

Supporting Information

Porous carbon nanosheets armoring 3D current collector towards ultrahigh mass loading for high-energy-density all-solid-state supercapacitor

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Calculation Methods

1. Three-electrode cells

The area-specific capacitance was calculated from the galvanostatic discharge profile, according to Eqn. S1.

$$C_a = (I \times \Delta t) / (\Delta U \times S) \quad \text{Eqn. S1}$$

where C_a (F cm⁻²) is the area-specific capacitance, I (A) is the discharge current, Δt (s) is the discharge time, ΔU (V) is the operation potential window, and S (cm²) is the area (1.0 cm × 1.0 cm) of the electrode.

2. Two-electrode solid-state asymmetric cells

As for the NCM2.0@C@Ni//AC device, the cathode's mass ratio and the anode were determined according to Eqn. S2.

$$m_+ / m_- = (C_- \times \Delta U_-) / (C_+ \times \Delta U_+) \quad \text{Eqn. S2}$$

where m (mg) is the mass of active materials, C (F g⁻¹) is the mass-specific capacitance, and ΔU (V) is the voltage window. The subscripts represent the cathode and the anode. The mass-specific capacitance (C_m) was calculated based on the total mass loading of both active materials (~62 mg cm⁻²). The area-specific capacitance (C_a) is calculated on Eqn. S1. The energy density and power density were calculated according to Eqns. S3 and S4, respectively.

$$E = (C \Delta U^2) / 2 \quad \text{Eqn. S3}$$

$$P = E / \Delta t \quad \text{Eqn. S4}$$

where E (μWh cm⁻² or Wh g⁻¹) is the energy density, P is the power density (μW cm⁻²

or $W \text{ kg}^{-1}$), C (F cm^{-2} or F g^{-1}) is the mass- or area-specific capacitance of the ASC, ΔU (V) is the voltage window, and Δt (s) is the discharge time.

3. Surface capacitance contribution

The proportion of the capacitance contribution can be quantitatively determined by Dunn's method, i.e., separating the current response (i) at a fixed potential (V) in the CV curve into a capacitive effect and a diffusion control reaction. The current (i) and the scan rate (ν) have a relationship shown in Eqn. S5.

$$i(U) = k_1\nu + k_2\nu^{1/2} \quad \text{Eqn. S5}$$

where i (A) is the current response value at a specific voltage (U), ν (V s^{-1}) is the scan rate, k_1 and k_2 are coefficients. k_1 and k_2 represent the capacitance contribution from the surface capacitance and semi-infinite linear diffusion of ions.

Table S1. Experimental conditions for all the NCM-based samples.

Samples	M^{x+} (mmol)	Urea (g)	DI (mL)	Mass loading (mg cm^{-2})
NCM0.5@Ni	0.5	0.338	30	3.2
NCM1.0@Ni	1.0	0.676	30	5.1
NCM2.0@Ni	2.0	1.352	30	12.9
NCM3.0@Ni	3.0	2.028	30	27.3
NCM0.5@C@Ni	0.5	0.338	30	5.2
NCM1.0@C@Ni	1.0	0.676	30	10.6
NCM2.0@C@Ni	2.0	1.352	30	23.1
NCM3.0@C@Ni	3.0	2.028	30	40.8

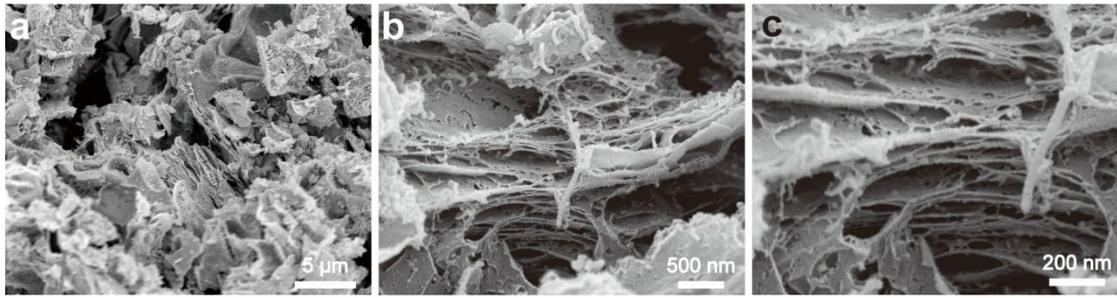


Figure S1. SEM images at different magnifications of the carbon nanosheets obtained without the Ni foams.

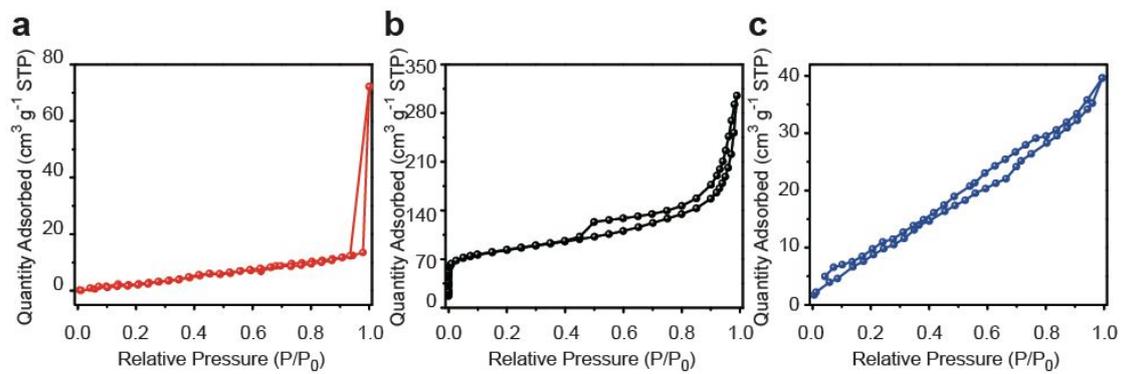


Figure S2. N₂ adsorption/desorption isotherms of (a) the pristine Ni foam; (b) carbon nanosheets; (c) C@Ni.

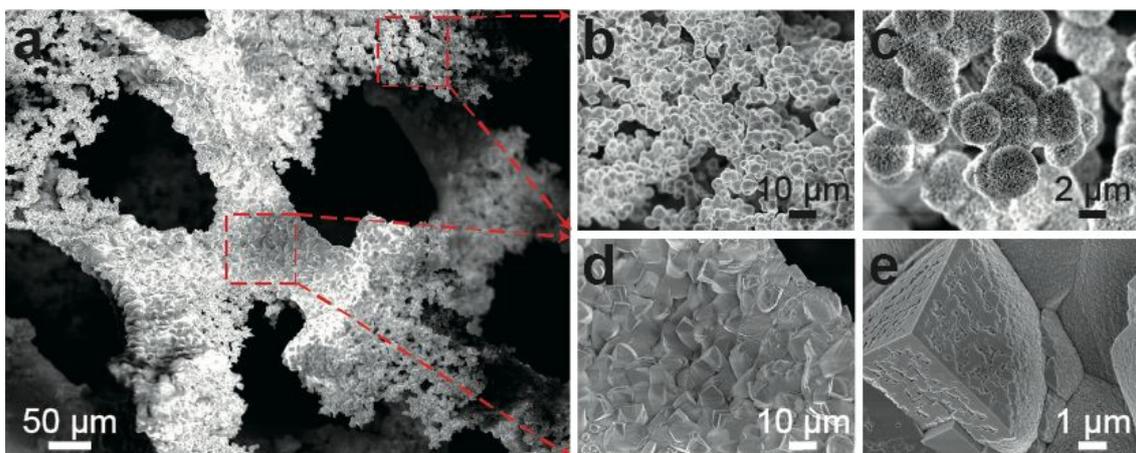


Figure S3. SEM images with different magnifications of NCM3.0@C@Ni

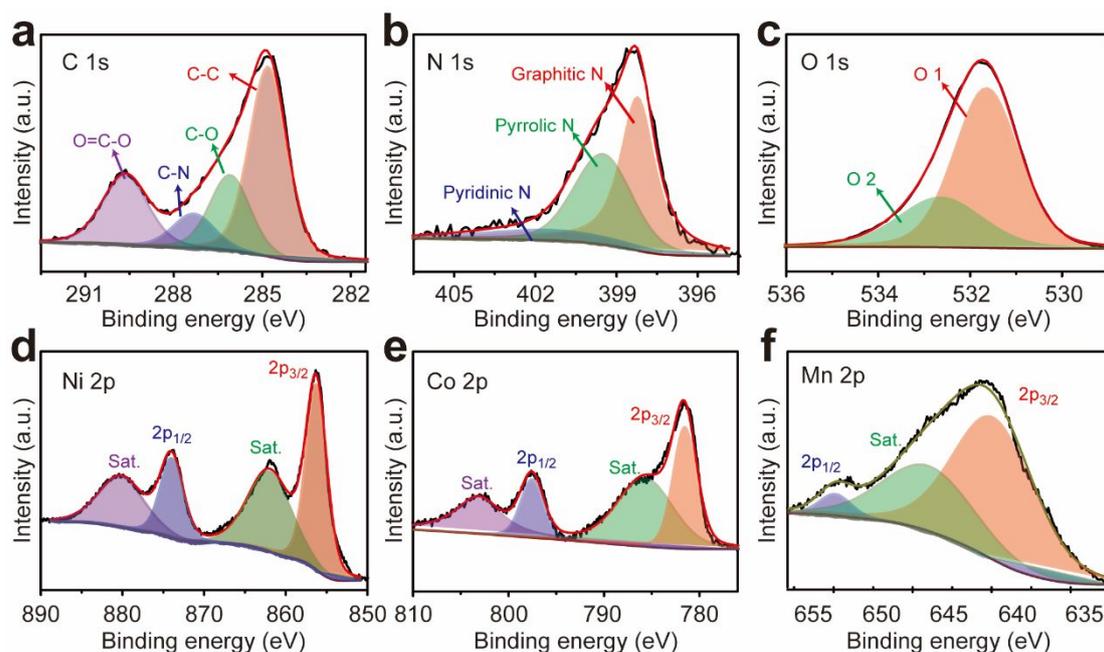


Figure S4. XPS spectra of the NCM2.0 nanoneedle deprived from the C@Ni: (a) C 1s; (b) N 1s; (c) O 1s; (d) Ni 2p; (e) Co 2p and (f) Mn 2p.

XPS spectra were obtained to elaborate further on the chemical composition and element valence of the NCM2.0 nanoneedle. Figure S4 presents the high-resolution XPS spectra of C 1s, N 1s, O 1s, Ni 2p, Co 2p, and Mn 2p fitted by the Gaussian method separately. Figure S4a confirms that the carbon element appears as the C-C, C-O, sp^3 C-N, and O=C-O¹⁻². The N 1s spectrum shows the existence of graphitic N (398.2 eV), pyrrole N (399.5 eV), and pyridine N (401.4 eV) (Figure S4b). The graphitic N dopes into the C₆ ring and bonds with sp^2 hybridized C atoms, providing a feasible way to transfer ions and charges². In Figure S4c, the peak at 531.7 eV belongs to the O 1s electron of transition metal hydroxides³; the peak at 532.6 eV corresponds to HOH in adsorbed water molecules¹. The XPS spectra of Ni 2p, Co 2p, and Mn 2p can be fitted with two prominent spin-orbit peaks and two satellite peaks (labeled as Sat.), confirming the presence of Ni²⁺, Co²⁺, and Mn²⁺ on the surface, respectively. This result is consistent with XRD, FTIR, and Raman.

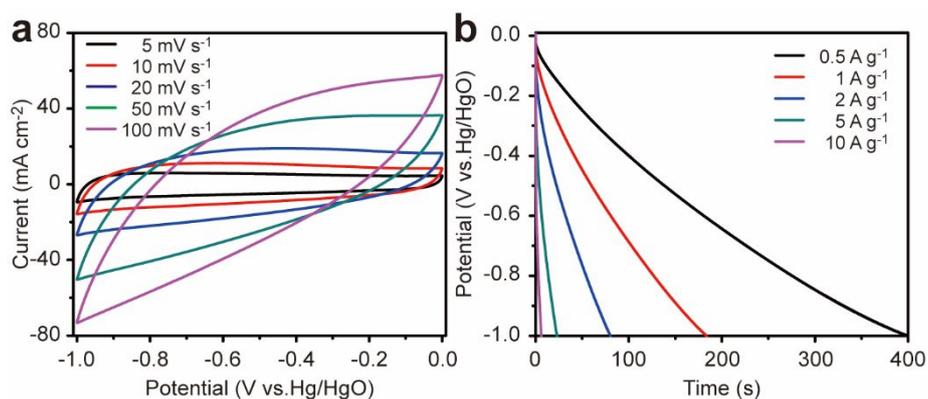


Figure S5. The performance of the AC single electrode: (a) CV curves at different scan rates and (b) GCD profiles at different current densities.

References

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- (3) Yin, C.; Yang, C.; Jiang, M.; Deng, C.; Yang, L.; Li, J.; Qian, D. A Novel and Facile One-Pot Solvothermal Synthesis of PEDOT-PSS/Ni-Mn-Co-O Hybrid as an Advanced Supercapacitor Electrode Material. *ACS Appl. Mater. Interfaces* **2016**, *8*, 2741-2752.