PROGRAM AND ABSTRACTS

October 2-3, 2025

OMU International Symposium on All-Solid-State Battery

Osaka Metropolitan University

Thursday, October 2, 2025

Venue : Osaka Metropolitan University, I-site Namba ※ +ONLINE

9:00	Registration (Onsite)		
9:30	**Opening**	Masahiro Tatsumisago (Professor Emeritus,Osaka Metropolitan University)	
9:40	**“Foundation Potentials for Massive-Scale Design of Solid Electrolytes”**	Shyue Ping Ong (University of California San Diego)	P.1
10:20	**“Challenges toward reproducibility in solid-state batteries: from materials to cells”**	Saneyuki Ohno (Tohoku University)	P.2
Break (10:50-11:05)			
11:05	**“Toward Practical High-Energy All-Solid-State Batteries: From Solid Electrolyte Materials to Sheet-Type Electrodes and Pouch Cells”**	Yoon Seok Jung (Yonsei University)	P.3
11:45	**“Electronic Functions and Applications of Solid State Ionic Devices”**	Takashi Tsuchiya (National Institute for Materials Science)	P.4
Lunch (12:15-13:15 / Luncheon Seminar 30minutes “Analysis and Evaluation Techniques for Lithium-Ion Batteries - From Raw Materials to Cell Evaluation -” — Takashi Sumoto (Shimadzu Corporation)			
13:15	**“High-Capacity Intercalation-type Cathodes using Anionic Redox for All-solid-state Fluoride-Ion Batteries”**	Kentaro Yamamoto (Nara Women’s University)	P.5
13:45	**“Interface issues of all-solid-state batteries”**	Yasutoshi Iriyama (Nagoya University)	P.6
14:15	**“Synthesis and Structural Analysis of Sulfide-Type Solid Electrolytes Synthesized by Liquid Phase Methods for All-Solid-State Batteries”**	Kazuhiro Hikima (Toyohashi University of Technology)	P.7
Break (14:45-15:05)			
15:05	**“All solid battery for sustainable mobilities”**	Hideki Iba (Toyota Motor Corporation)	P.8
15:45	**“Development of a dense all-solid-state chloride ion battery using solid electrolytes with low Young’s modulus”**	Atsushi Inoishi (Kyushu University)	P.9
16:15	**“Dry mixing and film formation for sulfide solid state battery electrodes”**	Arno Kwade (Technische Universität Braunschweig)	P.10
16:55	**Closing**	Fuminori Tokunaga (Vice President in charge of Research and Global Strategy ,Osaka Metropolitan University)	
Move venue			
18:00	**Networking event** Venue: Tennoden		

Friday, October 3, 2025

Venue : Osaka Metropolitan University, Nakamozu Campus, Conference Hall / Bldg. ia Smart Energy

13:30	Registration (Onsite)		
14:00	**“Development of solid electrolytes with reduction stability for all-solid-state sodium batteries”**	Akira Nasu (Hokkaido University)	P.11
14:20	**“Investigation of Li metal-sulfide solid electrolyte composite electrode for all-solid-state Li metal batteries”**	Hirokazu Kitaura (National Institute of Advanced Industrial Science and Technology)	P.12
14:40	**“Strain-free MXene electrodes for all-solid-state batteries”**	Kosuke Kawai (Tohoku University)	P.13
	Break (15:00-15:05)		
15:05	**Shortpresentation**		
15:45	**Posterpresentation**		
16:45	**Guided Tour of Bldg.ia Smart Energy**		
17:15	**Networking event** Venue: Nakamozu Campus,Conference Hall (Standing buffet)		

Foundation Potentials for Massive-Scale Design of Solid Electrolytes

Shyue Ping Ong[1]

[1] Aiiso Yufeng Li Family Department of Chemical and Nano Engineering, University of California, San Diego, 9500 Gilman Dr. Mail Code #0448, La Jolla, CA 92093-0448, USA

Foundation potentials (FPs)[1], i.e., machine learning interatomic potentials with near universal coverage of the entire periodic table, have emerged as a promising new tool in the study of materials. In this talk, I will provide a general overview of the capabilities of FPs, especially in the context of design of solid electrolytes for all-solid-state batteries. I will demonstrate how modern FPs trained on robust, diverse datasets such as MatPES,[2] are a highly effective surrogate for expensive density functional theory (DFT) calculations to access a wide range of PES-derived properties, such as the ionic conductivity, phase stability, electrochemical stability, phonon dispersion, elastic moduli, etc. I will also discuss approaches to fine-tune such FPs with a minimal amount of data,[2] resulting in significantly improved accuracies and efficiency for simulations at scale. I will share new insights obtained from FP-accelerated simulations with regards the differences in ionic conductivities between different structures and chemistries.

[1] Chen, C.; Ong, S. P. A Universal Graph Deep Learning Interatomic Potential for the Periodic Table. Nat Comput Sci 2022, 2 (11), 718–728. https://doi.org/10.1038/s43588-022-00349-3.
[2] Kaplan, A. D.; Liu, R.; Qi, J.; Ko, T. W.; Deng, B.; Riebesell, J.; Ceder, G.; Persson, K. A.; Ong, S. P. A Foundational Potential Energy Surface Dataset for Materials. arXiv 2025. https://doi.org/10.48550/arXiv.2503.04070.
[3] Qi, J.; Ko, T. W.; Wood, B. C.; Pham, T. A.; Ong, S. P. Robust Training of Machine Learning Interatomic Potentials with Dimensionality Reduction and Stratified Sampling. npj Computational Materials 2024, 10 (43), 1–11. https://doi.org/10.1038/s41524-024-01227-4.

Challenges toward reproducibility in solid-state batteries: from materials to cells

Saneyuki Ohno

Institute of Interdisciplinary Materials Research for Advanced Materials, Tohoku University, Japan

Solids are typically rigid, and the elements constituting matter generally remain immobile at room temperature. However, a unique class of materials exists that can conduct constituent ions like liquids, despite their solid nature; these are termed ionic conductors. Among inorganic solids exhibiting ionic conductivity, those that specifically transport alkali metal ions such as lithium or sodium around room temperature are of particular interest. These materials show promise as solid electrolytes, forming the cornerstone of all-solid-state batteries, which are safe and high-power candidates for next-generation energy storage devices. Our research team has been dedicated to elucidating the ionic transport mechanisms within solid electrolytes, thereby utilizing our understanding of material design principles.

Notably, even subtle changes in composition or crystal structure within a given material family can result in variations in ionic conductivity spanning several orders of magnitude [1]. Understanding the mechanisms underlying such drastic changes and identifying the governing factors that modulate ion transport in solids is essential for material design, potentially enabling enhanced performance across a wide range of materials. We classify these influences into three main categories: *static lattice effects*, which depend on static structural information; *dynamic lattice effects*, which are contingent upon dynamic structural information; and *synthetic effects*, which rely on the method of material synthesis, among others. Some recent achievements in materials design will be showcased to highlight the importance of a deeper understanding of ion transport mechanisms [2-3].

A key focus of the presentation is the challenge of reproducibility in evaluating the performance of solid electrolytes and solid-state batteries. Considerable variation in reported ionic conductivities has been observed, even for nominally identical compositions [4]. Such variation remains even in the samples prepared via an identical synthetic route. Inspired by the high yet largely varied ionic conductivity of Na_3SbS_4, we systematically examined sample-to-sample variability and its thermodynamic origins by modulating the concentration of native Na vacancies [5]. The presentation will also explore hidden variables associated with synthesis that critically impact transport properties [6].

[1] S. Ohno et al., *Prog. Energy*, 2, 022001 (2020)
[2] Z. Huang, S. Ohno et al., *ACS Mater. Lett.*, 6, 1732-1738 (2024)
[3] CW Lee, S. Ohno et al., *ACS Mater. Lett.*, 7, 620-626 (2025)
[3] S. Ohno et al., *ACS Energy Lett.*, 5, 910-915 (2020)
[4] M. Shimoda, S. Ohno et al., *Chem. Mater.*, 34, 5634-5643 (2022)
[5] A. Kizilaslan, S. Ohno, A. Miura et al., *ACS Energy Lett.*, 10, 156-160 (2025)

Toward Practical High-Energy All-Solid-State Batteries: From Solid Electrolyte Materials to Sheet-Type Electrodes and Pouch Cells

Yoon Seok Jung[1]

[1]Department of Chemical and Biomolecular Engineering, Yonsei University, Seoul 03722, South Korea

Solidifying electrolytes in rechargeable batteries using nonflammable inorganic materials offers a promising route toward achieving high energy density. This approach enables the use of high-capacity electrode materials such as Li metal and Si anodes, while significantly improving safety. Among various inorganic solid electrolyte (SE) candidates, sulfide and halide materials stand out due to their favorable mechanical deformability for manufacturing and sufficient ionic conductivity. For practical implementation, however, further advancements are required in key functional areas, including chemical and electrochemical stability. On the anode side, Li metal is prone to dendritic growth, which can lead to SE penetration and eventual cell failure—prompting intensive research on interfacial engineering. For the cathode side, while high-capacity materials such as sulfur or Li_2S are attractive, their significant volume change during charge-discharge cycling and insulating nature present substantial challenges. As an alternative, increasing operating voltages beyond 5 V represents a promising direction, though relevant studies remain limited.

In this presentation, I will share our recent efforts to develop advanced sulfide and halide SE materials, with a particular focus on chemical stability relevant to dry-room processing and strategies to extend electrochemical stability beyond 5 V. I will also present our approaches to enable Li metal and Si anodes. Finally, I will introduce our integration of these technologies into pouch-type all-solid-state batteries, especially those operating at 5 V.

[1] H. Kwak *et al.,* ACS Energy Lett. 2, 1776 (2022).
[2] Y. B. Song *et al.,* Adv. Energy Mater. 13, 2301142 (2023).

Electronic Functions and Applications of Solid State Ionic Devices

Takashi Tsuchiya[1-3], Makoto Takayanagi[1,3], Daiki Nishioka[4], Wataru Namiki[1],
Tohru Higuchi[3], and Kazuya Terabe[1]

[1]Research Center for Materials Nanoarchitectonics (MANA), National Institute for Materials Science (NIMS), Japan
[2]Department of Nanoscience & Nanoengineering, Waseda University, Japan
[3]Tokyo University of Science, Japan
[4]International Center for Young Scientists, NIMS, Japan

Solid state ionic devices have been attracting attention in a wide range of applications to energy devices (e.g., all-solid-state lithium ion batteries(ASS-LIBs), capacitors, sensors). While such energy devices are the primary and most promising applications of solid state ionics, high-density electronic carrier modulation, a feature of solid state ionics, makes it attractive beyond energy applications, further extending the application range of solid-state ionics into the electronics field. For example, we utilized solid state ionic devices in the form of field-effect transistors to tune the electronic carrier density of semiconducting materials to wide control of electronic properties with inorganic ion conductors, which is hardly obtained with conventional dielectrics [1,2]. The solid state ionic transistors have become a typical approach nowadays to explore novel electronic functions (e.g., optical bandgap, electron mobility, superconducting temperature, magnetization) of materials and their implementation as a solid device. The electronic carrier modulation of solid state ionic devices gives us, in turn, useful analytic capacity related to solid electrolyte interfaces. The space charge layer or electric double layer effect in solid electrochemical systems is a key topic in energy storage applications, and observing this effect based on conventional electrochemical measurements is not straightforward. We applied solid state ionic transistors to investigations on the electric double layer effect of solid electrolyte interfaces by utilizing a semiconducting diamond surface as a probe of electrical charges at the solid/solid interfaces [3]. Hall measurements with various solid electrolyte-based transistors evidenced that the electric double-layer effect strongly depends on the properties of the electrolyte and the very thin region from the interface. The unveiled features of the electric double layer at solid electrolyte interfaces were quantitatively discussed from the viewpoint of charge density and charging-discharging rate [4]. Electrical transport characteristics of solid-state ionic devices accompanying transient and nonlinear responses have been applied to physically implement materials-based information processing, including ionic makers for solving multi-armed bandit problems by mimicking spatiotemporal dynamics of feeding amoeba (natural computing)[5] or ion-gating reservoirs performing physical reservoir computing for time-series processing [6]. Redox-based electronic carrier modulation has achieved notable performance improvements in magnonic neuromorphic computing through magnetic property tuning [7].

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[2] T. Tsuchiya *et al.,* Adv. Mater. 26, 1087 (2014).
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[4] M. Takayanagi *et al.,* Mater. Today Phys. 31, 101006 (2023).
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High-Capacity Intercalation-type Cathodes using Anionic Redox for All-solid-state Fluoride-Ion Batteries

Kentaro Yamamoto

Faculty of Engineering, Nara Women's University, Kitauoya-nishimachi, Nara 630-8263, Japan

All-solid-state fluoride-ion batteries, which use fluoride-ions as carriers, are attracting attention as the next-generation batteries due to their theoretical high energy density and safety. Although simple metal/metal fluorides such as Cu/CuF_2 have been developed as conventional cathodes[1], these cathodes have disadvantages of the power density and cyclability due to the large volume change during charging/discharging. To solve these problems, intercalation-type cathodes, similar to electrode materials applied in lithium-ion batteries, have been developed[2]. However, they have the disadvantage of relatively small capacity due to the heavy formula weight, compared to metal/metal fluorides. Recently, intercalation-type cathode $La_{1.2}Sr_{1.8}Mn_2O_{7-\delta}F_2$ has been developed to provide high capacity involving O–O bond formation (anionic redox)[3]. In this study, $SrFeO_2$ and Ca-doped $SrFeO_2$ cathodes with infinite layer structure are introduced as new high-capacity intercalation-type cathodes involving O–O bond formation[4].

$Ca_{1-x}Sr_xFeO_2$ ($x = 0, 0.2, 0.4, 0.6, 0.8, 1.0$) was synthesized by a low-temperature reduction method using CaH_2. $Ca_{1-x}Sr_xFeO_2/La_{0.9}Ba_{0.1}F_{2.9}$/vapor grown carbon fiber (VGCF) was used as the composite cathode, $La_{0.9}Ba_{0.1}F_{2.9}$ as the electrolyte and $Pb/PbF_2/La_{0.9}Ba_{0.1}F_{2.9}$/VGCF as the composite anode to construct the electrochemical cell. Charge-discharge measurements were carried out in the cut-off voltage range of –1.5 to 3.0 V at 140°C. X-ray absorption spectroscopy (XAS) measurements of Fe *K*-edge, O *K*-edge, and F *K*-edge and resonant inelastic X-ray scattering (RIXS) measurements of O K-edge were performed on the samples after charging and discharging.

$SrFeO_2$ and Ca-doped $SrFeO_2$ cathodes showed voltage plateaus around –0.5 V and 1.5 V during charging, and $Ca_{0.8}Sr_{0.2}FeO_2$ among the cathodes delivered the maximum discharge capacity (580 mAh g^{-1}). The XAS for Fe *K*-edge revealed that the Fe is responsible for the charge compensation in the voltage plateau around –0.5 V while is not responsible for the charge compensation in the voltage plateau around 1.5 V. The XAS and RIXS measurements for O *K*-edge revealed that the charge compensation was carried out by the formation of O–O bonding, as which is observed in lithium-rich cathodes[5], in the voltage plateau around 1.5 V. The developed intercalation-type cathode materials using anionic redox significantly exceeds the capacity of conventional lithium-ion battery cathodes.

References

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Acknowledgement

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Interface issues of all-solid-state batteries

Yasutoshi Iriyama

Department of Materials Design Innovation Engineering, Graduate School of Engineering,
Nagoya University, Furo-cho, Chikusa-ku, Nagoya, Aichi, 464-8603

Sulfide-based all-solid-state batteries (SBs) are expected as an advanced power source for electric vehicles (EVs) because sulfide-based solid electrolytes (SEs) are high Li^+ conductive materials and are ductile. However, SEs have a narrow potential window[1] and are highly reactive with cathode materials, leading to a resistive interface. To overcome these problems, a coating layer, commonly amorphous Li–Nb–O (a-LNbO, ~10 nm in thickness), is formed on the cathode surface.[2]

A practical method to improve the energy density of SBs is to increase their charging voltages. However, high-voltage charging degrades SBs and several degradations occur inside SBs, such as deterioration of the cathode structure, degradation of the coating layer, and oxidation of SEs around the coating layer. Here, we investigated the degradation mechanism of a-LNbO coating layer on electrode preserved at high-voltage (~4.55 V vs. Li/Li^+). In the case of the a-LNbO coating layer, the following decomposition schemes have been proposed indirectly based on X-ray absorption fine structure (XAFS) analysis of Nb L_3-edge local structure variations at high voltages:[3]

$$LiNbO_3 \rightarrow Li_{1-2x}NbO_{3-x} + x\ Li_2O \qquad (1)$$
$$Li_2O \rightarrow 0.5O_2 + 2Li^+ + 2e^- \qquad (2)$$

In this study, we investigated gas generation from SBs with and without coating layers on the NCM. We will show that a larger amount of O_2 is generated from the a-LNbO coating layer at high-voltage charging. Additionally, Li extraction from the a-LNbO coating layer is confirmed using electrochemical X-ray photoelectron spectroscopy (XPS) analysis [4]. These results directly support the above degradation schemes. Based on these results, we propose that the design of coating layers that suppress O_2 generation is a key factor in stabilizing high-voltage charging of the SBs. In fact, we will show that an amorphous Li–Nb–P–O coating layer, known to improve the high-voltage stability of SBs,[4, 5] generates less O_2.

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[4] K. Yoshikawa, Y. Iriyama et al., *Adv. Sci.* **2024**, *11*, 2402528.
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This work was supported by NEDO SOLiD-Next (JPNP23005) and JSPS KAKENHI “Interface IONICS” JP19H05813&05814.

Synthesis and Structural Analysis of Sulfide-Type Solid Electrolytes Synthesized by Liquid Phase Methods for All-Solid-State Batteries

Kazuhiro Hikima[1], Atsunori Matsuda[1]

[1] Department of Electrical and Electronic Information Engineering, Toyohashi University of Technology, Japan

Sulfide-type solid electrolytes for all-solid-state lithium batteries are usually synthesized via mechanical milling or melt-quenching techniques. In recent years, liquid-phase synthesis has been introduced as a new route for synthesizing sulfide–type solid electrolytes, offering advantages such as scalability and the ability to fabricate homogeneous electrode composites. Liquid-phase synthesis can be broadly categorized into suspension and solution synthesis methods based on the state of the solution during the reaction process. In suspension synthesis, we previously reported the liquid-phase shaking method. A variety of sulfide-type solid electrolytes have been synthesized using the liquid-phase shaking method with organic solvents as a reaction medium, such as $Li_7P_2S_8I$ [1] and $Li_{10}GeP_2S_{12}$ [2]. For instance, in the case of $Li_7P_2S_8I$, surface analysis using X-ray photoelectron spectroscopy (XPS), high-energy X-ray scattering, and neutron total scattering revealed the formation of a stable surface layer derived from the organic solvent. This layer contributes significantly to the enhanced stability against lithium metal anodes compared to the mechanically milled sample.

On the other hand, the suspension synthesis is a time-consuming method, for example, $Li_{10}GeP_2S_{12}$, which takes more than 24 h. Therefore, we developed a solution synthesis method using excess sulfur with acetonitrile, tetrahydrofuran, and trace amounts of ethanol as solvents, which enables the rapid and scalable manufacturing of sulfide solid electrolytes, such as $Li_7P_3S_{11}$ [3], Li_6PS_5Cl [4], and $Li_{10}GeP_2S_{12}$-type solid electrolytes [2, 5].

Additionally, we proposed a liquid-phase method for fabricating electrode composites of active materials and solid electrolytes, utilizing a liquid-phase process [6]. Therefore, the liquid phase process is useful not only for the synthesis of sulfide-based solid electrolytes but also for the preparation of electrode composites for all-solid-state batteries.

Acknowledgement

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References

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All solid battery for sustainable mobilities

Hideki IBA
Toyota Motor Corporation

Sustainable Mobility

In order to realize a sustainable society, it is necessary to have vehicles with low fossil-fuel consumption and low carbon dioxide (CO2) emission. For that purpose, various efforts have been made such as making the vehicles smaller/lighter or lowering the fuel consumption of engines.

Since hybrid vehicles achieve a balance between fuel efficiency and driving performance, their types and sales have been increasing after Prius was released in 1997. Today's hybrid vehicles achieve fuel efficiency by getting a supply of gasoline and having an efficient exchange of energy with batteries.

Compared with hybrid vehicles that run only by getting a supply of gasoline, plug-in hybrid vehicles can deal with the diversification of primary energy by charging the storage batteries installed in the vehicles from the power supply of houses, etc., and can be expected to lower CO2 emission or energy cost. For plug-in hybrid vehicles, the larger the capacity of storage batteries are, the longer the distance they can run by batteries get and CO2 emission or energy cost can be reduced.

Although electric vehicles are being put into practical use in many vehicle models, further improvements in the performance of storage batteries are expected in order for them to be widely adopted in general vehicle use.

As mentioned above, storage batteries are a core technology for hybrid and electric vehicles, and it is no exaggeration to say that innovation in batteries is directly linked to innovation in vehicles.

Innovative battery

As the history of storage battery development shows, new principles and materials are critical to develop batteries with a revolutionary large amount of energy. The basic configuration of the battery is the electrode and the electrolyte. These days, there are candidates of new principles such as the all-solid battery which uses a solid electrolyte rather than the conventional liquid type, or the metal-air battery which uses metal for the electrode as it is, but there are many challenges to achieve practical use and a breakthrough by a long-term re-search is necessary.

Toward the realization of our dreams

I believe that creative/pioneering research based on science is indispensable for such breakthrough. This kind of research is difficult to conduct single-handedly by a private company since it requires not just a high academic level but also requires a long research period and the probability of success is low, so there are high expectations toward research at universities or national laboratories and handing over of the achievements by university-industry collaboration. Recent researches related to innovative battery have yielded numerous achievements by national projects, and some of them are directly related to research of batteries such as analysis on the crystal structure of solid electrolyte or the interface layer of all solid batteries.

Development of a dense all-solid-state chloride ion battery using solid electrolytes with low Young's modulus

Atsushi Inoishi[1], Liwei Zhao[1], Hidenori Miki[2], Hikari Sakaebe[1]

[1]Institute for Materials Chemistry and Engineering, Kyushu University, Japan
[2]Toyota Motor Corporation, Advanced Material Engineering Division, Japan

Chloride ion batteries that utilize conversion reactions at both electrodes possess a significantly higher theoretical energy density compared to lithium-ion batteries. Although chloride batteries have a lower energy density than fluoride batteries, they exhibit higher plasticity, making them well-suited for battery design. In chloride batteries, all components—cathode, electrolyte, and anode—are made of soft chloride materials, which is expected to allow better accommodation of volume changes. However, solid electrolytes with high chloride ion conductivity had not been developed. We have discovered that the perovskite-type $CsSn_{0.95}Y_{0.05}Cl_{3.05}$ (CSYC) exhibits a conductivity of 4.9 mS cm^{-1} at room temperature.[1] This material is a perovskite-type compound in the $CsSnCl_3$ system, which is monoclinic in its undoped state. However, when Y is doped into the B-site, the cubic structure becomes stabilized at room temperature. In this report, we present the development status of this material.

CSYC was synthesized by ball-milling stoichiometric amounts of CsCl, $SnCl_2$, and YCl_3.A 1.0 g precursor mixed with 60 g of 3 mm diameter zirconia balls was planetary ball-milled at 600 rpm for 12 h. The elastic modulus of the cold-pressed pellets was evaluated using an ultrasonic pulse-echo technique with 5 MHz frequency transducers. Cold-pressed pellets were prepared by pelletizing the powders in an Ar-filled glovebox under a pressure of 510 MPa at room temperature. An all-solid-state cell was fabricated using a $BiCl_3$ composite as the cathode ($BiCl_3$:CSYC:AB=4:5:1 wt%), a Sn composite as the anode (Sn:CSYC:AB=4:5:1 wt%), and CSYC as the solid electrolyte. Galvanostatic charge-discharge tests of the cell were performed at 25 °C at a current density of 20 µA cm^{-2} in an Ar-filled glovebox.

Various dopants were introduced at the Sn site in $CsSnCl_3$, and it was found that Y doping resulted in the highest ionic conductivity at room temperature (4.9 mS cm^{-1}). High ionic conductivity arises from the stabilization of the cubic perovskite structure, and even after one year of storage in a glove box, CSYC retained its structure. CSYC exhibited 95% relative density through cold pressing and demonstrated an extremely low Young's modulus of 8.2 GPa, indicating high plasticity. After charge-discharge cycling, the battery cross-section was observed using SEM. Figure 1 presents an SEM image of the interface of the battery after discharge-charge cycling. The cathode, electrolyte, and anode layers all appeared highly dense, with minimal cracks or voids observed.

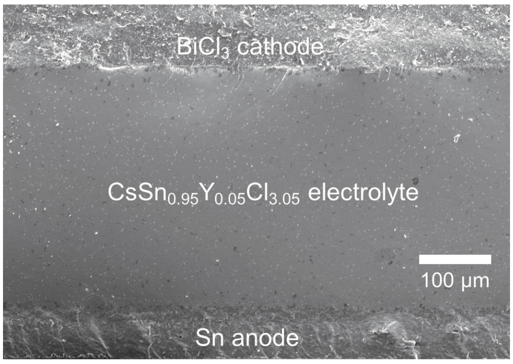

Fig.1 SEM image of the all-solid state chloride cell after cycling.

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Dry mixing and film formation for sulfide solid state battery electrodes

Arno Kwade[1], Finn Frankenberg[1], Carina Heck[1], Peter Michalowski[1]

[1]Technische Universität Braunschweig, Institute for Particle Technology, Volkmaroder Straße 5,
Braunschweig, 38104, Germany

While the effects of new solid electrolytes and active materials in cathode composites for solid-state batteries (SSBs) are widely studied, the influence of mechanical processing on their properties remains largely unexplored. This talk systematically investigates how dry mixing parameters, using planetary ball milling and high-intensive mixers, affect the production of composite cathodes composed of Li_6PS_5Cl (LPSCl), $LiNi_{0.83}Co_{0.11}Mn_{0.06}O_2$ (NCM) and carbon additive.

Overall, this talk focuses on the critical importance of precisely controlling mechanical processing parameters to optimize the structure and performance of composite cathodes in SSBs. Our results demonstrate that mechanical processing has a significant impact on the composite microstructure and subsequent cell performance by affecting the particle size of LPSCl, the distribution of the carbon additive and the formation of electrolyte – active material interphases and aggregates. Higher stress intensities and extended mixing times promote LPSCl comminution and the formation of composite aggregates, improving interfacial contact, evidenced by reduced overpotentials during cycling. Effective ionic conductivity also increases with mixing intensity and time, attributed to smaller LPSCl particles, improved aggregate formation, and reduced porosity. However, excessive mechanical stress, caused by high collision energies, results in undesirable effects such as increased strain in NCM and can strongly reduce LPSCl crystallite sizes, as revealed by XRD Rietveld refinement.

Combining experimental data with discrete element method (DEM) simulations shows that these microstructural changes can be estimated based on the stress intensity and specific energy input. Higher energy input increases heterocontact density, correlating with enhanced electrochemical performance. Subsequent dry film formation from premixed composites confirms that the premixing step has a significant impact on both final film quality and electrochemical performance. Optimized premixing leads to enhanced C-rate performance, with specific discharge capacities reaching ~180 mAh/g at 0.1 C, which are even higher than for the premixed composite. This improvement comes form the high shear forces applied in the calender gap, which enhance active material utilization, which was measured to increase from 88 % to 96 %.

Development of solid electrolytes with reduction stability for all-solid-state sodium batteries

Akira Nasu, Hiroaki Kobayashi, Matsui Masaki
Department of Chemistry, Faculty of Science, Hokkaido University, Japan

All-solid-state sodium batteries are one of the most promising battery systems combining high safety and high energy density. Development of appropriate solid electrolytes act as the bottleneck hindering the social implementation. The key functions are ionic conduction and the stability against active materials working as strong oxidizing or reducing agents. Notably, sulfides, such as Na_3PS_4 and Na_3SbS_4 systems, has gained attention due to its high ionic conductivity and high ductility, which facilitate cold sintering. Unfortunately, the sulfides lack electrochemical stability, and then electrochemical properties degraded owing to the interface reaction. In particular, the reduction by negative electrode materials is inevitable and the produced interphase often electronic conductivities [1]. Its electronic conductivities cause the supply of electron to the interface and reaching distribution equilibrium on inner potential. The process is tied to continuous reduction of the solid electrolytes.

The previous report [2] proposed that the solution preventing the degradation is a total substitution of centered elements from P to B. The Na_3BS_3 glass electrolytes formed thin and stable interphase against sodium metal. The interphase has Na_2S_x and borate without sodium compound Na-B. In a series of following studies, these electronic conductivities are derived from the sodium compounds. In other words, the degradation is inherently controllable by the selection of components elements without forming sodium compounds.

In this study, we proposed development of novel sulfides-based solid electrolytes via a synthesis technique with using Na_2S_x for compositional versatility. Alkaline poly sulfides work as the oxidant for the pure elements and homogeneous diffusion medium [3]. This process is reported to be applied for development of the sodium conductor by using Na_2S_x [4]. In addition, oxysulfides are homogeneously obtained by using the oxide starting material [5]. The strategic substitution in both cationic and anionic will be achieved in the process.

As the cationic substitution, the $Na_{11}Ge_2PS_{12}$-type ionic conductor is considered a candidate archetype owing to the high ionic conductivity and tolerance to cationic substitution. In the former part of the study, the $Na_{11}Ge_2PS_{12}$-type based on $Na_{12}Sn_2SiS_{12}$ has been developed and substituted from tin to group 4 and 5 elements. These heavy elements have similar ionic radii with tin, however there are limited reports about the sulfide's synthesis due to instability of the starting materials. By using the Na_2S_x and pure elements, we developed novel materials with the combination of the extended centered-cation groups.

As the anionic substitution, the Na_3PS_4-Na_3PO_4 pseudo binary system has been developed via Na_2S_x. The polyanionic units coordinated by a pair of anions often be regarded as enhancement factor for ionic conductivity and electrochemical stability [6]. According to homogeneous synthesis of oxysulfides via Na_2S_x, we evaluated the effect to the ionic conduction by the mixed anion units themselves.

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Investigation of Li metal-sulfide solid electrolyte composite electrode for all-solid-state Li metal batteries

Hirokazu Kitaura[1], Eiji Hosono[1], Misae Otoyama[2], Tomonari Takeuchi[2], Kentaro Kuratani[2]

[1]Research Institute for Energy Efficient Technologies,
National Institute of Advanced Industrial Science and Technology (AIST), Japan
[2]Research Institute of Electrochemical Energy,
National Institute of Advanced Industrial Science and Technology (AIST), Japan

Lithium metal is expected to be a promising negative electrode material because of its theoretical capacity of 3860 mAh g^{-1} and low potential of -3.04 V (vs. SHE). The use of lithium metal electrode has also been investigated for all-solid-state batteries with sulfide-based solid electrolytes. A major issue is short-circuits associated with stripping and deposition reactions of lithium metal under high current densities. Various efforts have been made to solve this issue, and Li metal sheets are used in many of them [1]. Therefore, the stripping and deposition reactions of Li metal occur in the two-dimensional plane where the Li metal sheet and solid electrolyte layer are in contact. This limited contact area may be one factor that facilitates short circuits. In this study, composite electrodes with three-dimensional contact were fabricated using Li metal powder and solid electrolyte powder, and the effects of increasing the contact area on short-circuit characteristics and Li stripping/deposition behaviors were investigated.

Li metal powder (p-Li) and $54Li_3PS_4 \cdot 46LiI$ (LPSI) solid electrolyte powder were used as electrode materials. The composite powder was made by mixing p-Li and LPSI in a weight ratio of LPSI/p-Li = 1.5 using a vortex mixer. The composite powder and stainless-steel current collector were stacked on both sides of the temporarily molded LPSI solid electrolyte layer, and the p-Li-LPSI | LPSI | p-Li-LPSI symmetric cell was fabricated by pressing at 360 MPa for 5 minutes. For comparison, the symmetric cell with Li-only electrodes of LPSI/p-Li = 0 was fabricated. Cross-sectional SEM observation of the composite electrode confirmed the three-dimensional contact between Li and LPSI within the electrode. Short-circuit current densities of symmetric cells at 25ºC were evaluated by a galvanostatic cycling test. As a result, the symmetric cell with the composite electrode showed a higher short-circuit current density than the cell with Li-only electrode. These results indicate that composite electrodes with three-dimensional contacts are effective in improving the short-circuit current density of all-solid-state Li metal batteries with sulfide solid electrolytes.

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Strain-free MXene electrodes for all-solid-state batteries

Kosuke Kawai[1]

[1]Institute of Multidisciplinary Research for Advanced Materials, Tohoku University, Japan

All-solid-state batteries with non-flammable inorganic solid electrolytes are a key technology to address the safety issues of lithium-ion batteries with flammable organic liquid electrolytes. However, conventional electrode materials suffer from substantial volume change during lithium-ion (de)intercalation, leading to the failure of the interface between the electrode materials and solid electrolytes and then severe performance degradation.[1] In this presentation, I introduce strain-free charge storage via an interface between a transition-metal carbide (MXene) and solid electrolytes.[2] MXene shows negligible structural change during lithium-ion (de)intercalation with sulfide and halide solid-state electrolytes. Scanning electron microscopy and X-ray diffraction confirm the ability of MXene to maintain a robust interface with the solid electrolyte. Furthermore, operando scanning transmission electron microscopy with electron energy loss spectroscopy visualizes Li^+ intercalation in MXene for the first time. A strain-free all-solid-state battery, which consists of MXene anode and disordered rocksalt $Li_{8/7}Ti_{2/7}V_{4/7}O_2$ cathode [3], demonstrates a long-term operation owing to the strain-free nature of both electrode materials (Fig. 1).

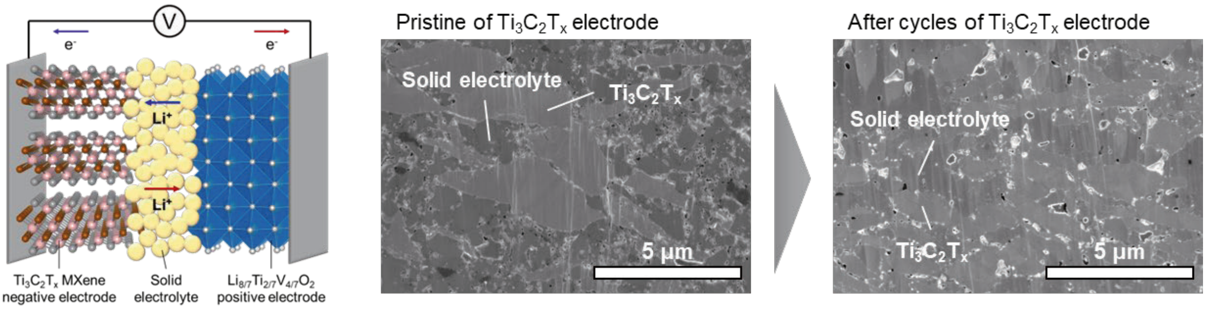

Fig. 1 Schematic illustration of strain-free all-solid-state battery and cross-sectional scanning electron spectroscopy images of MXene electrodes before and after charge/discharge cycling.

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