Supporting Information for

Constructing Robust Artificial Solid Electrolyte Interphase with Metal-Organic Framework for Stable Li Metal Anode

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Supplemental Experimental Procedures

Density Function Theory (DFT) Calculations. The DFT calculations are performed by the Vienna Ab initio Simulation Package (VASP) with the projector augmented wave (PAW) method. Generalized gradient approximation (GGA) of Perdew-Burke-Ernzerhof (PBE) functional is used for the exchange-functional. 20 Å of vacuum spacing in a direction perpendicular to the plane of the MOF is used to avoid interaction between units. The Brillouin zone integration is performed using 3 × 3 × 1 Monkhorst-Pack k-point sampling for a structure. The cut-off energy of the plane-wave basis is set at 400 eV. The self-consistent calculations apply a convergence energy threshold of 10^{-4} eV. The vibrational frequencies were computed to consider the zero-point energies, enthalpy, and entropy and ultimately calculate the free energies at room temperature (298.15 K).

Finite Element Modeling. Simplified model was adopted to investigate the concentration gradient of Li^+ in Li|Cu half-cell using the finite element solver of COMSOL Multiphysics. The simulations were performed using Electrodeposition model with the physics module "Tertiary Current Distribution". The ions flux was governed by the Nernst-Planck equation, while the current density, as a function of potential and Li^+ concentration, was given by the Butler-Volmer equation.

Supplementary Figures

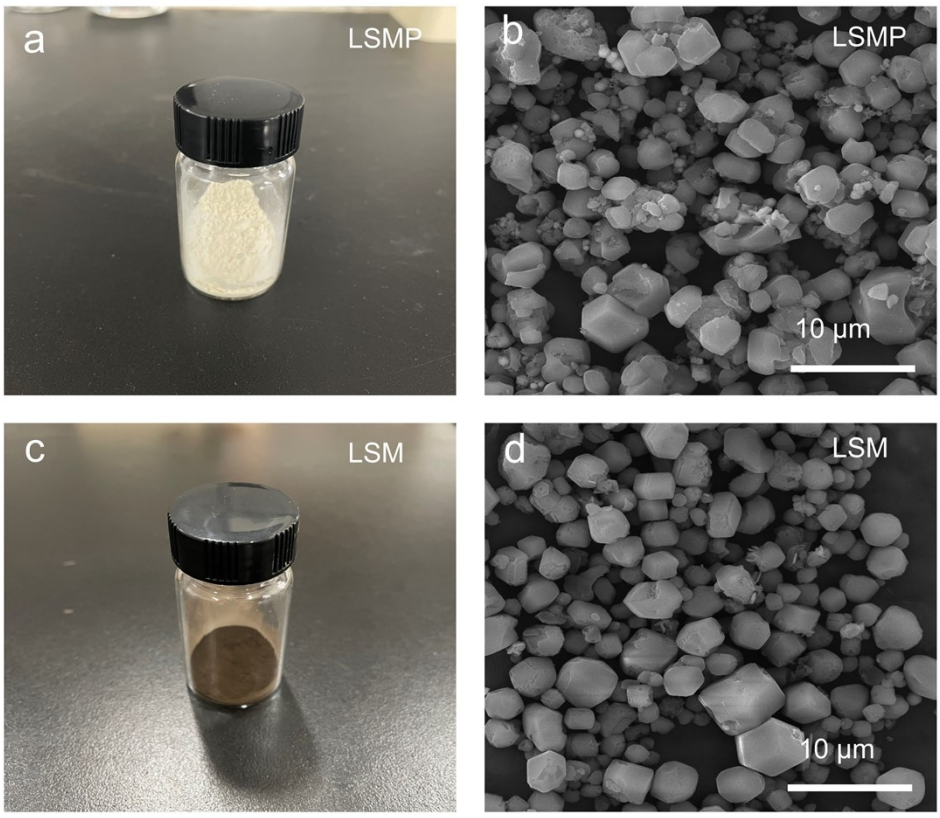

Figure S1. (a) Digital photos of LSMP; (b) SEM image of LSMP; (c) digital photos of LSM; (d) SEM image of LSM.

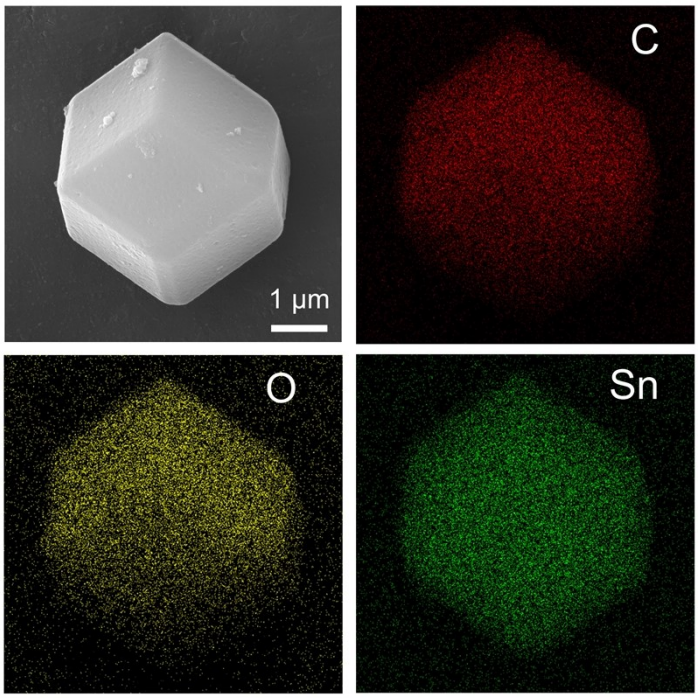

Figure S2. (a) SEM image of LSM; Elemental mapping images of LSM (b) C; (c) O and (d) Sn.

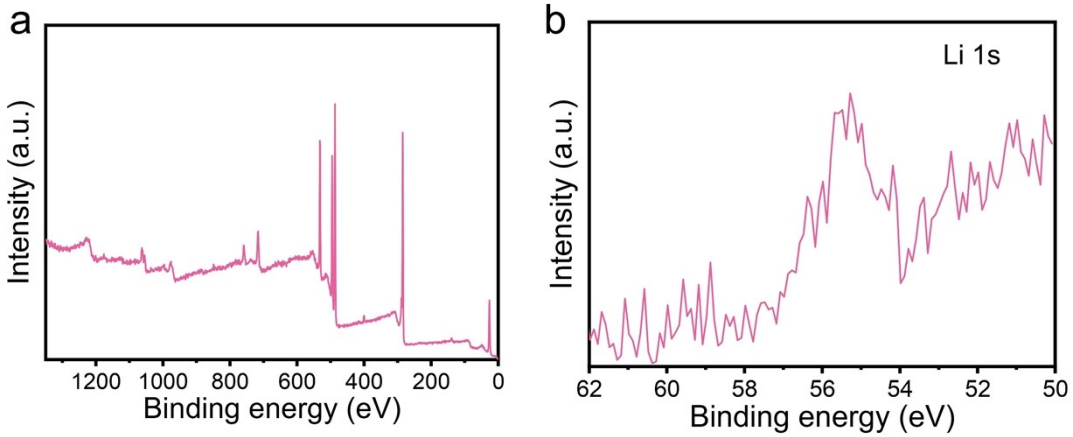

Figure S3. (a) XPS survey spectrum of LSM; (b) Li 1s XPS spectrum of LSM.

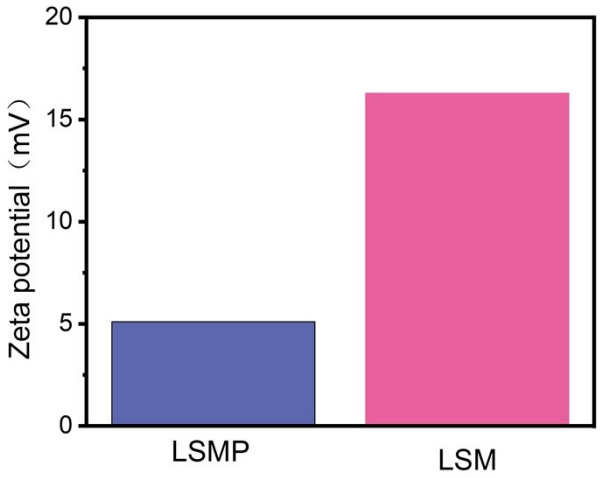

Figure S4. Zeta potential of LSMP and LSM.

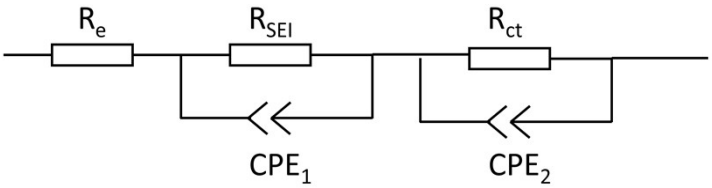

Figure S5. The equivalent circuit used to fit the Nyquist plots.

Supplementary Table

Table S1. Fitted R_{SEI} and R_{ct} values from the temperature-dependent EIS spectra

Temp (K)	R_{SEI} (Ω)		R_{ct} (Ω)	
	Cu-Li	LSM@Cu-Li	Cu-Li	LSM@Cu-Li
303	65.49	49.78	216.5	18.41
313	50.69	40.91	144.6	16.78
323	37.79	31.19	103.7	11.18
333	25.84	22.24	67.62	6.73
343	19.49	17.22	34.68	4.40