

Supplementary Information

Archetypical 2D Sheet-Like Cu₂MoS₄ for All-Solid-State Symmetric Pseudocapacitors with Ultra-Steady Performance Efficiency

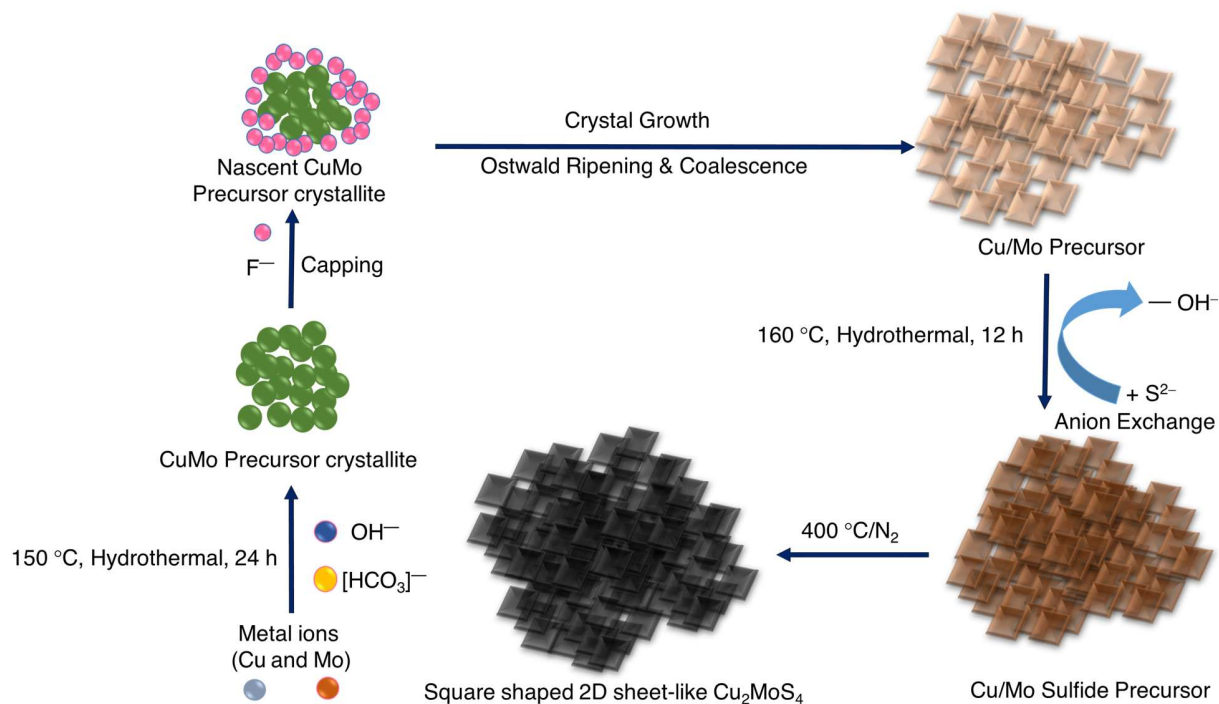
Yogesh Kumar Sonia and Sumanta Kumar Meher*

Materials Electrochemistry & Energy storage Laboratory, Department of Chemistry, Malaviya National Institute of Technology Jaipur, Rajasthan 302017, India

Email*: skmeher.chy@mnit.ac.in

Table S1. Attributions of the peaks in the survey XPS spectrum of Cu₂MoS₄.

Sl. No.	Binding Energy (eV)	Attributions
1	952.30	Cu 2p _{1/2}
2	932.50	Cu 2p _{3/2}
3	565.00	Cu 3p
4	531	O 1s
5	506	Mo 3s
6	411	Mo 3p
7	392	Mo 3p
8	284	S 2s
9	228.6	Mo 3d
10	161	S 2s
11	120	Cu 3s
12	74.25	Cu 3p
13	37.26	Mo 4p



Scheme S1. Plausible growth mechanism and formation of square shaped 2D sheet-like Cu₂MoS₄.^{s1}

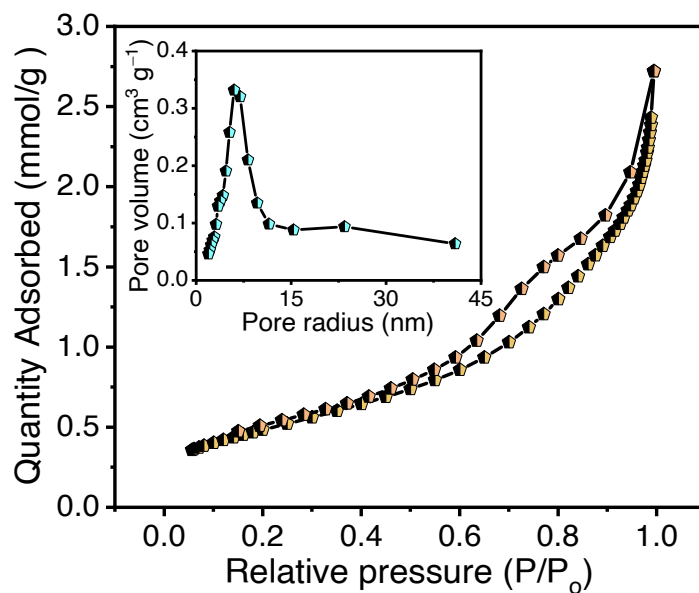


Fig. S1 BET N₂ adsorption-desorption isotherm of Cu₂MoS₄; inset shows the BJH pore size distribution plot of Cu₂MoS₄.

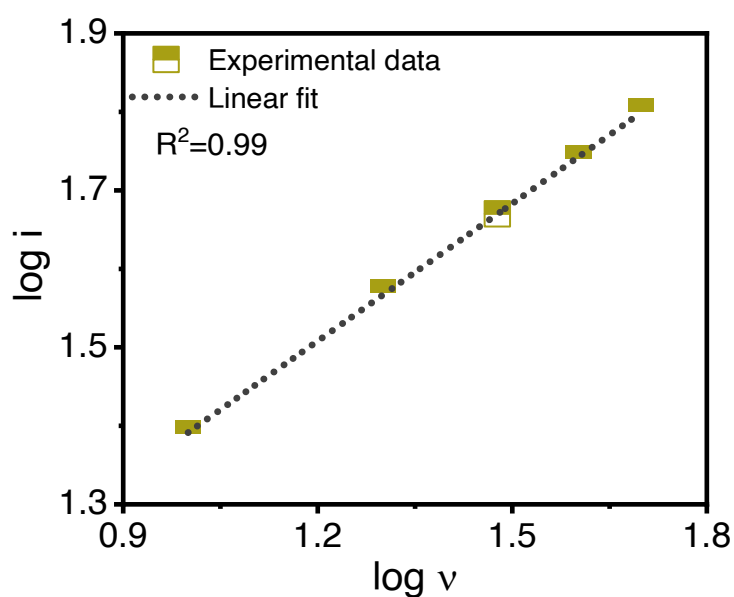


Fig. S2 $\log i$ vs. $\log v$ plot of Cu_2MoS_4 , when studied as the positive electrode material.

Table S2: Comparison of the specific capacitance of Cu_2MoS_4 with the reported Cu and Mo based oxides/sulfides in 3 electrode set up.

Sl. No.	Sample Name	Specific Capacitance @ Current density	References
1	Cu_2O	$144 \text{ F g}^{-1}@1 \text{ A g}^{-1}$	s2
2	RGO/ Cu_2O /Cu	$98 \text{ F g}^{-1}@1 \text{ A g}^{-1}$	s3
3	Cu_2O -GN	$416 \text{ F g}^{-1}@1 \text{ A g}^{-1}$	s4
4	CNT@CuS	$122 \text{ F g}^{-1}@1.2 \text{ A g}^{-1}$	s5
5	H-CuS	$536 \text{ F g}^{-1}@8 \text{ A g}^{-1}$	s6
6	CuS	$237 \text{ F g}^{-1}@0.5 \text{ A g}^{-1}$	s7
7	PPy/ MoO_3	$687 \text{ F g}^{-1}@1 \text{ A g}^{-1}$	s8
8	MoO_3 /C	$331 \text{ F g}^{-1}@1 \text{ A g}^{-1}$	s9

9	MoO ₃	603 F g ⁻¹ @1 A g ⁻¹	s10
10	MoS ₂	366 F g ⁻¹ @0.5 A g ⁻¹	s11
11	MoS ₂	231 F g ⁻¹ @1 A g ⁻¹	s12
12	MoS ₂ -rGO	387 F g ⁻¹ @1.2 A g ⁻¹	s13
13	Cu₂MoS₄	1055 F g⁻¹@1 A g⁻¹	This work

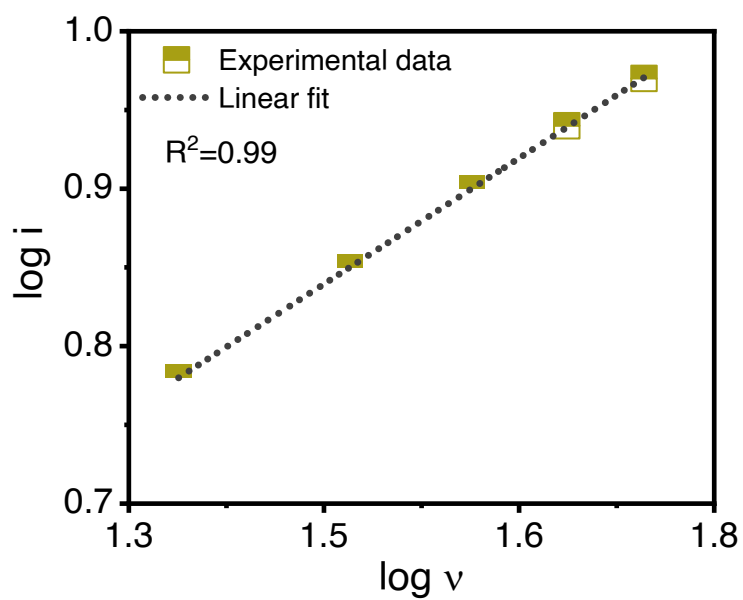


Fig. S3 $\log i$ vs. $\log v$ plot of Cu_2MoS_4 , when studied as the negative electrode material.

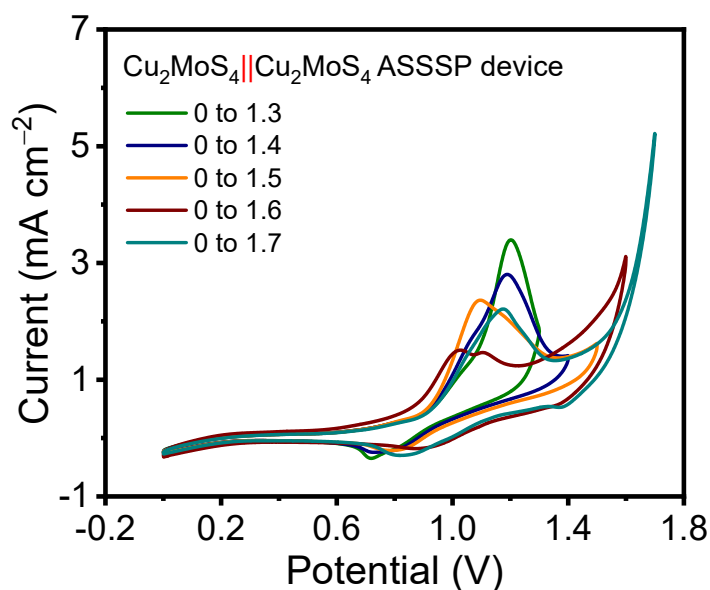


Fig. S4 Current vs. potential plots of $\text{Cu}_2\text{MoS}_4||\text{Cu}_2\text{MoS}_4$ ASSSP device at different potential windows.

Table S3. Comparison of the charge transfer resistance of Cu_2MoS_4 with the reported electrode materials (for symmetric supercapacitors) in 3 electrode set up; comparison of the energy density, power density and cyclic stability of the $\text{Cu}_2\text{MoS}_4 || \text{Cu}_2\text{MoS}_4$ ASSSP device with the reported symmetric supercapacitor devices.

SI. No.	Symmetric device	Charge transfer resistance in 3 electrode set up (R_{ct})	Energy density ($Wh\ kg^{-1}$)	Power density ($W\ kg^{-1}$)	Retention (%) in Cyclic Stability (No. of cycles)	References
1	$\text{Fe}_3\text{O}_4/\text{Graphene} \text{Fe}_3\text{O}_4/\text{Graphene}$	–	19.2	800.2	94% (3200)	s14
2	$\text{MnO}_2 \text{MnO}_2$	1.2 Ω	12.7	87	83% (3000)	s15
3	$\text{N, S co-doped PCFF} \text{N, S co-doped PCFF}$	–	16.3	147	79% (10000)	s16
4	$\text{NiS}_2 \text{NiS}_2$	–	7.97	500	90% (1500)	s17

5	MoS ₂ /CC MoS ₂ /CC	4.3 Ω	5.42	128	96.5% (5000)	s18
6	MnO ₂ MnO ₂	31.42 Ω	23	1923	92% (2200)	s19
7	MoS ₂ /CC MoS ₂ /CC	0.51 Ω	11.1	250	87.9% (1000)	s20
8	NiCo ₂ O ₄ NiCo ₂ O ₄	–	30.50	750	86% (500)	s21
9	CMS/Ni CMS/Ni	0.48 Ω	23.61	1000	87.7% (3000)	s22
10	CuCo ₂ O ₄ CuCo ₂ O ₄	1.89 Ω	16.87	8200	100.94 (3000)	s23
11	FeS FeS	5.15 Ω	2.56	726	91% (1000)	s24
12	f-CFP f-CFP	–	5.2	326	99% (5000)	s25
13	PGBC PGBC	0.51 Ω	6.68	100.2	84% (5000)	s26
14	HCP HCP	–	9.67	-	90.2% (10000)	s27
15	NCF NCF	–	1.35	2900	95.8% (1000)	s28
16	rGO-PMo ₁₂ rGO-PMo ₁₂	–	17.20	130	89% (5000)	s29
17	PANI@CNFs PANI@CNFs	10.5 Ω	10.04	225	81% (1000)	s30
18	HN-CNFs/GNs HN-CNFs/GNs	–	6.3	344.1	99% (5000)	s31
19	CuS/MnS@NF CuS/MnS@NF	–	2.54	174.7	78% (2500)	s32
20	Cu ₂ MoS ₄ Cu ₂ MoS ₄	0.45 Ω	30.53	5649	97.1 (10,500)	This work

Table S4. Comparison of the relaxation time of the $\text{Cu}_2\text{MoS}_4 \parallel \text{Cu}_2\text{MoS}_4$ ASSSPC device with the reported symmetric/asymmetric supercapacitor devices.

Sl. No.	Electrode Material	Symmetric/Asymmetric supercapacitor device	Relaxation time (τ_0)	Reference No.
1	Carbon/Carbon supercapacitors	Carbon \parallel Carbon	4.9 seconds	s33
2	CoFe_2O_4 thin film	$\text{CoFe}_2\text{O}_4 \parallel \text{CoFe}_2\text{O}_4$	174 milliseconds	s34
3	Ti-rich TiO_2 tubular nanolettuces	Ti-rich $\text{TiO}_2 \parallel \text{Ti-rich TiO}_2$	1.7 seconds	s35
4	3D cross-linked graphene	NPFG-0.3 \parallel NPFG-0.3	28.4 milliseconds	s36
5	Manganese oxide	$\text{MnO}_2 \parallel \text{NiCo}_2\text{O}_4$	14 milliseconds	s37
6	$\alpha\text{-Fe}_2\text{O}_3$ thin film	$\alpha\text{-Fe}_2\text{O}_3 \parallel \alpha\text{-Fe}_2\text{O}_3$	9 milliseconds	s38
7	V_2O_5 encapsulated MWCNTs	$\text{V}_2\text{O}_5/\text{MWCNT} \parallel \text{V}_2\text{O}_5/\text{MWCNT}$	5.6 milliseconds	s39
8	MnO_x/Au multilayers	$\text{MnO}_x/\text{Au} \parallel \text{MnO}_x/\text{Au}$	5 milliseconds	s40
9	Silicon nanowires (SiNWs)	SiNWs \parallel SiNWs	3.5 milliseconds	s41
10	Graphene on metal template	Metal graphene \parallel Metal graphene	1.8 milliseconds	s42
11	Polymer-derived carbyne	Carbyne \parallel Carbyne	1.2 milliseconds	s43
12	Graphene	Printable graphene \parallel Printable graphene	1 millisecond	s44
13	Cu_2MoS_4	$\text{Cu}_2\text{MoS}_4 \parallel \text{Cu}_2\text{MoS}_4$	0.5 millisecond	This work

REFERENCES

- s1 M. K. Paliwal, Y. K. Sonia and S. K. Meher, *Mater. Today Chem.* 2021, **22**, 100551.
- s2 L. Chen, Y. Zhang, P. Zhu, F. Zhou, W. Zeng, D. D. Lu, R. Sun and C. Wong, *Sci. Rep.* **2015**, *5*, 1–7.
- s3 X. Dong, K. Wang, C. Zhao, X. Qian, S. Chen, Z. Li, H. Liu and S. Dou, *J. Alloys Compds.* **2014**, *586*, 745–753.
- s4 W. Zhang, Z. Yin, A. Chun, J. Yoo, G. Diao, Y. S. Kim and Y. Piao, *J. Power Sources* **2016**, *318*, 66–75.
- s5 T. Zhu, B. Xia, L. Zhou and X. W. D. Lou, *J. Mater. Chem.* **2012**, *22*, 7851–7855.
- s6 Y. Liu, Z. Zhou, S. Zhang, W. Luo and G. Zhang, *Applied Surface Science*, **2018**, *442*, 711–719.
- s7 J. Zhang, H. Feng, J. Yang, Q. Qin, H. Fan, C. Wei and W. Zheng, *ACS Appl. Mater. Interfaces* **2015**, *7*, 21735–21744.
- s8 X. Wu, Q. Wang, W. Zhang, Y. Wang and W. Chen, *Mater. Lett.* **2016**, *182*, 121–124.
- s9 H. Ji, X. Liu, Z. Liu, B. Yan, L. Chen, Y. Xie, C. Liu, W. Hou and G. Yang, *Adv. Funct. Mater.* **2015**, *25*, 1886–1894.
- s10 N. Zhao, H. Fan, M. Zhang, J. Ma, Z. Du, B. Yan, H. Li and X. Jiang, *Chem. Eng. J.* **2020**, *390*, 124477.
- s11 L. Jiang, S. Zhang, S. A. Kulinich, X. Song, J. Zhu, X. Wang and H. Zeng, *Mater. Res. Lett.* **2015**, *3*, 177–183.
- s12 M. Isacfranklin, L. E. M. Princy, Y. Rathinam, L. Kungumadevi, G. Ravi, A. G. Al-Sehemi and D. Velauthapillai, *Energy Fuels* **2022**, *36*, 6476–6482.
- s13 M. Saraf, K. Natarajan and S. M. Mobin, *ACS Appl. Mater. Interfaces* **2018**, *10*, 16588–16595.
- s14 S. Su, L. Lai, R. Li, Y. Lin, H. Dai and X. Zhu, *ACS Appl. Energy Mater.* 2020, **3**, 9379–9389.

- s15 G. S. Gund, D. P. Dubal, N. R. Chodankar, J. Y. Cho, P. Gomez-Romero, C. Park, C. D. Lokhande, *Sci. Rep.* 2015, **5**, 1–13.
- s16 L. Chen, Z. Wen, L. Chen, W. Wang, Q. Ai, G. Hou, Y. Li, J. Lou, L. Ci, *Carbon* 2020, **158**, 456–464.
- s17 A. M. Patil, A. C. Lokhande, N. R. Chodankar, V. S. Kumbhar and C. D. Lokhande, *Mater. Des.* 2016, **97**, 407–416.
- s18 M. S. Javed, S. Dai, M. Wang, D. Guo, L. Chen, X. Wang, C. Hu and Y. Xi, *J. Power Sources* 2015, **285**, 63–69.
- s19 N. R. Chodankar, D. P. Dubal, G. S. Gund and C. D. Lokhande, *J. Energy Chem.* 2016, **25**, 463–471.
- s20 C. Zhou, J. Wang, X. Yan, X. Yuan, D. Wang, Y. Zhu and X. Cheng, *Ceram. Int.* 2019, **45**, 21534–21543.
- s21 Z. Cao, C. Liu, Y. Huang, Y. Gao, Y. Wang, Z. Li, Y. Yan and M. Zhang, *J. Power Sources* 2020, **449**, 227571.
- s22 S. Sahoo, K. Krishnamoorthy, P. Pazhamalai and S. J. Kim, *Nanoscale* 2018, **10**, 13883–13888.
- s23 L. Liao, H. Zhang, W. Li, X. Huang, Z. Xiao, K. Xu, J. Yang, R. Zou and J. Hu, *J. Alloys Compd.* 2017, **695**, 3503–3510.
- s24 S. S. Karade, P. Dwivedi, S. Majumder, B. Pandit, B. R. Sankapal, *Sust. Energy Fuels* 2017, **1**, 1366–1375.
- s25 P. Suktha, P. Chiochan, P. Iamprasertkun, J. Wutthiprom, N. Phattharasupakun, M. Suksomboon, T. Kaewsongpol, P. Sirisinudomkit, T. Pettong and M. Sawangphruk, *Electrochim. Acta* 2015, **176**, 504–513.
- s26 Y. Gong, D. Li, C. Luo, Q. Fu and C. Pan, *Green Chem.* 2017, **19**, 4132–4140.

- s27 Y. Wang, R. Liu, Y. Tian, Z. Sun, Z. Huang, X. Wu and B. Li, *Chem. Eng. J.* 2020, **384**, 123263.
- s28 K. Xiao, L. X. Ding, G. Liu, H. Chen, S. Wang and H. Wang, *Adv. Mater.* 2016, **28**, 5997–6002.
- s29 D. P. Dubal, J. Suarez-Guevara, D. Tonti, E. Enciso, P. Gomez-Romero, *J. Mater. Chem. A* 2015, **3**, 23483–23492.
- s30 C. Hu, X. Zhang, B. Liu, S. Chen, X. Liu, Y. Liu, J. Liu and J. Chen, *Electrochim. Acta* 2020, **338**, 135846.
- s31 X. Li, X. Chen, Y. Zhao, Y. Deng, J. Zhu, S. Jiang and R. Wang, *Electrochim. Acta* 2020, **332**, 135398.
- s32 M. Zhai, Y. Cheng, Y. Jin and J. Hu, *Int. J. Hydrogen Energy* 2019, **44**, 13456–13465.
- s33 C. Portet, P. L. Taberna, P. Simon, E. Flahaut and C. Laberty-Robert, *Electrochim. Acta* 2005, **50**, 4174–4181.
- s34 J. S. Sagu, K. G. U. Wijayantha and A. A. Tahir, *Electrochim. Acta* 2017, **246**, 870–878.
- s35 M. Qorbani, O. Khajehdehi, A. Sabbah and N. Naseri, *ChemSusChem* 2019, **12**, 4064–4073.
- s36 A. Kumar, C. S. Tan, N. Kumar, P. Singh, Y. Sharma, J. Leu, E. W. Huang, T. Winie, K. H. Wei and T. Y. Tseng, *RSC Adv.* 2021, **11**, 26892–26907.
- s37 S. J. Patil, J. S. Park, Y. B. Kim and D. W. Lee, *Energy Technol.* 2018, **6**, 1380–1391.
- s38 S. N. Khatavkar and S. D. Sartale, *AIP Conference Proceedings* 2021, **2335**, 040008.
- s39 B. Pandit, D. P. Dubal, P. Gómez-Romero, B. B. Kale and B. R. Sankapal, *Sci. Rep.* 2017, **7**, 1–12.
- s40 W. Si, C. Yan, Y. Chen, S. Oswald, L. Han and O. G. Schmidt, *Energy Environ. Sci.* 2013, **6**, 3218–3223.

- s41 D. Aradilla, P. Gentile, G. Bidan, V. Ruiz, P. Gómez-Romero, T. J. Schubert, H. Sahin, E. Frackowiak and S. Sadki, *Nano Energy* 2014, **9**, 273–281.
- s42 Z. K. Wu, Z. Lin, L. Li, B. Song, K. S. Moon, S.L. Bai and C. P. Wong, *Nano Energy* 2014, **10**, 222–228.
- s43 V. K. Mariappan, K. Krishnamoorthy, S. Manoharan, P. Pazhamalai and S. J. Kim, *Small* 2021, **17**, 2102971.
- s44 Z. S. Wu, Z. Liu, K. Parvez, X. Feng and K. Müllen, *Adv. Mater.* 2015, **27**, 3669–3675.