

Supporting Information

**Approaching high-performance lithium storage materials of
CoNiO₂ microspheres wrapped coal tar pitch-derived porous
carbon**

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1. Synthesis of CTP

Coal tar pitch-derived porous carbon (abbreviated as CTP) was prepared by template method. The nano-calcium carbonate powder (Adamas Reagent Co., Ltd, AR, particle size: 50-100 nm) and coal tar pitch with a mass ratio of 1:2 were mixed, then the tetrahydrofuran solvent (Adamas Reagent Co., Ltd, AR, 99.9%) was added to the mixture slowly with a vigorous stirring. Subsequently, the resultant solution was treated in an oil bath at 90°C with continuous stirring until the solvent completely evaporated, and the obtained solid mixture was ground into powder with particle size, and then was calcined at 900°C at a heating rate of 5 °C min⁻¹ for 3 h under N₂ atmosphere. After annealing, the samples were immersed in 3 M HCl (Aladdin Reagent Co., Ltd, 36-68 wt%) solution overnight to remove calcium carbonate, centrifuged and washed with deionized water (DI water) for several times, and then dried at 120°C to obtain CTP products.

2. Synthesis of CoNiO₂ microspheres

CoNiO₂ was synthesized by one-step hydrothermal method. First, 1 mmol Co(NO₃)₂·6H₂O (Adamas Reagent Co., Ltd, AR, 99%), 1 mmol Ni(NO₃)₂·6H₂O (Adamas Reagent Co., Ltd, AR, 99%), 3 mmol NH₄F (Adamas Reagent Co., Ltd, AR, 98%) and 5 mmol CO(NH₂)₂ (Adamas Reagent Co., Ltd, AR, 99%) were successively dissolved into 75 ml DI water with a stirring to obtain a uniform mixed solution. Then the solution was moved into the Teflon-lined autoclave and sealed. After held at 120 °C in an electric oven for 12 h, the autoclave was cooling to room temperature in air

and the precursors were gathered and washed with deionized water and ethanol for three times. The precursors were dried at 80 °C for over 12 h to obtain CoNiO₂ microspheres.

3. Synthesis of CNO@CTP

After the CTP and CoNiO₂ microsphere were mixed evenly by ball milling, the obtained samples were dissolved in absolute ethanol and stirred for 12 h and processed by ultrasound. After centrifugation and washing, the powder was dried at 60°C for 6 h, and then calcined in N₂ at 610°C for 3 h with a rising rate of 5°C min⁻¹ to acquire CoNiO₂@CTP powder. The proportion of CTP in the total sample is 0%, 3%, 5% and 10%, respectively, and the obtained samples are denoted as CNO, CNO@CTP3, CNO@CTP5 and CNO@CTP10.

3 Materials characterization

The crystalline structure of all samples was analyzed by X-ray diffraction (XRD, a Rigaku instrument with Cu K α radiation, $\lambda=0.15406\text{nm}$) with the scanning angle range of 2θ between 10°-90°. Raman spectrometer (Thermo Fischer DXR) was applied to test the molecular structure of the samples. The morphology and microstructure of the samples were observed by a ZEISS SUPRA 55 field emission scanning electron microscope (SEM), transmission electron microscopy (TEM, Talos F200X) and a high-resolution transmission electron microscope (HRTEM, JEOL, JEM-2100), respectively. X-ray photoelectron spectroscopy (XPS, ESCALAB 250Xi, Thermo Fisher Scientific) with an Al K α radiation was employed to investigate the valence states and bonding conditions of elements. The energy-dispersive spectroscopy (EDS) element mapping was performed using a JSM-6490LV instrument.

4 Electrochemical measurements

CR2025 coin-type cells were employed to investigate the electrochemical performance of all samples. The homogeneous slurry containing active material, acetylene black, polyvinylidene fluoride (PVDF) (80:10:10 in weight) and N-methylpyrrolidone (NMP) were painted on a copper foil and desiccated at 80 °C overnight under vacuum to form the working electrode. The loading mass of active material is about 2.0-2.5 mg cm⁻². The half-cell consists of working electrode and lithium foil, which separated by a porous polypropylene film, and 1 mol L⁻¹ LiPF₆ in EC: DMC (1:1 in volume) as the electrolyte. The cyclic voltammetry (CV) test was performed on a CHI 1000C (Shanghai, China) electrochemical workstation at a scanning rate of 0.5 mV s⁻¹ between 0.01 and 3.0 V. Electrochemical impedance spectroscopy (EIS) studies were carried out on a Zennium-E41103 electrochemical workstation with a frequency of 100 kHz to 0.01 Hz. The galvanostatic charge-discharge performance were conducted between 0.01 and 3.0 V (vs. Li⁺/Li) on Neware CT-4008T battery testing system (Shenzhen, China).

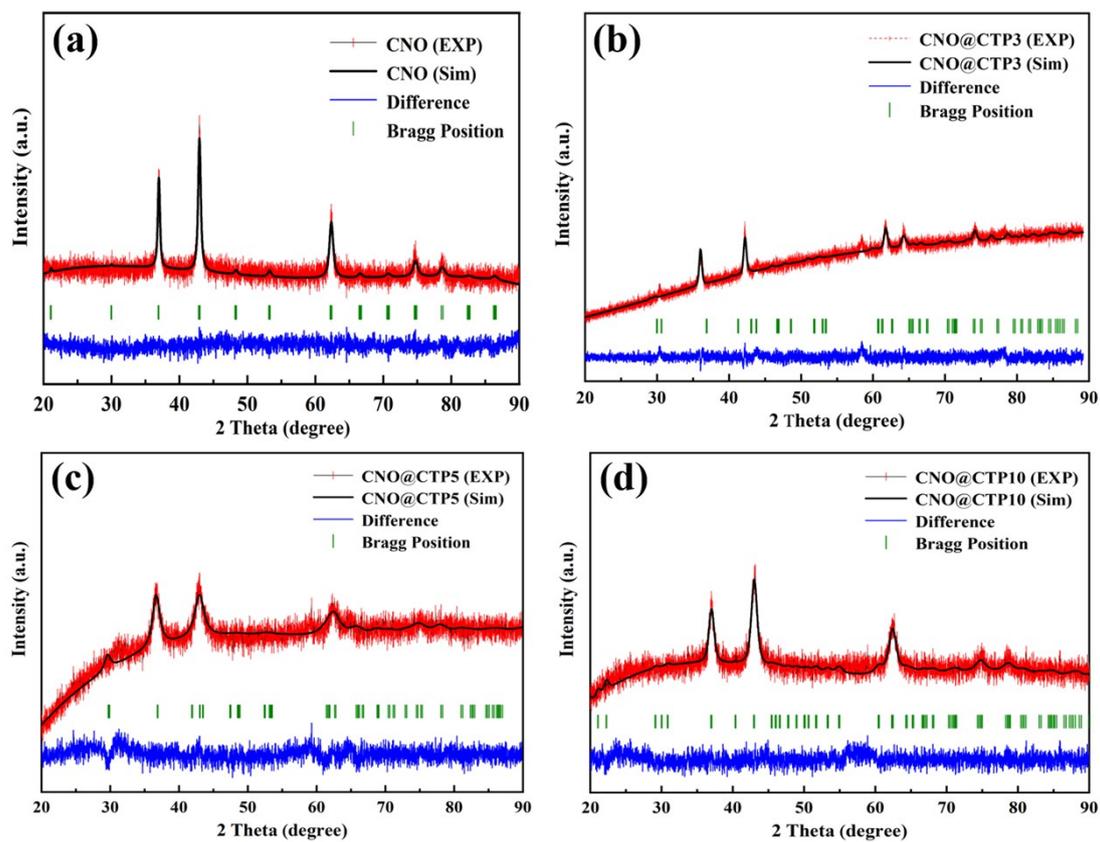


Fig. S1 The Rietveld refinement for samples prepared with (a) CNO, (b) CNO@CTP3, (c) CNO@CTP5 and (d) CNO@CTP10.

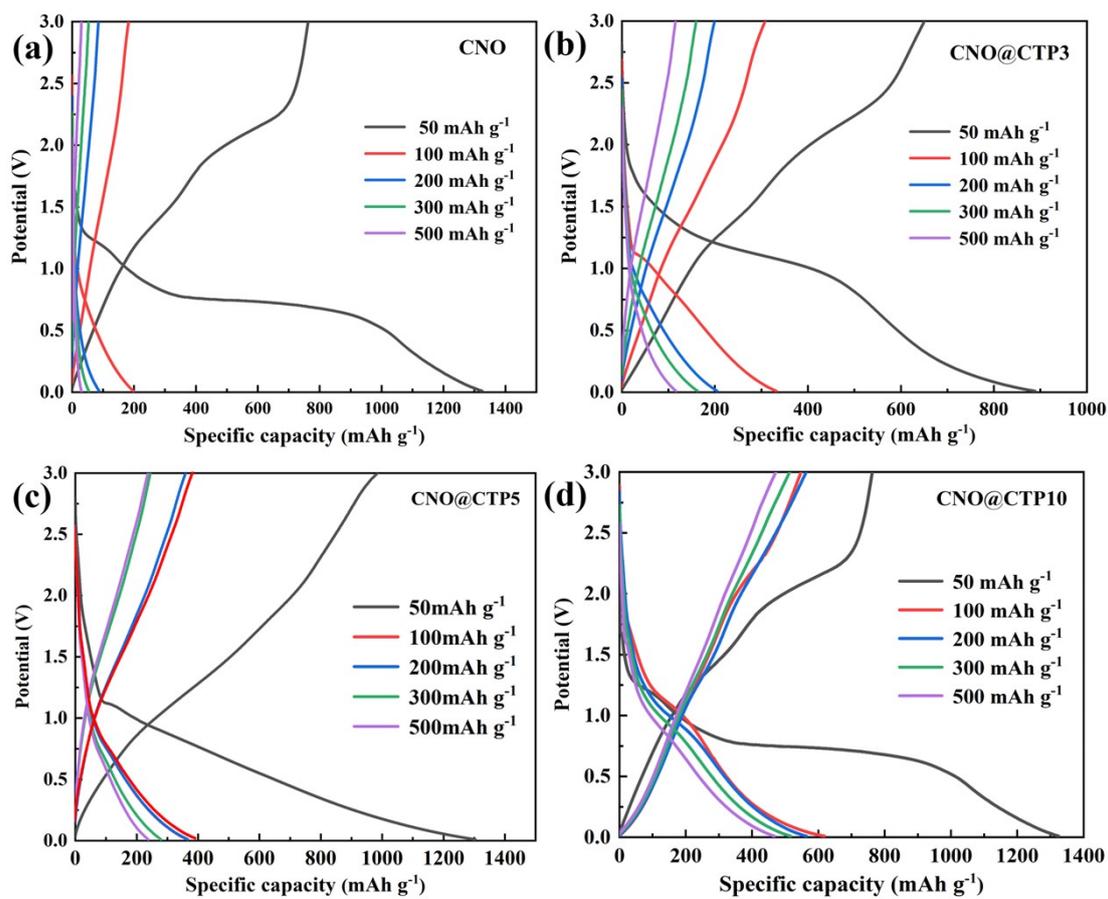


Fig. S2 GCD profiles of (a) CNO, (b) CNO@CTP3, (c) CNO@CTP5 and (d) CNO@CTP10 electrodes at different current densities.

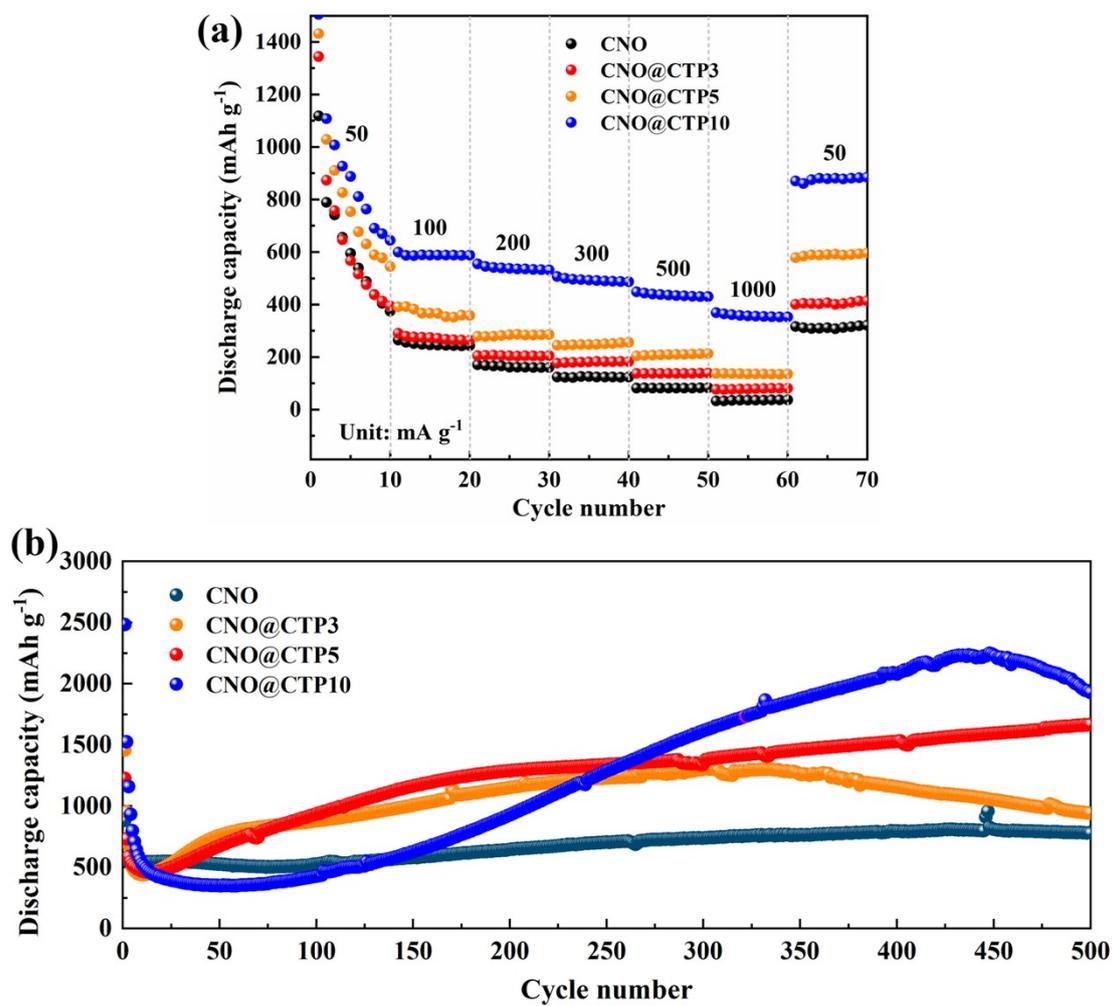


Fig. S3 (a) Rate capability. (b) Cycling property of pure CNO and CNO@CTP10 composites at 500 mA g⁻¹.

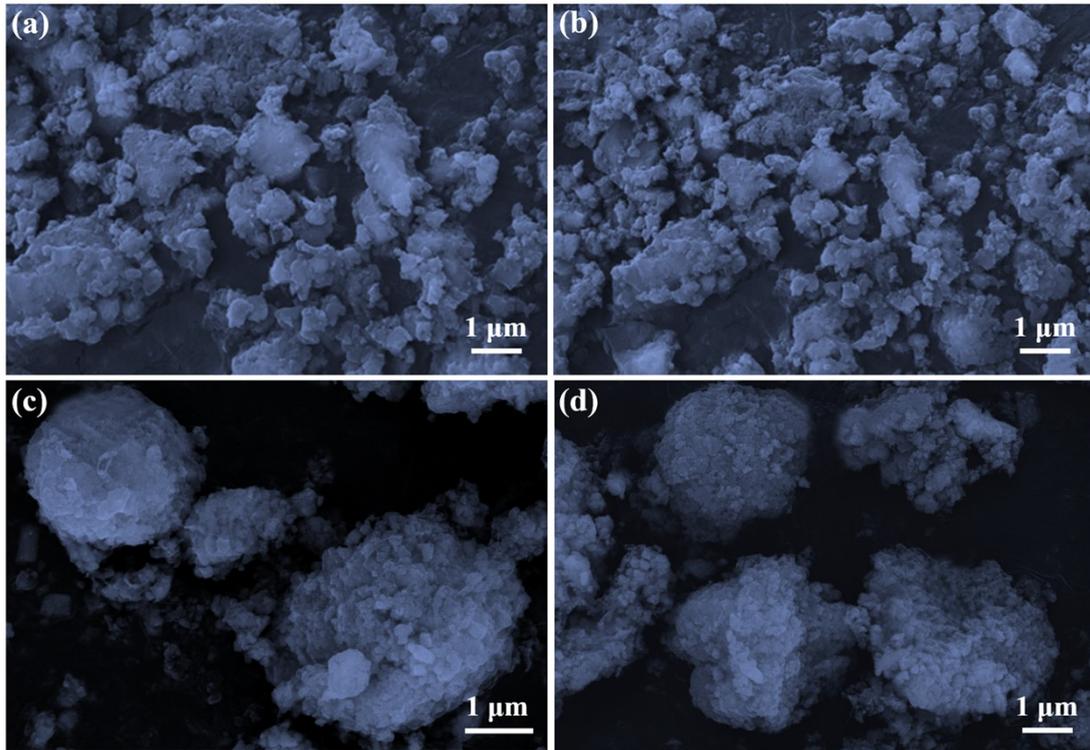


Fig. S4 SEM images of of (a,b) CNO and (c,d) CNO@CTP10 at 500 mA g⁻¹ after 500 cycles.