

SOLID-STATE

HYPE, HOPES, AND HURDLES

As conventional lithium-ion battery technology approaches its theoretical limits, researchers are studying alternative architectures with solid electrolytes.

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BATTERIES

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SOLID-STATE BATTERIES

The cornerstone technology that powers smartphones, electric vehicles (EVs), and various other modern devices is the lithium-ion battery (LIB). The widespread adoption of LIBs reflects their efficiency, scalability, and versatility. In its *World Energy Outlook 2022* report, the International Energy Agency identified LIBs as the fastest growing storage technology in the world.

To reach global net-zero carbon emissions by 2050, the report says that annual battery demand for EVs would need to grow from 0.16 TWh in 2020 to 14 TWh by 2050. Meeting that demand is critical to decarbonizing transportation and mitigating climate change. Major automakers have committed billions of dollars to embrace EVs and meet net-zero emissions goals. The switch away from internal combustion engines would comply with policies such as the European Union's plan to ban gasoline-powered vehicle sales by 2035.

Despite the widespread use and undeniable advantages of LIBs, they have several limitations. Safety is a perennial concern because short circuits can lead to fires. Conventional LIB technologies, moreover, are approaching theoretical energy limits. Such constraints underscore the need for innovations that meet the demands of automakers and deliver higher energy densities and faster charging speeds than today's LIBs.

Some automakers have EV models with battery packs that can provide roughly 600 km of range on a single charge.¹ But car buyers' range anxiety—the fear of running out of battery power while driving—is still one of the most significant barriers to EV adoption. Further increasing the driving range may not be feasible with conventional LIB technology. Figure 1 shows that approximately 20 kWh of energy is needed to drive an EV 100 km. Given that conventional LIBs with graphite anodes have energy densities of about 250 Wh/kg and that the ideal battery weight is below 500 kg, the farthest an EV can travel with an LIB is around 600 km.

Another drawback of EVs is the time it takes to recharge their batteries. Recently, some eye-catching claims have promised that certain vehicle models with new battery designs could recharge in about 10 minutes.² Such capability, however, comes with an important caveat: A full charge to 100% would require about a megawatt of power (see figure 1), which is comparable to the output of a small power plant.

Researchers are pursuing various battery architectures to replace or improve LIBs. Some work, for example, has focused on replacing lithium's graphite anode with lithium metal or a lithium-silicon mixture. A compelling next-generation solution for delivering high energy and high-power density with improved safety is the solid-state battery (SSB).³ The technology has drawn interest from established companies, such as

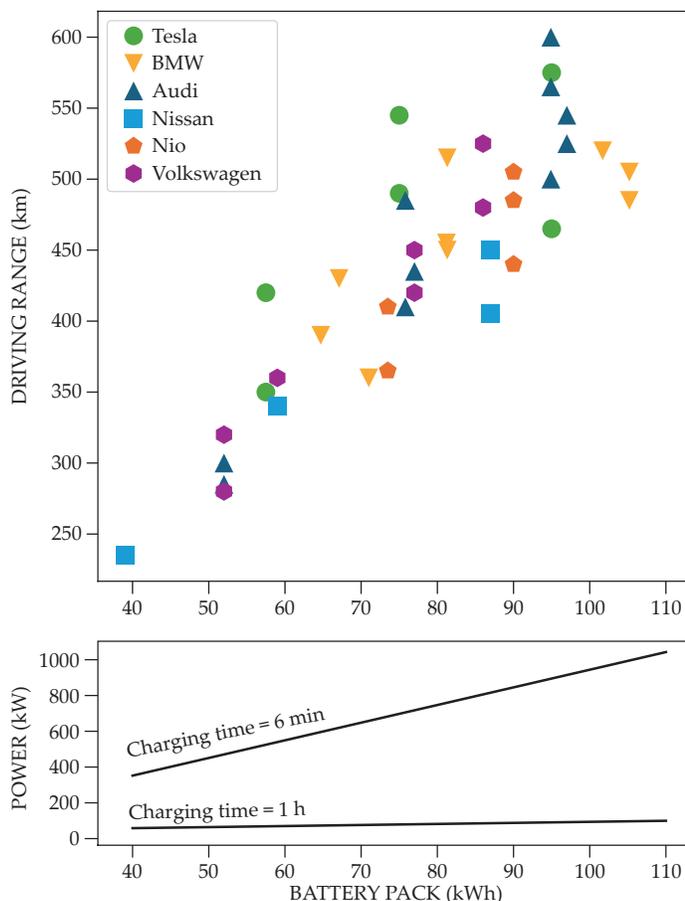


FIGURE 1. ELECTRIC VEHICLES (EVs) can currently drive 600 km or less (**top**) on a single charge.² To extend the vehicles' driving range with conventional lithium-ion technology, a battery pack would need to provide more than 110 kWh of energy, which would make it prohibitively heavy. Many EVs take at least an hour to charge (**bottom**) but require no more than about 100 kW of power. To charge a vehicle in just a few minutes requires 4–10 times as much power. (Figure adapted from ref. 1.)

Toyota and Samsung, and has spurred a wave of innovations among startups, including QuantumScape and Solid Power. They all have recently developed SSB prototypes that demonstrate encouraging performance metrics (see box 1 for metric definitions), and Toyota has claimed that SSBs could be commercialized as early as 2027. The developments suggest that SSB technology could be a promising alternative to LIBs.⁴

Unpacking battery basics

A battery consists of three main components: a cathode, an anode, and an electrolyte. The cathode, or positive electrode, is typically made from layered transition-metal oxides that contain nickel, manganese, and cobalt. Lithium ions slip in and out of the oxides' crystal structure during charging and discharging.

On the opposite side of the cathode is the anode, or negative electrode, which is commonly made of graphite with a honeycomb lattice structure. It has tightly spaced layers that can host lithium ions.

Between the two electrodes is the electrolyte, which allows lithium ions, but not electrons, to shuttle to and from the electrodes. In conventional LIBs, electrolytes consist of organic solvents such as ethylene carbonate and dimethyl carbonate. Those materials present a safety risk because of their high flammability.

SSBs have a similar architecture, but a solid material replaces the liquid electrolyte, as shown in figure 2. Solid electrolytes are nonflammable; allow for battery components to be more tightly packed, which can increase energy density; and permit the anode to be made from lithium metal, which has a higher energy density than graphite. To realize all the benefits, however, solid electrolytes must enable lithium ions to move at room temperature at least as fast as they do in liquid electrolytes.⁵

Quest for new solid electrolytes

The conventional thinking is that ions diffuse through liquid electrolytes faster than they do through solid ones. Yet lithium ions seem to defy that expectation in certain solid materials, known as superionic conductors.⁶ Inelastic neutron scattering studies have revealed that the energy of some crystal-lattice vibrations in those materials can be unusually low, which is associated with the high ionic mobility and liquid-like behavior of the mobile-ion sublattice.

Ion mobility can also be affected by crystal structure. As lithium ions diffuse through the electrolyte, they eventually encounter bottlenecks, which are regions where the diffusion pathway narrows and creates an energetic barrier to ion transport. To minimize the barrier, researchers have designed solid-electrolyte materials that have a large diffusion pathway. In addition, they can encourage ion mobility by minimizing the crystal structure's change in the coordination environment, which is a measure of the number of atoms in the vicinity of the mobile ions.

In recent decades, researchers have identified and studied many solid-state superionic materials. Figure 3 shows a timeline of the discoveries of three families of ceramic lithium-ion conductors. A breakthrough came in 2011 with the discovery of lithium germanium thiophosphate (LGPS).⁷ It has a room-temperature ionic conductivity of 12 mS/cm—many liquid electrolytes' conductivity is on the order of 10 mS/cm.

Since LGPS's discovery, even more conductive materials have emerged. Earlier this year, researchers made a lithium-strontium-antimony compound with an ionic conductivity of 42 mS/cm, which is higher than that of any other lithium-ion conductor.⁸

So, considering their measured conductivities, are solid electrolytes suitable alternatives to liquid electrolytes? The answer is complicated. In conventional LIBs, the liquid

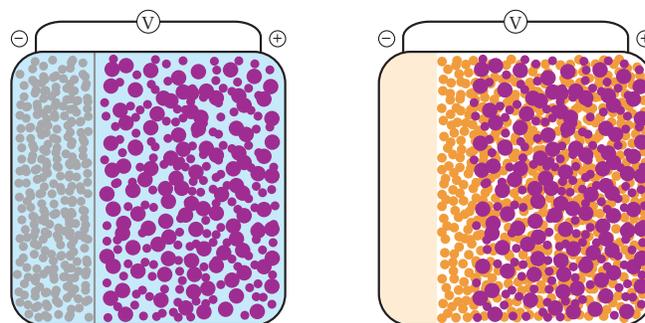


FIGURE 2. BATTERY SCHEMATICS. A conventional lithium-ion battery (LIB; **left**) consists of a graphite anode (gray particles), transition-metal oxide cathode (purple particles), and liquid electrolyte (blue background). During the discharge phase, positively charged lithium ions move through the electrolyte from the anode to the cathode while the electrons flow through the external circuit to power a device. During the charging phase, the sequence is reversed. A safer alternative to a liquid electrolyte is a solid-state one. By using a solid electrolyte (orange particles) and replacing the graphite anode with a lithium-metal one (light yellow), a solid-state battery (**right**) can provide approximately 40% higher energy density per weight than an LIB and 70% higher energy density per unit volume. (Figure adapted from ref. 16.)

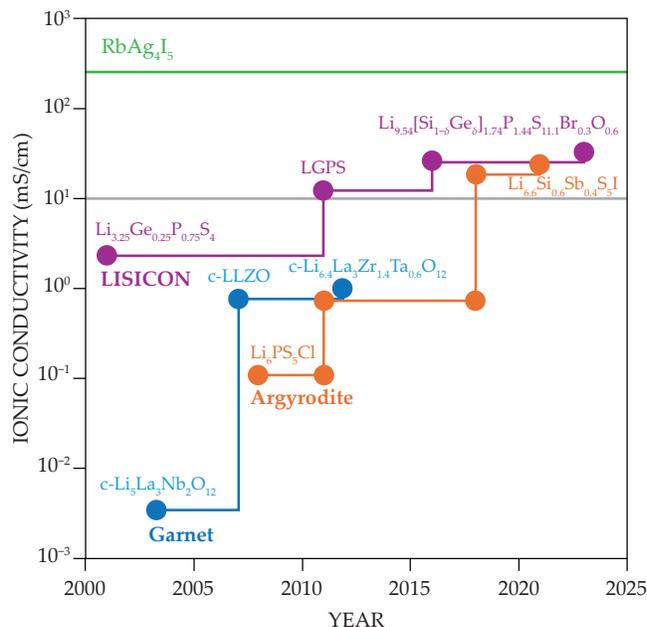


FIGURE 3. THREE FAMILIES of lithium-based materials—argpyrodite, garnet, and LISICON, or lithium superionic conductor—that could be used as electrolytes in solid-state batteries have been discovered over the past century. The gray line shows the ionic conductivity of lithium in the liquid electrolytes that are used in conventional lithium-ion batteries; the green line indicates the conductivity of rubidium silver iodide, which was discovered in 1967 and has the highest known ionic conductivity of any material. LGPS is lithium germanium thiophosphate, and LLZO is lithium lanthanum zirconium oxide.

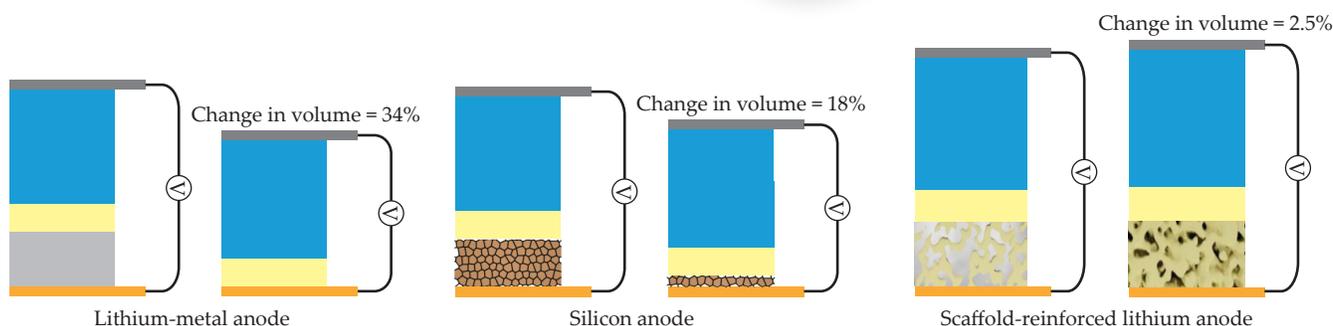


FIGURE 4. VOLUME CHANGES that occur during the charging and discharging of solid-state batteries (SSBs) degrade battery performance and reliability. The three types of SSBs shown here have the same cathode (blue) and solid electrolyte (yellow), but they have different anodes: **(left)** lithium metal, **(middle)** silicon, and **(right)** scaffold-reinforced lithium. The calculations assume that the cathode is made of entirely active material, the scaffold has a porosity of 70%, and the amount of charge per unit area of the batteries is 8 mAh/cm².

electrolyte can percolate through the porous cathode, which enables efficient ion transport and the full utilization of the cathode active material, the compound that produces the battery's electrical energy through chemical reactions. Compared with the electrodes in conventional LIBs, the ones in SSBs have higher tortuosity—a measure of the complexity of the ion's path—and other resistive heterogeneities, such as grain boundaries, which lead to lower ionic conductivities in real-world applications.

Those factors can significantly hinder the formation of a robust ion-conduction network, especially when the fraction of solid electrolyte in the electrode is low. To ensure effective ion transport, researchers must meticulously control the microstructures of both the solid electrolyte and the cathode-particle mixture.⁵ If SSBs are to compete with LIBs in terms of performance metrics such as areal loading and current density, they need to have an effective ionic conductivity—which takes tortuosity into account—of at least 10 mS/cm.

Although a few solid electrolytes, such as lithium argyrodites and LGPS derivatives, meet the ionic-conductivity benchmark, solid electrolytes fall short in other performance metrics, such as the electrochemical stability of the high-voltage cathodes and lithium-metal anodes. The search, therefore, continues for new lithium superionic conductors with high lithium-ion conductivity, chemical stability, and optimized solid-electrolyte composites that balance ionic and electronic conductivity, stability, and mechanical integrity.⁵

Energy density of solid-state batteries

Although solid electrolytes can significantly boost a battery's energy density by minimizing the battery's volume, the greatest gains come from replacing conventional graphite anodes with higher-capacity, low-electric-potential alternatives. Some candidates, such as lithium metal and silicon-based materials, offer dramatically higher capacities than graphite (see box 2). But they also introduce substantial challenges.

One major issue is the pronounced volume changes, plotted in figure 4, that the candidate materials undergo during charging and discharging. Lithium-metal anodes, for example,

expand and contract by 15–30%, whereas cathodes typically exhibit volume changes of 1–5%. Volume change creates significant mechanical strains at the interfaces between the active materials and the solid electrolyte. Those strains can lead to a loss of contact between the electrolyte and the cathode and to the cathode fracturing and fatiguing. Such failures can cascade to further issues, including microcracking, dendrite growth, and electrical shorting, all of which compromise the battery's performance and lifetime.

A promising approach to mitigating volume change of lithium-metal anodes involves the use of scaffolds. They provide a rigid framework for lithium ions to attach to the anode and strip evenly from it. The risk of dendrites plummets, and battery performance and reliability improves. Various startup companies are developing scaffold designs that enable stable lithium uptake and release.

Researchers have demonstrated continuous lithium-metal cycling at extremely high current density without dendrite formation in a symmetric battery cell, in which both the cathode and anode are made of a porous LLZO (lithium lanthanum zirconium oxide) scaffold that's separated by a dense LLZO layer. But the scaffold doesn't lead to much higher energy density compared with that of the scaffold-free battery. The scaffold is an inactive material that takes up precious space in the battery that would otherwise be used for materials that increase the battery's capacity.⁹

The problems of contact loss and particle fracture created by volume change could be alleviated by zero-strain cathode materials. They experience minimal structural changes during lithium insertion and removal and, in theory, could alleviate contact loss and particle fracture. Some researchers are encouraged by approaches to control volume change, but more work is needed to evaluate whether zero-strain cathode materials and novel battery architectures are viable pathways toward high-energy density SSBs.

An alternative to a lithium-metal anode is one made of silicon. It offers exceptional theoretical specific capacity—about 10 times as much as that of graphite—but during lithiation, silicon also undergoes significant volume expansion, which leads to

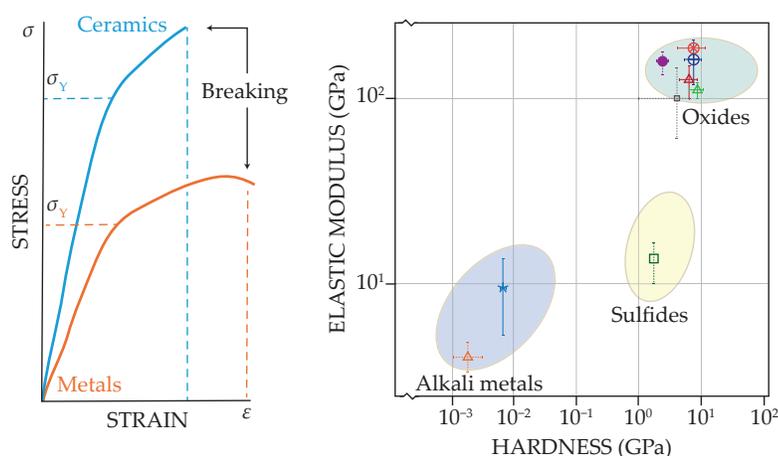


FIGURE 5. THE CONSTITUENT MATERIALS in solid-state batteries (SSBs) respond differently to applied pressure. **(left)** The horizontal lines correspond to the stress level in ceramics and metals as they enter the plastic regime, and the vertical lines correspond to each material's breaking point. The differences affect how readily the materials can stay in contact with one another and thus the battery's operation and reliability. **(right)** When choosing a solid electrolyte, researchers must balance a material's mechanical trade-offs. Oxide-based materials have a high elastic modulus and fracture toughness, which help them resist crack propagation. But those properties make it harder for the electrolyte to remain in contact with the alkali-metal cathode. More ductile materials, such as sulfide-based argyrodites, can deform plastically during electrode volume changes, which helps the electrolyte to maintain contact with the battery's active materials.

mechanical stress on the anode and other parts of the battery and to capacity fading (see figure 4). Companies such as Sila Nanotechnologies are advancing commercial solutions by introducing engineered silicon particles as partial or full replacements for graphite. Silicon-enhanced anodes reportedly are 20% of the weight of graphite and are 50% of its volume.¹⁰

Mechanical properties of solid electrolytes

The performance and durability of SSBs depend on the interfaces between ceramic particles, cathode active materials, and the lithium-metal anode. The interfaces are influenced by the materials' mechanical properties, including brittleness, ductility, and plasticity. Brittle materials break easily, ductile materials deform elastically, and plastic materials deform readily without hardening. To classify a material, researchers measure its stress-strain curve, which describes the material's deformation under stress. Some example curves are shown in figure 5.

Oxide-based electrolytes such as LLZO, for example, exhibit higher elastic moduli, hardness, and fracture toughness than do sulfide-based electrolytes. That means that the ceramic LLZO struggles to maintain contact with the cathode particles during battery cycling. Softer materials such as argyrodites deform plastically during volume changes, which helps the active materials maintain better contact with the electrolyte. Ceramic and glass electrolytes, however, often suffer from low fracture toughness, and their pairing with lithium metal in high-rate applications is limited.

The use of a lithium-metal anode in SSBs is hindered by filament growth and fractures caused by stress gradients and defects. In addition, battery cycling at high charge rates for long lifetimes remains challenging. Deformable solid electrolytes that ensure continuous contact during operation are preferred because they prevent mechanical failures, including crack formation, crack propagation, and filament growth.¹¹ Researchers have also proposed pressure control to maintain contact at interfaces, but that approach has been challenging to implement.

Box I. Battery performance metrics

- **Specific energy (Wh/kg):** the amount of energy a battery can store per unit mass, either of the active material or of the entire battery cell.
- **Energy density (Wh/L):** the amount of energy that a battery can store per unit volume.
- **Specific capacity (mAh/g):** the amount of electric charge that a battery can deliver per unit mass of the active material in the electrodes. Specific capacity is related to specific energy: specific energy (Wh/kg) = specific capacity (mAh/g) × voltage (V).
- **Power density (W/L):** the amount of power a battery can deliver per unit volume. It indicates how quickly a battery can be charged or discharged.
- **Areal density or loading (mAh/cm²):** the amount of charge per unit area of the electrode. The higher the areal density, the higher the battery's specific capacity, but such a battery will require higher ionic and electronic conductivity to sustain its high charge and discharge currents.
- **C-rate:** a measure of how quickly a battery is charged or discharged relative to its capacity. Rates of 0.5 C, 1 C, and 2 C mean that the battery is fully charged or discharged in two hours, one hour, and half an hour, respectively.

Box 2. Anode comparison: Graphite and lithium

The theoretical capacity of graphite, the most commonly used anode material in lithium-ion batteries, can be calculated according to the intercalation of lithium into graphite. It proceeds via the equation $\text{Li}^+ + \text{e}^- + \text{C}_6 \rightarrow \text{LiC}_6$. Each unit of graphite (C_6) can accommodate one lithium ion (Li^+). The molar mass of graphite C_6 is $12 \text{ g/mol} \times 6 = 72 \text{ g/mol}$. One mole of lithium corresponds to one mole of electrons, which equals 96485 C/mol (Faraday's constant). Each mole of graphite, therefore, can store 96485 C . Normalized by the molar weight, graphite's specific capacity is

$${}^{96485/72} \text{ C/g} = {}^{96485/72} \times {}^{1000/3600} \text{ mAh/g} = 372 \text{ mAh/g.}$$

With the same steps, the theoretical specific capacity of lithium metal can be calculated. The only difference is that the corresponding reaction is $\text{Li}^+ + \text{e}^- \rightarrow \text{Li}$. Each lithium atom contributes one electron. The molar mass of lithium is 6.94 g/mol . The specific capacity of a lithium-metal anode, therefore, is

$${}^{96485/6.94} \text{ C/g} = {}^{96485/6.94} \times {}^{1000/3600} \text{ mAh/g} = 3860 \text{ mAh/g.}$$

That capacity is about 10 times as large as graphite's capacity.

Exploring alternatives

Much of the focus of this article has been on ceramic electrolytes, but they're not the only option for SSBs. Polymer electrolytes made of materials in the polyethylene oxide family, for example, have been extensively studied and commercialized in certain applications.¹²

The company Blue Solutions has developed SSBs that capitalize on polymer electrolytes' unique advantages, including the ease with which they are processed and their excellent contact with cathode particles, which helps to reduce interfacial resistance. Polymer electrolytes, however, still face limitations. Their low ionic conductivity necessitates elevated operating temperatures—typically around 80°C —for adequate performance.

A persistent challenge for all types of solid electrolytes is their chemical stability when they're paired with advanced electrode materials. As with liquid electrolytes, the wide number of possible next-generation anodes and cathodes creates compatibility issues. LGPS, for example, offers high ionic conductivity but suffers from chemical instability and reacts with cathodes and anodes to form interphases that degrade performance over time. Even LLZO, which is known for its superior chemical stability, faces challenges such as forming reliable interfaces.

An intriguing approach to overcoming chemical instability is the use of dual, or bilayer, electrolyte systems. In those configurations, solid electrolytes are tailored to stabilize their

respective interfaces, and each layer plays a specialized role to enhance the overall electrochemical stability of the battery cell. Although promising, that strategy has its own set of challenges, including maintaining adequate ion transport and chemo-mechanical stability at the layer interfaces.

Looking ahead, advances in AI offer exciting opportunities to accelerate the discovery of new battery materials. By learning from vast datasets of known compounds, AI models can predict promising candidates with chemical stability and tailored ionic conductivity. Landmark efforts such as Google DeepMind's discovery of more than 2 million new crystal structures¹³ and Microsoft Research's development of a generative AI capable of proposing materials with targeted properties¹⁴ mark significant strides toward truly AI-driven materials discovery.

AI models, however, are not yet able to reliably handle structures with compositional disorder or partial occupancies. As a result, the models may incorrectly identify compounds with the same structure but different arrangements of atoms as distinct "new" materials, when, in fact, the compounds all correspond to the same experimentally determined structure with partial occupancy. In addition, AI-based algorithms for automatic identification of new compounds from x-ray diffraction analysis still require further refinement to unambiguously distinguish novel phases.¹⁵

The integration of AI with multiscale modeling and experimental data is paving the way for digital twins—virtual replicas of batteries that can simulate electrochemical behavior in diverse conditions. AI-enhanced digital twins not only enable optimization of battery design and operation but also offer promising strategies for addressing challenges such as maintaining robust electrolyte–electrode interfaces without the need for external pressure. As the field advances, such data-driven approaches may be critical to unlocking the full potential of solid-state batteries.

Sokseiha Muay and Yang Shao-Horn have a research collaboration with Toyota.

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