

Supporting Information

LAGP | Li interface modification through wetted polypropylene interlayer for solid
state Li-ion and Li-S batteries

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Synthesis of LAGP membrane

LAGP was synthesized through a typical solid state reaction method. Required amount of high purity chemicals Li_2CO_3 (99%), GeO_2 (99%), Al_2O_3 (99%) and $\text{NH}_4\text{H}_2\text{PO}_4$ (98%) (all from Aldrich) were ball milled using zirconia balls in 2-propanol for about 48 h using a Pulverisette 6, Fritsch, Germany. After the evaporation of the solvents at room temperature, the mixture was heated from room temperature to 700 °C in a platinum crucible and held at this temperature for 2 h to release the volatile compounds and then cooled down to room temperature. The resultant powder was grounded again for 24 h using zirconia balls in 2-propanol. After the evaporation of the solvent the powder was heated in a platinum crucible from room temperature to 1350 °C with a heating rate of 1°C per min and held at that temperature for 1 h. The melted LAGP was quickly cooled to 500 °C and held at that temperature for 2 h, followed by cooling down to room temperature. The as synthesized LAGP was then crystallized at 850 °C for 10 h. Finally the sintered LAGP was shaped to pellets of 14 mm diameter and 200 μm in thickness.

Structural characterizations

The phase purity and crystal structure of the prepared samples were examined by X-Ray Diffraction (XRD) using Bruker D8 advance Davinci diffractometer equipped with $\text{Cu K}\alpha$ radiation source ($\lambda = 1.5418 \text{ \AA}$) at a generator voltage of 40 kV and current of 30 mA with a scanning speed of 2 °/min (2θ). The surface morphology of the samples were characterized using field-emission scanning electron microscope (FESEM MIRA3 LMU) with EDX attachment for elemental composition analysis, TESCAN instrumentation. X-ray photoelectron spectroscopy (XPS) spectra were acquired on a Thermo Fisher Scientific Multilab 2000 spectrometer fitted with non-monochromatic $\text{Al K}\alpha$ radiation (1486.6 eV) as the X-ray source operated at a power of 150 W (12.5 mA, 12 kV).

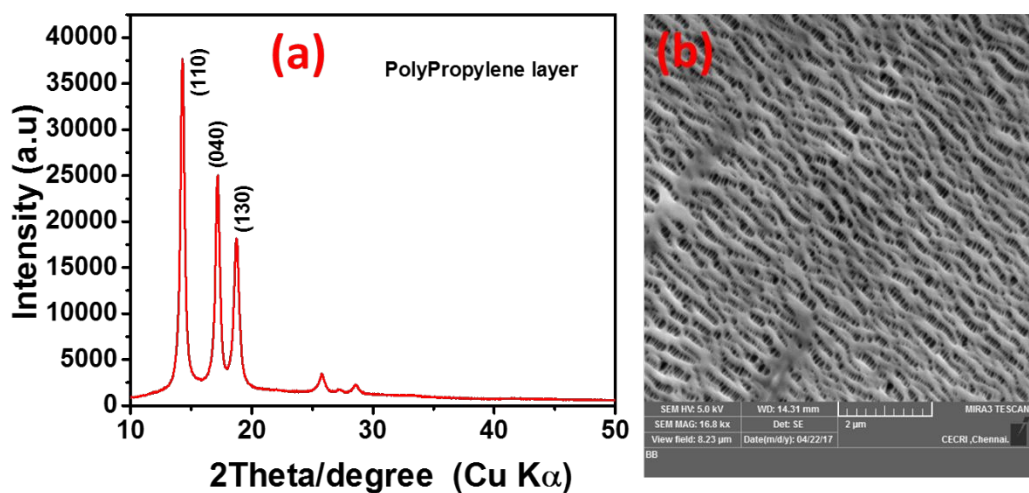


Figure S1. (a, b) The XRD and SEM of PP layer

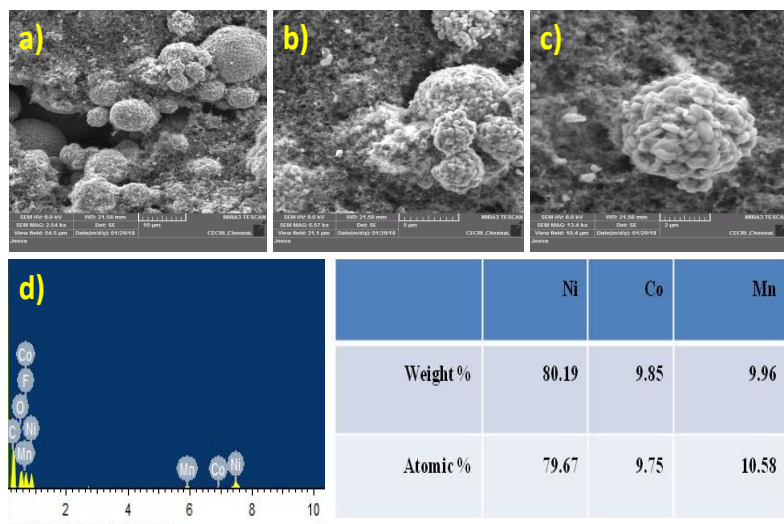


Figure S2. (a-c) SEM of LiNi_{0.8}Mn_{0.1}Co_{0.1}O₂ cathode, (d) EDAX and weight and atomic percent of LiNi_{0.8}Mn_{0.1}Co_{0.1}O₂.

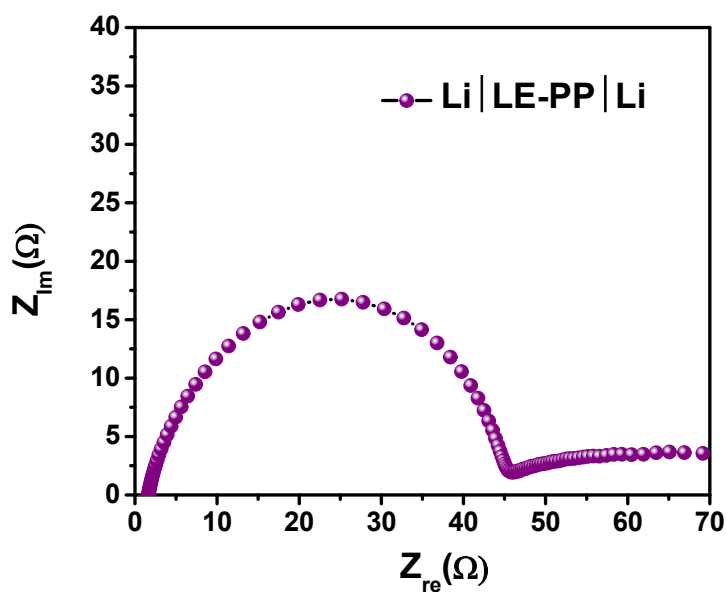


Figure S3. The impedance plot of Li | LE-PP | Li symmetric cell

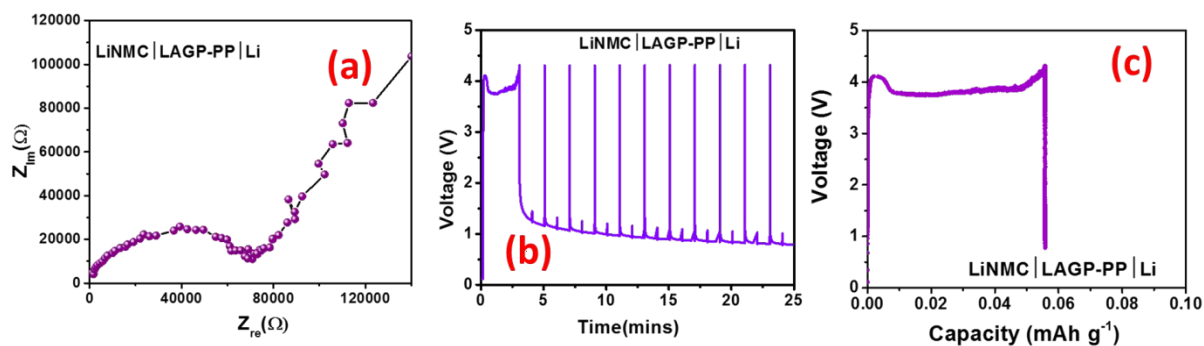


Figure S4. The electrochemical behavior of LiNMC | LAGP-PP | Li cell without wetting the separator with liquid electrolyte. (a) impedance, (b, c) cycling profile where (b) voltage Vs time and (c) voltage Vs capacity.

Synthesis of S@AB composite

60 mg of Sulfur was sonicated in 50 ml of DMF solvent until the sulfur dissolved into the DMF solvent. 40 mg of Acetylene black was added into the above solution and the mixture was

further sonicated for 2h. The resultant mixture was then transferred into 1000 ml distilled water, where the color changed into white indicate the formation of colloidal sulfur. The solution was stirred overnight till the colloidal sulfur was absorbed by the carbon. The solution was vacuum filtrated and dried at 65 °C overnight. The composite was labelled as S@AB.

Electrochemical characterizations for Li-S battery

The electrochemical characterization was performed by coating a composite mixture of 80% S@AB with 10 %SP Carbon and 10% PVDF binder on carbon paper and further vacuum dried at 80 °C in an oven overnight to remove the excess solvent. The total electrode weight was 3 mg/cm². Lithium metal foils (thickness 700 µm) were used as counter and reference electrode. LAGP membrane with 200 µm thickness was used as a solid electrolyte. The electrolyte was 1 M bis (trifluoromethane) sulfonamide lithium salt (LiTFSI, Sigma-Aldrich) dissolved in 1, 2-dimethoxyethane (DME, Sigma-Aldrich) and 1,3Dioxalane (DOL, Sigma-Aldrich) in 1:1 V /V% solvents. Coin cell were assembled in argon filled glove box (where O₂, H₂O levels restricted to ≤1ppm). The cells were galvano statically cycled in the voltage range of 1.7- 2.7 V. All the experiments were carried out in VMP3Z Biologic multichannel galvanostat/ potentiostat. The cyclic voltammetry was performed at 0.01mV scan speed within 1.7- 2.7 V and electrochemical impedance studies were performed over a frequency range of 400 kHz to 50 mHz. The capacities are calculated based on mass of sulfur in the electrode.