Supplementary Information

Direct Writing 1.8 V All-Solid-State Flexible Asymmetric Microsupercapaitors

Yaling Wang,[a] MengYuan Ran,[a] Ming Zhu[a] and Lei Li *[b]

[a]College of Energy Engineering, Xi'an University of Science and Technology, Xi'an,710054, China
[b]State Key Laboratory for Mechanical Behavior of Materials, Xi'an Jiaotong University, Xi'an Jiaotong University, Xi'an, 710049, China

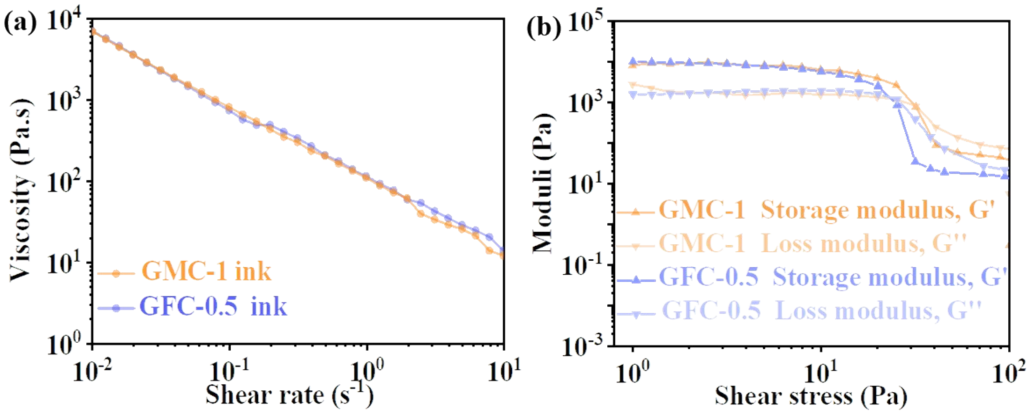

Fig. S1 The rheological properties of the inks.

(a) Apparent viscosities of GMC-1 and GFC-0.5 inks at a shear rate from 0.01 to10 s^{-1}. (b) Storage and loss modulus of GMC-1 and GFC-0.5 inks at a shear stress from 1 to 100 Pa.

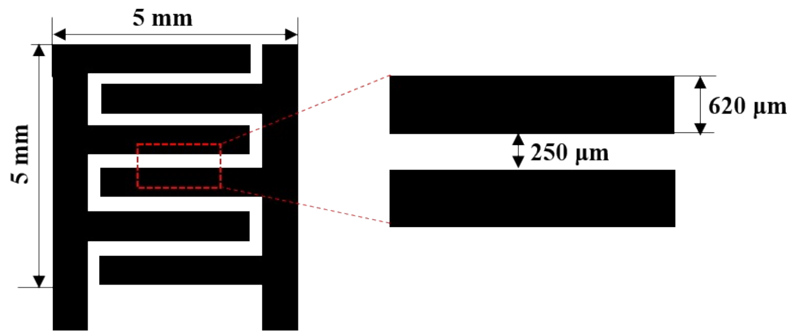

Fig. S2 Dimensions of the printed electrodes for GMC//GFC AMSCs.

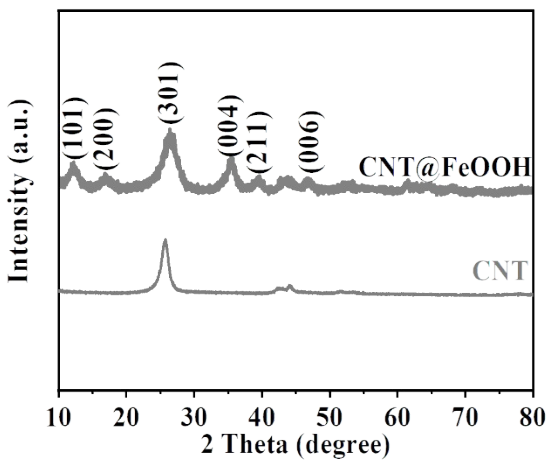

Fig. S3 and Fig. S4 Characterization of electrode components.

Fig. S3 XRD patterns of CNT and CNT@FeOOH.

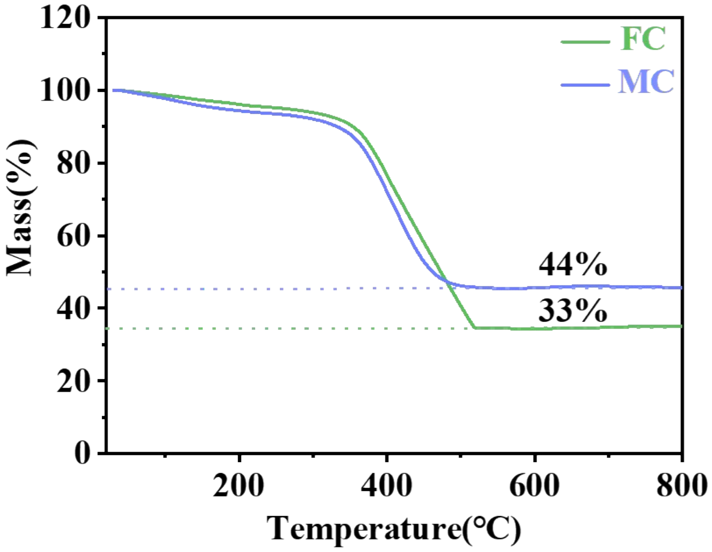

Fig. S4 TGA analysis of MC and FC.

Fig. S5-S9 and Table S1 Electrochemical properties of electrodes and devices.

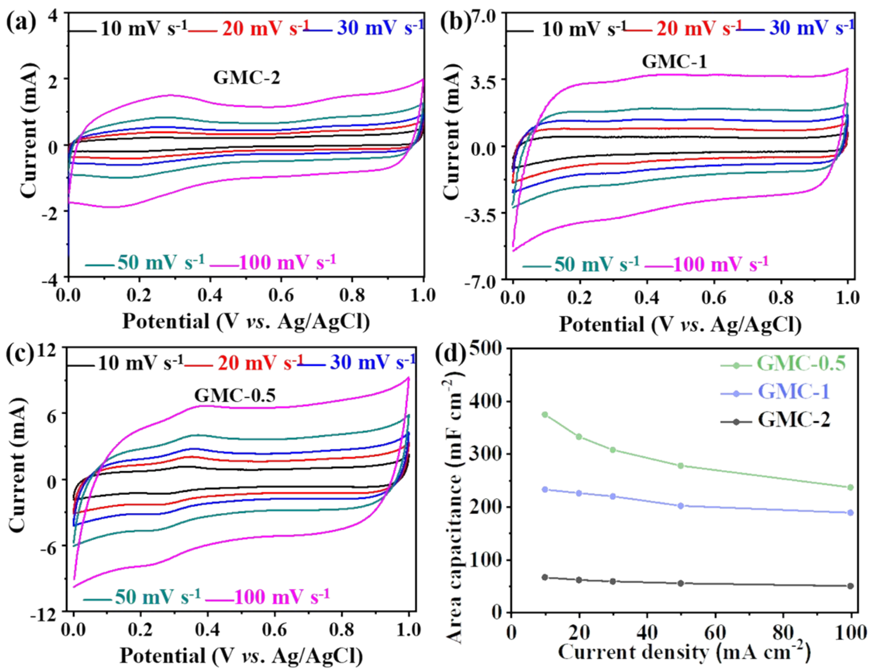

Fig. S5 (a-c) CV curves of GMC-2, GMC-1 and GMC-0.5 electrodes at various scan rates. (d) Specific areal capacitance of GMC-2, GMC-1 and GMC-0.5 electrodes at different scan rates of GMC-X.

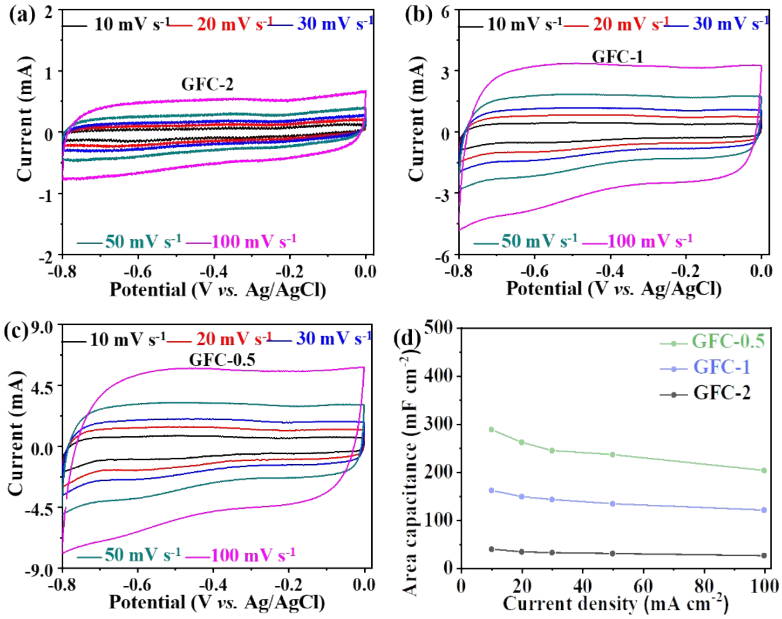

Fig. S6 (a-c) CV curves of GFC-2, GFC-1 and GFC-0.5 at various scan rates. (d) Specific areal capacitance of GFC-2, GFC-1 and GFC-0.5 at different scan rates.

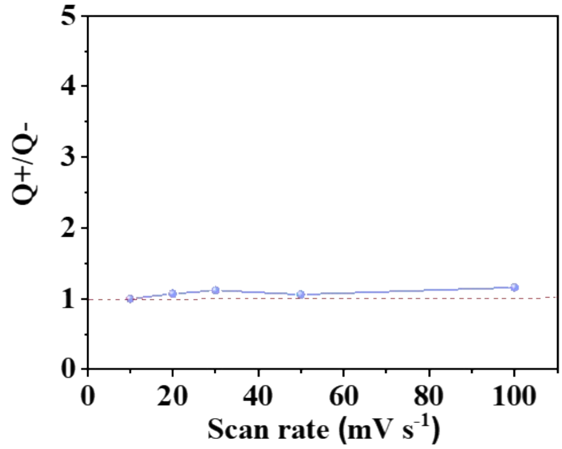

Fig. S7 The Q_{+}/Q_{-} for GMC-1 and GFC-0.5 electrodes at various scan rates.

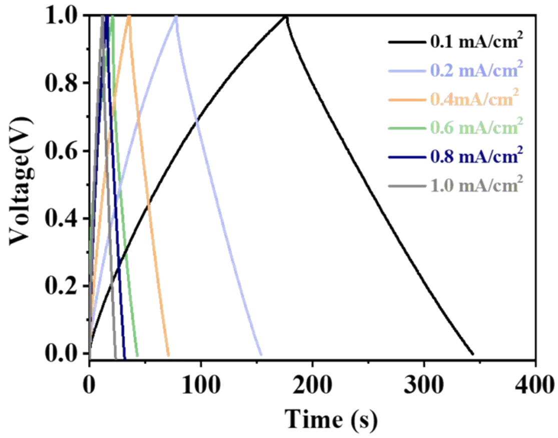

Fig. S8 GCD curves of GMC MSC at different current densities.

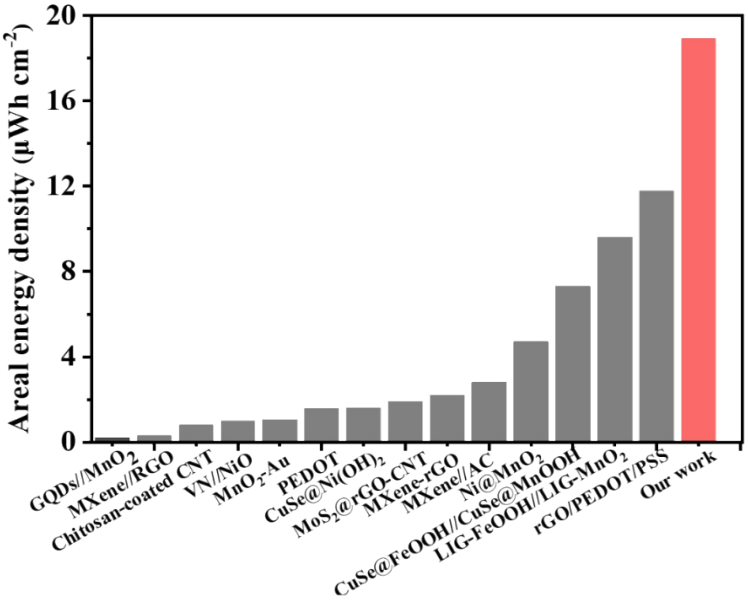

Fig. S9 Comparison of the areal energy density between GMC//GFC AMSC and other reported MSC and AMSC.[1-14] Ref. S1: GQDs//MnO_2, Ref. S2: MXene//RGO, Ref. S3: Chitosan-coated CNT, Ref. S4: VN//NiO, Ref. S5: MnO_2-Au, Ref. S6: PEDOT, Ref. S7: CuSe@$Ni(OH)_2$, Ref. S8: MoS_2@rGO-CNT, Ref. S9: MXene-rGO, Ref. S10: MXene//AC, Ref. S11: Ni@MnO_2, Ref. S12:CuSe@FeOOH//CuSe@MnOOH, Ref. S13:LIG-FeOOH//LIG-MnO_2, Ref. S14: rGO/PEDOT/PSS.

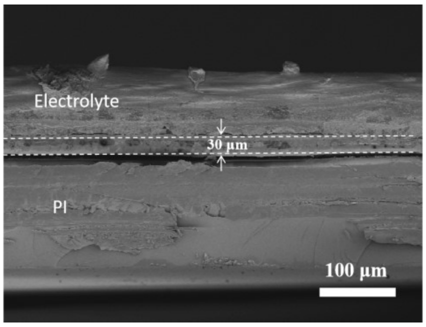

Fig S10 The cross-sectional SEM images of the printed pseudocapacitive electrodes for AMSCs.

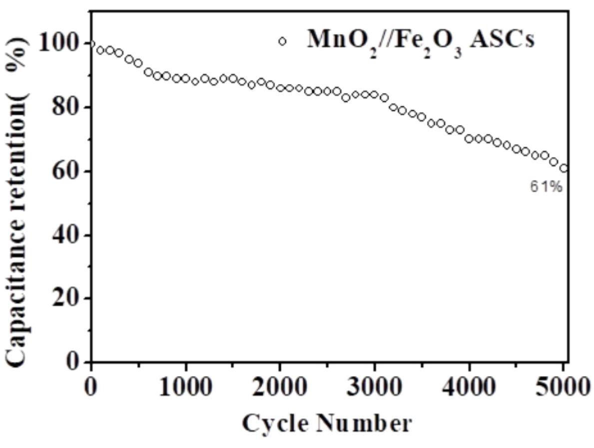

Fig. S11 Cycling performance of MnO_2//Fe_2O_3 ASC at a current density of 0.4 mA cm^{-2}.

Table S1 the Q_+/Q_- at various scan rates for GMC-X and GFC-Y.

Samples / Q+/Q- / Scan rate	GMC-2 /GFC-2	GMC-2 /GFC-1	GMC-2 /GFC-0.5	GMC-1 /GFC-2	GMC-1 /GFC-1	GMC-1 /GFC-0.5	GMC-0.5 /GFC-2	GMC-0.5 /GFC-1	GMC-0.5 /GFC-0.5
10	2.1	0.5	0.28	7.57	1.8	**1**	12.2	2.9	1.62
20	2.3	0.5	0.29	8.54	1.9	**1.07**	12.6	2.8	1.59
30	2.3	0.5	0.29	8.77	1.9	**1.12**	12.3	2.7	1.57
50	2.3	0.5	0.28	8.54	1.88	**1.06**	11.8	2.6	1.47
100	2.4	0.5	0.30	9.4	1.95	**1.16**	11.8	2.46	1.46

Table S2 Comparison of electrochemical performance of various MSCs and AMSCs.

Electrode material	Current collector	Electrolyte	Voltage window (V)	Areal Capacitance ($mF\ cm^{-2}$)	Areal Energy density ($\mu Wh\ cm^{-2}$)	References
GQDs//MnO_2	Au	Na_2SO_4	0-1.0	1.1	0.2	Ref.S1
MXene//RGO	-	PVA/H_2SO_4	0-1.0	2.4	0.3	Ref.S2
Chitosan-coated CNT	-	PVA/H_2SO_4	0-0.8	6.1	0.8	Ref.S3
VN//NiO	-	KOH	0-1.8	23.3	1	Ref.S4
MnO_2-Au	-	PVA/$LiClO_4$	0-0.8	11.9	1.1	Ref.S5
PEDOT	Au/Cr	H_2SO_4	0-0.8	21.3	1.8	Ref.S6
CuSe@Ni$(OH)_2$	Au	PVA/LiC	0-1.0	13.6	0.38	Ref.S7
MoS_2@rGO-CNT	-	PVA/H_2SO_4	0-1.0	13.7	1.9	Ref.S8
MXene-rGO	-	PVA/H_2SO_4	0-0.6	61	3.1	Ref.S9
MXene//AC	Au/Cr	PVA/Na_2SO_4	0-1.6	7.8	2.8	Ref.S10
Ni@MnO_2	Au	PVA/CH3COOLi	0-0.8	52.9	4.7	Ref.S11
CuSe@FeOOH//CuSe@MnOOH	Au	PVA/LiCl	0-1.3	20.5	4.8	Ref.S12
LIG-FeOOH//LIG-MnO_2	-	PVA/LiCl	0-1.8	21.9	9.9	Ref.S13
rGO/PEDOT/PSS.	Au	PVA/H_3PO_4	0-1.0	84.7	13.1	Ref.S14
GMC//GFC	-	**PVA/LiCl**	**0-1.8**	**42**	**18.9**	**Our work**

Reference

1 W.-W. Liu, Y.-Q. Feng, X.-B. Yan, J.-T. Chen and Q.-J. Xue, *Adv. Funct. Mater.,* 2013, 23, 4111-4122.

2 C. Couly, M. Alhabeb, K.L. Van Aken, N. Kurra, L. Gomes, A.M. Navarro-Suárez, B. Anasori,

H.N. Alshareef and Y. Gogotsi, *Adv. Electron. Mater.,* 2018, 4, 1700339.
3 Y. Yang, L. He, C. Tang, P. Hu, X. Hong, M. Yan, Y. Dong, X. Tian, Q. Wei and L. Mai, *Nano Res.,* 2016, 9, 2510-2519.
4 E. Eustache, R. Frappier, R.L. Porto, S. Bouhtiyya, J.F. Pierson and T. Brousse, *Electrochem. Commun.,* 2013, 28, 104-106.
5 H. Hu, Z. Pei, H. Fan and C. Ye, *Small*, 2016, 12, 3059-3069.
6 Y. Diao, Y. Lu, H. Yang, H. Wang, H. Chen and J.M. D'Arcy, *Adv. Funct. Mater.,* 2020, 2003394.
7 J. Gong, J.-C. Li, J. Yang, S. Zhao, Z. Yang, K. Zhang, J. Bao, H. Pang, and M. Han, *ACS Appl. Mater. Inter.,* 2018, 10, 38341-38349.
8 W. Yang, L. He, X. Tian, M. Yan, H. Yuan, X. Liao, J. Meng, Z. Hao and L. Mai, *Small,* 2017, 13, 1700639.
9 C.J. Zhang, M.P. Kremer, A. Seral-Ascaso, S.-H. Park, N. McEvoy, B. Anasori and Y. Gogotsi, *Adv. Funct. Mater.,* 2018, 28, 1705506.
10 Y. Xie, H. Zhang, H. Huang, Z. Wang, Z. Xu, H. Zhao, Y. Wang, N. Chen and W. Yang, Nano Energy, 2020, 74, 104928.
11 Y. Lin, Y. Gao and Z. Fan, *Adv. Mater.* 2017, 29, 1701736.
12 J.-C. Li, J. Gong, X. Zhang, L. Lu, F. Liu, Z. Dai, Q. Wang, X. Hong, H. Pang and M. Han, *ACS Appl. Energy Mater.,* 2020, 3, 3692-3703.
13 L. Li, J. Zhang, Z. Peng, Y. Li, C. Gao, Y. Ji, R. Ye, N.D. Kim, Q. Zhong, Y. Yang, H. Fei, G. Ruan and J.M. Tour, *Adv. Mater.,* 2016, 28, 838-845.
14 Y. Liu, B. Weng, Q. Xu, Y. Hou, C. Zhao, S. Beirne, K. Shu, R. Jalili, G.G. Wallace, J.M. Razal and J. Chen, *Adv. Mater. Technol.,* 2016, 1, 1600166.