Computational and Data Science for Next-generation Batteries with Safety and High Performance

To address the threat of climate change, Japan and other countries have embraced the goal of becoming carbon neutral and reducing their greenhouse gas emissions to zero by 2050. High-performance rechargeable batteries are expected to provide a decisive advantage in achieving this goal. Solid-state batteries in particular are expected to be widely used in electric vehicles, and the race to develop them is taking off around the world. To use the powers of computer science and data science to help develop solid-state batteries, Yoshitaka Tateyama's research group has been making steady progress in the quest to understand the underlying mechanisms of suitable materials for these new batteries. Their “Fugaku Battery & Fuel Cell”*1 project falls under the “Program for Promoting Research on the Supercomputer Fugaku” rubric.

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*1 Research project name:
“Computational and Data Science Study for ET Revolution by Development of Next-Generation Battery and Fuel Cell” (Principal Investigator: Yoshitaka Tateyama)

Solid-state Batteries to Make Electric Vehicles Mainstream

Electric vehicles themselves do not emit carbon dioxide while running. So, if the electricity used to charge them is obtained from renewable energy sources, the total carbon dioxide emissions should be drastically less than with gasoline-powered vehicles. One of the key factors in the adoption of electric vehicles is developing safe, efficient batteries. At present, electric vehicles are equipped with lithium-ion batteries similar to those used in smartphones and other portable devices. But the liquid electrolytes used in these lithium-ion batteries are flammable and require strict safety measures. To mitigate these problems, both academic and industrial researchers are working on battery designs

with solid electrolytes instead of the liquid ones.

“Solid-state batteries are not only safer; they can also be more efficient. With energy and power densities greater than those of conventional lithium-ion batteries, solid-state batteries are expected to extend the cruising range of electric vehicles, improve acceleration, and shorten recharging times,” says Tateyama.

But developing solid-state batteries is not an easy task. The biggest challenge is the need for a solid electrolyte material that conducts lithium ions well. In both conventional and solid-state batteries, lithium ions travel between the positive and negative electrodes during charging and discharging (Fig.1). The organic liquid electrolyte in a conventional battery allows lithium ions to flow through freely. But the solid electrolyte particles in a solid-state

battery impede the lithium ions' ability to pass through, limiting the battery's charge-discharge performance.

Researchers are therefore investigating a range of candidate materials to find a solid electrolyte that will allow lithium ions to pass through faster. Physically testing the immense number of candidate materials, however, would require enormous time and effort. That's where Tateyama and his team come in. They are using supercomputers like Fugaku to search for solid electrolyte material candidates.

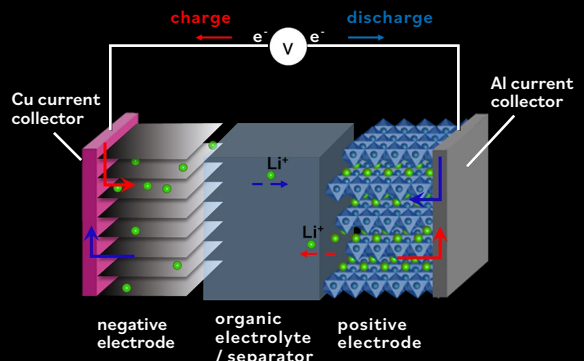
Using Machine Learning to Search for Promising Solid Electrolytes

To efficiently search for materials that can pass lithium ions very quickly, Randy Jalem of Tateyama's group devised a new approach to the calculations (Fig. 2). First, based on structures found in a database of known materials, tens of thousands of hitherto unknown materials are generated on a computer. Next, using clues such as the shape of the pathway for lithium ions and the types of elements around the pathway, promising candidates are selected from among the many materials generated. Then, machine learning with

Fig. 1 Lithium-ion Battery Designs

In solid-state batteries, the conventional organic liquid electrolyte (a) between the positive and negative electrodes is replaced with a solid electrolyte material (b). Both designs operate the same way, with lithium ions transfering back and forth between the positive and negative electrodes when charging or discharging the battery.

(a) Conventional Li-ion Battery



(b) Solid-state Battery

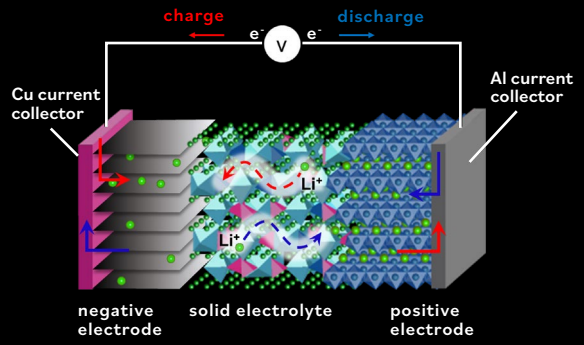


Fig. 2 **The computational process for discovering solid electrolyte candidates and its inventor, Randy Jalem.**

The Steps to Discovery:
(1) Rearrange the atoms of known crystal structures to generate tens of thousands of solid electrolyte candidates.
(2) Narrow down the candidates based their lithium ion pathways and the types of elements around the pathways.
(3) Use Bayesian optimization to select the remaining candidates with higher lithium ion conductivity.
The remaining candidate materials discovered through this process were actually synthesized in the laboratories of the Tokyo Institute of Technology.

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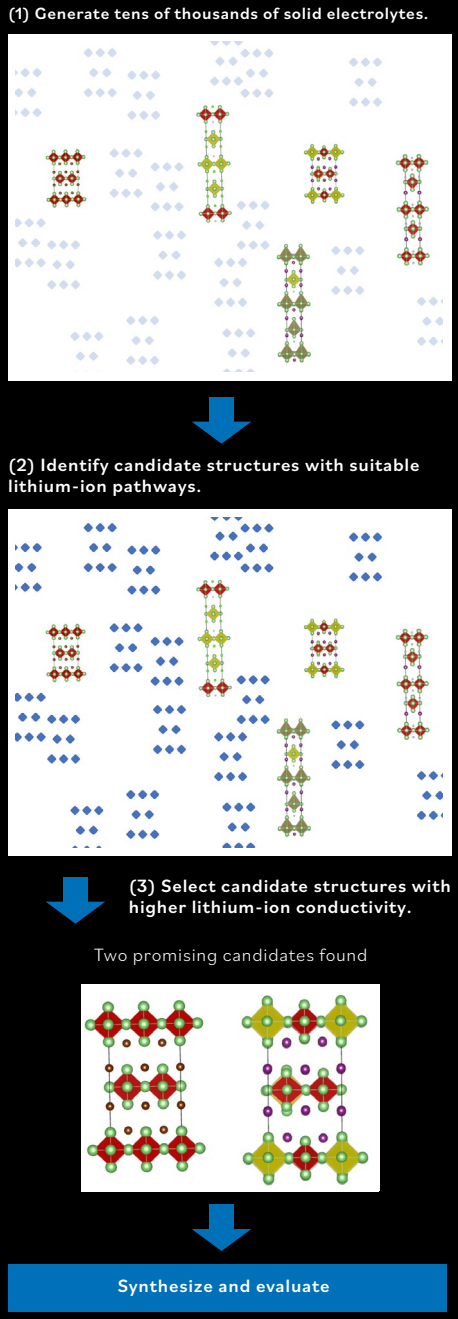


Bayesian optimization is used to select the materials that should allow the lithium ions to pass through the fastest.

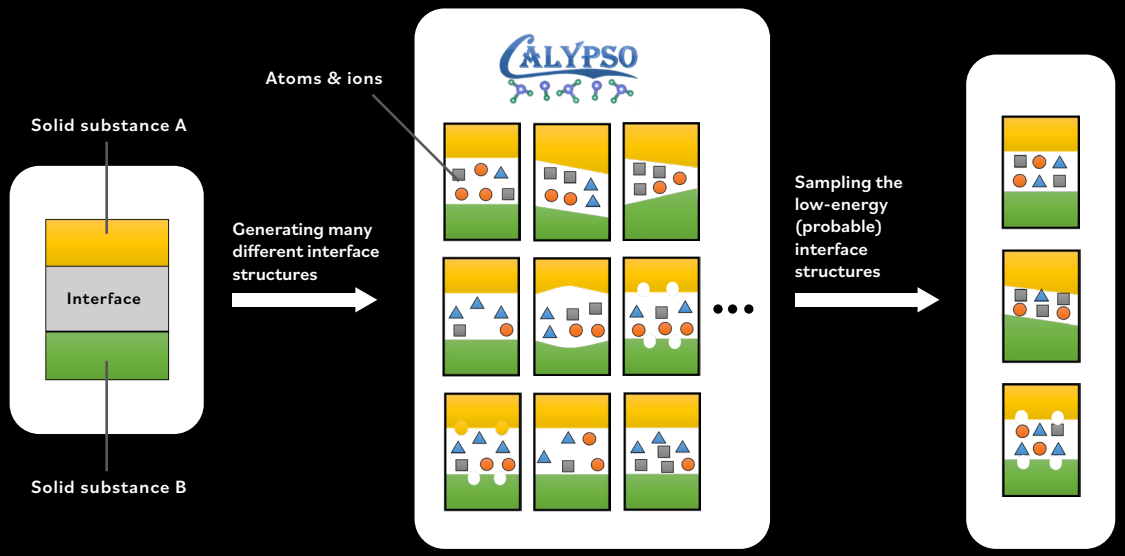
Jalem says, “With this approach, I found two promising candidates. Those two will be synthesized in a lab and tested for their performance.” The results show that large-scale computations can speed up research by selecting candidate materials logically.

Jalem enthuses, “When we were using the K computer, the materials search calculations took weeks, because we have to iterate through loop statements so many times. With Fugaku's huge number of high-performance nodes, we can run the calculations much faster. So, we could run a search with many more candidate materials.”

Fig. 3

Heterogeneous Interface CALYPSO Method

First, in the interface section between the two different types of solids (A and B), various arrangements of atoms and ions from these two solids are used to generate tens of thousands of different interface structures. While the energy of each candidate interface structure is being calculated, the positions of its atoms and ions are optimized. From these, the structures with the lower energies, meaning more probable structures, are selected.



Massive Searches for Interfaces to Maximize Battery Performance

Another major issue in the development of solid-state batteries is controlling the interface between the solid electrolyte and each electrode material. Researching and developing the coating layers is particularly important. “Actually, solid-state battery performance tends to degrade over time with repeated charging and discharging. We know that a thin coating of a different material applied to the surface of the electrode material can effectively prevent this degradation, but we don't exactly know how that works. So, the strategy has not been quite established which material is best to use or how thick that coating should be,” explains Tateyama. To make the best use of lab testing resources, he is using computer science to better understand how lithium ions and electrons travel through the materials that comprise the electrodes, the coating layer, and the solid electrolyte.

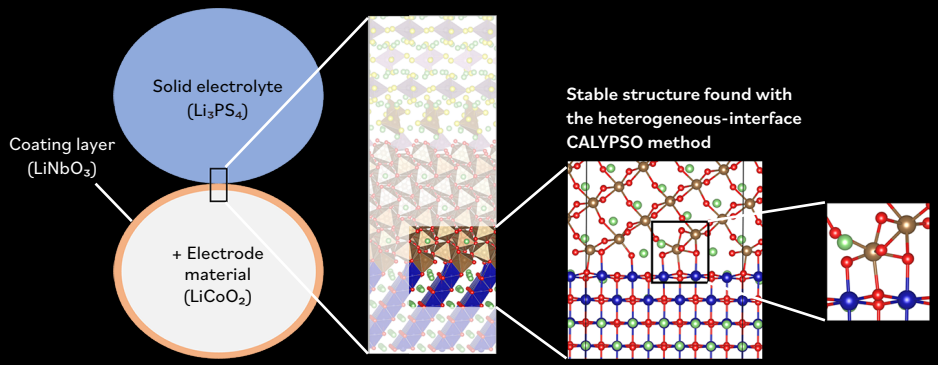
Tateyama's group developed an AI-based CALYPSO (Crystal structure AnaLYsis by Particle Swarm Optimization, Fig. 3) method to predict the structures at the “heterogeneous interface” where the atoms and ions of two different materials adjoin in structures with varying arrangements. The CALYPSO method generates tens of thousands of these atomic interface structures, and efficiently calculates the energy of each to predict which structures are more stable and therefore more likely to occur. Using this method, Tateyama was able to find stable structures for both the electrode-to-coating interface and the

coating-to-solid electrolyte interface (Fig. 4). This was Tateyama and his team's game changing work with computer simulations to automate searches for stable interface structures. The computing power of Fugaku enabled them to search an enormous number of these structures. The team is now studying the behavior of the lithium ions and electrons in the structures they discovered to better understand how the coating layer suppresses degradation.

“Controlling the interface as well as the coating is key to developing solid-state batteries. So, we'd like to help formulate guidelines for interface design by showing what's actually happening at the interface. To calculate the energy of the interface structure, we use a precise method called “first-principles calculations”,[*2] which requires a huge amount of calculating for even a single structure. But with Fugaku, we can compute the energies of many structures at once in parallel, so our research can move quickly,” says Tateyama. The combination

*2 First-principles calculation: A calculation that follows the fundamental laws of physics without the use of empirical parameters. In this case, it refers to calculating a property such as the energy of a material according to quantum mechanics.

Fig. 4 Coating Layer Research



The coating layer is sandwiched between an electrode material on one side (in this example, the positive electrode material), and the solid electrolyte material on the other. The particles comprising the electrode material and the solid electrolyte material are a few micrometers in size, but the coating layer is much thinner, only a few nanometers. The heterogeneous interface CALYPSO method was used to find possible interface materials with a stable structure on both the electrode-facing side and the solid electrolyte-facing side. The conductivity of the structures in those candidate interface materials was then studied to determine how easily lithium ions and electrons could pass through them. The right side of the figure shows an example of the calculated interface structures between the solid electrolyte material and the coating layer. In practice, the interfaces between many different materials were studied.

of laboratory testing and Fugaku's calculations will greatly accelerate the research and development of solid-state batteries. Computer science and data science will help create high-performance solid-state batteries, which in turn will help us achieve carbon neutrality as electric vehicles become the norm.

About the

Researcher

Yoshitaka Tateyama decided to become a scientist in elementary school upon reading the biography of physicist Lev Landau and being captivated by his forthright attitude toward science. Tateyama says his motto concerning work is “to do groundbreaking research”. To that end, he values actively communicating with people from all walks of life. There is a lot to be learned from conversations, and sometimes the seeds of future research can be found through interactions with experimenters. A nice drink or an enjoyable meal can grease the wheels of communication. But for now, the COVID-19 pandemic has put such opportunities on hold.

Associated Research Projects

• “Computational and Data Science Study for ET Revolution by Development of Next-Generation Battery and Fuel Cell” (hp200131/hp210173)
Principal Investigator: Yoshitaka Tateyama, National Institute for Materials Science

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