Electrochemical Performance of Garnet based Lithium Metal Battery with Interface Modification.

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Supporting Information:

Figure S1:



Figure S1: XRD pattern of pristine NMC (red). Li₂SiO₃ (blue) and composite cathode (black)

Figure S2:



Figure S2: XRD pattern of pristine LLZA(red). Li₂SiO₃ (blue) and LS-LLZA (black)





Figure S3: Raman Spectrum of LLZA (blue) and LS-LLZA (red)

Figure S4:



Component	Equivalent circuit	Capacitance	Resistance (Ω cm ²)
	parts		
LLZA	R1, R2, Q2	$\sim 10^{-12} \mathrm{F}$	114
LS-LLZA	R3, Q3	~10 ⁻⁵ F	1308
NMC-LS	R4, Q4	~10 ⁻⁵ F	42.6
NMC	W5, Q5	~ 10 ⁻⁵ F	

Table 1: Equivalent circuit and parameter of NMC||LS-LLZA-LS||NM

Figure S5:



Figure S5: Cole-Cole plot of LS-LLZA-LS. The impedance data's were fitted with the equivalent circuit shown as inset in which R_1 corresponds to bulk resistance, R_2 corresponds to grain boundary impedance and Q_2 corresponds to constant phase element of LLZA, R_3 and Q_3 corresponds to charge transfer resistance and double layer capacitance on LS||LLZA interface.





Figure S6: Cole-Cole plot of Li||Au||LLZA||Au||Li symmetric cell after heat-treatment at 180 °C

Figure S7:



Figure S7: R_1 corresponds to bulk resistance, R_2 grain boundary impedance and Q_2 corresponds to constant phase element of LLZA, R_3 and Q_3 corresponds to charge transfer resistance and constant phase element of LS||LLZA and Au||LLZA interface, R_4 and Q_4 corresponds to charge transfer resistance and constant phase element of LS||NMC and Au||Li interface and W_5 and Q_5 corresponds to diffusion impedance inside of cathode.

Figure S8:



Figure S8: Charge-Discharge capacity of NMC||LLZA||Li all-solid-state cell without any interface modification.





Figure S9a: Cole-Cole plot of Li||GPE||LLZA||GPE||Li symmetric cell measured at 25 °C.

Figure S9b:



Figure S9b: Galvanostatic cycling performance of Li||GPE||LLZA||GPE||Li symmetric cell at a current density of 0.4 mAcm⁻². The inset shows the zoom-in figure with a stable voltage response.

Figure S9c:



Figure S9c: Galvanostatic cycling of Li||LLZA||Li symmetric cell without any interface modification. Spike like structure are observed because of large polarization between LLZA membrane and Li metal resulting in poor contact.