Supplementary Data

Analyzing the Effect of Nano-Sized Conductive Additive Content on Cathode Electrode Performance in Sulfide All-Solid-State Lithium-Ion Batteries

Jae Hong Choi [1], Sumyeong Choi [1], Tom James Embleton [1], Kyungmok Ko [1], Kashif Saleem Saqib [1], Jahanzaib Ali [1], Mina Jo [1], Junhyeok Hwang [1], Sungwoo Park [1], Minhu Kim [1], Mingi Hwang [1], Hees oo Lim [1] and Pilgun Oh *[1,2]

1 Department of Smart Green Technology Engineering, Pukyong National University, 45, Yongso-ro, Nam-gu, Busan 48547, Re public of Korea

2 Department of Nanotechnology Engineering, Pukyong National University, 45, Yongso-ro, Nam-gu, Busan 48547, Republic of Korea

*Correspondence: E-mail addresses: poh@pknu.ac.kr; Tel.: +82-51-629-6387; Fax: +82-51-629-6388

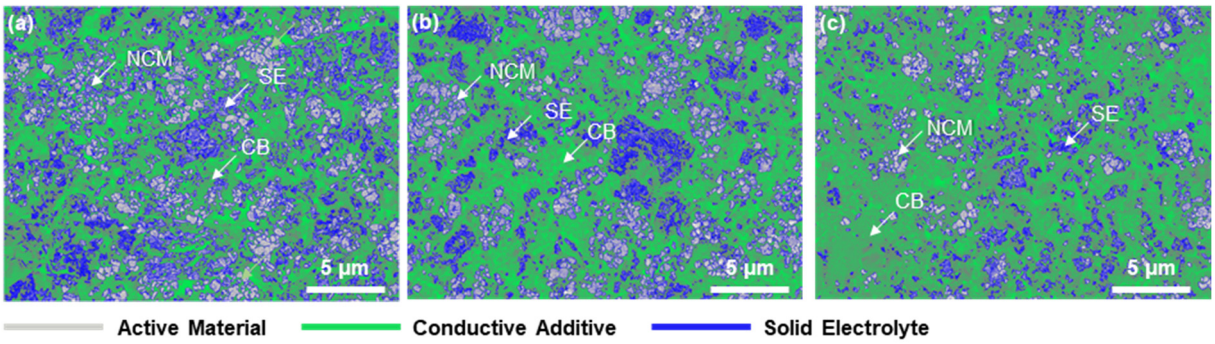

Figure S1. Scanning electron microscopy (SEM) images of the surface of (a) 3 wt%, (b) 5 wt%, and (c) 7 wt% carbon black (CB) composites after pelletizing with the look-up table (LUT) color gradient altered to better display the 3 components of the cathode composite.

Image Processing Methods

ImageJ software was used to process the scanning electron microscopy images. The images were edited using the Color-LUT function to directly map the new colors that were selected onto the previously gray gradient of the SEM imaging. The colors were automatically mapped and had no user input aside from the color selection.

Table S1. The properties and impact of applying of various carbon additives, as characterized in previous work.

	Properties				Impact	
Carbon Additive	Dispersion	Surface Area	Graphiticity	Stability with SSE	Key advantages	Key disadvantage
Carbon Black	High	High	Low	Low	Good electrical network	Large CB/SE contact
Carbon Nanofiber	Low	Low	High	High	Low CNF/SE contact	Poor electrical network
Carbon Nanotube	Medium	Medium	High	Low	Capable of coating on active material	Agglomeration difficulties

Table S2. The charge–discharge capacities, te irreversible capacities, and coulombic efficiencies (CEs) of 3 wt%, 5 wt%, and 7 wt% CB composites conducted at 0.5 C for the 1st, 10th, 20th, 30th, 40th, and 50th cycles.

Electrochemical Analysis	3wt% CB						5wt% CB						7wt% CB					
	1st Cycle	10th Cycle	20th Cycle	30th Cycle	40th Cycle	50th Cycle	1st Cycle	10th Cycle	20th Cycle	30th Cycle	40th Cycle	50th Cycle	1st Cycle	10th Cycle	20th Cycle	30th Cycle	40th Cycle	50th Cycle
Charge Capacity (mAh/g)	176	132	121.8	112	104	97	182	131	120	111	104	98	190	134	119	105	88.6	70.3
Discharge Capacity (mAh/g)	145	130	119.8	111	103	96	149	130	119	110	103	97.1	153	132	117	102	85.9	68.4
Irreversible Capacity (mAh/g)	30.5	2.26	1.985	1.58	1.23	1.1	33.2	1.4	1.03	1.1	1	0.87	36.6	2	2.4	2.7	2.68	1.93
Coulombic Efficiency (%)	82.6	98.3	98.37	98.6	98.8	99	81.7	99	99.1	99	99	99.1	80.8	98	98	97	97	97.3

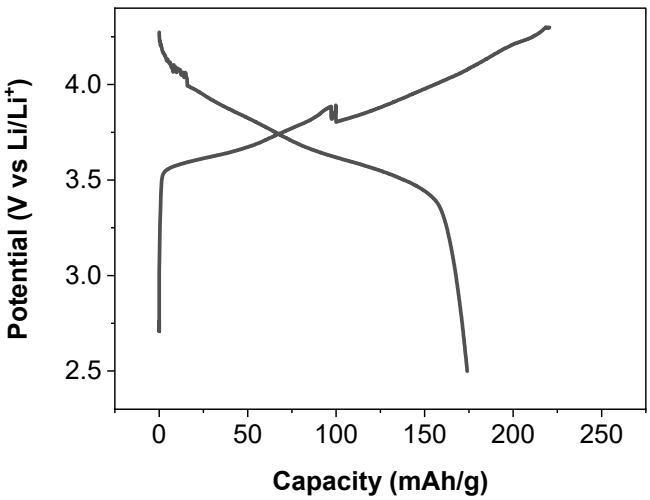

Figure S2. (a) Electrochemical performance of 0 wt% CB composite for the formation (initial charge–discharge) voltage profiles in a voltage range of 2.5–4.3 V at 0.05C.

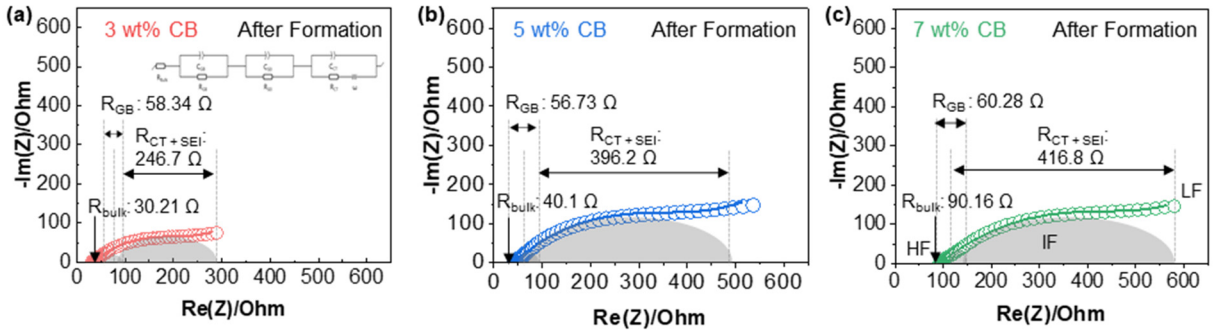

Figure S3. Nyquist plots (0.1 Hz –100 kHz) and their fittings for (a) 3 wt%, (b) 5 wt%, and (c) 7 wt% CB composites comparing results obtained after formation at 0.05C.

Table S3. Solid electrolyte bulk resistance (Rb), cathode electrolyte interphase resistance (RGB), and charge transfer resistance (RCT) of 3 wt%, 5 wt%, and 7 wt% CB composites after formation at 0.05C determined via electrochemical impedance spectroscopy (EIS).

Sample		R_{bulk} (Ω)	R_{GB} (Ω)	R_{CT+SEI} (Ω)
After formation	3 wt% CB	30.2	58.3	246.7
	5 wt% CB	40.1	56.7	396.2
	7 wt% CB	90.2	60.3	416.8

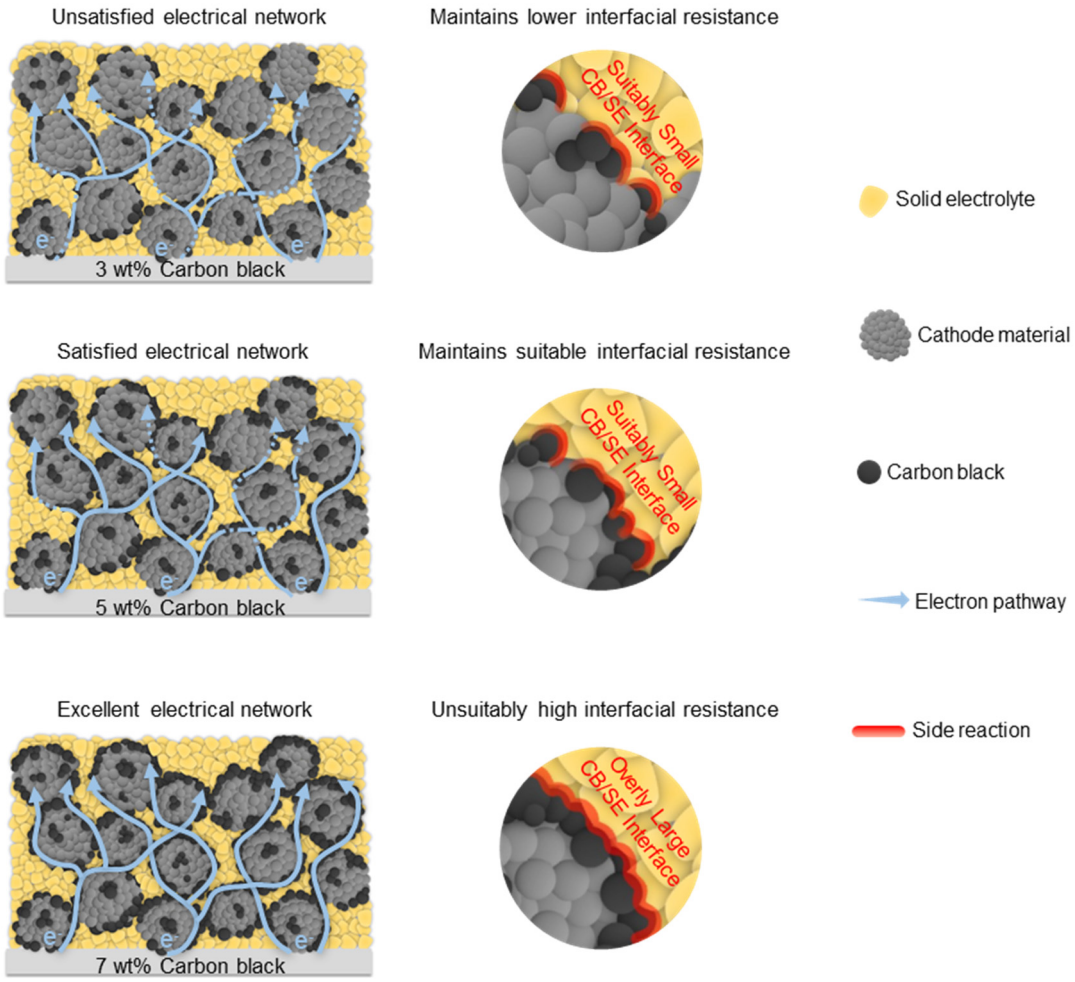

Figure S4. Scheme showing the importance of electrical conductivity of the cathode electrode and the corresponding interfacial resistance between carbon materials and solid electrolyte from inclusion of 3 wt%, 5 wt%, and 7 wt% CB.