Electronic Supplementary Information

Mixed-Metal Cu-BTC MOFs as a Strong Adsorbent for Molecular Hydrogen at Low Temperature

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Scanning Electron Microscopy Imaging (SEM): The SEM of microcrystalline sample were conducted by dispersing the sample material into a double-sided adhesive conductive carbon tape that was attached to a flat aluminum sample holder**.** The sample were imaged with Quattro ESEM. SEM image indicating the specific region in which EDS was collected to quantify the ratio of metals present.

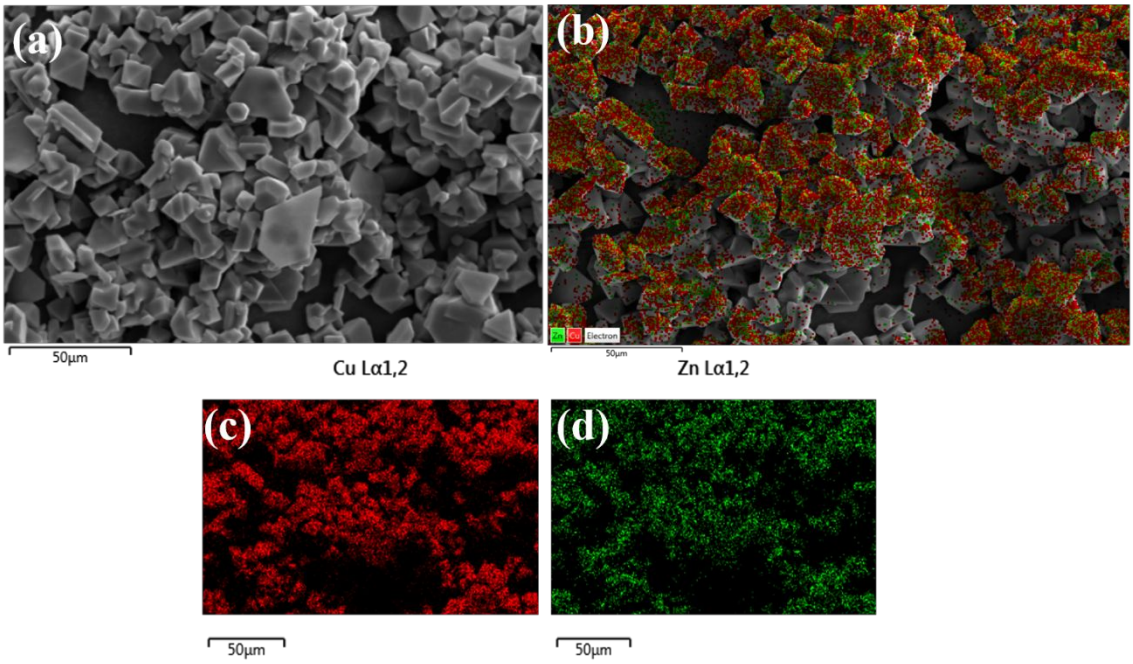

Figure S1. (a) SEM image of Zn-Cu-BTC. (b) EDS mapping of Zn-Cu-BTC with overlay of Cu(red) and Zn(green) as maps shown in (c and d).

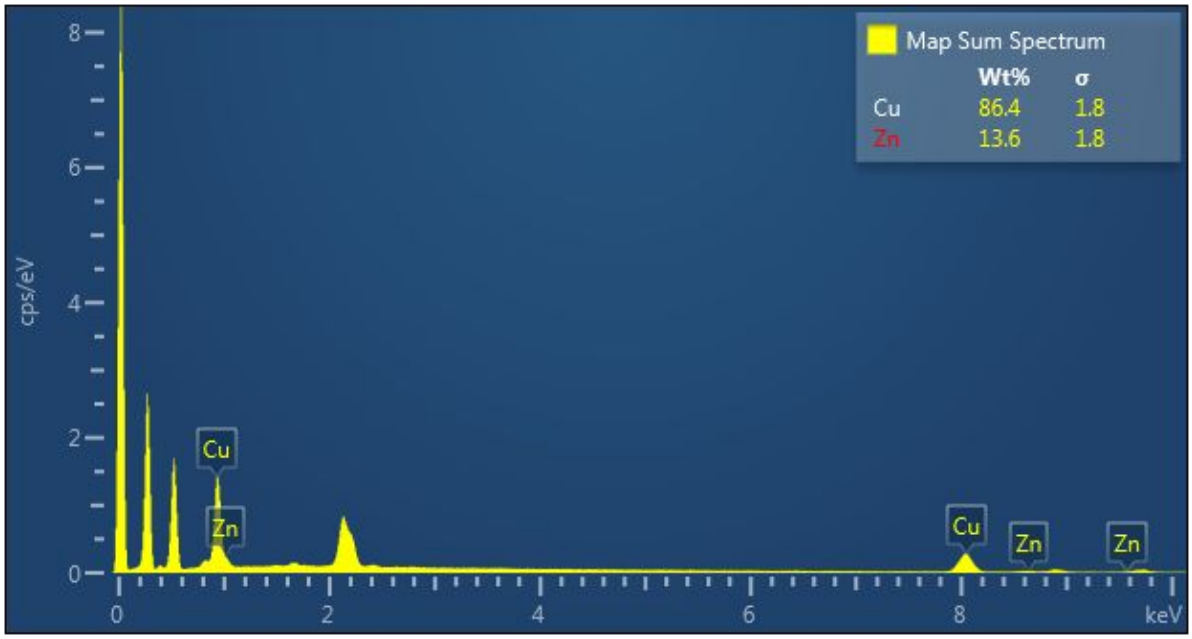

Figure S2. The EDX shows the ratio of metal found in the regions (indicated in EDS mapping figure S1) of Zn-Cu-BTC sample.

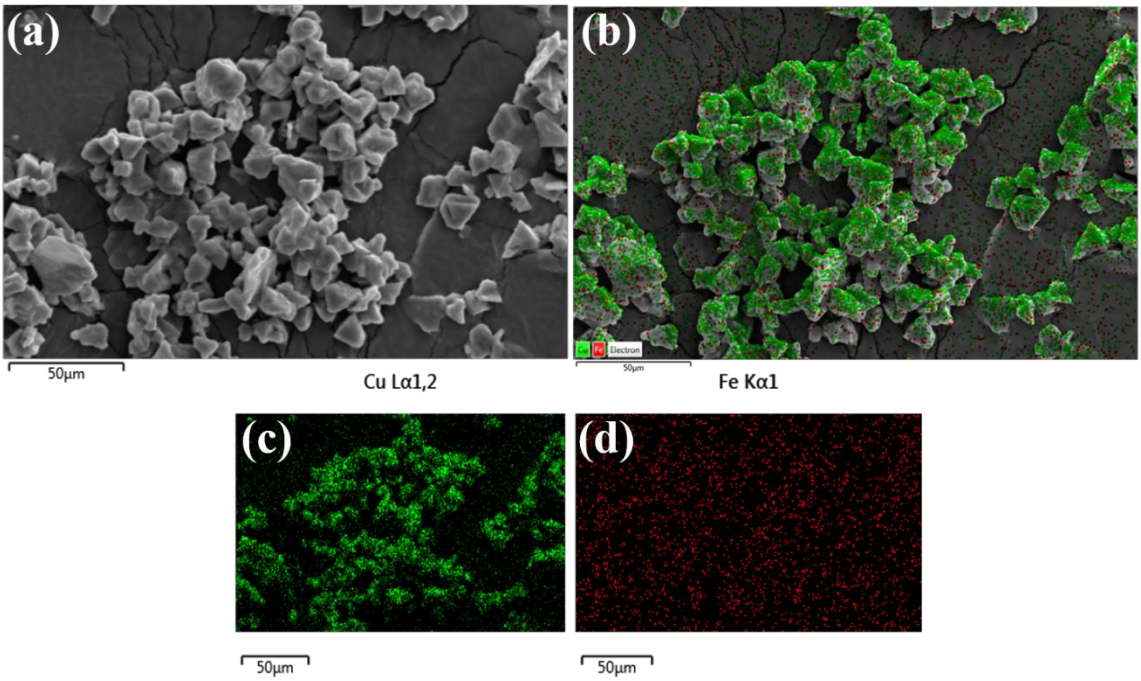

Figure S3. **.** (a) SEM image of Fe-Cu-BTC. (b) EDS mapping of Fe-Cu-BTC with overlay of Cu(red) and Fe(green) as maps shown in (c and d).

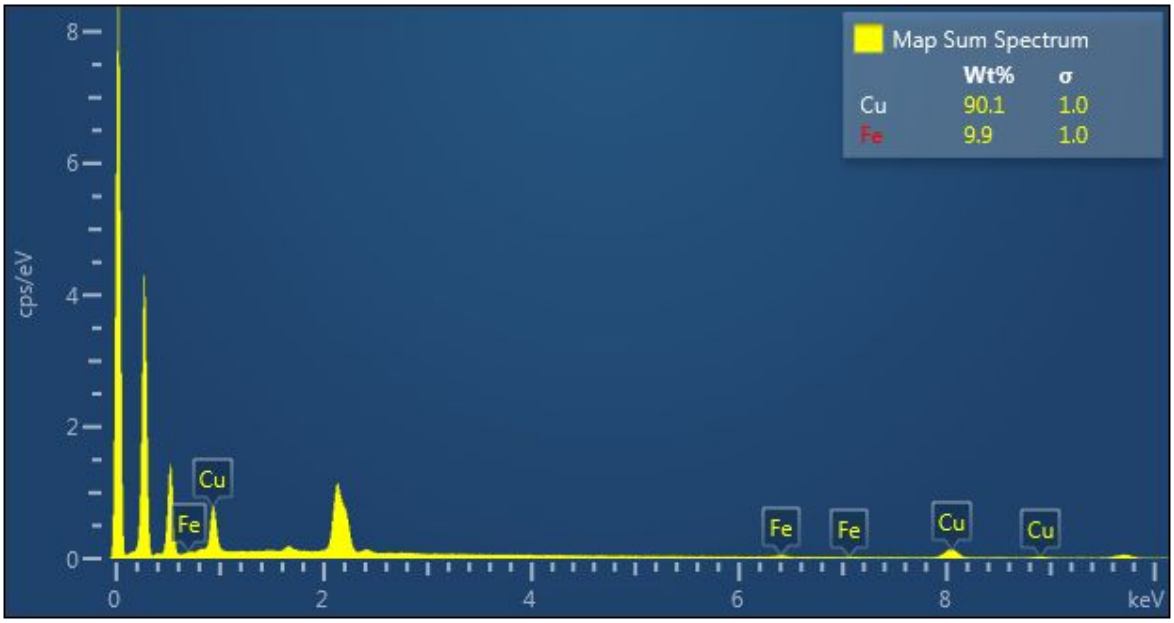

Figure S4. The EDX shows the ratio of metal found in the regions (indicated in EDS mapping figure S3) of Fe-Cu-BTC sample.

Thermogravimetric analysis (TGA):

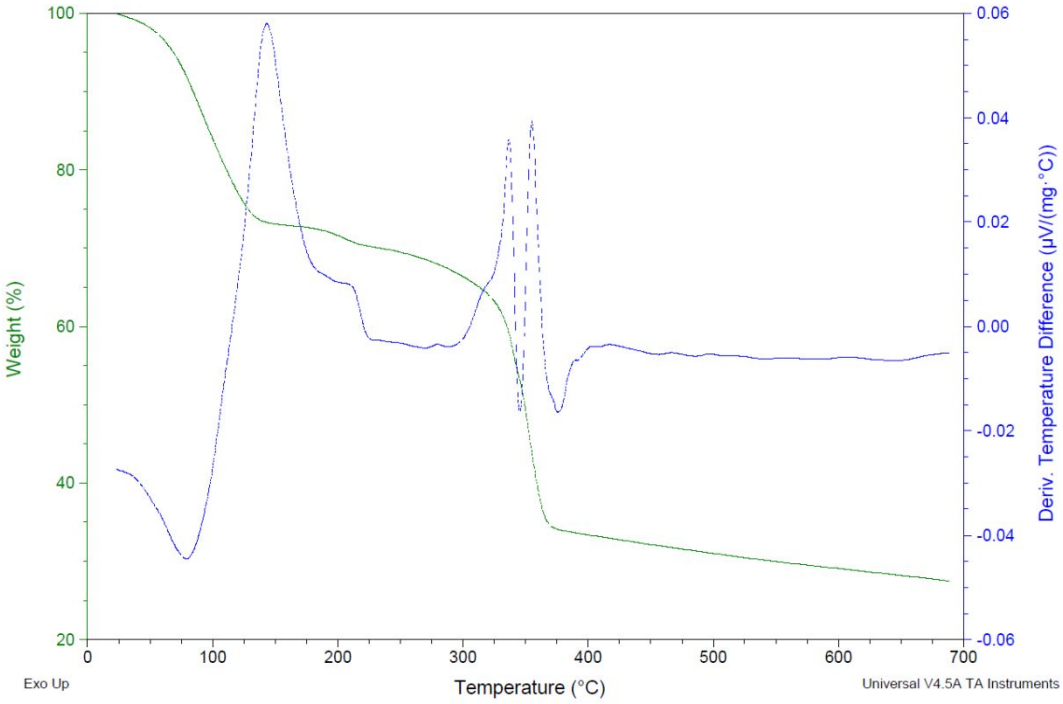

Figure S5. TGA analysis of Cu-BTC (as synthesized).

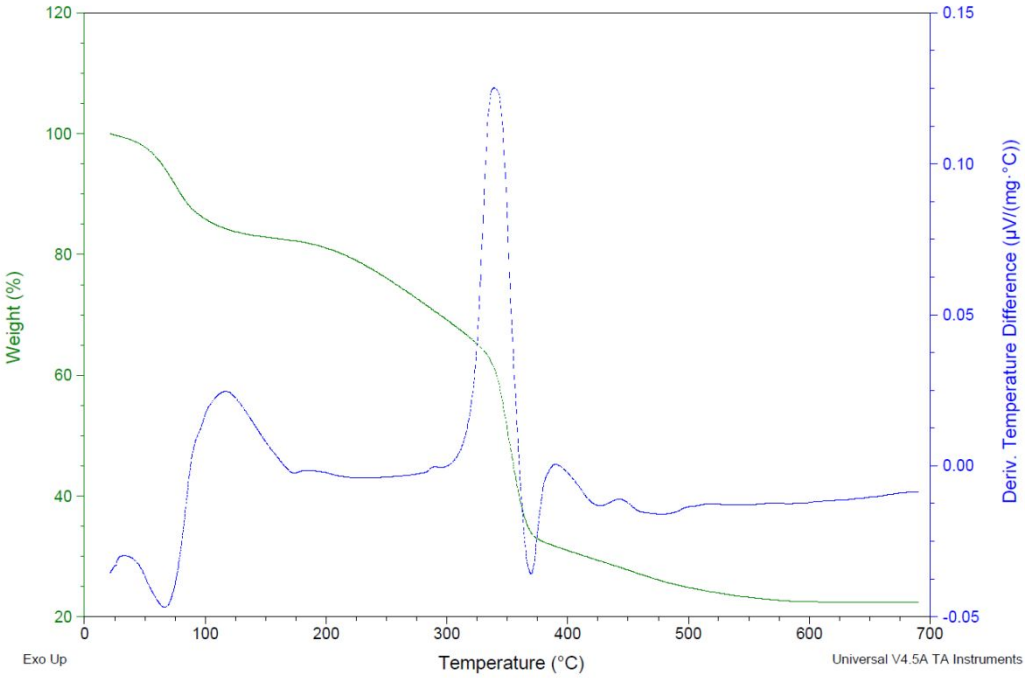

Figure S6. TGA analysis of Co-Cu-BTC (DCM exchanged).

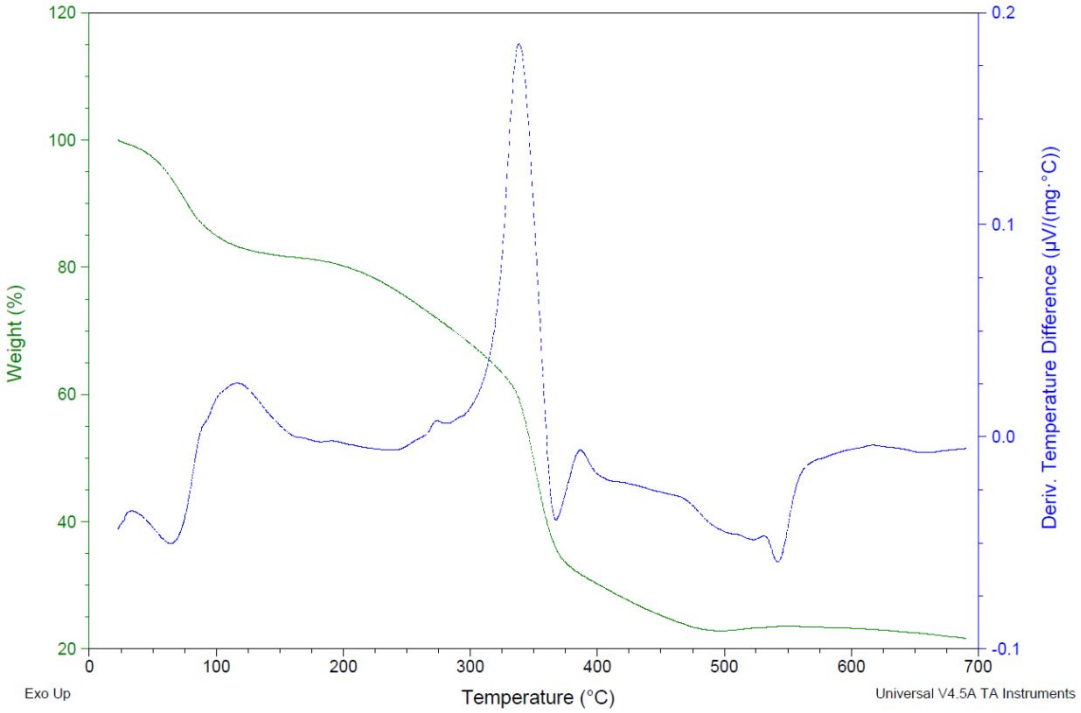

Figure S7. TGA analysis of Fe-Cu-BTC (DCM exchanged).

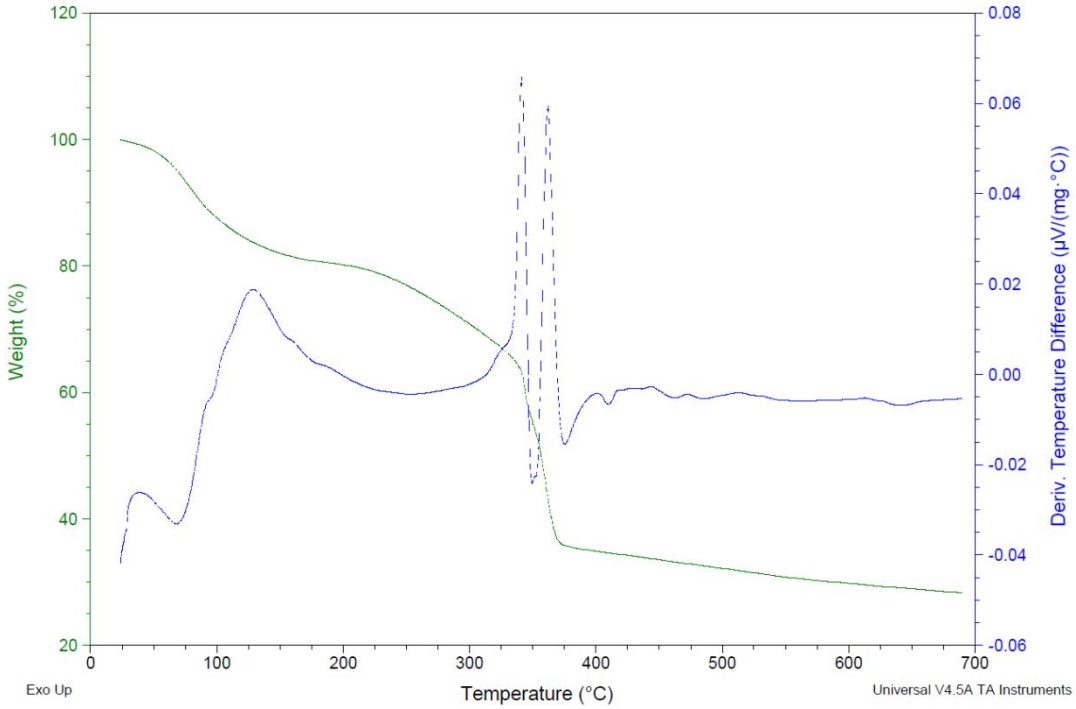

Figure S8. TGA analysis of Ni-Cu-BTC (DCM exchanged).

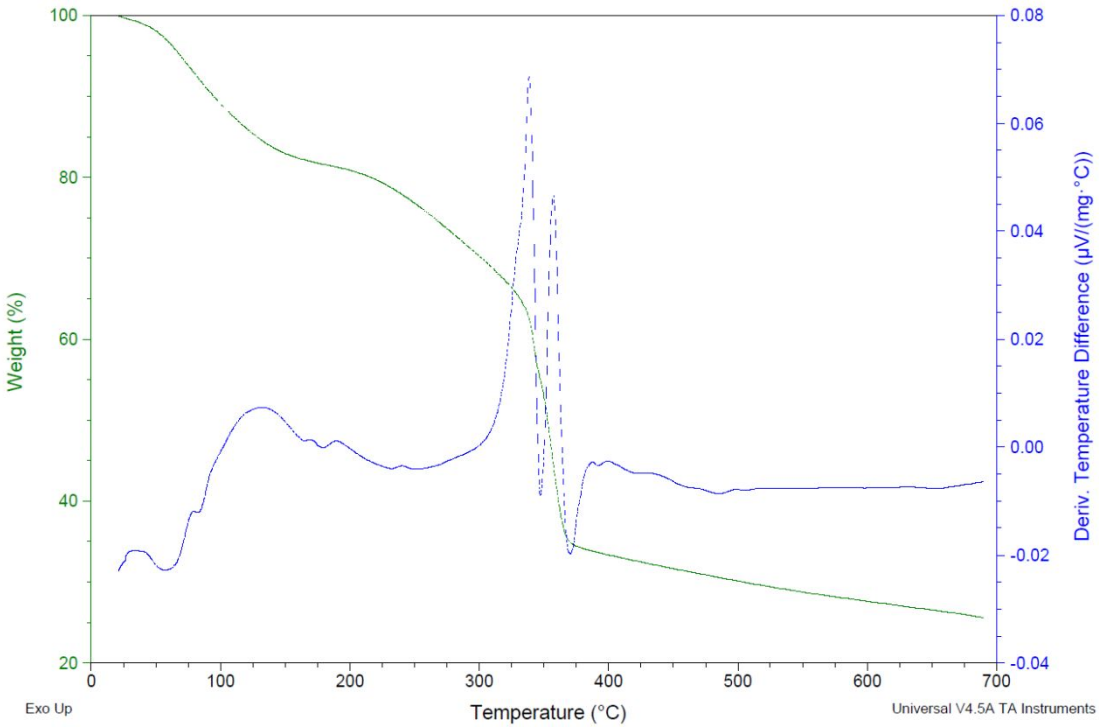

Figure S9. TGA analysis of Zn-Cu-BTC (DCM exchanged).

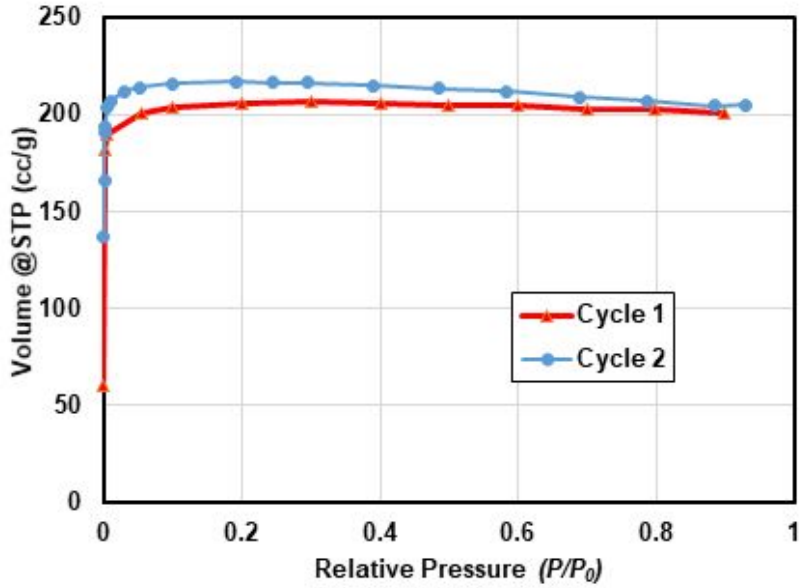

Figure S10. BET measurement of Ni-Cu-BTC after cyclic adsorption/desorption of Hydrogen. (Note: Area in cycle 1= 828 m^2/g and in cycle 2= 895 m^2/g).

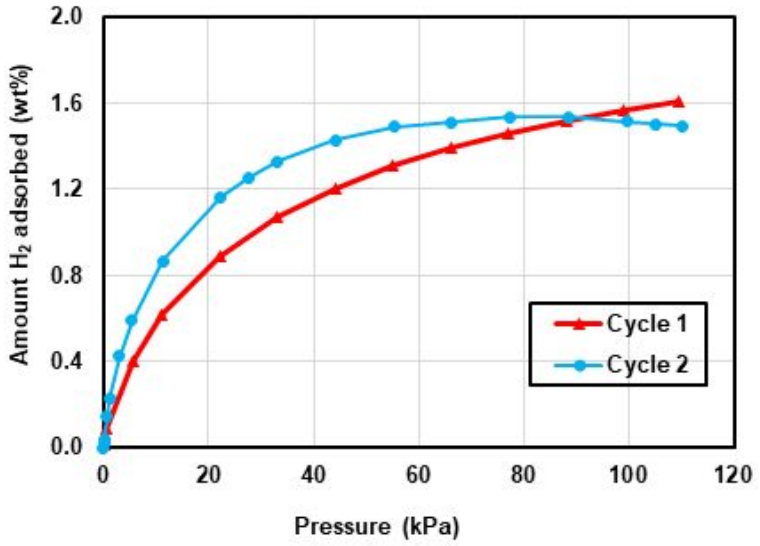

Figure S11. Cyclic adsorption/desorption of Hydrogen on Ni-Cu-BTC.
(Note: Hydrogen adsorption at 110 kPa in cycle 1 = 1.61 wt% and in cycle 2 = 1.49 wt%).

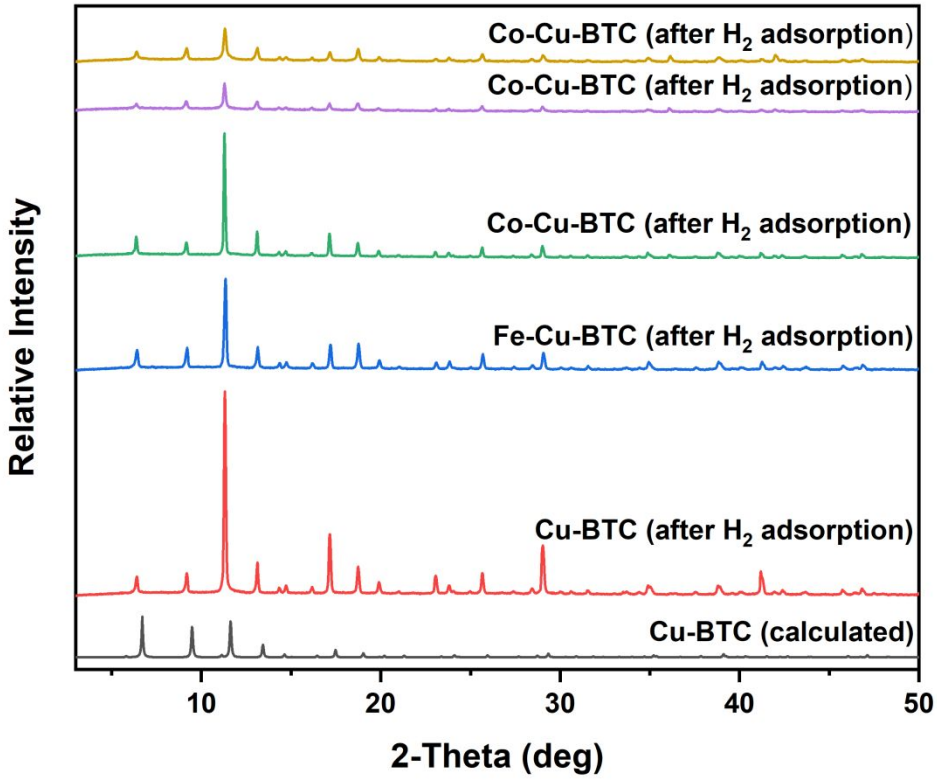

Figure S12. PXRD pattern of Cu-BTC (calculated), Cu-BTC (after H_2 adsorption) and M-Cu-BTC (after H_2 adsorption).

Table S1: H_2 sorption data for Cu-BTC using Quantachrome Autosorb iQ-C-MP at 77 K.

Pressure (kPa)	Adsorbed Volume (cc/g) @STP	Wt.%
0.0651	0.9306	0.008309
0.1148	1.7039	0.015213
0.5695	7.1859	0.06416
1.1163	11.9405	0.106612
5.5826	34.9801	0.312322
11.0028	51.3645	0.458612
21.9892	72.1523	0.644217
33.0505	85.2877	0.761497
44.1071	94.1696	0.8408
54.923	100.5032	0.89735
65.9358	105.0757	0.938176
76.9028	108.4406	0.96822
87.8822	110.9808	0.9909
98.7918	112.807	1.007205
109.2568	113.7541	1.015662

Table S2: H_2 sorption data for Ni-Cu-BTC using Quantachrome Autosorb iQ-C-MP at 77 K.

Pressure (kPa)	Adsorbed Volume (cc/g) @STP	Wt.%
0.0632	1.3065	0.011665
0.5545	9.5752	0.085493
5.5207	45.2176	0.403729
11.1841	69.049	0.616509
22.2171	99.7042	0.890216
33.1015	119.8654	1.070227
44.1538	134.9067	1.204524
55.0118	146.3398	1.306605
66.0284	155.6773	1.389976
77.1278	163.3796	1.458746
88.0814	169.8753	1.516744
98.9856	175.4107	1.566167
109.4226	180.0945	1.607987

Table S3: H_2 sorption data for Zn-Cu-BTC using Quantachrome Autosorb iQ-C-MP at 77 K.

Pressure (kPa)	Adsorbed Volume (cc/g) @STP	Wt.%
0.0022	0.0358	0.00032
5.5345	46.7733	0.417619
10.907	69.9746	0.624773
22.4914	102.5602	0.915716
33.4391	122.7899	1.096338
44.6613	137.9963	1.23211
55.297	149.1839	1.331999
66.4173	158.5332	1.415475
77.6712	166.2144	1.484057
88.8854	172.7061	1.542019
100.0461	178.4765	1.59354
110.8205	182.9953	1.633887

Table S4: H_2 sorption data for Fe-Cu-BTC using Quantachrome Autosorb iQ-C-MP at 77 K.

Pressure (kPa)	Adsorbed Volume (cc/g) @STP	Wt.%
0.0592	1.0853	0.00969
5.7255	40.8304	0.364557
11.2532	62.1381	0.554804
23.348	93.4122	0.834038
34.7663	113.6735	1.014942
46.4595	129.6269	1.157383
58.2206	142.6756	1.273889
69.1405	152.9108	1.365275
80.7508	162.301	1.449116
92.3022	171.0824	1.527521
104.2679	179.0861	1.598983
114.7966	185.6938	1.65798
110.122	182.7507	1.631703

Table S5: H_2 sorption data for Co-Cu-BTC using Quantachrome Autosorb iQ-C-MP at 77 K.

Pressure (kPa)	Adsorbed Volume (cc/g) @STP	Wt.%
0.0612	1.1746	0.010488
5.2223	38.4574	0.34337
11.0765	59.8173	0.534083
21.9812	84.2673	0.752387
33.3734	99.6875	0.890067
43.8835	108.8136	0.97155
55.0452	115.4892	1.031154
66.31	119.7913	1.069565
77.3366	122.5363	1.094074
88.4572	124.2836	1.109675
99.5937	125.0684	1.116682
109.8914	125.1707	1.117596

Table S6: N_2 adsorption data for Ni-Cu-BTC at 77 K.

Relative Pressure	Amount adsorbed (cc/g) @ STP
3.32E-06	59.9944
1.17E-03	181.3121
5.03E-03	189.9998
5.36E-02	200.8047
1.01E-01	203.2944
1.99E-01	205.345
3.00E-01	206.04
4.00E-01	205.7813
5.00E-01	204.9898
6.00E-01	204.2632
7.00E-01	202.9607
7.99E-01	202.0569
8.99E-01	200.4901

Table S7: N_2 adsorption data for Zn-Cu-BTC at 77 K.

Relative Pressure	Amount adsorbed (cc/g) @ STP
3.14E-06	66.0136
1.22E-03	204.9672
5.13E-03	214.1443
4.67E-02	225.6907
1.00E-01	229.8888
1.98E-01	233.8115
2.98E-01	235.978
3.98E-01	238.053
4.98E-01	238.5771
5.98E-01	239.4727
6.98E-01	240.9111
8.05E-01	242.8489
9.06E-01	242.3217

Table S8: N_2 adsorption data for Co-Cu-BTC at 77 K.

Relative Pressure	Amount adsorbed (cc/g) @ STP
3.15E-06	55.0299
6.08E-04	175.749
5.10E-03	189.0781
5.30E-02	199.63
1.00E-01	202.5339
1.98E-01	205.2087
3.00E-01	206.6305
4.01E-01	207.1988
5.02E-01	206.9261
6.02E-01	207.3909
7.02E-01	207.1147
8.03E-01	206.7822
9.02E-01	205.9641

Table S9: N_2 adsorption data for Cu-BTC at 77 K.

Relative Pressure	Amount adsorbed (cc/g) @ STP
4.99E-04	195.9417
5.13E-03	214.501
4.72E-02	227.5828
9.67E-02	232.1124
2.02E-01	236.9369
3.03E-01	239.9826
3.96E-01	242.1347

5.03E-01	243.9255
6.03E-01	245.8189
7.02E-01	248.8121
7.99E-01	254.378
8.97E-01	260.4328

Table S10: N_2 adsorption data for Fe-Cu-BTC at 77 K.

Relative Pressure	Amount adsorbed (cc/g) @ STP
6.18E-04	170.8769
5.04E-03	185.67
5.15E-02	198.2872
9.88E-02	202.6972
2.03E-01	208.163
3.05E-01	211.7958
4.05E-01	213.9368
5.06E-01	216.6195
6.06E-01	219.0389
7.06E-01	222.0785
8.07E-01	223.5877
9.07E-01	225.734

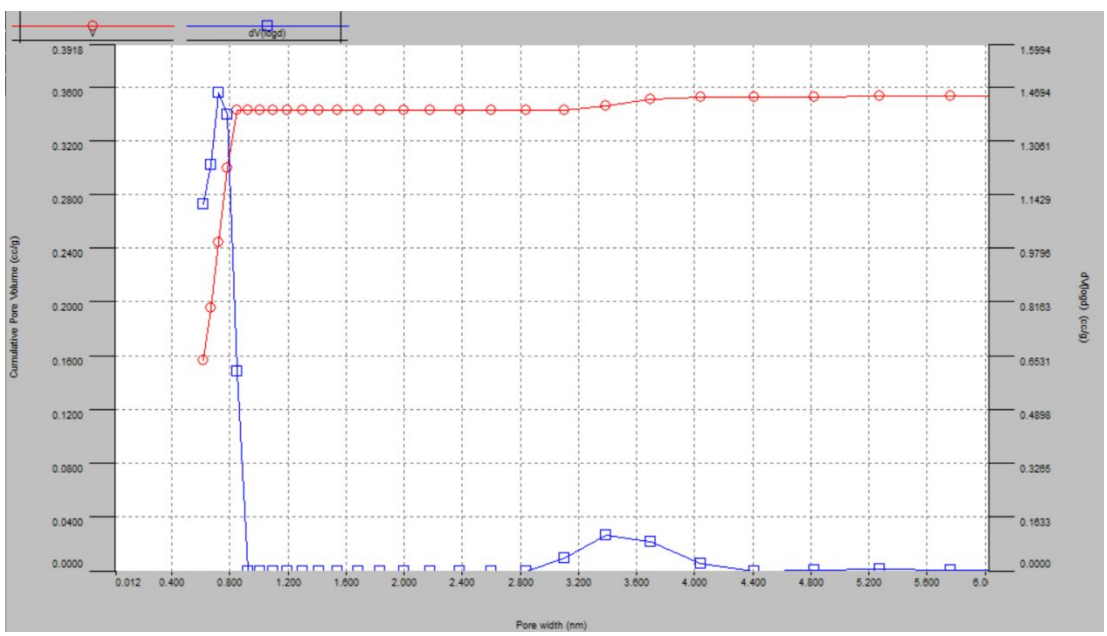

Figure S12. Pore size distribution of Zn-Cu-BTC

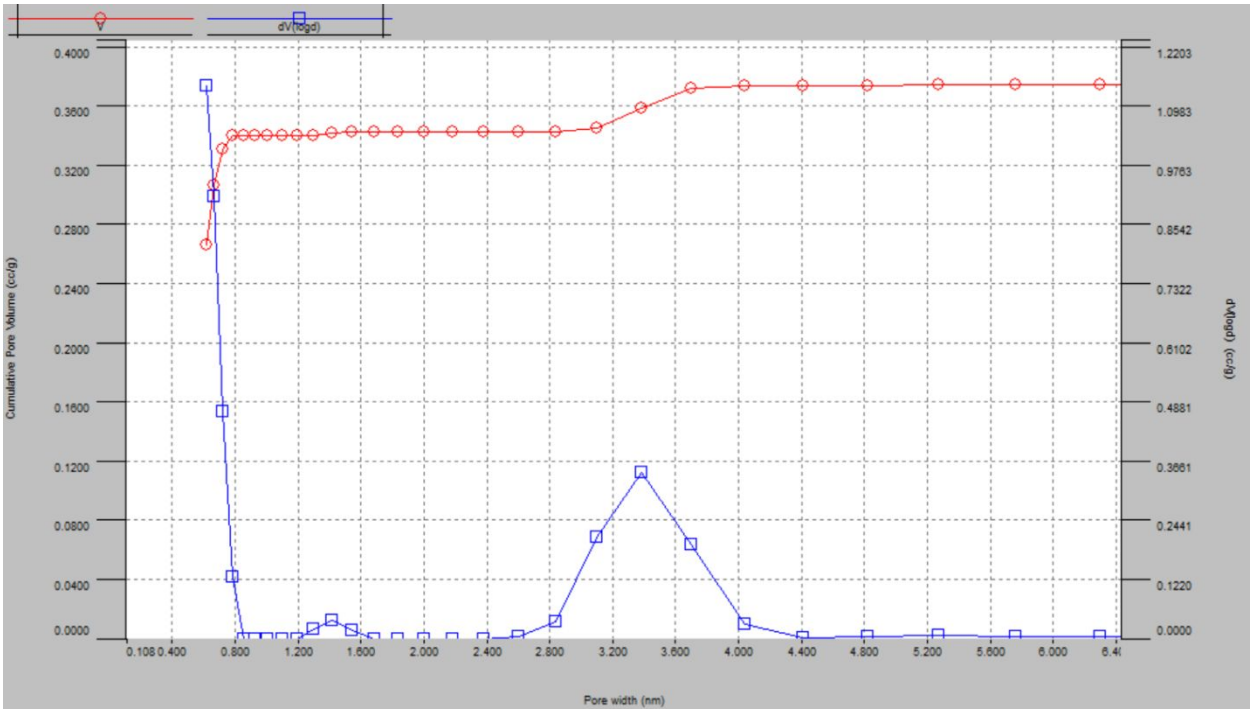

Figure S13. Pore size distribution of Cu-BTC

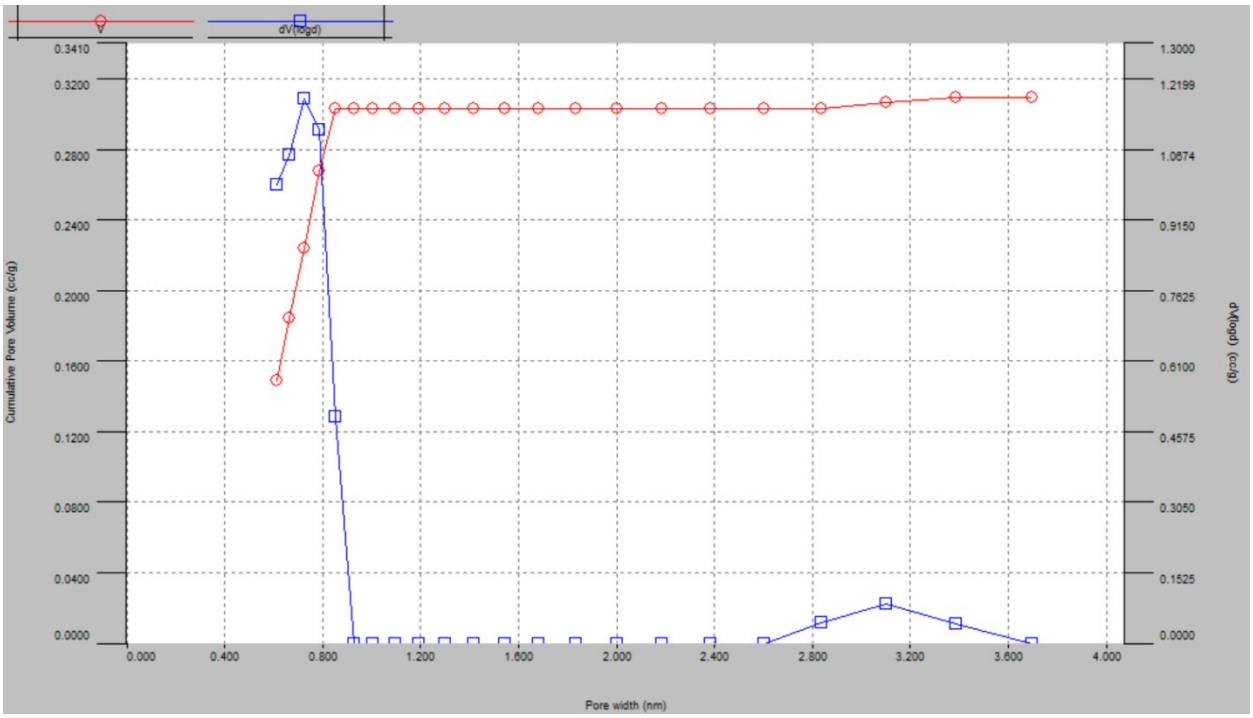

Figure S14. Pore size distribution of Ni-Cu-BTC

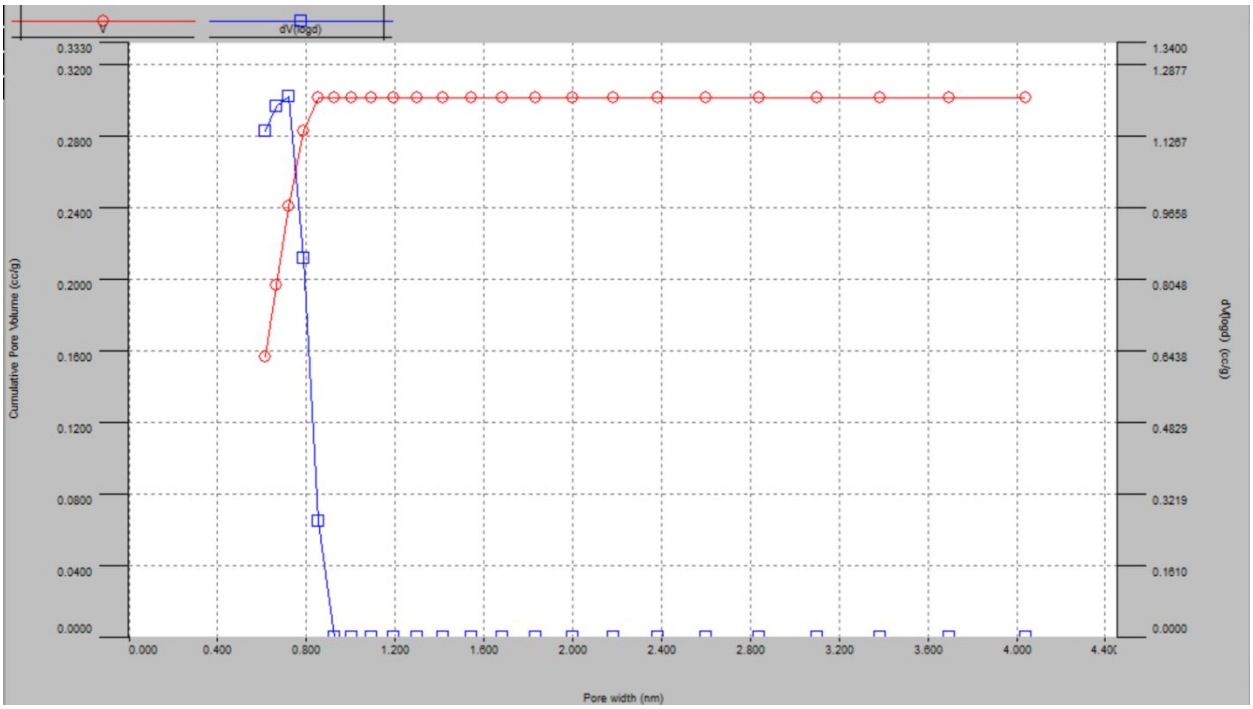

Figure S15. Pore size distribution of Co-Cu-BTC

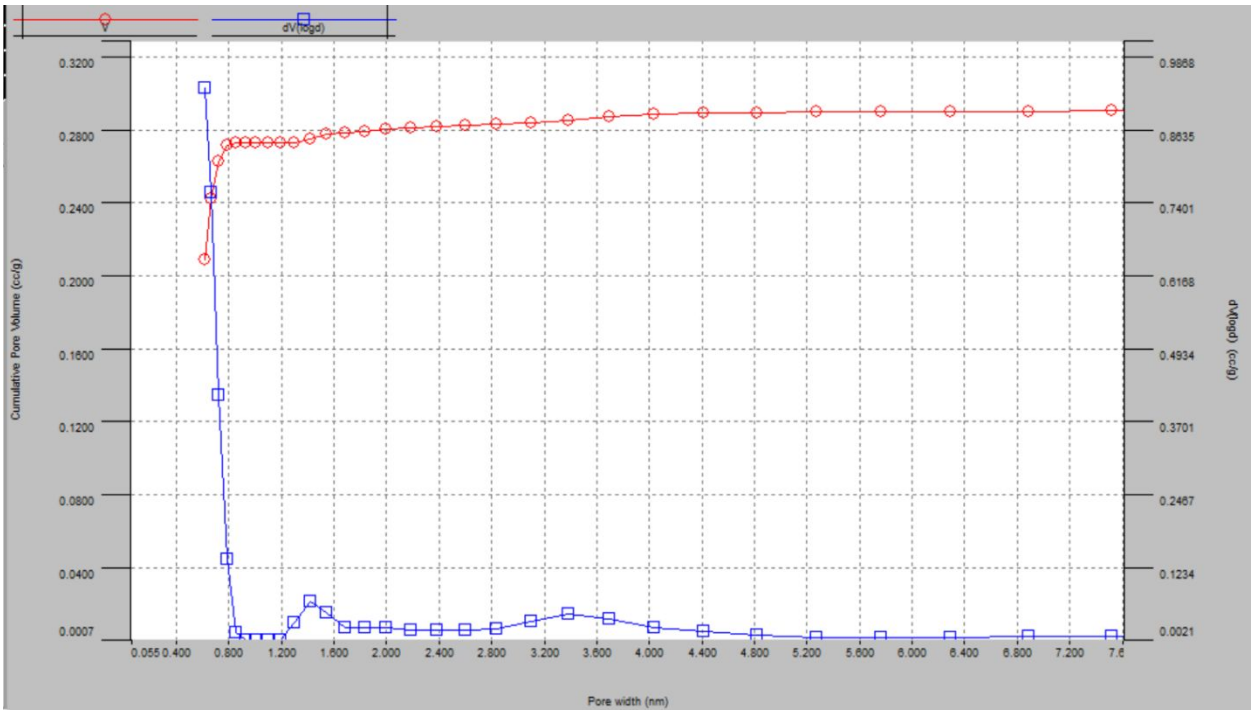

Figure S16. Pore size distribution of Fe-Cu-BTC