

## Supplementary Materials

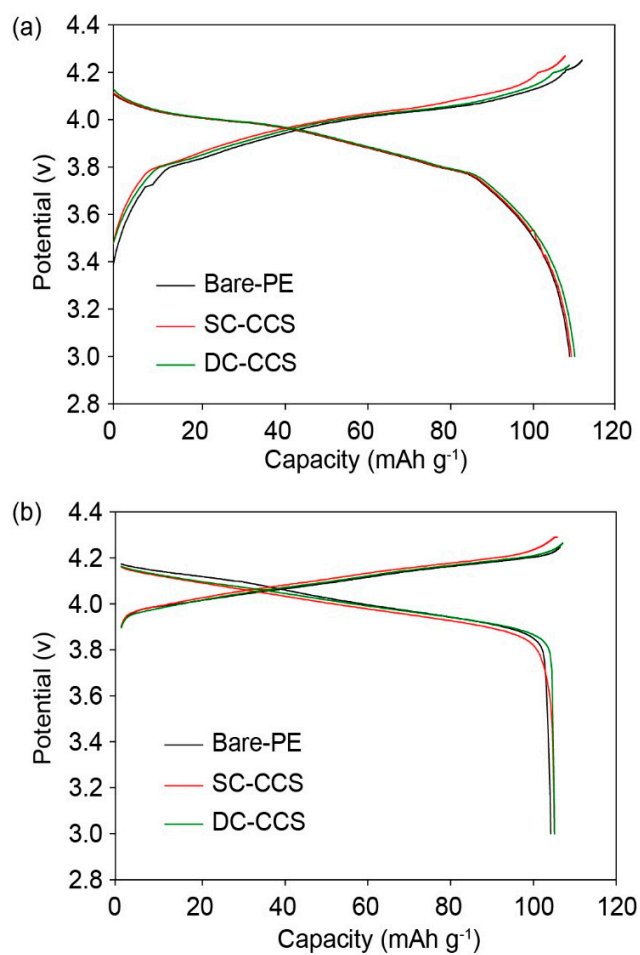
# Synergistic Effect of Dual-Ceramics for Improving the Dispersion Stability and Coating Quality of Aqueous Ceramic Coating Slurries for Polyethylene Separators in Li Secondary Batteries

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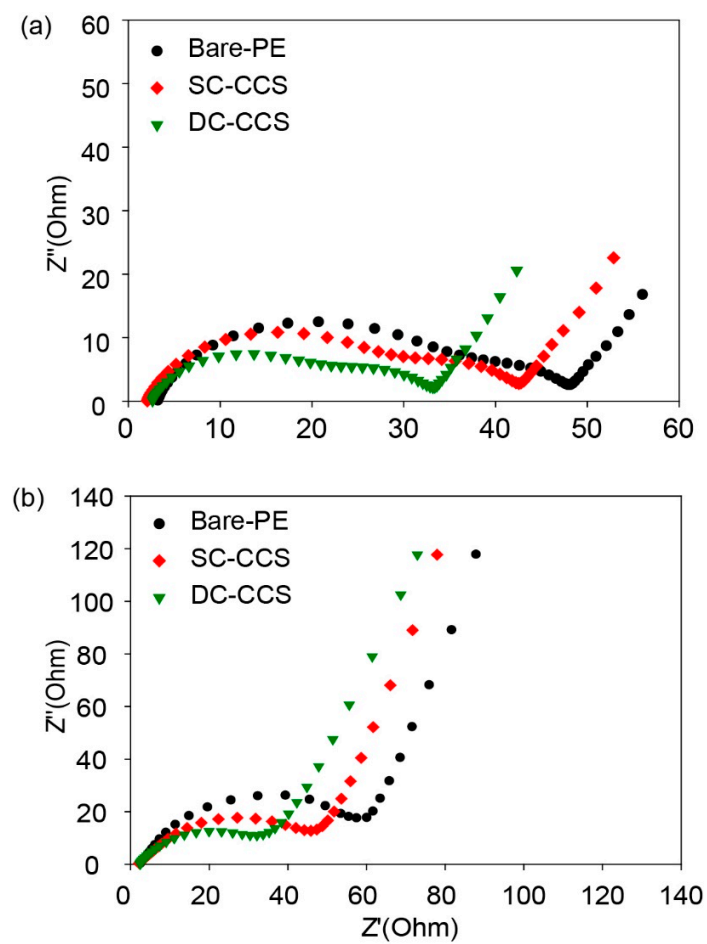
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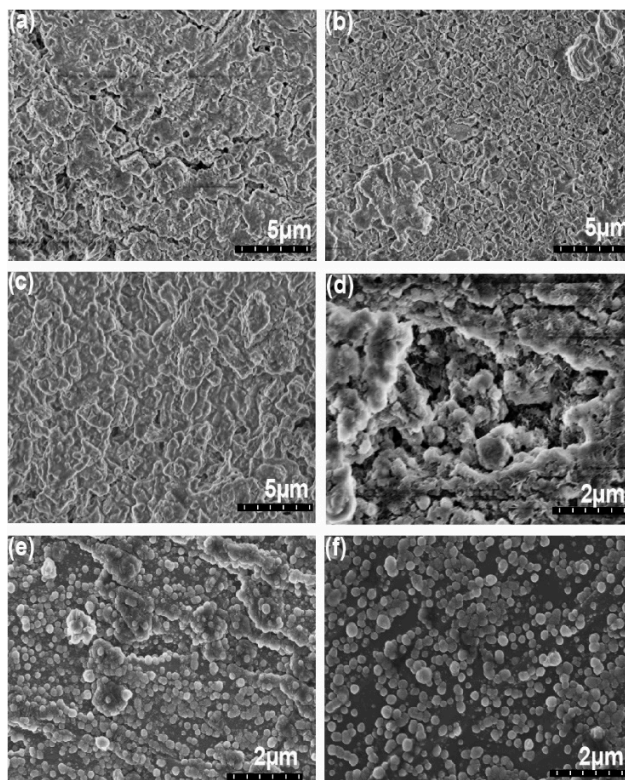
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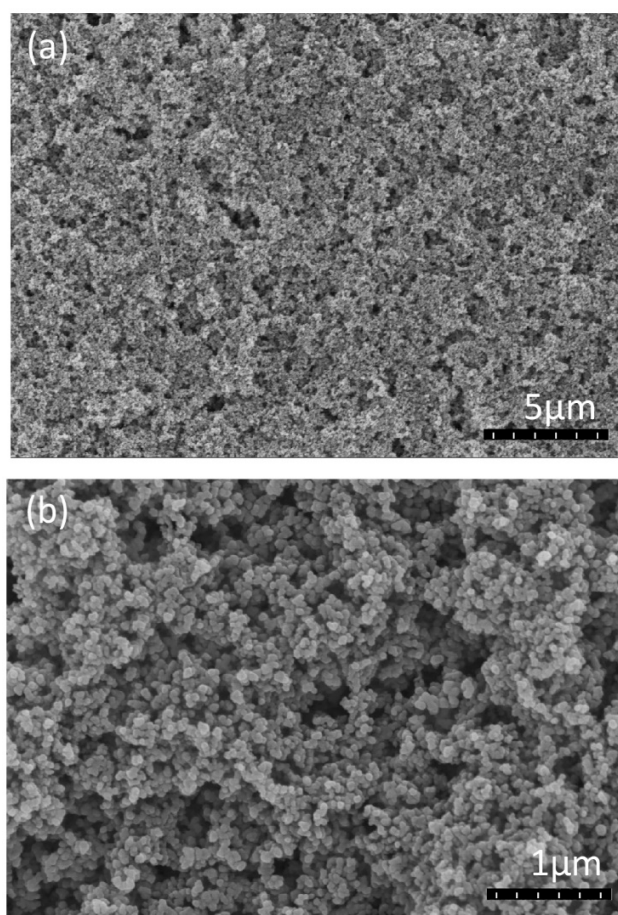
**Figure S1.** Potential profiles during the precycling (at a  $C/5$  rate between 3.0 and 4.3 V vs.  $\text{Li/Li}^+$ ) of (a) half cells (LMO/Li metal) and (b) full cells (LMO/graphite) (charging rate = discharging rate =  $C/5$ , i.e.,  $0.257 \text{ mA cm}^{-2}$ ).



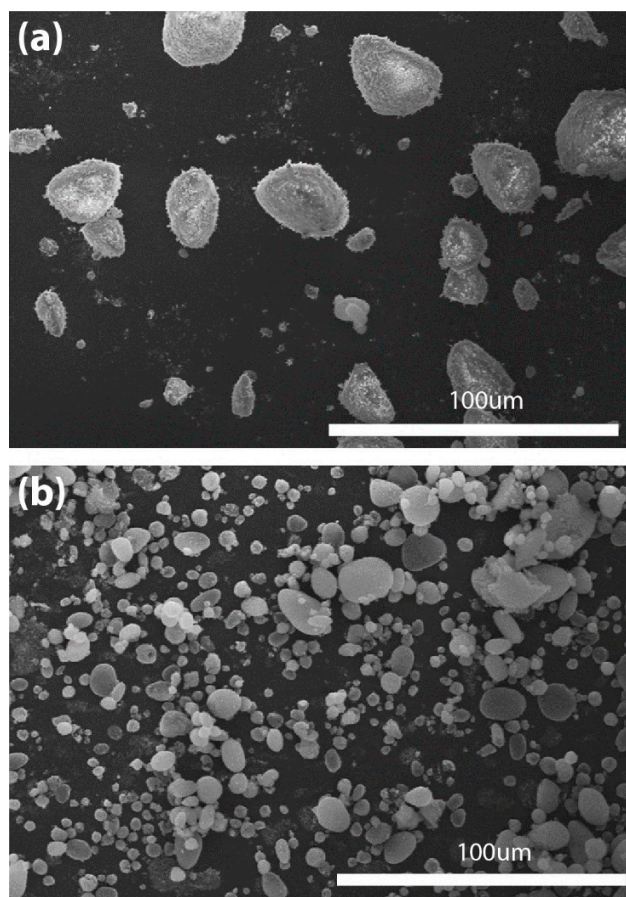
**Figure S2.** Nyquist plots of (a) full cells (LMO/graphite) and (b) half cells (LMO/Li metal) containing bare PE, SC-CCS, and DC-CCS PE separators after precycling, corresponding to Figure S1.



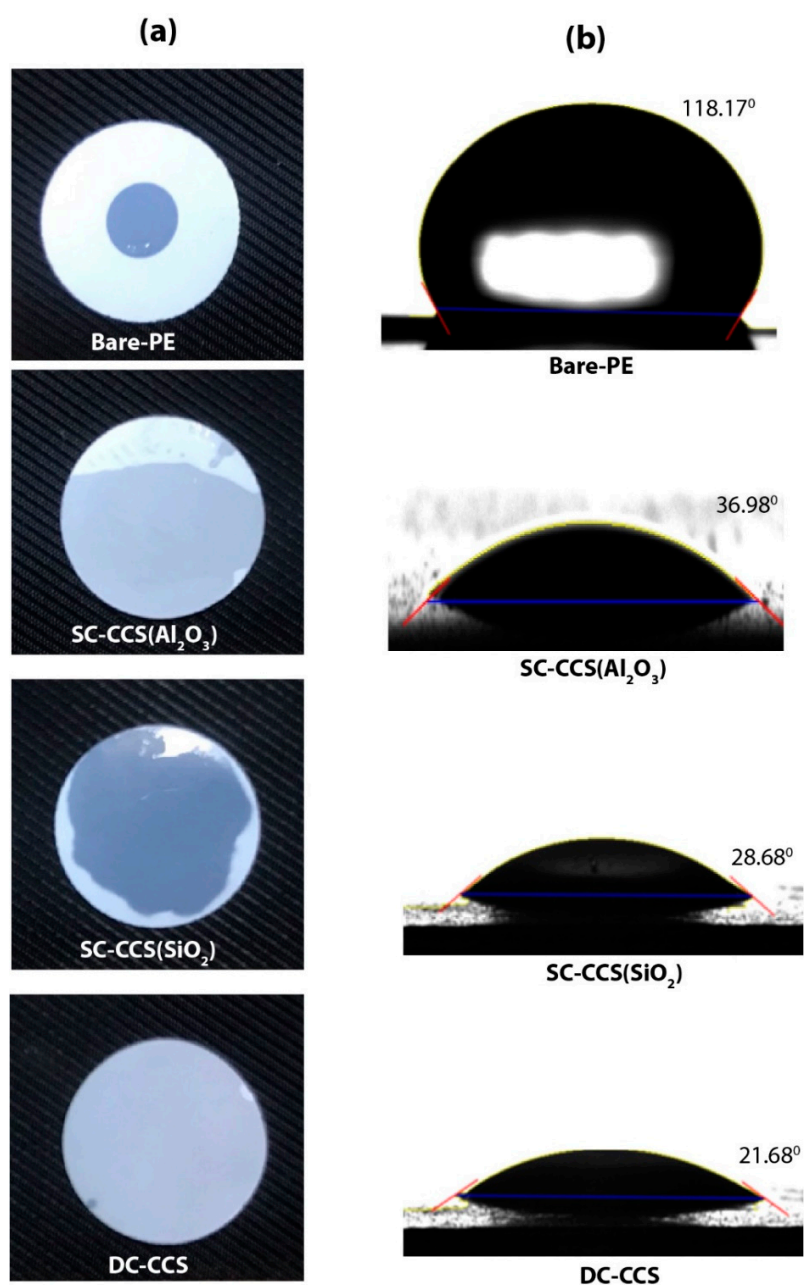
**Figure S3.** Surface SEM images of Li metal disassembled from half cells (LMO/Li metal) containing (a) bare PE, (b) SC-CCSs, and (c) DC-CCSs after cycle number 25, shown in Figure 7c. Surface SEM images of Li metal disassembled from Li/Li symmetric cells containing (d) bare PE, (e) SC-CCSs, and (f) DC-CCSs after plating at  $0.2 \text{ mA cm}^{-2}$  for 30 min.



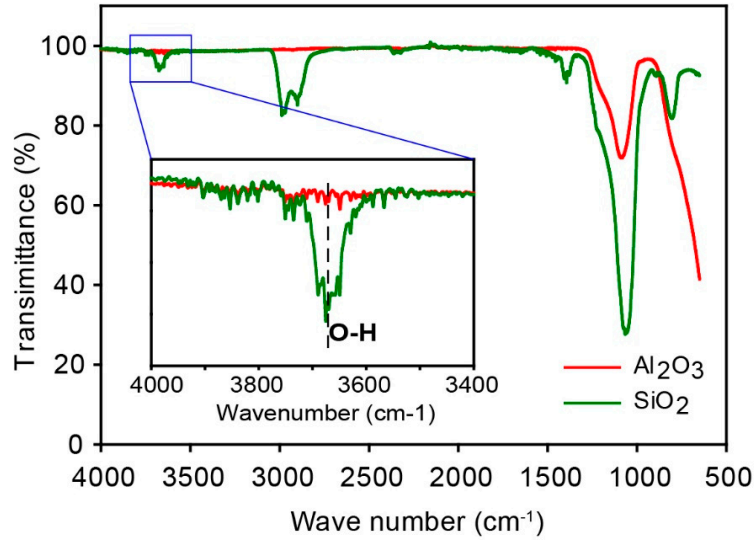
**Figure S4.** SEM images of SC-CCSs containing SiO<sub>2</sub>.



**Figure S5.** SEM images of pure (a)  $\text{Al}_2\text{O}_3$  and (b)  $\text{SiO}_2$ .



**Figure S6.** (a) Digital camera images of bare PE, SC-CCS( $\text{Al}_2\text{O}_3$ ), SC-CCS( $\text{SiO}_2$ ), and DC-CCSs surfaces 5 min after pouring a drop of liquid electrolyte. (b) Contact angle images of separator surfaces with a drop of D.I. water.



**Figure S7.** FT-IR spectra of SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> showing the absorption bands of surface hydroxyl groups (–OH) on particle surfaces.

**Table S1.** Calculation of the Li transference number ( $t_+$ ) of each separator system

Cell	Polarization time (h)	$\Delta V$ (mV)	$R_0$ ( $\Omega$ )	$R_{ss}$ ( $\Omega$ )	$I_0$ (mA)	$I_{ss}$ (mA)	$t_+$
Bare-PE	5	10	1.348	1.383	0.0198	0.0065	0.33
SC-CCS(Al <sub>2</sub> O <sub>3</sub> )	5	10	1.511	2.174	0.0156	0.0062	0.40
SC-CCS(SiO <sub>2</sub> )	5	10	1.700	2.522	0.0190	0.0082	0.43
DC-CCS	5	10	1.554	1.681	0.0158	0.0070	0.45

The Li transference number ( $t_+$ ) is determined by the following equation [1]:

$$t_+ = \frac{I_{ss} (\Delta V - I_0 R_0)}{I_0 (\Delta V - I_{ss} R_{ss})}$$

where  $I_{ss}$  is the steady-state current,  $I_0$  is the initial current,  $\Delta V$  is the applied potential,  $R_e$  is the electrolyte resistance, and  $R_{ss}$  and  $R_0$  are the electrode resistances after and before the polarization, respectively.



## Reference

[1] Evans, J.; Vincent, C.A.; Bruce, P.G. Electrochemical measurement of transference numbers in polymer electrolytes. *Polymer* **1987**, *28*, 2324-2328.