

Supplementary Material

Binary anion and cation co-doping enhance sulfide solid electrolyte performance for all-solid-state lithium batteries

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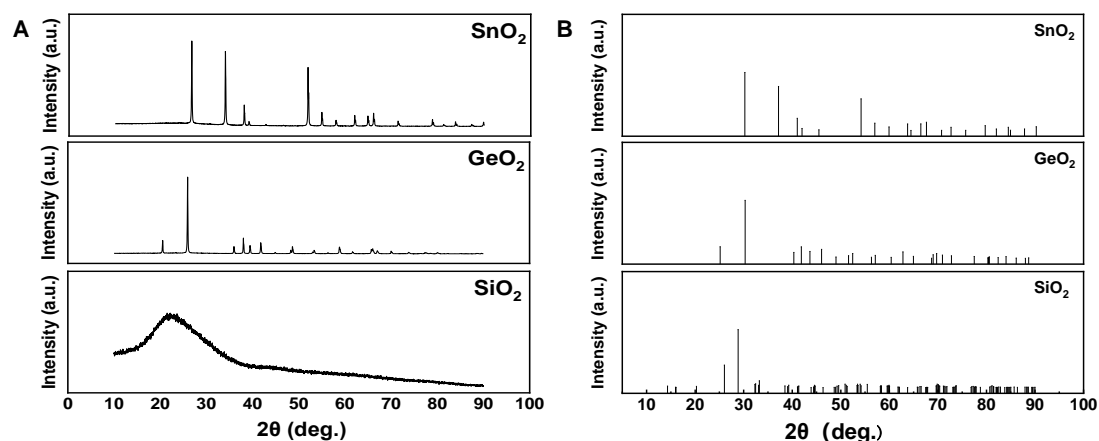


Figure S1. XRD patterns and Standard spectrum of oxides (SiO₂, GeO₂, SnO₂)

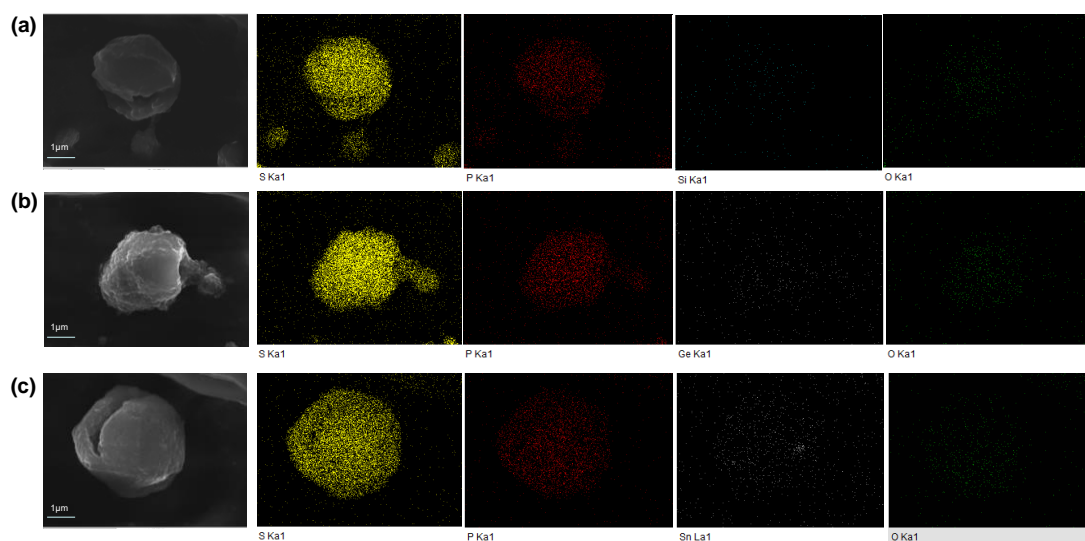


Figure S2. SEM images and EDS maps of the treated sulfide electrolyte particles of (a) LPS-0.05SiO₂; (b) LPS-0.05GeO₂; (c) LPS-0.05SnO₂.

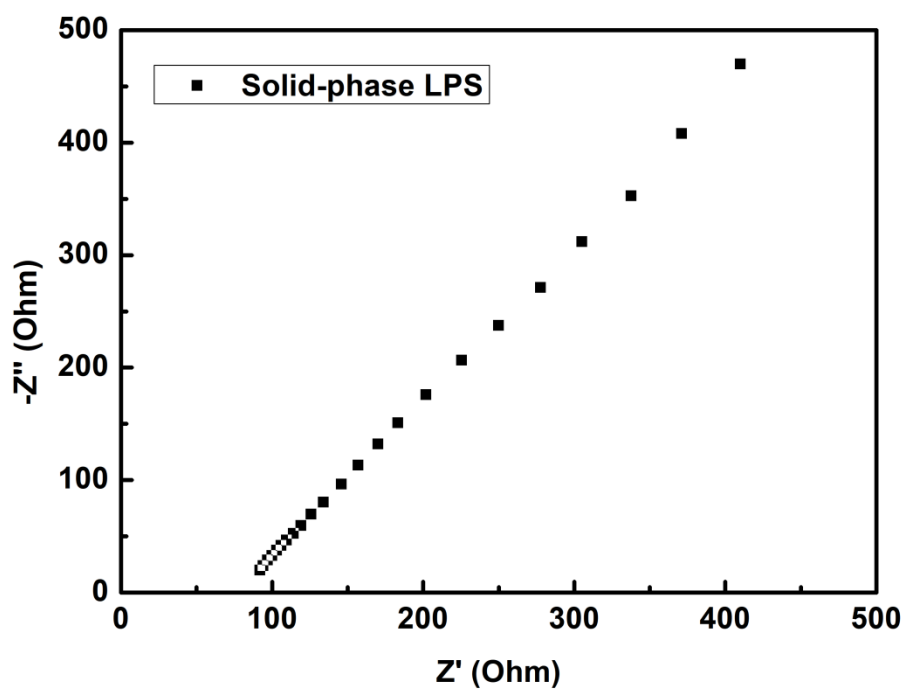


Figure S3. Nyquist plots for the LPS solid electrolytes measured at room temperature.

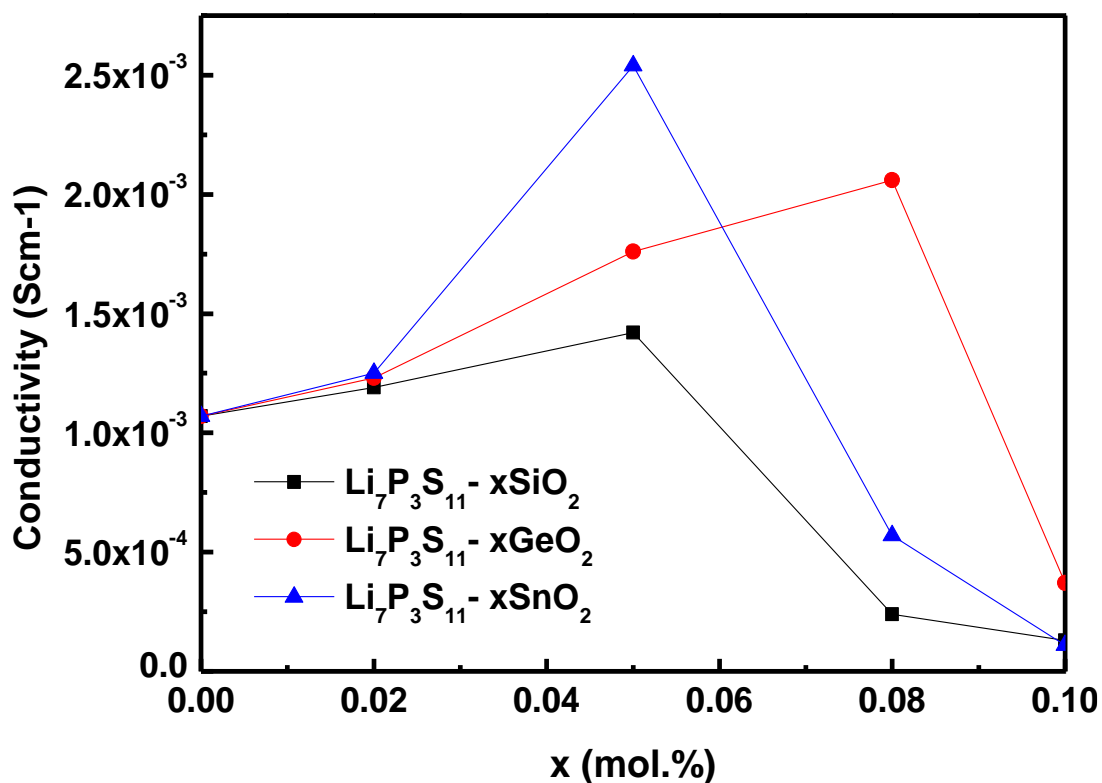


Figure S4. Relationship between the doping amount and ionic conductivities of samples

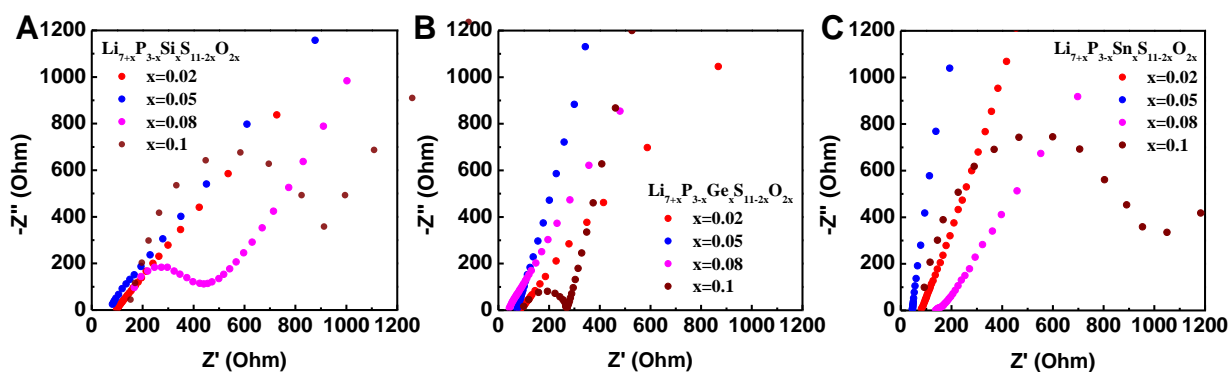


Figure S5. The high-frequency range of Nyquist plots for the solid electrolytes measured at room temperature. (A) LPS- x SiO₂ ($x=0.02, 0.05, 0.08, 0.1$). (B) LPS- x GeO₂ ($x=0.02, 0.05, 0.08, 0.1$). (C) LPS- x SnO₂ ($x=0.02, 0.05, 0.08, 0.1$).

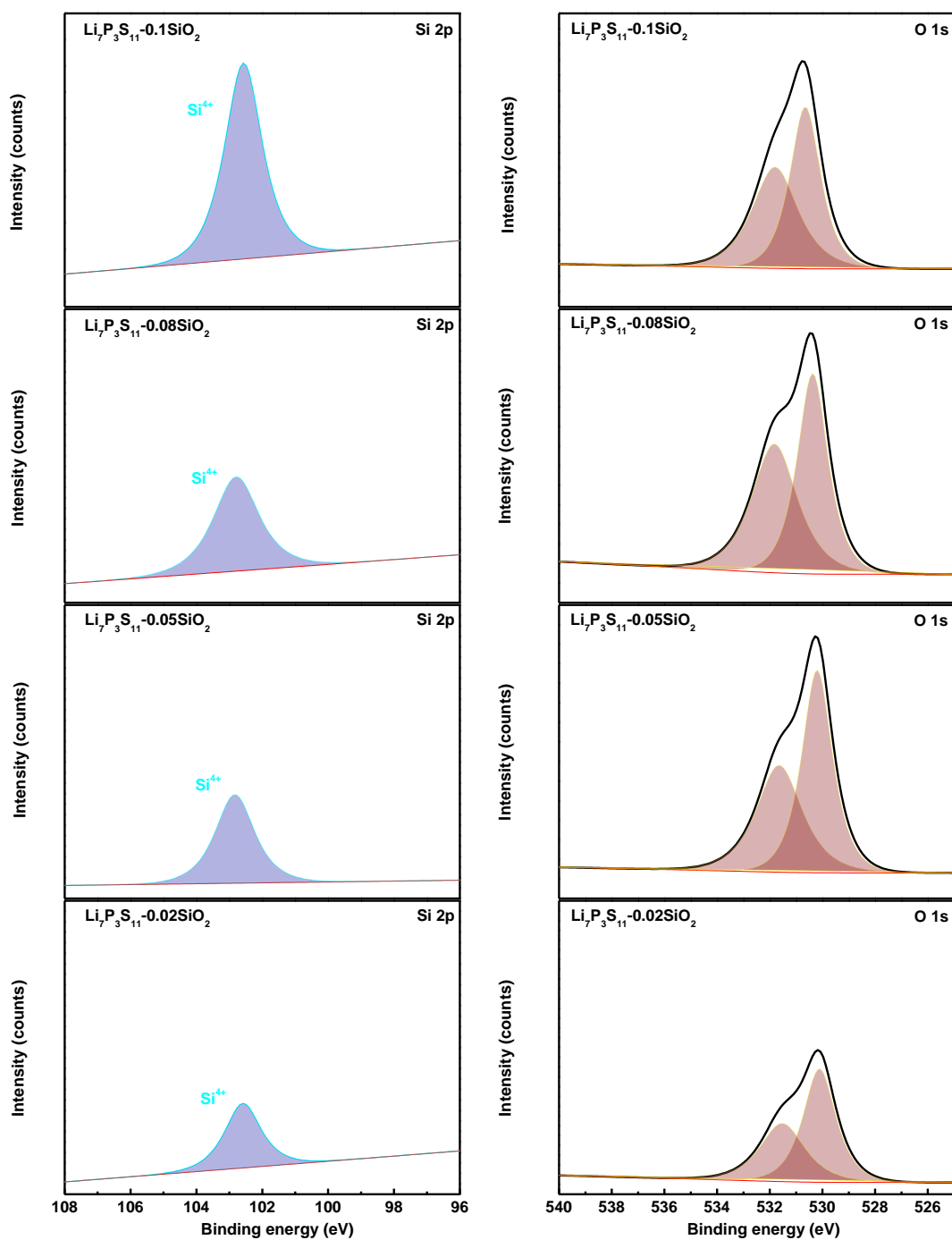


Figure S6. XPS detail spectra of Si 2p and O 1s for LPS, LPS-0.02SiO₂, LPS-0.05SiO₂, LPS-0.08SiO₂ and LPS-0.1SiO₂.

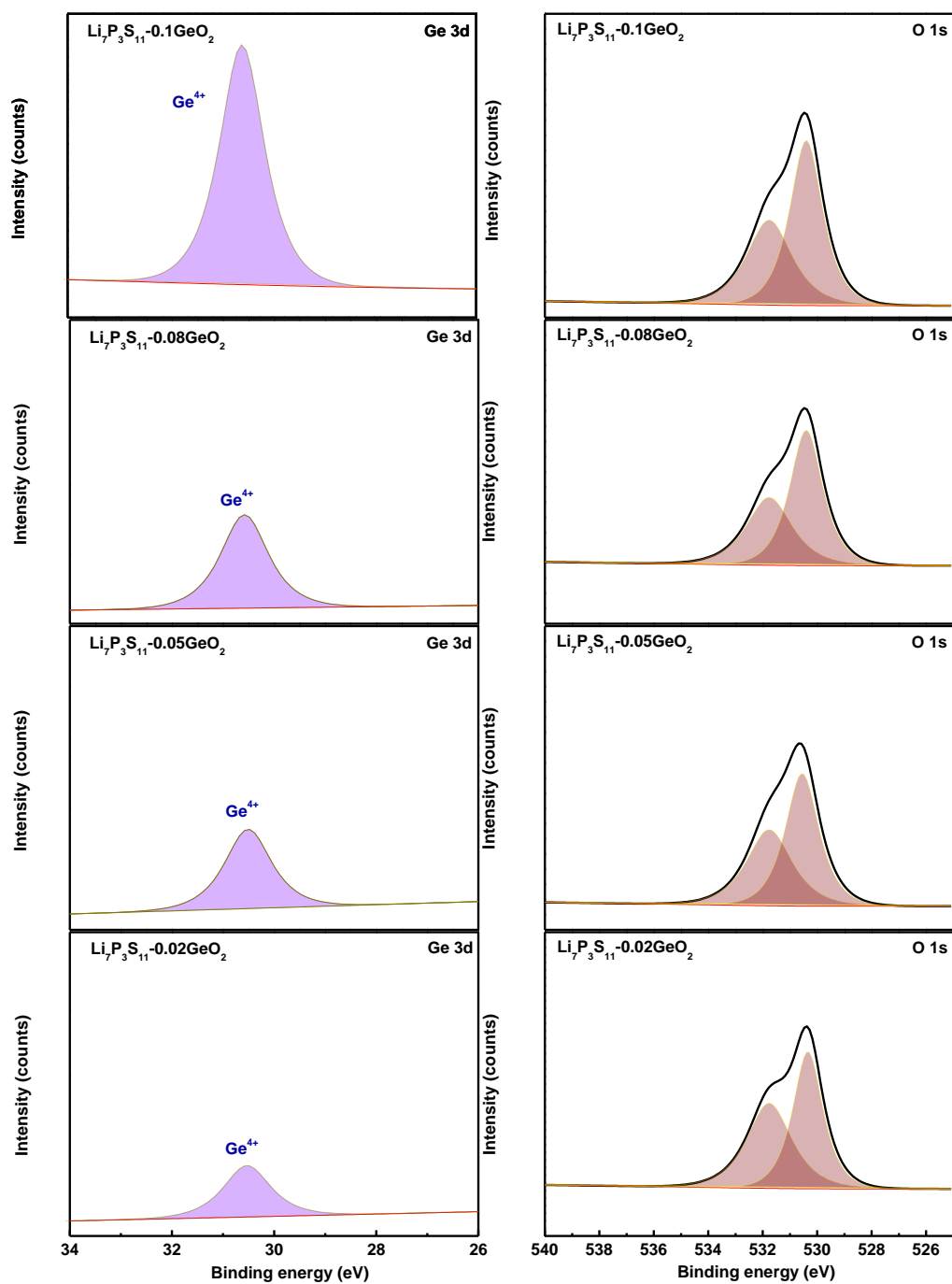


Figure S7. XPS detail spectra of Si 2p and O 1s for LPS, LPS-0.02GeO₂, LPS-0.05GeO₂, LPS-0.08GeO₂ and LPS-0.1GeO₂.

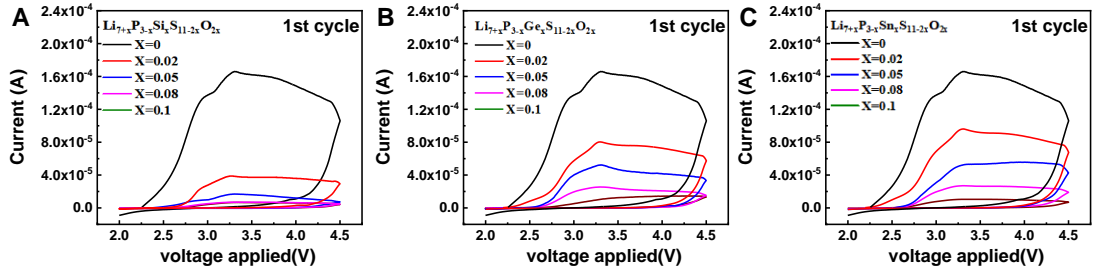


Figure S8. CV curves of the Li/SSEs/SSEs+AB cells within the voltage range from 2 to 4.5 V (vs Li/Li⁺), the first cycle curves of (A) LPS-xSiO₂ (x=0, 0.02, 0.05, 0.08, 0.1), (B) LPS-xGeO₂ (x=0, 0.02, 0.05, 0.08, 0.1), (C) LPS-xSnO₂ (x=0, 0.02, 0.05, 0.08, 0.1).

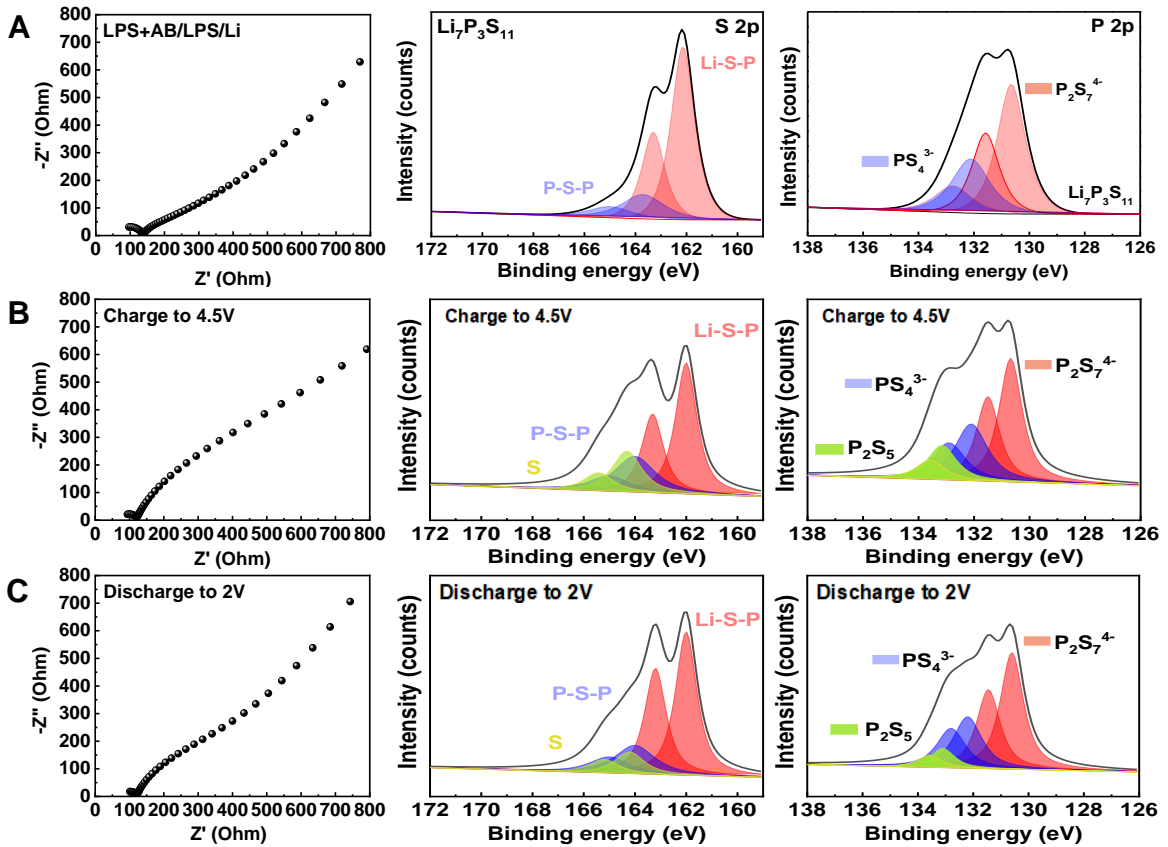


Figure S9. Nyquist plots and XPS spectra of LPS+AB/LPS/Li cell with pristine electrolytes LPS (A) Before cycling , (B) Charge to 4.5 V, (C) Discharge to 2 V.

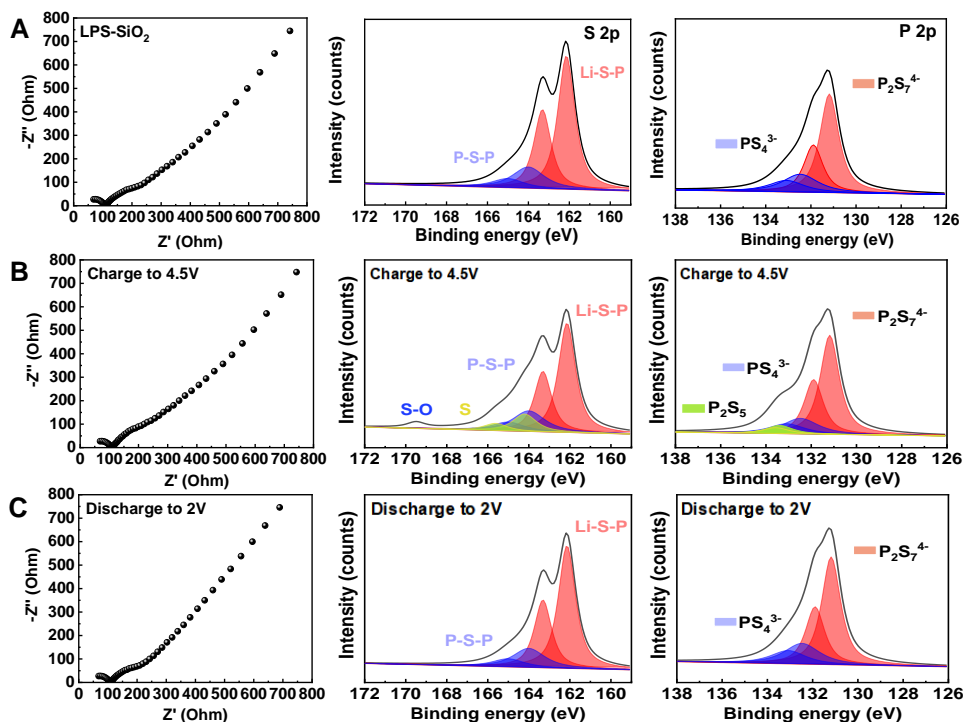


Figure S10. Nyquist plots and XPS spectra of cell with electrolytes LPS-SiO₂ (A) Before cycling , (B) Charge to 4.5 V, (C) Discharge to 2 V.

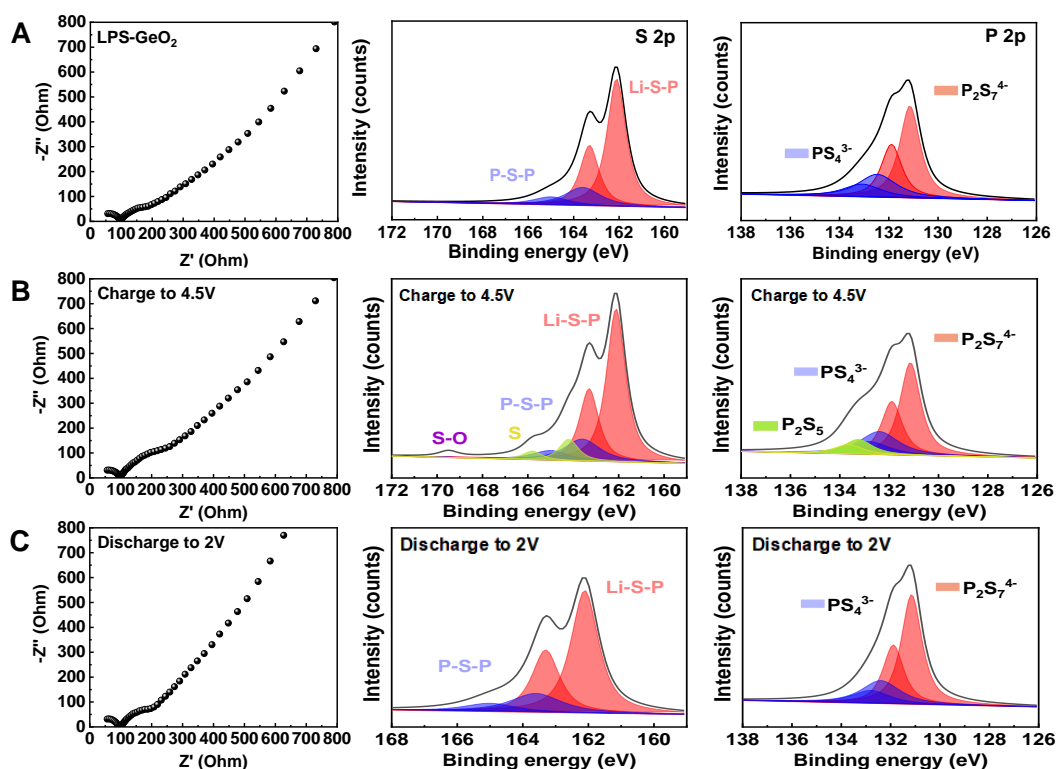


Figure S11. Nyquist plots and XPS spectra of cell with electrolytes LPS-GeO₂ (A) Before cycling , (B) Charge to 4.5 V, (C) Discharge to 2 V.

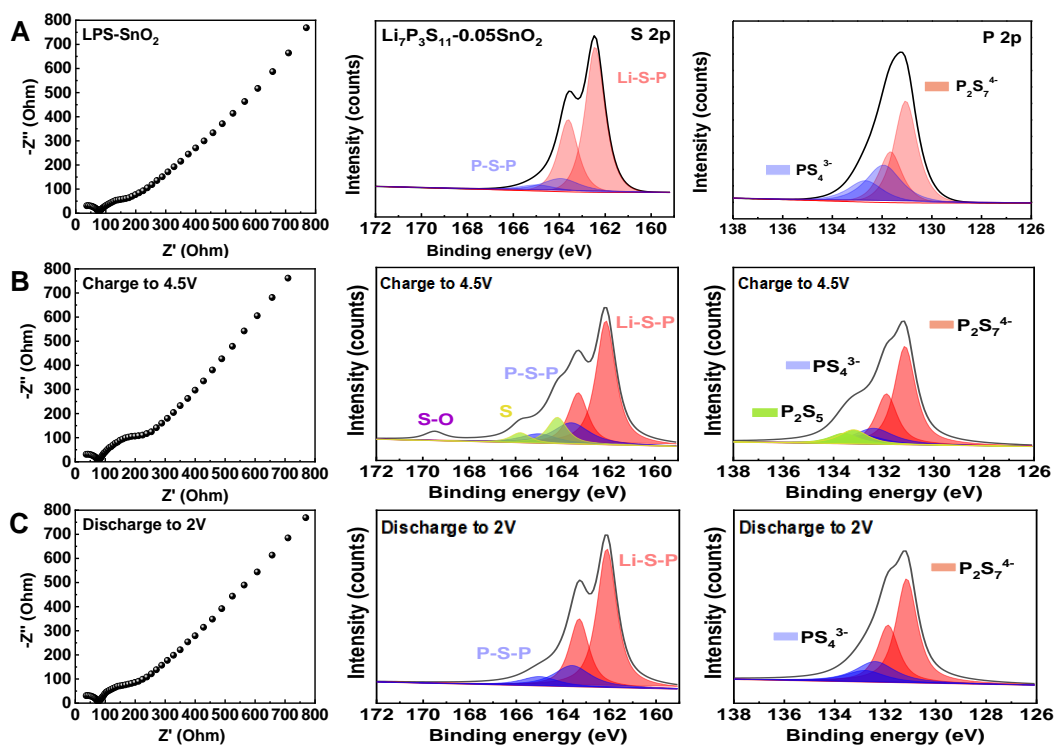


Figure S12. Nyquist plots and XPS spectra of cell with electrolytes LPS-SnO₂ (A) Before cycling , (B) Charge to 4.5 V, (C) Discharge to 2 V.

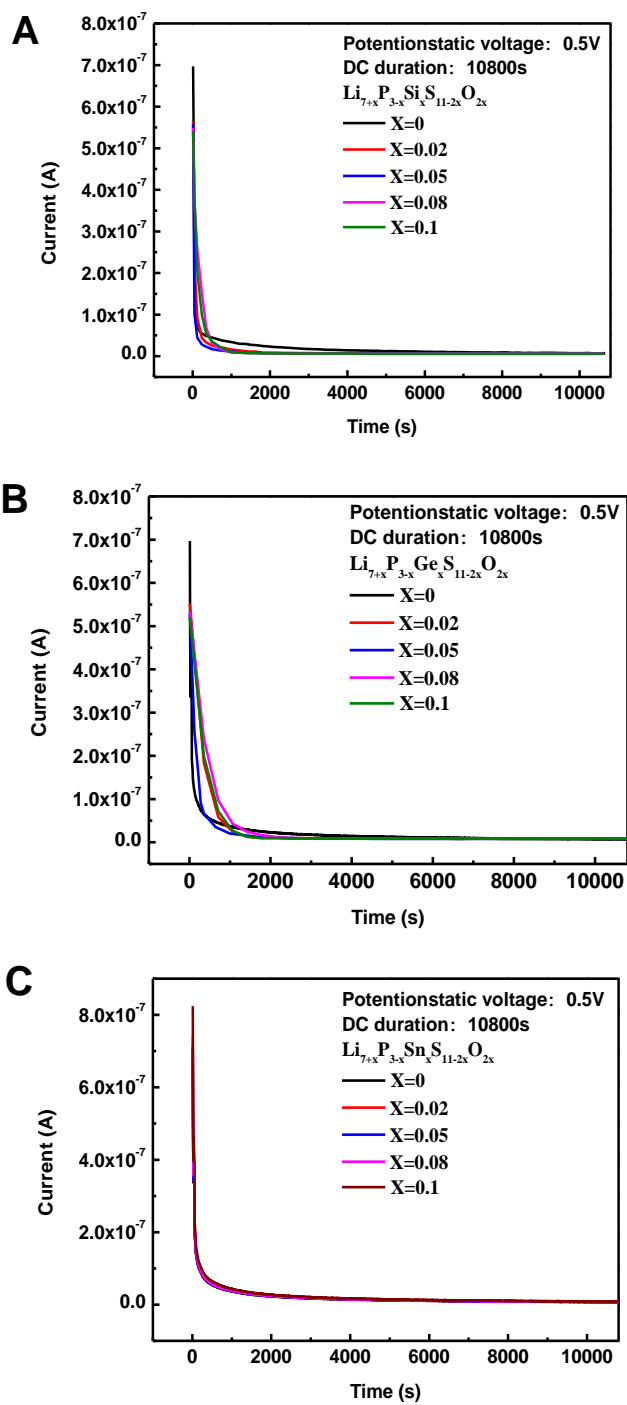


Figure S13. Current-time curve of the SS/SSEs/SS cells with the potentiostatic voltage 0.5V. (A) LPS-xSiO₂ (x=0, 0.02, 0.05, 0.08, 0.1). (B) LPS-xGeO₂ (x=0, 0.02, 0.05, 0.08, 0.1). (C) LPS-xSnO₂ (x=0, 0.02, 0.05, 0.08, 0.1).

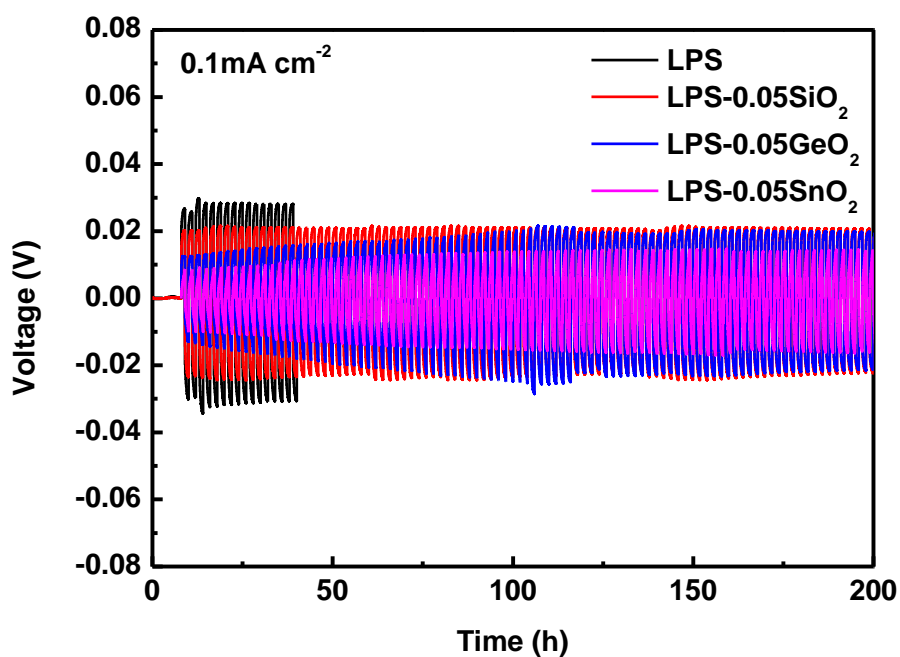


Figure S14. Galvanostatic charge/discharge voltage profiles of the Li/SSEs/Li symmetric cells at 0.1 mA cm^{-2} at room temperature.

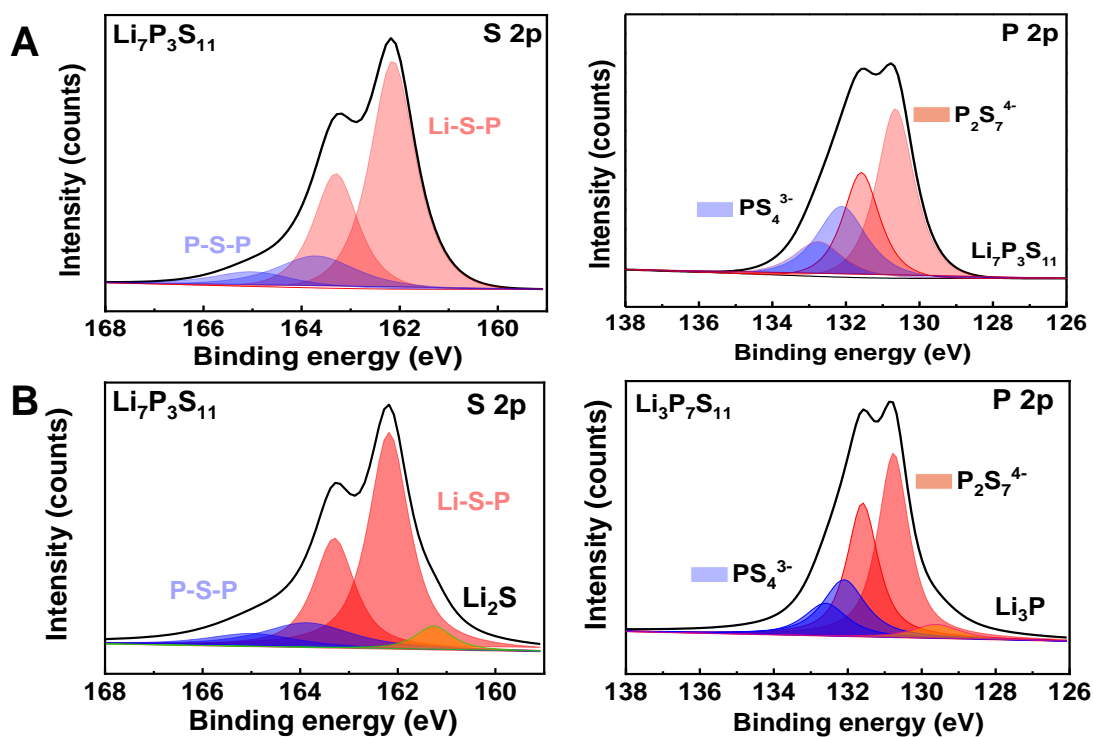


Figure S15. XPS detail spectra of pristine LPS electrolyte (A) Before cycling, (B) After five cycles

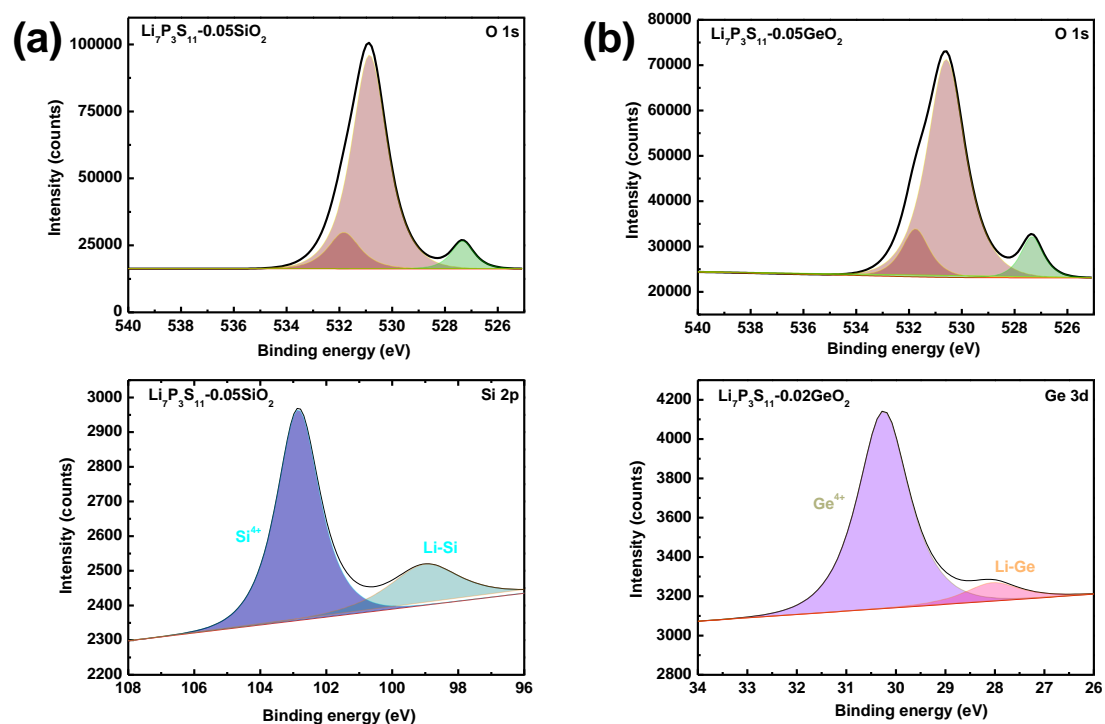


Figure S16. XPS detail spectra of (a) LPS-0.05SiO₂ and (b) LPS-0.05GeO₂ sulfide electrolyte after five cycles.

Supplementary Table S1. The lattice constant change of LPS-xMO₂ electrolytes with different doping conditions

	space group	a (Å)	b (Å)	c (Å)
LPS	P1	12.4502	6.0483	12.5215
LPS-0.02SiO₂	P1	12.4506	6.0486	12.5218
LPS-0.05SiO₂		12.4513	6.0491	12.5225
LPS-0.08SiO₂		12.4517	6.0498	12.5231
LPS-0.1SiO₂		-	-	-
LPS-0.02GeO₂		12.4539	6.0521	12.5244
LPS-0.05GeO₂	P1	12.4572	6.0562	12.5272
LPS-0.08GeO₂		12.4601	6.0611	12.5298
LPS-0.1GeO₂		-	-	-
LPS-0.02SnO₂	P1	12.4566	6.0531	12.5257
LPS-0.05SnO₂		12.4617	6.0597	12.5296
LPS-0.08SnO₂		12.4691	6.0654	12.5334
LPS-0.1SnO₂		-	-	-

Supplementary Table S2. Electronic conductive of treated electrolytes with different doping conditions

Electronic conductive ($\times 10^{-10} \text{ Scm}^{-1}$)					
	$x=0$	$x=0.02$	$x=0.05$	$x=0.08$	$x=0.1$
LPS- $x\text{SiO}_2$	12	11.75	11.44	10.76	10.18
LPS- $x\text{GeO}_2$	12	12.67	12.39	12.21	11.97
LPS- $x\text{SnO}_2$	12	12.19	12.28	12.73	14.19