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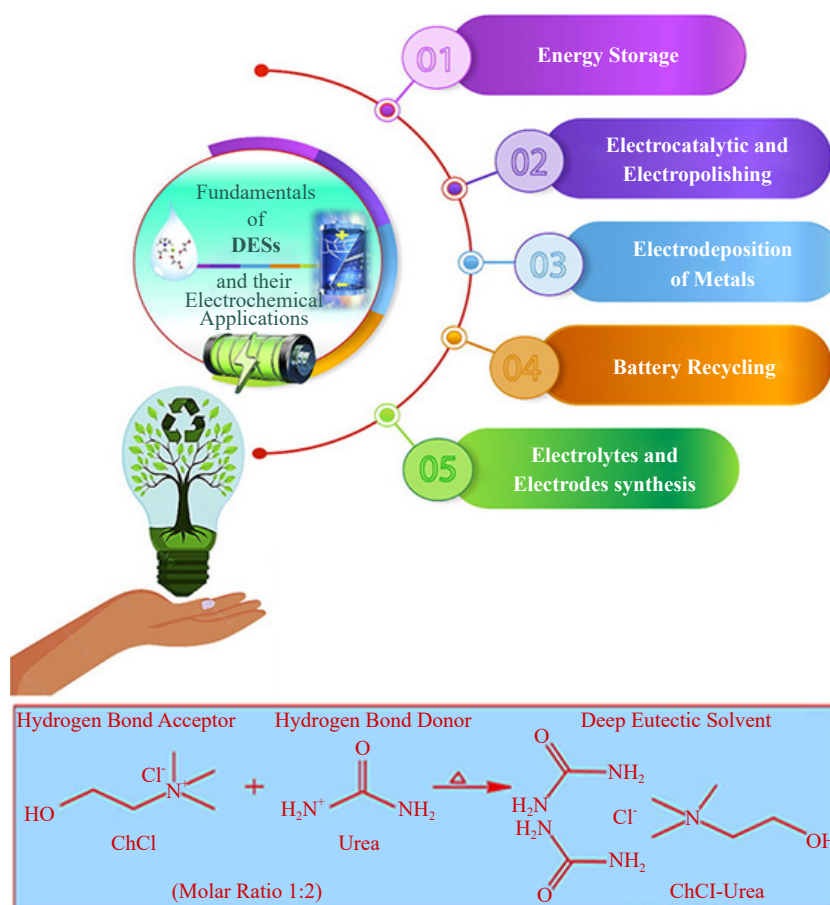
The Roles of Deep Eutectic Solvents in Batteries for Sustainable Energy Storage

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Graphical Abstract



Abstract: The escalating demand for sustainable and efficient energy storage solutions has catalyzed the exploration of advanced materials and technologies in battery systems. Deep Eutectic Solvents (DES), a novel class of ionic liquids, have emerged as promising candidates due to their unique physicochemical properties, including low volatility, high thermal stability, and tunable solvation capabilities. This paper investigates the roles of DES in enhancing the performance and sustainability of batteries, particularly focusing on lithium-ion and sodium-ion technologies. DES-based electrolytes offer significant advantages, such as improved ionic conductivity and enhanced electrochemical stability, which are critical for the longevity and efficiency of battery systems. Furthermore, the eco-friendly nature of DES, derived from biodegradable and non-toxic components, aligns with the overarching goal of developing green and sustainable energy storage solutions. This study synthesizes recent advancements in DES research, evaluates their practical applications in current battery technologies, and discusses future directions for integrating DES in next-generation energy storage systems. The findings underscore the potential of DES to revolutionize battery technology, contributing to a sustainable energy future through enhanced battery performance and reduced environmental impact.

Keywords: Dendrite-free electrolyte systems, sustainable energy storage, dendrite suppression, electrolyte additives, lithium-ion batteries, solid-state batteries

1. Introduction

As the global demand for sustainable energy solutions continues to rise, the need for efficient, cost-effective, and environmentally friendly energy storage systems becomes increasingly critical. Traditional lithium-ion batteries, although widely used, face challenges such as limited raw material availability, safety concerns, and environmental impact. To address these issues, researchers are exploring alternative materials and technologies that can enhance the sustainability of energy storage systems. One promising avenue of research is the use of Deep Eutectic Solvents (DES) in batteries.¹

DES are a class of ionic solvents formed by mixing two or more components, typically a hydrogen bond donor and a hydrogen bond acceptor, in such proportions that the mixture exhibits a significantly lower melting point than either of the individual components. This unique property makes DES highly versatile and suitable for a wide range of applications, including electrochemistry and materials science. In the context of batteries, DES offer several advantages that could potentially revolutionize energy storage technology.²

1.1 Background of the study

The primary motivation for investigating DES in battery applications is their potential to overcome the limitations associated with conventional electrolytes. Traditional electrolytes used in lithium-ion batteries, such as organic carbonates, pose several challenges, including flammability, toxicity, and limited electrochemical stability. In contrast, DES are generally non-flammable, exhibit low toxicity, and possess high thermal and chemical stability, making them attractive candidates for safer and more sustainable battery electrolytes.³

Furthermore, DES are relatively easy to synthesize from inexpensive and abundant materials, aligning with the goal of reducing the overall cost of battery production and environmental impact. The tunable nature of DES, achieved by varying the components and their ratios, allows for the customization of their physical and chemical properties to meet the specific requirements of different battery systems.⁴

1.2 Research problem

Despite the promising characteristics of DES, their application in batteries is still in the early stages of development. Several fundamental challenges need to be addressed to fully realize their potential. These challenges include understanding the mechanisms of ion transport within DES, optimizing their electrochemical performance, and ensuring their long-term stability and compatibility with other battery components.

The research problem, therefore, lies in systematically exploring the roles of DES in batteries to identify the key factors that influence their performance and sustainability. This involves investigating various DES formulations,

characterizing their properties, and evaluating their performance in different battery configurations.

1.3 Objectives

The primary objectives of this research are to:

- Synthesize and characterize a range of DES formulations suitable for battery applications.
- Investigate the ion transport mechanisms and electrochemical behavior of DES-based electrolytes.
- Evaluate the performance of DES in various battery systems, including lithium-ion, sodium-ion, and solid-state batteries.
- Assess the environmental impact and sustainability of DES in comparison to traditional electrolytes.
- Identify the key challenges and opportunities for the commercialization of DES-based batteries.

1.4 Significance of the study

The significance of this study lies in its potential to contribute to the development of safer, more sustainable, and cost-effective energy storage solutions. By advancing the understanding of DES in batteries, this research could pave the way for the next generation of battery technologies that are not only efficient and high-performing but also environmentally friendly and economically viable.

Furthermore, the successful integration of DES in batteries could have far-reaching implications for various sectors, including renewable energy, electric vehicles, and portable electronics, by enhancing the reliability and sustainability of energy storage systems. This, in turn, would support global efforts to transition towards cleaner and more sustainable energy sources, addressing pressing environmental and economic challenges.

2. Properties of DES

DES are an emerging class of ionic liquids that have garnered significant interest in various fields, including energy storage. DES are formed by mixing a hydrogen bond donor and a hydrogen bond acceptor, resulting in a eutectic mixture with unique physicochemical properties. Their application in batteries is particularly promising for sustainable energy storage solutions.^{4,5}

DES are known for their low volatility, high thermal stability, and high ionic conductivity, making them suitable for use in battery electrolytes. These properties contribute to safer and more durable battery systems. Additionally, the wide electrochemical window of DES allows for their application in high-voltage batteries, enhancing energy density and performance. This paper explores the properties of DES and their roles in enhancing the sustainability of batteries.

2.1 Low volatility and high thermal stability

One of the most advantageous properties of DES is its low volatility and high thermal stability. Unlike traditional organic solvents, DES does not evaporate easily, reducing the risk of leakage in batteries. This non-volatile nature contributes to the overall safety of battery systems, minimizing the potential for thermal runaway—a critical issue in high-energy-density batteries. The high thermal stability of DES allows them to operate effectively under a wide range of temperatures, enhancing the durability and lifespan of batteries.⁶

One critical area of research is the relevance of low concentrations of DES in energy storage applications. The current literature suggests that while low concentrations of DES can demonstrate certain beneficial properties, their effectiveness in practical applications remains underexplored. For DES to be viable in real-world battery systems, a deeper understanding of their behavior at various concentrations is essential. This includes their ionic conductivity, electrochemical stability, and overall impact on battery performance.⁷

A comprehensive analysis of existing studies highlights the potential and limitations of DES in battery applications. For instance, research has shown that DES can improve the ionic conductivity and thermal stability of batteries. However, these studies often use concentrations that may not be representative of practical scenarios. To bridge this gap, it is crucial to conduct experiments that mimic real-world conditions, including varying DES concentrations and

assessing their impact on battery performance over extended periods.

2.2 Ionic conductivity

DES exhibits high ionic conductivity, which is essential for efficient ion transport within batteries. This property ensures that ions can move freely through the electrolyte, facilitating fast charging and discharging processes. High ionic conductivity is crucial for maintaining the performance of batteries, especially in applications requiring rapid energy delivery, such as electric vehicles and grid storage. The ionic nature of DES contributes to their superior conductivity compared to conventional organic solvents.⁸

DES are frequently compared to ionic liquids (ILs) due to their similar properties. However, DES offer distinct advantages, such as lower cost and easier preparation. A comparative analysis reveals that while ILs have been extensively studied and applied in energy storage, DES offer a more sustainable and potentially more efficient alternative. Future research should focus on direct comparisons of DES and ILs in identical experimental setups to provide clearer insights into their relative benefits and limitations.

2.3 Wide electrochemical window

The wide electrochemical window of DES is another key property that makes them suitable for battery applications. This broad electrochemical stability range allows DES to be used in high-voltage batteries without undergoing decomposition. A wide electrochemical window ensures that DES can accommodate a variety of electrode materials, including those that operate at high potentials. This versatility is particularly beneficial for developing advanced battery chemistries that aim to achieve higher energy densities.⁹

To accurately assess the potential of DES in batteries, a standardized methodology for evaluating their performance is necessary. This includes consistent measurements of ionic conductivity, electrochemical stability, and thermal properties across different DES formulations and concentrations. Such standardization will enable more reliable comparisons and facilitate the identification of the most promising DES for specific energy storage applications.

2.4 Biodegradability and low toxicity

Environmental sustainability is a critical consideration in modern battery technology. DES are often biodegradable and exhibit low toxicity, making them environmentally friendly alternatives to traditional solvents. The low toxicity of DES reduces health risks associated with battery manufacturing, use, and disposal. Additionally, the biodegradability of DES ensures that they do not persist in the environment, aligning with global efforts to minimize ecological impact and promote green technologies.

DES are often touted as green solvents due to their low toxicity and biodegradability. However, the environmental impact of DES varies significantly depending on their constituents. For example, DES composed of choline chloride and ethylene glycol may not be as biocompatible as other mixtures. Therefore, it is important to carefully evaluate the greenness of each DES formulation and consider the entire lifecycle of the solvent in the context of sustainability.¹⁰

3. Roles of DES in batteries for sustainability

3.1 Enhanced safety and longevity

The low volatility and high thermal stability of DES enhance the safety profile of batteries, reducing the likelihood of accidents and improving longevity. By mitigating the risk of thermal runaway and leakage, DES contributes to developing safer battery systems that can operate reliably over extended periods.^{11,12}

3.2 Improved performance and efficiency

High ionic conductivity is critical for optimizing the performance and efficiency of batteries. DES enables faster ion transport, improving charging and discharging rates. This property is particularly advantageous for applications

requiring high power output and rapid energy exchange, such as electric vehicles and renewable energy storage.¹³

3.3 Support for high-voltage applications

The wide electrochemical window of DES supports the use of high-voltage electrode materials, paving the way for batteries with higher energy densities.¹⁴ This capability is essential for next-generation batteries that aim to deliver greater energy storage capacity while maintaining stability and performance.

3.4 Environmental and health benefits

The biodegradability and low toxicity of DES offer significant environmental and health benefits. By replacing traditional, hazardous solvents with DES, the battery industry can reduce its ecological footprint and minimize health risks.^{15,16} This shift towards greener solvents aligns with the principles of sustainable development and responsible innovation.

The environmental impact of DES varies significantly depending on their specific constituents. While DES are often promoted as green solvents due to their low toxicity and biodegradability, it is essential to recognize that not all DES are equally environmentally friendly. For example, a DES composed of choline chloride (ChCl) and ethylene glycol (EG) is not the most biocompatible option. Therefore, categorizing all DES as inherently green solvents can be misleading.¹⁷ A more nuanced approach is necessary, evaluating each DES formulation on a case-by-case basis to accurately determine its environmental friendliness.¹⁸

4. Specific DES mixtures utilized for energy storage

4.1 Choline Chloride-Ethylene Glycol (ChCl:EG)

One of the widely studied DES mixtures is Choline Chloride-Ethylene Glycol (ChCl:EG). This mixture is known for its high ionic conductivity and wide electrochemical window, making it suitable for battery applications. However, the biocompatibility of ChCl:EG is a concern due to the toxicity of ethylene glycol. The ratio of ChCl to EG significantly affects the physicochemical properties of the mixture. Higher concentrations of EG lead to lower viscosity and higher ionic conductivity, but also reduce the environmental friendliness of the DES.¹⁹

4.2 Choline Chloride-Urea (ChCl:Urea)

Choline Chloride-Urea (ChCl:Urea) is another DES mixture used in energy storage. This mixture is notable for its low cost and biodegradability. The hydrogen bond interactions between ChCl and Urea result in a stable eutectic mixture with good thermal stability. The ratio of ChCl to Urea impacts the melting point and viscosity of the DES, with higher Urea concentrations leading to lower melting points and viscosity. This mixture has been used in electrochemical capacitors and batteries, providing a balance between performance and sustainability.

4.3 Choline Chloride-Glycerol (ChCl:Glycerol)

Choline Chloride-Glycerol (ChCl:Glycerol) is recognized for its excellent biocompatibility and environmental safety. This DES mixture has been utilized in lithium-ion batteries as an electrolyte. The ratio of ChCl to Glycerol affects the ionic conductivity and viscosity of the mixture. Higher concentrations of Glycerol increase the viscosity but enhance the safety and biocompatibility of the DES. The ChCl:Glycerol mixture is particularly advantageous for applications requiring non-toxic and biodegradable solvents.

4.4 Impact of hydrogen bond donor (HBD) to hydrogen bond acceptor (HBA) ratios

The ratio of HBD to HBA in DES mixtures plays a crucial role in determining their physicochemical properties and suitability for energy storage applications. The following are the key impacts of varying HBD to HBA ratios.

4.4.1 Viscosity and ionic conductivity

The viscosity of DES mixtures is heavily influenced by the HBD to HBA ratio. Higher concentrations of HBD typically result in lower viscosity, which enhances ionic conductivity. This is critical for battery applications where efficient ion transport is necessary for fast charging and discharging processes. For example, in ChCl:EG mixtures, increasing the concentration of EG (HBD) reduces viscosity and increases ionic conductivity.

4.4.2 Thermal stability

Thermal stability is another important property affected by the HBD to HBA ratio. DES mixtures with higher concentrations of HBA generally exhibit higher thermal stability, making them suitable for applications involving high-temperature operations. In ChCl:Urea mixtures, the stability of the eutectic mixture is maintained across a wide temperature range, ensuring the durability of batteries under varying thermal conditions.

4.4.3 Environmental impact

The environmental impact of DES mixtures is closely tied to the choice and ratio of HBD and HBA. Mixtures like ChCl:Glycerol are favored for their biocompatibility and biodegradability, making them environmentally friendly alternatives to traditional solvents. Adjusting the ratio to optimize performance while maintaining low toxicity is essential for developing green solvents for energy storage.

5. DES in battery technologies

5.1 DES-based electrolytes

DES have emerged as a promising electrolyte in the development of advanced battery technologies, such as lithium-ion, sodium-ion, and redox flow batteries. DES are composed of a mixture of two or more components, which, when combined, exhibit lower melting points than their components. This unique property endows DES with several advantages over traditional electrolytes.

In lithium-ion batteries, DES-based electrolytes offer high ionic conductivity and excellent electrochemical stability, which are crucial for the efficient lithium ions transport between the anode and cathode during charge and discharge cycles. These properties enhance the battery performance, including higher energy densities, improved charge/discharge rates, and extended cycle life. Moreover, DES-based electrolytes exhibit a wider electrochemical window, making them suitable for high-voltage applications where traditional electrolytes might degrade.

Similarly, in sodium-ion batteries, DES can address some of the challenges associated with the larger ionic radius of sodium compared to lithium. The high ionic conductivity of DES facilitates efficient sodium ion transport, which can mitigate issues related to slower kinetics and lower energy densities typically seen in sodium-ion batteries. Additionally, the electrochemical stability of DES helps in maintaining the structural integrity of the battery components over prolonged cycling.

In redox flow batteries, DES-based electrolytes can enhance the solubility of active materials, leading to higher energy densities and better overall performance. The tunable nature of DES allows for the customization of electrolyte properties to optimize battery efficiency and longevity.

5.2 DES in electrode fabrication

The use of DES in electrode fabrication presents significant advantages in the synthesis and modification of battery electrodes. DES acts as an excellent solvent and co-solvent in the preparation of electrode materials, enabling the formation of uniform and high-capacity electrode structures.

During the synthesis of electrode materials, DES facilitates the uniform dispersion of active materials and binders, leading to the formation of homogenous and defect-free electrodes. This uniformity is critical for achieving high electrochemical performance and ensuring consistent battery operation. Furthermore, DES can aid in the formation

of nanostructured electrode materials, which offer larger surface areas and enhanced electrochemical reactivity, contributing to higher energy and power densities.

In the modification of existing electrode materials, DES can be used to introduce functional groups or dopants that enhance the electrochemical properties of the electrodes. For instance, DES can be employed to incorporate conductive polymers, carbon nanomaterials, or metal oxides into electrode structures, thereby improving conductivity, stability, and capacity retention.

5.3 DES for battery recycling

Battery recycling is a crucial aspect of sustainable battery technology, and DES has shown significant promise in this area. The selective dissolution and recovery capabilities of DES make them ideal for extracting valuable metals from spent batteries, contributing to a circular economy and reducing environmental impact.

DES can selectively dissolve metals such as lithium, cobalt, nickel, and manganese from battery cathodes, allowing for efficient separation and recovery. This selective dissolution is facilitated by the complexation properties of DES, which can form stable complexes with specific metal ions. The recovered metals can then be purified and reused in the new battery production, reducing the reliance on virgin raw materials and minimizing waste.

Moreover, the use of DES in battery recycling processes can reduce the need for harsh chemicals and energy-intensive procedures typically associated with conventional recycling methods. DES is often composed of non-toxic and biodegradable components, making them an environmentally friendly alternative for battery recycling.

6. Sustainability and environmental impact

The use of Deep Eutectic Solvents in batteries represents a significant advancement in pursuing sustainable and environmentally friendly energy storage solutions. This section explores the sustainability and environmental impact of DES in batteries, focusing on green chemistry, life cycle assessment, and economic viability.

6.1 Green chemistry

Deep Eutectic Solvents are synthesized from renewable and non-toxic components, making them an ideal choice in line with the principles of green chemistry. Traditional solvents used in battery production often pose significant environmental hazards, including toxicity and non-renewability. In contrast, DES are typically composed of natural substances like choline chloride and various hydrogen bond donors (e.g., urea, glycerol). These components are not only abundant but also biodegradable, reducing the environmental footprint from their inception.

The benign nature of DES helps in minimizing hazardous waste production and lowers the risks associated with handling and disposal. Furthermore, the synthesis of DES generally occurs at ambient temperatures and pressures, which reduces energy consumption during production. This contrasts with the high energy demands of producing conventional organic solvents, thereby contributing to a reduction in greenhouse gas emissions.

6.2 Life cycle assessment

Conducting a comprehensive life cycle assessment (LCA) of DES-based batteries provides a clearer picture of their environmental benefits. LCAs evaluate the environmental impact of the extraction of raw materials through production, usage, and disposal. For DES-based batteries, LCAs consistently show a lower environmental impact compared to conventional batteries.

Key findings from LCA studies indicate that the use of DES in batteries significantly reduces the carbon footprint. This reduction stems from lower energy requirements during solvent production and decreased emissions associated with material synthesis. Additionally, the biodegradability of DES ensures that post-use disposal has minimal adverse effects on ecosystems, contrasting sharply with the persistence of traditional solvents in the environment.

Moreover, the sustainable resource utilization of DES is evident in their ability to be synthesized from by-products of other industries. For example, the use of waste glycerol from biodiesel production as a component in DES aligns with

circular economy principles, ensuring that resources are reused and recycled effectively.

6.3 Economic viability

Economic viability is a critical factor in the widespread adoption of new technologies. DES offer cost-effectiveness both in production and application within batteries, making them a competitive alternative to traditional solvents. The raw materials for DES are generally inexpensive and widely available, which reduces the overall production costs.

In the context of battery manufacturing, DES can simplify the production process by eliminating the need for extensive purification steps required by conventional solvents. This simplification leads to lower operational costs and enhances production efficiency. Additionally, the long-term stability and performance of DES-based electrolytes can extend the lifespan of batteries, offering better value over time.

The economic benefits extend to potential reductions in regulatory and disposal costs. Given their non-toxic nature, DES-based batteries may face fewer regulatory hurdles related to hazardous materials, and their environmentally benign disposal could lower end-of-life management costs.

6.4 Market adoption and future prospects

The integration of DES in battery technology is poised to enhance market adoption of sustainable energy storage solutions. As the global demand for batteries continues to rise, particularly with the expansion of electric vehicles and renewable energy storage, DES-based batteries offer a promising path forward. Their combined environmental and economic advantages can drive a shift towards greener battery technologies.

Looking ahead, ongoing research and development efforts are likely to further optimize the performance of DES in batteries. Innovations in DES formulations and their applications could lead to even greater efficiencies and broader applicability across various types of batteries.

7. Future perspectives

7.1 Research and development

The future of batteries for sustainable energy storage lies in the continued research and development of DES. Ongoing studies are focused on optimizing DES formulations and exploring new combinations to enhance battery performance. Collaboration between academia and industry is crucial in accelerating the development and commercialization of DES-based batteries. By leveraging the expertise and resources of both sectors, researchers can more effectively address challenges such as electrolyte stability, ion conductivity, and cycling stability. Additionally, advancements in DES chemistry hold the potential to revolutionize battery design, leading to safer, more efficient, and environmentally friendly energy storage solutions.

7.2 Technological Integration

The successful integration of DES into existing battery technologies is essential for realizing their full potential in sustainable energy storage systems. However, this integration presents technical challenges related to compatibility, scalability, and performance optimization. To overcome these hurdles, ongoing efforts in materials science and engineering are focused on developing novel electrode materials, membrane separators, and cell designs tailored for DES-based batteries. By harnessing the latest advancements in nanotechnology, solid-state chemistry, and manufacturing processes, researchers aim to overcome technical barriers and enable the seamless integration of DES into various battery systems. This integration will not only enhance the energy density and efficiency of batteries but also improve their safety, reliability, and lifecycle sustainability.

7.3 Policy and regulation

The widespread adoption of DES-based batteries relies on supportive policies and regulations that incentivize

investment, innovation, and market deployment. Governments play a crucial role in shaping the regulatory landscape through initiatives such as research funding, tax incentives, and procurement mandates. By providing financial incentives for research and development, policymakers can stimulate innovation and accelerate the commercialization of DES-based battery technologies. Furthermore, the establishment of industry standards and certification programs ensures the safety, reliability, and interoperability of DES-based batteries across diverse applications and markets. By fostering a conducive regulatory environment, policymakers can promote the sustainable development of energy storage technologies and drive the transition towards a cleaner, more resilient energy future.

8. Conclusion

In conclusion, DES hold significant promise for revolutionizing the field of energy storage, particularly in the context of batteries. The unique properties of DES, such as their low cost, non-toxicity, high stability, and tunability, make them attractive candidates for various battery applications. Through this paper, we have explored the roles of DES in batteries for sustainable energy solutions.

Firstly, DES offer opportunities for enhancing battery performance. By serving as electrolytes or electrolyte additives, DES can improve the conductivity and stability of batteries, leading to enhanced energy density, cycling stability, and safety. Additionally, DES-based electrolytes can enable the use of high-voltage cathode materials and facilitate the development of next-generation battery technologies.

Moreover, DES contribute to sustainability in battery production and operation. The environmentally benign nature of DES, coupled with their abundance and low-cost synthesis from natural compounds, aligns with the principles of green chemistry. This not only reduces the environmental footprint of battery manufacturing but also addresses concerns related to resource depletion and toxic waste generation.

Furthermore, DES have the potential to enable the recycling and reuse of battery materials. Their ability to dissolve a wide range of metal salts and oxides makes them suitable for extracting valuable components from spent batteries, thus promoting a circular economy in the battery industry. This approach not only conserves resources but also reduces the reliance on raw materials and minimizes the environmental impact of battery disposal.

However, despite the numerous advantages offered by DES, several challenges and barriers remain to be addressed. These include the optimization of DES formulations for specific battery chemistries, the scale-up of DES production, and the development of efficient separation and purification methods for recycling applications. Additionally, regulatory frameworks and standards need to be established to ensure the safety and compatibility of DES-based battery technologies.

In conclusion, Deep Eutectic Solvents represent a promising avenue for enhancing the performance and sustainability of batteries. Their unique properties and environmental benefits position them as key contributors to the future of energy storage. Continued research and development, along with supportive policies, will pave the way for the widespread adoption of DES-based batteries, addressing both environmental and performance challenges in the quest for sustainable energy solutions.

Conflict of interest

The author declares no competing financial interest.

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