

Supplementary Information

Multifunctional copper sulphide nanoflowers for superior supercapacitors and microwave absorption

Maiyong Zhu*, Kai Zhang

School of Materials Science & Engineering, Jiangsu University, Zhenjiang, 212013, P. R. China

Material characterization

The comprehensive characterisation of the samples was accomplished using multiple techniques: X-ray diffraction (XRD, Cu K α radiation, $\lambda=1.5406$ Å) was performed with a scan range of 5–80° at a rate of 8°/min to determine crystal structure and phase composition; Multiscale morphological observation and quantitative elemental analysis were performed using scanning electron microscopy (SEM) and transmission electron microscopy (TEM) combined with energy-dispersive spectroscopy (EDS); X-ray photoelectron spectroscopy (XPS) and Fourier transform infrared spectroscopy (FTIR) were employed to analyse the chemical states of surface elements and functional groups respectively; the specific surface area and pore structure of the material were systematically evaluated via BET nitrogen adsorption. Magnetic parameters were measured using a VSM (HH-15), while electromagnetic parameters were recorded via a vector network analyser (VNA, N5224 A) to elucidate electromagnetic wave absorption characteristics. Coaxial annular samples with an inner diameter of 3.04 mm and an outer diameter of 7 mm were prepared by loading 30 wt% absorber into paraffin wax.

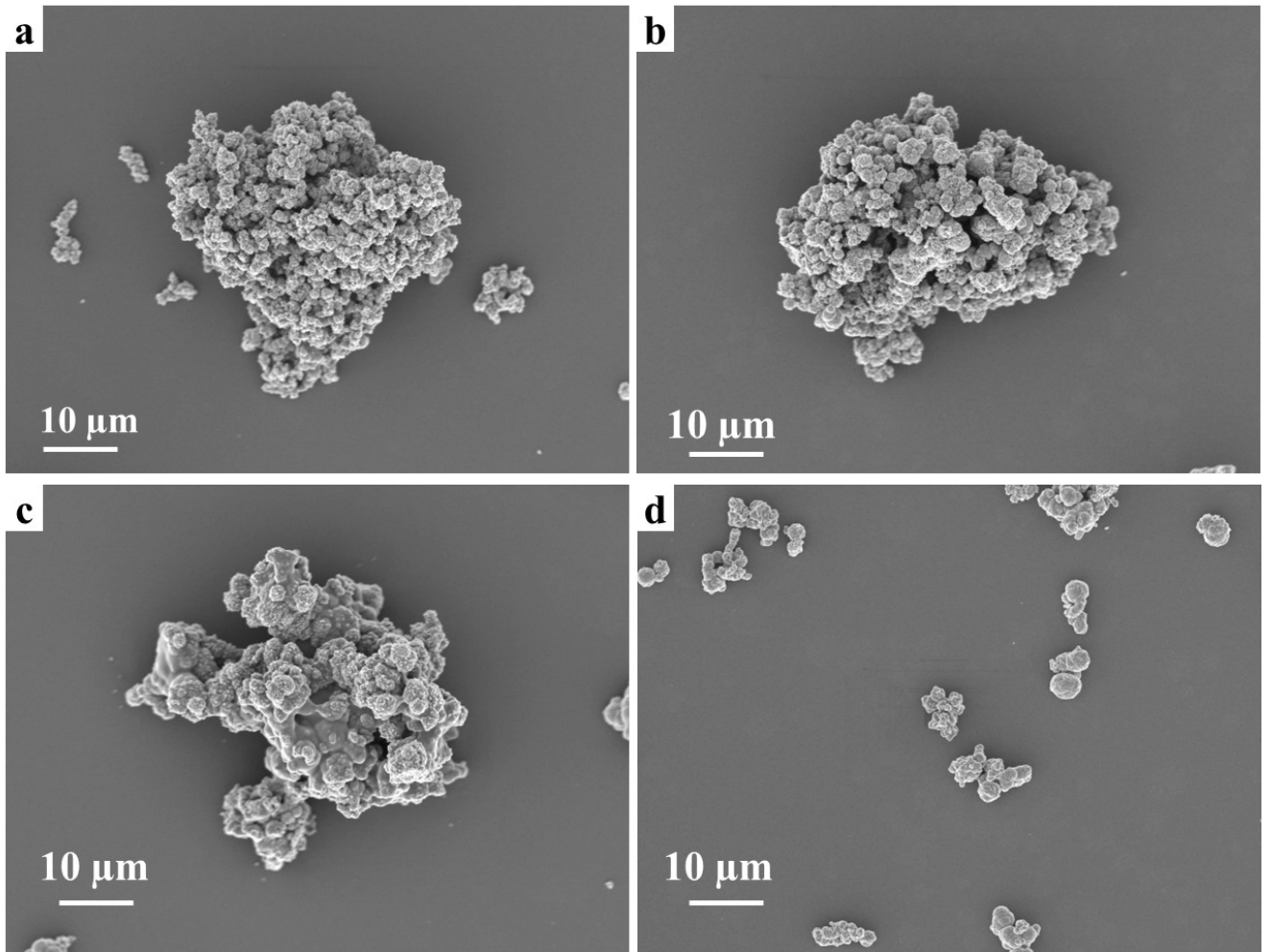


Figure S1 SEM images of Cu_xS_y : (a) Cu_xS_0 ; (b) Cu_xS_{20} ; (c) Cu_xS_{30} ; (d) Cu_xS_{60} .

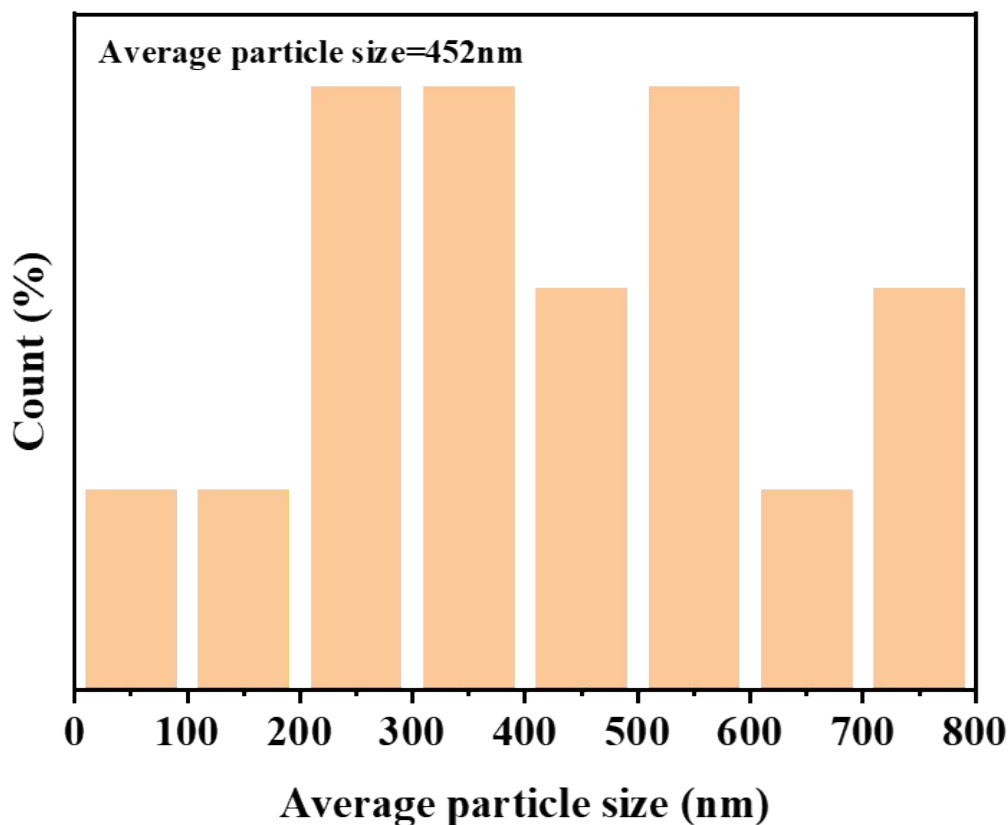


Figure S2 Average particle size analyzer of Cu_xS-40.

Preparation of Electrode Sheets

It is imperative to acknowledge the significance of electrode composition in determining the efficacy of supercapacitors. Electrode sheets were prepared according to the following procedure: PVDF (binder), acetylene black (conductive agent), and the active material were combined in a mass ratio of 1:1.5:7.5. We then ground the mixture in a mortar while adding NMP solvent incrementally until a homogeneous slurry was achieved. The slurry was applied in a uniform manner to the surface of a pristine 1×1 cm² foam nickel substrate. Subsequently, the nickel foam was subjected to vacuum-drying in an oven at a temperature of 60°C for a duration of 12 hours. After removal, the foam nickel was pressed at 18 MPa for 40s to ensure the material adhered firmly to the foam nickel surface. The pressed foam nickel was weighed in order to ensure that the activity of the material was maintained at a level of between 3–4 mg.cm⁻². Prior to the implementation of electrochemical testing, the prepared electrode underwent an activation process in a 1 M KOH solution for a duration of approximately 3 hours. The preparation of the activated carbon electrode followed a similar procedure to the working electrode, differing only in the replacement of the active material with activated

carbon.

Electrochemical performance testing

The average crystallite size (D) was calculated from the XRD data using the Scherrer equation:

$$D = \frac{K\lambda}{\beta \cos \theta} \quad (1)$$

where K is the shape factor (0.89), λ is the X-ray wavelength (0.15406 nm), β is the full width at half maximum (FWHM) of the diffraction peak in radians, and θ is the Bragg angle.

In electrochemical testing within a three-electrode system, the working electrode comprises the fabricated electrode, with the counter electrode and reference electrode being a platinum sheet and a saturated calomel electrode (SCE), respectively. The electrolyte consists of a 1 M KOH aqueous solution. For two-electrode system testing, the fabricated electrode serves as the cathode and is assembled with an activated carbon electrode as the anode to form the device. The electrolyte remains 1 M KOH aqueous solution. The mass loading of the cathode and anode is determined according to the following formula:

$$\frac{m_+}{m_-} = \frac{C_- \times \Delta V_-}{C_+ \times \Delta V_+} \quad (2)$$

The mass of the positive and negative active materials is denoted as m_+ and m_- , respectively, in milligrams (mg), while C_+ and C_- correspond to the specific capacitance values of the positive and negative materials measured at the same current density in the three-electrode test system in Farad per gram ($F g^{-1}$). The electrochemical windows of the positive and negative electrode materials are defined as ΔV_+ and ΔV_- in volts (V), respectively, characterizing their respective operating voltage intervals.

Specific capacitance (C), energy density (E) and power density (P) are calculated according to the following equations:

$$C = \frac{I\Delta t}{m\Delta V} \quad (3)$$

$$E = \frac{C\Delta V^2}{2 \times 3.6} \quad (4)$$

$$P = \frac{3600 \times E}{\Delta t} \quad (5)$$

where Δt , I, ΔV and m represent the discharge time (t), current density ($A g^{-1}$), operating voltage window (V) and mass of active material loaded on the electrode (g), respectively.

The relationship between peak current (i) and scan rate (v) can be calculated by the following

equation:

$$i = ab^v \#(6)$$

In the quantitative analysis of the electrochemical energy storage mechanism, Eqs. a and b are adjustable parameters that can be determined by experimental fitting. Among them, the value of b is the key parameter to reveal the control mechanism of charge storage behavior: when $b = 0.5$, the charge storage process follows the diffusion control mechanism; when $b = 1$, the charge storage process is dominated by the capacitance control mechanism.

The ratio of capacitive control and diffusion control can be calculated by the following equation:

$$i = k_1 v + k_2 v^{\frac{1}{2}} \#(7)$$

where i represents the peak current, v represents the scan rate, and k_1 and k_2 are constants.

Microwave Absorption Testing

Reflected loss

$$RL = 20 \log \left| \frac{Z_{in} - Z_0}{Z_{in} + Z_0} \right| \quad (8)$$

Impedance matching

$$Z_{in} = Z_0 \sqrt{\frac{\mu_r}{\epsilon_r} \tanh \left(j \frac{2\pi f d \sqrt{\mu \epsilon}}{c} \right)} \quad (9)$$

Z_{in} represents the impedance of the absorber material itself, whilst Z_0 denotes the impedance of free space. The degree of impedance matching is characterised by the normalised characteristic impedance Z ($Z = |Z_{in}/Z_0|$).

Decay constant

$$\alpha = \frac{\sqrt{2}\pi f}{c} \sqrt{(\mu'' \epsilon'' - \mu' \epsilon') + \sqrt{(\mu'' \epsilon'' - \mu' \epsilon')^2 + (\epsilon' \mu'' + \epsilon'' \mu')^2}} \quad (10)$$

Table S1. equivalent circuit fitted values for impedance.

Sample	$R_s(\Omega)$	$R_{ct}(\Omega)$	$W_o(S \cdot s^{1/2})$
Cu _x S-0	1.498	88.73	0.215
Cu _x S-20	1.267	452.1	0.1958
Cu _x S-30	1.115	221.4	0.1952
Cu _x S-40	1.264	133.8	0.3526
Cu _x S-60	1.303	100.4	0.1806