Electronic Supplementary Material (ESI) for ChemComm.
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Electronic Supplementary Information

for:

A Montmorillonite-Modification Strategy Enabling Long Cycling Stability of Dual-Ion Batteries

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Experimental Section

Preparation of Li-MMT. Firstly, commercial calcium-based montmorillonite and Li_2CO_3 (Sigma-Aldrich) were added to distilled water in a mass ratio of 50:3, stirred at 60 °C for 12 h, followed by centrifugation and finally drying at 100 °C for 12 h.

Materials Characterization. The samples were characterized by field-emission SEM (Hitachi SU8010), XRD (Bruker AXS D8-Focus, Cu), TEM (SUPRA 55, Germany), TGA (STA449F3) and XPS (ESCALAB 250XI). The energy-dispersive X-ray spectroscopy (EDX) mapping images of electrodes were collected using the JEOL-2100F Plus.

Preparation of Cathodes. MCMB and Li-MMT were firstly mixed by ultrasonic stirring in alcohol at a mass ratio of 8:1, and then dried at 80^{o}C to obtain the MCMB@MMT. The samples with MCMB and MMT mass ratio of 5:1 were also treated in the same way. The cathode slurry was made by mingling 80wt% sample, 10wt% carbon black, 10wt% polyvinylidene fluoride (PVDF, Sigma Aldrich, purity >99%) in N-methyl-2-pyrrolidone (NMP, Sigma Aldrich, purity >99%), which was then casted on an aluminum foil and then dried at 100^{o}C for 12 hours.

Electrochemical Measurements. The working electrode and Li foil were separated with a polypropylene membrane as the separator. Galvanostatic charge–discharge (GCD) was measured using a battery test system (Wuhan Landian Electronic Co., Ltd., China). CV were performed with a CHI660C electrochemical workstation. EIS was recorded on a Gamry Reference 3000. The electrolyte is 4 M $LiPF_6$ in EMC.

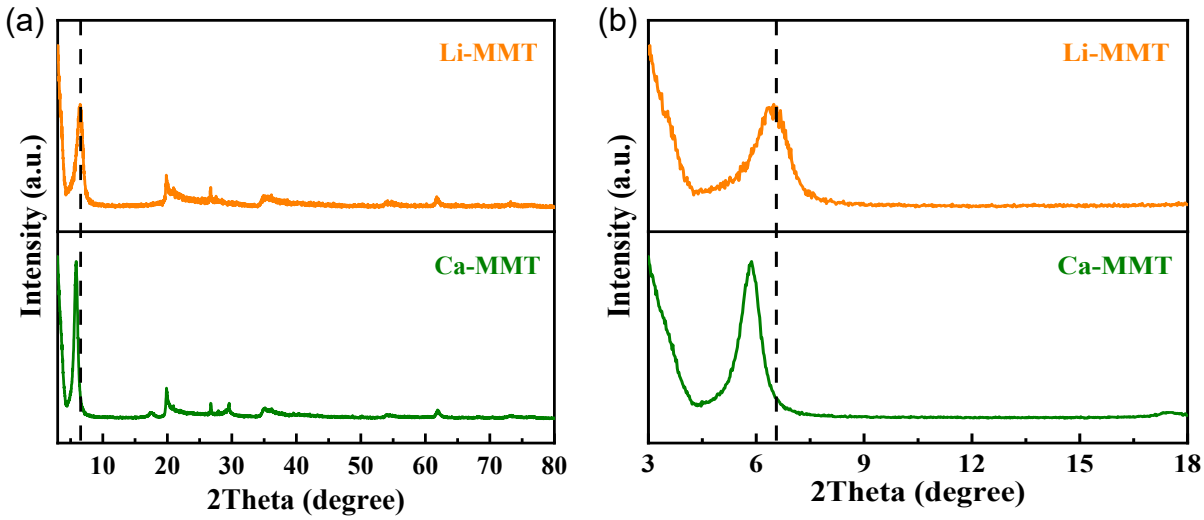

Figure S1. XRD patterns of Li-MMT and Ca-MMT.

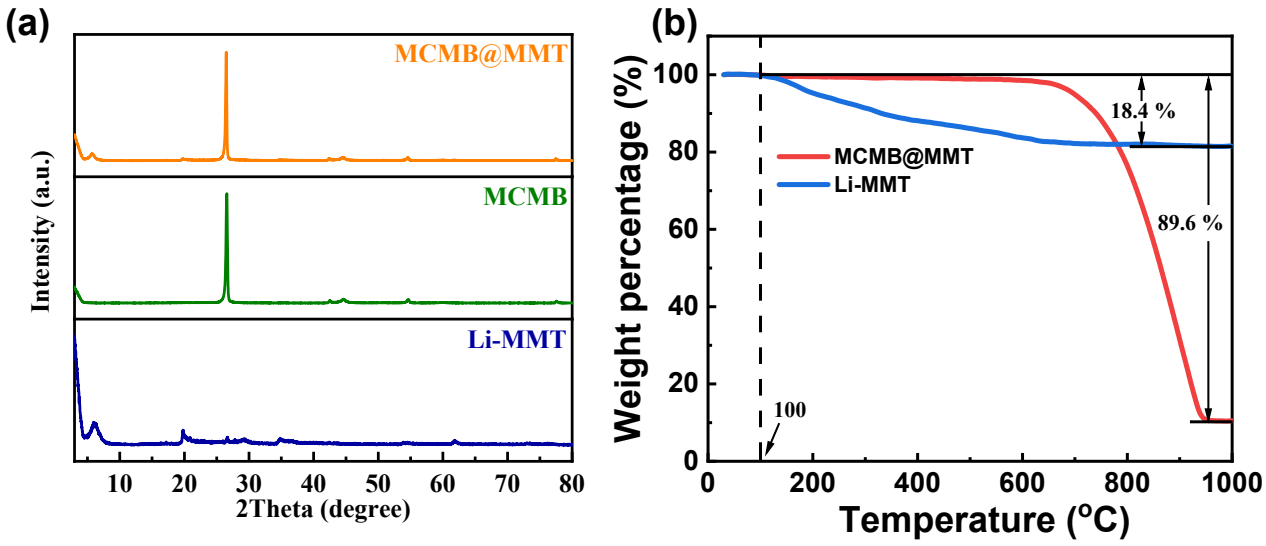

Figure S2. (a) XRD patterns of MCMB@MMT, MCMB and Li-MMT. (b) TGA and DSC curves of the MCMB@MMT and Li-MMT.

The TGA analysis indicates that the weight loss (18.4%) of MMT from 100 °C to 600 °C is mainly from the interlayer water and surface functional groups of MMT. MCMB@MMT shows the weight loss of 89.6% after 650 °C due to the combustion of MCMB and the decomposition of MMT. By calculation, the weight percent of MMT in the MCMB@MMT composite is determined to be 12.7% (**Figure 2b**, ESI†).

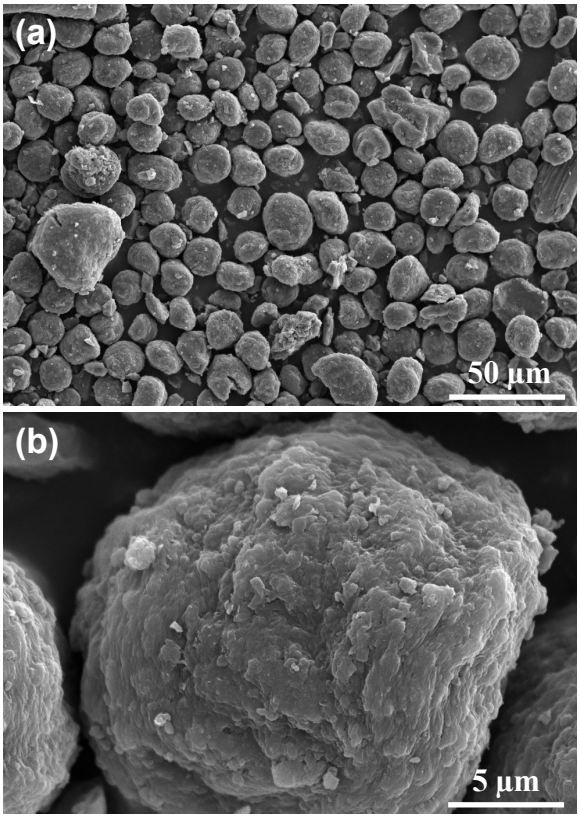

Figure S3. SEM image of MCMB.

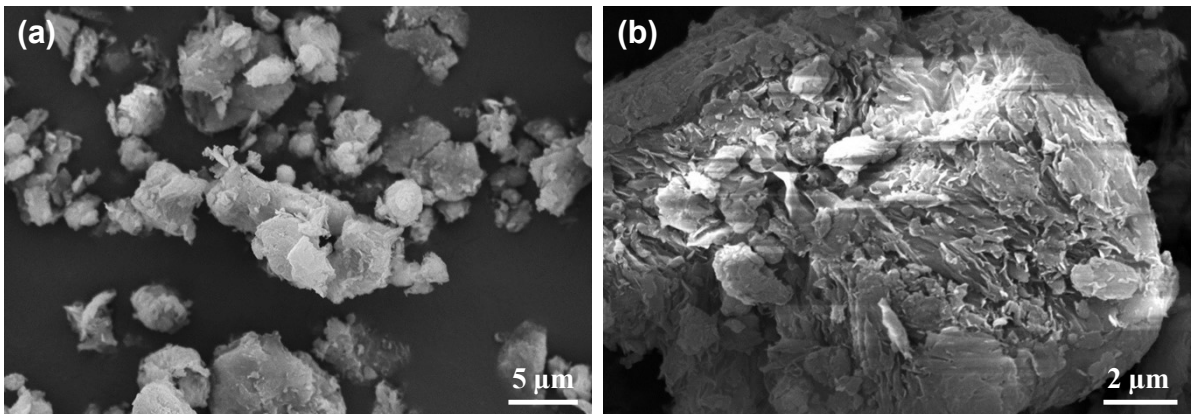

Figure S4. SEM image of Ca-MMT.

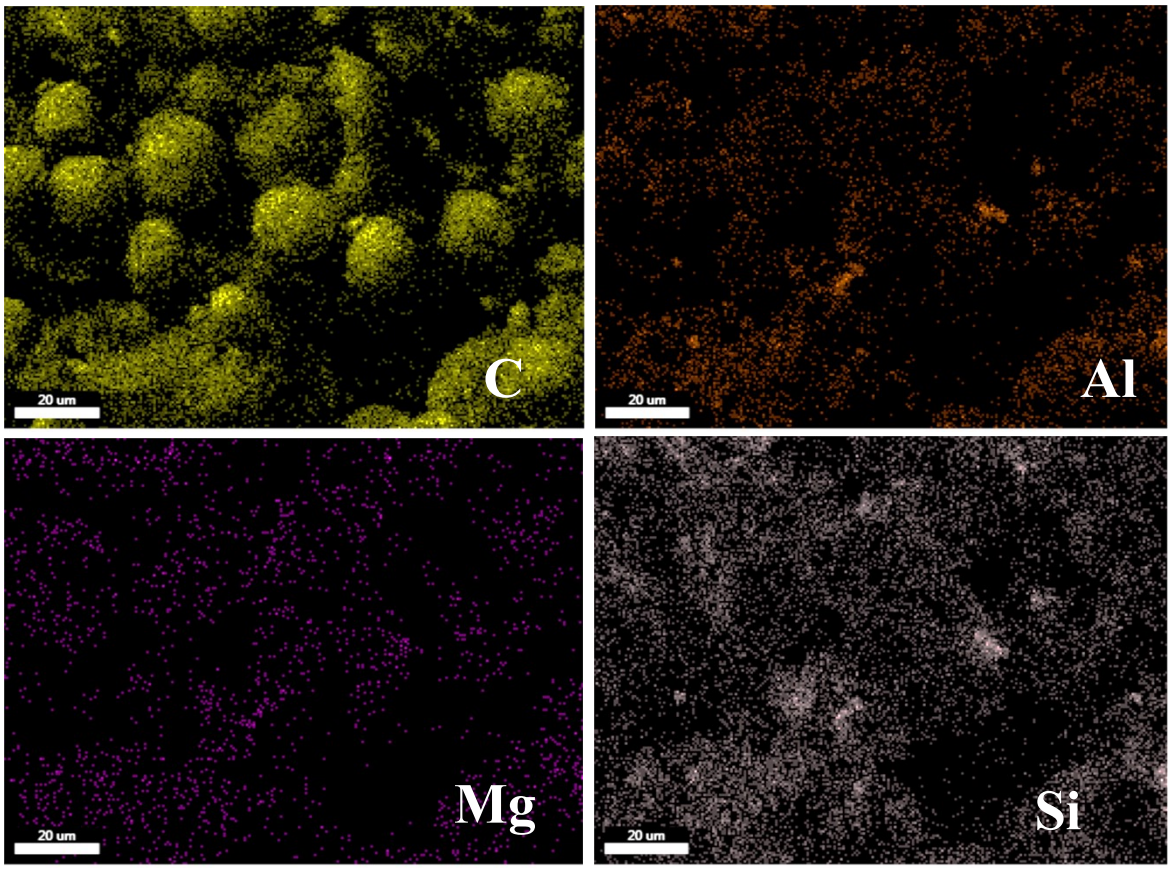

Figure S5. Element mappings of MCMB@MMT of C, Al, Mg, and Si.

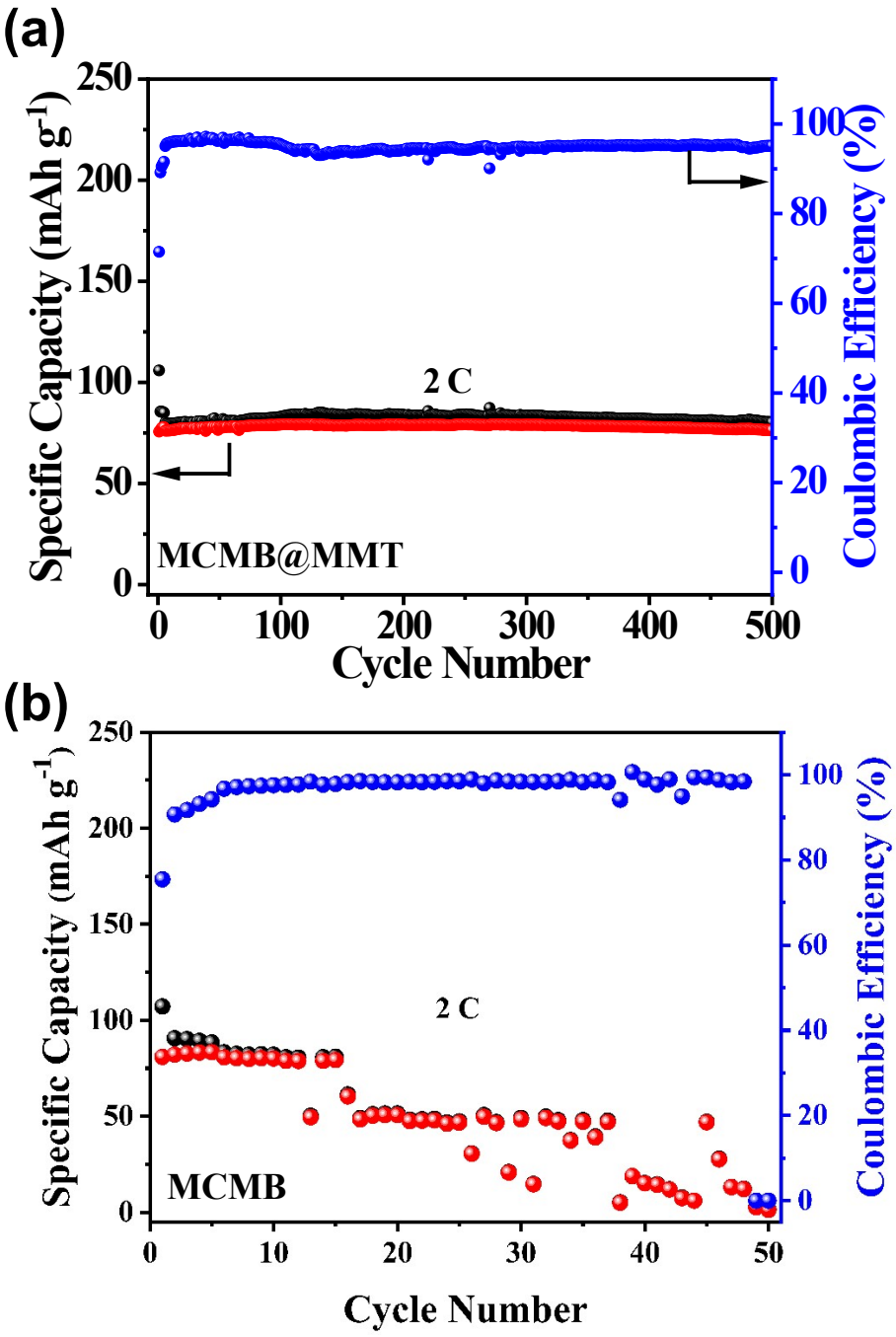

Figure S6. Cycling performance of (a) MCMB@MMT and (b) MCMB at a rate of 2 C.

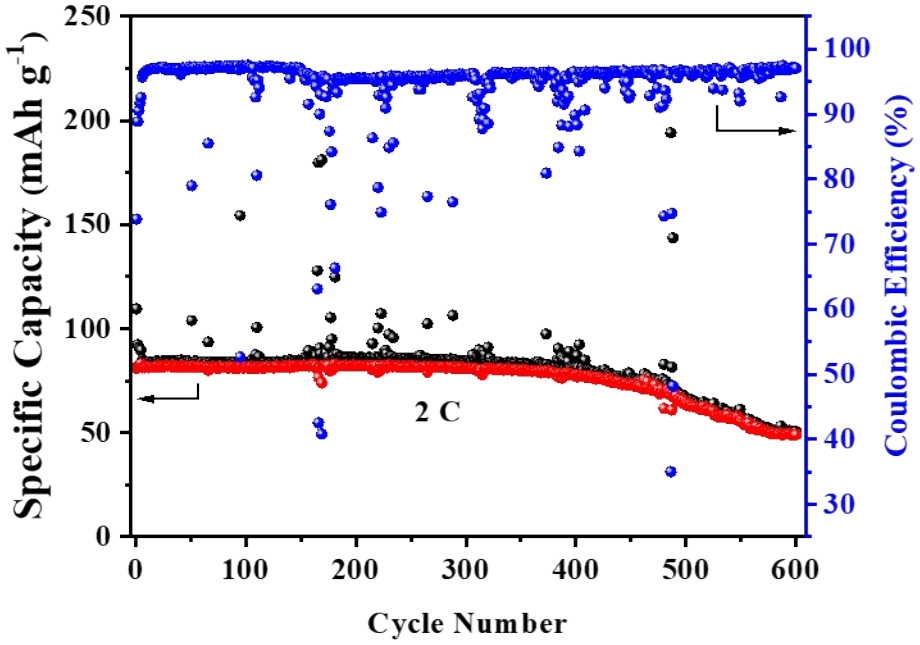

Figure S7. Cycling performance MCMB@MMT of at 2 C with the mass ratio (15 : 1) of MCMB and MMT.

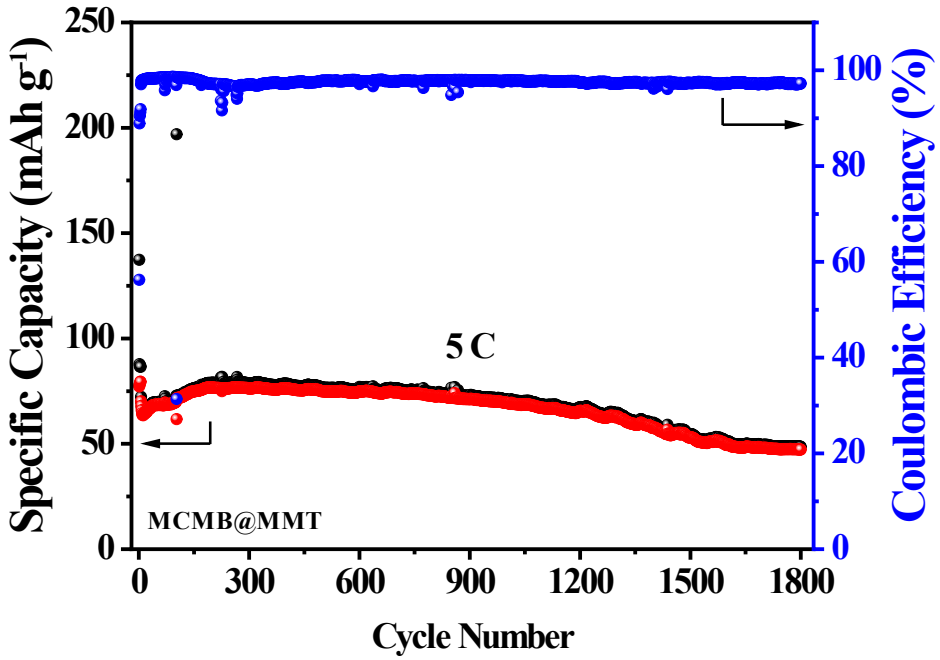

Figure S8. Long-term cycling performance of MCMB@MMT at a rate of 5 C.

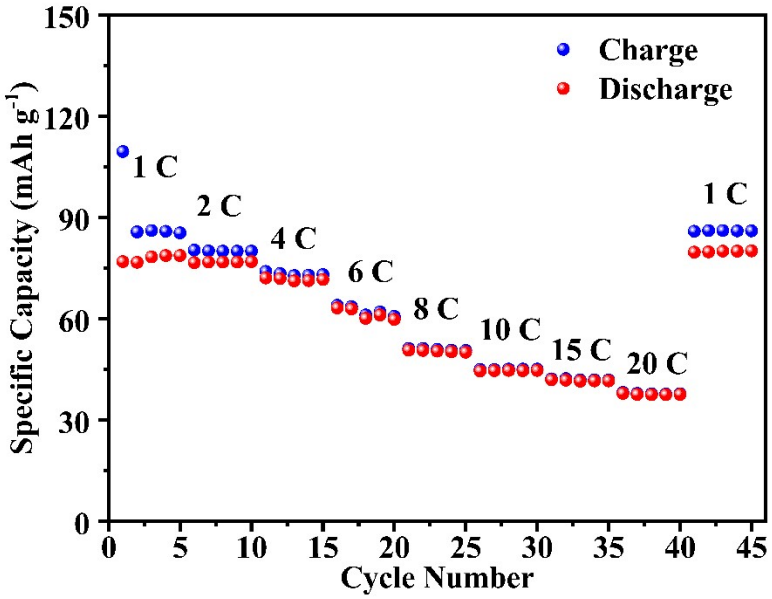

Figure S9. Rate capability of the MCMB@MMT cathode.

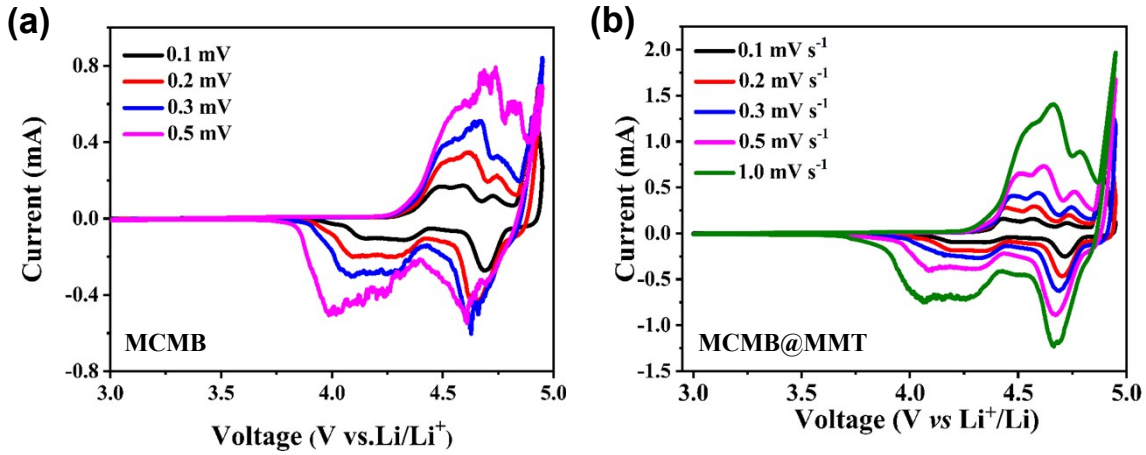

Figure S10. CV curves of (a) MCMB and (b) MCMB@MMT at various scan rates.

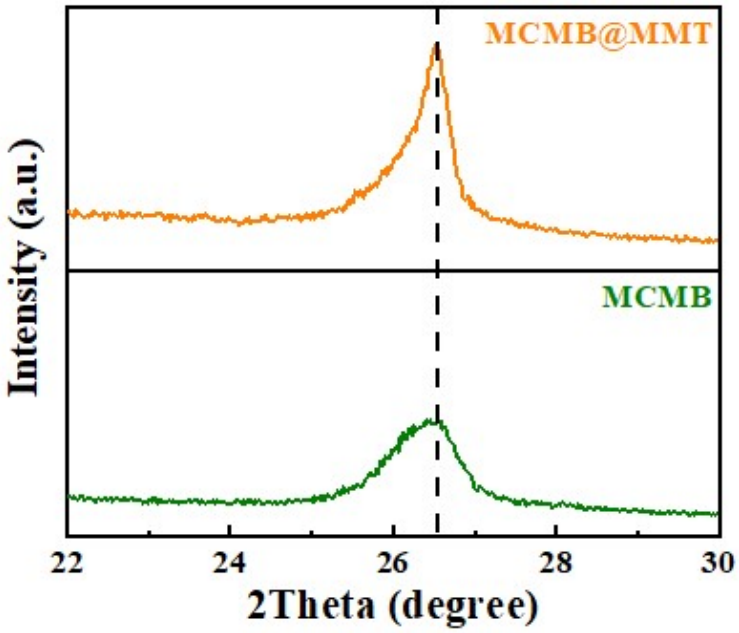

Figure S11. XRD patterns of the MCMB and MCMB@MMT electrodes after 50 cycles at 2 C.

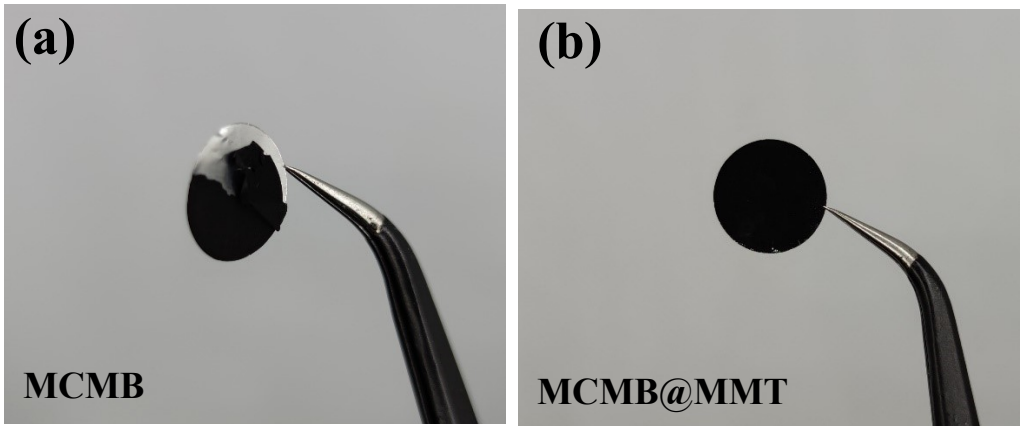

Figure S12. Photographs of the MCMB and MCMB@MMT electrodes after 50 cycles at 2 C.

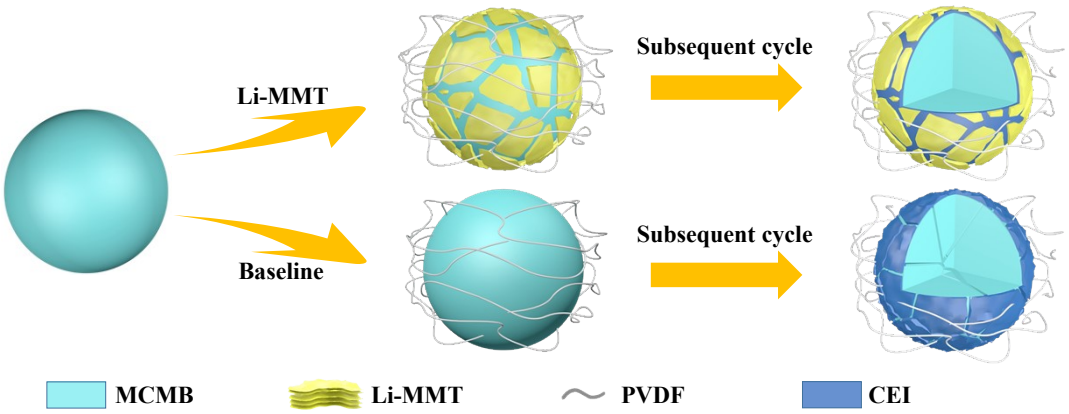

Figure S13. Illustration of the evolution of CEI during cycling with and without the MMT additive.