

Korea-EU Horizon Europe Researchers Networking Forum on Energy & Advanced Mobility

Taehyo Kim

Principal Researcher

KITECH

**Energy Devices
(ASSBs, PEMFCs)**

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I. Introduction

Mission

Contributing to national industrial development through **the development and commercialization of fundamental industrial technologies in the manufacturing sector**, as well as by supporting small and medium-sized enterprises (SMEs) and mid-sized companies and promoting the dissemination of technological achievements.

Univ.

Research Institute

Industrial field



Basic

Fundamental Industrial

Industrial Technology

Applied Research

Commercialization

산업핵심 선도기술개발

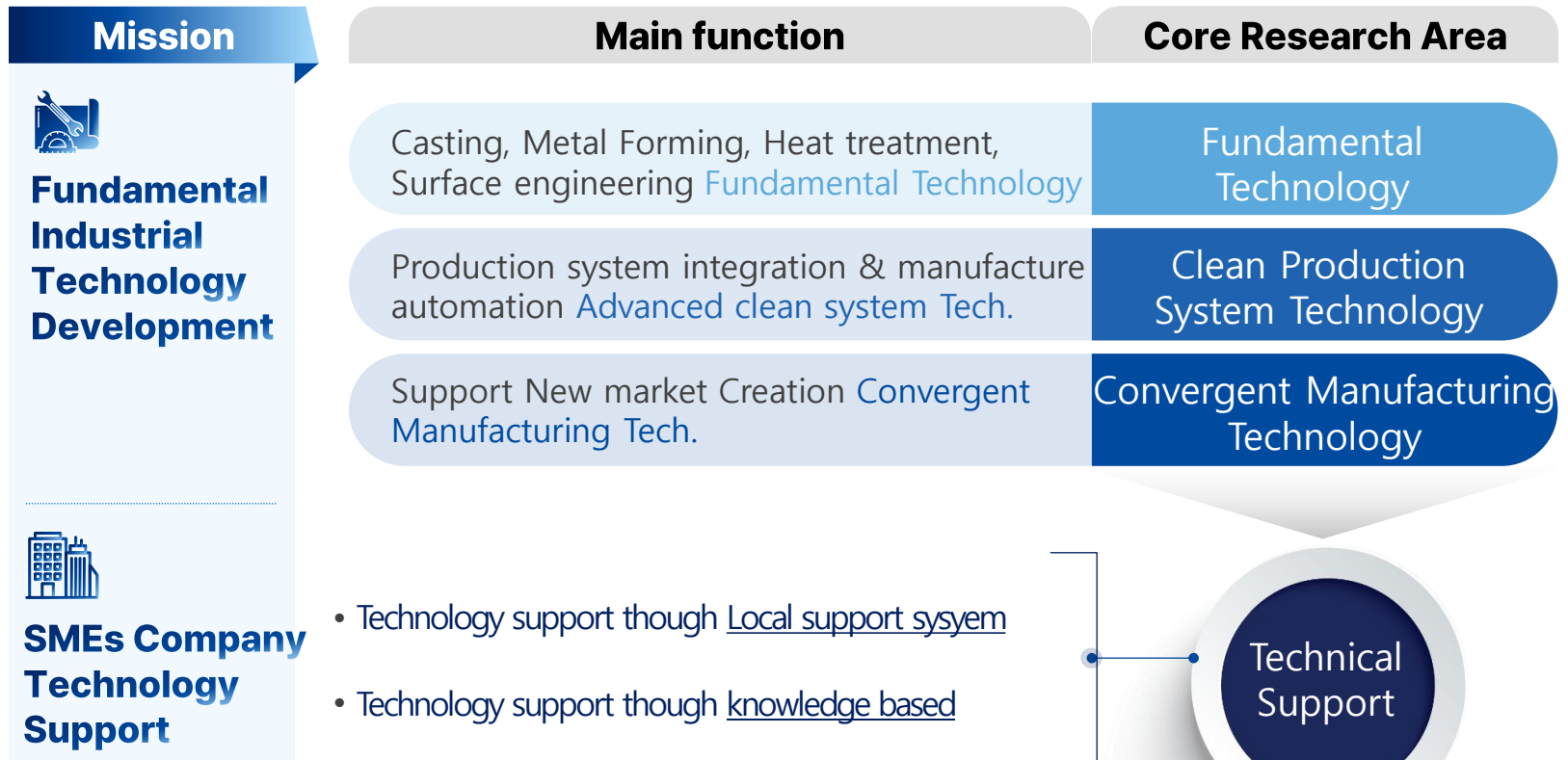
기업 공통 수요 기술개발

기술/인재/인프라 활용 기술서비스

중소·중견기업 기술지원 및 기술이전

I. Introduction

Focusing on R&D three key areas and commercialization, as well as developing SMEs through technical support



I. Introduction

Electric Vehicles(EVs) Necessity



자료: 연합뉴스

- CO₂ emission of EV about 1/3 the average CO₂ equivalent of Internal Combustion Engine (ICE) Vehicles.
- Reducing CO₂ by **2 tons** per EV/year

I. Introduction

Lithium Ion Batteries Issues

Low energy density

LIBs Energy density limit
: 250 ~ 300 Wh/kg

EV



500km

The short mileage of EVs

ISE



600km

Source: Samsung SDI.

Low safety

Risk of ignition
and explosion

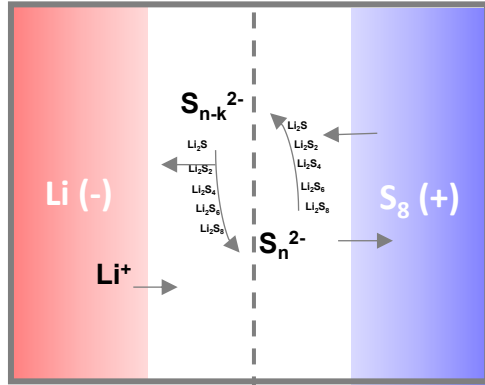


Source: Tesla Model S.

**New high-performance secondary batteries
that surpass LIBs are needed**

I. Introduction

Next generation batteries



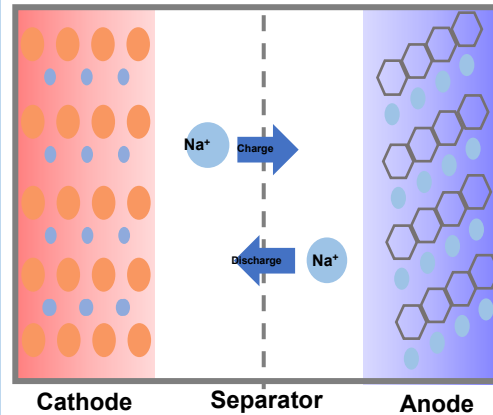
Lithium-Sulfur (Li-S) battery

Pros

- High capacity
- Low manufacturing costs

Cons

- H_2S gas generation
- Low battery life



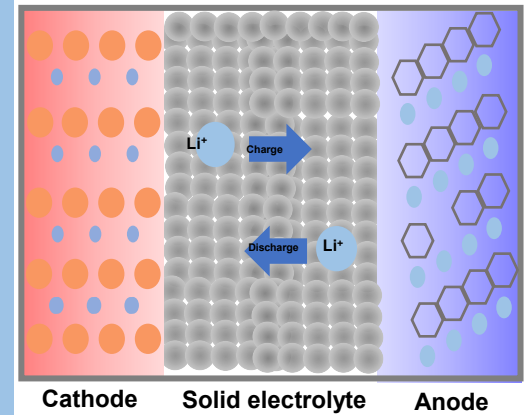
Sodium-Ion (Na-Ion) battery

Pros

- High capacity
- Low manufacturing costs

Cons

- H_2S gas generation
- Low battery life



All-Solid-State battery

Pros

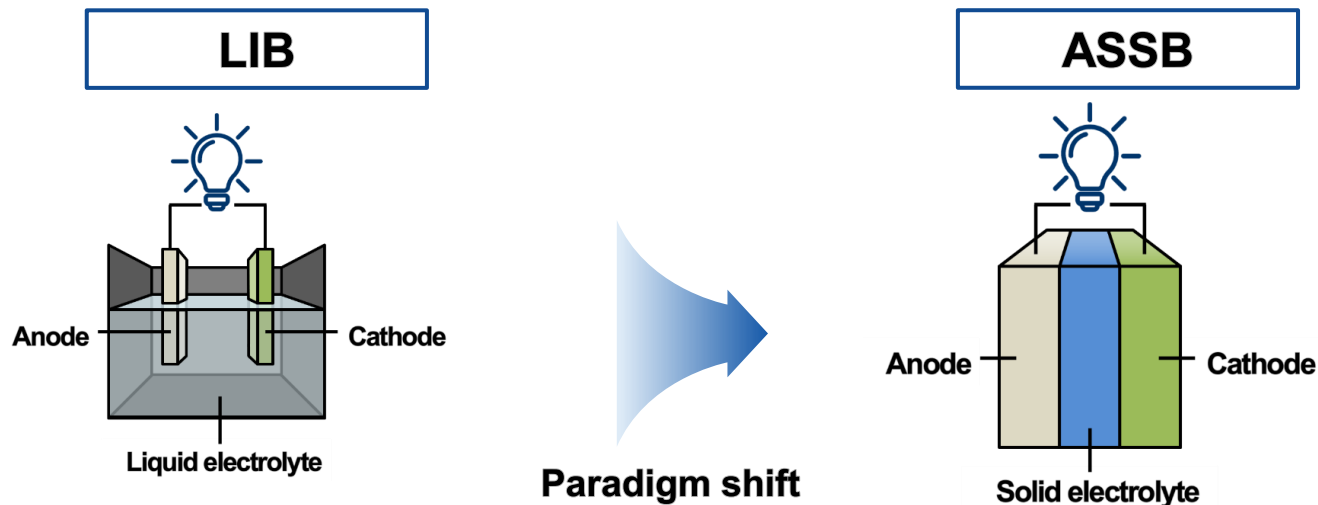
- High stability
- High energy density

Cons

- Low ionic conductivity
- Interfacial resistance

I. Introduction

All-Solid-State Batteries(ASSBs) Necessity



❖ Liquid electrolyte

- ✓ Leakage
- ✓ Narrow range of operation temperature
- ✓ Deformation, expansion, and explosion upon heating

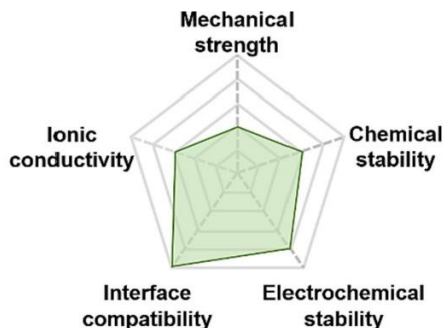
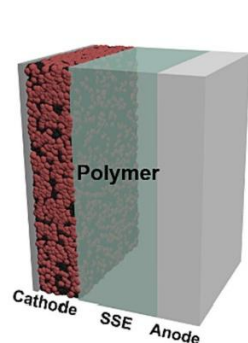
❖ Non-explosive

- ✓ **Good safety**
- ❖ Smaller and more lightweight
 - ✓ **High energy density**
- ❖ Problems
 - ✓ Low ionic conductivity

Dramatically improved **safety** and **energy density**
Research is needed to improve **ionic conductivity**

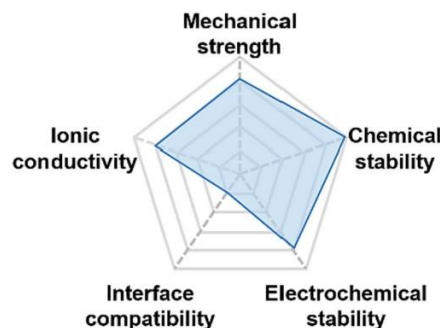
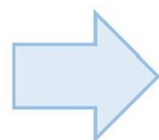
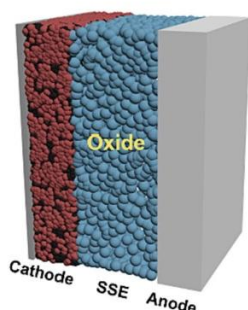
I. Introduction

Performance of Different Solid Electrolyte (SE) Materials



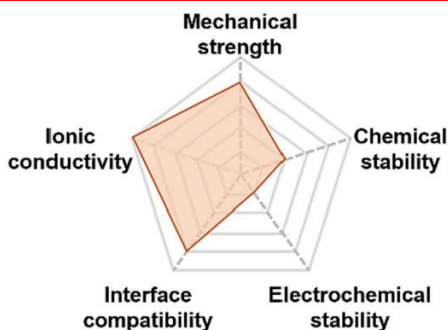
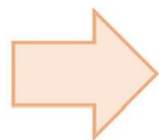
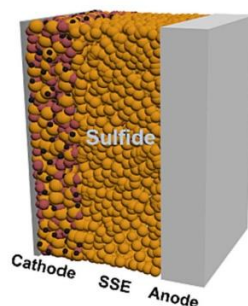
Polymer SEs

- ✓ **Good interfacial stability**
- ✓ **High electrochemical stability**
- ✓ **Low mechanical strength**
- ✓ **Low ion conductivity**



Oxide SEs

- ✓ **Good chemical stability**
- ✓ **High electrochemical stability**
- ✓ **High contact resistance**

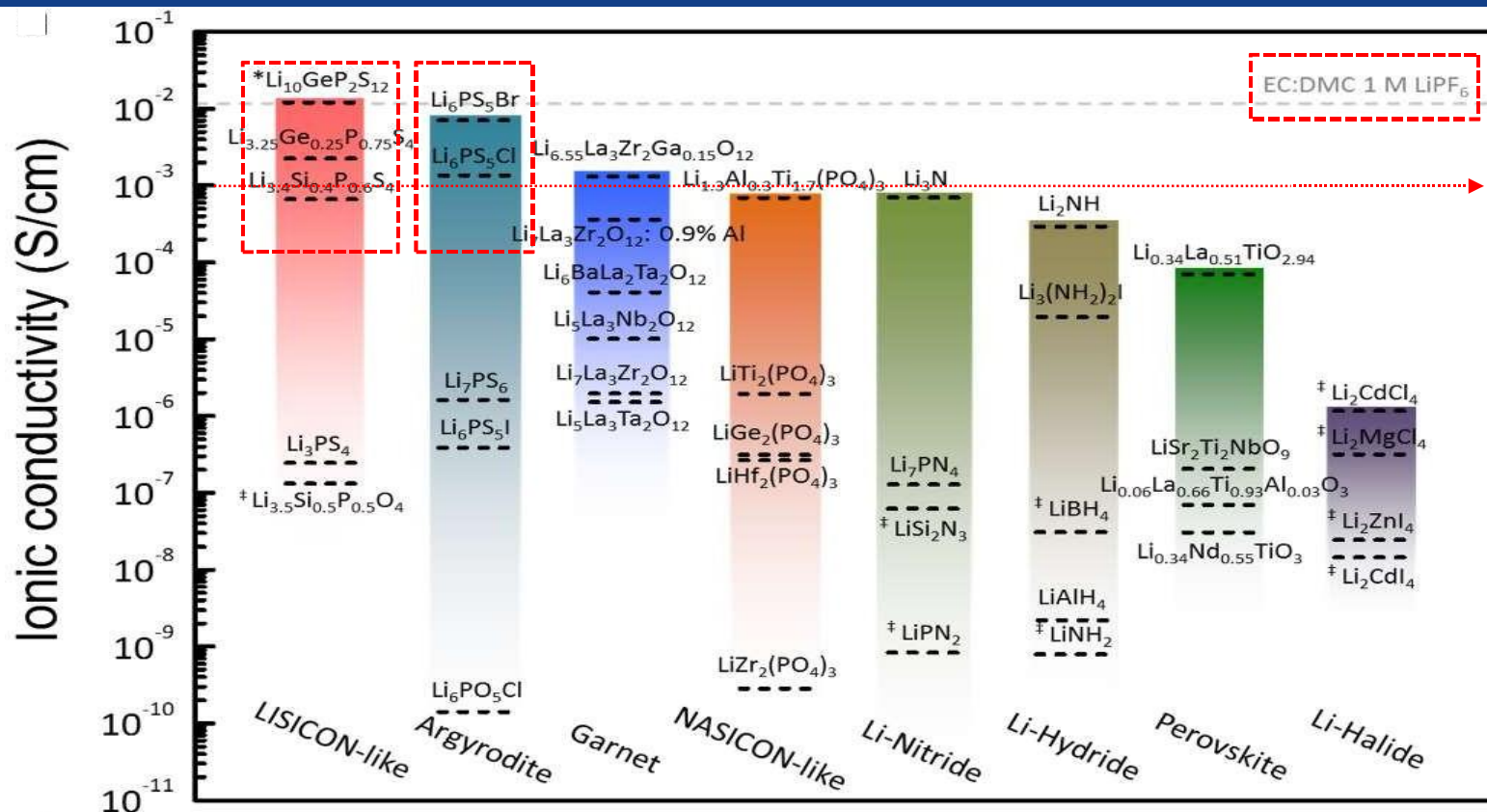


Sulfide SEs

- ✓ **Good ion conductivity**
- ✓ **High mechanical strength**
- ✓ **Poor air stability**
- ✓ **Li-dendrites growth**

I. Introduction

Ion conductivity



(1) Ion conductivity of solid electrolyte in RT.

LISICON-like (LISICON, thio-LISICON and $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$), Argyrodite (LPSCI), Garnet, NASICON-like, Li-nitride, Li-hydride, Perovskite and Li-halide.

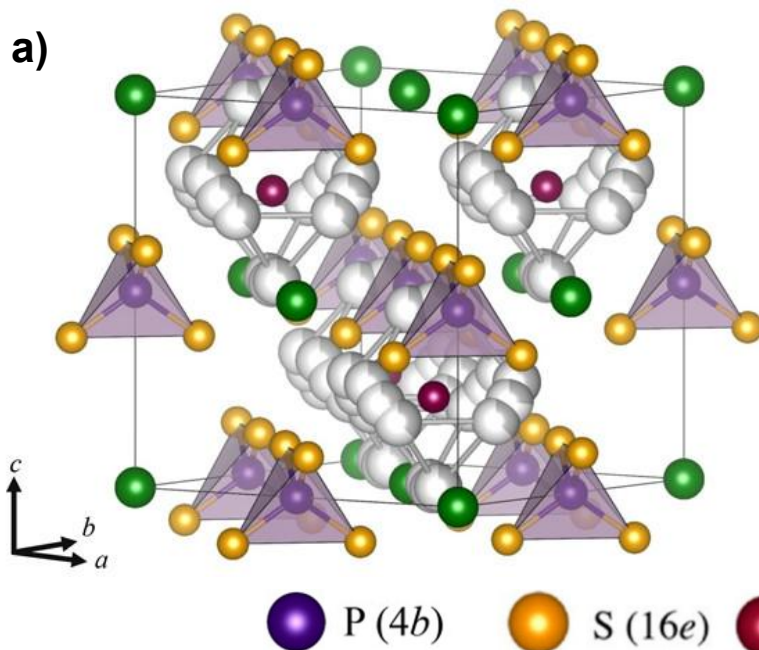
(2) Liquid Electrolyte (EC:DMC 1 M LiPF_6) $\sim 10^{-2}$ S/cm

(3) LiGPS & LiPSX higher ionic conductivity ($> \sim 10^{-3}$ S/cm)

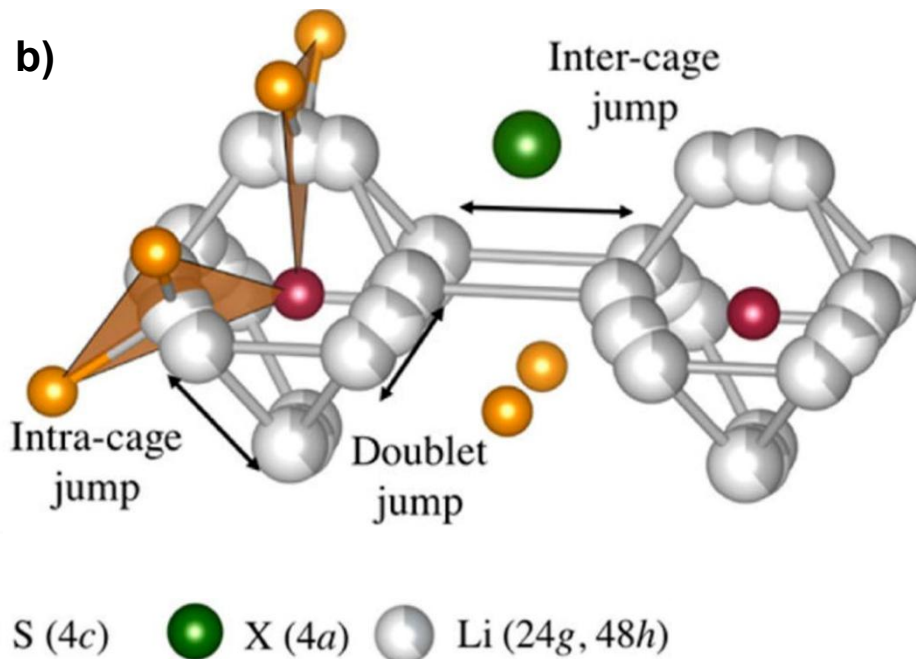
II. Theme I_Al-Cl Doping

Motivation of Al-Cl Doping

Structure of argyrodite $\text{Li}_6\text{PS}_5\text{X}$



(a) Kraft, Marvin A., et al. *Journal of the American Chemical Society* 140.47 (2018): 16330-16339.
(b) Park, Jae-Sang, Chang-Heum Jo, and Seung-Taek Myung. *Energy Storage Materials* (2023): 102869.



✓ Li ions migrate in three possible jump processes

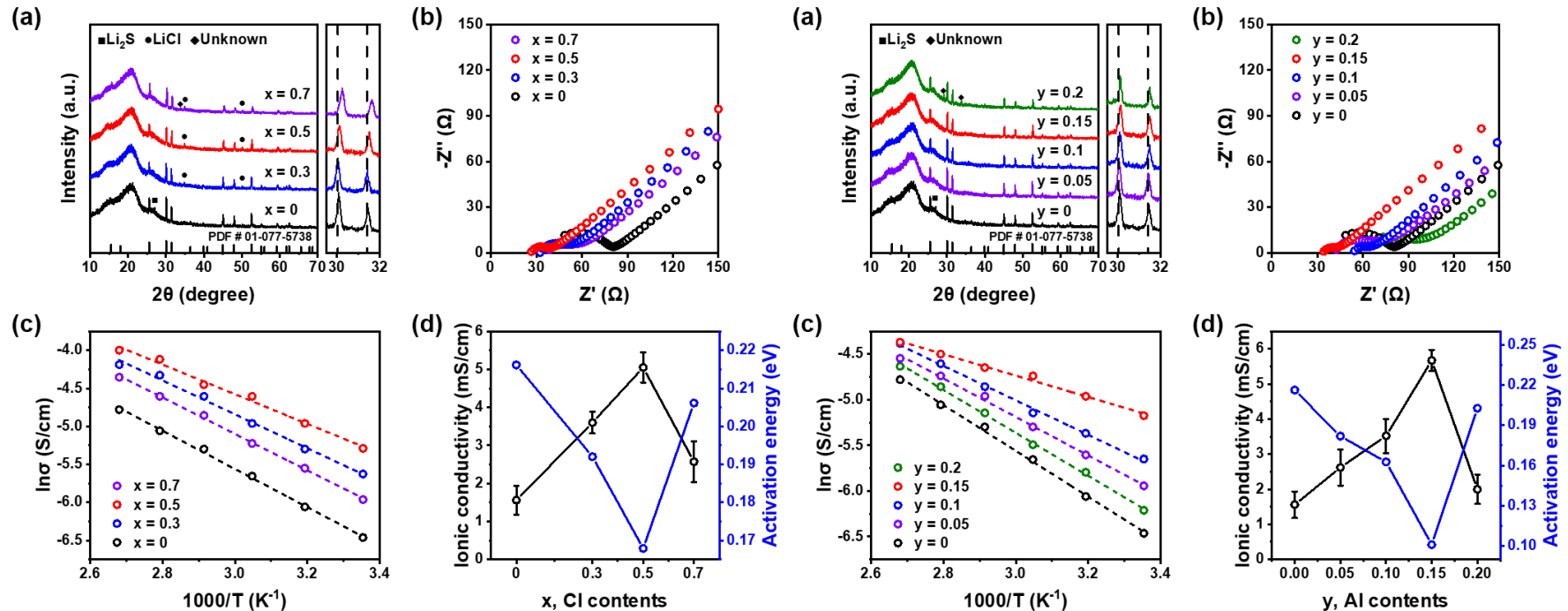
- 1) Inter-cage jump
- 2) Intra-cage jump
- 3) Doublet jump

- ✓ Aliovalent doping of the Li^+ site with Al^{3+}
→ generates Li^+ vacancies
- ✓ Cl^- substitution at mixed $\text{S}^{2-}/\text{Cl}^-$ sites
→ induces vacancies on the Li sites

II. Theme I_Al-Cl Doping

Cl and Al Doped Solid Electrolytes

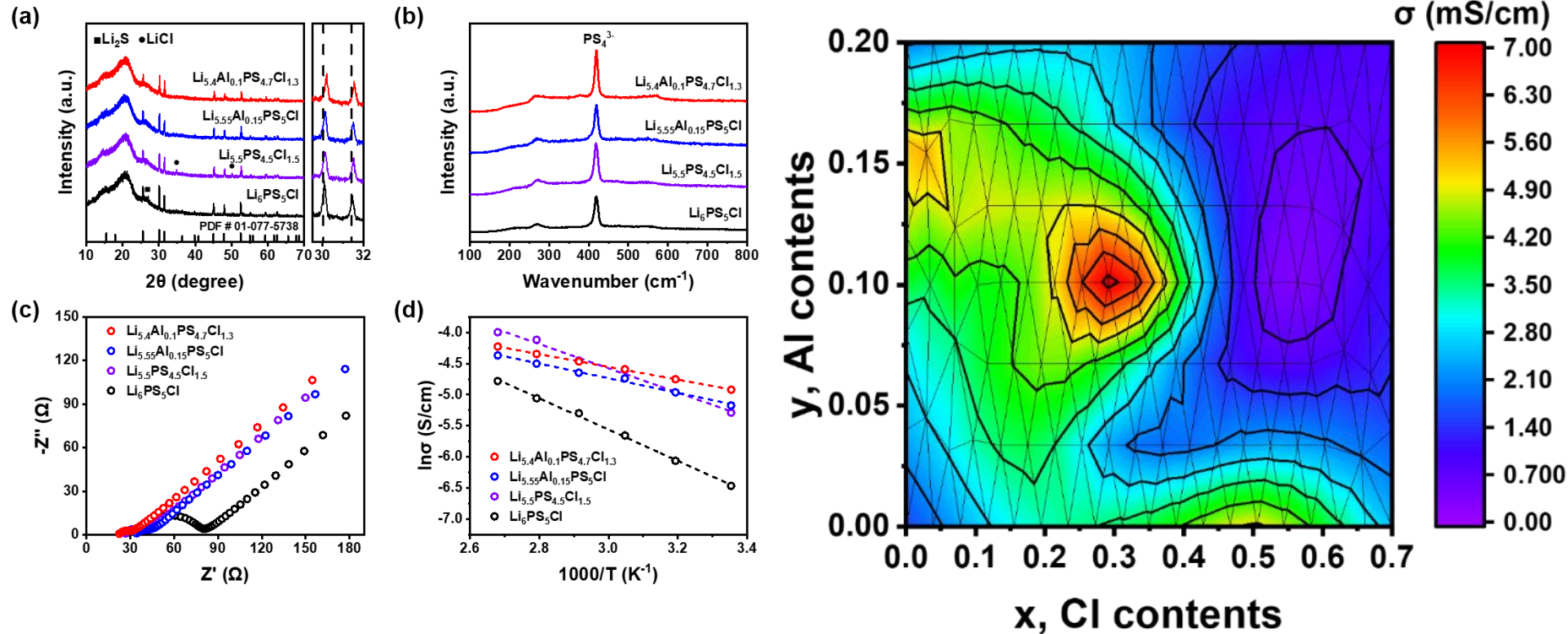
Kim et al. *Nanomaterials* 12.24 (2022): 4355.



- ✓ Cl doping causes the disorder of the $\text{Cl}^-/\text{S}^{2-}$ site, and the ionic conductivity can be improved by forming a vacancy site in Li due to the increase in the disorder of the $\text{Cl}^-/\text{S}^{2-}$ site.
- ✓ The vacancy of Li^+ sites is increased by substituting a cation with a radius similar to that of Li^+ ions. This means that the ionic conductivity was improved by forming a diffusion pathway.

II. Theme I_Al-Cl Doping

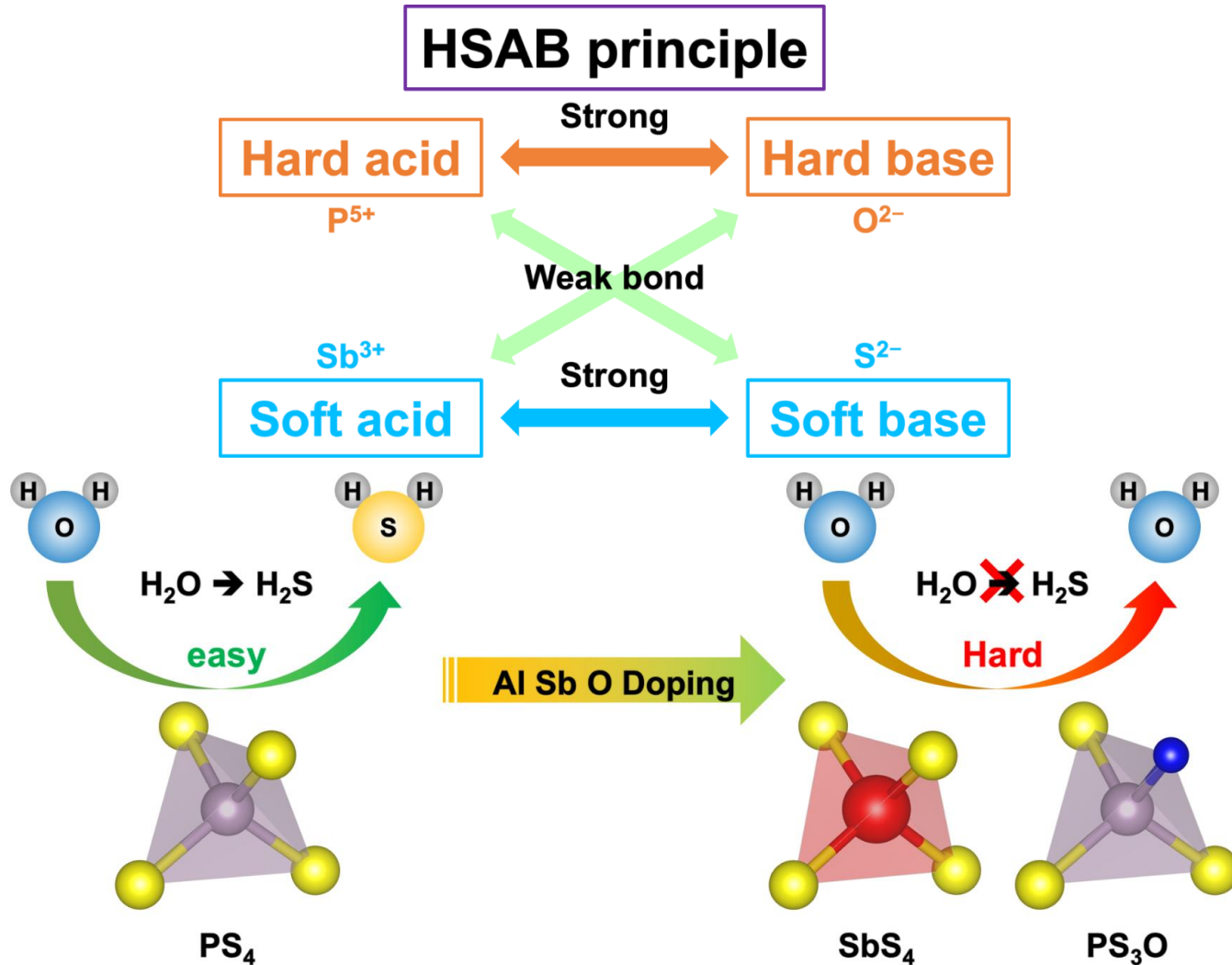
Cl-Al Doped Solid Electrolytes



- ✓ The two effects work together to lower the E_a and increase the ionic conductivity and Li diffusion, provided that the solubility limit of the lattice for either dopant is not exceeded.

II. Theme I_Al-Sb-O-Cl Doping

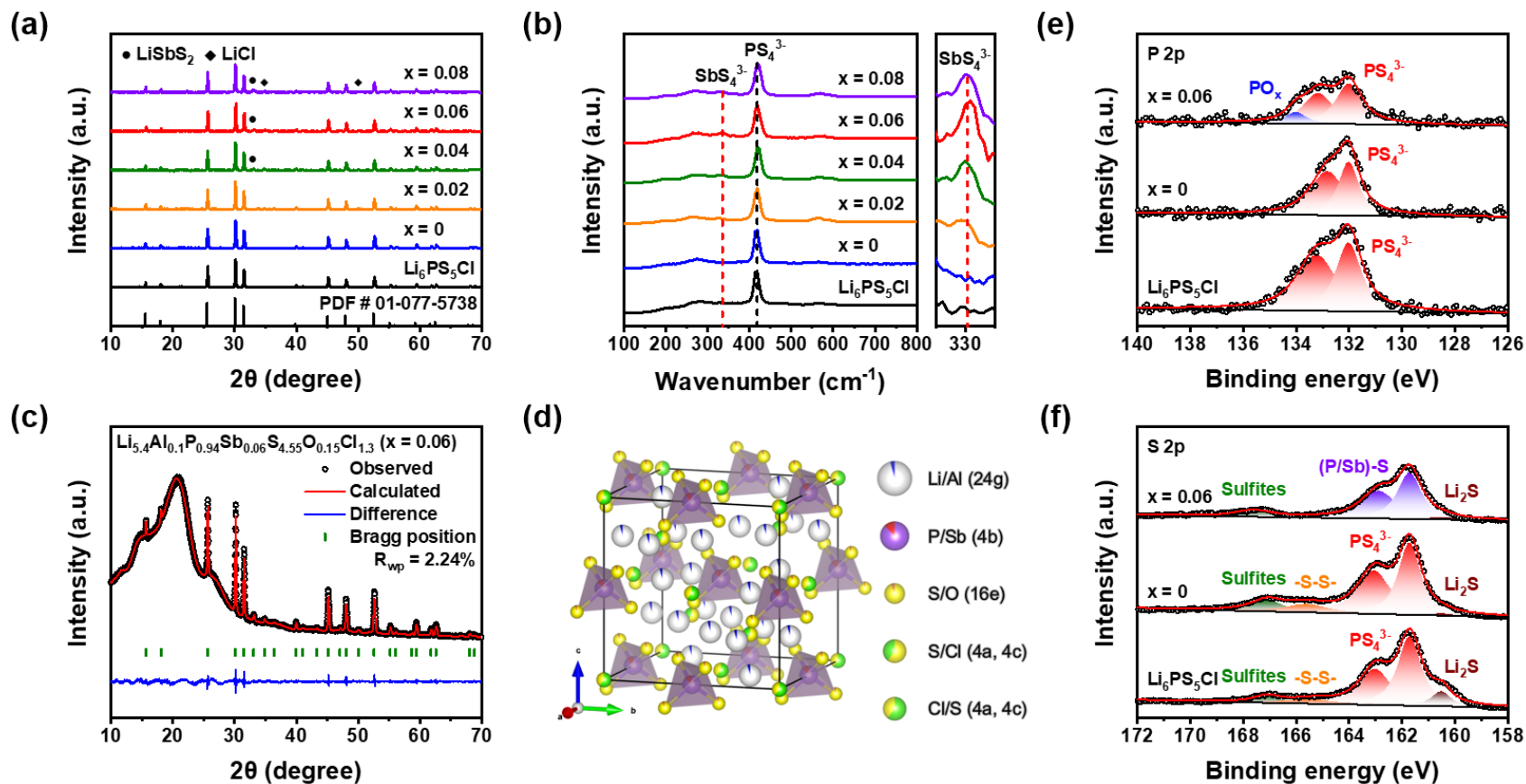
Motivation of Sb-O Doping



II. Theme I_Al-Sb-O-Cl Doping

Structural Analysis of Sb-O Doped Solid Electrolytes

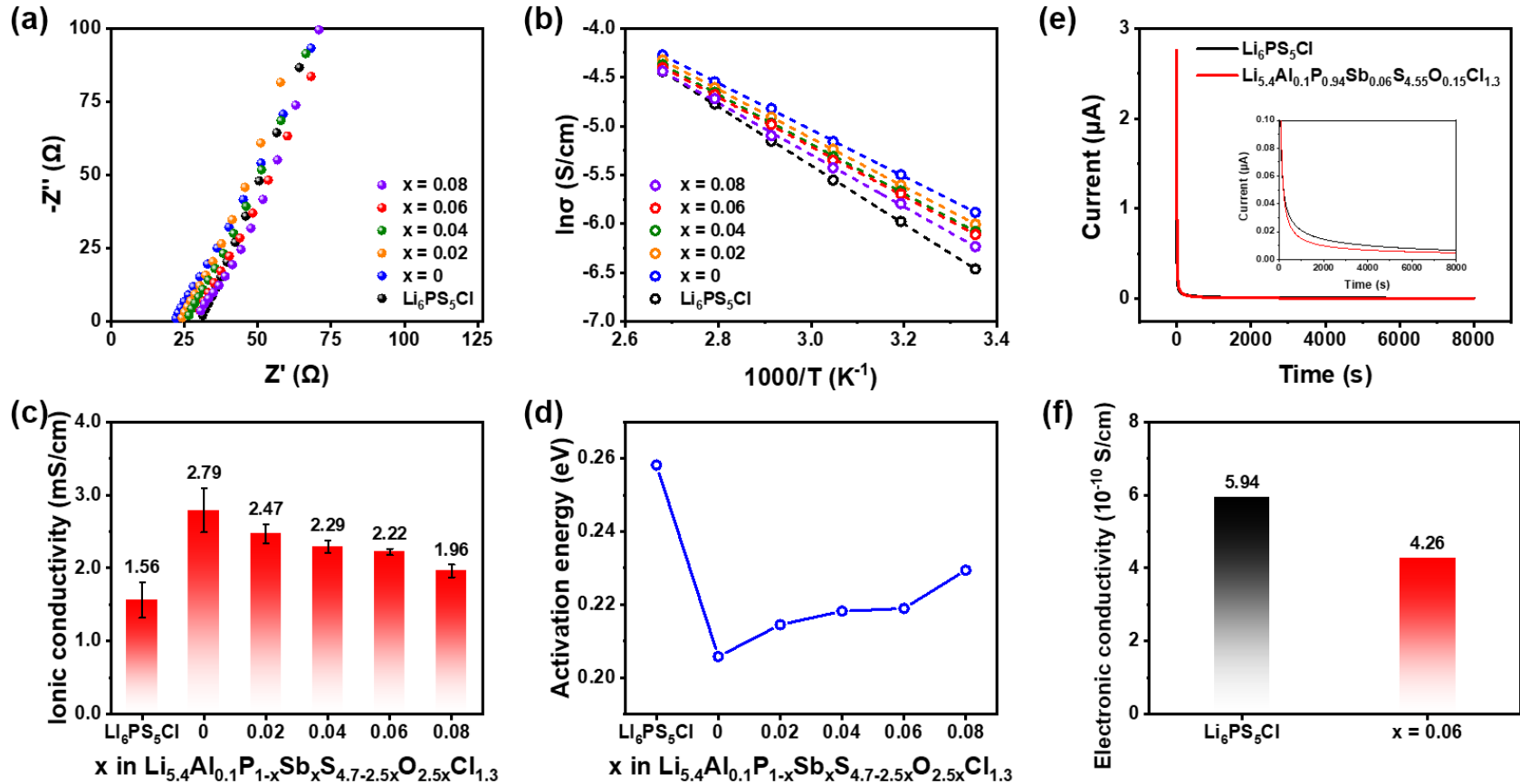
Kim et al, *Chemical Engineering Journal*, 497, (2024) 154426.



- ✓ Structural analyses revealed successful Al-Sb-O-Cl multiple-doping in $\text{Li}_6\text{PS}_5\text{Cl}$ electrolyte.
- ✓ Sb and O replaced P and S without damaging the argyrodite structure.

II. Theme I_Al-Sb-O-Cl Doping

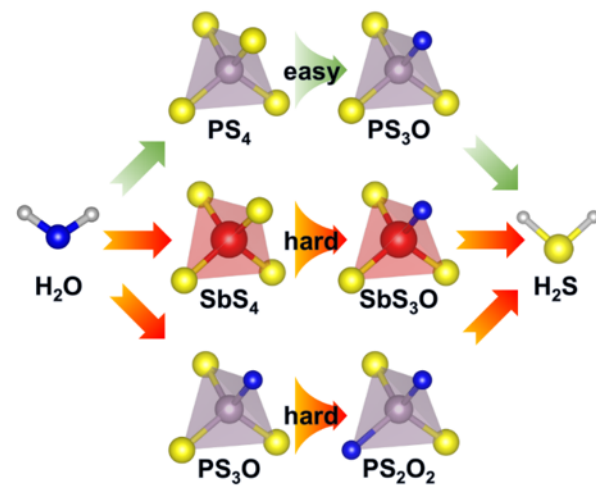
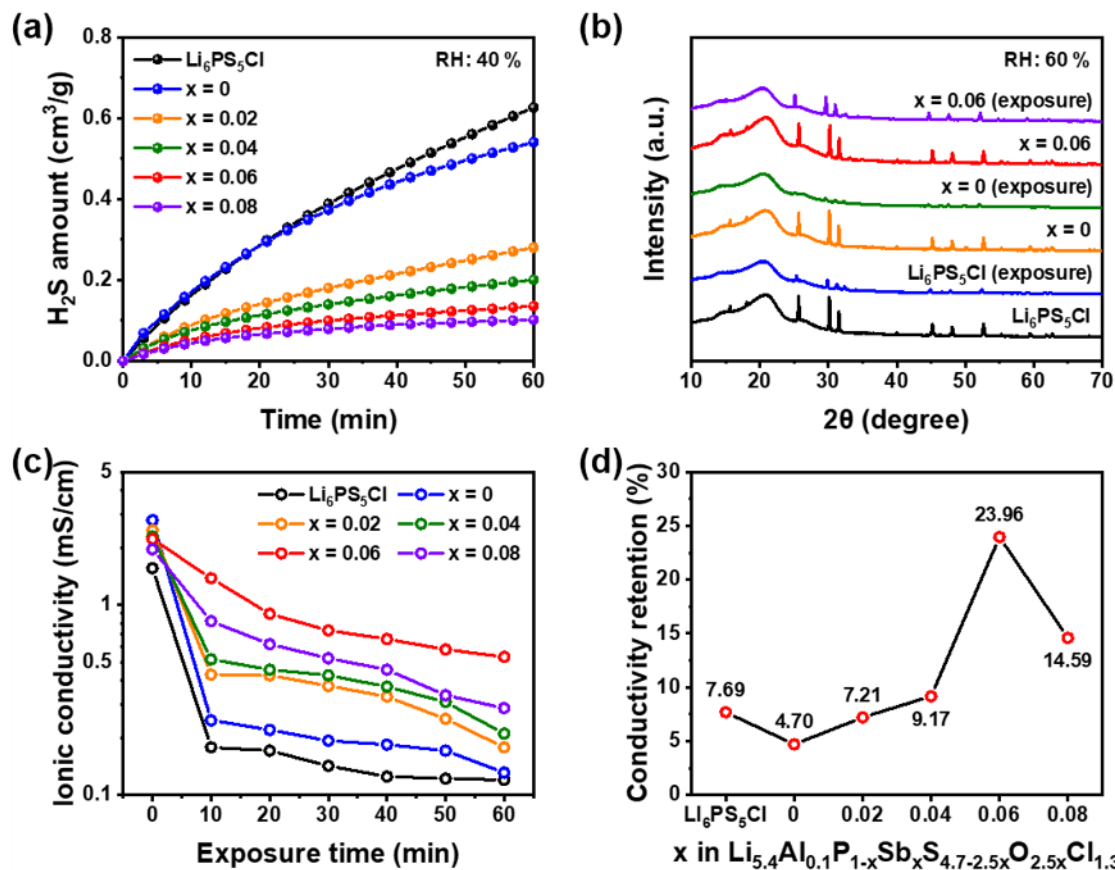
Electrochemical Properties of Sb-O Doped Solid Electrolytes



- ✓ Elevated levels of Sb_2O_5 doping may impede the ionic conduction pathway in the electrolyte, as the introduction of oxygen into the structure tends to decrease conductivity.

II. Theme I_Al-Sb-O-Cl Doping

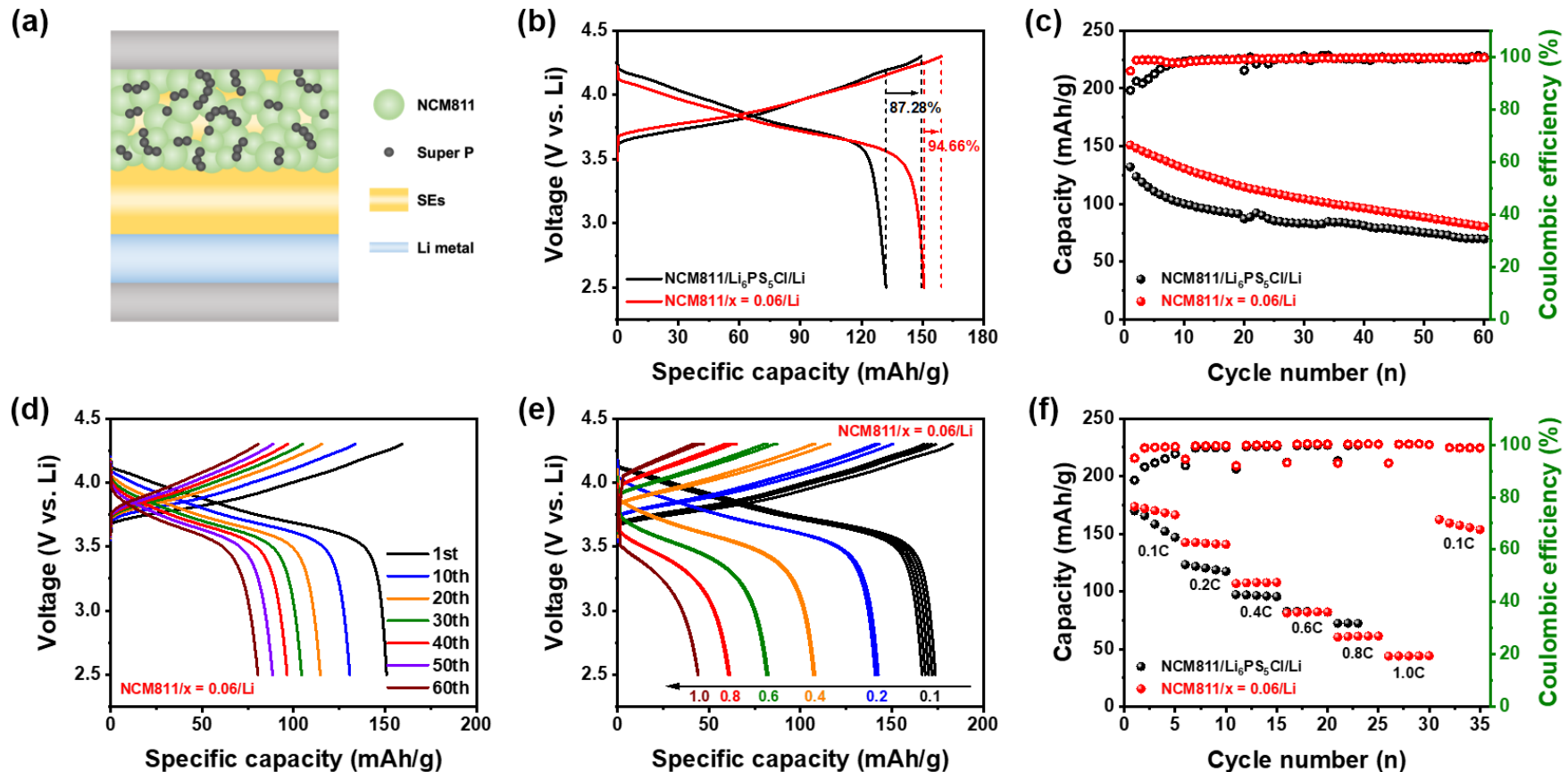
Air-Stability of Sb-O Doped Solid Electrolytes



- ✓ This suggests that the release of H₂S experiences rapid suppression with increasing Sb₂O₅ content.
- ✓ Air-stability is significantly improved when Sb and O are introduced into the structure.

II. Theme I_Al-Sb-O-Cl Doping

Full Battery Performance

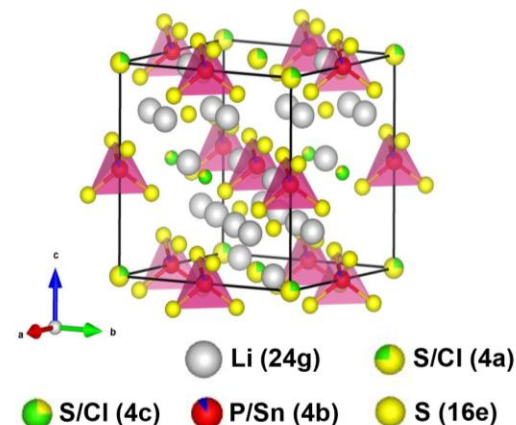
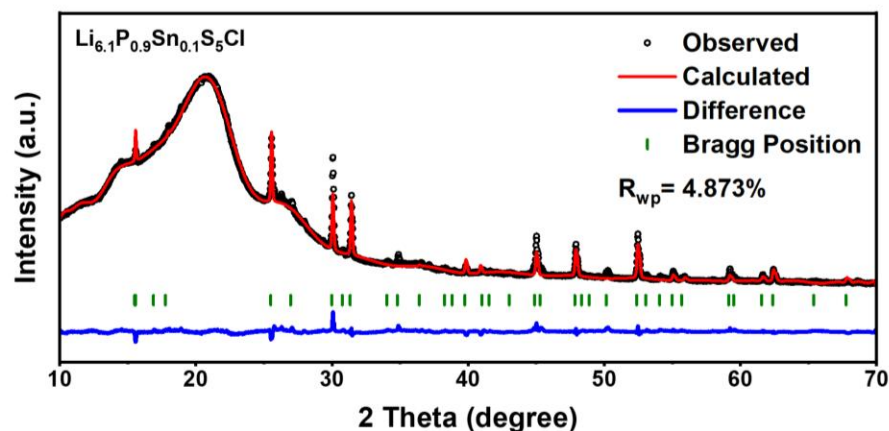
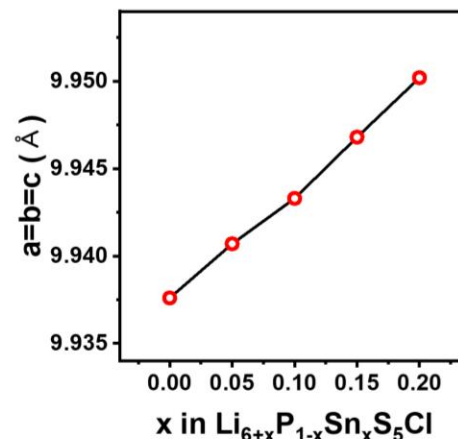
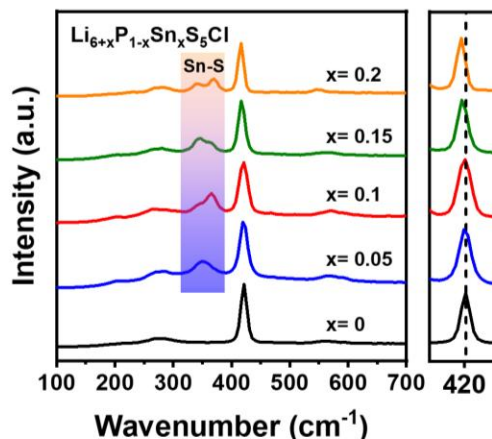
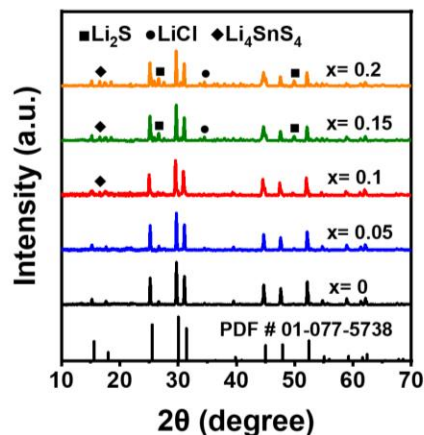


- ✓ The superior battery performance is attributed to the better bare Li metal compatibility with $x = 0.06$.
- ✓ The existence of Sb can mitigate the growth of Li dendrite and promote Li diffusion.

III. Theme II_Sn Doping

Structural Analysis of Sn Doped Solid Electrolytes

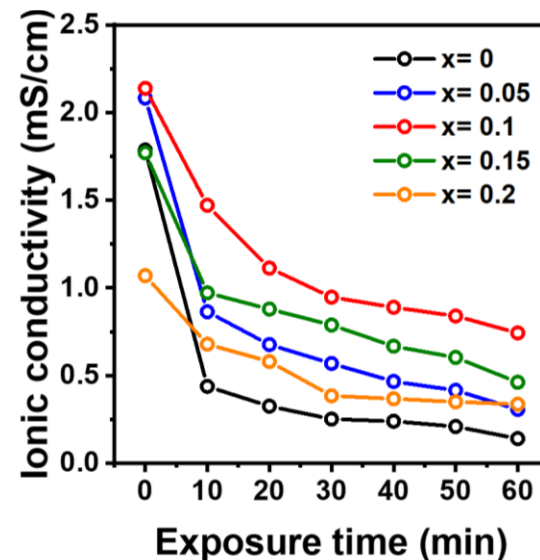
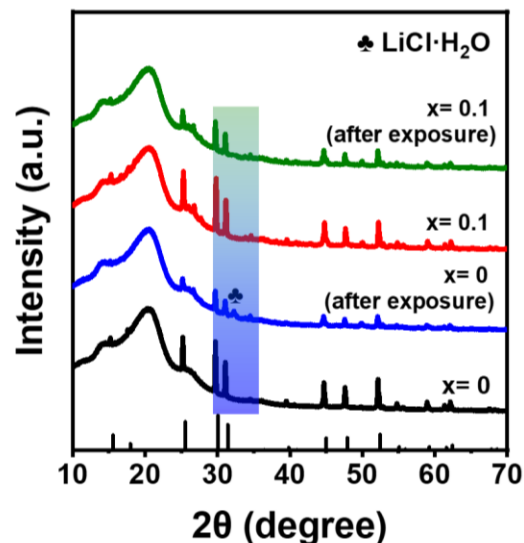
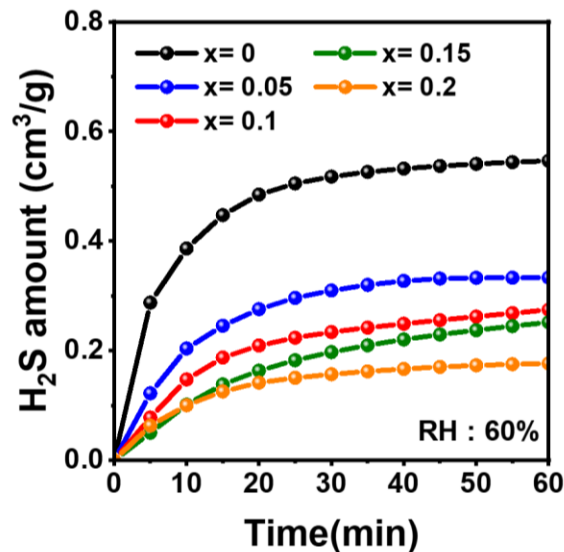
Kim et al, *ACS Applied Energy Materials*, 7 (2024), 9451-9459



- ✓ Structural analyses revealed successful Sn-doping in $\text{Li}_6\text{PS}_5\text{Cl}$ electrolyte.
- ✓ Sn replaced P without damaging the argyrodite structure.

III. Theme II_Sn Doping

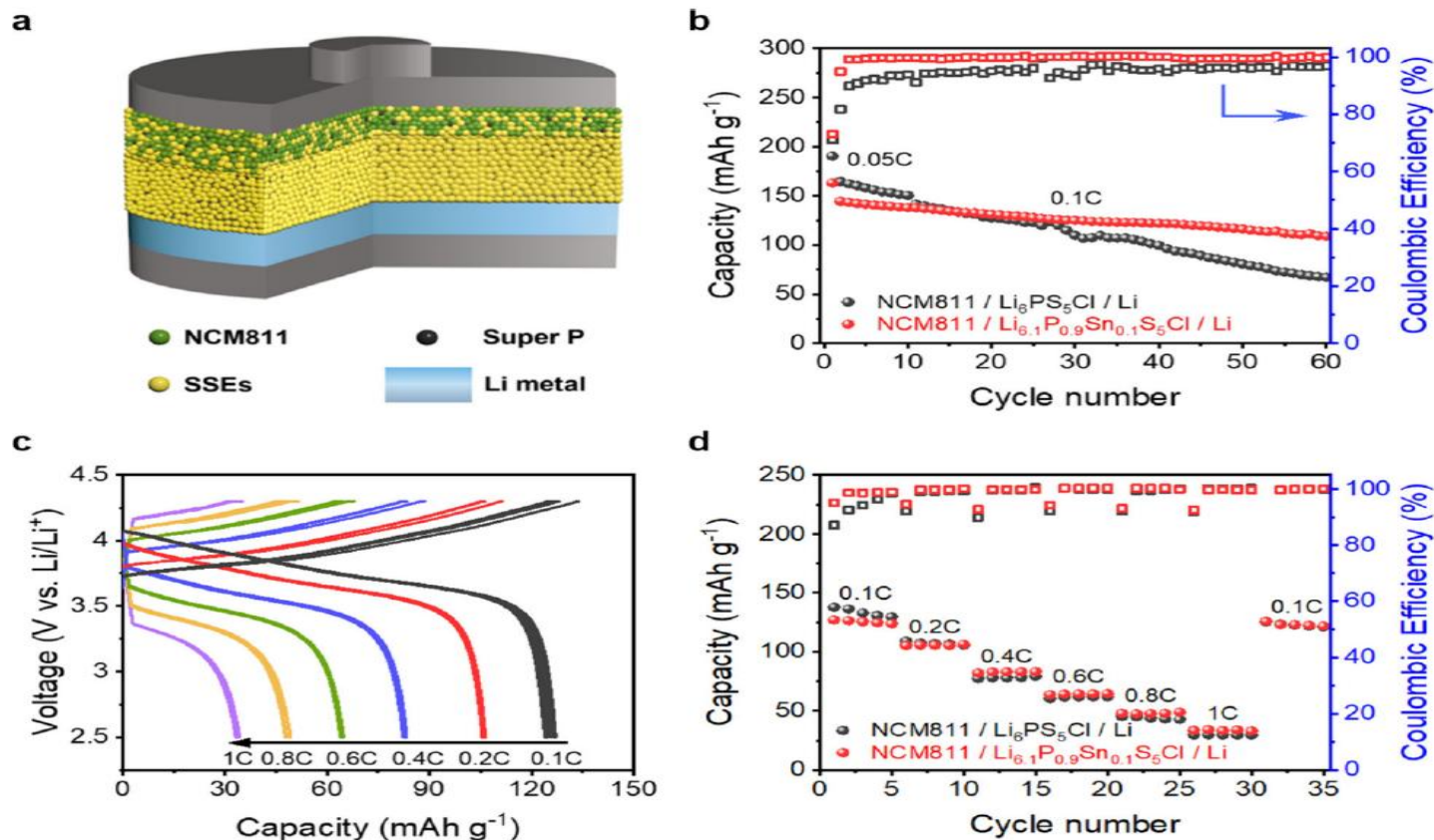
Air-Stability of Sn Doped Solid Electrolytes



- ✓ This suggests that the release of H₂S experiences rapid suppression with increasing SnS₄ content.
- ✓ Air-stability is significantly improved when Sn is introduced into the structure.

III. Theme II_Sn Doping

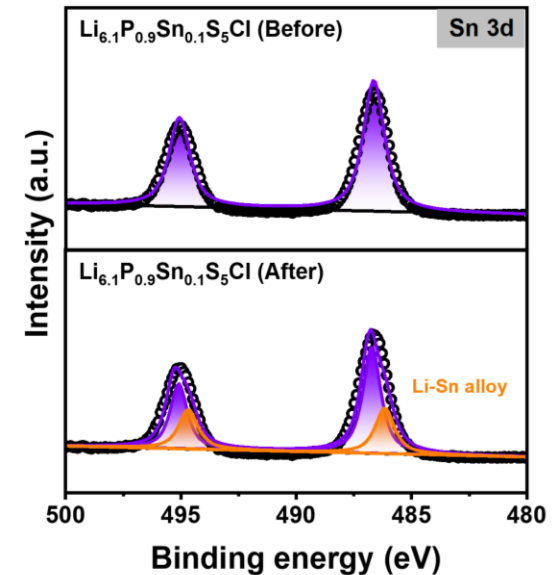
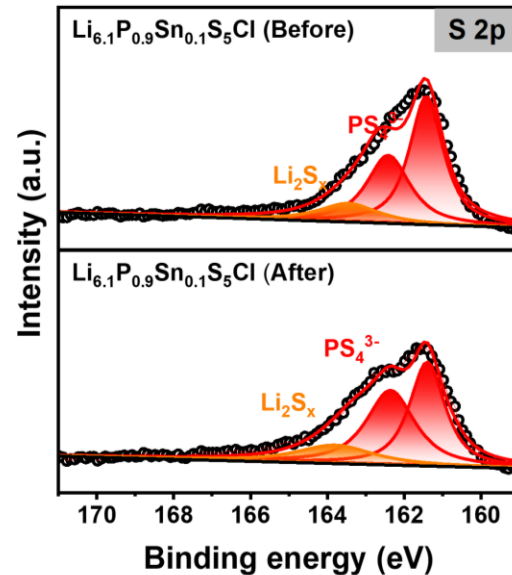
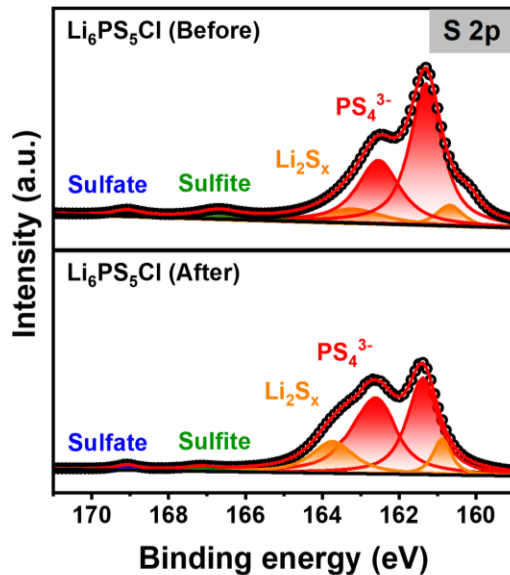
Full Battery Performance



- ✓ The superior battery performance is attributed to the better bare Li metal compatibility with Li_{6.1}P_{0.9}Sn_{0.1}S₅Cl.
- ✓ The existence of Sn can mitigate the growth of Li dendrite and promote Li diffusion.

III. Theme II_Sn Doping

XPS spectra of Sn Doped Solid Electrolytes

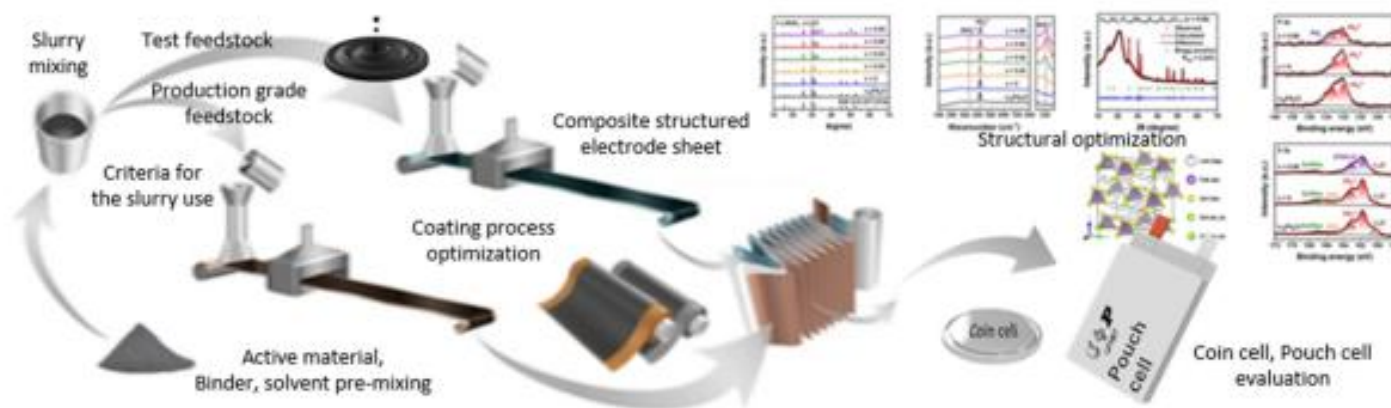


✓ Li-Sn alloy can promote the even deposition of Li and ensure a consistent Li^+ flow at the interface.

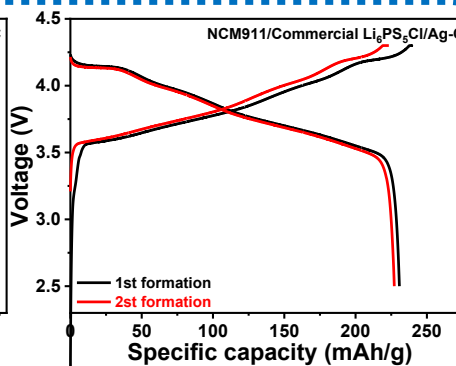
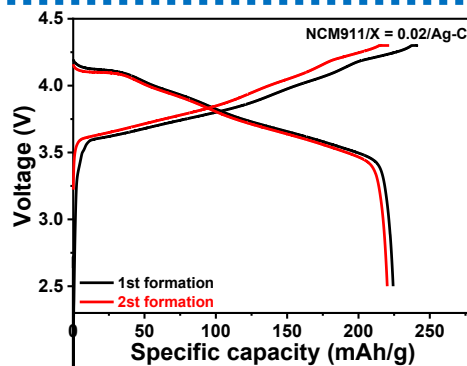
IV. Research Plan

Automation ASSB electrode production

“Development of a hyper-connected Ai-driven autonomous manufacturing system with integrated operation for next-generation (Thio-) ASSB electrode production.”



Pouch Cell test

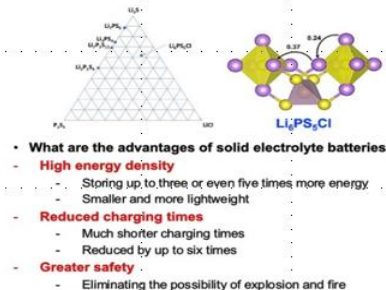
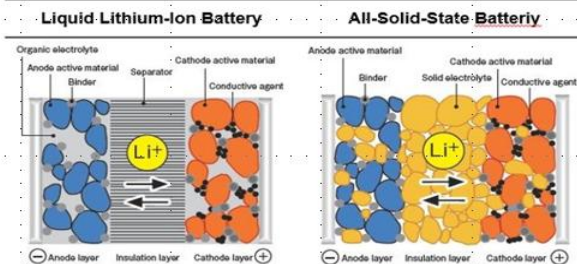


Secondary lithium ion battery

기술내용

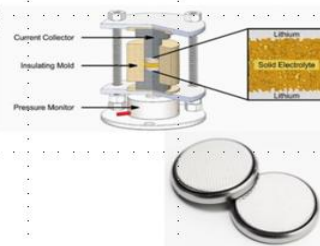
All-Solid-State Battery

<Comparison between lithium-ion battery and all-solid-state battery>

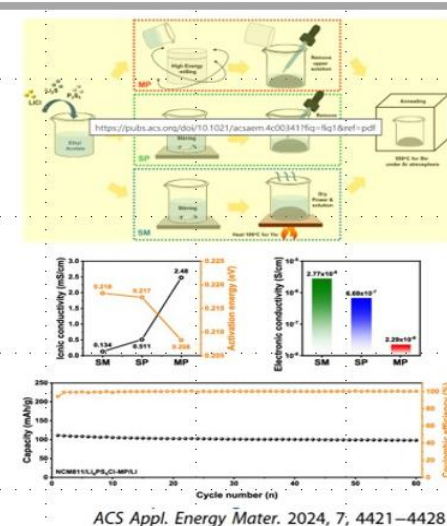


수행 연구내용

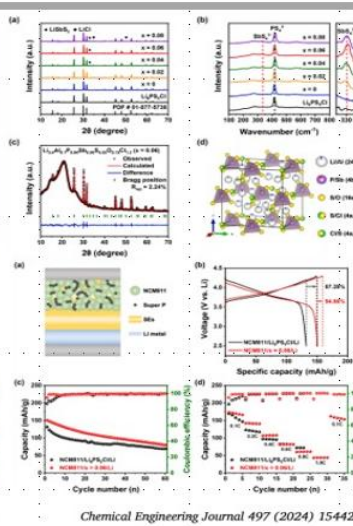
- 전고체전지용 전해질 설계 및 합성
- 황화물계 고체전해질 설계 및 합성
- 대기안정형 고체전해질 합성
- 고안정성 고체전해질 제조
- 전고체전지 셀 제작
- 리튬이차전지 평가
- 고체전해질 이온전도도 측정
- 전고체전지 셀 특성 분석



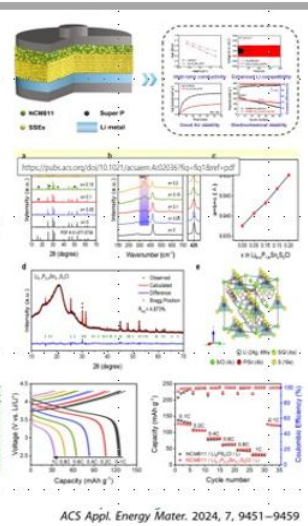
연구성과 대기 안정형 황화물계 전해질 제조 기술 개발



ACS Appl. Energy Mater. 2024, 7, 4421–4428



Chemical Engineering Journal 497 (2024) 154426



ACS Appl. Energy Mater. 2024, 7, 9451–9459

연구실적

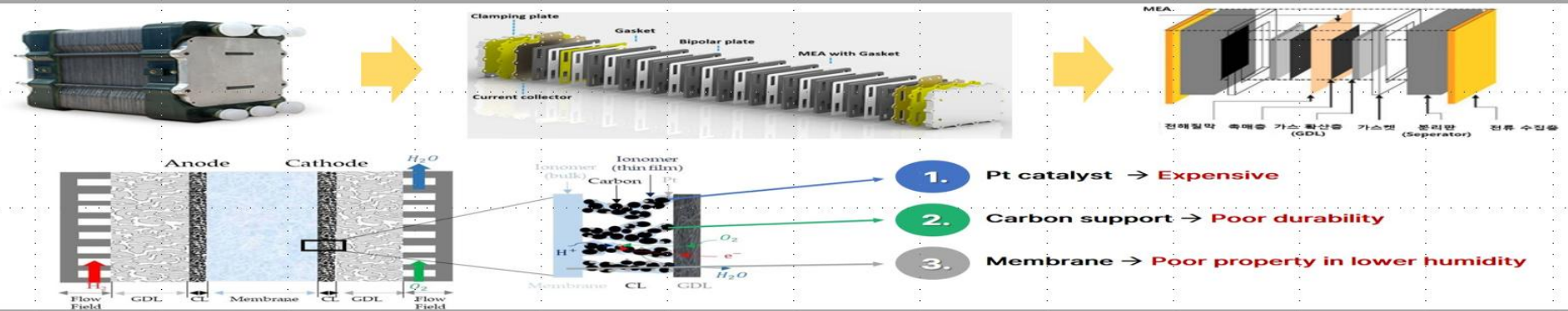
주요수행과제

- 50G 이상 고에너지를 적용한 저가 고이온 전도성 다성분계 설파이드 공정기술개발
- 친환경 합성법을 통한 전기차용 고체 이온 전도 물질 제조 및 공정기술 개발

연구실적 (SCI(우수)논문 3편, 특허 등록&출원 6건)

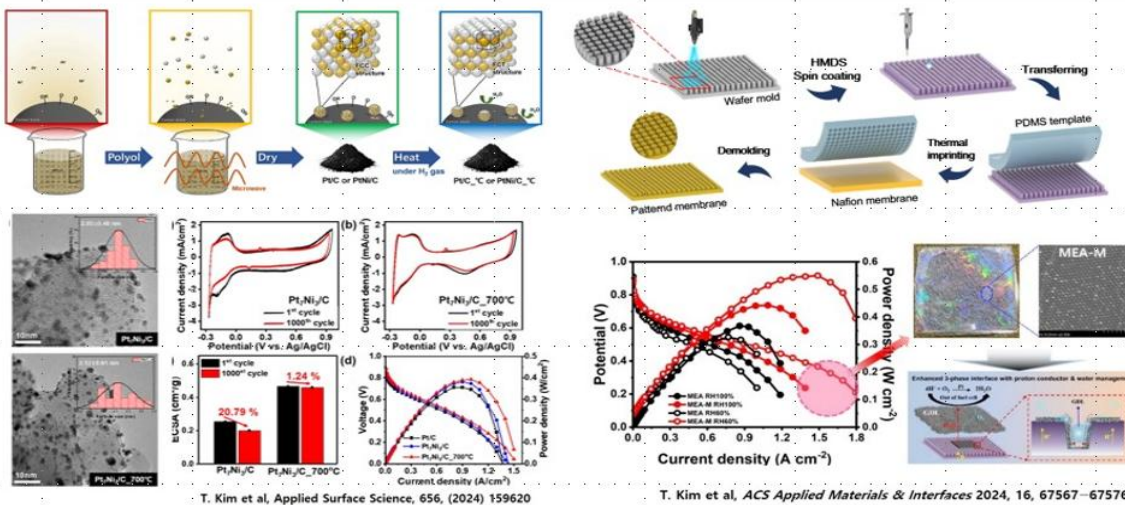


기술내용



연구성과

고출력 고내구성 MEA 핵심 소재 기술 개발



연구실적

주요수행과제

- 수소 생태계 확립을 위한 원스탑 플랫폼 사업
- 수소 산업 기업지원 혁신기반구축 조성사업
- 고분자 전해질 연료전지용 고내구성 복합 소재 분리판 제조 기술 개발

연구실적 (SCI(우수)논문 3편, 특허 등록&출원 3건)



Facilities

■ ASSBs



Milling machine



Tube furnace



Glove box for electrolyte synthesis



Glove box for cell fabrication



Cell test systems

■ PEMFCs



Clean room



Ultrasonic spray coater



Hot Press for MEA



Hot Press for bipolar plate



PEMFCs single cell test systems

