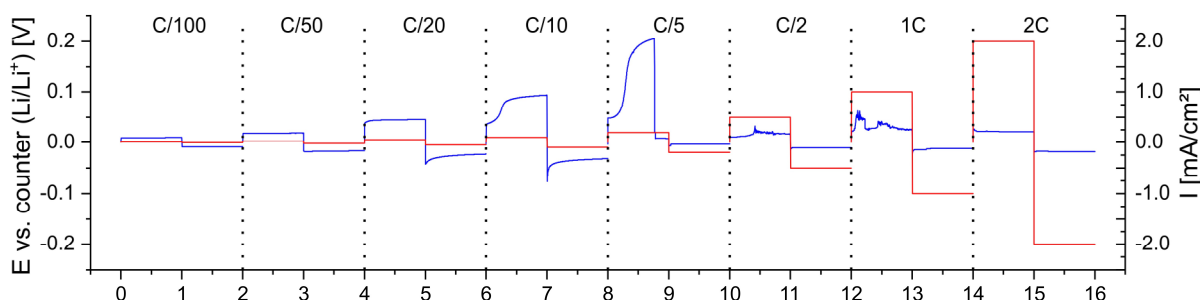


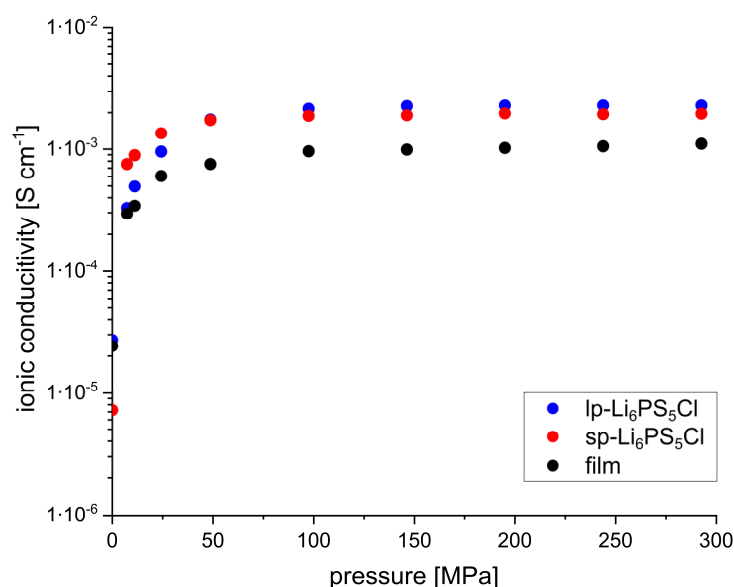
# Rational Optimization of Cathode Composites for Sulfide-Based All-Solid-State Batteries

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**Figure S1.** critical current density of  $\text{Li}_6\text{PS}_5\text{Cl}$  electrolyte membrane. A current density of 1C corresponds to  $1 \text{ mA/cm}^2$ . The measurement was conducted by sandwiching a  $300 \mu\text{m}$  thick electrolyte membrane in-between two Li electrodes of 16 mm diameter and applying 10 MPa of pressure.



**Figure S2.** ionic conductivity vs. pressure plots for powder pellets using lp-, sp- $\text{Li}_6\text{PS}_5\text{Cl}$ , and the prepared electrolyte films.

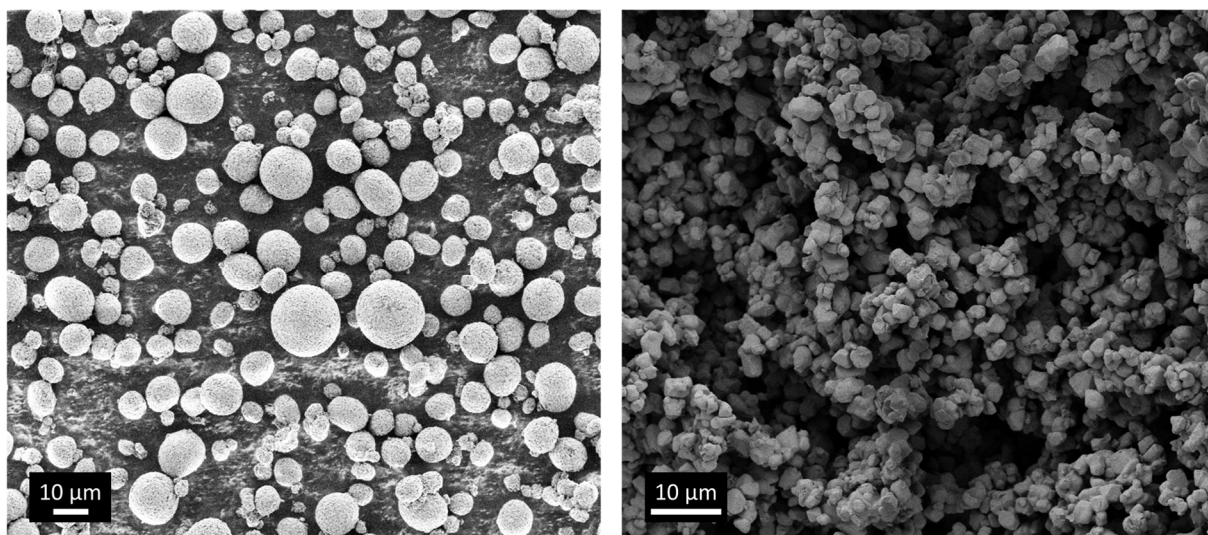


Figure S3. SEM micrographs of p-NMC811 (left) and s-NMC811 (right).

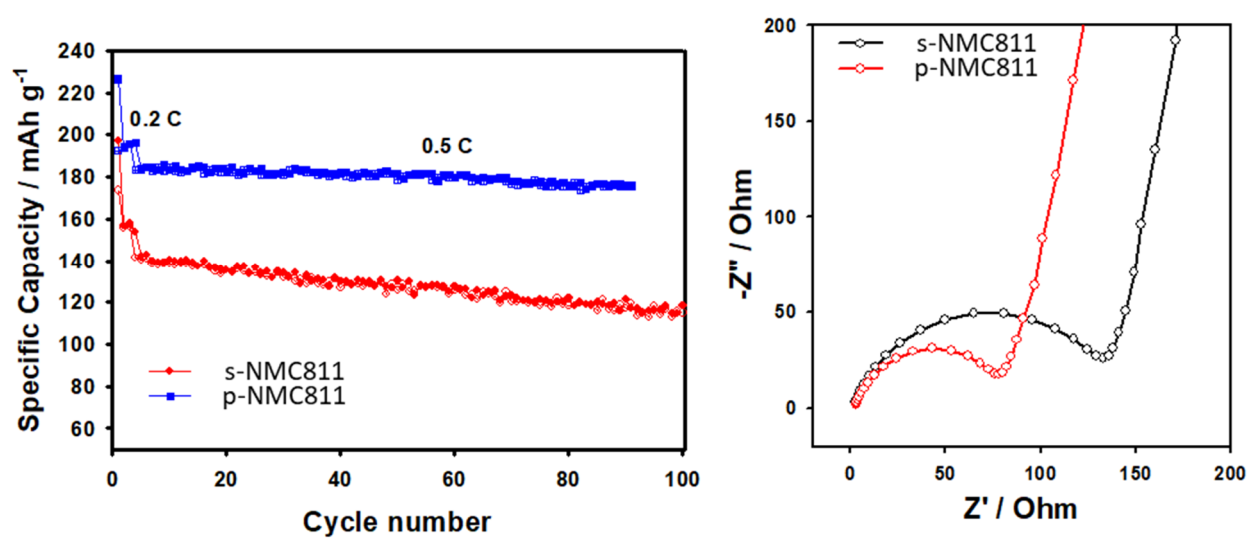
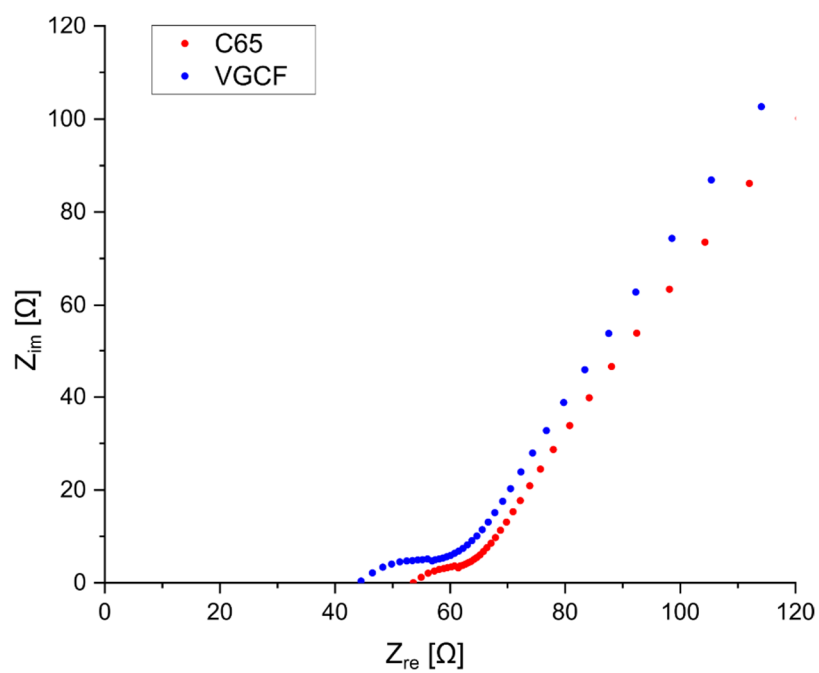
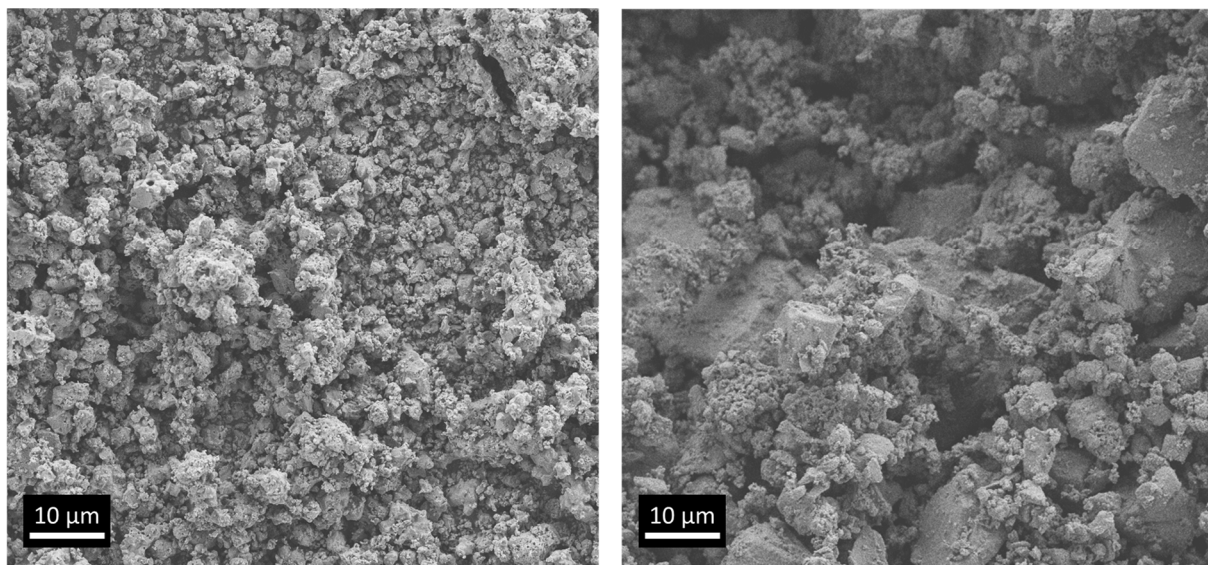


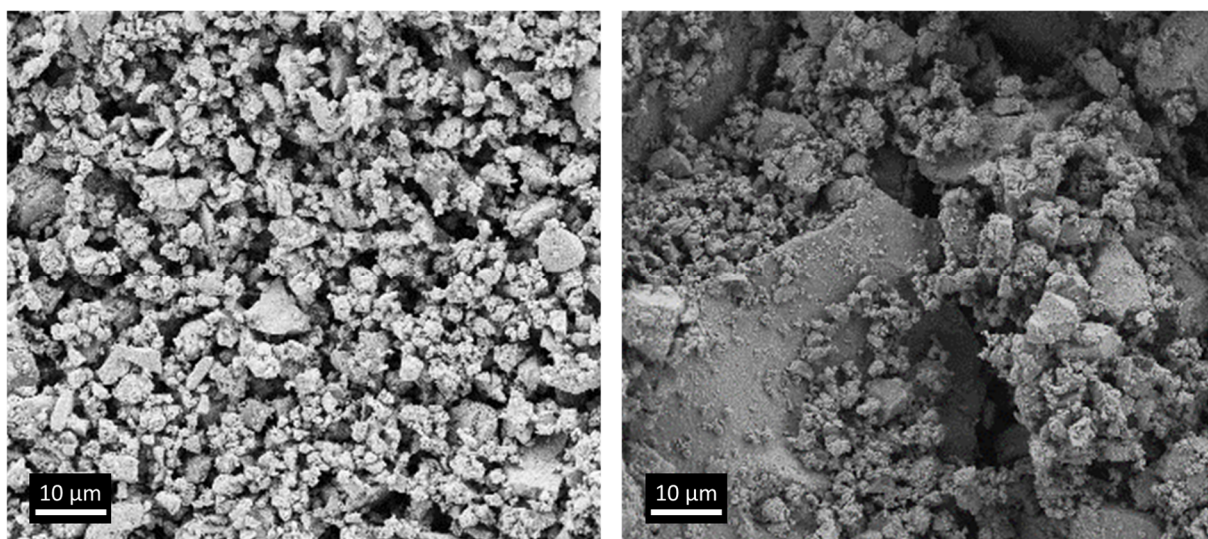
Figure S4. cycling performance of s-NMC811 and p-NMC811 in 1 M LiPF<sub>6</sub> in EC/DEC 1:1 vol./vol.).



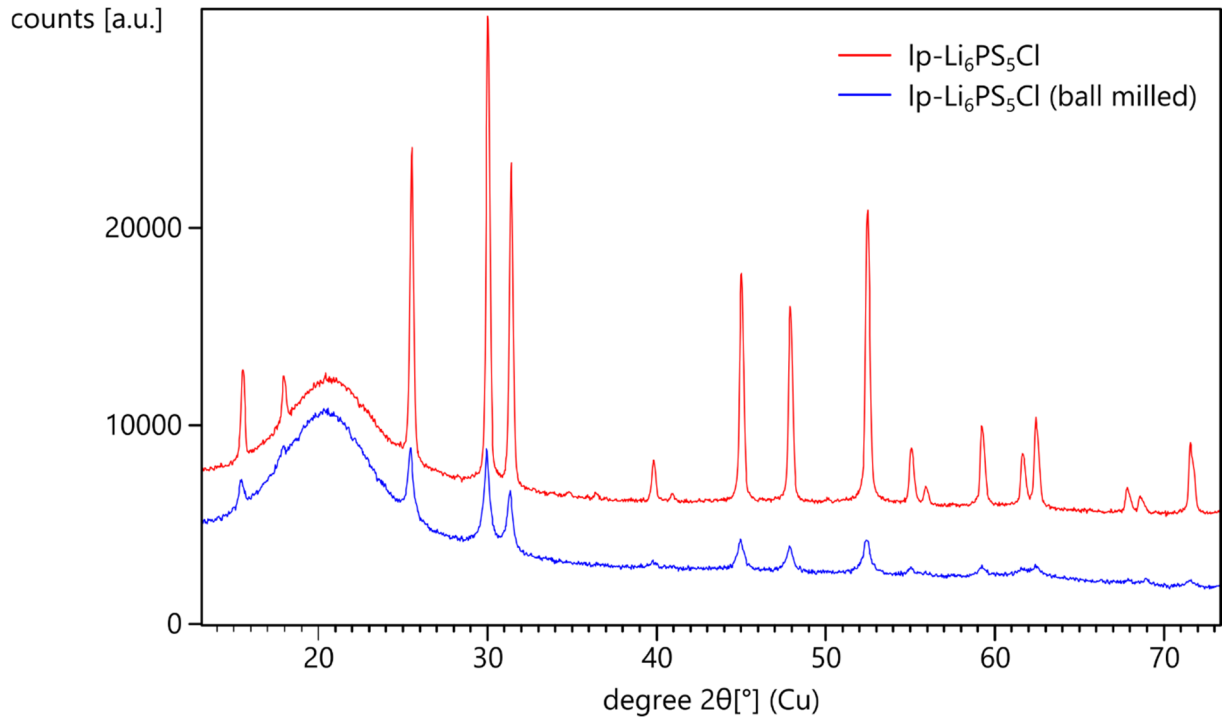
**Figure S5.** EIS data before cycling of electrodes, with C65 and VGCF as conductive additives.



**Figure S6.** SEM micrographs of sp-Li<sub>6</sub>PS<sub>5</sub>Cl (left) and lp-Li<sub>6</sub>PS<sub>5</sub>Cl (right).



**Figure S7.** SEM micrographs of lp-Li<sub>6</sub>PS<sub>5</sub>Cl and lp-Li<sub>6</sub>PS<sub>5</sub>Cl (ball milled).



**Figure S8.** XRD pattern of lp-Li<sub>6</sub>PS<sub>5</sub>Cl and lp-Li<sub>6</sub>PS<sub>5</sub>Cl (ball milled).

**Table S1.** list of all composite cathode formulations used in this work.

	AM	SE	CA	BM	AM:CA:SE:BM [wt./wt.]	AM:CA:SE:BM [vol./vol.]	AM:SE [vol./vol.]
1	p-NMC811	sp-Li <sub>6</sub> PS <sub>5</sub> Cl	C65	-	67:3:30:0	40:5:55:0	42:58
2	s-NMC811	sp-Li <sub>6</sub> PS <sub>5</sub> Cl	C65	-	67:3:30:0	40:5:55:0	42:58
3	s-NMC811	sp-Li <sub>6</sub> PS <sub>5</sub> Cl	VGCF	-	67:3:30:0	40:5:55:0	42:58
4	s-NMC811	lp-Li <sub>6</sub> PS <sub>5</sub> Cl	C65	-	67:3:30:0	40:5:55:0	42:58
5	s-NMC811	lp-Li <sub>6</sub> PS <sub>5</sub> Cl (ball-milled)	C65	-	67:3:30:0	40:5:55:0	42:58
6	s-NMC811	sp-Li <sub>6</sub> PS <sub>5</sub> Cl	C65	-	64:3:33:0	38:5:57:0	40:60
7	s-NMC811	sp-Li <sub>6</sub> PS <sub>5</sub> Cl	C65	-	73:3:24:0	48:5:47:0	50:50
8	s-NMC811	sp-Li <sub>6</sub> PS <sub>5</sub> Cl	C65	-	79:3:18:0	57:5:38:0	60:40
9	s-NMC811	sp-Li <sub>6</sub> PS <sub>5</sub> Cl	C65	-	85:3:12:0	66:5:29:0	70:30
10	s-NMC811	sp-Li <sub>6</sub> PS <sub>5</sub> Cl	C65	-	90:3:7:0	77:5:18:0	80:20
11	s-NMC811	sp-Li <sub>6</sub> PS <sub>5</sub> Cl	C65	SBS	74.5:2.5:20.5:2.5	48:5:39:8	55:45
12	s-NMC811	sp-Li <sub>6</sub> PS <sub>5</sub> Cl	C65		76:3:21:0	52:5:43:0	55:45
13	s-NMC811	sp-Li <sub>6</sub> PS <sub>5</sub> Cl	C65		81:3:16	60:5:35	63:37