Supplementary Information (SI) for Journal of Materials Chemistry A.

Supporting information

Enhanced Rate and Cycle Performance of All-Solid-State Batteries with an Ionic and Electronic Conductive Composite Strategy

Kyoungwan Kim,[‡ab], Seung Ho Choi,[‡a] Chaeyeon Ha,[b] Hyun-seung Kim,[a] Woosuk Cho,[a] Young-Jun Kim,[*b] KyungSu Kim[*a]

[a]*Advanced Batteries Research Center, Korea Electronics Technology Institute, Seongnam 13509, Republic of Korea*

[b]*SKKU Advanced Institute of Nanotechnology (SAINT), Sungkyunkwan University, Suwon 16419, Republic of Korea*

[*]Corresponding author.

E-mail address: kimkyungsu@keti.re.kr (K. Kim)

E-mail address: yjkim68@skku.edu (Y. Kim)

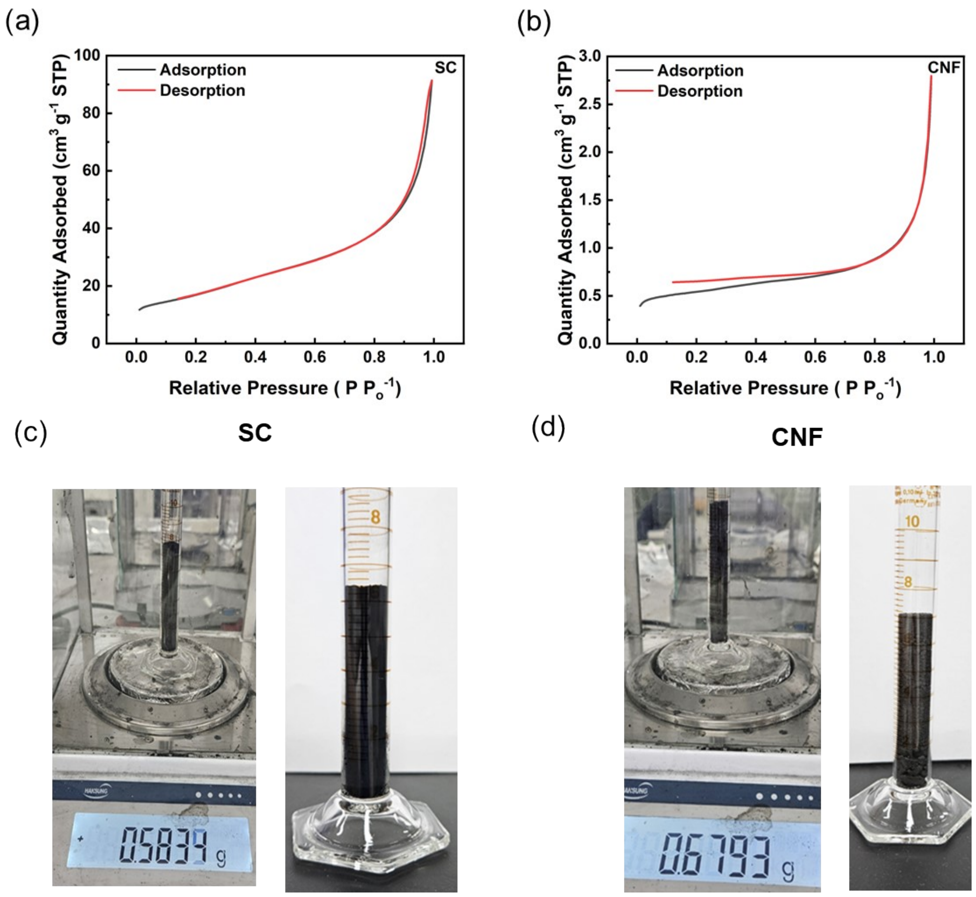

Fig. S1. (a) Super C, (b) CNF N_2 gas adsorption/desorption isotherm. (c) Super C, (d) CNF Tap density.

Table S1. Comparison of Specific surface area, Total volume in pores and Tap density between Super C and CNF.

BET Analysis	Morphology	Specific surface area (m^2 g^{-1})	Total volume in pores (cm^3 g^{-1})	Tap density (g cc^{-1})
SC	Particle	59.84	0.14	0.094
CNF	Fiber	1.84	0.0035	0.097

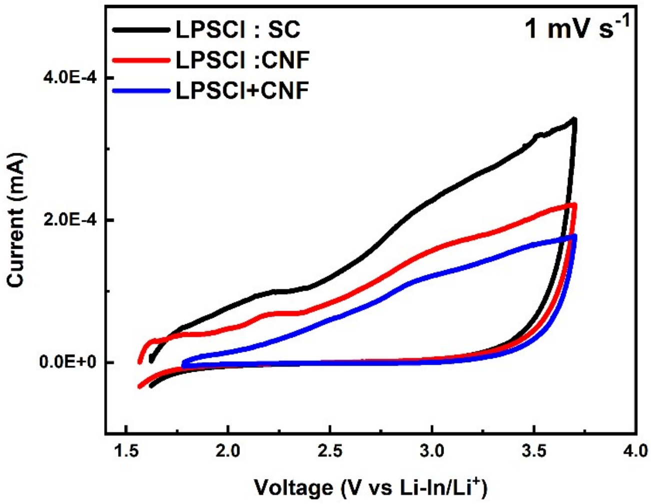

Fig S2. Cyclic voltammetry recorded from the pellet electrode with the weight ratio of solid electrolyte to carbon additive with 14.6: 0.4.

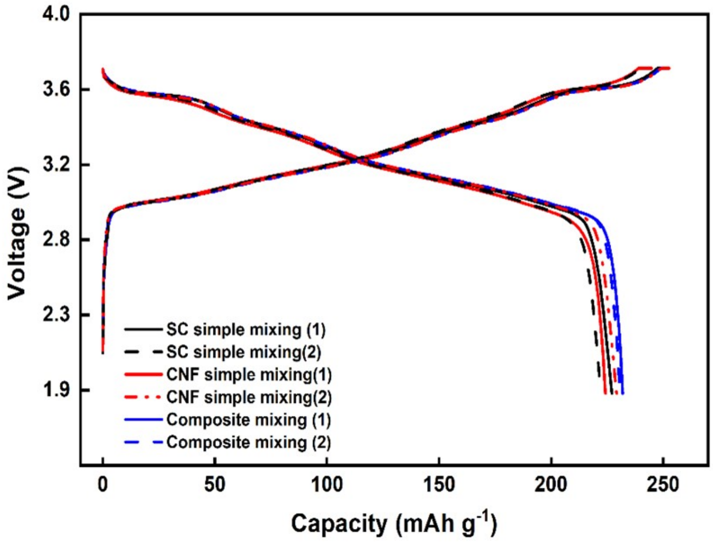

Fig S3. Electrochemical characterization of SC simple mixing, CNF simple mixing, and composite mixing at 60°C: formation voltage profile 0.1 C.

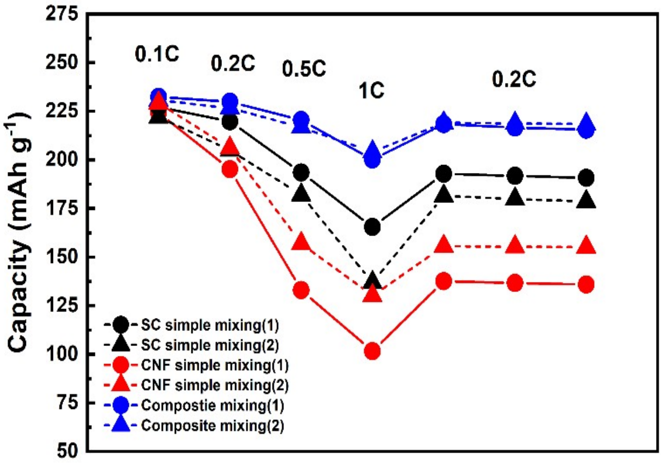

Fig S4 . Rate performances of SC simple mixing, CNF simple mixing, and composite mixing cathodes at 60 °C under various C-rates (0.1C, 0.2C, 0.5C, and 1C).

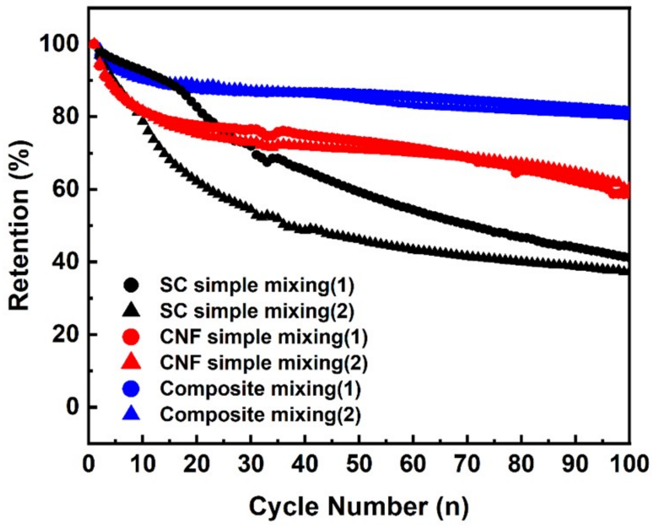

Fig S5. Electrochemical characterization of SC simple mixing, CNF simple mixing, and composite mixing at 60°C: cycle performance at high C-rates (0.5C).

Table S2. Comparison of battery performances using carbon nanofiber additives as a conductive agent.

Cathode electrode (ratio)	Areal loading (mg cm^{-2})	Initial discharge capacity (mAh g^{-1})	C-rate	Cycle #/ Capacity retention (%)	Temperature (^{o}C)	Reference
NCM622/Li_3PS_4/CNF/binder (67.2:28.8:2:2 wt%)	7.6	140	0.5 C	50 / 90	60	S1
NCA/Li_6PS_5Cl/VGCF (70:25:5 wt%)	14.0	158	0.64 C	100 / 75.6	25	S2
NCM622/Li_6PS_5Cl/VGCF (60:35:5 wt%)	11.3	123	0.5 C	-	55	S3
NCM811/Li_6PS_5Cl/CNF (64:27:9 wt%)	8.5	194.7	0.5 C	50 / 78.8	30	S4
NCM522/Li_2S-P_2S_5/CNF (60:35:5 wt%)	10	128	-	50 / 62.5	-	S5
NCM811/Li_6PS_5Cl/CNF (66.6:28.6:4.8 wt%)	11.4	191.8	0.5 C	50 / 67.4	30	S6
NCM811/Li_6PS_5Cl/CNF (70:30:5 wt%)	11.4	150	0.5 C	50 / 90.1	30	S7
NCM622/Li_3PS_4/VGCF (70:30:3 wt%)	8.9	144	0.1 C	100 / 45.1	25	S8
NCM711/Li_6PS_5Cl/CNF (75.6:19.4:3 wt%)	22.6	170.0	0.1 C	100 / 75.3	-	S9
$LiNi_{0.9}Co_{0.05}Mn_{0.05}O_2$/$Li_6PS_5Cl$/CNF (85:14.6:0.4 wt%)	7.5	231.9	0.5 C	100 / 81.6	60	This work

References

[S1] T. Ates, M. Keller, J. Kulisch, T. Adermann and S. Passeriniaoi, *Energy Storage Mater.*, 2019, **17**, 204−210.

[S2] J. Gao, J. Hao, Y. Gao, X. Sun, Y. Zhang, D. Song, Q. Zhao, F. Zhao, W. Si, K. Wang, T. Ohsaka, F. Matsumoto, J. Wu and H. Xie, *eTransportation*, 2023, **17**, 100252.

[S3] Y. Huh, H. G. Lee, C.-M. Cho, J.-W. Park, B. G. Kim, Y.-J. Lee, H. Park, Y.-C. Ha, J.-H. Choi, J. Yi, S.-M. Lee, J. I. Lee and J.-H. Park, *Batteries & Supercaps* 2023, **6**, e202300036.

[S4] J. H. Choi, S. Choi, T. J. Embleton, K. Ko, K. S. Saqib, M. Jo, J. Hwang, S. Park, Y. Son and P. Oh, *Batteries*, 2023, **9**, 590.

[S5] T. Shi, Y.-Q. Zhang, Q. Tu, Y. Wang, M. C. Scott and G. Ceder, *J. Mater. Chem. A*, 2020, **8**, 17399.

[S6] J. H. Choi, S. Choi, T. J. Embleton, K. Ko, K. S. Saqib, J. Ali, M. Jo, J. Hwang, S. Park, M. Kim, M. Hwang, H. Lim and P. Oh, *Nanomaterials*, 2023, **13**, 3065.

[S7] K. S. Saqib, T. J. Embleton, J. H. Choi, S.-J. Won, J. Ali, K. Ko, S. Choi, M. Jo, S. Park, J. Park, W. Kaveevivitchai, Y. Son, W.-J. Lee, P. Oh, *ACS Appl. Mater. Interfaces*, 2024, **16**, 47551−47562.

[S8] F. Walther, S. Randau, Y. Schneider, J. Sann, M. Rohnke, F. H. Richter, W. G. Zeier, Jürgen Janek, *Chem. Mater.*, 2020, **32**, 6123−6136.

[S9] N. Lee, J. Lee, T. Lee, J. Oh, I. Hwang, G. Seo, H. Kim and J. W. Choi, *ACS Appl. Mater. Interfaces*, 2023, **15**, 34931−34940.