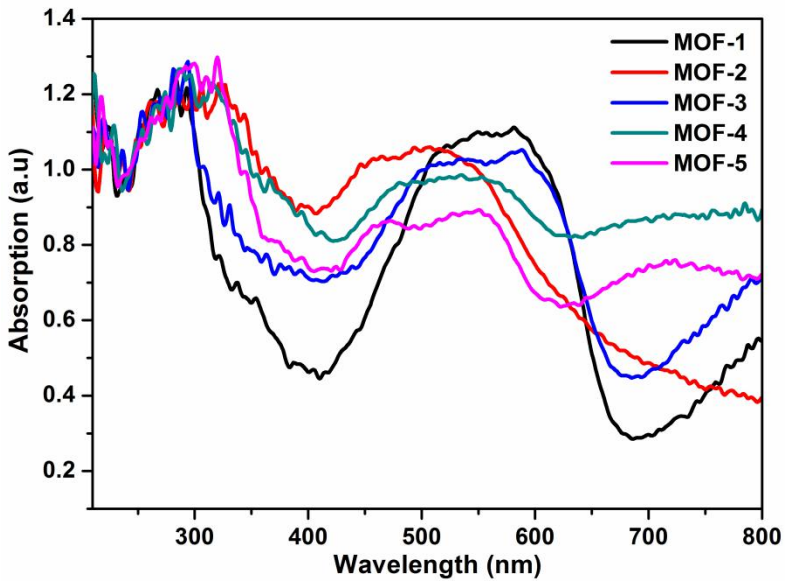


Fig. S6 The UV-vis spectra of complexes **1-5**.

Fig. S7

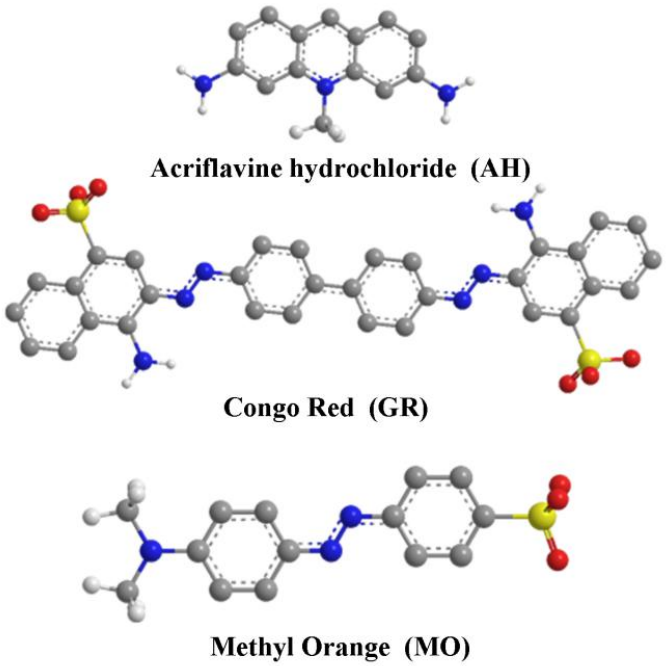


Fig. S7 The different structures and sizes of three organic dyes.

Fig. S8

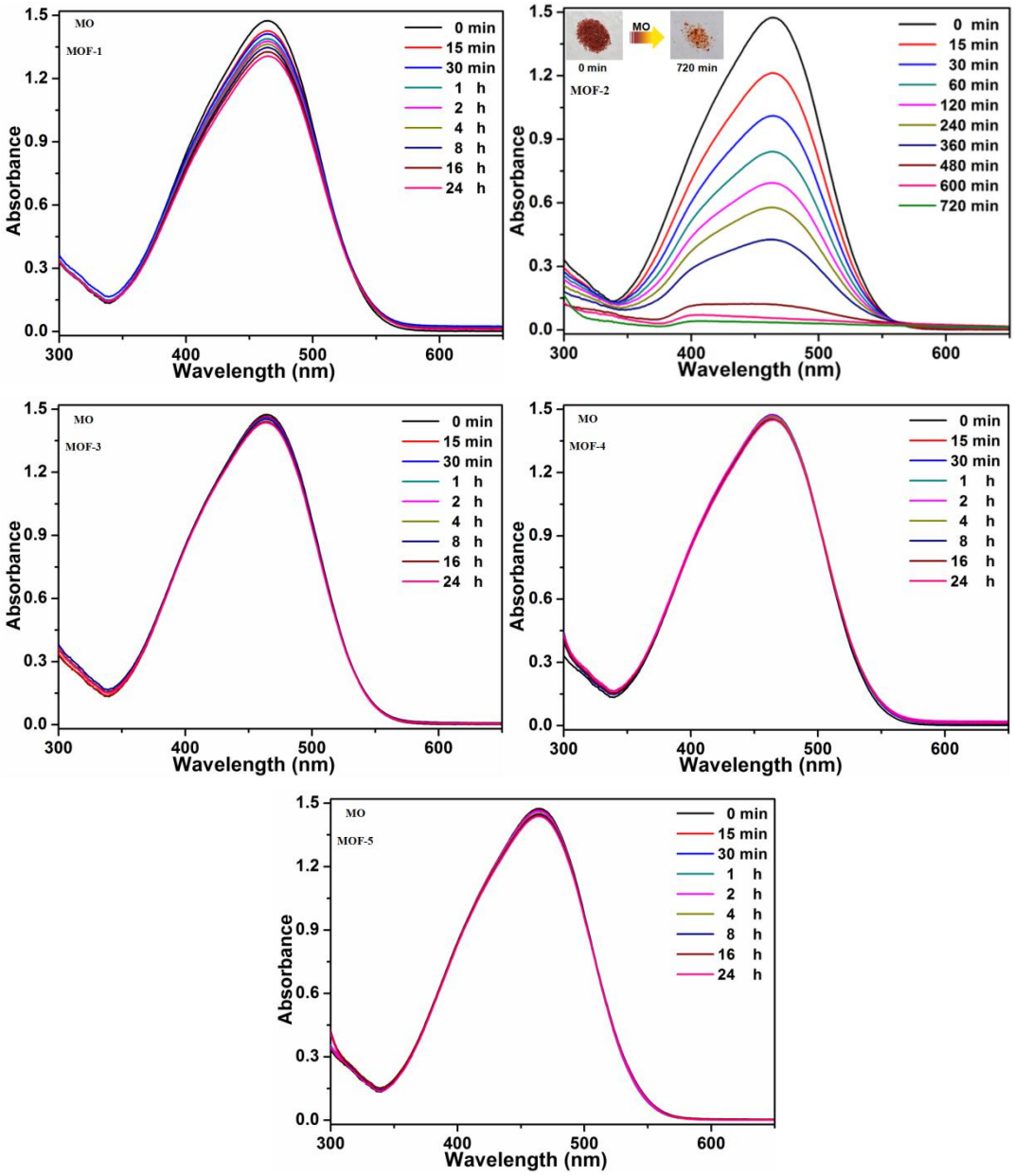


Fig. S8 UV-vis spectra for the uptake of MO from aqueous solutions at various time intervals for **1**-**5**, respectively.

Fig. S9

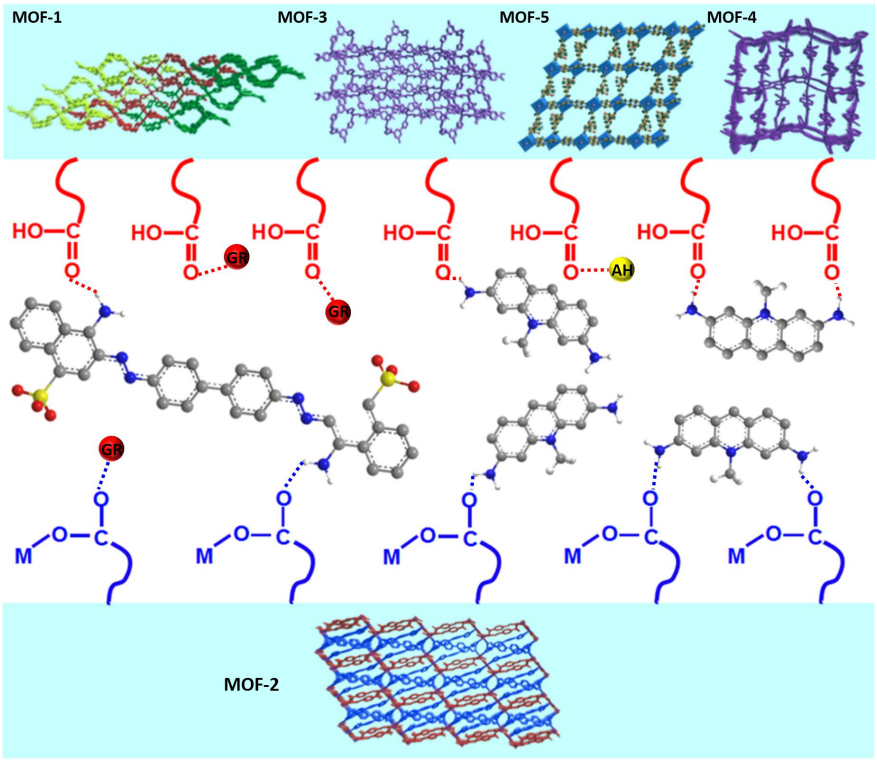
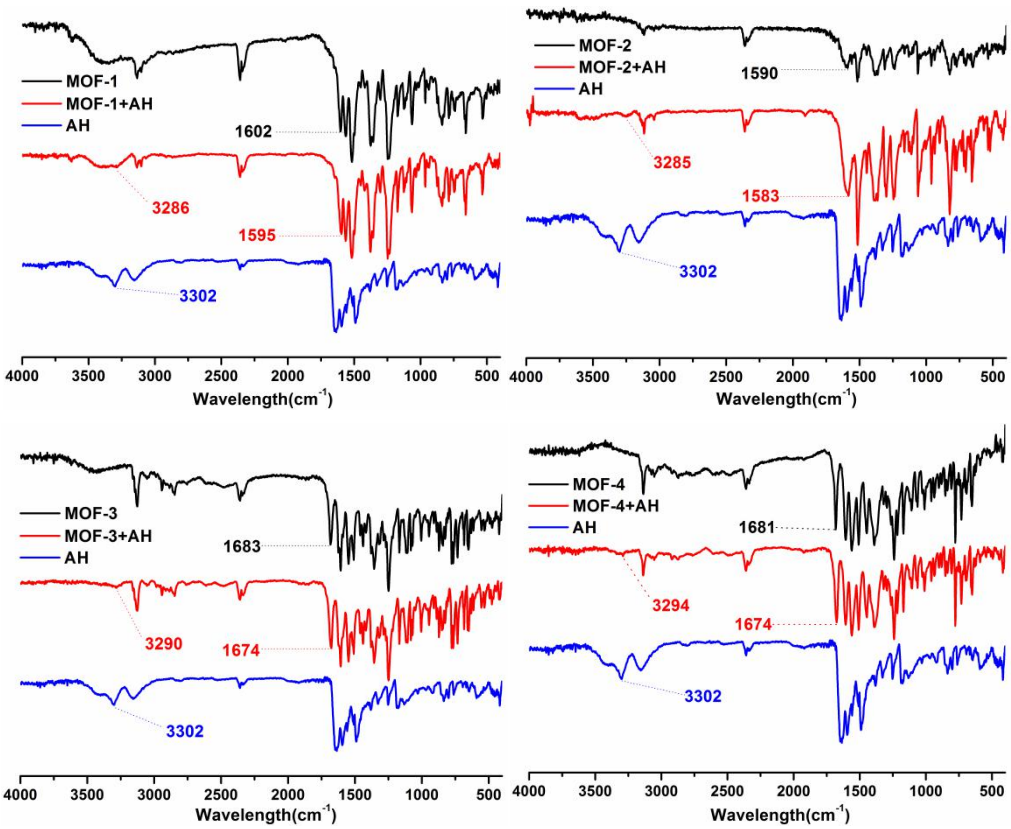


Fig. S9 The possible mechanism between two amino organic dyes and MOFs.

Fig. S10



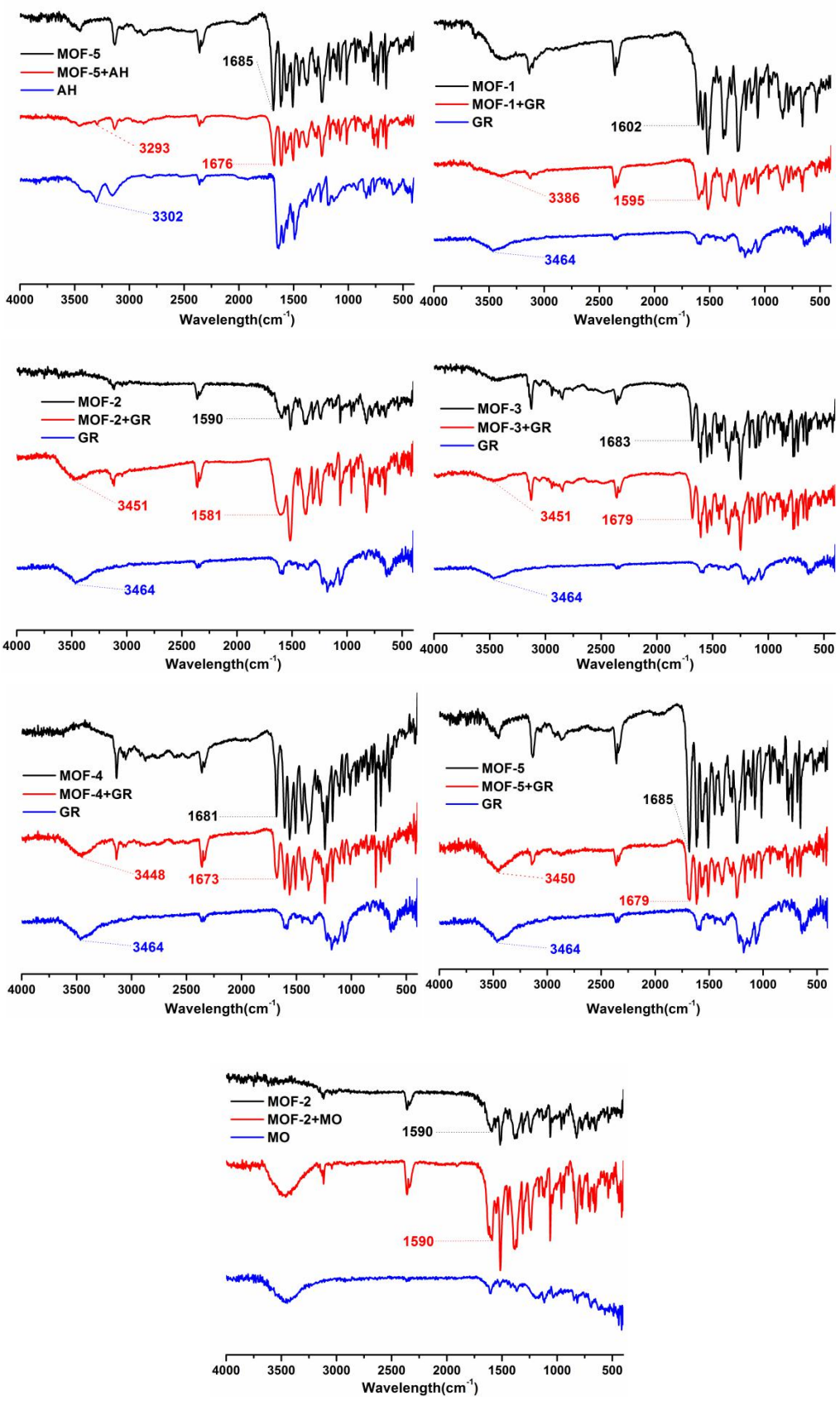


Fig. S10 IR spectra of complexes **1-5** before and after the absorption of dyes.

Fig. S11

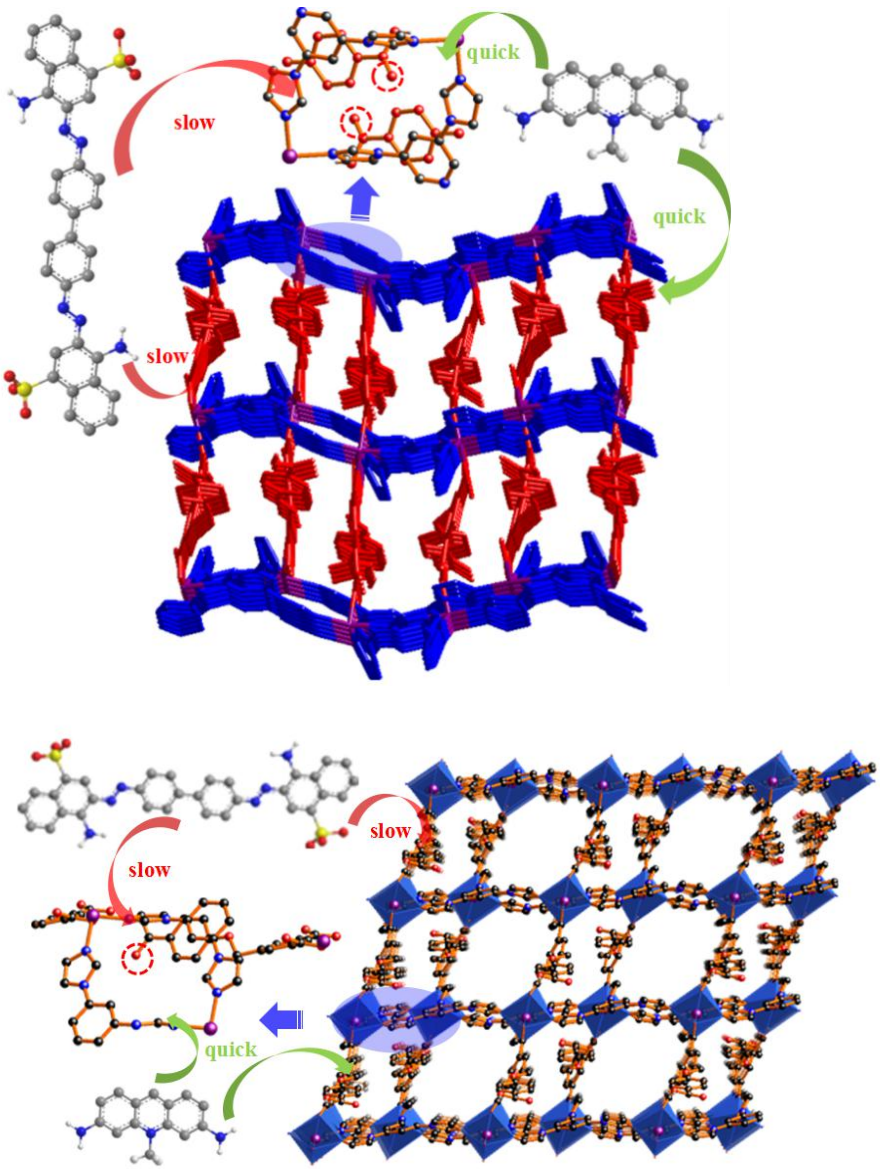


Fig. S11 The different adsorption capacity between GR and AH in MOF-**4** and MOF-**5**.

Fig. S12

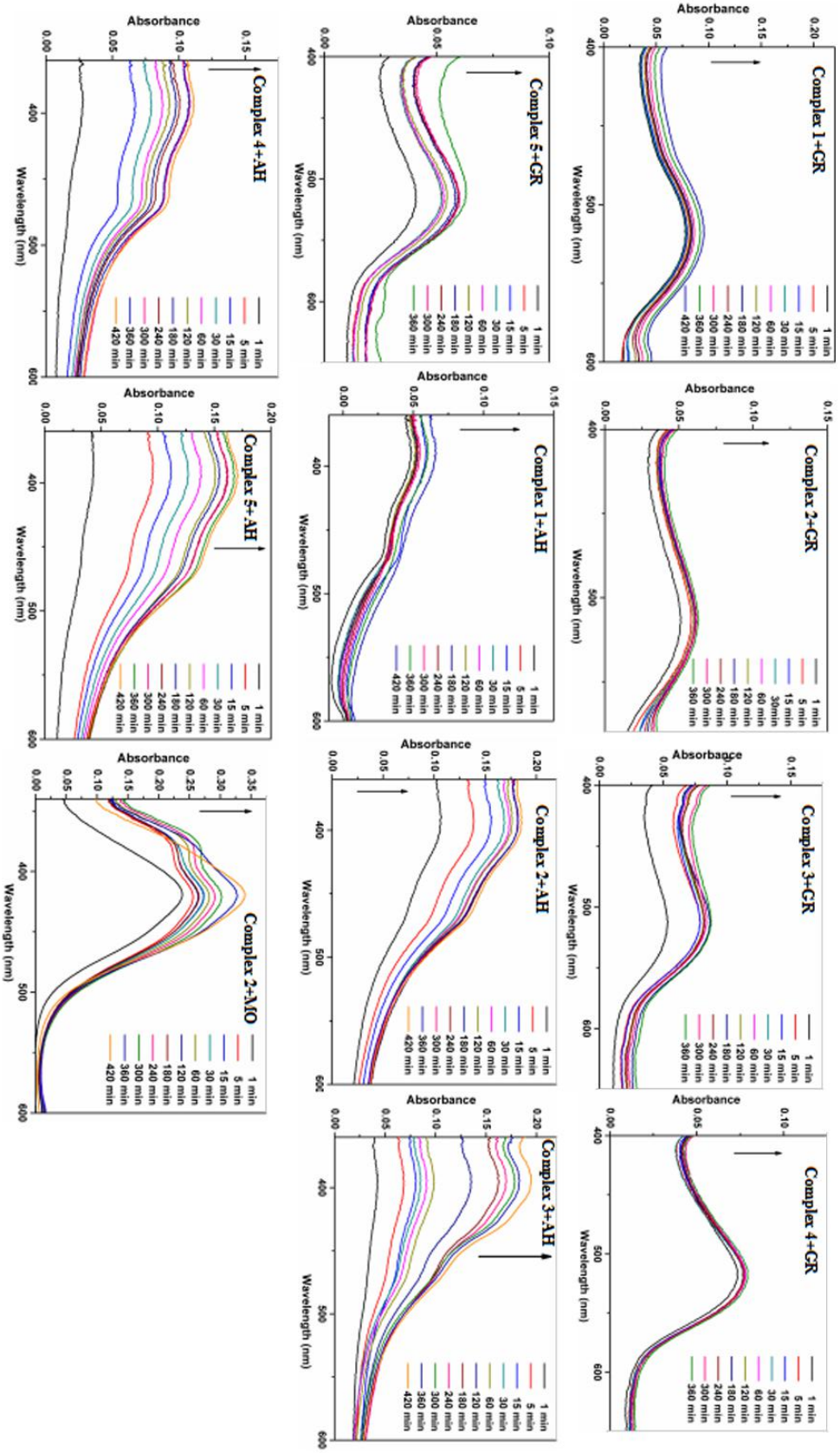


Fig. S12 UV-vis spectra for the adsorption-release process of GR, AH and MO from complexes **1**-**5**, respectively.

Fig. S13

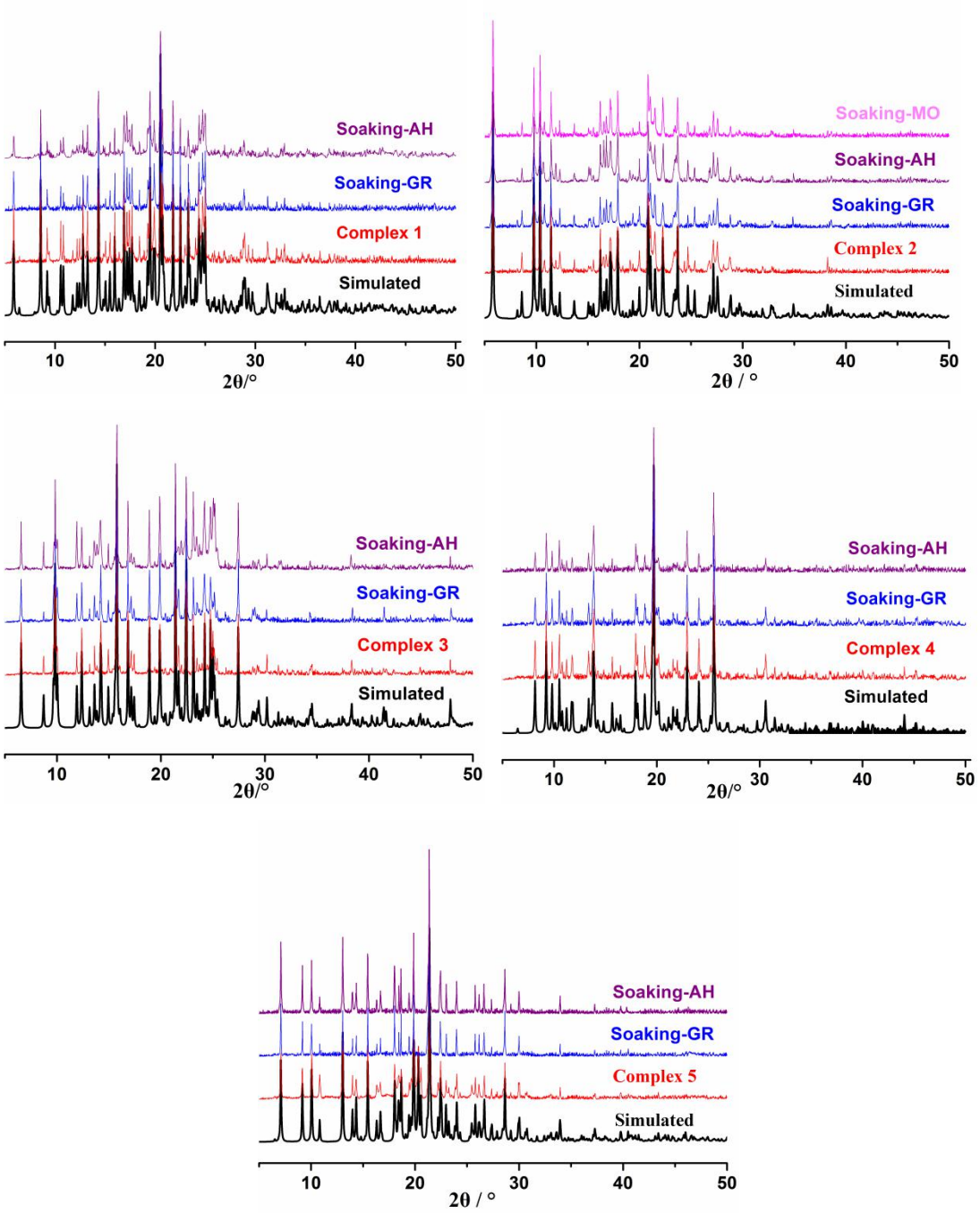


Fig. S13 Comparison of PXRD for complexes **1-5** after dye adsorption.