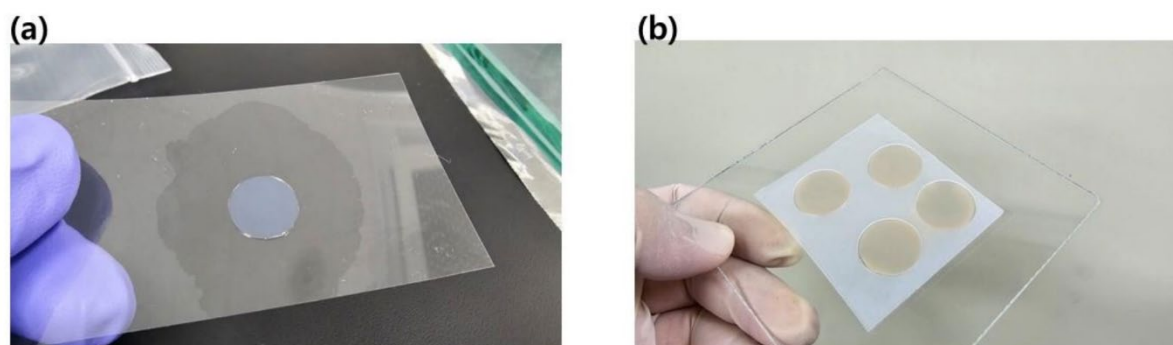
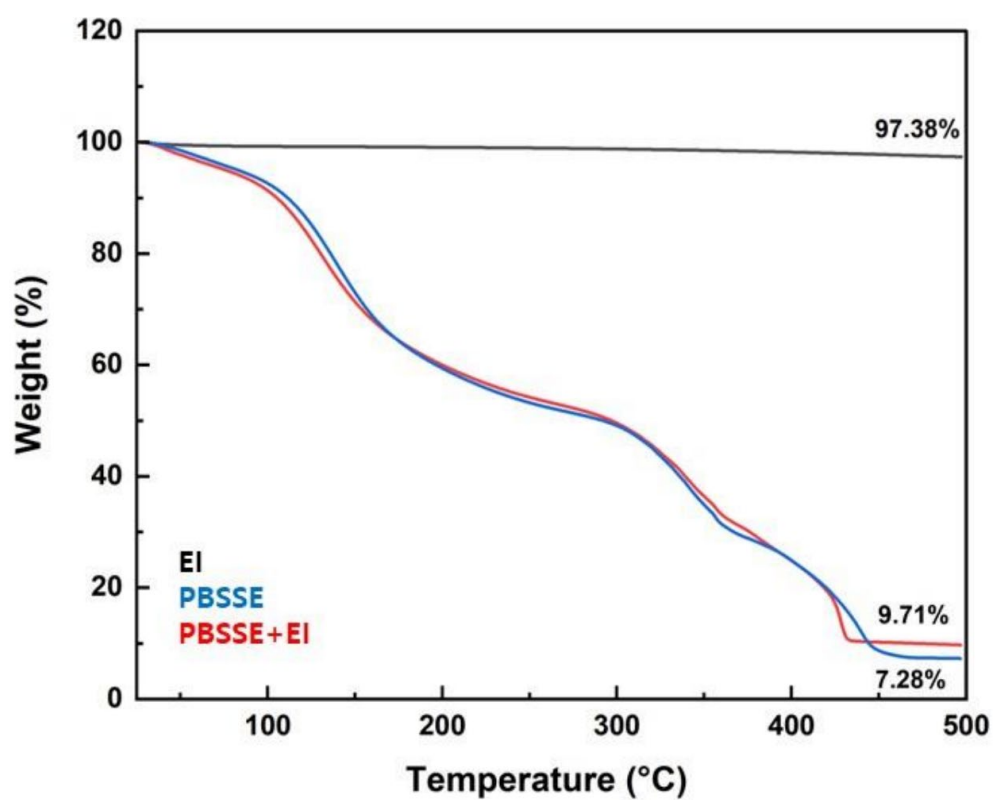


**Table S1.** Particle size analysis data of the pure illite and expanded illite (EI).

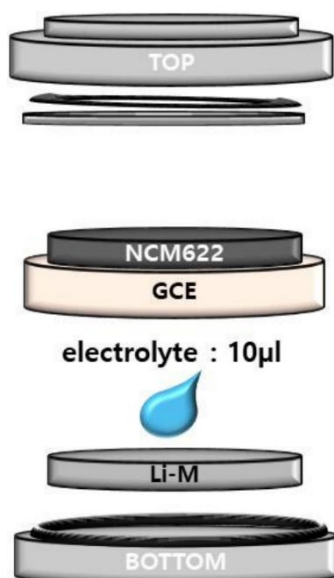
	Pure illite	EI
Median size ( $\mu\text{m}$ )	3.81	3.41
Mean size ( $\mu\text{m}$ )	5.28	8.26
Mode size ( $\mu\text{m}$ )	4.79	4.18
St. Dev. ( $\mu\text{m}$ )	11.05	26.55
Diameter on cumulative %	(2)10.00 (%) - 1.12 ( $\mu\text{m}$ ) (5)50.00 (%) - 3.81 ( $\mu\text{m}$ ) (9)90.00 (%) - 8.44 ( $\mu\text{m}$ )	(2)10.00 (%) - 0.84 ( $\mu\text{m}$ ) (5)50.00 (%) - 3.41 ( $\mu\text{m}$ ) (9)90.00 (%) - 9.05 ( $\mu\text{m}$ )



**Figure S1.** Photo images of the (a) PBSSE without the EI (white color) and (b) PBSSE with 4 wt% of the EI (yellow color).



**Figure S2.** TGA curves of the EI (black line), PBSSE without the EI (blue line), and PBSSE with 4 wt% of the EI (red line).



**Figure S3.** Coin-cell configuration for the all-solid-state Li-ion batteries (ASSLIBs).

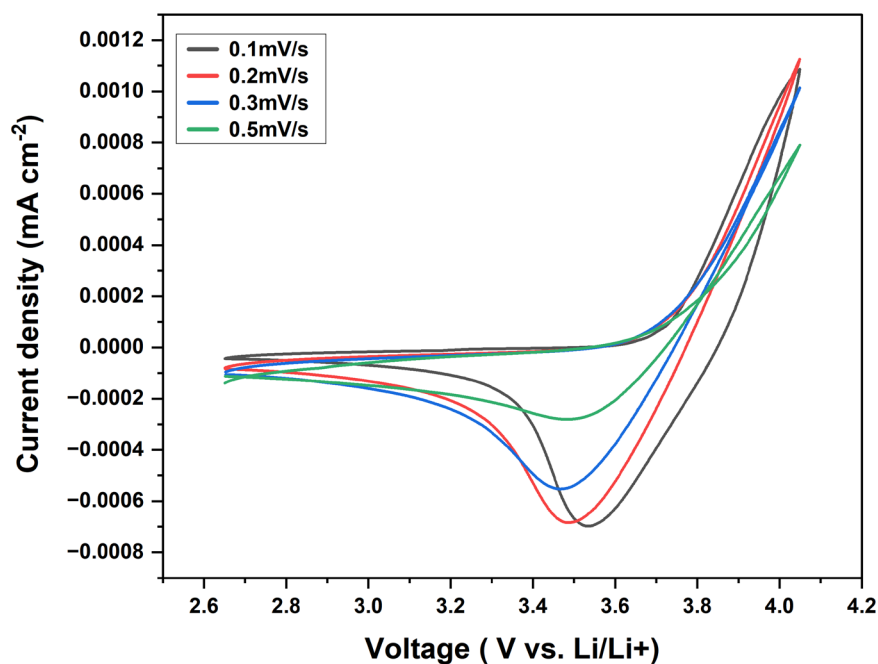


Figure S4. Cyclic voltammogram of the ASSLIBs tested at different scan rates from 0.1 to 0.5 mV s<sup>-1</sup>.

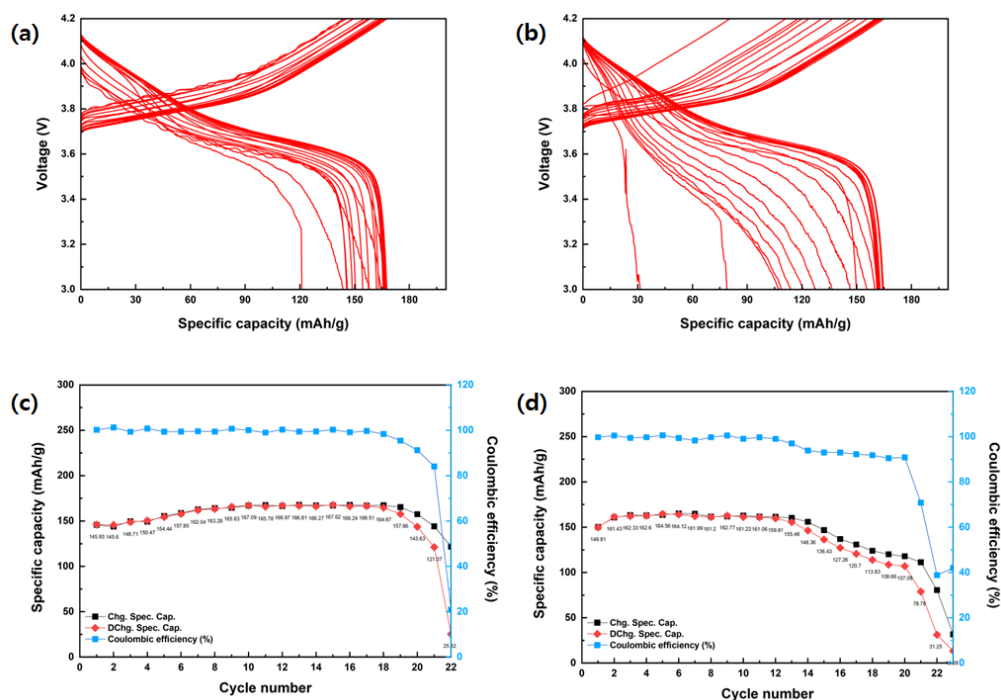


Figure S5. Charge and discharge curves of the ASSLIBs assembled (a) without the EI and (b) with 8% of the EI. Cyclability of the ASSLIBs (c) without the EI and (d) with 8% of the EI.