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Structural and Electrical Investigations of Lithium Borate Glasses Modified with Different Electrolytes for High Ionic Conductivity

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ABSTRACT: Lithium borate-based glass electrolytes have emerged as promising materials for solid-state electrochemical applications due to their excellent glass-forming ability, structural flexibility, and tunable ionic conductivity. Among various systems, $Li_2O–B_2O_3$ glasses modified with additional network formers such as SiO_2, P_2O_5 and SeO_2 exhibit remarkable enhancement in electrical, dielectric, and thermal properties. This review critically summarizes recent progress in the synthesis, structural characterization, ionic transport mechanisms, dielectric behaviour, and thermal stability of $Li_2O–B_2O_3$ based glass systems. Emphasis is placed on the role of mixed former effects, network depolymerization, and dopant-induced disorder in facilitating Li^+ ion migration. Comparative analysis of conductivity, activation energy, and stability reported in the literature is presented. The review highlights SeO_2-modified lithium borate glasses as strong candidates for next-generation solid-state batteries and high-temperature electrochemical devices.

KEYWORDS: $Li_2O–B_2O_3$ glasses, solid electrolytes, ionic conductivity, dielectric properties, mixed former effect, SeO_2, SiO_2 and P_2O_5 modification.

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I. INTRODUCTION

The rapid expansion of portable electronics, electric vehicles, and renewable energy technologies has intensified the demand for safe, efficient, and thermally stable energy storage systems. Conventional liquid electrolytes suffer from inherent limitations such as leakage, flammability, and restricted operating temperature ranges. These challenges have driven extensive research into solid-state electrolytes, which offer improved safety and long-term reliability.

Glassy solid electrolytes are particularly attractive due to their isotropic nature, absence of grain boundaries, and ease of compositional modification. Lithium borate ($Li_2O–B_2O_3$) based glasses have gained significant attention owing to their excellent glass-forming ability and adaptable structural framework. In pure borate glasses, ionic conductivity is relatively low; however, the incorporation of Li_2O introduces non-bridging oxygen (NBOs) that enhance Li^+ ion mobility. Further improvements in conductivity and stability can be achieved by introducing secondary network formers such as SiO_2, P_2O_5 and SeO_2, which tailor the glass network and optimize conduction pathways.

This review provides a comprehensive overview of the structural and electrical investigations of $Li_2O–B_2O_3$ based glasses modified with SiO_2, P_2O_5 and SeO_2, emphasizing structure–property relationships and application prospects.

II. RELATED WORK

Extensive studies over the past several decades have explored lithium borate glasses as solid electrolytes. Early investigations established that $Li_2O–B_2O_3$ glasses conduct ions through thermally activated hopping of Li^+ ions between

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interstitial sites created by borate network depolymerization. Subsequent research demonstrated that the addition of mixed network formers significantly alters short-range order and enhances ionic transport.
SiO_2 addition improves network rigidity and chemical durability while maintaining reasonable conductivity. P_2O_5 enhances amorphous character and introduces additional NBOs, leading to improved dielectric response. More recent studies have shown that SeO_2 modification substantially increases ionic conductivity due to higher polarizability, reduced activation energy, and increased free volume. These findings confirm that mixed former lithium borate glasses exhibit superior electrochemical performance compared to binary systems.

III. SIGNIFICANCE OF THE SYSTEM

This article reviews the structural and electrical investigations of $Li_2O–B_2O_3$ based glass electrolytes modified with SiO_2, P_2O_5 and SeO_2 for enhanced ionic conductivity and thermal stability. The related research work reported by various scientists on lithium borate glass systems is discussed in Section II. Section IV presents the commonly used glass preparation techniques and characterization methods adopted in these studies. Section V summarizes and analyses the structural, electrical, dielectric, and thermal properties of the modified glass systems along with their conduction mechanisms. Finally, Section VI discusses the application potential, future research directions, and concluding remarks of the reviewed studies.

IV. METHODOLOGY

Materials

Lithium Oxide (Li_2O)
The primary lithium source used in the reported studies on $Li_2O–B_2O_3$ based glass electrolytes is lithium carbonate (Li_2CO_3) of analytical grade purity. Li_2CO_3 is commonly employed due to its thermal stability, ease of decomposition, and controlled release of Li_2O during melting. It plays a crucial role as a network modifier by introducing mobile Li^+ ions and creating non-bridging oxygens within the borate glass network.

Boron Oxide (B_2O_3)
Boron oxide, generally obtained from high-purity boric acid (H_3BO_3), serves as the primary glass former in lithium borate systems. B_2O_3 provides excellent glass-forming ability and forms structural units such as BO_3 and BO_4, which significantly influence network connectivity and ionic transport behaviour.

Silicon Dioxide (SiO_2)
Silicon dioxide (SiO_2) is widely used as a secondary network former in $Li_2O–B_2O_3$ based glass electrolytes. SiO_2 enhances the rigidity and stability of the glass network by forming strong Si–O–Si and Si–O–B linkages. Its incorporation increases the degree of cross-linking, improves chemical durability, and raises the glass transition temperature. Although excessive SiO_2 content may reduce ionic conductivity due to increased network compactness, optimized concentrations promote structural stability while maintaining sufficient Li^+ ion mobility. SiO_2 also suppresses crystallization, thereby preserving the amorphous nature essential for uniform ionic transport.

Phosphorus Pentoxide (P_2O_5)
Phosphorus pentoxide (P_2O_5) acts as an effective glass former and network modifier in lithium borate glass systems. The presence of PO_4 structural units introduces additional non-bridging oxygens, which facilitate Li^+ ion migration and enhance ionic conductivity. P_2O_5 improves glass-forming ability and increases structural disorder, resulting in enhanced amorphousness. However, higher P_2O_5 concentrations may lead to moisture sensitivity due to hygroscopic nature. Therefore, controlled addition of P_2O_5 is essential to achieve a balance between high conductivity, dielectric stability, and chemical durability.

Selenium Dioxide (SeO_2)
Selenium dioxide (SeO_2) is introduced as a network modifier in $Li_2O–B_2O_3$ based glasses to enhance ionic conductivity and dielectric performance. SeO_2 increases the polarizability of the glass network due to the formation of Se–O bonds, which lowers the activation energy for Li^+ ion transport. The incorporation of SeO_2 also increases free volume and structural flexibility, thereby facilitating faster ionic motion. Compared to traditional modifiers, SeO_2 significantly improves electrical conductivity without severely compromising thermal stability, making SeO_2-modified lithium borate glasses highly suitable for solid-state electrolyte and electrochemical applications.

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Methods

Thermal Analysis
Thermal stability of the glass systems is examined using differential thermal analysis (DTA) and thermogravimetric analysis (TGA). These techniques are employed to determine glass transition temperature, crystallization behaviour, and thermal decomposition characteristics. The effect of dopants on thermal resistance and stability is analysed based on reported thermal profiles.

Differential Scanning Calorimetry (DSC)
Differential scanning calorimetry is frequently employed to determine thermal parameters such as glass transition temperature (Tg), crystallization temperature (Tc), and thermal stability range. DSC helps in understanding the effect of SiO_2, P_2O_5, and SeO_2 incorporation on structural relaxation and phase stability of lithium borate glasses.

Raman Spectroscopy
Raman spectroscopy is used to complement FTIR analysis by providing detailed information on short-range structural units such as BO_3, BO_4, PO_4, SiO_4, and SeO_3 groups. Changes in peak position and intensity reveal network polymerization, mixed former effects, and dopant-induced structural disorder.

Nuclear Magnetic Resonance (NMR) Spectroscopy
Solid-state NMR techniques, particularly ^{11}B and ^{31}P NMR, are widely used to quantify the relative fractions of BO_3/BO_4 units and phosphate environments. NMR provides direct evidence of network modification and its correlation with ionic conductivity enhancement.

Glass Preparation Technique
Most studies reviewed employ the conventional melt-quenching technique for glass synthesis. Stoichiometric amounts of precursor materials are thoroughly mixed and melted in platinum or alumina crucibles at temperatures ranging from 1000 to 1200 °C. The homogeneous melt is then rapidly quenched between stainless steel or brass plates to obtain amorphous glass samples. The quenched glasses are typically annealed near the glass transition temperature to relieve internal stresses.
Alternative synthesis routes such as sol–gel processing and mechanochemical milling are also reported. The sol–gel method enables molecular-level mixing and lower processing temperatures, while mechanochemical synthesis promotes defect generation and nano structuring, which can enhance ionic transport.

Structural Characterization
X-ray diffraction (XRD) is widely used to confirm the amorphous nature of the prepared glass samples, characterized by broad diffraction halos. Fourier transform infrared (FTIR) and Raman spectroscopy are employed to identify structural units such as BO_3, BO_4, SiO_4, PO_4, and SeO_3 and to analyse network modification effects. Scanning electron microscopy (SEM) and atomic force microscopy (AFM) are used to study surface morphology and homogeneity.

Electrical and Dielectric Measurements
Electrical properties are mainly investigated using impedance spectroscopy over a wide frequency range (typically 1 Hz to 1 MHz) and temperature range. DC ionic conductivity is calculated from impedance data and analysed using Arrhenius plots to determine activation energy. AC conductivity behaviour is interpreted using Jonscher's universal power law to identify ion transport mechanisms such as correlated barrier hopping.
Dielectric parameters, including dielectric constant and dielectric loss, are evaluated as a function of frequency and temperature to understand polarization effects and relaxation behaviour.

V. EXPERIMENTAL RESULTS AND DISCUSSION

Influence of SiO_2, P_2O_5, and SeO_2 on Glass Properties
The reviewed studies on $Li_2O–B_2O_3$ based glass electrolytes demonstrate that the incorporation of SiO_2, P_2O_5, and SeO_2 significantly influences the structural, electrical, dielectric, and thermal characteristics of the glass system. These modifiers alter the glass network connectivity, free volume, and lithium ion transport pathways. The results reported in the literature are collectively discussed below by correlating compositional variation with observed material properties.

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Structural Characteristics

X-ray diffraction (XRD) patterns reported in most studies exhibit broad halos without sharp crystalline peaks, confirming the amorphous nature of all prepared glass compositions. The addition of SiO_2 and P_2O_5 enhances glass-forming ability and suppresses crystallization by increasing network connectivity through Si–O–B and P–O–B linkages. FTIR and Raman spectroscopic analyses reveal systematic changes in structural units, such as an increase in BO_4 units at the expense of BO_3 groups with modifier addition.

The presence of SeO_2 introduces Se–O bonds that increase structural disorder and polarizability. This mixed former effect leads to a more flexible glass network, which is favourable for ionic conduction. NMR studies further confirm the redistribution of borate and phosphate structural units, supporting the observed electrical behaviour.

Ionic Conductivity Behaviour

The DC ionic conductivity increases with the introduction of P_2O_5 and SeO_2 due to the formation of non-bridging oxygens and enhanced lithium ion mobility. Maximum conductivity is typically observed at optimized modifier concentrations, beyond which conductivity decreases due to excessive network rigidity or ion trapping. SeO_2-modified glasses consistently exhibit lower activation energy compared to SiO_2-rich compositions, indicating easier Li^+ ion hopping.

Arrhenius plots confirm thermally activated conduction behaviour, and the calculated activation energies decrease with increasing structural disorder and free volume.

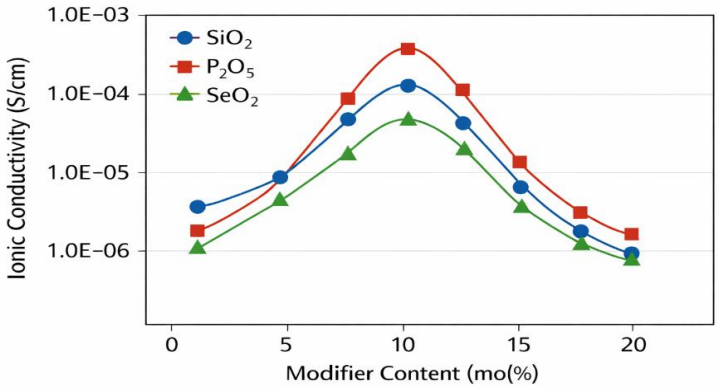

Figure 1. Variation of ionic conductivity of Li_2O–B_2O_3 based glasses with SiO_2, P_2O_5, and SeO_2 content

Dielectric and Relaxation Properties

Dielectric studies reveal that the dielectric constant decreases with increasing frequency and increases with temperature, indicating space charge polarization effects at low frequencies. The inclusion of SeO_2 significantly enhances dielectric constant due to its high polarizability. Dielectric loss analysis shows relaxation peaks that shift toward higher frequencies with temperature, confirming ion hopping mechanisms.

Modulus spectroscopy analysis suppresses electrode polarization effects and provides insight into relaxation dynamics. The non-Debye relaxation behaviour observed in most compositions is attributed to the distribution of relaxation times caused by structural heterogeneity.

Thermal Stability Analysis

Thermal analysis using DTA, DSC, and TGA demonstrates that the addition of SiO_2 improves thermal stability by increasing glass transition and crystallization temperatures. P_2O_5 enhances glass-forming ability but may slightly reduce moisture resistance at higher concentrations. SeO_2-modified glasses exhibit moderate thermal stability with improved electrical performance, indicating a trade-off between conductivity and thermal resistance.

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Table 1.
General trends in thermal behaviour of modified $Li_2O–B_2O_3$ glass systems

Sr. No	Modifier	Tg Trend	Thermal Stability	Electrical Impact
1	SiO_2	Increases	High	Moderate Conductivity
2	SeO_2	Slight decrease	Moderate	Highest Conductivity
3	P_2O_5	Moderate increase	Good	High conductivity

Conduction Mechanism
The conduction mechanism in these glass electrolytes is primarily governed by lithium ion hopping between non-bridging oxygen sites. Jonscher's universal power law is widely used to describe AC conductivity behavior, indicating correlated barrier hopping as the dominant transport mechanism. The presence of mixed glass formers enhances conduction pathways by disrupting long-range order and increasing structural flexibility.

Comparative Performance Evaluation
A qualitative performance comparison across reported compositions suggests that glasses containing optimized amounts of P_2O_5 and SeO_2 exhibit the best balance between ionic conductivity and structural stability. Excessive modifier content leads to network over-modification, resulting in reduced conductivity and thermal performance.

Discussion and Implications
The reviewed results clearly indicate that controlled modification of $Li_2O–B_2O_3$ glasses with SiO_2, P_2O_5, and SeO_2 is an effective strategy for tailoring glass electrolyte properties. SiO_2 enhances mechanical and thermal stability, P_2O_5 improves glass formation and ionic transport, while SeO_2 significantly lowers activation energy and enhances conductivity.
From an application perspective, these materials show strong potential for use in solid-state lithium batteries, electrochemical sensors, and energy storage devices. Optimized compositions provide a favourable balance between conductivity, stability, and processability, which is critical for practical deployment.

V. CONCLUSION AND FUTURE WORK

This review has systematically analysed the structural, electrical, dielectric, and thermal properties of $Li_2O–B_2O_3$ based glass electrolytes modified with SiO_2, P_2O_5, and SeO_2, as reported in the literature. The reviewed studies demonstrate that controlled modification of lithium borate glass networks plays a crucial role in tailoring ionic conductivity and stability, which are essential for solid-state electrochemical applications.
The incorporation of SiO_2 enhances network rigidity, glass-forming ability, and thermal stability by increasing cross-link density, while P_2O_5 contributes to improved amorphousness and ionic transport through the formation of non-bridging oxygens. SeO_2 acts as an effective network modifier by increasing polarizability and free volume, thereby reducing activation energy and significantly enhancing lithium ion mobility. An optimal balance between these modifiers results in glass systems exhibiting high ionic conductivity along with acceptable thermal and mechanical stability.
The conduction mechanism in these glasses is primarily governed by thermally activated lithium ion hopping, supported by impedance, dielectric, and modulus spectroscopy analyses reported in the literature. Mixed glass former effects arising from the simultaneous presence of borate, silicate, phosphate, and selenite units further contribute to improved conduction pathways and relaxation dynamics.
From an application perspective, $Li_2O–B_2O_3$ based glasses modified with suitable concentrations of SiO_2, P_2O_5, and SeO_2 show strong potential as solid electrolytes for lithium-ion batteries, electrochemical sensors, and other energy storage devices. However, excessive modifier content can lead to network over-rigidity or structural instability, highlighting the importance of compositional optimization.
Future research should focus on systematic compositional tuning to achieve an optimal trade-off between ionic conductivity and thermal stability. Advanced characterization techniques such as solid-state NMR, synchrotron-based spectroscopy, and in-situ impedance analysis are recommended to gain deeper insight into ion transport mechanisms. Additionally, computational approaches including molecular dynamics simulations and density functional theory can provide atomistic understanding of lithium migration pathways. Long-term stability studies, interface compatibility with

electrode materials, and scalable fabrication methods should also be explored to facilitate practical implementation of these glass electrolytes in next-generation solid-state energy storage systems.

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