## Electronic Supporting Information

# Suppressing the Voltage-fading of Layered Lithium Rich Cathode Materials via an Aqueous Binder for Li-ion Batteries

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## Experimental section

**Sample preparation** $Li_{1.14}Ni_{0.18}Mn_{0.62}O_2$ was synthesized by a molten salt method. All the raw materials were of analytical grade. $Li_2CO_3$, $Ni(CH_3COO)_2\cdot 4H_2O$ and $Mn(CH_3COO)_2\cdot 4H_2O$ were chosen as starting materials. 2.327 g $Li_2CO_3$ (5% excess to compensate the evaporation of Li at high sintering temperature), 2.489 g $Ni(CH_3COO)_2\cdot 4H_2O$, 7.353 g $Mn(CH_3COO)_2\cdot 4H_2O$ and a large excess of KCl salt were mixed by ball milling. The weight ratio of KCl of the target product was 10. The mixtures were calcined in alumina crucibles in a muffle furnace. The mixture was heated at 450 °C for 6 hours and further heated at 800 °C for 12 hours. The heating rate of each process was 5 °C/min. After cooling down to room temperature in the muffle furnace naturally, the product was washed 6 times with deionized water to remove the residual $K^+$ and $Cl^-$ and then filtered, and then dried at 100 °C in an oven for 12 hours.

## Materials characterization

The morphologies of the prepared materials were observed by field emission scanning electron microscopy (SEM) on a Zeiss Sigma operated at 15 kV. Transmission electron microscopy (TEM, JEOLJEM-2100 microscopy) was used to identify the morphologies and detailed structure information. Powder X-ray diffraction (XRD) patterns were acquired on a Rigaku D/Max-Ⅳ X-ray diffractometer employing Cu Kα radiation operated at 40 kV and 40 mA. Data were collected over 2θ range from 10

° to 90 ° at a scan rate of 1 °/min. Rietveld refinement was conducted by a PDXL program (Rigaku Corporation, PDXL 2.1). Inductively coupled plasma-optical emission spectrometry (ICP-OES, Thermo Electron IRIS Intrepid II XSP) was used to analyze the chemical composition. The dissolubility of guar gum (GG) is investigated by add certain amount of GG in the electrolyte (EC: DMC=1:1, 1M LiPF6).

## Electrochemical tests

The charge-discharge measurements were performed using CR2025-type coin cells. 80 wt% of the synthesized material, 10 wt% of carbon black (BLACK PEARL 2000) as conductive agent, and 10 wt% of guar gum (Hongjian, Shanghai, China) as binder were mixed in deionized water to get slurry. For comparison, LLRO material, carbon black and PVDF with a weight ratio of 8:1:1 were mixed in N-methyl-2-pyrrolidone (NMP) solvent to get slurry. Then the slurry was coated onto an aluminium foil and dried in vacuum for 12 hours at 100 °C. The diameter of the electrodes were 12 mm, and the mass loading of active material was kept at 1.5±0.2 mg. The CR2025-type coin cells were assembled in an argon-filled glove box using a lithium foil as counter electrode, Celgard2300 film as separator. The electrolyte solution was 1 M $LiPF_6$ in ethylene carbonate (EC)/dimethyl carbonate (DMC) (1:1 by volume) (Guangzhou Tinci Materials Technology Co., Ltd.). The cells were charged and discharged between 2.0 V and 4.8 V galvanostatically on a Land-

CT2001A battery tester (Wuhan, China) at 30 °C. The calculated capacities were based on the weight of active material. In order to determine the capacity and electrochemical activity of GG and PVDF, pure GG or PVDF were coated onto the aluminium foil and dried in vacuum for 12 hours at 100 °C. After that, CR2025-type coin cells using the electrodes with pure GG or PVDF were assembled, lithium foil as counting electrode. The coin cells were charged and discharged between 2.0 V and 4.8 V galvanostatically on a Land-CT2001A battery tester (Wuhan, China) at 30 °C. CV tests were carried out between 2.0 and 4.8 V with a scanning rate of 0.2 mV/s on a CHI660D electrochemical workstation (Chenhua, Shanghai, China) .

In order to characterize the cycled LLRO material, the cells cycled 200 times were disassembled in the argon-filled glove box. The separated electrodes were washed by DMC several times to remove the electrolyte, and then the electrodes were took out of the glove box and washed with ethanol several times.

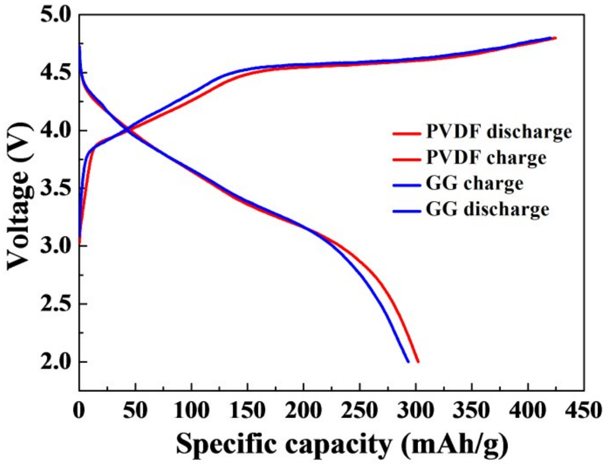

Fig. S1 The initial charge-discharge curves of the electrodes with PVDF or GG as binder (20 mA/g).

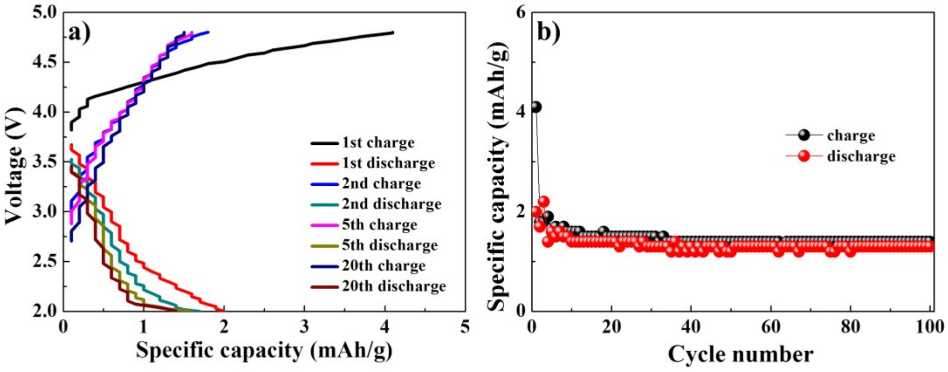

Fig. S2 The charge-discharge curves of the electrode with pure GG at 40 mA/g (a), the cycle performance of electrode with pure GG at 40 mA/g (b).

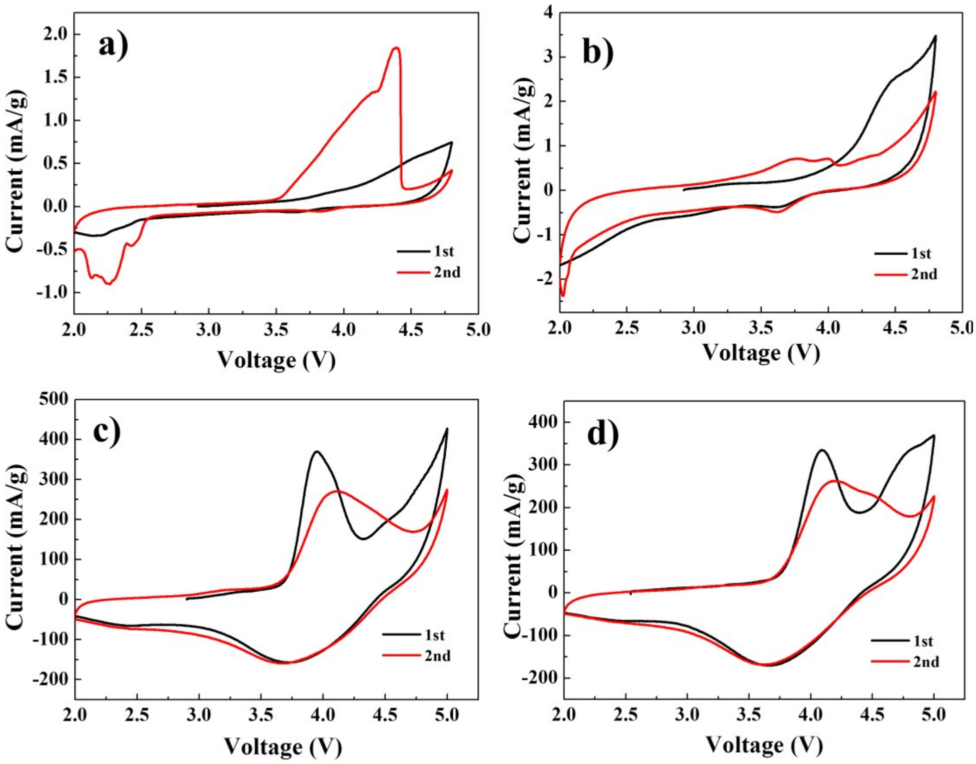

Fig. S3 CV curves of the pure PVDF binder (a) and GG binder (b) at scanning rate of 0.2 mV/s; CV curves of the electrodes with PVDF binder (c) or GG binder (d).

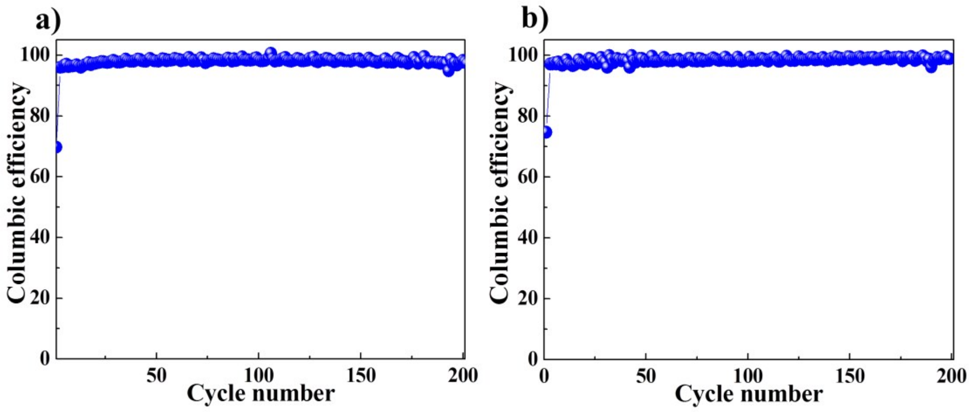

Fig. S4 The columbic efficiency of the electrodes with PVDF binder (a) or GG binder (b) at 100 mA/g.

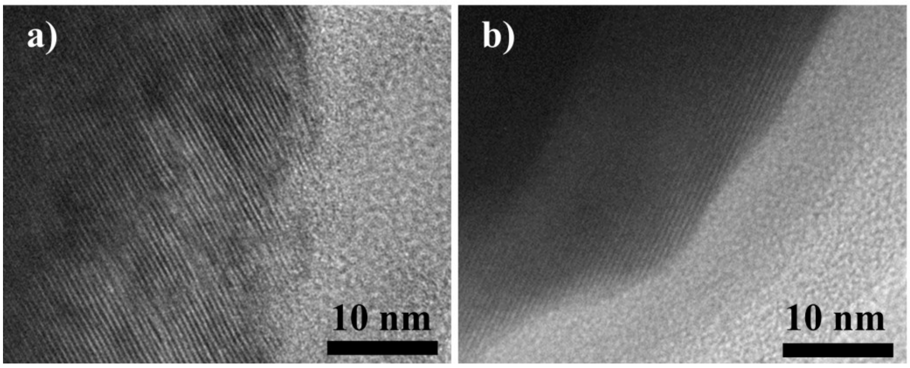

Fig. S5 The HRTEM images of the particles after 200 cycles with PVDF (a) or GG (b) as binder.