Compositionally Variant Bimetallic Cu-Mn Oxysulfide Electrodes with Meritorious Supercapacitive Performance and High Energy Density

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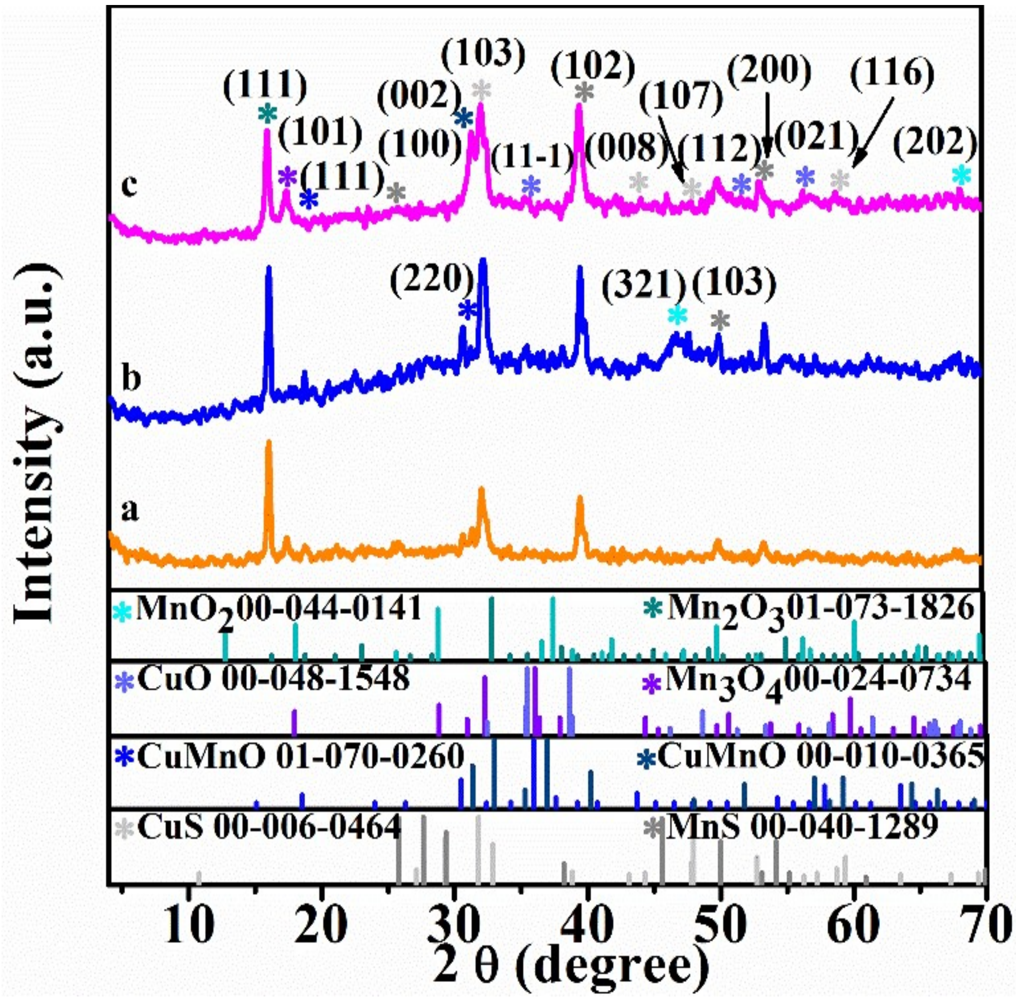

Figure S1: XRD patterns of (a) C_1M_3OS, (b) C_1M_1OS, and (c) C_3M_1OS nanocomposites with their corresponding reference cards.

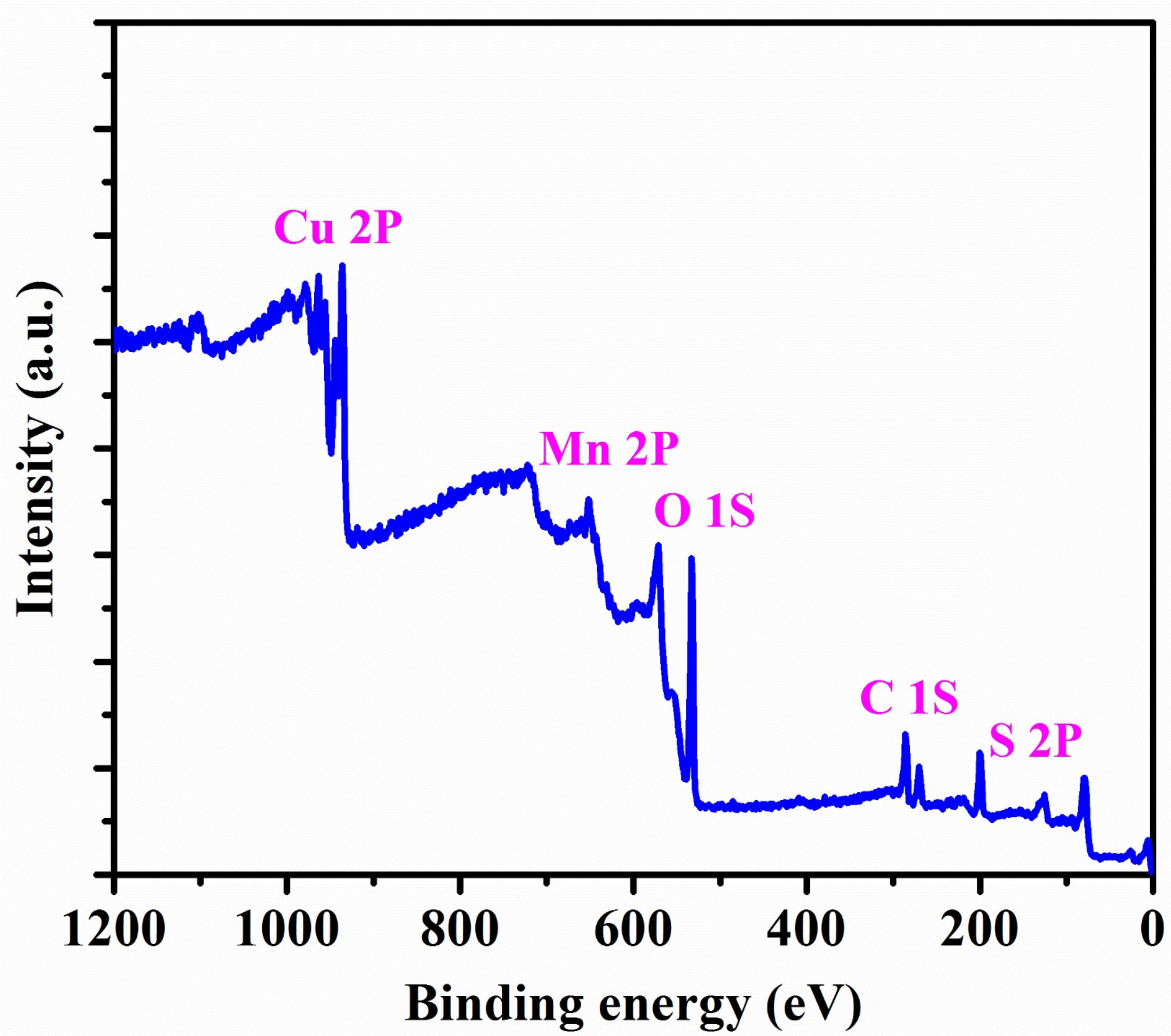

Figure S2: X-ray photoelectron spectroscopy survey scan of the C_3M_1OS nanocomposite.

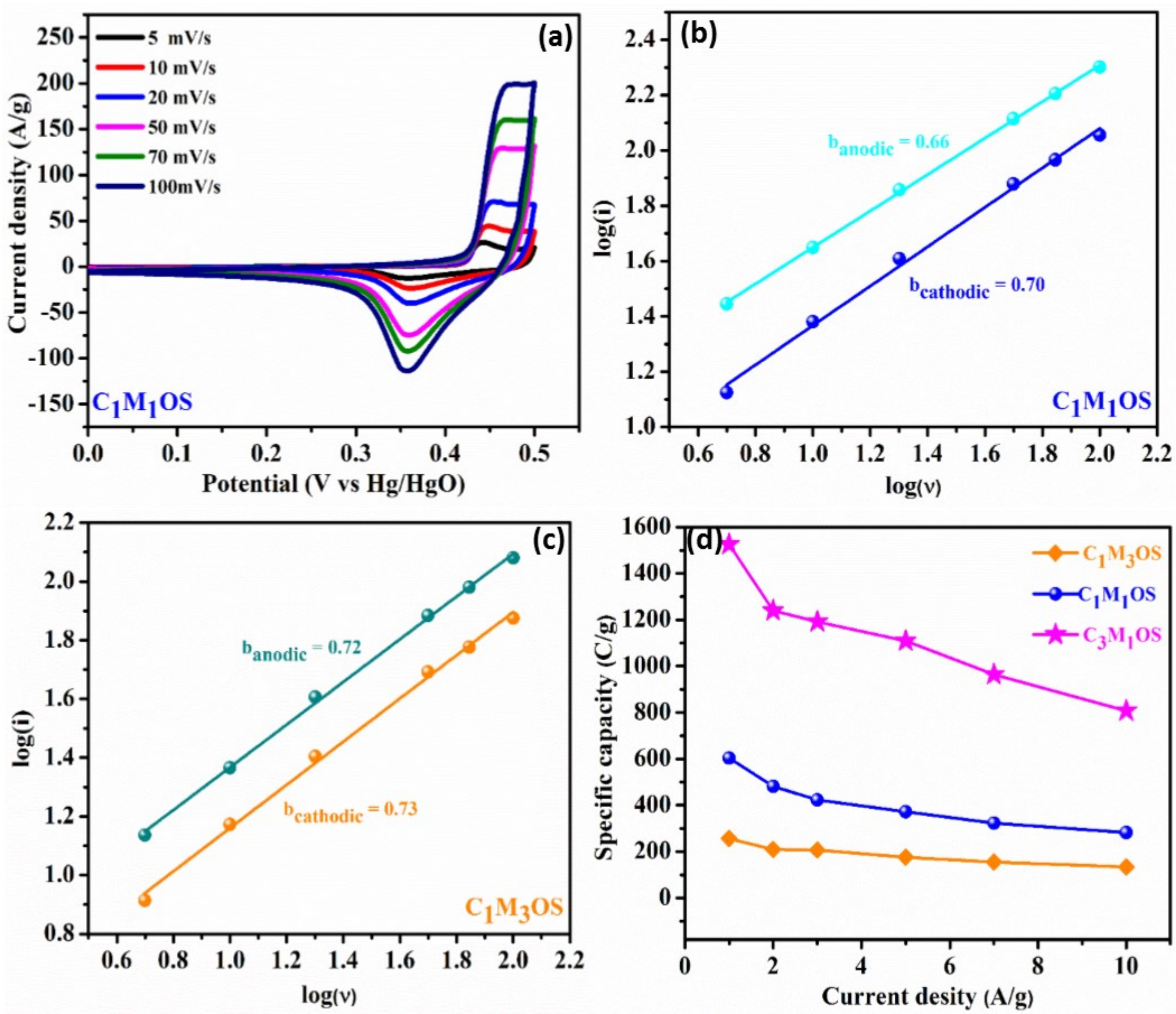

Figure S3: Three-electrode electrochemical measurements: (a) CV profile of C_1M_1OS at various scan rates (5−100 mV/s), The reliance of cathodic and anodic currents on the applied scan rate for (b) C_1M_1OS, (c) C_1M_3OS, and (d) specific capacitance recorded at different several scan rates for C_1M_3OS, C_1M_1OS, and C_3M_1OS electrodes in 2 M KOH.

The electrochemical active surface area (ECSA) was calculated from a series of cyclic voltammograms in the non-faradaic region at different scan rates (3–100 mV/s), as shown in Fig.

7a. The ECSA values can be obtained using the equation $ECSA = \frac{C_{DL}}{C_s}$, where Cs is the specific capacitance of a flat surface of the material per unit area. This specific capacitance is usually between 20 and 60 μF/cm^2 for metal-based materials in alkaline electrolytes; the average value of 40 μF/cm^2 is usually used and Cdl is the electrochemical double-layer capacitance of the Flat surface. [1–3] Based on the obtained Cdl values from the slopes in **Figure S4 b,d,f,** the ESCA was found to be 32, 25, and 17 cm2 C_3M_1OS, C_1M_1OS, and C_1M_3OS electrodes , respectively. Confirming the superior activity of the fabricated composites and the outstanding electrochemical performance of the C_3M_1OS electrode.

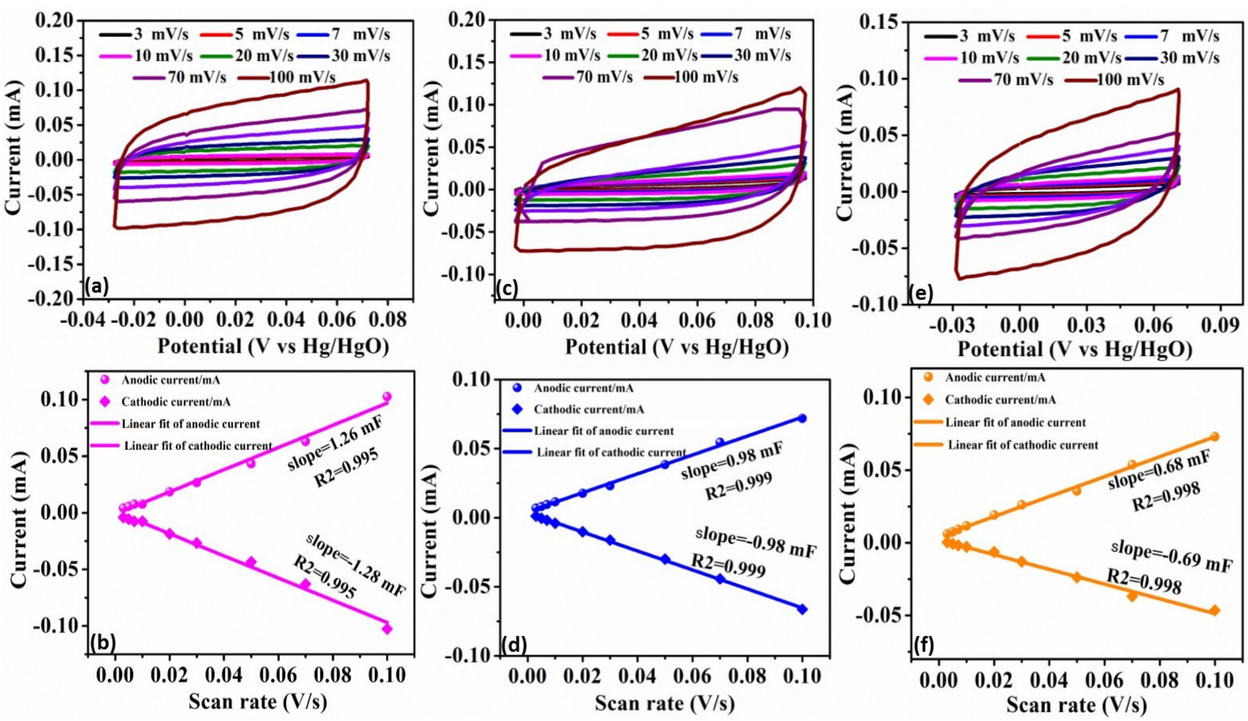

Figure S4: The electrochemical surface area measurements: cyclic voltammograms at different scan rates (3–100) mV/s, anodic and cathodic currents as a function of scan rate of the (a, b) C_3M_1OS, (c, d) C_1M_1OS, and (e, f) C_1M_3OS electrodes.

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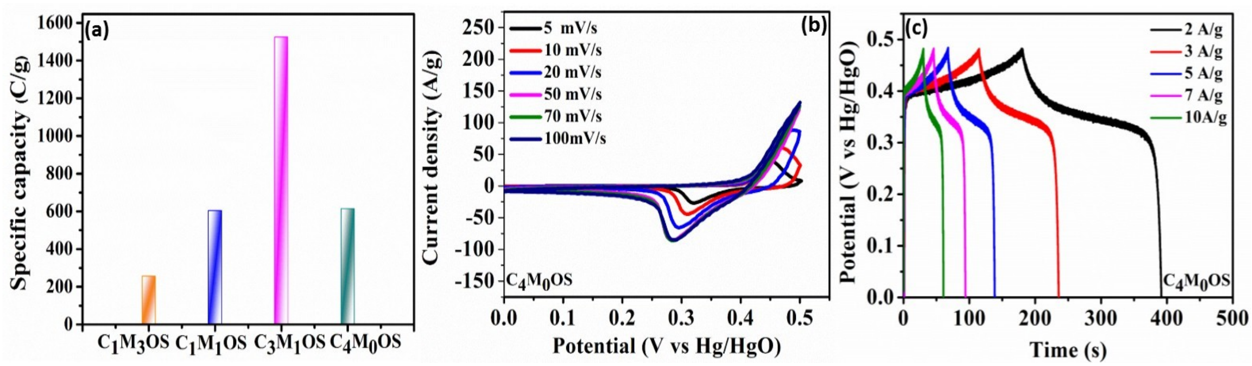

Figure S5: Three-electrode electrochemical measurements: (a) specific capacity of the C_1M_3OS, C_1M_1OS, C_3M_1OS and C_4M_0OS electrodes at a current density of 1 A/g. (b) CV profiles at various scan rates (5−100 mV/s), and (c) GCD profiles at various current densities (2−10 A/g) of C_4M_0OS electrode.

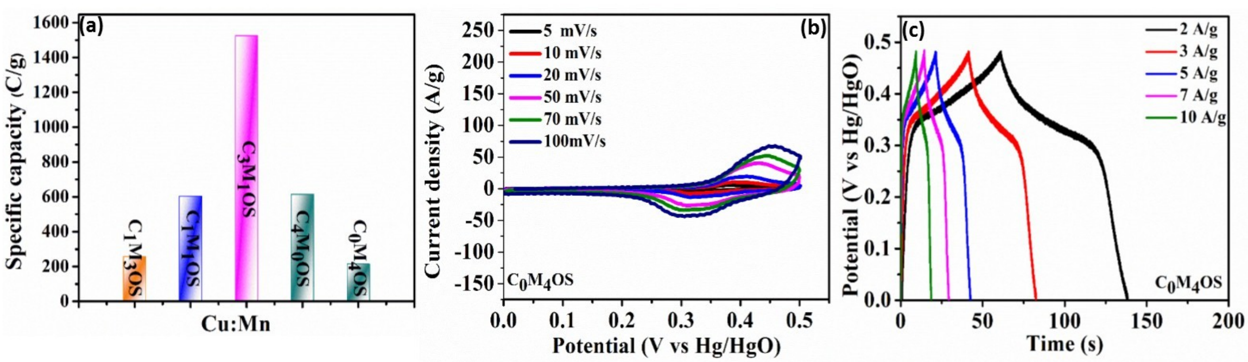

Figure S6: Three-electrode electrochemical measurements: (a) specific capacity of the C_1M_3OS, C_1M_1OS, C_3M_1OS, C_4M_0OS and C_0M_4OS electrodes at a current density of 1 A/g. (b) CV profiles at various scan rates (5−100 mV/s), and (c) GCD profiles at various current densities (2−10 A/g) of C_0M_4OS electrode.

Material characterization of C_3M_1OS **electrode after cycling**

As for the morphology of the electrode after cycling, SEM image (**Figure S7a**) showed the retaining of the nanocube-like structure covered with nanoneedles with the presence of some aggregates. Additionally, EDX and XPS analysis shown in **Figure S7b,c** confirmed the presence of all chemical elements (Cu, Mn, S, and O) after cycling, demonstrating the stability of the C_3M_1OS electrode.

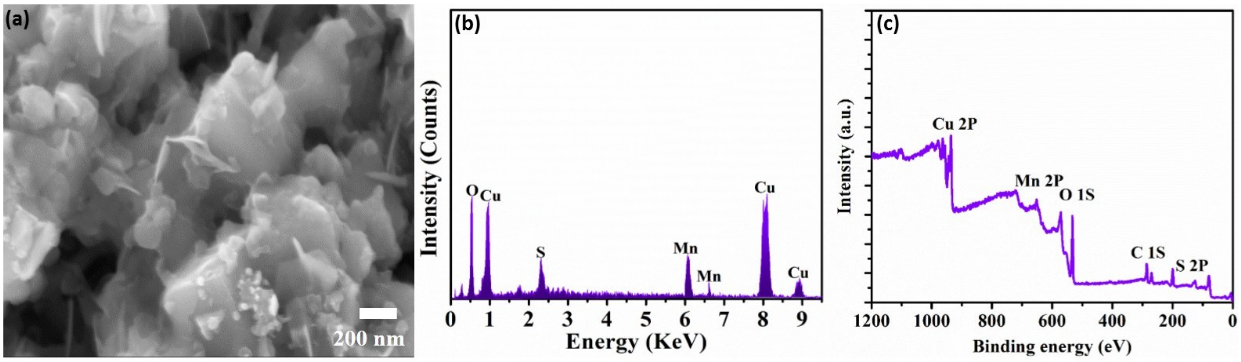

Figure S7: (a) field-emission scanning electron microscopy (FESEM) image, (b) the EDX spectrum and (c) X-ray photoelectron spectroscopy survey scan of C_3M_1OS electrode after cycling.

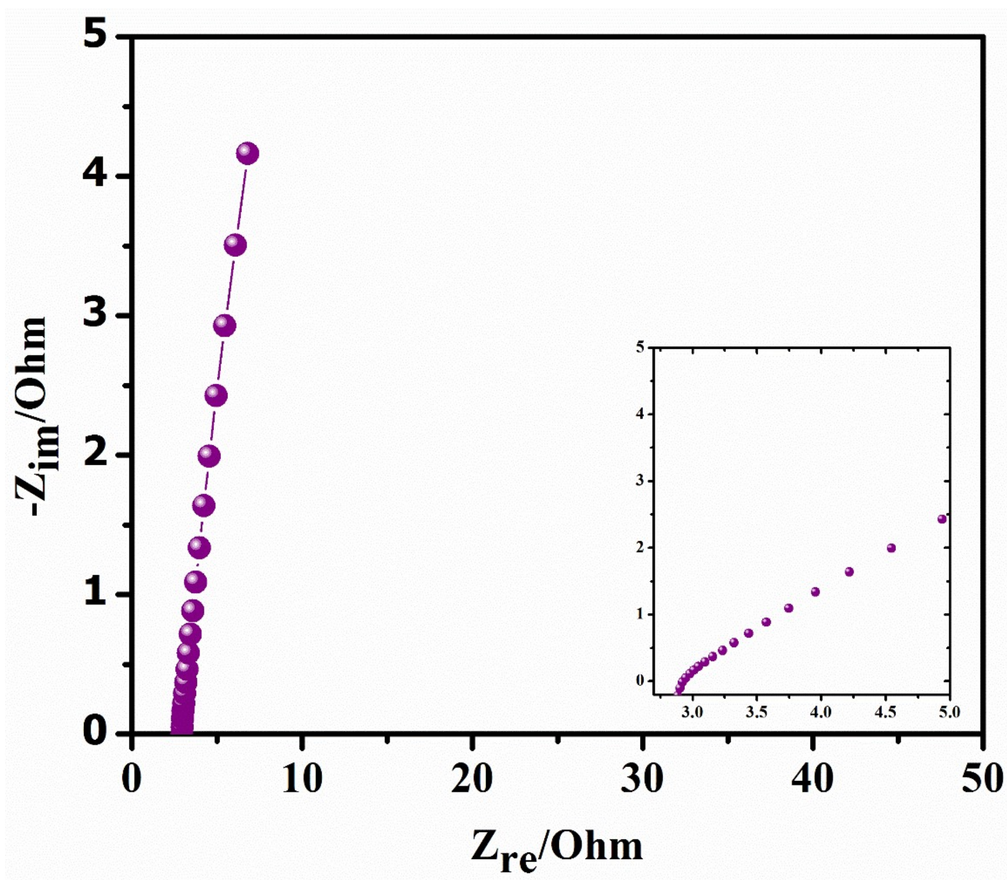

Figure S8: Electrochemical characteristics of C_3M_1OS//AC asymmetric device: (a) Nyquist plots for the designed device.

Table S1 Bader net atomic charge

Element	**C_1M_3OS**	**C_1M_1OS**	**C_3M_1OS**
Cu	0.413269	0.480593	0.514096
Cu	---	0.490029	0.532672
Cu	---	---	0.895732
Mn	1.041738	1.61466	1.75898
Mn	1.577323	1.614569	---
Mn	1.577179	---	---
O	-1.329480	-1.268808	-1.117837
O	-1.329707	-1.268761	-1.117261
S	-0.975155	-0.831103	-0.712779
S	-0.975167	-0.83118	-0.753602
No of electrons	56	60	64

Bader net atomic charge =ZVAL-Bader population