Supporting Information

Electrochemically triggered rational design of bismuth copper sulfide for wearable all-sulfide semi-solid-state supercapacitor with a wide operational potential window (1.8 V) and ultra-long life

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S1. Material characterization

To study the surface morphology and elemental mappings of the prepared samples, field-emission scanning electron microscope (FE-SEM, Carl Zesis) equipped with energy-dispersive X-ray spectroscopy (EDS) analysis was employed, and inductively coupled plasma spectroscopy (ICP, Direct Reading Echelle ICP, LEEMAN) was performed. Intrinsic morphology was investigated by using a transmission electron microscope (TEM). The crystal structure of the prepared samples was investigated using X-ray diffraction (XRD, Mac Science M18XHF-SRA, Cu K α radiation; $\lambda = 1.5406$ Å). The existing oxidation states of elements in the optimized sample were investigated using X-ray photoelectron spectroscopy (XPS, K-alpha (Thermo Electron)) analysis. High-resolution Raman (HR-Raman) spectroscopy (inVia Raman microscope) was performed.

S2. Calculations

$$\frac{2I\int v(t) dt}{m \times V}$$
(S1)
$$C_{ts} = \frac{I \times \Delta t}{m \times V}$$
(S2)
$$P = \frac{E}{\Delta t}$$
(S3)

where ' C_{ts} ' is the specific capacity (mAh g⁻¹), ' $\int v(t) dt$, is an integral area (cm²), 'V' is the potential window (V), ' C_s ' is the specific capacitance (F g⁻¹), and 'E' and 'P' are the energy density (Wh kg⁻¹) and power density (W kg⁻¹), respectively. ' Δt ' is referred to as the discharge time (s), '*m*' is referred to as the mass of the active electrode (g), and '*I*' is a current (A). ^{S1}



Fig. S1. TEM images of the (a-c) BCS-0.5, (d-f) BCS-1, and (g-f) BCS-2. The insets inside the HR-TEM images show the corresponding SAED patterns, respectively.



Fig. S2. Pie charts of the detected atomic ratio percentages from ICP spectroscopy analysis for the (a) BCS-1 and (b) NS. CV curves at different scan rates for the (c) BCS-0.5 and (d) BCS-2 electrodes. GCD curves at different current densities for the (e) BCS-0.5 and (f) BCS-2 electrodes.



Comparative (g) Raman spectra and (h) XRD patterns of the BCS-1 electrode before and after cycling.

Fig. S3. Cycling stability performance of the ASSSC device at 2 and 6 A g^{-1} .

Reference

[S1] M. Nagaraju, S. C. Sekhar, S. J. Arbaz, and J. S. Yu, Solvothermal-derived nanoscale spinel bimetallic oxide particles rationally bridged with conductive vapor-grown carbon fibers for hybrid supercapacitors, *Appl. Surf. Sci.*, 2021, **563**, 150223.