Electronic Supplementary Information (ESI)

Argyrodite sulfide coated NCM cathode for the improved interfacial contact in normal-pressure operational all-solid-state batteries

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Fig. S1 1st charge–discharge voltage profile of NCM523 with liquid electrolyte.
Fig. S2 BET analysis of $\text{N}_2$ adsorption–desorption isotherm for NCMS23.
Fig. S3 Results of liquid-phase synthesis of Li$_6$PS$_5$Cl precursors in solvent. Digital photographs of (a) without dibutyl ether and (b) with dibutyl ether (during synthesis without stirring).
Fig. S4 Nyquist plots of impedance spectra for (a) LPSCI_BM, (b) LPSCI(LP)1, and (c) LPSCI(LP)2 with elevating temperature from RT to 110 °C at intervals of 20 °C.
**Fig. S5** Nyquist plots of impedance spectra for (a) LPSCI_BM, (b) LPSCI_LP1, and (c) LPSCI_LP2 with heat-treatment at 180 °C.
Fig. S6  Chronoamperometry results of (a) LPSCI_BM, (b) LPSCI_LP1, and (c) LPSCI_LP2 with an applied voltage of 0.1, 0.2, 0.3, 0.4, and 0.5 V. (d) Linear fits.
Fig. S7 SEM images of (a) NCM523_BM, (b) NCM523_LP1, and (c) NCM523_LP2 with corresponding EDS mapping of Co, Mn, O, P, and Cl.
Fig. S8 (a) Digital photograph of 10 g base coating process. SEM images of (b) 1 g base and (c) 10 g base of NCM523_LP2 with corresponding EDS mapping of Ni, Co, Mn, O, P, S, and Cl.
**Fig. S9** Ni 2p XPS spectra of NCM523 and NCM523_LP2.
Fig. 510 Rietveld refinement results of the XRD patterns of NCM523_LP2.
**Fig. S11** 1\textsuperscript{st} charge–discharge voltage profiles of all-solid-state cells with composite cathode featuring NCM523, NCM523_LP1, and NCM523_LP2.
Fig. S12 Fitted values for impedance spectra of all-solid-state cells with (a) NCM523, and (b) NCM523_LP2 in Fig. 4e.
Fig. S13  Electrochemical characterization of all-solid-state cells employing bare NCM523 and NCM523_LP2 with variable fabrication pressure. (a, c) 1st charge–discharge voltage profiles. Cross-sectional SEM images of composite cathode with (b) NCM523, and (d) NCM523_LP2 applying variable fabrication pressure. Red arrows indicate micro cracks in cathode active material.
**Fig. S14** Corresponding EDS mapping of S in Fig. 5a and b. Pristine state of composite cathode with (a) NCM523, and (b) NCM523_LP2. Red arrows indicate intimate contacts.