Electronic Supplementary Information (ESI)

A single-crystalline Co_3O_4 nanoparticles-assembled three-dimensional chain as ultra-stable magnesium-ion battery cathode under different temperatures

Qian Ding[a], Tianli Han[a], Xirong Lin[b], Ting Zhou,[a] Jinyun Liu[*a] and Huigang Zhang[*c]

Experimental

Synthesis of Co_3O_4

All chemicals were used directly without further purification and were purchased from Aladdin. Ultrapure water was used in all synthesis processes. The 3D hierarchical Co_3O_4 was synthesized by a hydrothermal method. In a typical process, 1.76 g $Co(CH_3COO)_2.4H_2O$, 3.0 g $CO(NH_2)_2$ and 5 g dimethyl diallyl ammonium chloride were dissolved in 55 mL ultrapure water at room temperature. After stirring for 5 min, a uniform pink solution was obtained. The mixed solution was transferred to a 50 mL Teflon-lined high-pressure reactor and heated at 120 ℃ for 12 h. After reaction, it was cooled to room temperature, and the sample was collected, washed by alternating centrifugation with water and ethanol, and dried at 80 ℃ overnight. Finally, the sample was calcined in an air atmosphere at 450 ℃ for 2 h by using a ramp rate of 5 ℃ min^{-1}.

Characterization

The samples were characterized by scanning electron microscopy (SEM, Hitachi S-4800) and transmission electron microscopy (TEM, HT-7700). X-ray photoelectron

spectroscopy (XPS, ESCALAB 250Xi) was used to analyze the composition, and X-ray diffraction (XRD, Bruker D8 Advance) was used to study the crystalline structure. The lattice spacing was studied on high-resolution TEM (HRTEM). Energy dispersive X-ray spectroscopy (EDS), and elemental mapping was employed for measuring elemental distribution. The surface area and pore-size distribution were measured by using Micrometrics ASAP 2460 analyzer.

Electrochemical tests

Electrochemical performance was investigated by using a 2032-typed button cell system. The active material (60 wt%), conductive carbon black (30 wt%) and polyvinylidene fluoride (PVDF, 10 wt%) were evenly mixed and dispersed in n-methylpyrrolidone (NMP, 6.54 wt %). Then, the uniform black slurry was evenly coated on the carbon paper carbon paper collector by scraping method, dried in vacuum for 12 h, and cut into discs with a diameter of 12 mm. The 0.4 M $(PhMgCl)_2$-$AlCl_3$/THF was used as electrolyte. The Cycling experiments were tested on a battery tester (NEWARE, CT-4008), and cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) were performed on an electrochemical workstation (CHI-660E).

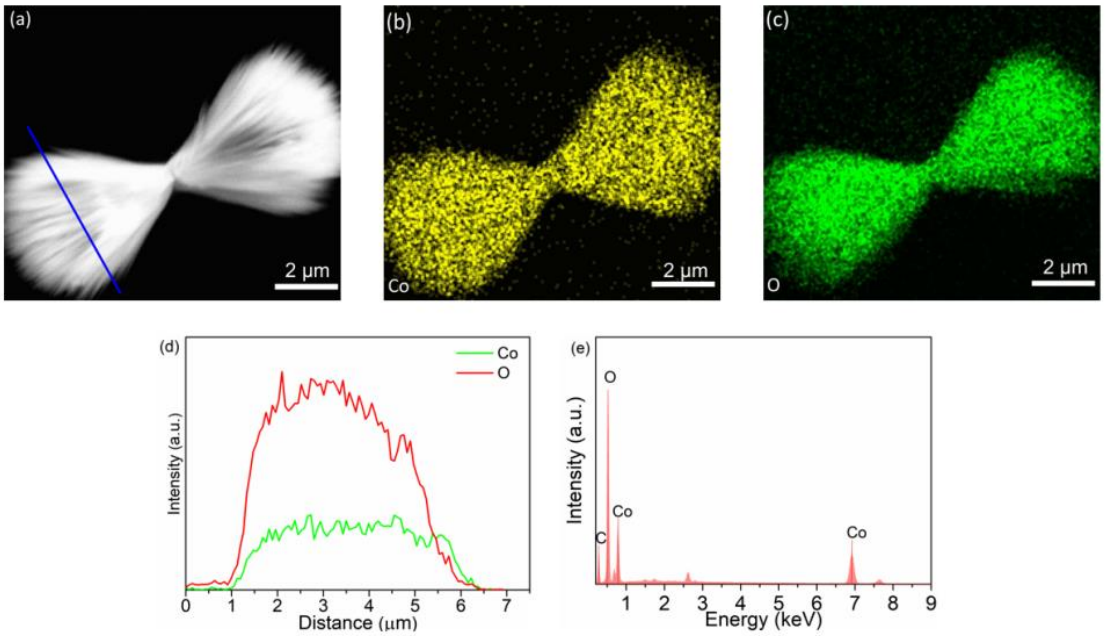

Fig. S1 (a) SEM and (b-c) mapping images of Co_3O_4. The line on (a) indicates the scanning path. (d) line-scanning curves and (e) EDS spectrum.

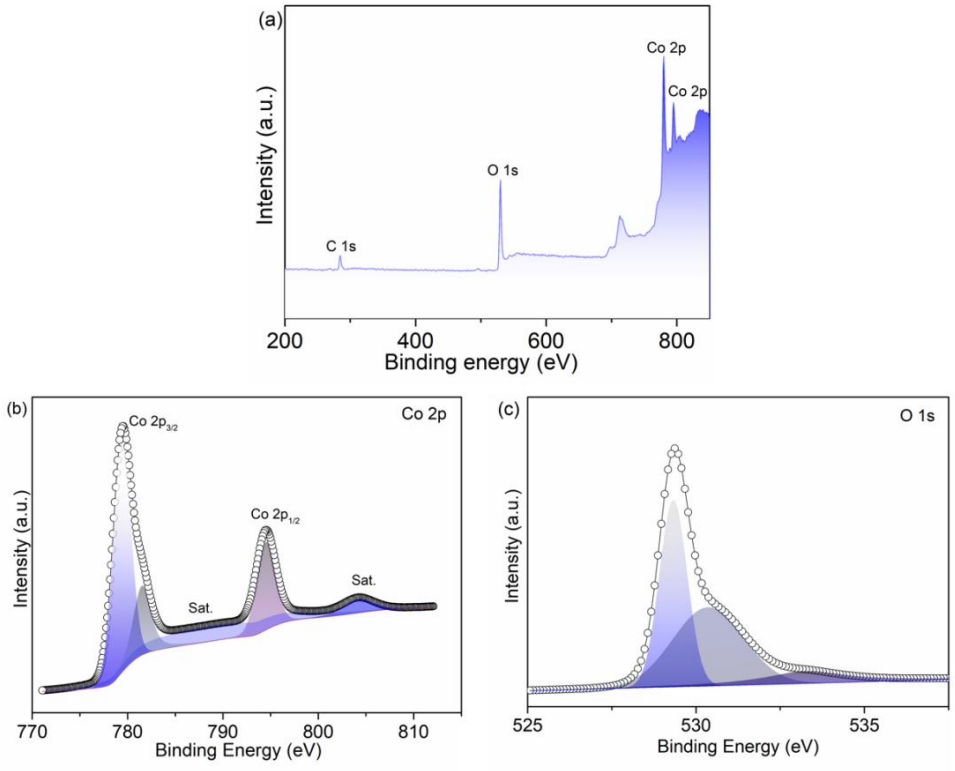

Fig. S2 XPS spectra of Co_3O_4: (a) survey spectrum, (b) Co 2p and (c) O 1s.

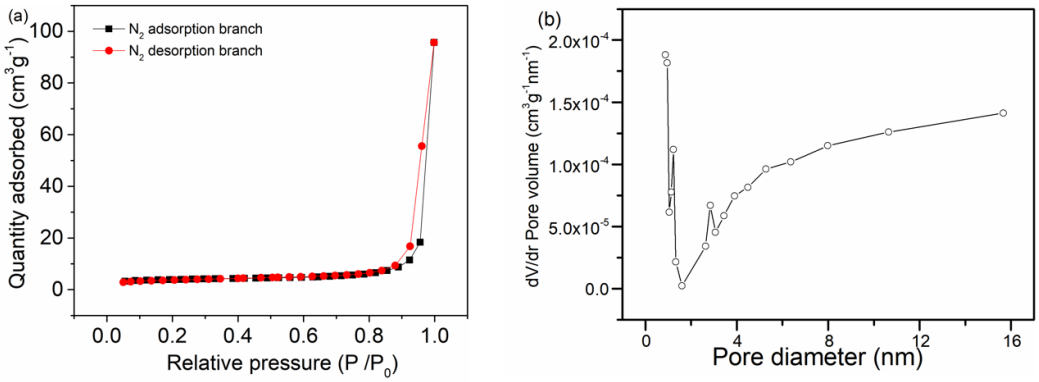

Fig. S3 (a) Adsorption-desorption isothermal and (b) pore-size distribution of Co_3O_4.

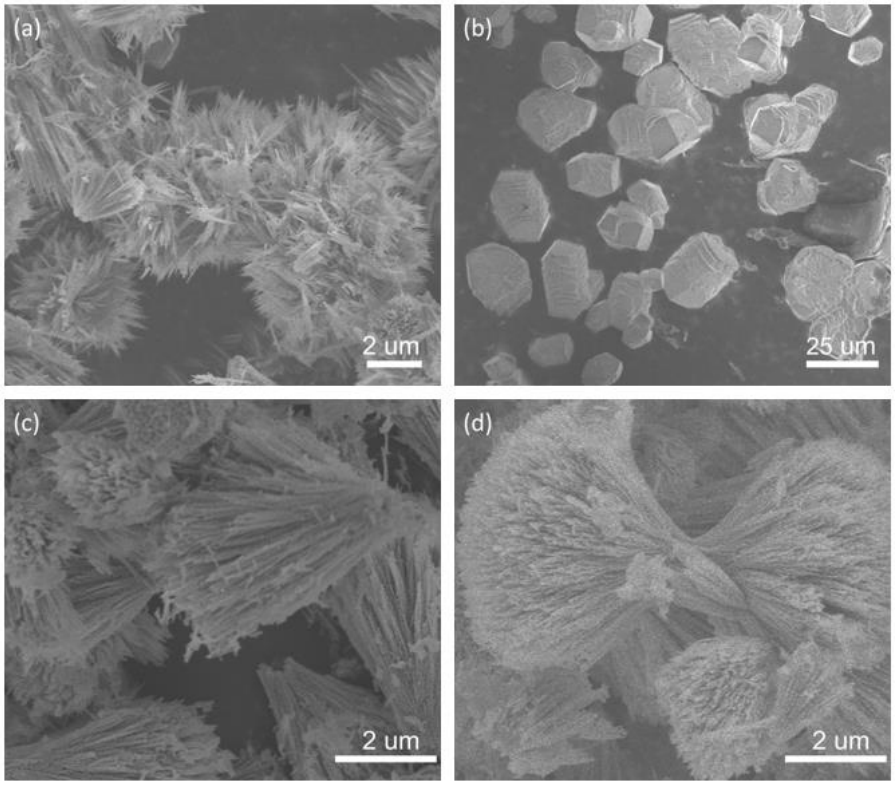

Fig. S4 SEM images of the Co_3O_4 prepared under different hydrothermal conditions: (a) 100 ºC and (b) 140 ºC for 12 h; 120 ºC for (c) 10 and (d) 14 h.

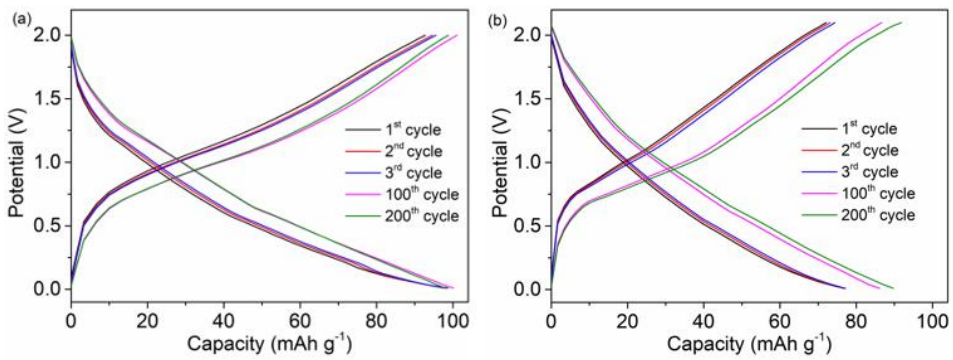

Fig. S5 (a) Charge-discharge profiles of 3D single-crystalline nanoparticles-chained Co_3O_4 charging at 100 mA g^{-1} and discharging at 200 mA g^{-1}. (b) Charge-discharge profiles of Co_3O_4 charging at 200 mA g^{-1}, and discharging at 100 mA g^{-1}.

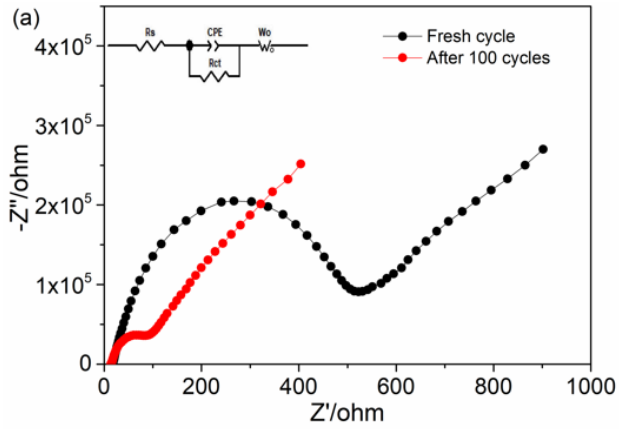

Fig. S6 EIS spectra of the Co_3O_4 cathode before and after cycling at 100 mA g-1.

Table S1. Comparison on the performance of some nanocomposite-based cathodes.

Material	Preparation approach	Rate (mA g^{-1})	Cycle number	Capacity (mAh g^{-1})	Ref.
F-doped TiO_2	Hydrothermal synthesis	20	50	63.7	1
CNT@VS_4	Solvothermal method	500	800	76.3	2

$MgMn_{1.8}Sr_{0.2}O$	Self-propagating combustion	100	10	59	3
$CuCo_2S_4$/CuS@MWCNTs	Hydrothermal synthesis	300	1000	38.7	4
MnO_2/MXene-V_2C	Etching method	100	100	76.7	5
Co_3O_4	Hydrothermal synthesis	100	200	111.7	This study

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