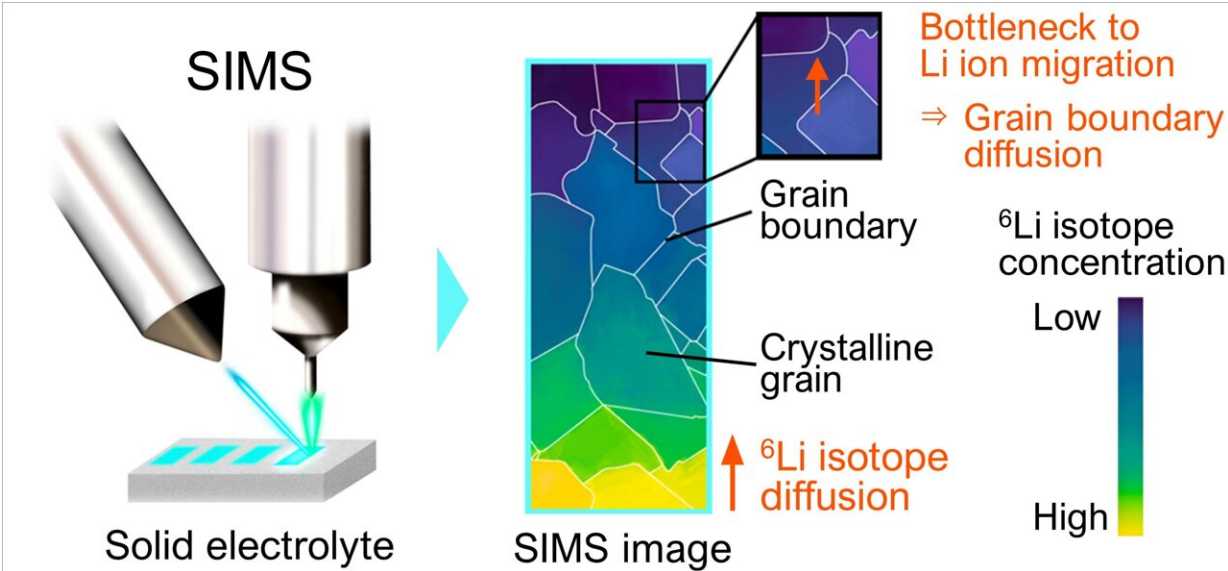


Imaging grain boundaries that impede lithium-ion migration in solid-state batteries

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SIMS imaging of lithium-ion migration within a solid electrolyte specimen. Grain boundaries resistant to ionic diffusion have created an uneven ^6Li distribution. Quantitative analysis found that ionic diffusion across these boundaries was 10,000 times slower than diffusion through the grains. Credit: Naoaki Kuwata National Institute for Materials Science

A NIMS research team has developed a new technique to image grain boundaries obstructing lithium-ion migration in solid-state batteries—a promising type of next-generation battery.

Solid-state batteries—next-generation rechargeable batteries—are intended to be safer and have higher energy densities than conventional lithium-ion batteries by replacing liquid organic electrolytes with [solid electrolytes](#). A major issue in current solid-state battery R&D is the obstruction of lithium-ion migration at the interfaces between active materials and solid electrolytes and at the grain boundaries within solid electrolytes.

These obstructions lower charge/discharge rates and reduce [energy density](#) in batteries. A solid electrolyte is composed of crystalline grains and the boundaries between them. Existing ionic conductivity evaluation methods had only been able to measure average ionic conductivity across a solid electrolyte and were unable to quantify ionic conductivity at individual grain boundaries and identify boundaries restricting ionic migration.

This research team succeeded in imaging and quantifying ionic migration/diffusion at individual grain boundaries within a solid electrolyte using [secondary ion mass spectrometry](#) (SIMS). SIMS enables the imaging of chemical element distribution across a solid electrolyte specimen by sputtering the surface of the specimen with a focused primary ion beam and collecting and analyzing ejected secondary ions.

The team first replaced a portion of a stable lithium isotope, ${}^7\text{Li}$ (mass number: 7, natural abundance: 92%), constituting an electrolyte specimen with another lithium isotope, ${}^6\text{Li}$ (mass number: 6, natural abundance: 8%), at the edge of the specimen using an isotope exchange technique.

The team then observed the diffusion of ${}^6\text{Li}$ within the specimen using SIMS. Because it was impossible to image and quantify the distribution of fast-diffusing ${}^6\text{Li}$ using conventional SIMS, the team significantly slowed ${}^6\text{Li}$ diffusion by cooling the specimen (i.e., cryo-SIMS), enabling

the team to precisely measure the ^6Li distribution and identify grain boundaries acting as bottlenecks to ionic [migration](#).

The cryo-SIMS technique can be used to directly observe lithium-ion [diffusion](#), identify interfaces/[grain boundaries](#) acting as bottlenecks among the many interfaces/boundaries existing in a solid-state battery, and determine the causes of these obstructions. This approach is expected to contribute to the development of higher-performance [solid-state batteries](#).

The work is [published](#) in the *Journal of Materials Chemistry A*.

More information: Gen Hasegawa et al, Visualization and evaluation of lithium diffusion at grain boundaries in $\text{Li}_{0.29}\text{La}_{0.57}\text{TiO}_3$ solid electrolytes using secondary ion mass spectrometry, *Journal of Materials Chemistry A* (2023). [DOI: 10.1039/D3TA05012B](https://doi.org/10.1039/D3TA05012B)

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