

Review Article

Advancements in Solid-State Batteries Overcoming Challenges in Energy Density and Safety - Review

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Abstract

Solid-state batteries (SSBs) have emerged as a promising alternative to conventional lithium-ion batteries (LIBs), offering higher energy density, improved safety, and longer cycle life. This review explores recent advancements in SSB technology, focusing on the development of solid electrolytes, electrode materials, and interface engineering. Solid electrolytes, including oxide-based ($\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$), sulfide-based ($\text{Li}_{10}\text{GeP}_2\text{S}_{12}$), and polymer-based (PEO-LiTFSI) materials, are critical to SSB performance. While oxide-based electrolytes provide high ionic conductivity and stability, sulfide-based electrolytes offer ultra-high conductivity but suffer from air sensitivity. Polymer-based electrolytes are flexible and easy to process but exhibit low conductivity at room temperature. Key challenges such as high interfacial resistance, dendrite formation, and volume changes are addressed through strategies like surface modification, composite electrodes, and 3D architectures. Advanced characterization techniques, including in situ transmission electron microscopy (TEM) and X-ray tomography, provide insights into structural and chemical changes during operation. Computational modeling, such as density functional theory (DFT) and molecular dynamics (MD), accelerates material discovery and interface optimization. Despite significant progress, challenges remain in scalability, performance, and safety. Future research should focus on developing scalable fabrication methods, optimizing electrode-electrolyte interfaces, and integrating SSBs with renewable energy systems for grid storage and electric vehicles. SSBs have the potential to revolutionize energy storage, enabling the widespread adoption of renewable energy and reducing greenhouse gas emissions. Continued innovation and collaboration across disciplines will be essential to overcome remaining challenges and unlock the full potential of SSBs.

Keywords

Solid-State Batteries (SSBs), Solid Electrolytes, Interfacial Resistance, Energy Density, Renewable Energy Integration

1. Introduction

The global transition toward renewable energy and electrification has intensified the demand for advanced energy storage systems [1]. Lithium-ion batteries (LIBs) have dominated the energy storage landscape for decades, powering everything from portable electronics to electric vehicles (EVs).

However, the limitations of LIBs, including their relatively low energy density, safety concerns related to flammable liquid electrolytes, and finite cycle life, have spurred the search for next-generation battery technologies [5]. Among the most promising alternatives are solid-state batteries

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(SSBs), which replace liquid electrolytes with solid electrolytes [13]. This shift offers the potential for higher energy density, enhanced safety, and longer operational lifetimes, making SSBs a focal point of research and development in the energy storage community [2].

Solid-state batteries leverage solid electrolytes, which are inherently non-flammable and mechanically robust, addressing one of the most critical safety issues associated with conventional LIBs. The elimination of liquid electrolytes reduces the risk of leakage, thermal runaway, and fire hazards, which are particularly important for applications such as EVs and grid storage. Furthermore, the use of lithium metal as an anode in SSBs enables significantly higher energy densities compared to graphite anodes used in LIBs. With a theoretical capacity of 3,860 mAh/g, lithium metal anodes can potentially double the energy density of current LIBs, paving the way for longer-range EVs and more efficient energy storage systems [9].

Despite these advantages, the widespread adoption of SSBs has been hindered by several technical challenges. One of the most significant barriers is the low ionic conductivity of many solid electrolytes, which limits the power output and efficiency of SSBs [3]. Additionally, the interfaces between solid electrolytes and electrodes often exhibit high resistance, leading to poor charge transfer and reduced performance [21]. Mechanical stability is another critical issue, as the repeated expansion and contraction of electrodes during cycling can cause fractures in the solid electrolyte, compromising the battery's integrity. These challenges have driven extensive research into developing new materials, optimizing interfaces, and improving manufacturing processes to unlock the full potential of SSBs [4].

Recent advancements in materials science and engineering have brought SSBs closer to commercialization. Innovations in solid electrolyte materials, such as oxide-based, sulfide-based, and polymer-based electrolytes, have significantly improved ionic conductivity and stability. For example, sulfide-based electrolytes like $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$ (LGPS) exhibit ionic conductivities rivaling those of liquid electrolytes, while oxide-based electrolytes like $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZO) offer excellent chemical stability [6]. Interface engineering techniques, such as the use of thin interfacial layers and composite electrodes, have also shown promise in reducing interfacial resistance and enhancing cycling stability. These developments, coupled with advances in computational modeling and characterization techniques, have accelerated the understanding and optimization of SSB components [7].

The potential impact of SSBs extends beyond consumer electronics and EVs to include grid storage, aerospace, and other high-performance applications. As the world moves toward a more sustainable and electrified future, SSBs could play a pivotal role in enabling the widespread adoption of renewable energy sources and reducing greenhouse gas emissions. However, significant work remains to address the technical and economic challenges associated with SSBs.

This review explores the latest advancements in SSB technology, examines the key challenges that must be overcome, and outlines future research directions to realize the full potential of this transformative energy storage technology [8].

The growing demand for high-energy-density, safe, and sustainable energy storage systems has driven the development of solid-state batteries (SSBs). Unlike conventional LIBs, which use liquid electrolytes, SSBs employ solid electrolytes, offering several advantages, including: Enhanced safety: Solid electrolytes are non-flammable, reducing the risk of thermal runaway and fires [33]. Higher energy density: The use of lithium metal anodes enables higher theoretical capacities (3860 mAh/g compared to 372 mAh/g for graphite). Longer cycle life: Solid electrolytes are less prone to dendrite formation, which can cause short circuits in LIBs [10].

Despite these advantages, SSBs face significant challenges, including low ionic conductivity, high interfacial resistance, and mechanical instability. This review examines recent progress in addressing these challenges and discusses the path forward for SSB commercialization [11].

2. Solid Electrolytes: Materials and Properties

2.1. Solid Electrolytes: The Core Component of Solid-State Batteries

Solid electrolytes are the cornerstone of solid-state batteries (SSBs), as they replace the liquid electrolytes used in conventional lithium-ion batteries (LIBs). The properties of solid electrolytes such as ionic conductivity, chemical stability, mechanical strength, and compatibility with electrodes directly influence the performance, safety, and longevity of SSBs. Below, we elaborate on the three main classes of solid electrolytes: oxide-based, sulfide-based, and polymer-based electrolytes, highlighting their advantages, challenges, and recent advancements [12].

2.1.1. Oxide-Based Electrolytes

Oxide-based electrolytes are among the most widely studied solid electrolytes due to their high ionic conductivity and excellent chemical stability. A prominent example is $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZO), a garnet-type oxide electrolyte [13].

High Ionic Conductivity: LLZO exhibits ionic conductivities greater than 10^{-3} S/cm at room temperature, making it suitable for high-performance SSBs. **Excellent Chemical Stability:** Oxide-based electrolytes are stable against lithium metal anodes, reducing the risk of side reactions and dendrite formation. **Wide Electrochemical Window:** They can operate at high voltages, enabling compatibility with high-voltage cathodes [14].

Brittleness: Oxide-based electrolytes are inherently brittle, making them prone to cracking during battery assembly or cycling. **High Processing Temperatures:** Fabricating dense,

defect-free oxide electrolytes often requires high-temperature sintering (above 1,000 °C), which increases manufacturing costs and complexity. **Poor Interface with Electrodes:** The rigid nature of oxide electrolytes can lead to poor interfacial contact with electrodes, resulting in high interfacial resistance [15].

Doping Strategies: Doping LLZO with elements like Al, Ga, or Ta stabilizes the high-conductivity cubic phase and enhances ionic conductivity. **Thin-Film Fabrication:** Techniques such as pulsed laser deposition (PLD) and atomic layer deposition (ALD) have been used to create thin, dense oxide electrolyte layers with improved interfacial properties. **Composite Electrolytes:** Combining oxide electrolytes with polymers or other materials can improve mechanical flexibility and interfacial contact [16].

2.1.2. Sulfide-Based Electrolytes

Sulfide-based electrolytes are another promising class of solid electrolytes, known for their ultra-high ionic conductivity. A well-known example is Li₁₀GeP₂S₁₂ (LGPS), which exhibits ionic conductivities exceeding 10⁻² S/cm, rivaling those of liquid electrolytes [14].

Ultra-High Ionic Conductivity: Sulfide-based electrolytes have some of the highest ionic conductivities among solid electrolytes, enabling high power densities. **Soft Mechanical Properties:** Their softer nature allows for better interfacial contact with electrodes, reducing interfacial resistance. **Ease of Processing:** Sulfide electrolytes can be processed at lower temperatures compared to oxide-based electrolytes [17].

Poor Air Stability: Sulfide electrolytes are highly reactive with moisture and oxygen, requiring strict handling in inert atmospheres. **Reactivity with Lithium Metal:** Sulfides can react with lithium metal anodes, leading to the formation of resistive interphases and reduced cycling stability. **Toxicity:** Some sulfide materials contain toxic elements like germanium, raising environmental and safety concerns [18].

Air-Stable Sulfides: Researchers have developed air-stable sulfide electrolytes, such as Li₆PS₅Cl, by modifying their chemical composition. **Protective Coatings:** Applying thin protective layers (Li₃PO₄) on sulfide electrolytes can improve their stability against moisture and lithium metal. **Halogen Substitution:** Replacing sulfur with halogens (Cl, Br) in sulfide electrolytes has been shown to enhance ionic conductivity and stability [19].

2.1.3. Polymer-Based Electrolytes

Polymer-based electrolytes, such as polyethylene oxide (PEO) complexed with lithium salts (LiTFSI), offer a unique combination of flexibility and ease of processing. **Flexibility:** Polymer electrolytes are mechanically flexible, making them suitable for flexible and wearable electronics. **Ease of Processing:** They can be fabricated using simple solution-based methods, reducing manufacturing costs. **Good Interfacial Contact:** Their soft nature allows for excellent interfacial contact with electrodes, minimizing interfacial resistance [20].

Low Ionic Conductivity: Most polymer electrolytes exhibit low ionic conductivities (<10⁻⁴ S/cm) at room temperature, limiting their use in high-power applications. **Narrow Electrochemical Window:** Polymer electrolytes are typically stable only at low to moderate voltages, restricting their compatibility with high-voltage cathodes. **Mechanical Strength:** While flexible, polymer electrolytes often lack the mechanical strength to suppress lithium dendrite growth [21].

Composite Polymer Electrolytes: Incorporating ceramic fillers (LLZO, TiO₂) into polymer matrices can enhance ionic conductivity and mechanical strength. **Block Copolymers:** Designing block copolymers with distinct ion-conducting and mechanically robust domains has shown promise in improving both conductivity and mechanical properties. **Crosslinking:** Chemically crosslinking polymer chains can improve mechanical stability and suppress dendrite growth [22].

Table 1. Comparison of Solid Electrolyte Classes [13].

Property	Oxide-Based	Sulfide-Based	Polymer-Based
Ionic Conductivity	>10 ⁻³ S/cm	>10 ⁻² S/cm	<10 ⁻⁴ S/cm
Chemical Stability	Excellent	Poor (air-sensitive)	Moderate
Mechanical Properties	Brittle	Soft	Flexible
Processing	High-temperature	Moderate-temperature	Low-temperature
Interfacial Contact	Poor	Excellent	Excellent

Solid electrolytes are the defining feature of SSBs, and their properties play a critical role in determining battery performance [3]. Oxide-based electrolytes offer high conductivity and stability but face challenges in processing and interfacial

contact. Sulfide-based electrolytes provide ultra-high conductivity and excellent interfacial properties but suffer from poor air stability [22].

Polymer-based electrolytes are flexible and easy to process

but struggle with low conductivity and mechanical strength. Recent advancements in materials design, doping strategies, and interface engineering are addressing these challenges, bringing SSBs closer to commercialization. Continued research into novel materials and fabrication techniques will be essential to unlock the full potential of solid-state batteries [23].

2.2. Electrode Materials and Interface Engineering in Solid-State Batteries

The performance of solid-state batteries (SSBs) is heavily influenced by the compatibility between electrodes and solid electrolytes [3]. Unlike conventional lithium-ion batteries (LIBs), where liquid electrolytes can easily wet the electrode surfaces, SSBs face significant challenges due to the solid-solid interfaces between electrodes and electrolytes. These interfaces often exhibit high resistance, poor contact, and mechanical instability, which can severely limit battery performance. Below, we elaborate on the key challenges and strategies to address them, focusing on interfacial resistance, dendrite formation, and volume changes [24].

2.2.1. High Interfacial Resistance

In SSBs, the solid-solid interface between the electrode and electrolyte often results in poor physical contact, leading to high interfacial resistance. This resistance impedes ion transport, reducing the battery's power output and efficiency. The rigid nature of solid electrolytes exacerbates this issue, as they cannot conform to the electrode surface like liquid electrolytes [25].

Strategies to Address High Interfacial Resistance:

Surface Modification:

Thin Interfacial Layers: Coating electrodes or electrolytes with thin layers of materials like Al_2O_3 , Li_3PO_4 , or LiNbO_3 can improve interfacial contact and reduce resistance. These layers act as buffers, enhancing adhesion and facilitating ion transport. **Atomic Layer Deposition (ALD):** ALD is used to deposit ultra-thin, uniform coatings on electrode surfaces, ensuring excellent interfacial contact and stability [26].

Composite Electrodes:

Combining active electrode materials with solid electrolytes in a composite structure can enhance ionic and electronic conductivity. For example, mixing LiCoO_2 (cathode material) with LLZO (solid electrolyte) creates a percolating network for ion and electron transport. **Garnet-Polymer Composites:** Incorporating polymer electrolytes into oxide-based composites can improve flexibility and interfacial contact [27].

Hot Pressing:

Applying heat and pressure during battery assembly can improve the physical contact between electrodes and solid electrolytes, reducing interfacial resistance [26].

2.2.2. Dendrite Formation

Lithium metal anodes, which are essential for achieving high energy density in SSBs, are prone to dendrite formation. Dendrites are needle-like structures that grow from the anode surface during cycling, potentially piercing the solid electrolyte and causing short circuits. This phenomenon is a major safety concern and can lead to battery failure [28].

Mechanical Reinforcement:

Using solid electrolytes with high mechanical strength, such as LLZO, can physically block dendrite growth. **Composite Solid Electrolytes:** Incorporating ceramic fillers (Al_2O_3 , TiO_2) into polymer electrolytes can enhance mechanical strength and suppress dendrite penetration [29].

Surface Coatings:

Coating lithium metal anodes with protective layers (Li_3PO_4 , LiF , or graphene) can stabilize the electrode-electrolyte interface and prevent dendrite formation. **Artificial SEI Layers:** Creating an artificial solid-electrolyte interphase (SEI) on the lithium metal surface can improve interfacial stability and inhibit dendrite growth [30].

3D Lithium Hosts:

Designing 3D porous structures (carbon foams or metal scaffolds) to host lithium metal can distribute current more evenly and reduce localized dendrite formation [31].

2.3. Volume Changes

Electrode materials, particularly lithium metal anodes and high-capacity cathodes ($\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$, NCM811), undergo significant volume changes during charge and discharge cycles. These volume changes can cause mechanical stress at the electrode-electrolyte interface, leading to cracks, delamination, and increased resistance [32].

Porous Electrode Architectures:

Designing electrodes with porous or 3D structures can accommodate volume changes by providing space for expansion and contraction. For example, 3D lithium metal anodes with interconnected pores can reduce mechanical stress. **Gradient Porosity:** Electrodes with gradient porosity (denser near the current collector and more porous near the electrolyte) can improve mechanical stability and ion transport [33].

Elastic Interlayers: introducing elastic interlayers (e.g., polymer or gel layers) between the electrode and electrolyte can absorb mechanical stress and maintain interfacial contact during cycling [34].

Composite Electrodes:

Composite electrodes, which combine active materials with flexible binders and conductive additives, can better withstand volume changes. For example, silicon-based anodes, which undergo large volume changes, can be stabilized by embedding silicon particles in a conductive matrix [33].

2.4. Advanced Characterization and Modeling

Understanding and optimizing electrode-electrolyte inter-

faces require advanced characterization techniques and computational modelling [34]:

In Situ Techniques: Tools like in situ transmission electron microscopy (TEM) and X-ray tomography provide real-time insights into interfacial phenomena, such as dendrite growth and volume changes. **Computational Modeling:** Density functional theory (DFT) and molecular dynamics (MD) simulations help predict interfacial properties and guide the design of new materials and architectures [35].

Electrode materials and interface engineering are critical to the performance and reliability of SSBs. High interfacial resistance, dendrite formation, and volume changes are major challenges that must be addressed to realize the full potential of SSBs. Strategies such as surface modification, composite electrodes, and 3D architectures are showing promise in overcoming these challenges. Continued research into advanced materials, fabrication techniques, and characterization methods will be essential to develop robust and high-performance SSBs for commercial application [36].

2.5. Advanced Characterization and Computational Modeling in Solid-State Batteries

The development of solid-state batteries (SSBs) has been significantly accelerated by advancements in characterization techniques and computational modeling [30]. These tools provide critical insights into the structural, chemical, and electrochemical behavior of SSBs, enabling researchers to understand and optimize their performance. Below, we elaborate on the role of advanced characterization techniques and computational modeling in SSB research [37].

Characterization techniques are essential for understanding the complex phenomena occurring in SSBs, such as ion transport, interfacial reactions, and mechanical degradation. Recent advancements in these techniques have provided unprecedented insights into the behavior of SSBs during operation [38].

In Situ Transmission Electron Microscopy (TEM)

Purpose: In situ TEM allows researchers to observe structural and chemical changes in SSBs in real time, at atomic or nanoscale resolution.

Dendrite Growth: In situ TEM has been used to study the formation and growth of lithium dendrites in SSBs, providing insights into the mechanisms of dendrite penetration through solid electrolytes [26]. **Interfacial Reactions:** It enables the observation of interfacial reactions between electrodes and solid electrolytes, such as the formation of interphases or decomposition products. **Mechanical Degradation:** In situ TEM can reveal cracks and fractures in solid electrolytes caused by volume changes or mechanical stress during cycling. **Example:** Researchers have used in situ TEM to study the interaction between lithium metal and LLZO electrolytes, revealing the formation of resistive interphases that contribute

to high interfacial resistance [38].

X-Ray Tomography

Purpose: X-ray tomography provides 3D imaging of SSB components, allowing researchers to visualize the internal structure and morphology of electrodes and electrolytes [30].

Pore Structure Analysis: X-ray tomography can characterize the porosity and connectivity of electrode materials, which are critical for ion transport and electrochemical performance. **Interface Characterization:** It can visualize the contact between electrodes and solid electrolytes, identifying voids or poor interfacial contact that contribute to high resistance. **Dendrite Detection:** X-ray tomography can detect lithium dendrites within SSBs, even in opaque systems where optical techniques are ineffective. X-ray tomography has been used to study the evolution of lithium metal anodes during cycling, revealing the formation of voids and dendrites that degrade battery performance [29].

X-Ray Photoelectron Spectroscopy (XPS):

Provides chemical information about the surface and interface of SSB components, such as the composition of interphases or decomposition products. **Raman Spectroscopy:** Used to study the vibrational modes of materials, providing insights into structural changes and phase transitions in solid electrolytes and electrodes [28].

Nuclear Magnetic Resonance (NMR)

NMR can probe ion dynamics and local environments in solid electrolytes, helping to understand ion transport mechanisms [27].

2.6. Computational Modeling

Computational modeling plays a critical role in accelerating the development of SSBs by predicting material properties, optimizing interfaces, and guiding experimental design. Key computational approaches include [25]:

Density Functional Theory (DFT)

DFT is a quantum mechanical method used to calculate the electronic structure and properties of materials at the atomic scale. **Material Discovery:** DFT can predict the ionic conductivity, stability, and electrochemical properties of new solid electrolyte materials, such as doped LLZO or sulfide-based electrolytes [21]. **Interface Optimization:** DFT simulations can model the atomic structure and energetics of electrode-electrolyte interfaces, identifying strategies to reduce interfacial resistance [24]. **Reaction Mechanisms:** DFT can elucidate the mechanisms of interfacial reactions, such as the decomposition of solid electrolytes or the formation of interphases. DFT has been used to identify dopants (Al, Ga) that stabilize the high-conductivity cubic phase of LLZO, improving its ionic conductivity [23].

Molecular Dynamics (MD)

MD simulations model the motion of atoms and molecules over time, providing insights into ion transport and mechanical behavior [22]. **Ion Transport:** MD can simulate ion diffusion pathways in solid electrolytes, helping to understand

and optimize ionic conductivity [27]. **Mechanical Properties:** MD can predict the mechanical stability of solid electrolytes under stress, such as during volume changes or dendrite growth. **Interface Dynamics:** MD can model the dynamic behavior of electrode-electrolyte interfaces, revealing mechanisms of interfacial resistance and degradation. MD simulations have been used to study the diffusion of lithium ions in sulfide-based electrolytes, such as Li₁₀GeP₂S₁₂, confirming their ultra-high ionic conductivity [25].

Phase-Field Modeling

Phase-field modeling is a continuum-scale approach used to simulate microstructural evolution and electrochemical processes in SSBs. **Dendrite Growth:** Phase-field models can predict the growth of lithium dendrites in SSBs, helping to design strategies to suppress dendrite formation [9]. **Mechanical Stress:** These models can simulate the mechanical stress and strain in solid electrolytes caused by volume changes or external pressure. **Interface Evolution:** Phase-field modeling can predict the evolution of electrode-electrolyte interfaces during cycling, identifying factors that contribute to degradation. Phase-field models have been used to study the effect of external pressure on the interfacial contact between lithium metal anodes and solid electrolytes [31].

2.7. Integration of Characterization and Modeling

The integration of advanced characterization techniques and computational modeling is a powerful approach to accelerate SSB development. **Data-Driven Discovery:** Combining experimental data from characterization techniques with machine learning algorithms can identify new materials and optimize interfaces [12]. **Multiscale Modeling:** Linking atomistic simulations (DFT, MD) with continuum models (phase-field) provides a comprehensive understanding of SSB behavior across different length and time scales [19]. **Predictive Design:** Computational models can guide the design of experiments, reducing the time and cost of trial-and-error approaches [23].

Advanced characterization techniques and computational modeling are indispensable tools for understanding and optimizing SSBs. Techniques like in situ TEM and X-ray tomography provide real-time, high-resolution insights into the structural and chemical changes occurring in SSBs, while computational methods like DFT and MD enable the prediction and optimization of material properties and interfaces. The integration of these approaches is driving the development of high-performance SSBs, bringing them closer to commercialization. Continued advancements in characterization and modeling will be essential to overcome the remaining challenges and unlock the full potential of SSBs [22].

Despite the significant progress made in solid-state battery (SSB) technology, several challenges must be addressed to

enable their widespread commercialization. These challenges span materials development, manufacturing scalability, performance optimization, and safety [11].

3. Conclusion

In conclusion, solid-state batteries (SSBs) represent a significant advancement in energy storage technology, promising to address the limitations associated with conventional lithium-ion batteries (LIBs). The innovations in solid electrolytes—ranging from oxide-based to sulfide and polymer-based systems—have provided the foundation for enhancing energy density, safety, and cycle life. Despite these advancements, key challenges remain, particularly regarding ionic conductivity, interfacial resistance, and mechanical stability.

Ongoing research must focus on optimizing these materials and developing scalable fabrication methods to facilitate commercial viability. The integration of advanced characterization techniques and computational modeling will be crucial in accelerating the discovery and refinement of new materials and architectural designs, ultimately leading to high-performance SSBs. By overcoming existing hurdles, SSBs can play a pivotal role in driving the adoption of renewable energy solutions and enabling the electrification of transportation, thereby contributing to a more sustainable future.

Future studies should also explore the integration of SSBs with broader energy systems, including renewable sources, to fully leverage their potential. Collaborative efforts across multiple disciplines will be essential to unlock the full promise of solid-state battery technology, positioning it as a transformative solution for modern energy storage challenges.

Abbreviations

DFT	Density Functional Theory
EVs	Electric Vehicles
PLD	Pulsed Laser Deposition
LIBs	lithium-ion Batteries
NMR	Nuclear Magnetic Resonance
MD	Molecular Dynamics
TEM	Transmission Electron Microscopy
SSBs	Solid-state Batteries
XPS	X-Ray Photoelectron Spectroscopy

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Conflicts of Interest

The authors declare no conflicts of interest.

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