Electrochemistry
The Electrochemical Society of Japan
Received: March 31, 2025
Accepted: April 21, 2025
Published online: April 29, 2025
Issued: June 3, 2025
https://doi.org/10.5796/electrochemistry.25-71059
Review (Invited Paper) *Electrochemistry*, **93(6)**, 062003 (2025)

The 71st special feature "New Progress of Batteries and Fuel Cells"

Global Trends in Battery Research and Development: The Contribution of the Center for Advanced Battery Collaboration†

Kei NISHIKAWA,[a,*,§] Takuya MASUDA,[a,§] Yibin XU,[a] Yoshitaka TATEYAMA,[a,§] Shoichi MATSUDA,[a,§] Shuji NAKANISHI,[a,b,§] Shotaro HANADA,[b,§§] Yoshiharu MUKOUYAMA,[b,c,§] Toshihiko MANDAI,[a,§] Naoaki KUWATA,[a,§] Tsuyoshi OHNISHI,[a] and Kiyoshi KANAMURA[a,§§§]

[a] *Center for Advanced Battery Collaboration, Research Center for Energy and Environmental Materials, National Institute for Materials Science, 1-1 Namiki, Tsukuba, Ibaraki 305-0044, Japan*
[b] *Research Center for Solar Energy Chemistry, Graduate School of Engineering Science, The University of Osaka, 1-3 Machikaneyama, Osaka 560-8531, Japan*
[c] *Division of Science, College of Science and Engineering, Tokyo Denki University, Ishizaka, Hatoyama, Saitama 350-0394, Japan*

* *Corresponding author: NISHIKAWA.Kei@nims.go.jp, TEL: +81 29 860 4387*

ABSTRACT

The Center for Advanced Battery Collaboration (ABC) was established at the National Institute for Materials Science with the support of COI-NEXT, Japan Science and Technology Agency (JST). ABC aims to create a platform for collaboration between academia and industry for battery research and development with a special focus on developing battery simulation protocols through advanced characterization, data science and theoretical calculations. These protocols not only support the development of advanced Li batteries, Li-air batteries, sodium batteries, magnesium batteries, and all-solid-state batteries but also promote the innovation of new battery technologies in collaboration with industries. This article provides an overview of the projects in ABC and highlights the efforts and achievements of each research team.

Keywords : Rechargeable Batteries, Research Trend, Center for Advanced Battery Collaboration

1. Introduction

Rechargeable batteries are used in various devices, including cars, cell phones, and laptops, significantly shaping modern life, especially after the commercialization of lithium-ion batteries (LIBs). Recently, large-scale LIBs have been applied to electric vehicles and energy storage systems as part of efforts to reduce carbon dioxide emissions.[1] Another crucial role of rechargeable batteries is to power Internet of Things (IoT) systems, which are vital for healthcare and disaster prevention information networks. Sustainable development goals (SDGs) are widely recognized as key objectives for building a sustainable human society.[2] In this project, next-generation rechargeable batteries, such as all-solid-state batteries, advanced lithium batteries, LIBs, lithium–air batteries, sodium-ion batteries (SIBs), and magnesium metal batteries, have been developed to address the major SDGs (Fig. 1). However, the development of optimal and commercially viable rechargeable batteries is usually time-consuming. To accelerate battery research and development (R&D), another crucial target of this project is to establish a comprehensive battery research platform. This platform enables the simulation of cell and electrode performance based on the measurements of various electrochemical parameters of the active materials, ionic conductivities of the electrolytes, and physical and electronic parameters of the binders and conductive additives. The developed simulation program can predict the electrochemical behaviors of electrodes, cells, and pack batteries. By simulating the discharge and charge characteristics of porous electrodes and cells with various parameters, the platform provides valuable information on material selection, electrode design, and cell configuration to achieve overall battery improvement, thereby supporting the R&D of rechargeable batteries.

In addition, we have developed an automatic experimental system (the Smart Labo System), which can be used for rapid optimization of battery materials and electrode composition, significantly reducing the time required for battery R&D.

Furthermore, data science and computational science have been applied to nanoscale design. Data science aids in identifying new materials suitable for solid electrolytes (SEs), cathode, etc. The database has already been prepared and is accessible to researchers outside the National Institute for Materials Science (NIMS). Computational science can provide valuable information on the

†A part of this paper has been presented in the 65th Battery Symposium in 2024 (Presentation #1A01, #1A19).

§ECSJ Active Member
§§ECSJ Student Member
§§§ECSJ Fellow
K. Nishikawa orcid.org/0000-0002-7718-7606
T. Masuda orcid.org/0000-0001-7462-2177
Y. Xu orcid.org/0000-0001-8600-8748
Y. Tateyama orcid.org/0000-0002-5532-6134
S. Matsuda orcid.org/0000-0002-0640-3404
S. Nakanishi orcid.org/0000-0002-3313-2689
S. Hanada orcid.org/0009-0003-9656-8054
Y. Mukouyama orcid.org/0000-0003-3057-2467
T. Mandai orcid.org/0000-0002-2403-7794
N. Kuwata orcid.org/0000-0002-0736-6967
T. Ohnishi orcid.org/0000-0002-2333-7752
K. Kanamura orcid.org/0000-0002-0939-6748

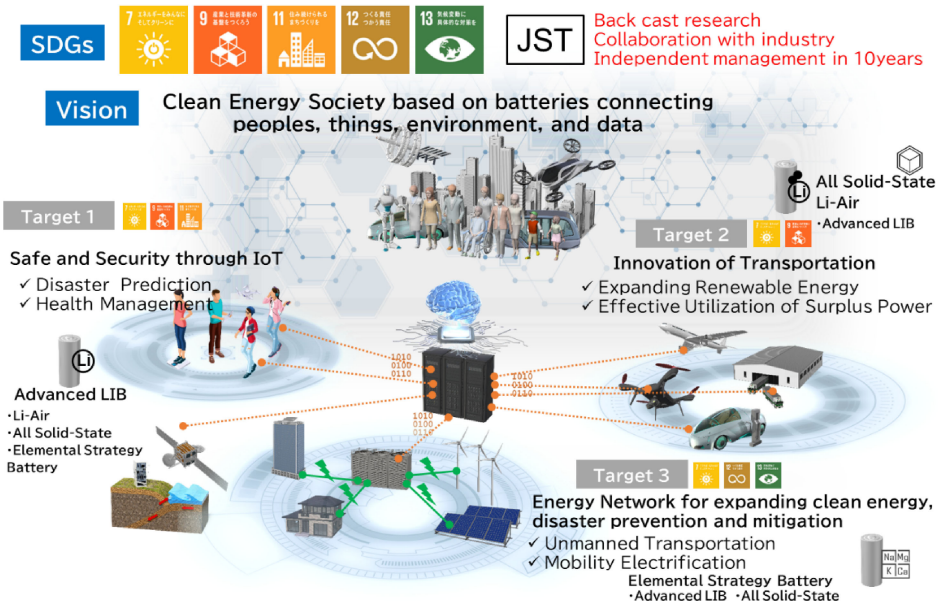

Figure 1. Targets of the COI-NEXT projects based on SDGs.

Li^+ pathway in SEs, the solid electrolyte interphase (SEI), and so on. Thus, the platform covers the research from the nanometer to the centimeter scale in batteries. In other words, it provides a multiscale simulation approach, combined with specific measurements, data, and computational science, for the rapid development of rechargeable batteries in both industry and academia.

2. Research and Development of Batteries in the World

Research on next-generation batteries has been conducted through various national projects. In the United States, the Joint Center for Energy Storage Research led by Argonne National Laboratory (ANL) focused on the development of multivalent ion and lithium-sulfur batteries. This research project was one of the largest in the physical chemistry field with funding of $240 million over 10 years. The Battery500 Consortium, a research initiative based on the Pacific Northwest National Laboratory, focuses on the development of lithium-metal batteries with a specific energy of 500 Wh/kg. Additionally, Li-Bridge is an industry-government collaboration committed to the development of a domestic supply chain for lithium batteries. As part of this project, ANL has published the "National Blueprint for Lithium Batteries 2021–2030," which outlines a roadmap to maintain long-term competitiveness.[3]

The European Battery Innovation project, involving 12 participating countries and primarily led by Germany, had a total budget of up to 2.9 billion euros supported by BATT4EU through Horizon Europe. This large-scale initiative integrated industry, government, and academia, focusing on strengthening supply chains, recycling, and life cycle assessment (LCA). The primary targets are LIBs and solid-state batteries.[4] Battery 2030+ aim to develop high-performance batteries with long cycle life and enhanced safety. This initiative not only develops battery materials but also focuses on long-term goals related to manufacturing and recycling. In this project, the research groups from Karlsruhe Institute of Technology, Technical University of Denmark, and Uppsala University have developed simulation techniques for the characterization and life prediction of batteries. In the United Kingdom, the Faraday Battery Challenge supported R&D in the Faraday Institution, covering all stages from basic research to commercialization of next-generation batteries. This project, with a total budget of £610 million from 2017 until 2025, targeted LIBs, all-solid-state batteries, lithium-sulfur batteries, and multivalent ion batteries.[5] The research team at Imperial College London works on battery modeling and simulation.

In Japan, the Green Technology of Excellence (GteX) project was launched in 2023 and supported by JST. GteX has 7 battery research teams, focusing on advanced LIBs, sulfide-based all-solid-state batteries, oxide-based all solid-state batteries, SIBs, Mg batteries, Li-sulfur batteries, and Li-air batteries. These teams bring together researchers from various disciplines, supported by a system that fosters collaboration across teams.[6] In addition, the New Energy and Industrial Technology Development Organization provides RISING 3, SOLiD-Next, and Green Innovation Fund for battery R&D, especially for electric vehicles. These projects emphasize not only material research but also practical applications, with a strong awareness of social implementation.

The development of new materials for solid and liquid electrolytes is essential for high-performance lithium batteries and next-generation batteries. Especially, new sulfide solid electrolytes are very important in fabricating all-solid-state batteries. Highly concentrated electrolytes are promising to improve the performances of LIBs, lithium-air batteries and Li-S batteries. Mg batteries generally need new electrolyte systems to stabilize the operation of Mg electrode and cathode. New solid electrolytes with high F^- ion conductivity are required for fluoride batteries. In the case of SIBs, new cathode and anode materials, electrolytes, and other components are also required. Various solid and liquid electrolytes have been developed through the abovementioned Japanese projects, attempting to find new materials that can drive significant innovations in the battery industry. In addition, some projects focus on establishing supply chains, recognizing the critical role batteries play in economic security.

The COI-NEXT project at NIMS is unique as it integrates simulation technologies, special measurement and evaluation methods, and some next-generation battery research under one roof. Additionally, the project in Europe includes a simulation technology for battery design and prediction of battery life. However, the project including simulation technology, advanced measurement and evaluation technologies, and some next generation battery research under one-roof is not present except for the COI-NEXT project conducting in NIMS. In addition, considering that most national projects on next-generation batteries do not include LIBs, another target of the COI-NEXT project is to establish a platform for the R&D of rechargeable batteries, especially LIBs.

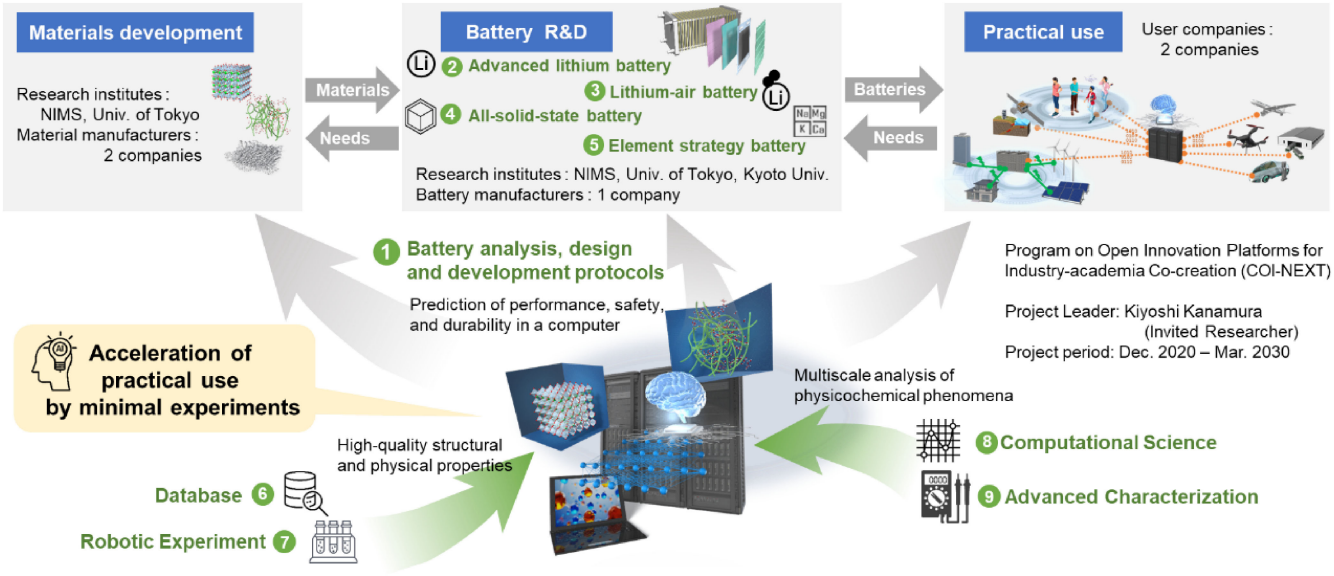

Figure 2. Schematic of ABC organization.

3. Center for Advanced Battery Collaboration

NIMS has established the Center for Advanced Battery Collaboration (ABC), which is supported by the COI-NEXT program. The center consists of nine research teams: the Protocol Team, Advanced Li Battery Team, Li-Air Battery Team, All-Solid-State Battery Team, Elemental Strategy Team, Data Resource Team, Smart Laboratory Team, Computational Calculation Team, and Advanced Measurement Team. All teams are managed by permanent researchers in NIMS, with a project director and a manager overseeing all teams. Moreover, the University of Tokyo and Kyoto University have joined this center to support innovative electrolyte research and synchrotron-based characterization, respectively. In addition, five companies supported the research center, focusing on the commercialization of fundamental research. Figure 2 demonstrates the schematic of the ABC organization. The most important aspect of this center is that all teams share the primary goal of establishing "protocols" for battery research. These "protocols" serve as tools not only for battery research but also for battery design based on the intrinsic parameters of the active materials, electrolytes, separators, and three-dimensional (3D) electrode structures. The project director oversees the development of these "protocols" and their applications within the center. Ultimately, ABC aims to publish the 'protocols' for commercial use in the Japanese battery industries.

4. Development of a Protocol for Rechargeable Batteries

As aforementioned, developing "protocols" is the primary purpose of this project. The ideal "protocol" is a multiscale model based on the experimental results from the atomic scale to the millimeter scale, theoretical computational calculations, and data-driven techniques, including artificial intelligence (AI)–based machine learning (ML) technology. We now focus on the electrode structure to simulate the charge and discharge curves. Newman et al.[7] proposed a one-dimensional simulation model, and several researchers have used the Newman model to predict battery performance.[8,9] However, battery electrodes are "composites" with a complicated 3D structure, composed of active material particles, nanocarbon as a conductive agent, and binders. The chemical species in the electrolyte can transfer to the electrode pores, and ionic conduction in the electrode significantly influences the electrochemical characteristics. Considering that LIBs have been used in various applications, the "protocol" aims to propose the suitable electrode structure for each application. Inoue and Kawase[10] reported that the 3D structure affects the ionic conduction of battery electrodes. To incorporate such 3D structural characteristics into a simulation scheme, it is important to obtain the real 3D electrode structure experimentally. The focused-ion-beam scanning electron microscopy (FIB-SEM)-based auto-slicing technique, especially plasma FIB-SEM (P-FIB), can examine extremely large sample areas. In this project, P-FIB was used to obtain a 3D structure with a 300-µm width and a 50-nm pitch from 4000 sliced images (Fig. 3). These images were reconstructed to analyze the interaction in the electrode pores, and the tortuosity and porosity were evaluated using AVIZO (Thermo Fischer Scientific Co.). In this project, the 3D structure of the electrode was used to estimate the ionic conductivity of the electrode, which was then validated through four-electrode cell measurements. In addition, the other parameters, such as viscosity, diffusion coefficient, transference number of the electrolyte, exchange current density, and diffusion coefficient of the active materials were also obtained from a series of experiments. Figure 4 shows the typical experimental techniques, including single particle measurement and ionic conductivity measurement using a four-electrode cell. The simulation of the charge/discharge curves will be performed based on the finite element calculation technique using COMSOL Multiphysics (COMSOL Inc.) combined with our experimental results and real 3D images.

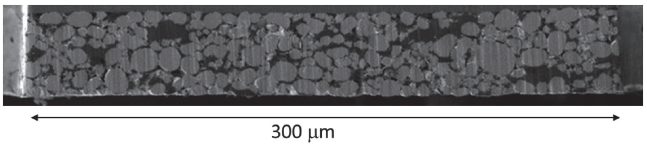

Figure 3. Cross-sectional image of LIB positive electrode obtained from P-FIB slicing.

However, analyzing the real 3D images obtained from the auto-sliced FIB takes a long time. Recently, ML techniques have demonstrated the potential to accelerate electrode structure analysis, where only a few images are required to estimate the 3D parameters of an electrode. Accelerating this type of R&D is also important from the perspective of contributing to industry.

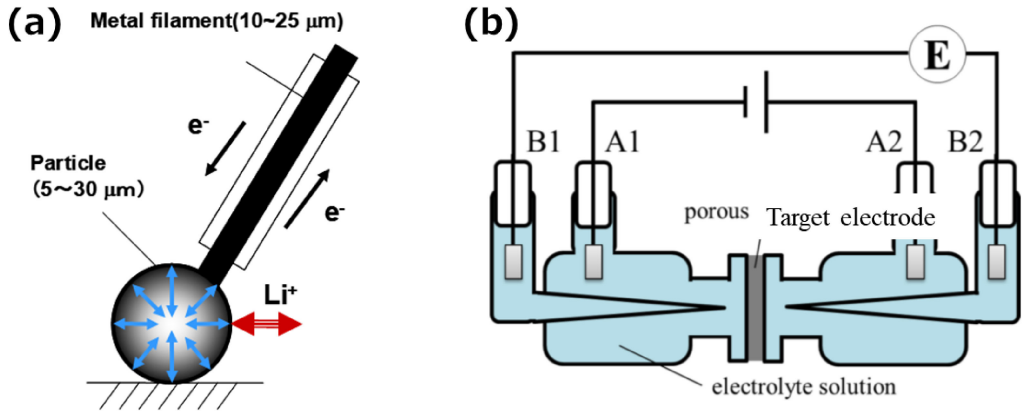

Figure 4. Schematic of the experimental setup for measuring some parameters of battery materials: (a) single-particle measurement setup for electrode active materials and (b) four-electrode cell for the ionic conductivity measurement of the composite electrode.

The manufacturing process of LIB electrodes is also crucial for the battery industry. The battery electrode is fabricated by mixing active materials, conductive agents, binders, and solvents, followed by casting the mixture on a current collector, drying, and pressing. Nowadays, the process is established through many years of experience. However, the physical chemistry aspects of each process have not been described theoretically. The experimental parameters were varied for optimization, and the experimental results were shared in the "Materialize" project conducted at Tohoku University supported by JST. The "Materialize" project will simulate the battery manufacturing process based on theoretical calculations. Establishing this process protocol will have a significant impact not only on the battery industry but also across several engineering fields.

5. Measurement System Development

5.1 Formation of $LiCoO_2$–Li_3BO_3 composite electrode on an SE sheet and *operando* chemical analysis

$LiCoO_2$ fine powder was applied to a $Li_{6.6}La_3Zr_{1.6}Ta_{0.4}O_{12}$ (LLZT) sheet precoated with a Nb-thin protection layer. The composite was then annealed at a relatively low temperature after the wet chemical treatment with aqueous LiOH and H_3BO_3 solutions to form a $LiCoO_2$–Li_3BO_3 electrode on the LLZT sheet.[11] After binding a Li foil on the other LLZT sheet side, the electrochemical reactions at the $LiCoO_2$-Li_3BO_3 composite electrode were investigated using a laboratory-based hard X-ray photoelectron spectroscopy (HAXPES) apparatus equipped with a bias application system.[11]

The first charge/discharge cycle clearly showed an irreversible capacity, probably due to the substance instability; however, after a few cycles, the charge/discharge process became reversible with a capacity density of ~85 mAh/g and a Coulombic efficiency of ~100 % (Fig. 5a). The photoelectron spectrum in the Co 2p region obtained in the pristine state exhibited a characteristic $LiCoO_2$ profile, which comprised a sharp main peak and a shoulder corresponding to the Co^{3+} species and the final state effect, respectively (Fig. 5b).[11] After charging (extracting Li ions from $LiCoO_2$), the shoulder broadened to a higher binding energy due to the oxidation of Co^{3+} to Co^{4+} (Fig. 5b). In addition, the main peak shifted to a lower binding energy as the delithiation of $LiCoO_2$ caused an electronic state transition from p-type semiconducting to metallic. After subsequent discharging (insertion of Li ions into $Li_{1-x}CoO_2$), the shoulder returned to its original width due to the reduction of Co^{4+} to Co^{3+} (Fig. 5b). These reversible interconversions were successfully observed during the successive charge/discharge cycles.

After calibrating the photoelectron spectra, using the B 1s peak corresponding to Li_3BO_3 at 191.5 eV as a standard, the Co^{3+} peaks shifted consistently with the cell voltage during the charge/discharge cycles (Fig. 5c). This shows that the Fermi level of $LiCoO_2$ shifts in accordance with the cell voltage, and Li_3BO_3 acts as a bulk electrolyte throughout the charge/discharge cycle. The analysis area and information depth addressed by the *operando* HAXPES were relatively large because the Cr-Kα source (5414.9 eV) used in this project generated an X-ray beam with a spot size of φ200 μm. However, the width and symmetry of the Co^{3+} peak were maintained during the charge/discharge cycle, confirming that $LiCoO_2$ and Li_3BO_3 were uniformly dispersed and well-connected to each other in the composite electrode.

5.2 *Operando* nanomechanical mapping of electrode active materials

Understanding the nanoscale and mesoscale mechanical properties, as well as the morphologies of the electrode active materials in response to the state of charge, is essential for designing highly durable cells that can manage the volume change of the active materials. An *operando* bimodal atomic force microscopy (AFM) system was developed to perform nanomechanical mapping, as well as topographic imaging (Fig. 6a), of the electrode active materials in an all-solid-state cell configuration during charge/discharge cycles. The system was used to analyze amorphous Si electrodes that underwent notoriously severe volume change during lithiation/delithiation.[12]

Upon lithiation, Young's modulus drastically changed from ~80 to ~55 GPa due to the formation of lithium silicides, Li_xSi, in the initial stage and then monotonically decreased to ~40 GPa with increasing capacity density, i.e., Li content x in Li_xSi (Figs. 6b and 6c). The correlation between Young's modulus and capacity density agreed well with those estimated by theoretical calculations and nanoindentation measurements, confirming the reliability of this technique.[12] Collaboration is now ongoing with a multiscale modeling approach to mitigate mechanical degradation.

6. Data Science

Data-driven research can facilitate battery development by enabling efficient discovery and optimization of materials. By analyzing large datasets, researchers can identify patterns and predict properties, such as ionic conductivity, to accelerate innovation. ML models reduce the reliance on time-intensive experiments, streamlining the screening and design of advanced materials. This approach also uncovers critical compositional or structural features to optimize existing materials, thereby enhancing their performance. To further accelerate the exploration of new battery materials, we have compiled data from relevant research papers to develop an AtomWork Battery database.[13] In addition, we

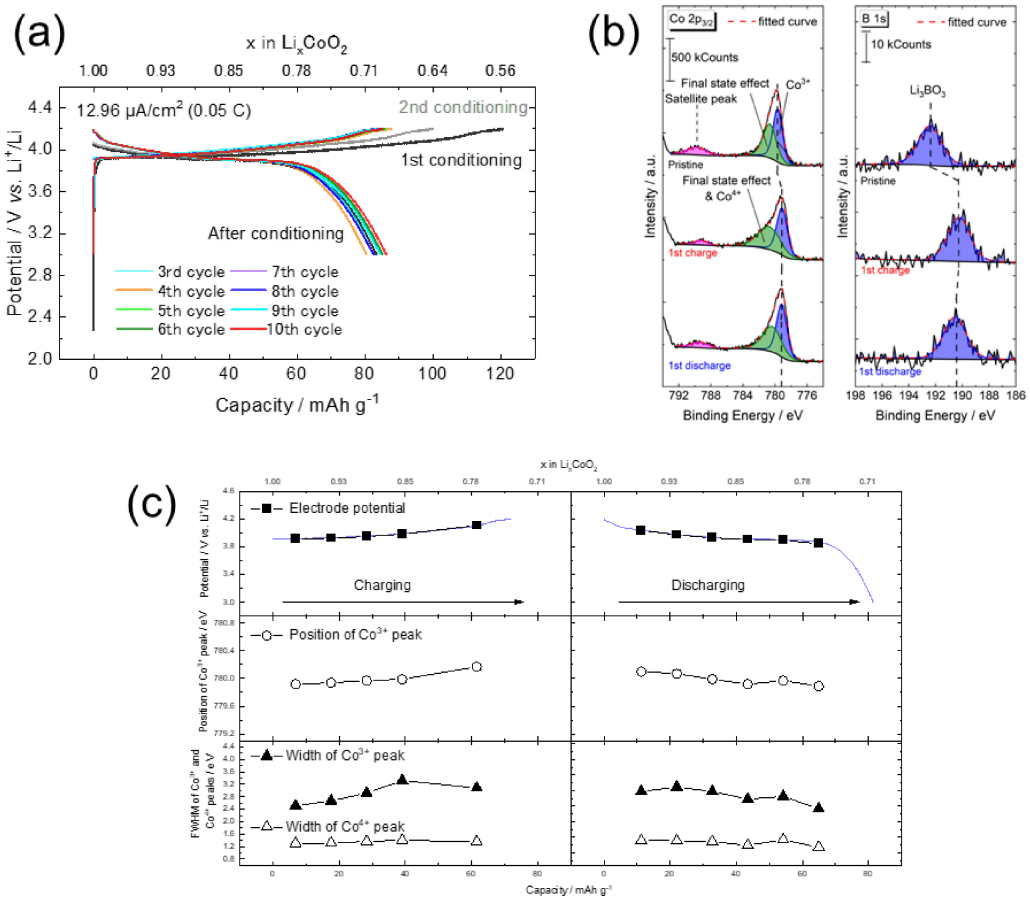

Figure 5. (a) Galvanostatic charge/discharge potential profiles of the $LiCoO_2$–Li_3BO_3/LLZT/Li cell; (b) Co 2p and B 1s photoelectron spectra of the $LiCoO_2$–Li_3BO_3 composite electrode in the pristine state and after charging/discharging; (c) (top) galvanostatic charge/discharge potential profiles, (middle) positions of Co^{3+} peak after calibration with the B 1s peak, and (bottom) full width at half maxima of Co^{3+} and Co^{4+} peaks.[11]

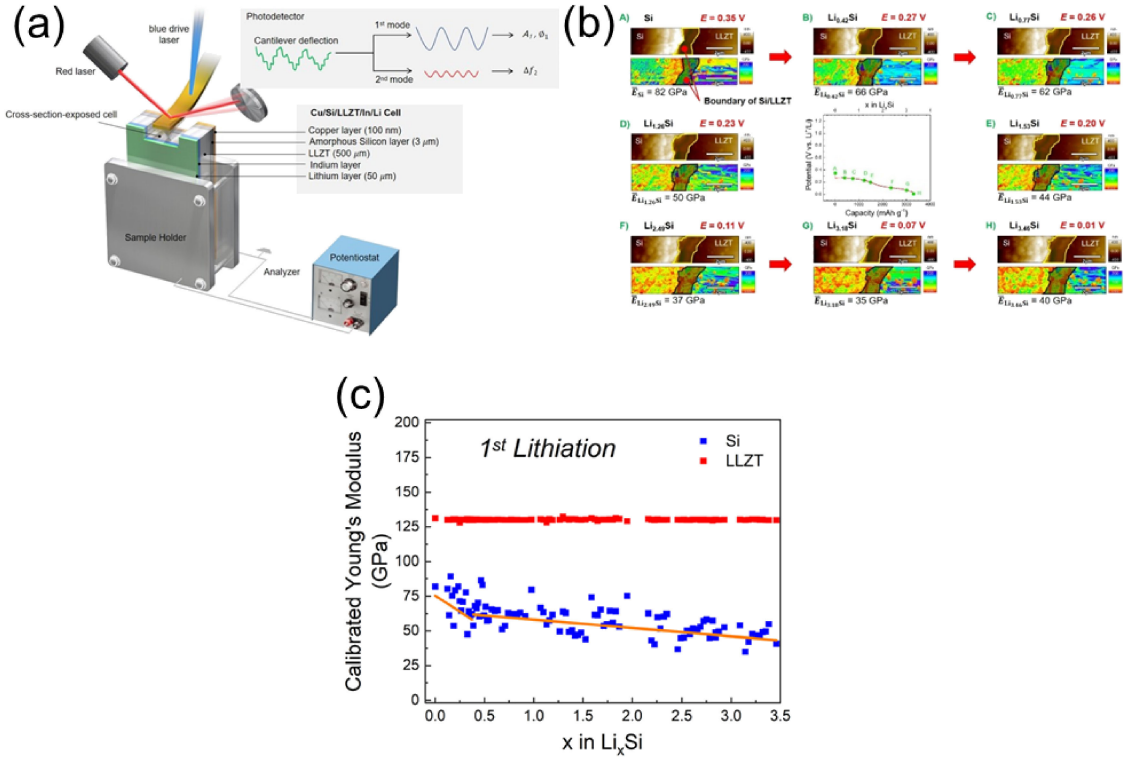

Figure 6. (a) Schematic of the *operando* bimodal AFM; (b) topography images and Young's modulus maps of the Si thin film during lithiation; (c) average Young's moduli of the Si thin film.[12]

Table 1. Number of data entries of the AtomWork Battery database in 2023.

	Solid electrolyte	Cathode material	Total
Paper	538	350	888
Material	2,721	1,122	3,843
Chemical system	461	214	670
Substance	583	252	812
Battery cell	222	1,344	1,566
Material property	23,184	495	23,679
Charge/discharge capacity	1,798	15,667	17,465
Curve	6,504	9,694	16,198
Data point of the curve	246,705	770,915	1,017,620

created AtomWork Creator ML tools for property prediction and material design.

6.1 Battery material database

The AtomWork Battery database contains data on SEs and cathode-active materials extracted from relevant research papers. Using natural language processing, we identified and retrieved papers related to these materials from a collection of over 350,000 publications. Data extraction and curation are performed by highly trained editors, who systematically extract material information, such as chemical composition, phase composition, synthesis processes, and properties, as well as battery-related information, including battery composition and charge/discharge capacities. The database includes the material properties, such as ionic conductivity, diffusion coefficients, and activation energy. The current number of data entries in the database is summarized in Table 1. The database is updated annually, with approximately 400 new papers added each year.

6.2 ML tools for battery material design

AtomWork Creator is a Python package developed using ML methods for property prediction and material design. Its main functions include the following.

1) Encoding/Decoding of Periodic Descriptors: These are the original descriptor sets representing the chemical compositions of the materials.
2) Prediction of Material Properties: AtomWork Creator leverages ML models to forecast material properties.
3) Design of Chemical Compositions: AtomWork Creator identifies the optimal chemical composition of a material to achieve the best properties.

By default, the package includes ML models trained on AtomWork Battery data to predict ionic conductivity and design materials with high ionic conductivity. Users can also integrate their own datasets to predict other material properties, making the tool highly adaptable to various research needs.

7. Computational Science Team

This team has four targets: (1) creation of a database regarding the self-diffusivity (tracer diffusion coefficient) of SEs to contribute to the battery protocol developed by the Protocol Team; (2) development of new workflows, techniques, and codes for high-throughput self-diffusivity calculations at the density functional theory (DFT)–based molecular dynamics (MD) level; (3) elucidation of ion and electron transfer mechanisms through DFT calculations; (4) collaboration with the experimental teams.

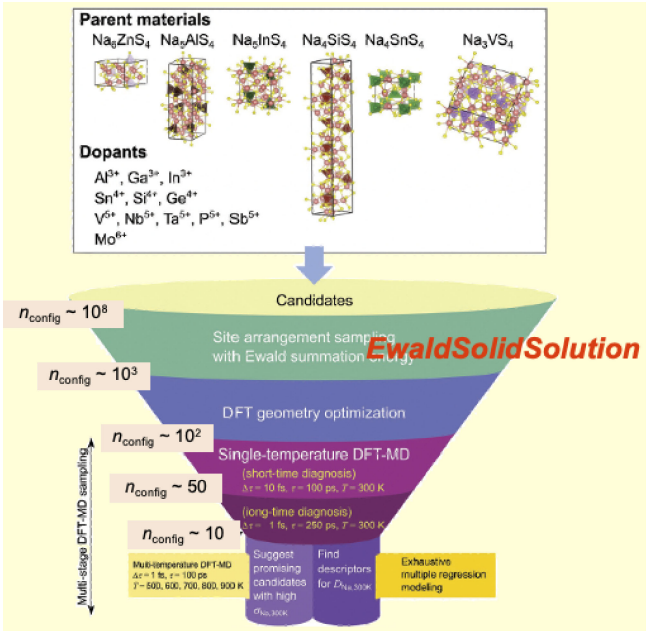

Figure 7. High-throughput flow for ion self-diffusivity screening at the DFT-MD level, with EwaldSolidSolution calculations.

In this section, we especially focus on the targets (1) and (2). In conventional SE searches, the crystal structure is predefined, and materials are selected through ion exchange using genetic algorithms or Bayesian optimization. Our approach, though more brute force, is computationally effective through parallelization, allowing us to construct a workflow that can equally consider various crystal structures.[14]

For example, we first extracted all 7 Na-ion sulfide $Na_nM_mS_x$ (M = Metal) from the ICSD database and used them as "mother" crystals (See Fig. 7). We then considered the doping of M′ metal ions with a different valence (m′) into the metal ion M position. Because of charge neutrality, Na-ion defects corresponding to the doping amount were introduced. If the M′ ion with a higher valence than the M ion is chosen, Na-ion vacancies are introduced. Four doping levels (1/8, 1/4, 3/8, and 1/2) yield 170 possible compositions of $Na_nM_mM'_{m'}S_x$. Converting this into the number of ion configurations, a total of approximately 100 million possible structures are generated, given the degrees of freedom in the positions of dopant M′ ions and the Na-ion vacancies in the supercell with around 200 ions used in this study.

Because it is practically impossible to perform 100 million DFT calculations, we developed a point-charge-based Ewald energy (E_{Ewald}) calculation code, called *EwaldSolidSolution*, as a surrogate model (a proxy for DFT calculations).[15] The model selects stable configurations and removes unstable ones based on the electrostatic energy for each given ion configuration. With the code parallelization, the process is fast enough to finish in approximately half a day for around 100 million configurations. Unlike Bayesian optimization, the EwaldSolidSolution strategy is to exhaustively screen the material parameter space using parallelized calculations. Five low-energy structures obtained for each composition from the surrogate model screening were subjected to DFT geometry optimization. Convex hull analysis is performed, and the metastable structures with a hull energy of less than 100 meV/atom are selected for the next step. The 170 $Na_nM_mM'_{m'}S_x$ structures remaining at this stage were finally screened for the Na-ion self-diffusivity, which is related to ionic conductivity. The static DFT calculations of the activation energy using the nudged elastic band method is not appropriate to such complex atomic structures. Therefore, from the viewpoint of

the concerted dynamics of ions, the DFT-MD approach is employed to directly calculate the self-diffusivity based on mean-squared displacements.

This DFT-MD sampling is very time-consuming, and it is virtually impossible to compare all 170 structures at the same level. Therefore, a short diagnostic flow was introduced to perform self-diffusivity screening by increasing the MD time step by a factor of 10 (to 10 fsec) and sampling for a short time (around 100 psec level). This is a reasonable approximation unless the period of vibrations in the ionic framework related to Na-ion diffusion is slower than this time interval. After narrowing down the targets to 44 structures based on the short-time diagnosis, the Na-ion self-diffusivity at room temperature was obtained by DFT-MD sampling at the 250-ps level using the conventional 1 fsec as the time step. The Na ion conductivity at room temperature was obtained through the Nernst-Einstein law. At this stage, the number of structures was narrowed down to 27. It is imperative to investigate the correlation between the so-called long- and short-time diagnoses. Our assessment confirms that a reasonable level of positive correlation exists. In other words, the structures with a conductivity of 1 mS/cm or higher in the short-time diagnosis also have high ionic conductivity in the long-time diagnosis. Throughout the flow explained above, the search for SEs with high ionic conductivity can be accelerated by considering the realistic concerted dynamics. The flow and code have been extended to cover a wider material parameter space.[16–18]

8. Smart Labo System

The application of data-driven approaches for material discovery has gained considerable attention in the research of rechargeable batteries. Rather than relying on experience and intuition to select new materials, data science techniques can significantly reduce the time and cost involved in finding the most suitable materials. Our research group has developed an automated robotic experimental system to study battery electrolytes.[19] Based on a liquid-handling dispenser and microplate transfer technologies established in biochemistry, a microplate-based electrochemical cell (E-microplate) was created. In this design, electrodes and electrolytes are placed inside the wells of a 96-well microplate, which mimics the environment of a conventional coin-type cell. Using the E-microplate, we constructed an automated experimental robot (Fig. 8) comprising a noncontacting-type liquid-handling dispenser, a multichannel electrochemical analyzer, and a robotic arm for transporting the microplate. For automated operation, an E-microplate with electrodes and stock solutions was prepared by filling it with additive solutions in the storage magazine. Once the solution formulation recipe was input into the automated experimental robot, the E-microplate was transported from the storage magazine to the liquid-dispensing stage by the robotic arm. The prescribed additive solutions were then dispensed into each well of the E-microplate. Once the dispensing process was completed, the E-microplate was transferred to the electrochemical measurement unit, where the battery properties of each well were evaluated. The used E-microplate is then transported to the disposal box. By continuously executing this sequence of operations, the automated experimental robot achieves a high-throughput measurement of over 1000 samples per day. Although this system provides superior experimental throughput, careful experimental design is critical for efficiently exploring the electrolyte compositions in the large search space. For example, selecting 5 of 20 candidate compounds results in over 10^7 possible combinations, making it impractical to comprehensively evaluate all possibilities, even with robotic experiments. To efficiently identify the optimal electrolyte additive combinations within such an extensive search space, the application of data-driven methodologies proves highly effective. In fact, our research group has successfully discovered electrolyte compositions that improve the cycle life of lithium-air batteries using the Bayesian optimization technique.[20] To further enhance the interaction between data-driven methodologies and automated robotic experiments, we developed NIMO (NIMS Orchestation System) (Fig. 9) to accommodate multiple AI methods with varying exploratory objectives while using a unified data format.[21] Currently, NIMO supports several AI methods as standard implementations, including Bayesian optimization via PHYSBO,[22] BLOX nonobjective search

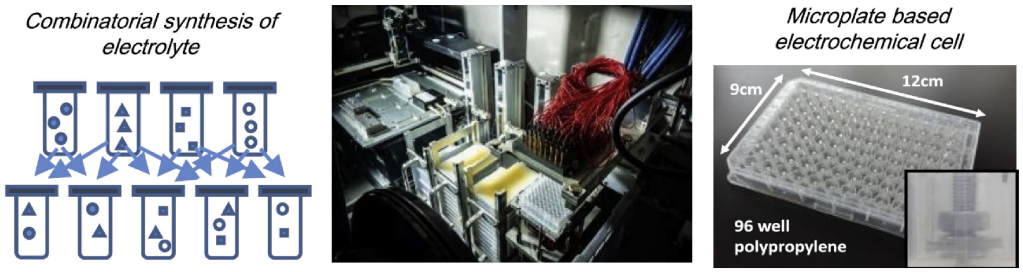

Figure 8. Schematic and photograph of data-driven high-throughput automated robotic experiments to search multicomponent electrolytes.

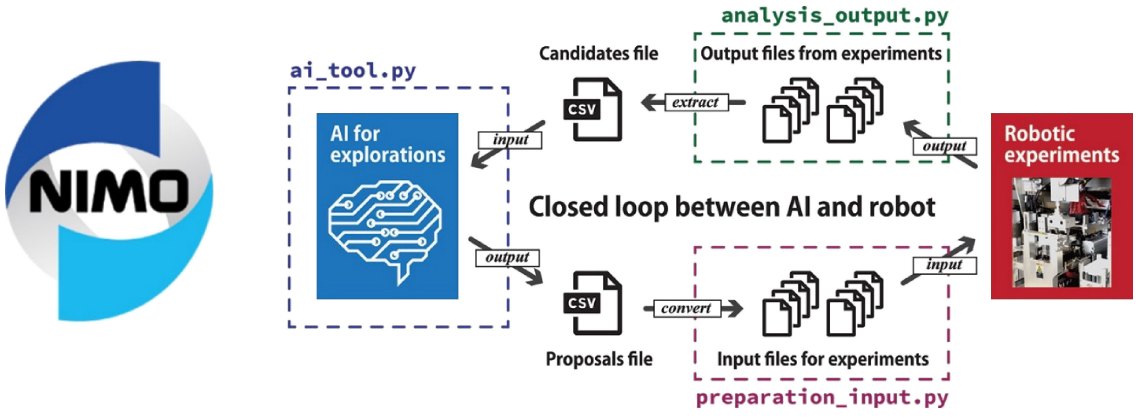

Figure 9. Procedures of using NIMO to achieve a closed loop between AI methods and robotic experiments.

method,[23] and random sampling. By introducing a unified data format, NIMO facilitates the seamless integration of new algorithms for autonomous experiments.

9. Next-Generation Battery Research

9.1 Advanced LIBs

The research focuses on an advanced LIB comprising a high-voltage and high-capacity cathode, a nonflammable electrolyte, and a Si-based or Li metal anode. As a promising anode material for next-generation batteries, Li metal offers a very large capacity. However, its practical application is restricted by the large irreversible capacity induced by the side reactions during cycling, as well as the formation of nonuniform morphology (so-called dendrite formation). In addition, the charging and discharging processes induce electrodeposition and dissolution reactions, respectively, leading to large volume changes during cycling and presenting challenges for battery design. To develop a Li metal anode for a long-life battery, it is crucial to understand the electrodeposition and dissolution mechanisms of Li. Several researchers have reported the morphological variation of electrodeposited Li metal by surface and cross-sectional SEM observation.[24,25] As mentioned in the Protocol section, the P-FIB system is highly capable of capturing 3D images. The 3D structure of electrodeposited and cycled Li metal is very meaningful information for understanding the deposition and dissolution mechanisms. Figure 10 shows the 3D structure of electrodeposited Li metal on Cu obtained in a 4 mol dm^{-3} LiFSI-DME electrolyte at 0.5 mA cm^{-2} for 4 h.[25] Pores are observed in the electrodeposited layer, and the porosity depends on the applied current density. In addition, by increasing the current density, the deposit size becomes smaller. The sphericity is also affected by the current density. These parameters relate to the side reaction between the electrodeposited Li metal and electrolyte. This study provides insights into the morphological variation of Li metal during the charge/discharge cycle. In addition, a Li metal anode is necessary for the interface design because Li metal is a very reactive metal that decomposes electrolyte species. The decomposition deteriorates battery reversibility and causes electrolyte depletion. To restrict the side reaction, an artificial layer is introduced on the Li metal surface.[26] The layer needs both hardness and softness against expansion and contraction. In this project, a polymer gel with hydrogen bonds[27] was introduced to the Li metal anode, which effectively inhibits the undesirable side reactions and improves the cycling performance. Our future work will focus on optimizing the artificial layer for various applications.

9.2 Rechargeable magnesium battery (RMB) and sodium ion battery (SIB)

Elemental strategic batteries, based on widely available elements with low resource constraints and geopolitical risks, are expected to serve as emergency and large-scale power sources to mitigate output fluctuations in renewable solar and wind power generation. In this project, we aim to develop innovative materials and fabrication techniques that will pave the way for commercial RMBs and SIBs.

9.2.1 RMBs

Rechargeable batteries based on magnesium metal negative electrodes are of particular interest due to the fascinating properties of magnesium metal.[28] A considerable challenge that hinders the launch of RMBs in the battery market is the absence of electrolyte materials perfectly compatible with the highly reactive magnesium metal. Conventional electrolyte solutions can trigger undesired side reactions with magnesium, which causes polarization and inactivates the reversible magnesium plating/stripping reactions.[29,30] Inhomogeneous passivation due to the native film on the bulk magnesium metal induces 3D deposition behaviors, which may result in short-circuit.[31–33]

To prevent the above undesired events related to the instability of magnesium metal–electrolyte interface, we fabricated an artificial interface on magnesium via a simple galvanostatic replacement reaction using $ZnCl_2$, in which Cl plays a pivotal role in etching the native-passivated surface.[34] A subsequent spontaneous replacement reaction between Zn^{2+} and the activated magnesium resulted in the formation of the homogeneous Mg^{2+}-conductive Mg–Zn–Cl-integrated functional interface. The $ZnCl_2$-modified magnesium electrodes exhibited excellent magnesium plating/stripping performance (Fig. 11a), thereby greatly enhancing the cycle life of the battery. In addition, $ZnCl_2$ modification is effective in improving the areal capacities, rate capability, and compatibility with the PTFE-based thin separator (Figs. 11b–11d).[33]

A combination of various analytical techniques was employed to reveal the mechanism underlying the positive effect of $ZnCl_2$ modification. Grazing-incidence X-ray diffraction, position-resolved electron diffraction, and electron energy loss spectroscopy, SEM observation and subsequent electron backscatter diffraction analysis showed that the amorphous nature and hybrid hierarchical composition of the Mg–Zn–Cl-integrated interface and favorable Mg^{2+} conductive characteristics were responsible for the exceptionally stable plating/stripping performance.[33]

The Zn-based interface is also effective in preserving the electrochemical activity of magnesium under a dry air atmosphere, whereas the non-modified interface does not show any reversible electrochemical reactions under the same experimental conditions.[35] The techniques developed in this study will open the possibility of high-energy-density practical RMB materialization by adapting the existing separator materials and manufacturing systems used for LIBs.

9.2.2 SIBs

SIBs are also regarded as alternative energy storage options because of the abundant natural availability of their component materials.[36] Although practical SIBs have already been in the market for electric vehicles and bicycles,[37,38] further development is still required for SIBs to achieve performance comparable to or better than current LIBs. In this context, the storage performance of SIBs is often overestimated in the literature due to oversight of the engineering aspects and the performance difference between the half-cell and full-cell configurations. As this COI-NEXT program aims to accelerate battery research and shorten the lead time for battery fabrication, we investigated the effect of engineering aspects on the performance of SIB full-cells.

Commercially available, well-studied compounds of $Na_3V_2(PO_4)_3$ (NVP) and hard carbon (HC) have been adopted as positive and negative electrode materials, respectively. Upon systematic comparative survey concerning the electrode composition and the capacity balance between the positive and negative electrodes (N/P ratio), the optimum composition of NVP and HC electrodes are proposed, and the significant detrimental effects of reactive Na^+ consumption by full-cell operation on the energy density of SIBs are clarified.[39]

By adopting adequate pre-sodiation techniques to compensate for the loss of Na^+, SIB full-cells with a prospective energy density of 100 Wh kg^{-1} have been achieved using conventional NVP and HC. This study provides guidelines for designing a practical full-cell SIB configuration with high-energy-density (Fig. 12).

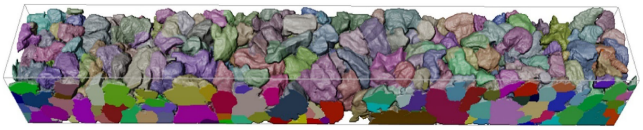

Figure 10. 3D structure of electrodeposited Li metal obtained in a 4 mol dm^{-3} LiFSI-DME electrolyte at 0.5 mA cm^{-2} for 4 h.

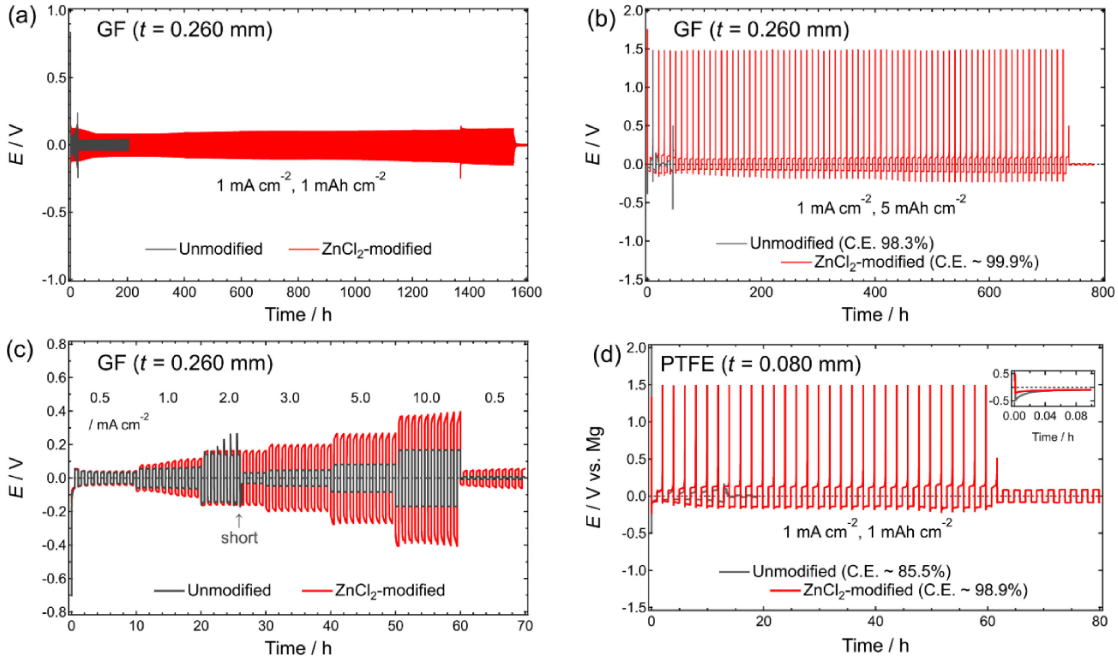

Figure 11. (a) Long-term galvanostatic cycling profiles of symmetric cells; (b) galvanostatic cycling profiles of asymmetric cells measured at a large areal capacity of 5 mAh cm^{-2}; (c) polarization curves of the rate capability test; (d) galvanostatic cycling profiles of asymmetric cells with a typical PTFE membrane filter, reproduced from Ref. 33 with permission from Elsevier (2024 Copyright CC BY-NC-ND).

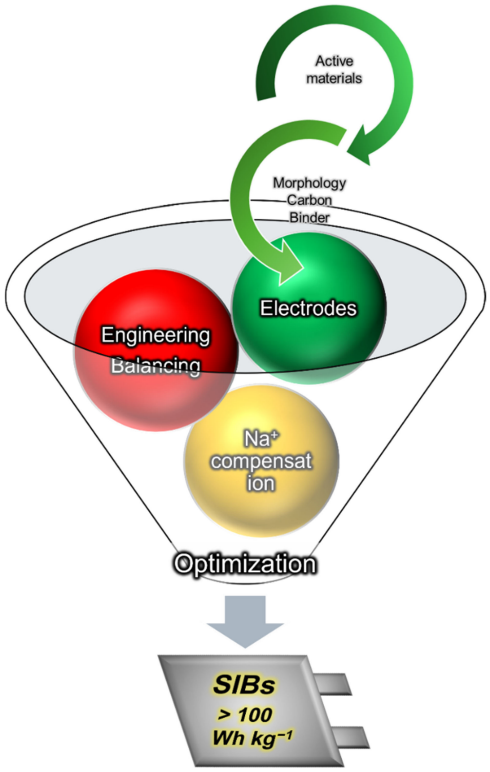

Figure 12. Schematic of the strategy to achieve high-performance SIBs.

9.3 Li-air battery

In lithium–oxygen batteries with nonaqueous electrolytes, Li_2O_2 deposits within the porous cathode via oxygen (O_2) reduction during discharge. Ideally, during charging, Li_2O_2 decomposes oxidatively to release O_2. However, in practice, electrode reactions, material transport, and the physical properties of Li_2O_2 interact in a highly complex manner, increasing the charging voltage and hindering stable charge/discharge cycling. Consequently, establishing distinct correlations between the charge/discharge cycle performance and experimental conditions is challenging, which complicates the development of design guidelines for these batteries. To address these challenges, model-based development has emerged as an effective approach, enabling quantitative analysis of the relationships between factors and outcomes.[40,41] In this context, we developed a finite element model that incorporates two distinct discharge reaction pathways (Fig. 13a), side reactions, and delithiation reactions during charging.[42,43]

Figure 13b presents a specific example of our modeling approach to evaluate the impact of the oxygen diffusion coefficient (D_{O2}). The results indicate that higher D_{O2} values lead to more extensive deposition of Li_2O_2 within the porous cathode, which enhances the cycling performance. A detailed analysis reveals that increasing D_{O2} improves the O_2 concentration within the porous cathode, thereby favoring the solution-mediated pathway. This is consistent with previous research, which reported that larger D_{O2} values enhanced battery performance.[44] Another key example involves simulating O_2 evolution behavior during charging, where the O_2 evolution efficiency decreases not only due to the progression of the side reactions but also during delithiation—a primary charging reaction. To address these complexities, we developed an improved model that treats LiO_2 as an intermediate in the charging reaction. This advancement enables the simultaneous prediction of charge/discharge cycle characteristics and O_2 evolution efficiency.[43] Figure 13c shows the calculated distribution of the four critical chemical species ($Li_2O_2^{surf}$, $Li_2O_2^{sol}$, LiO_2^{surf}, and LiO_2^{sol}) within the porous cathode. Although obtaining such detailed information experimentally is challenging, the simulation platform visualizes the spatiotemporal distribution of the chemical species under various conditions, clarifying their contribution to the voltage profiles and O_2 evolution behavior.

In real experimental systems, it is infeasible to independently evaluate the effects of parameters, such as the porosity and pore size of the porous cathode or the solution properties of the electrolyte, on

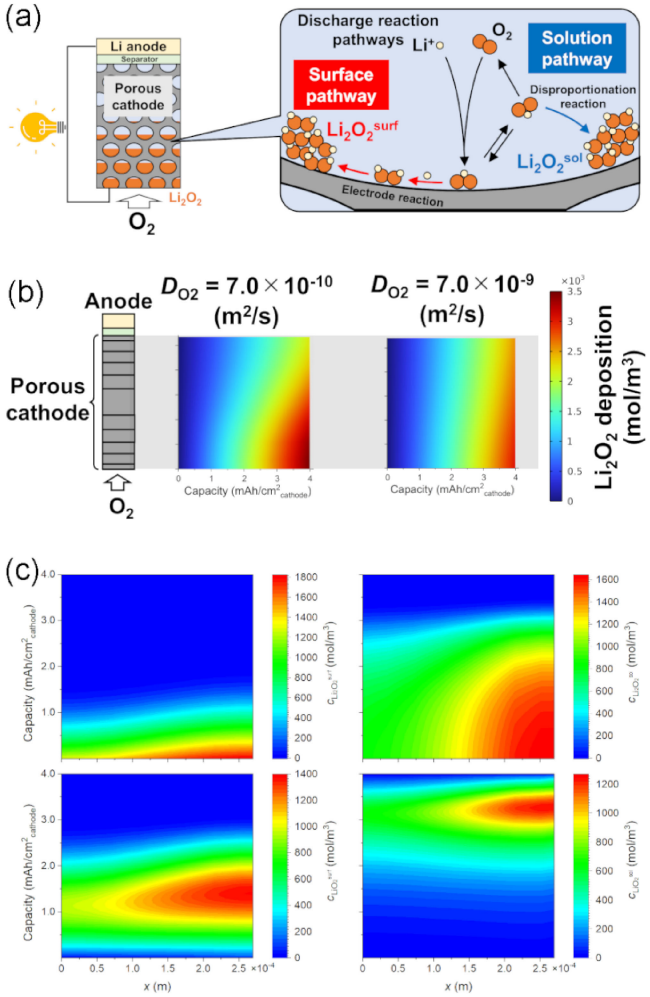

Figure 13. (a) Two discharge reaction pathways, (b) Impact of the oxygen diffusion coefficient (D_{O2}) on the discharge reaction, adapted with permission from Ref. 42, Copyright 2023 American Chemical Society. (c) Calculated distribution of four critical chemical species ($Li_2O_2^{surf}$, $Li_2O_2^{sol}$, LiO_2^{surf}, and LiO_2^{sol}) within the porous cathode. Adapted from Ref. 43 licensed under CC BY 4.0.

battery performance. To address this limitation, we developed a foundational model for quantitative analysis of their effects. This model is expected to facilitate the development of design guidelines for new materials, while also offering in-depth insights into the phenomena observed in experimental studies.

9.4 All-solid-state batteries

Solid-state batteries have attracted considerable attention because of their potential to improve efficiency and reliability. A key factor in advancing this technology is to improve the performance of SEs. Our research focuses on the precise measurement of the diffusion coefficient of lithium and the identification of the critical factors that contribute to the SE performance. For this purpose, we develop a methodology for measuring lithium diffusion using materials with ideal structures, especially single crystals. The target materials are SEs with an A-site defective perovskite structure. The perovskite-type lithium lanthanum titanate, $Li_{0.29}La_{0.57}\square_{0.14}TiO_3$ (LLTO), exhibits an ionic conductivity of 1×10^{-3} S cm^{-1} at 298 K.[45–47] A similar structure, lithium lanthanum niobate, $Li_{0.08}La_{0.31}\square_{0.61}NbO_3$ (LLNbO), with more vacancies (□) than LLTO, is expected to form fast diffusion pathways. Based on this, large single crystals of LLNbO that are stable in air have been prepared, and their ionic conductivity can reach 2×10^{-4} S cm^{-1}.[48–50]

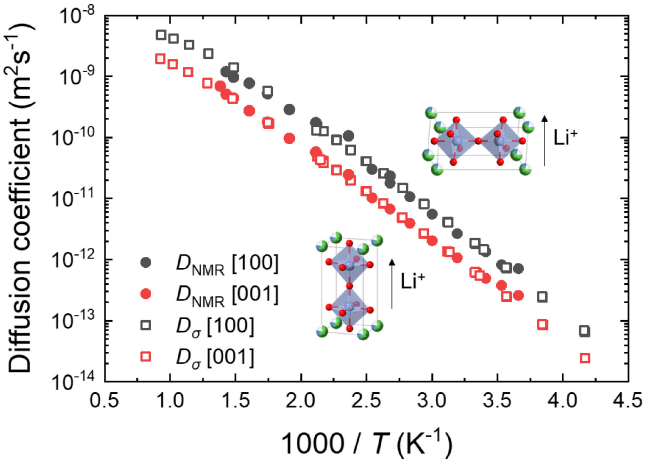

Figure 14. Diffusion coefficients determined from PFG-NMR (D_{NMR}) and ionic conductivity (D_σ) of LLNbO single crystals in the [100] (black) and [011] (red) directions. Reprinted from Ref. 51.

The diffusion coefficients of large single crystals of LLNbO were determined by integrating pulsed field gradient nuclear magnetic resonance (PFG-NMR) and high-frequency impedance methods. This approach effectively eliminates the grain boundary effects and facilitates the experimental clarification of the anisotropy of the bulk diffusion coefficients.

Figure 14 shows the temperature dependence of the diffusion coefficient (D_{NMR}) determined by PFG-NMR and the conductivity diffusion coefficient (D_σ) obtained from the ionic conductivity via the Nernst–Einstein law.[51] The D_{NMR} of LLNbO is 1.2×10^{-12} m^2 s^{-1} (293 K) in the [100] direction. The D_{NMR} values are anisotropic, with the [100] and [001] directions differing by a factor of about 3. The D_{NMR} of LLNbO [100] is comparable in magnitude to that of LLTO, indicating that the perovskite crystal structure of LLNbO is suitable for fast diffusion. The difference in ionic conductivity between LLTO and LLNbO is attributed to the number of carriers. The D_{NMR} and D_σ values are in excellent agreement, indicating that the Li^+ ions diffuse independently.

For an ideal crystal structure, the activation energies of the hopping Li^+ in the [100] and [001] directions should be different, resulting in different diffusion coefficients. However, the activation energy, which is within 0.32–0.33 eV near 300 K, exhibits no anisotropy in any orientation. This observation suggests that the presence of defects, such as La in the La-poor layer, within the actual LLNbO single crystals can act as pathways for the diffusion in the [001] direction. Consequently, we conclude that the average crystal structure, along with the defects in medium-range order, plays a pivotal role in the dynamics of the long-range ions. Using these methodologies, we have developed experimental techniques to elucidate the diffusion mechanism of SEs. which will support our future work in improving the performance of SEs and solid-state batteries.

10. Conclusion

The goal of ABC is to establish a platform for collaboration between academia and industry for battery research and development. Combining the advanced characterization, data science, smart laboratory, theoretical calculation with developing battery simulation protocols, ABC will support not only R&D of advanced Li batteries, Li-air batteries, sodium batteries magnesium batteries and all-solid-state batteries but also in Japan but also the innovation of new battery technologies. In addition, this project is being carried out under one roof, which has a major advantage over national projects overseas.

Kei Nishikawa (Team Leader, Center for Advanced Battery Collaboration, Research Center for Energy and Environmental Materials, National Institute for Materials Science)

Kei Nishikawa was born in 1980. He graduated from Graduate School of Energy Science, Kyoto University in September 2006, and earned Doctor of Energy Science, He worked in Tokyo Metropolitan University, Ecole Polytechnique, and JAXA as postdoctoral researchers. He joined NIMS in 2011 and was promoted to principal researcher in 2022. His research interests are electrodeposition of metals and secondary batteries. Hobby: Handball.

Takuya Masuda (Director, Research Center for Energy and Environmental Materials (GREEN), NIMS)

Takuya Masuda was born in 1978. He graduated from Graduate School of Science, Hokkaido University in March 2006, and earned Doctor of Science. He worked at Arizona State University as a postdoctoral fellow in 2006–2007, and at Hokkaido University as an assistant professor in 2007–2010. He joined NIMS in 2010 and was promoted to group leader in 2018 and director in 2024. His research interests center on in situ/*operando* observation of interfacial phenomena. Hobby: golf.

Yibin Xu (Group Leader, Center for Basic Research on Materials, NIMS)

Yibin Xu received her Ph.D. in Engineering in 1994 from the Shanghai Institute of Ceramics, Chinese Academy of Sciences, and a Ph.D. in Information Science in 2007 from Nagoya University. After working at Shanghai Institute of Ceramics, National Industrial Research Institute of Nagoya, and CTI Co., Ltd., she joint National Institute for Materials Science in 2002. Her recent research interests include the construction of materials big data and the machine learning-aided design and optimization of functional inorganic materials.

Yoshitaka Tateyama (Chief Invited Researcher, NIMS/Professor, Institute of Science Tokyo)

Born in 1970. He earned Ph.D from Graduate School of Science, The University of Tokyo in March, 1998. He has then worked in NIMS since 1998, and was a center director for 2023–2024. In 2023, he partially moved to Tokyo Institute of Technology, now Institute of Science Tokyo, as a professor. His research interest is computational materials science on electrochemical devices such as battery.

Shoichi Matsuda (Team Leader, Automated Electrochemical Experiments Team, Center for Green Research on Energy and Environmental Materials, National Institute for Material Science)

Shoichi Matsuda was born in 1987. He studied chemistry in The University of Tokyo and received his PhD in 2015. After that, he joined NIMS as an ICYS (International Center for Young Scientists) researcher and then got a tenured position in 2017.

Shuji Nakanishi (Professor, Graduate School of Engineering Science, The University of Osaka)

Shuji Nakanishi was born in 1973. He received his BS (1997), MS (1999) from Osaka University. Then, he worked as Assistant Professor in Osaka University (2000–2008). He received his PhD degree from Osaka University in 2002. He served as Associate Professor in The University of Tokyo (2008–2015). In 2015, he was promoted to a Professor in The University of Osaka.

Shotaro Hanada (Ph.D student, Graduate School of Engineering Science, The University of Osaka)

He was born in 2000. He graduated from Graduate School of Engineering Science, Osaka University in March 2025. Currently, he is engaged as Ph.D student. His research interests directed to development of secondary batteries. Hobby: Running.

Yoshiharu Mukouyama (Professor, Tokyo Denki University, NIMS Special Researcher)

Yoshiharu Mukouyama was born in 1973. He earned a Ph.D. in Science from Osaka University in 2001. He joined Tokyo Denki University the same year and became a Professor in 2017. In 2022, he was appointed as a Visiting Researcher at NIMS, and in 2025, he served as a Special Researcher at NIMS. His research interests include batteries and electrocatalysts. Hobby: ice hockey.

Toshihiko Mandai (Team Leader, Center for Advanced Battery Collaboration, Research Center for Energy and Environmental Materials, National Institute for Materials Science (NIMS))

Toshihiko Mandai received his PhD from Chiba University in 2012, and worked as a postdoc and an assistant professor at universities. In 2019, he joined NIMS and was promoted to principal researcher in 2024. His research interests include coordination complex for electrolyte applications. Hobby: cooking, swimming.

Naoaki Kuwata (Group Leader, Solid-State Battery Ionics Group, GREEN, NIMS)

He graduated from Graduate School of Chemistry, Hokkaido University in 2000, and earned his Ph.D. Principal Researcher at NIMS in 2019. Concurrently serving as Visiting Professor at the Graduate School of Chemical Science and Engineering, Hokkaido University in 2021. In his present position from 2024. Specialty: electrochemistry and solid state ionics. Hobby: Manga.

Tsuyoshi Ohnishi (Group leader, Battery Interface Control Group, Research Center for Energy and Environmental Materials, National Institute for Materials Science (NIMS))

Tsuyoshi Ohnishi was born in 1971. He graduated from Department of Materials Science and Engineering, Tokyo Institute of Technology in March 1999, and earned Doctor of Engineering. He worked in Lawrence Berkeley National Laboratory and Stanford University as Postdoctoral researcher in 1999–2002. He worked in the University of Tokyo as Research Associate in 2002–2008 and moved to NIMS in 2008. His research interests are thin film synthesis and applications. Hobby: Thin film synthesis.

Kiyoshi Kanamura (Emeritus Professor, Faculty of Urban Environmental Sciences, Tokyo Metropolitan University)

Kiyoshi Kanamura was born in 1957. He graduated from Graduate School of Engineering, Kyoto University in 1982, and earned Doctor of Science, He worked in Kyoto University in 1984–1998 in Electrochemical Energy Conversion System. He moved to Tokyo Metropolitan University as Associate Professor in 1998 and Professor in 2002. He was awarded Takei award from The Electrochemical Society of Japan in 2022. His research interests are battery and fuel cell. Hobby: golf.

Acknowledgment

This work is supported by Japan Science and Technology Agency COI-NEXT program "Center for Advanced Battery Collaboration" Grant Number JPMJPF2016. The authors would like to thank Dr. Iba and Dr. Nagamine for their valuable suggestions and advice.

The APC for the publication of this paper was supported by The Committee of Battery Technology, ECSJ.

CRediT Authorship Contribution Statement

Kei Nishikawa: Investigation (Equal), Writing – original draft (Lead), Writing – review & editing (Lead)
Takuya Masuda: Funding acquisition (Supporting), Investigation (Equal), Project administration (Supporting), Writing – original draft (Equal)
Yibin Xu: Data curation (Lead), Investigation (Equal), Writing – original draft (Equal)
Yoshitaka Tateyama: Investigation (Equal), Writing – original draft (Equal)
Shoichi Matsuda: Investigation (Equal), Writing – original draft (Equal)
Shuji Nakanishi: Investigation (Equal), Writing – original draft (Equal)
Shotaro Hanada: Investigation (Equal), Writing – original draft (Supporting)
Yoshiharu Mukouyama: Investigation (Equal), Writing – original draft (Supporting)
Toshihiko Mandai: Investigation (Equal), Writing – original draft (Equal)
Naoaki Kuwata: Investigation (Equal), Writing – original draft (Equal)
Tsuyoshi Ohnishi: Investigation (Equal)
Kiyoshi Kanamura: Conceptualization (Lead), Funding acquisition (Lead), Project administration (Lead), Writing – review & editing (Equal)

Conflict of Interest

The authors declare no conflict of interest in the manuscript.

Funding

Japan Science and Technology Agency: JPMJPF2016

References

1. K. Kanamura, *Electrochemical Science for a Sustainable Society, A Tribute to John O'M. Bockris* (Ed. K. Uosaki), Springer, New York, p. 175 (2017).
2. https://www.un.org/sustainabledevelopment/development-agenda-retired/ referred on 10 February, 2025.
3. https://www.anl.gov/li-bridge referred on 13 March, 2025.
4. https://www.ipcei-batteries.eu/about-ipcei referred on 13 March, 2025.
5. https://www.faraday.ac.uk/ referred on 13 March, 2025.
6. https://www.jst.go.jp/gtex/en/field/storage.html referred on 13 March, 2025.
7. J. Newman and K. E. Thomas-Alyea, *Electrochemical Systems* (3rd ed.), John Wiley & Sons, Inc. (2003).
8. J. C. Forman, S. J. Moura, J. L. Stein, and H. K. Fathy, *J. Power Sources*, **210**, 263 (2012).
9. T.-S. Dao, C. P. Vyasarayani, and J. McPhee, *J. Power Sources*, **198**, 329 (2012).
10. G. Inoue and M. Kawase, *J. Power Sources*, **342**, 476 (2017).
11. R. P. Putra, K. Matsushita, T. Ohnishi, and T. Masuda, *J. Phys. Chem. Lett.*, **15**, 490 (2024).
12. T. Iwama, T. Ohnishi, and T. Masuda, *Electrochemistry*, **91**, 117005 (2023).
13. Y. Xu, Y.-J. Wu, H. Li, L. Fang, S. Hayashi, A. Oishi, N. Shimizu, R. Caputo, and P. Villars, *Sci. Technol. Adv. Mater.*, **25**, 2403328 (2024).
14. S.-H. Jang, Y. Tateyama, and R. Jalem, *Adv. Funct. Mater.*, **32**, 2206036 (2022).
15. S.-H. Jang, R. Jalem, and Y. Tateyama, *J. Phys. Chem. A*, **127**, 5734 (2023).
16. S.-H. Jang, R. Jalem, and Y. Tateyama, *J. Mater. Chem. A*, **12**, 20879 (2024).
17. K. Kawai, S.-H. Jang, Y. Igarashi, K. Yazawa, K. Gotoh, J. Kikkawa, A. Yamada, Y. Tateyama, and M. Okubo, *Angew. Chem. Int. Ed.*, **64**, e202410971 (2025).
18. Y. Tateyama, S.-H. Jang, and R. Jalem, *Denki Kagaku*, **91**, 150 (2023).
19. S. Matsuda, K. Nishioka, and S. Nakanishi, *Sci. Rep.*, **9**, 6211 (2019).
20. S. Matsuda, G. Lambard, and K. Sodeyama, *Cell Rep. Phys. Sci.*, **3**, 100832 (2022).
21. R. Tamura, K. Tsuda, and S. Matsuda, *Sci. Technol. Adv. Mater.: Methods*, **3**, 2232297 (2023).
22. Y. Motoyama, R. Tamura, K. Yoshimi, K. Terayama, T. Ueno, and K. Tsuda, *Comput. Phys. Commun.*, **278**, 108405 (2022).
23. K. Terayama, M. Sumita, R. Tamura, D. T. Payne, M. K. Chahal, S. Ishihara, and K. Tsuda, *Chem. Sci.*, **11**, 5959 (2020).
24. K.-H. Chen, K. N. Wood, E. Kazyak, W. S. Lepage, A. L. Davis, A. J. Sanchez, and N. P. Dasgupta, *J. Mater. Chem. A*, **5**, 11671 (2017).
25. Y. Peng and K. Nishikawa, *Cell Rep. Phys. Sci.*, **6**, 102439 (2025).
26. Y. Peng, R. Tamate, and K. Nishikawa, *ChemElectroChem*, **11**, e202400278 (2024).
27. R. Tamate, Y. Peng, Y. Kamiyama, and K. Nishikawa, *Adv. Mater.*, **35**, 2211679 (2023).
28. C.-H. Shin, H.-Y. Lee, C. Gyan-Barimah, J.-H. Yu, and J.-S. Yu, *Chem. Soc. Rev.*, **52**, 2145 (2023).
29. Z. Lu, A. Schechter, M. Moshkovich, and D. Aurbach, *J. Electroanal. Chem.*, **466**, 203 (1999).
30. T. Mandai, M. Yao, K. Sodeyama, A. Kagatsume, Y. Tateyama, and H. Imai, *J. Phys. Chem. C*, **127**, 10419 (2023).
31. X. Liu, A. Du, Z. Guo, C. Wang, X. Zhou, J. Zhao, F. Sun, S. Dong, and G. Cui, *Adv. Mater.*, **34**, 2201886 (2022).
32. T. Mandai and H. Somekawa, *Batter. Supercaps*, **5**, e202200153 (2022).
33. T. Mandai, U. Tanaka, and M. Watanabe, *Energy Storage Mater.*, **67**, 103302 (2024).
34. S. S. Kim and K. A. See, *ACS Appl. Mater. Interfaces*, **13**, 671 (2021).
35. T. Mandai and M. Watanabe, *J. Mater. Chem. A*, **11**, 9755 (2023).
36. K. M. Abraham, *ACS Energy Lett.*, **5**, 3544 (2020).
37. A. Rudola, A. J. R. Rennie, R. Heap, S. S. Meysami, A. Lowbridge, F. Mazzali, R. Sayers, C. J. Wright, and J. Barker, *J. Mater. Chem. A*, **9**, 8279 (2021).
38. Q. Zhou, Y. Li, F. Tang, K. Li, X. Rong, Y. Lu, L. Chen, and Y.-S. Hu, *Chin. Phys. Lett.*, **38**, 076501 (2021).
39. T. Mandai, U. Tanaka, and S. Kimura, *Adv. Energy Sustain. Res.*, **5**, 2400059 (2024).
40. P. Albertus, G. Girishkumar, B. McCloskey, R. S. Sánchez-Carrera, B. Kozinsky, J. Christensen, and A. C. Luntz, *J. Electrochem. Soc.*, **158**, A343 (2011).
41. U. Sahapatsombut, H. Cheng, and K. Scott, *J. Power Sources*, **227**, 243 (2013).
42. Y. Mukouyama, S. Hanada, T. Goto, and S. Nakanishi, *J. Phys. Chem. C*, **127**, 10459 (2023).
43. S. Hanada, S. Nakanishi, and Y. Mukouyama, *Electrochem. Commun.*, **165**, 107752 (2024).
44. M. Mehta, V. Bevara, and P. Andrei, *J. Power Sources*, **286**, 299 (2015).
45. Y. Inaguma, *Next Generation Batteries: Realization of High Energy Density Rechargeable Batteries* (Ed. K. Kanamura), Springer Singapore, Singapore, p. 187 (2021).
46. G. Hasegawa, N. Kuwata, K. Hashi, Y. Tanaka, and K. Takada, *Chem. Mater.*, **35**, 3815 (2023).
47. G. Hasegawa, N. Kuwata, T. Ohnishi, and K. Takada, *J. Mater. Chem. A*, **12**, 731 (2024).
48. Y. Fujiwara, K. Hoshikawa, and K. Kohama, *J. Cryst. Growth*, **433**, 48 (2016).
49. X. Hu, C. A. J. Fisher, S. Kobayashi, Y. H. Ikuhara, Y. Fujiwara, K. Hoshikawa, H. Moriwake, K. Kohama, H. Iba, and Y. Ikuhara, *Acta Mater.*, **156**, 379 (2018).
50. Z. Yang, R. E. Ward, N. Tanibata, H. Takeda, M. Nakayama, and R. Kobayashi, *Solid State Ionics*, **366–367**, 115662 (2021).
51. N. Kuwata, G. Hasegawa, K. Hashi, Y. Fujiwara, and H. Iba, *65th Battery Symposium in Japan*, Abstr., p. 1A19 (2024).