Technische Universität München

Influence of Mg on the Structure and Electrolyte/electrode Interface in All-solid-state Lithium Battery

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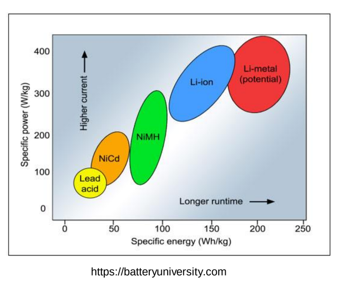
Background

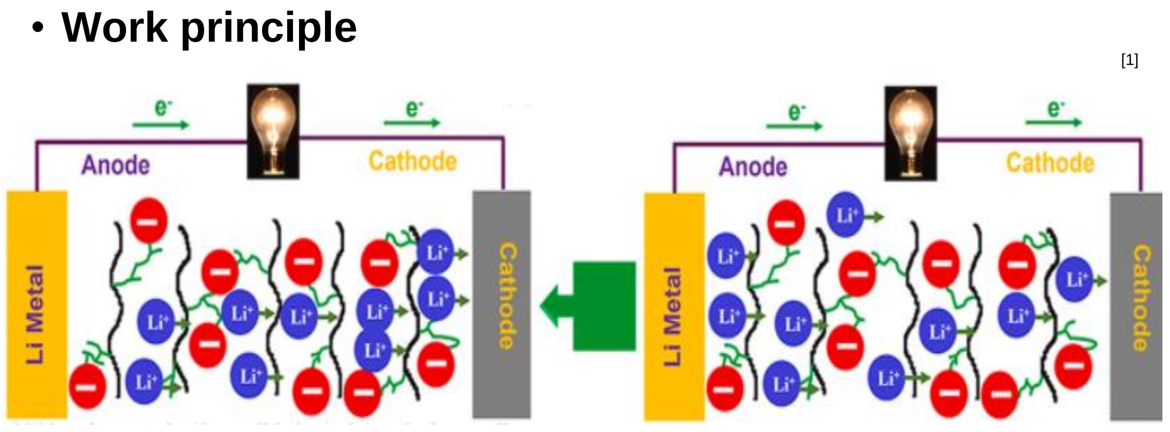
- **Lithium battery**



- Fossil fuel depletion
- Environmental pollution
- need clean energy

+ Lithium ion battery
+ Wide range application
+ High specific energy

- **Work principle**



Two electrode along with electrolyte embedded in between.

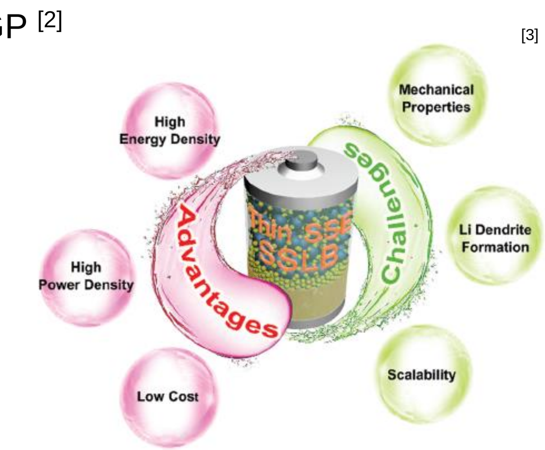
i. During charges and discharges, ions shuttle between two electrode.
ii. Discharging, the anode undergoes oxidation, the ion flow from anode to cathode; Charging reverse the direction.
iii. Li dendrites growth is a crucial challenge of battery.

All-solid-state lithium battery(ASSLB)

Solid state electrolyte

e.g., PEO, PAN, garnet LLZO, LAGP [2]

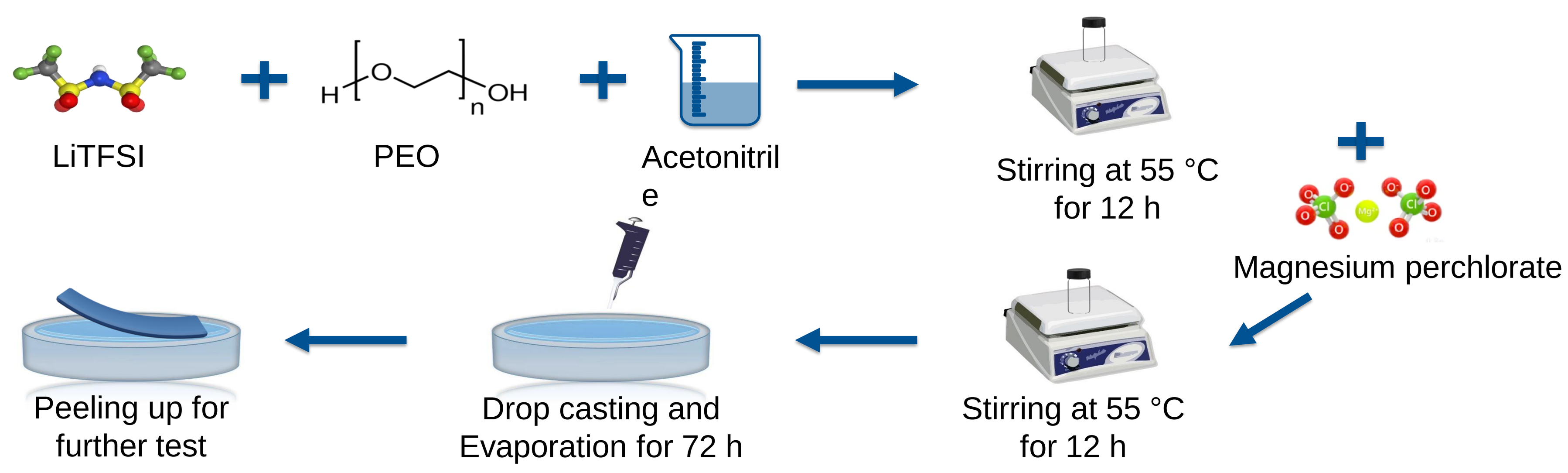
+ High thermal stability
+ Easy to synthesize
+ Flexibility
- Low conductivity at room temperature
- Interfacial instability



Aim of this work

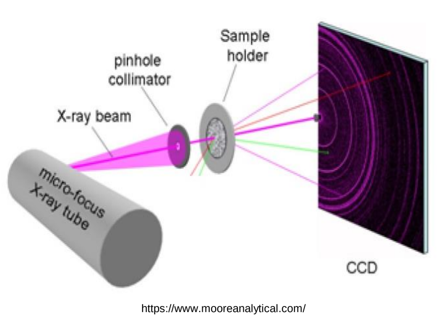
- Solve the low ionic conductivity and inhomogeneity interface formation problem of PEO electrolyte.
- Introduce $Mg(ClO_4)_2$ to modify structure and increase ionic conductivity.
- Construct a Li^+ conducting SEI between the electrolyte/electrode interface.

Sample preparation

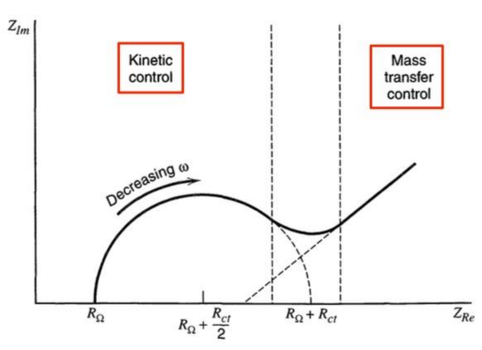


Characterization

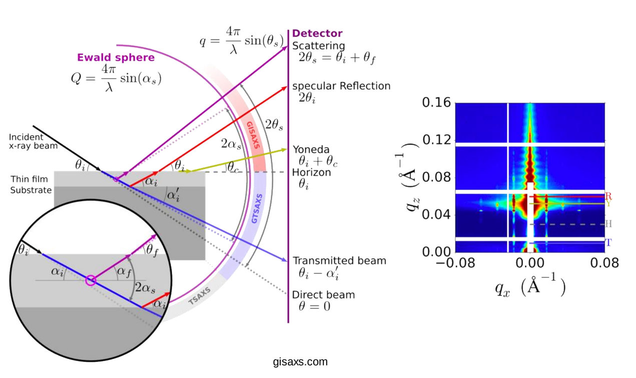
X-ray Diffraction



Electrochemical impedance spectroscopy



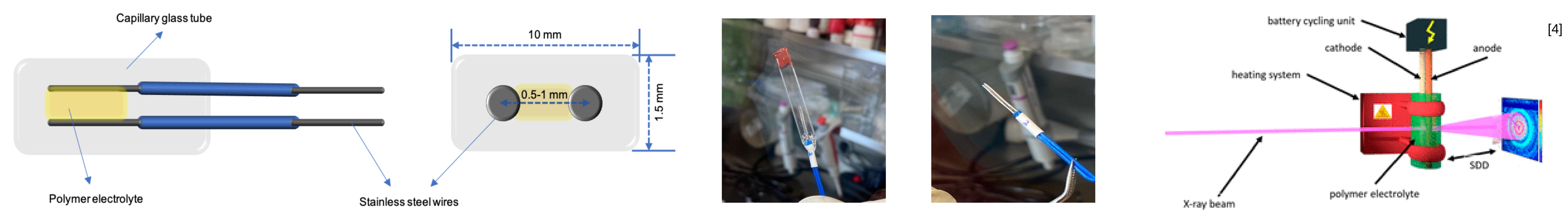
Small-Angle X-ray Scattering



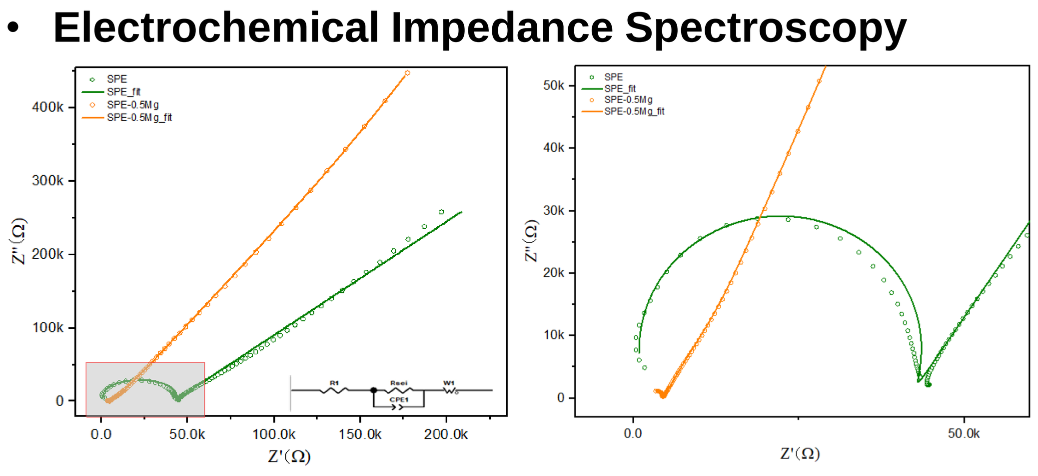
- Nondestructive small angle X-ray scattering method.
- Can follow the structural evolution of electrolytes.
- Use micro capillary cell as set up.

Results and discussion

- **Set up for measurement**

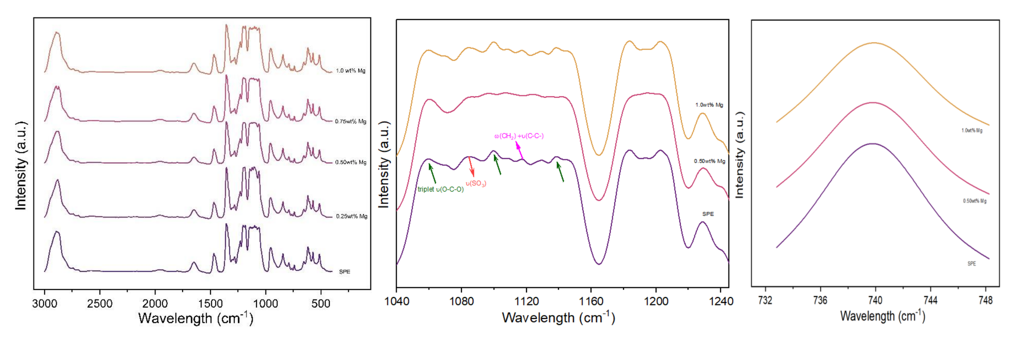


- **Electrochemical Impedance Spectroscopy**



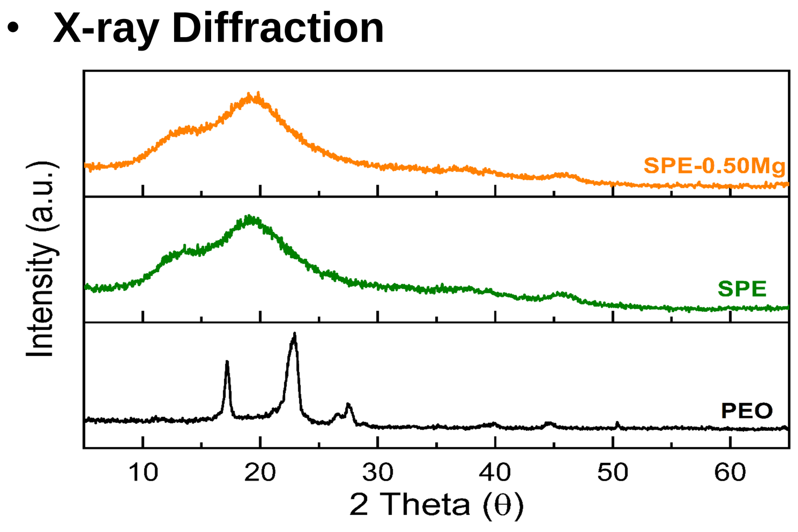
- A maximum conductivity of 3.79 x 10^{-4} S cm^{-1} with 0.5 wt% Mg
- Higher than the baseline sample (2.24 x 10^{-6} S cm^{-1})

- **Fourier-transform infrared spectroscopy**



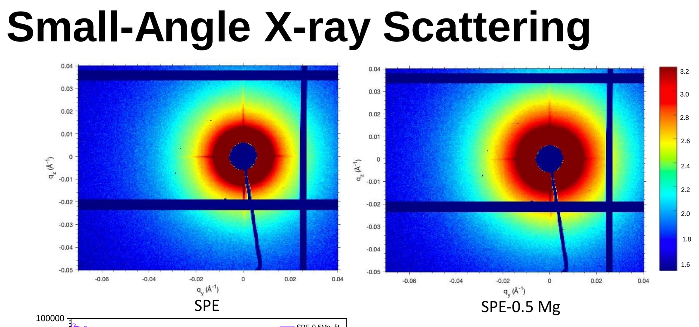
- Red-shift at peaks from 1040-1150 cm^{-1}
- High intermolecular interaction between $TFSI^-$ and Mg^{2+}
- Decrease intensity at 740 cm^{-1} means increase of free Li^+

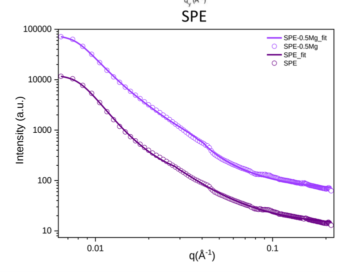
- **X-ray Diffraction**



- Crystalline peaks of PEO disappeared
- Broaden halo region indicates the amorphicity increase of electrolyte

- **Small-Angle X-ray Scattering**





- Radial cut on 2D image
- Fit the curve with polyelectrolyte model

Conclusion

- Solid electrolyte has higher thermal stability but suffers from the low conductivity and unstable interface problem.
- In this work, the PEO-based polymer electrolyte with inorganic additives is fabricated.
- $Mg(ClO_4)_2$ can promote the dissociation of Li^+-$TFSI^-$ ion pairs and increase the amount of mobility Li^+ ions.
- The formation of SEI layer can be detected during cycling with neutron reflectometry technology in the future.

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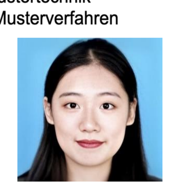

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Chair for Functional Materials

Reference

[1] Irfan, M., et al. "Recent advances in high performance conducting solid polymer electrolytes for lithium-ion batteries." Journal of Power Sources **486**.
[2] Li, Z., et al. "Mitigating Interfacial Instability in Polymer Electrolyte-Based Solid-State Lithium Metal Batteries with 4 V Cathodes." ACS Energy Letters **5**(10): 3244-3253.
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[4] Möhl, G.E., et al. "In Operando Small-Angle X-ray Scattering Investigation of Nanostructured Polymer Electrolyte for Lithium-Ion Batteries." ACS Energy Letters **3** (7): 1525-1530.