

Supporting Information

High-performance Li-ion battery driven by hybrid Li storage mechanism in three-dimensional architected ZnTiO₃/CeO₂ microsphere anode

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1. Materials synthesis

For the synthesis of ZnTiO₃-CeO₂ heterostructure, 2.19 g Zn(CH₃COO)₂ were dispersed in 50 mL ethylene glycol with continuous magnetic stirring for 24 h (solution A). At the same time, 3.4 g C₁₆H₃₆O₄Ti was dripped into 50 ml anhydrous ethanol and stirred for 12h to make it mix evenly (solution B). Then, solution A was slowly poured into solution B, and added Ce(NO₃)₃·6H₂O until uniformly mixed (solution C). The quality of Ce(NO₃)₃·6H₂O is quantified according to the proportion of the coating, the coated samples of 3 wt%, 6 wt% and 9 wt%, were recorded as ZTO-CO1, ZTO-CO2 and ZTO-CO3 in order. Subsequently, the as-prepared solution C was transferred into a 100 mL Teflon-lined stainless-steel autoclave after stirring for 24 h and heated at 180 °C for 6 h. The obtained product was collected and washed with ethanol for several times and subsequently dried at 70 °C overnight. Then, the as-prepared ZnTiO₃-CeO₂ composites were calcined at 600 °C for 3 h under atmosphere. Meanwhile, bare ZnTiO₃ materials as the baseline were prepared by a similar synthetic route without Ce(NO₃)₃·6H₂O.

2. Material characterization and electrochemical measurement

The crystal structure of pure ZnTiO₃ and ZnTiO₃-CeO₂ were carried on X-ray powder diffraction (XRD) with Cu Ka radiation. The morphologies of the two synthesized materials were characterized via field emission scanning electron microscopy (ZEISS-SUPRA55 SAPPHIRE) and transmission electron microscopy (TEM, JEOL-2100F). The electrodes were fabricated by mixing 70 wt% active

materials, 20 wt% acetylene black and 10 wt% polyvinylidene, respectively, in NMP solution. The obtained slurries were coated onto copper foil, and then dried at 100 °C under vacuum for 12 h. The CR2025 coin cells were assembled in an argon-filled glove box by using lithium foil as the counter electrode and Celgard 2500 membrane as the separator and containing a liquid electrolyte of 1M LiPF₆ in EC and DMC (1:1, v/v). The batteries tests were performed on between 0.01 and 3.0 V. Cycle voltammetry (CV) at 0.5 mV s⁻¹ between 0.01 and 3.0 V and electrochemical impedance spectroscopy (EIS) in a frequency range of 10 mHz–100 kHz with AC voltage amplitude of 5mV were conducted on the VSP electrochemical workstation (Bio-logic, FRA).

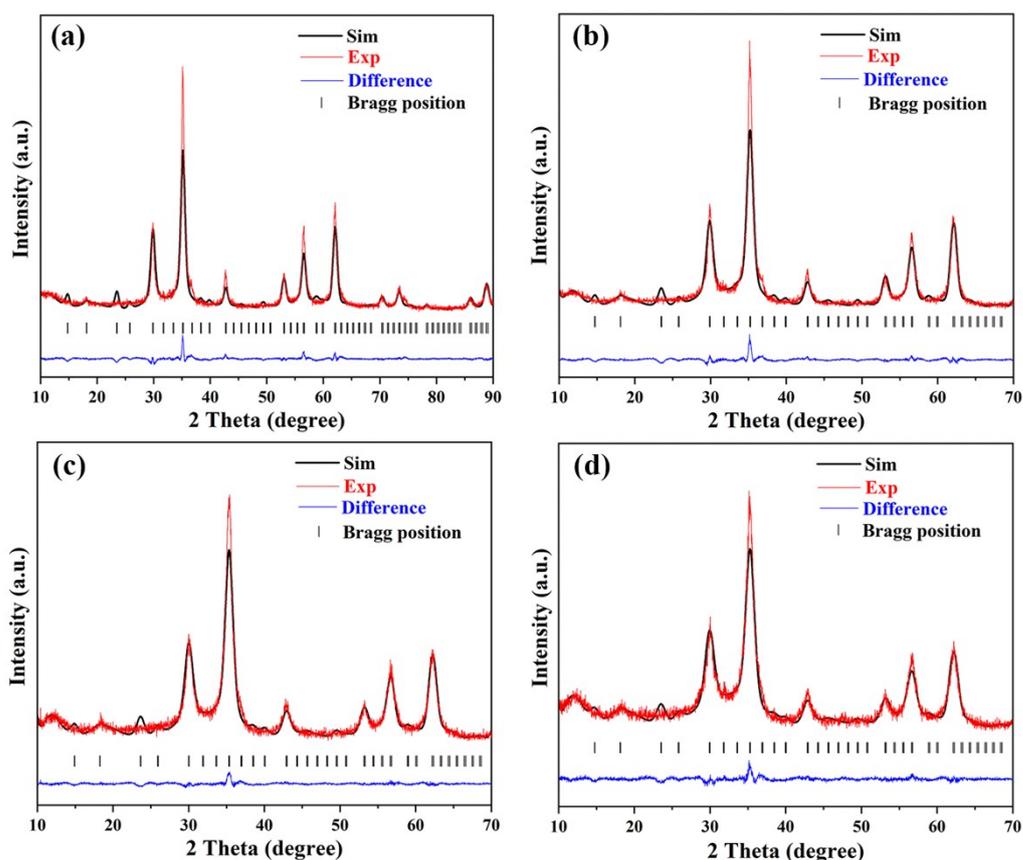


Figure S1. XRD Rietveld refinement profiles of (a) ZTO, (b) ZTO-CO1, (c) ZTO-CO2 and (d) ZTO-CO3.

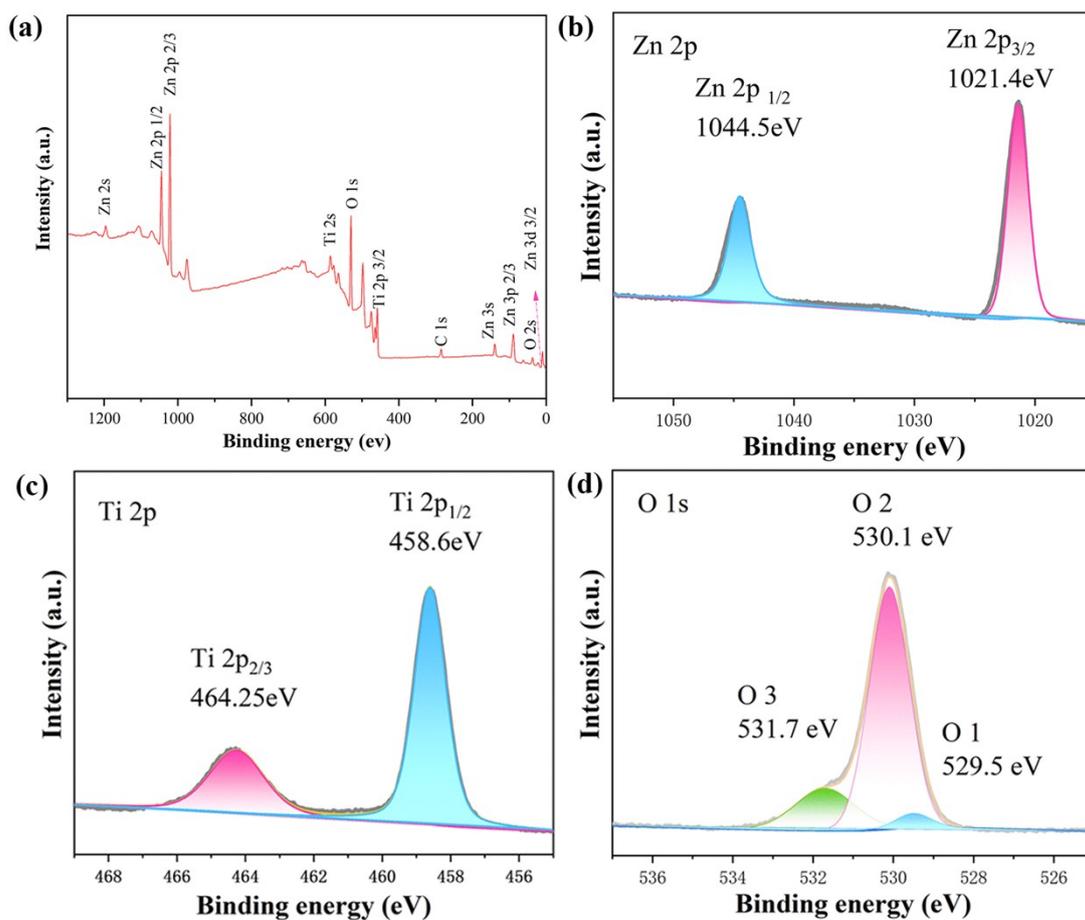


Figure S2. (a) XPS survey spectrum of ZTO and corresponding high-resolution XPS spectra of (b) Zn 2p, (c) Ti 2p and (d) O1s.

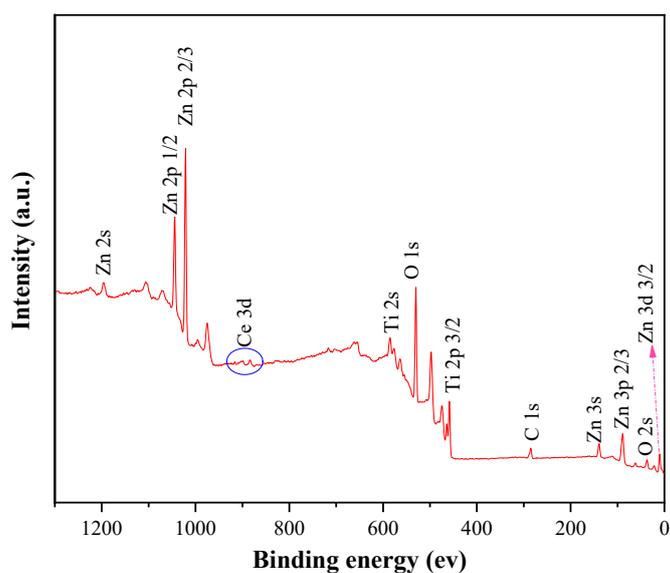


Figure S3. XPS survey spectrum of ZTO-CO2 composite.

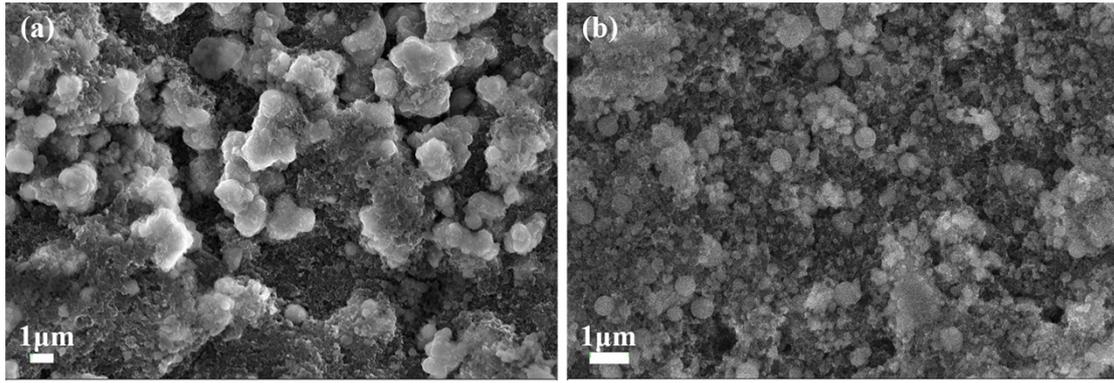


Figure S4. SEM images of (a) ZTO and (b) ZTO-CO₂ cycled at 0.5 A g⁻¹ after 500 cycles.