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All-Redox Hybrid Supercapacitors Based on Carbon Modified Stacked Zinc Cobaltite Nanosheets

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Supporting Information

1. Electrode preparation and assembly

For the preparation of the electrodes, the active material was blended with PVDF and carbon black in a ratio of 8:1:1. PVDF acted as a binder and carbon black worked as a conductive element. The three materials were blended properly in a mortar pestle with the help of N-Methyl-2-pyrrolidone (NMP) to get uniform slurry. The slurry was then coated onto the Nickel foam cut in circular form with area of 1 cm^2. The electrode was then kept for drying overnight at 60 ℃ in a vacuum oven. The separator (Whatman paper) wetted by 6M KOH electrolyte was sandwiched between the two electrodes and pressed using a hydraulic press to get the desired cell configuration. For symmetric configuration, two similar electrodes were taken. For asymmetric configuration, two different electrodes of different materials were chosen.

2. Electrochemical measurements

All the electrochemical measurements have been performed in 2-electrode configuration. The electrochemical investigations (CV and GCD) of the pure $ZnCo_2O_4$ and the composite $ZnCo_2O_4$/AC have been performed in symmetric configuration with a voltage range of 0-1V. For asymmetric configuration, the voltage window was optimized to be 0-1.6V. The EIS analysis was carried out for 10^5-0.1 Hz of frequency at open-circuit voltage. The Formulae used for determining various parameters such as specific capacitance, energy density, and power density has been provided in **Table S2**.

Table S1: Formulae used for finding various structural parameters.

Parameter	Formula	Terms used
Bragg's law	$2d\sin\theta = \lambda$	'd' is inter-planar spacing, 'θ' is bragg's diffraction angle, λ is the wavelength of X-ray
Interplanar spacing	$d = \frac{1}{\sqrt{\frac{h^2}{a^2}+\frac{k^2}{b^2}+\frac{l^2}{c^2}}}$	(hkl) are miller indices of lattice plane, (a, b, c) are lattice parameters of the crystal

Crystallite size, D (Schherrer equation)	$D = \frac{K.\lambda}{\omega \times \cos\theta}$	K(~0.94) is a constant, λ is the wavelength of X-ray, 'ω' is the FWHM, 'θ' is bragg's diffraction angle
Cell volume, V	$V = abc \sin\beta$	$a, b, and\ c$ are lattice parameters of the crystal structure. 'β' is the lattice angle
Micro strain, ε	$\varepsilon = \frac{\omega \times \cos\theta}{4}$	'ω' is the FWHM, 'θ' is bragg's diffraction angle
Dislocation density, δ	$\delta = \frac{1}{D^2}$	'D' is the crystallite size

Table S2: Formulae used for electrochemical investigation.

Parameter	**Formula**	**Terms used**
Specific capacitance, C_{sp} (from CV)	$C_{sp} = \frac{\int IdV}{m \times \nu \times dV}$	'I' is the current, dV is the potential window, 'm' is the mass of total active material, 'ν' is the scan rate.
Specific capacitance, C_{sp} (from GCD)	$C_{sp} = \frac{I \times \Delta t}{m \times dV}$	'I' is the current, 'dt' is the discharging time, 'm' is the mass of total active material.
Energy density, E_d (from GCD)	$E_d = \frac{C_{sp} \times (dV)^2}{7.2}$	'C_{sp}' is the specific capacitance, dV is the voltage window.
Power density, P_d (from GCD)	$P_d = \frac{E_d \times 3600}{\Delta t}$	'E_d' is the energy density, Δt is the discharging time.
Coulombic efficiency, η	$\eta = \frac{t_d \times 100}{t_c}$	't_d' is the discharging time and 't_d' is the charging time.
Response time, τ (from EIS)	$\tau = \frac{1}{\nu}$	'ν' is the frequency corresponding to phase angle $\theta = 45°$.

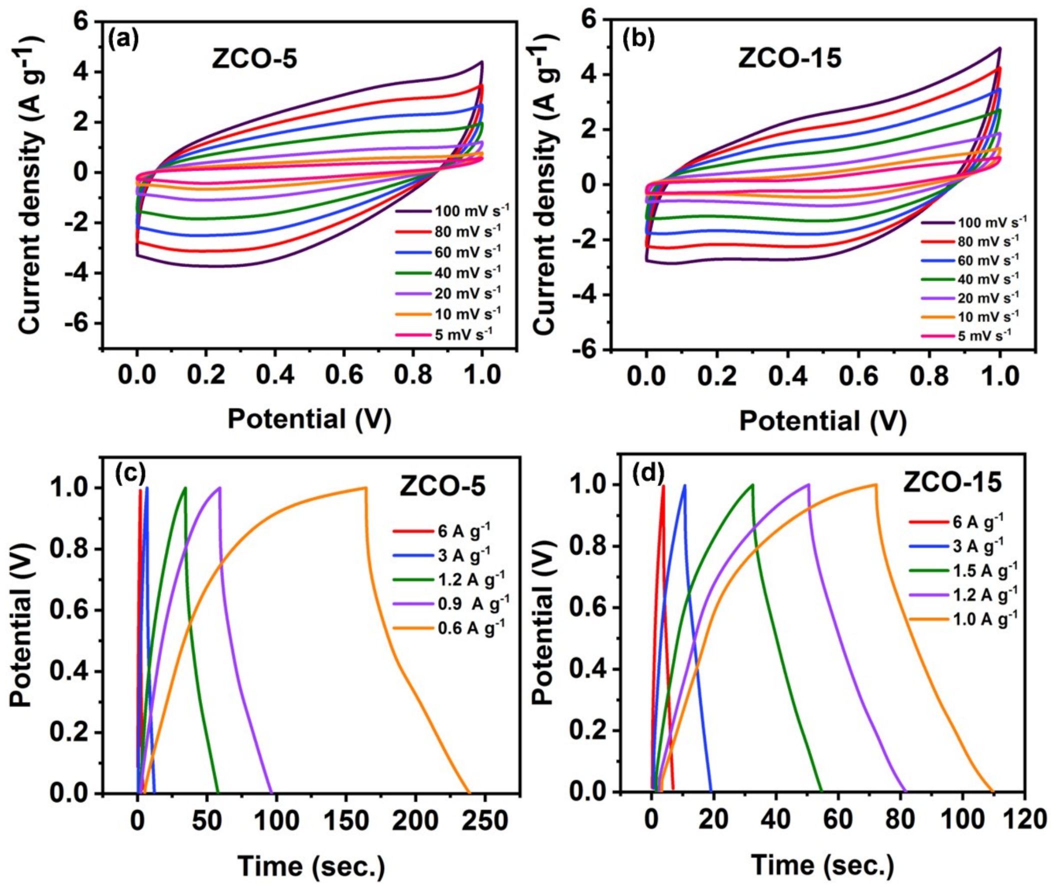

Fig. S1: (a, b) CV of ZCO-5 and ZCO-15 at various scan rates; **(c, d)** GCD of ZCO-5 and ZCO-15 at various currents.

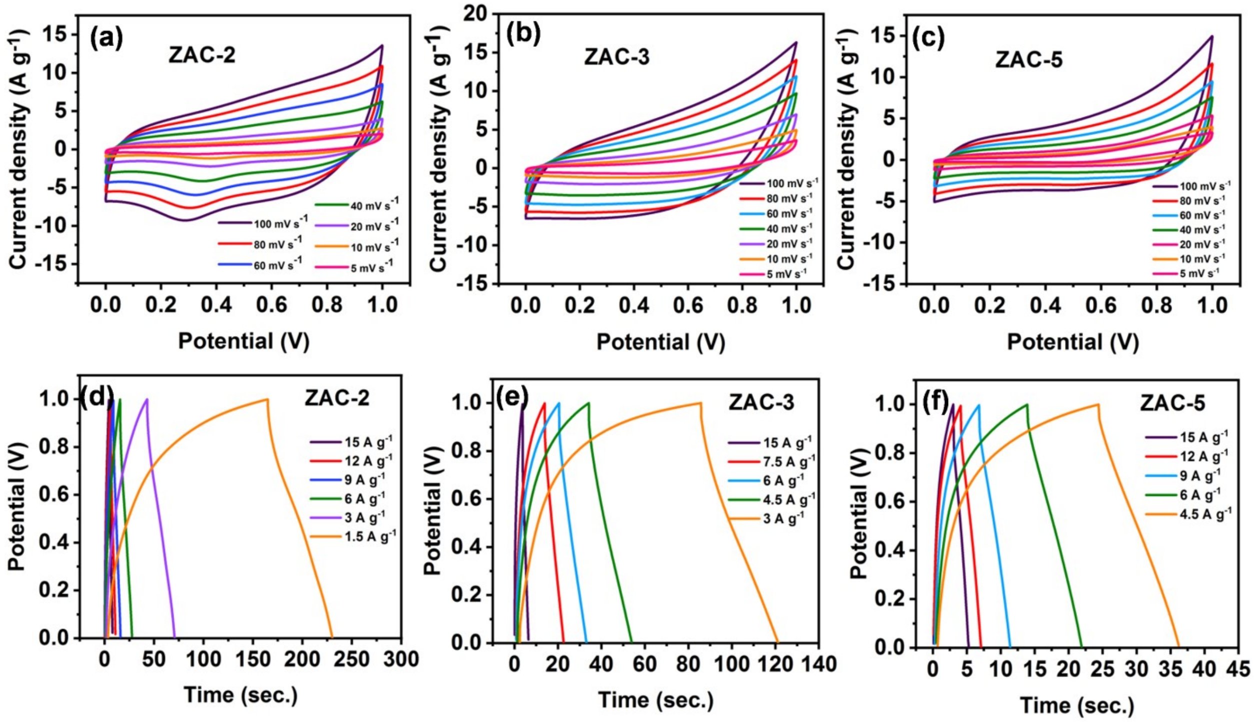

Fig. S2: (a-c) CV of ZAC-2, ZAC-3, and ZAC-5 at various scan rates; **(d-f)** GCD of ZAC-2, ZAC-3, and ZAC-5 at various currents.

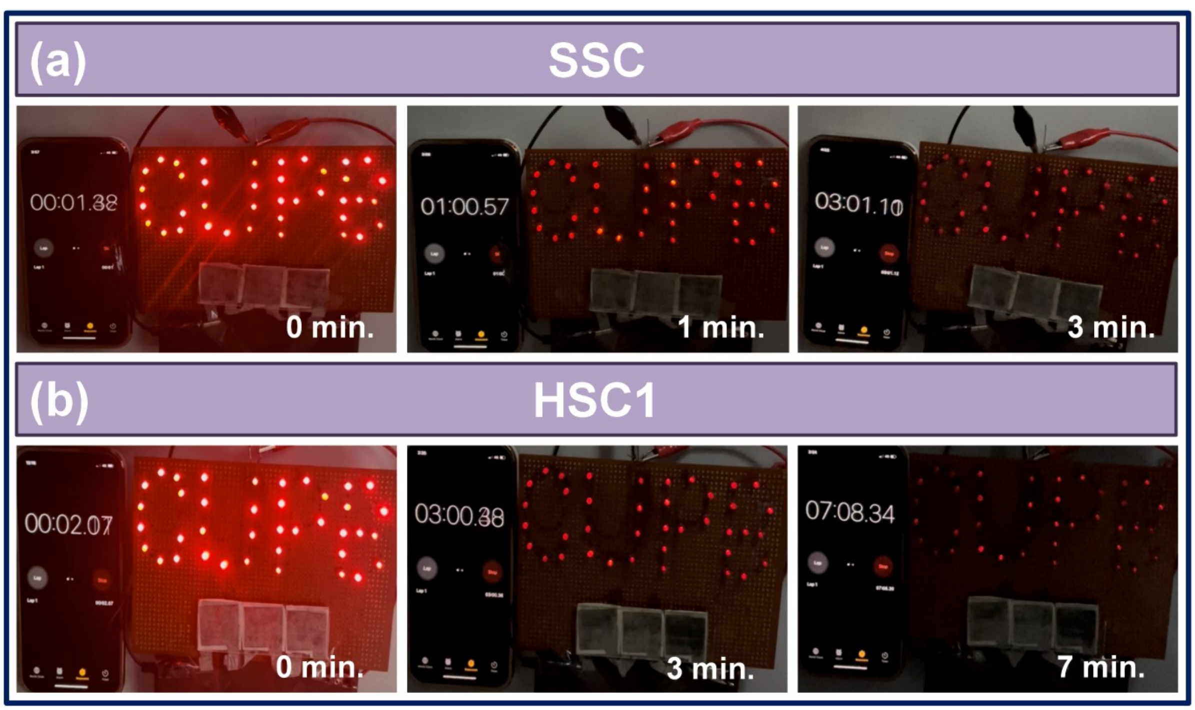

Fig S3: Photographs of illuminated LED panel for **(a)** SSC and **(b)** HSC1 at different times.

3. Three-electrode measurements of ZNCO-10

For 3-electrode analysis, the prepared ZNCO-10 electrode was taken as working electrode, Ag/AgCl as a reference electrode, and platinum electrode was used as the counter electrode. The cyclic voltammetry (CV) and constant current charge/discharge (GCD) analysis was performed to investigate the nature of the electrode material. The CV curves were measured at various scan rates (10–100 mV s^{-1}), and the GCD performance was tested in the potential window density from 0 to 0.4 V at different currents ranging from 2 to 12 A g^{-1}. All these measurements were conducted at room temperature. **Fig. S4(a)** represents the CV curves of the optimized ZNCO-10 with obvious redox peaks showing pseudocapacitive behavior of the electrode material. **Fig. S4(b)** represents the GCD curves for the sample with obvious charge plateaus.

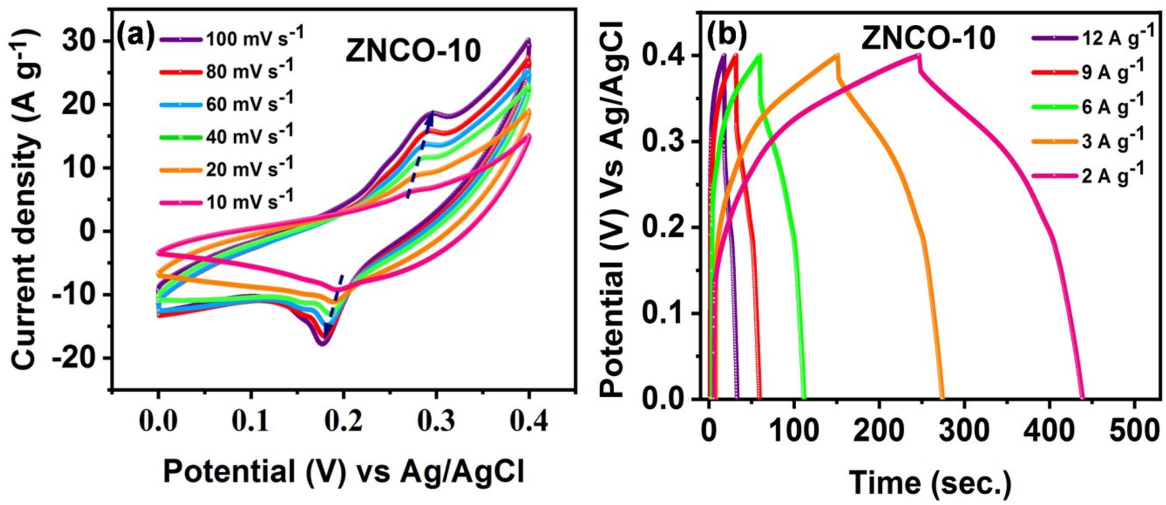

Fig. S4: (a) CV of ZNCO-10 at different scan rates; **(b)** GCD of ZNCO-10 at different current densities.