Supplemental Materials

A 700 Wh kg⁻¹ rechargeable pouch type lithium battery

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1. Experiments and Methods

Battery materials preparation. Ni_{0.12}Co_{0.12}Mn_{0.76}CO₃ precursor was purchased from Zhongwei New Materials Co., Ltd. Li₂CO₃ was purchased from Innochem Co., Ltd. They were mechanically mixed at a mass ratio of 1:1.4 and reacted at 500 °C for 5 hours under an oxygen atmosphere. The products were then preserved at 850 °C for 12 hours and annealed at 500 °C for 5 hours to obtain the target LRM. PVDF and LATP coating separator were purchased from Tianmulake Excellent Anode Materials Co., Ltd. The carbonate ester composed of 1M LiPF₆ in ethylene carbonate (EC) and dimethyl carbonate (DEC) (3:7 by volume) was used as the electrolytes. Both Li foils (with 20 μm lithium foil on 6 μm Cu both sides) and Li tablets (Φ16.5 mm, 1mm thickness) were received from the Tianqi Lithium Corporation.

Electrode fabrication and cell assembling. LRM electrode was prepared by coating the slurry on a 9 μm Al foil (one side coating of coin-cell and two sides coating of full-cell). The strict mass stoichiometric ratio of the slurry is LRM: Super P: polyvinylidene fluoride (PVDF) = 8:1:1. For the pouch cell, the ratio is 96: 2: 2. The area capacity of the LRM cathode is 10 mAh cm⁻² (calculated by 300 mAh g⁻¹). Then the coin-cell electrode was cast into Φ12 mm tablets, and the pouch cell electrode was punched into rectangular pieces (73×60 mm). All electrodes were dried in a vacuum oven for 24 h (120 °C) before assembly. Li foils are used in pouch cell and Li tablets (Φ16.5 mm, 1mm thickness) are used in coin-cells. Li foils were punched into 75×62 mm rectangular pieces in the dry room (dewpoint -60 °C) before assembly. CR2032-type coin cells were assembled to evaluate the cycling performance and energy density of LRM and Li material systems. All cells were assembled/disassembled in a glovebox filled with argon gas (H₂O and O₂< 0.1ppm). Pouch cells were produced in a dry room (dewpoint -60 °C) by using a semi-automatic cell-manufacturing line, including the process of mixing, coating, drying, rolling, cutting, lamination, welding, electrolyte injection, and packaging.

Measurement and characterization. The LRM | Li coin cells were cycled in a series of different voltage ranges: 2.0-4.8 V, 1.5-4.8 V, 1.25-4.8 V, and 1.0-4.8 V. The Li | Cu cells were cycled under a 1 mAh cm⁻² capacity and a 0.5 mA cm⁻² current density. The Li | Li cells were cycled under 1 & 10 mAh cm⁻² specific surface capacities and a 0.5 mA cm⁻² current density. All pouch cells were sandwiched in a bakelite clamping device and tested by the Land BA2100A Battery Test System (Wuhan, China) 0.05C in the first cycle and 0.1C in subsequent cycles under 25°C (1C = 10 mAh

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cm⁻²). The cells showed in Figure 4(a, b) were charged to 4.7 V for the first cycle and 4.8 V for rest cycles. Parallel cells showed in Figure S3 were cycled at 1.25 V-4.78 V for all cycles. All the energy density of batteries was calculated based on the data of the first discharging process. X-Ray Diffraction (XRD, Bruker, D8 ADVANCE) was performed to analyze the structural stability of LRM cathodes under various lithiation states.

2. Supplemental Figures and Captions

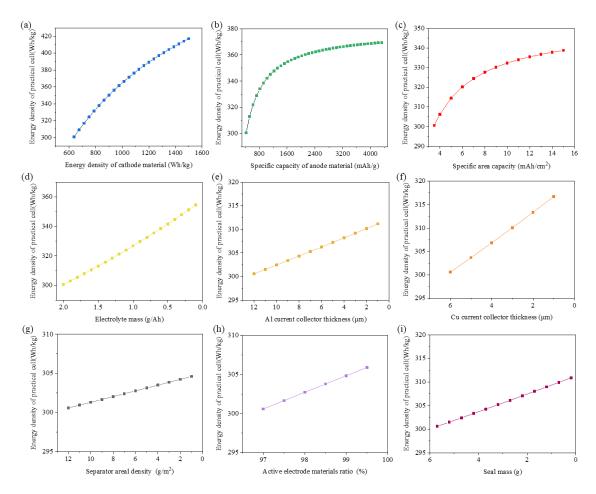


Fig. S1. The increase of energy density in practical lithium batteries by increasing active electrode materials' energy density and decreasing auxiliary materials mass. (a) energy density of cathode materials, (b) Specific capacity of anode materials, (c) specific area capacity, (d) the ratio of electrolyte mass and battery capacity, (e) cathode current collector thickness, (f) anode current collector thickness, (g) separator areal density, (h) active materials ratio in electrode paste, (i) packaging materials mass.

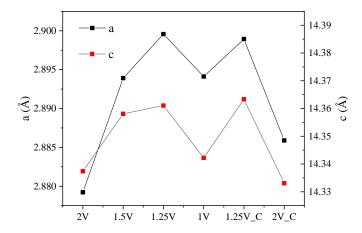


Fig. S2. The XRD refined results of LRM cathode at different cut-off voltage. The variation of a axis and c axis shows that the stability of structure at various cut-off voltage, 2V, 1.5V, 1.25V, 1V, 1.25 C (after 10 cycles), 2V C (9 cycles with 1.25V then stops at 2V).

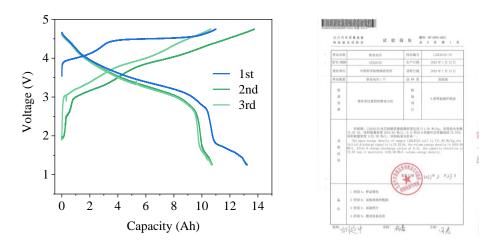


Fig. S3. Certificated results of the pouch cells by the China North Vehicle Research Institute.