## Supplementary Material

## Synergetic Interplay of Nitrogen and Sulfur-rich Copper bi-linker 2D cubic layered MOF Composite with MXene for Improved hybrid Supercapacitor Application

## FTIR and Thermal Analysis

Fourier transform infrared spectrum was measured using ATR in the range of 400-4000 cm<sup>-1</sup>. The FTIR spectrum of Cu-SIP-MOF and 5-sulfoisophhalic acid monosodium salt is presented in Fig. S1 and S. Cu-SIP-MOF frequencies was observed at 3229 cm<sup>-1</sup>, 1653 cm<sup>-1</sup>, 1604 cm<sup>-1</sup>, 1549 cm<sup>-1</sup>, 1437 cm<sup>-1</sup>, 1364 cm<sup>-1</sup>, 1244 cm<sup>-1</sup>, 668 cm<sup>-1</sup>, 492 cm<sup>-1</sup>. The shifting of symmetric and asymmetric vibrations of 5-sulfoisophhalic acid monosodium salt from 1702 cm<sup>-1</sup>, 1618 cm<sup>-1</sup>, 1447 cm<sup>-1</sup>, 1499 cm<sup>-1</sup> to 1653 cm<sup>-1</sup>, 1604 cm<sup>-1</sup>, 1549 cm<sup>-1</sup>, 1437 cm<sup>-1</sup> indicates binding of metal and ligand. Moreover, broad band at 3229 cm<sup>-1</sup> indicates presence of water molecules.

TGA of **Cu-SIP-MOF** (Fig.S3) demonstrates that at first stage nine non-coordinated water molecules were released corresponding to weight loss of 10.3 % (calculated 5.5 %). Experimental and theoretical difference in first weight loss may be attributed to removal of SO<sub>2</sub> along with water molecules. After release of water molecules, MOF remains stable up to 260 °C. Then successive weight loss between 260°C to 450°C corresponds to the removal of organic ligands such as 5-Sulfoisophthalic acid (SIP) and 4,4-bipyridine. The baseline was stable after 450°C, indicating the stability of residual product which may be attributed to 5 moles of CuO 13.8 % (calculated 13.6 %).



Fig. S1. FTIR spectrum of Cu-SIP-MOF.

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Fig. S2. FTIR spectrum of 5-sulfoisophthalic acid (SIP).



Fig. S3. TGA of Cu-SIP-MOF.



Fig. S4. Scanning electron micrograph of Cu-SIP-MOF

### Single crystal XRD of Cu-SIP-MOF:

An appropriate Cu-SIP-MOF crystal was chosen, and data collection was carried out at 293 K using D8-QUEST diffractometer fitted with a graphite-monochromatic Mo-Kα radiation source. Direct techniques were used to solve the structure using SHELXS-2013 [1-3] and full-matrix least-squares approaches on F2 were used to improve it using SHELXL-2013 [4]. Aqua ligands' H atoms were found in a distinct map and optimized at desire. Following their location from various maps, the remaining H atoms were handled as riding atoms with O-H distances of 0.82 Å and C-H distances of 0.93 Å. The subsequent protocols were employed in our analysis: gathering of data: Bruker APEX3 [5]; The programs was utilized for molecular graphics: MERCURY programs [6]; GUI for other software related to structure solution: WinGX [7, 8]. Table S1 provides information on data gathering and crystal structure calculations.



Fig. S5. An infinite 3D supramolecular network in Cu-SIP-MOF.



Fig. S6. Simulated XRD diffractogram of Cu-SIP-MOF



Fig.S7. b-value for Cu-SIP-MOF (A), CM-100 (B), CM-300 (C), Specific Capacitance of Cu-SIP-MOF, CM-100, CM-200 & CM-300 at various current densities (D)



**Fig.S8.** (A) Bar graph of diffusive and capacitive contribution for Cu-SIP-MOF (B) Diffusive, capacitive and experimental current Cu-SIP-MOF at 10 mV/s and (C) 50 mV/s (D) Bar graph of diffusive and capacitive contribution for CM-100 (E) Diffusive, capacitive and experimental current CM-100 at 10 mV/s and (F) 50 mV/s. (G) Graph between percentage contribution and scan rates (H) Diffusive and capacitive participation of current at 10 mV/s (I) Graph between diffusive and capacitive participation of current at scan rate (80 mV/s)



Fig.S9. (A)  $R^2$ -value for and CM-200//AC (B) b-value for and CM-200//AC

Empirical formula	$C_{108}H_{114}Cu_3N_{12}O_{60}S_6$
Formula weight	2923.09
Crystal system	Monoclinic
Space group	<i>C2/c</i>
<i>a (</i> Å)	53.485 (6)
<i>b (</i> Å)	11.0654 (12)
<i>c (</i> Å)	22.520 (2)
β (°)	113.926 (6)
$V(Å^3)$	12183 (2)
Z	4
$D_{\rm c}$ (g cm <sup>-3</sup> )	1.594
$\mu$ (mm <sup>-1</sup> )	0.73
θ range (°)	2.2-27.1
Measured refls.	225002
Independent refls.	15367
<i>R</i> <sub>int</sub>	0.077
S	1.08
R1/wR2	0.063/0.146
$\Delta \rho_{max} / \Delta \rho_{min} \ (e {\rm \AA}^{-3})$	0.90/-0.88
CCDC	2311394

 Table S1 Crystal data and structure refinement parameters.

Cu1-N1	2.046(2)	Cu1-N2	2.040(2)	Cu1-N6	2.009(2)
Cu1-N7 <sup>iii</sup>	2.015(2)	Cu1-O1	2.505(2)	Cu1-O8	2.687)2)
Cu2-N3	2.041(2)	Cu2-N4	2.000(3)	Cu2-N5 <sup>iv</sup>	2.014(3)
Cu2-O15	2.851(2)				

Table S2 Selected bond distances(Å)

Symmetry codes: (iii) x, y-1, z;(iv) x, y+1, z.

D-H····A	D-H	Н…А	D…A	D-H···A
C4—H4…O23 <sup>v</sup>	0.93	2.56	3.191 (5)	125
C10—H10…O22	0.93	2.53	3.154 (5)	125
C12—H12…O15 <sup>ii</sup>	0.93	2.31	3.030 (4)	134
C13—H13…O21 <sup>ii</sup>	0.93	2.60	3.497 (5)	163
C14—H14····O30 <sup>vi</sup>	0.93	2.57	3.220 (5)	128
C21—H21…O15 <sup>vii</sup>	0.93	2.47	3.244 (4)	140
C23—H23…O17 <sup>viii</sup>	0.93	2.53	3.307 (4)	141
C27—H27…O8 <sup>iv</sup>	0.93	2.50	3.243 (4)	137
C28—H28…O1 <sup>iv</sup>	0.93	2.51	3.220 (4)	133
O4—H4A…O26	0.82	1.77	2.572 (4)	164

Table S3 Hydrogen bond parameters (Å, °)

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O6—H6A…O22	0.82	1.76	2.574 (5)	169
O11—H11A…O28	0.82	1.79	2.598 (4)	171
013—H13A…O23 <sup>ix</sup>	0.82	1.78	2.580 (4)	163
O18—H18…O24 <sup>iii</sup>	0.82	1.78	2.576 (4)	164
O21—H21A…O30 <sup>x</sup>	0.82	1.82	2.639 (5)	172
O22—H22A…O16	0.81 (2)	2.36 (5)	3.139 (8)	162
O23—H23B⋯O2 <sup>v</sup>	0.82 (2)	2.20 (9)	2.728 (4)	122
O23—H23B…S1 <sup>v</sup>	0.82 (2)	2.83 (6)	3.583 (4)	153
O24—H24A…O25 <sup>xi</sup>	0.82 (2)	2.01 (2)	2.803 (5)	163
O24—H24B…O7 <sup>xii</sup>	0.82 (2)	2.23 (2)	3.031 (5)	164
O25—H25B…O5	0.84 (2)	1.97 (2)	2.810 (4)	178
O25—H25A⋯O2 <sup>v</sup>	0.83 (2)	2.35 (3)	3.129 (5)	157
O26—H26B····O14 <sup>xiii</sup>	0.81 (2)	2.15 (2)	2.945 (5)	164
O26—H26A…O27 <sup>xiv</sup>	0.83 (2)	2.01 (2)	2.820 (5)	167
O27—H27B…O19 <sup>xv</sup>	0.84 (2)	2.01 (3)	2.823 (4)	161
O27—H27A⋯O9 <sup>iv</sup>	0.84 (2)	2.19 (3)	2.988 (5)	160
O28—H28A…O29	0.83 (2)	2.18 (5)	2.704 (6)	122
O29—H29A…O12 <sup>xvi</sup>	0.85 (2)	2.13 (3)	2.908 (4)	151
O29—H29B…O10 <sup>vi</sup>	0.84 (2)	2.12 (3)	2.908 (4)	157
O30—H30B…O16 <sup>xvii</sup>	0.85 (2)	2.07 (4)	2.830 (6)	147
O30—H30A…O9 <sup>vi</sup>	0.85 (2)	1.95 (4)	2.714 (4)	148

Symmetry codes: (ii) -x+1, y, -z+1/2; (iii) x, y-1, z; (iv) x, y+1, z; (v) -x+3/2, -y+1/2, -z+1; (vi) -x+1, y, -z+3/2; (vii) -x+1, y-1, -z+1/2; (viii) x, -y+1, z+1/2; (ix) x, y, z+1; (x) x, y, z-1; (xi) -x+3/2, y+1/2, -z+1/2; (xii) x, -y+1, z-1/2; (xiii) -x+3/2, y+1/2, -z+3/2; (xiv) -x+3/2, y-1/2, -z+3/2; (xv) x, y+1, z+1; (xvi) -x+1, -y+1, -z+2; (xvii) -x+1, -y, -z+1.

MOF	Electrolyte	Qs (Cg <sup>-1</sup> )	Es (Wh kg-1)/	Ps (W kg <sup>-1</sup> )	Ref
Cu-MOF	6 M KOH	181	22.6	124	[9]
Cu-CAT	3M KCl	134	2.6	200	[10]
Ni-MOF	3 M KOH	161	57.3	160	[11]
Fe-MOF	6 M KOH	112.4	40	799	[12]
Cu-M-CNTs	1M KOH	348.6	27.7	1640	[13]
(MC) HMRL-1/R	1M Na <sub>2</sub> SO <sub>4</sub>	-	57.2	4380	[14]
Cu-MOF/G	6 М КОН	-	34.5	1350	[15]
MOF-5/V <sub>2</sub> CT <sub>x</sub>	ЗМ КОН	923	48.75	920	[16]
Cu–BTC/N– MXene	1M KOH	961	65.23	923	[17]
CuCo <sub>2</sub> S <sub>4</sub> / mxene	6М КОН	992.3	66.8	895.1	[18]
CM- 200//AC	1 М КОН	683.69	62	2330.4	This Work

**Table S4:** A comparison of present work with various synthesized MOFs.

#### Eq. S2-10

The equation 2&3 represent the relation between b-value and peak current.

$$i = av^{b} - - - (2)$$
  
 $\log (i) = \log (a) + blog (v) - - - (3)$ 

Dunn's equation (eq.4) can be utilized to investigate kinetic mechanisms to differentiate between capacitive and diffusive processes.

$$i(V) = k_1 v + k_2 v^{1/2} - \cdots (4)$$

From CV profile, specific capacity (Qs) and specific capacitance (Cs) were calculated using equation 5 & 6. For non-symmetric GCD curve due to quasi-reversible faradaic reaction, the ratio of faradaic reaction charge/voltage does not remain constant and changes with time. Hence, equation 7 to 10 were used to determined Cs, Qs, Es and Ps.





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