Supporting Information

Battery-capacitor hybrid yarn device with excellent flexibility and high electrochemical performance

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Experimental section

Preparation of CoNi-LDHs@CBY battery-type yarn electrode

Generally, the carbon-based yarn substrate was added into concentrated HNO_3 solution and soaked for 24 h, then washed with ultrapure water until neutral. Subsequently, 0.280 g of $Ni(CH_3COO)_2 \cdot 4H_2O$, 0.328 g of $Co(NO_3)_2 \cdot 6H_2O$ and 0.148 g NH_4F were mixed into 60 mL ultrapure water and stirred until completely dissolved. Then, the pre-treated carbon-based yarn substrate was suspension submerged in the mixed solution through a special mold, and oven at different temperatures (T=150°C, 180°C and 200°C) for 48 h. The obtained yarn was washed by ultrapure water and labelled as CoNi-LDHs@CBY yarn electrodes (150°C, 180°C and 200°C).

Preparation of S-CNPs@CBY capacitor-type yarn electrode

Typically, 3.00 g of C_2H_5NS was dispersed in 10 mL of ultrapure water. Then, the above pre-treated carbon-based yarn substrate was immersed in the solution and stirred for 24 h in 60°C. Followed, the precipitate yarn was washed, dried and annealed at 400°C for 2 h under Ar atmosphere, named as S-CNPs@CBY capacitor-type yarn electrode.

Fabrication of battery-capacitor hybrid yarn device

Firstly, for the battery-capacitor hybrid yarn device was employed CoNi-LDHs@CBY battery-type cathode and S-CNPs@CBY capacitor-type anode, respectively. Then, the electrodes were soaked in the prepared aqueous polyvinyl alcohol (PVA)/KOH gel electrolyte for 24 h, and evenly intertwined with each other. After coating the PVA/KOH gel on the electrodes once more, the battery-capacitor hybrid yarn device were obtained.

Material characterizations and electrochemical measurements

The Morphological, phase composition and structure were characterized by the scanning electron microscopy (SEM, Regulus8220, Hitachi, Japan), Transmission electron microscopy (TEM, JEM-2100, JEOL, Japan), X-ray diffraction (XRD) pattern (D8 Advance, Bruker) with Cu K_α radiation (λ=1.54056 Å), and X-ray

photoelectron spectroscopy (XPS, ESCALAB-250Xi, America) with AI-Kα X-ray source, respectively. Cyclic voltammetry (CV), galvanostatic charge-discharge (GCD), and electrochemical impedance spectroscopy (EIS) were carried out by an electrochemical workstation (CHI 660E, Chenhua, shanghai). In the three-electrode system, obtained activity yarn, Pt foil and Ag/AgCl were used as the working, counter and reference electrode in 1 M KOH solution, respectively.

The areal capacitance Cs (mF cm^{-2}), energy density E (μWh cm^{-2}) and power density P (mW cm^{-2}) is calculated by followed equations:

$$Cs = (I \times \Delta t)/(S \times \Delta U) \quad (1)$$

$$E = \frac{1}{2} Cs \Delta U^2 \Big/ 3.6 \quad (2)$$

$$P = 3.6E/\Delta t \quad (3)$$

$$S = 2\pi RL \quad (4)$$

Where R and L are the yarn radius and length, I (mA)/S (cm^2) denotes the current density, ΔU (V) represents the output voltage and Δt (s) is the discharge time.

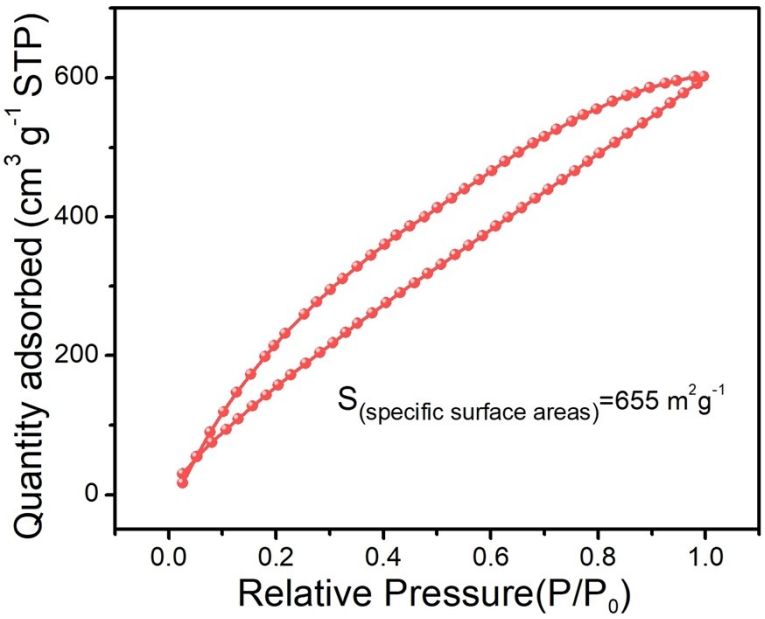

Fig. S1. N_2 adsorption-desorption curves and specific surface areas of CoNi-LDHs@CBY yarn electrode.

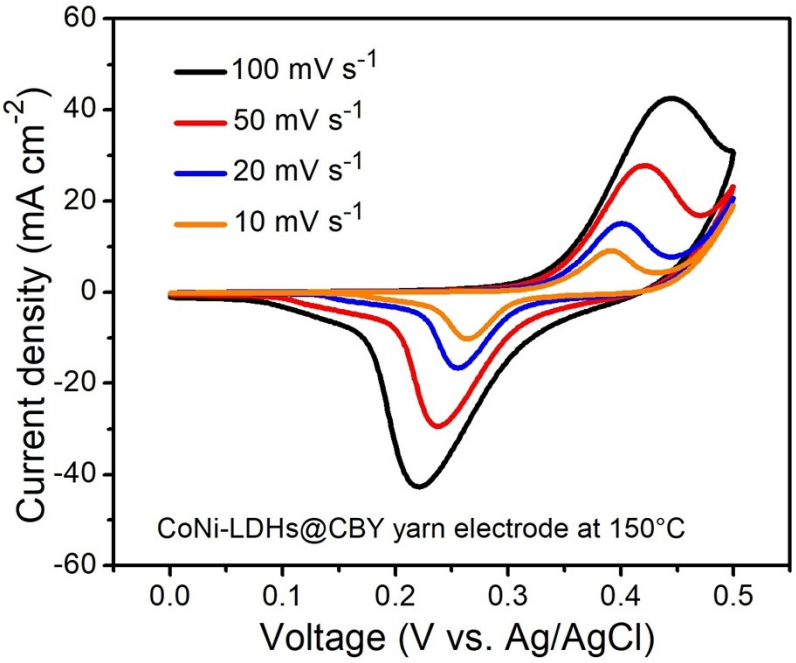

Fig. S2. CV curves of CoNi-LDHs@CBY yarn electrode at 150°C.

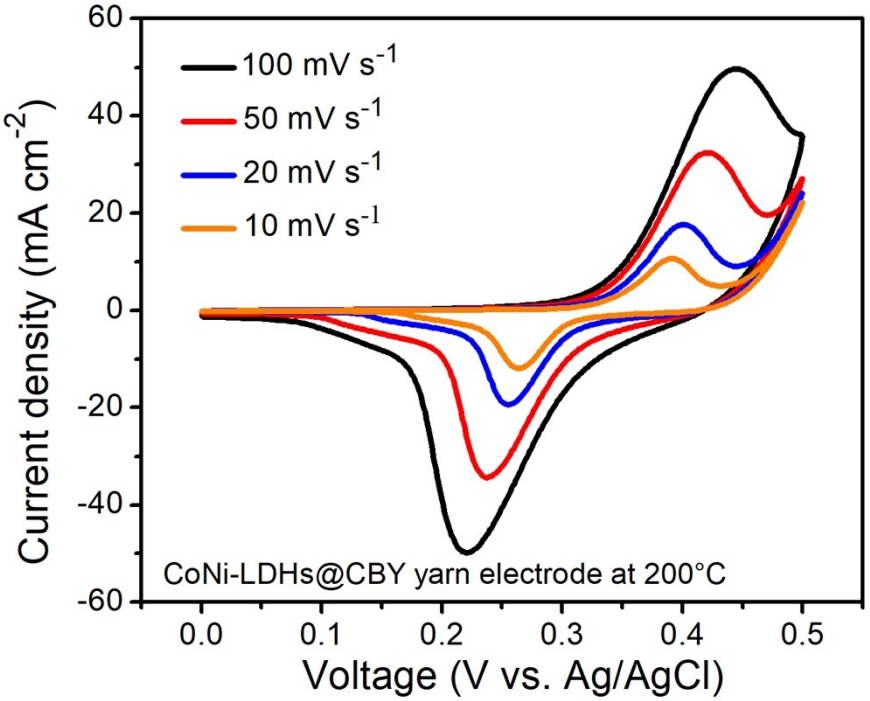

Fig. S3. CV curves of CoNi-LDHs@CBY yarn electrode at 200°C.

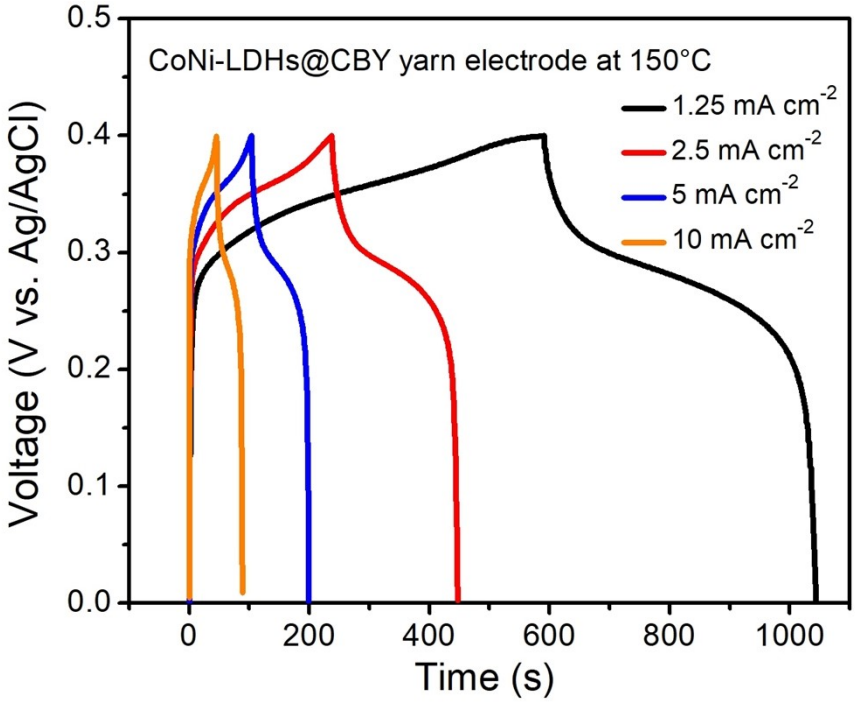

Fig. S4. GCD profiles of CoNi-LDHs@CBY yarn electrode at 150°C.

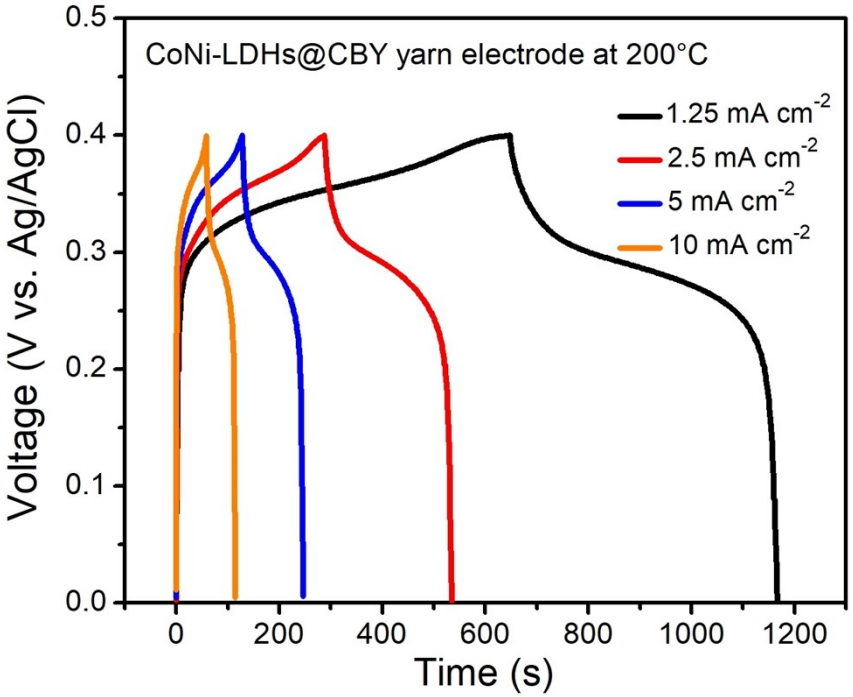

Fig. S5. GCD profiles of CoNi-LDHs@CBY yarn electrode at 200°C.

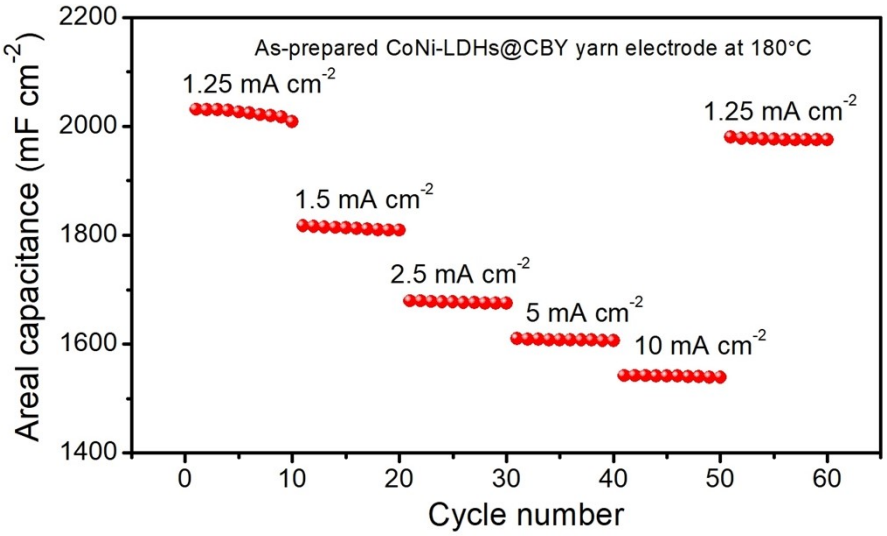

Fig. S6. Rate performance of the as-prepared CoNi-LDHs@CBY yarn electrode at 180°C.

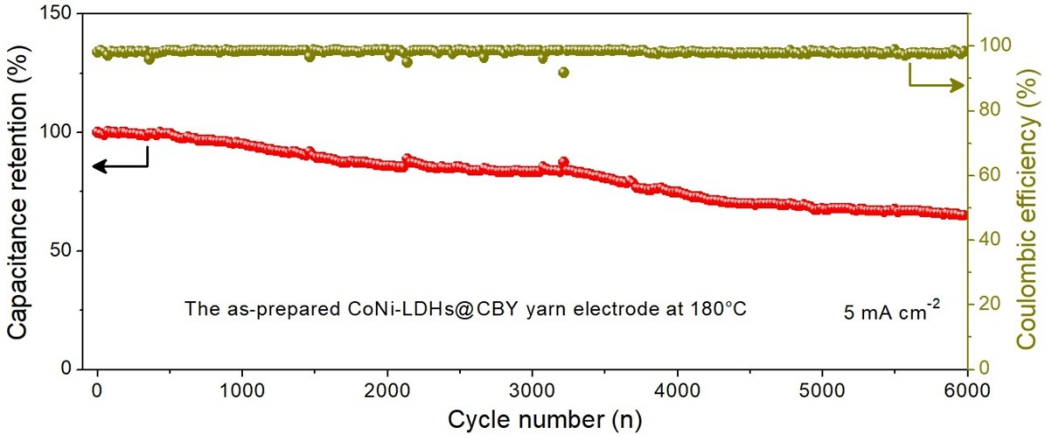

Fig. S7. Cycling performance of the as-prepared CoNi-LDHs@CBY yarn electrode at 180°C.

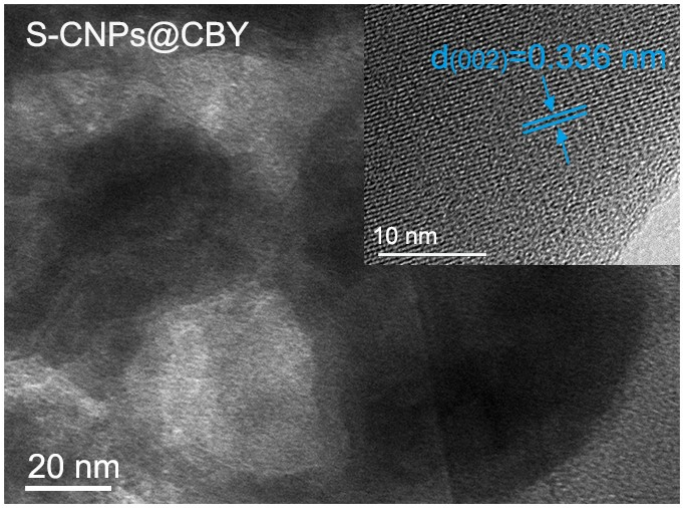

Fig. S8. The TEM and HRTEM images of S-CNPs@CBY yarn electrode.

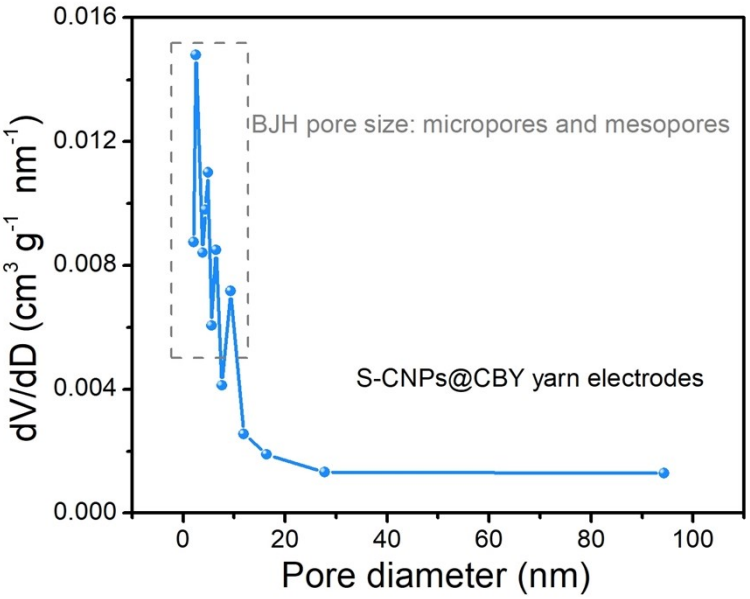

Fig. S9. The Barrett-Joyner-Halenda (BJH) pore size distribution profiles of S-CNPs@CBY yarn electrode.

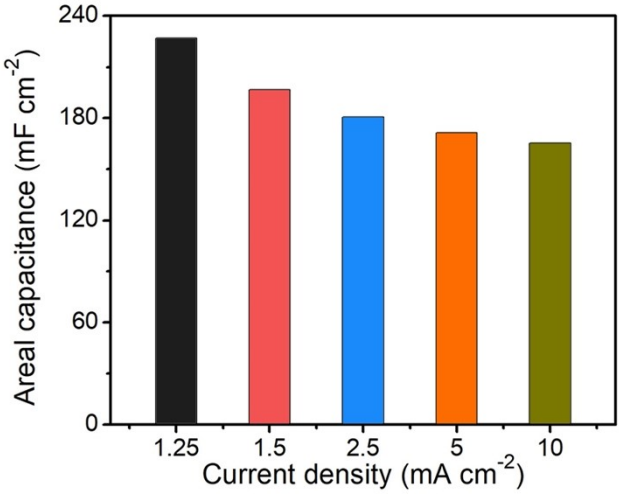

Fig. S10. The areal capacitance of the flexible CoNi-LDHs@CBY//S-CNPs@CBY device.

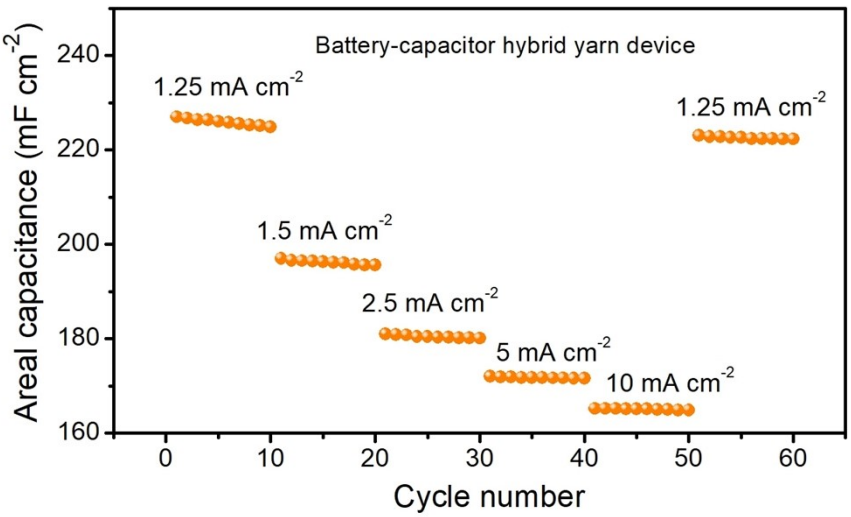

Fig. S11. Rate performance of the CoNi-LDHs@CBY//S-CNPs@CBY device.

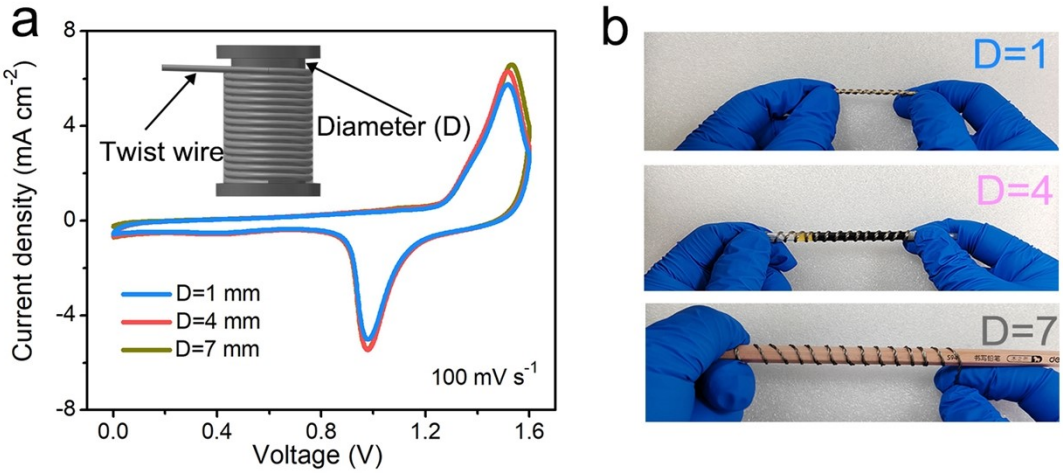

Fig. S12. (a) CV curves and (b) photograph of the flexible CoNi-LDHs@CBY//S-CNPs@CBY device under different twist twine diameters.