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2025 • 1

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ENHANCING HYDRONIUM ION MOBILITY IN GRAPHENE OXIDE-BASED PROTON EXCHANGE MEMBRANES

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Abstract. This study delves into the atomistic insights surrounding the use of task-specific Deep Eutectic Solvents (DES) to enhance hydronium ion mobility in Graphene Oxide (GO)-based Proton Exchange Membranes (PEMs). Employing advanced computational modeling techniques, the research meticulously examines the optimized structures and interaction energies of DES, hydronium ions, water molecules, GO, and their various complexes. The findings reveal a strategic alignment of hydronium ions and water molecules between the GO layers and DES, indicating robust interactions that significantly facilitate proton transport. Notably, DES demonstrates favorable energetic profiles, while the complex comprising GO, DES, water, and hydronium ions showcases markedly enhanced stability relative to their isolated counterparts. This enhanced stability underscores the efficacy of DES as potent additives for improving the performance of PEMs. The study's insights offer invaluable guidance for the rational design of task-specific solvents and the development of customized membranes. By highlighting the strong interactions and favorable energetics within these complexes, the research provides a foundational understanding that can be leveraged to optimize PEM performance, ultimately advancing the field of energy materials and fuel cell technologies.

Keywords: proton exchange membrane, graphene oxide, hydronium ion, mobility, density functional theory, quantum chemical calculations.

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ГРАФЕН ОКСИДІ НЕГІЗІНДЕГІ ПРОТОН АЛМАСУ МЕМБРАНАЛАРЫНДА ГИДРОНИЙ ИОНДАРЫНЫҢ ҚОЗГАЛҒЫШТЫҒЫН АРТТЫРУ

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Аннотация. Бұл зерттеу графен Оксиді (GO) негізіндеңі протон алмасу мембранныңда (PEMs) гидроний иондарының қозғалғыштығын арттыру үшін тапсырмаға тән терең әвтективалық еріткіштерді (DES) пайдалану туралы атомистік түсініктерді зерттейді.

Жетілдірілген есептеу модельдеу әдістерін қолдана отырып, зерттеу DES, гидроний иондарының, су молекулаларының, GO және олардың әртүрлі кешендерінің онтайланырылған құрылымдары мен өзара әрекеттесу энергияларын мұқият зерттейді. Нәтижелер гидроний иондары мен су молекулаларының арасындағы стратегиялық сәйкестікі, GO қабаттары мен DES протондардың тасымалдануын айтартықтай жөнілдететін сенімді өзара әрекеттесулерді көрсетеді.

Бір қызығы, DES қолайлы энергетикалық профильдерді көрсетеді, ал GO, DES, су және гидроний иондарынан тұратын кешен оқшауланған аналогтарымен салыстырғанда айтартықтай жақсартылған тұрақтылықты көрсетеді. Бұл жақсартылған тұрақтылық DES-тің PEMs өнімділігін арттыру үшін қоспалар ретіндегі тиімділігін көрсетеді. Зерттеу нәтижелері тапсырмаға арналған еріткіштерді ұтымды жобалау және теңшелген мембранның әзірлеу бойынша баға жетпес ұсыныстар береді. Осы кешендердегі өзара әрекеттесулер мен қолайлы энергетиканы көрсете отырып, зерттеу PEM өнімділігін онтайланыру үшін пайдаланылуы мүмкін іргелі түсініктерді қамтамасыз етеді, сایып келгенде, энергетикалық материалдар мен отын ұшықтары технологиялары саласын ілгерілетеді.

Түйін сөздер: протон алмасу мембранасы, графен оксиді, гидроний ионы, козғалғыштығы, тығыздықтың функционалдық теориясы, кванттық химиялық есептеулер.

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ПОВЫШЕНИЕ ПОДВИЖНОСТИ ИОНОВ ГИДРОНИЯ В ПРОТОНООБМЕННЫХ МЕМБРАНАХ НА ОСНОВЕ ОКСИДА ГРАФЕНА

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Аннотация. Это исследование раскрывает атомистические идеи, связанные с использованием специальных глубоких эвтектических растворителей (DES) для повышения подвижности ионов гидрония в протонообменных мембранах на основе оксида графена (GO). Используя передовые методы компьютерного моделирования, в ходе исследования были тщательно изучены оптимизированные структуры и энергии взаимодействия DES, ионов водорода, молекул воды, GO и их различных комплексов. Полученные данные свидетельствуют о стратегическом расположении ионов гидрония и молекул воды между слоями GO и DES, что указывает на устойчивые взаимодействия, которые значительно облегчают перенос протонов. Примечательно, что DES демонстрирует благоприятные энергетические характеристики, в то время как комплекс, состоящий из GO, DES, воды и ионов гидрония, демонстрирует заметно повышенную стабильность по сравнению с их изолированными аналогами. Эта повышенная стабильность подчеркивает эффективность DES в качестве мощных добавок для улучшения характеристик PEMS.

Результаты исследования являются бесценным руководством для рационального подбора растворителей для конкретных задач и разработки индивидуальных мембран. Подчеркивая тесное взаимодействие и благоприятную энергетику в этих комплексах, исследование дает фундаментальное понимание, которое может быть использовано для оптимизации производительности ПЭМ, что в конечном итоге приведет к развитию области энергетических материалов и технологий топливных элементов.

Ключевые слова: протонообменная мембрана, оксид графена, ион гидрония, подвижность, теория функционала плотности, квантово-химические расчеты.

Introduction

Fuel cells stand as a beacon of innovation in the realm of energy conversion, offering a highly efficient alternative to traditional combustion-based power generation methods. These electrochemical current sources facilitate the direct conversion of fuel and oxidizer energy into electrical energy, circumventing the inefficiencies inherent in combustion processes and boasting energy efficiency levels that can soar to remarkable heights, ranging from 70% to 90%. The unique design of fuel cells, featuring specialized porous electrodes (anode and cathode), catalyzed predominantly by palladium, enables the efficient conversion of chemical energy from hydrogen and oxygen into electrical energy, heat, and water (Jiao, et al, 2021). With their low emission footprint, fuel cells emerge as environmentally friendly energy generation solutions, emitting only water vapor and minimal amounts of carbon dioxide when pure hydrogen is utilized as the fuel.

Among the diverse array of fuel cell technologies, proton exchange membrane fuel cells (PEMFCs) have garnered significant attention due to their advantageous characteristics, including high proton ion conductivity, flexibility in fuel input, and compact design. Proton exchange membranes (PEMs), serving as the heart of PEMFCs, facilitate the transportation of protons through hydrophilic channels, thereby enabling efficient electrochemical reactions at the anode and cathode interfaces (Peighambardoust, et al, 2021; Wakizoe, et al, 1995; Sopian, et al, 2006; Le, et al, 2008; Cheddie, et al, 2005).

Graphene oxide (GO)-based proton exchange membranes emerge as promising candidates to address the aforementioned challenges, offering high efficiency in energy conversion and low ohmic resistance. In our previous work (Kemelbekova, 2023) we investigated the ion transport in poly(acrylic acid) and graphene oxide-based proton exchange membranes, which provided the foundation for the current study focused on enhancing membrane performance using deep eutectic solvents (DES). In addition to serving as efficient separators of electrochemical reactions, these membranes play a pivotal role in enhancing proton conductivity within fuel cells (Myrzabekova, 2014; Ren, 2020; Kahraman, 2024; Um, 2000; Luo, 2018; Boettner, 2002; Majlan, 2018). To further enhance the performance of PEMs, researchers have explored the incorporation of various solvents, including deep eutectic solvents (DESs), ionic liquids, and benzimidazole derivatives, as substitutes for water in facilitating proton transportation. DESs, in particular, represent a novel generation of ionic liquids characterized by their cost-effectiveness, environmentally friendly nature, and ease of synthesis.

The utilization of DESs in PEMs necessitates careful consideration of several factors, including thermal stability, viscosity, and the method of integration into the PEM matrix. Notably, DESs composed of ethylene glycol (EGL) and sodium halides exhibit favorable proton conductivity ranges for PEMFC applications (Janssen, 2001; Sivertsen, 2005; Pandey, 2017; He, 2014; Huang, 2018; Peng, 2016). However, gaining insights into the formation mechanisms of DESs and their structural organization within

GO-based PEMs at the molecular level is imperative to optimize fuel cell operations and develop PEMs with enhanced functional characteristics.

Computer modeling and simulations emerge as indispensable tools for elucidating the formation mechanisms of DESs and their applications in PEMs for hydronium ion transportation. By leveraging computational techniques, researchers can expedite the exploration of DES candidates with desirable properties, thereby reducing both time and costs associated with experimental endeavors (Xu, 2023; Wong, 2018; Zhang, 2019; Cheddie, 2005; Moreira, 2009). Furthermore, Density Functional Theory (DFT) calculations offer a platform for gaining detailed insights into the formation and applications of DESs in PEMs at the molecular level, paving the way for the development of innovative fuel cell technologies with enhanced efficiency and sustainability.

In this work, we delve into the atomistic insights into task-specific deep eutectic solvents, focusing on their role in enhancing hydronium ion mobility in graphene oxide-based proton exchange membranes for fuel cell applications. Through computational modeling and simulations, coupled with experimental validation, we aim to unravel the intricate molecular mechanisms underlying the formation and functionality of task-specific DESs within PEMs, thereby advancing the frontier of fuel cell technology towards greater efficiency and sustainability.

Materials and Methods

The investigation into the quantum chemical and structural properties of task-specific deep eutectic solvents (DESs) and their interaction with graphene oxide-based proton exchange membranes (PEMs) was conducted using computational simulations. Herein, the representative model of DESs comprised a mixture of choline chloride and urea in a 1:2 ratio. At the same time, PEM model consisted of graphene oxide (GO), water, and hydronium ions.

In this study, DFT calculations were employed to investigate the interactions between choline chloride, urea, and their mixture with hydronium ions in the presence of graphene oxide. The software Gaussian09 Rev. E.01, utilizing the PM6 method, was utilized for these calculations, which were further visualized using the GaussView 5.0 program package.

The first step in the computational analysis involved the optimization of initial structures. Pure choline chloride, pure urea, and the task-specific deep eutectic solvent (DES) were subjected to DFT calculations to obtain their optimized structures. Subsequently, complexes involving choline chloride, urea, and DES with hydronium ions and graphene oxide were generated and further optimized.

Following the structure optimization, various parameters were calculated to characterize the complexes. Additionally, frequency calculations were performed to verify the stability of the optimized structures. The absence of negative frequencies indicates that the structures correspond to the global energy minimum, ensuring their reliability for further analysis.

The structural and energetic parameters obtained from the DFT calculations were then analyzed to gain a deeper understanding of the quantum chemical and structural properties of the DESs and their interactions with the PEMs (Proton Exchange Membranes).

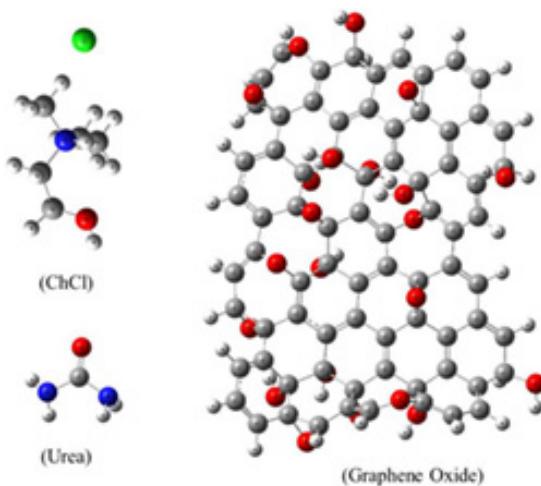


Fig 1. 3D representation of ChCl, Urea, and Graphene Oxide

To aid in the visualization and interpretation of the computational results, Figure 1 was generated to represent all the optimized structures and complexes. Visual representation of the molecular structures offers an intuitive understanding of their spatial arrangement and interactions, facilitating the interpretation of the computational data.

Results and Discussions

The study delves into the structural optimization and orbital energies of Choline Chloride (ChCl), Urea, and Deep Eutectic Solvent (DES) formed by their combination. Through molecular modeling, the researchers explored the configurations and electronic properties of these compounds, shedding light on their potential applications in various fields. This analysis aims to dissect the implications of the findings and their significance in the realm of chemical research and application.

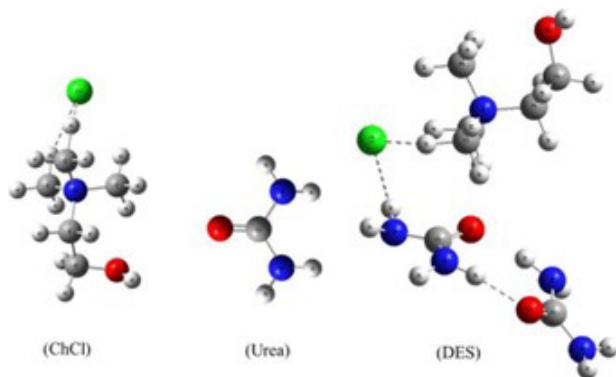


Fig 2. Optimized structures of ChCl, Urea, and DES

Table 1 – Energies of ChCl, Urea, and DES

	ChCl	Urea	DES
Energy (kJ/mol)	-277.87	-205.79	-791.73

The structural optimization of molecules such as ChCl (choline chloride), urea, and DES (deep eutectic solvent) through computational methods offers invaluable insights into their preferred configurations and the intricate network of intermolecular interactions governing their behavior. In Figure 2, a detailed depiction of these optimized structures reveals the spatial arrangement of atoms and the formation of molecular complexes. Notably, in the case of ChCl, the chloride ion tends to position itself proximal to the nitrogen atom of choline. This arrangement suggests a favorable interaction between the chloride ion and the choline moiety, likely facilitated by electrostatic forces and hydrogen bonding.

Upon the addition of two urea molecules to ChCl, the formation of DES occurs. In this complex, the chloride ion assumes a pivotal role, sandwiched between the nitrogen atom of choline and one urea molecule. Simultaneously, the second urea molecule interacts with the first one, forming a cohesive molecular structure. This arrangement underscores the intricate nature of DES formation, where multiple molecules coordinate to create a stable solvent environment.

To quantitatively assess the stability of these molecular species, optimized energies are computed and presented in Table 1. The energy associated with a single ChCl molecule is determined to be -277.87 kJ/mol, reflecting the stability of this individual component within the system. Similarly, the energy of a lone urea molecule is calculated to be -205.79 kJ/mol, indicating its inherent stability in isolation.

However, the most intriguing observation arises from the formation of DES. When one ChCl molecule associates with two urea molecules to form the DES complex, a significant decrease in energy is noted, resulting in an energy value of -791.73 kJ/mol. This substantial reduction in energy compared to the sum of individual components underscores the remarkable stability achieved within the DES structure. The emergence of such a stable configuration highlights the synergistic effects arising from the interactions between ChCl and urea molecules. These interactions likely involve a combination of hydrogen bonding, electrostatic forces, and van der Waals interactions, which collectively contribute to the enhanced stability of the DES complex.

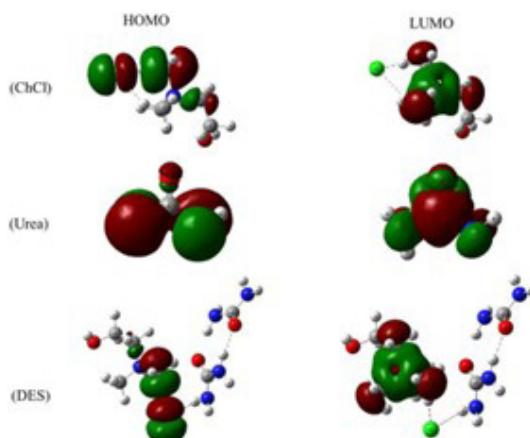


Fig 3. HOMO-LUMO orbitals of ChCl, Urea, and DES

The analysis of Highest Occupied Molecular Orbital (HOMO) and Lowest Unoccupied Molecular Orbital (LUMO) energies provides crucial information about the electronic structure and reactivity of the molecules (Figure 3). In ChCl, the HOMO is observed at -8.41 eV, primarily populated around the chloride ion, indicating its involvement in electron donation or sharing. Conversely, the LUMO at 0.30 eV is populated around the nitrogen, carbon, and carboxy groups of choline, suggesting potential sites for electron acceptance or interaction. Urea exhibits different orbital characteristics, with a HOMO at -10.66 eV, predominantly populated around nitrogen atoms and carbons. This suggests its potential as an electron donor. The LUMO at 1.00 eV is distributed around nitrogen, carbon, and oxygen atoms, indicating its ability to accept electrons. Upon the formation of DES, changes in orbital energies reflect the altered electronic interactions within the system. The HOMO of DES is found to be -8.65 eV, with significant population around the chloride ion, indicating its involvement in the electron-sharing process. The LUMO at 0.046 eV is primarily populated around choline, suggesting its role in electron acceptance.

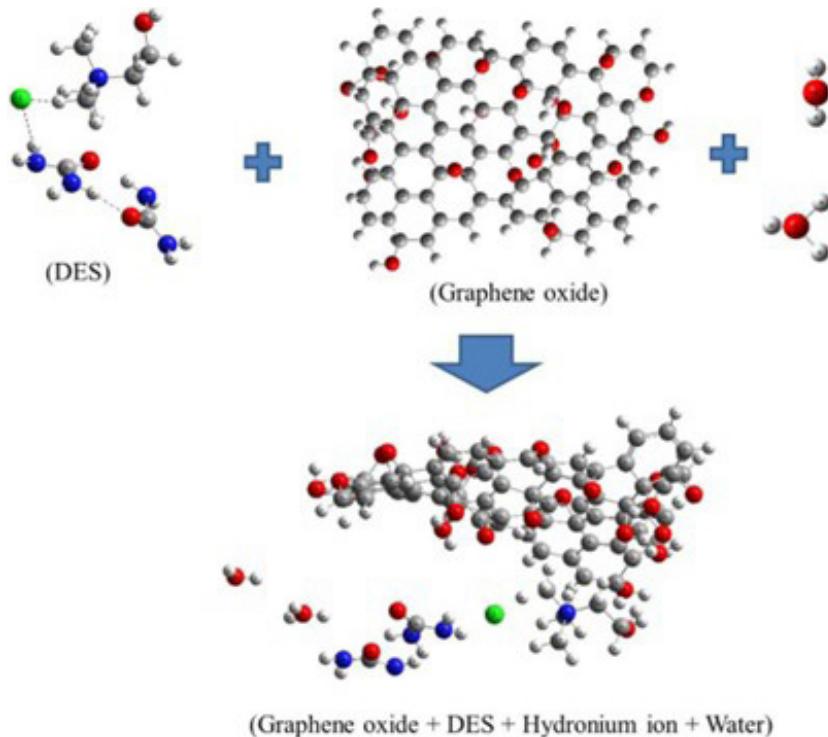


Fig 4. Optimized structures of DES, hydronium ion, water, GO, and GO+DES complex in the presence of hydronium ion and water

The study delves into the atomistic insights of task-specific Deep Eutectic Solvents (DES) and their role in enhancing hydronium ion mobility in Graphene Oxide-based Proton Exchange Membranes (PEMs) (Figure 4). Through computational modeling, the researchers investigated the optimized structures and energies of DES, hydronium

ion, water, Graphene Oxide (GO), and their complexes. This analysis aims to elucidate the molecular interactions and energy landscapes within these systems, shedding light on their potential applications in PEM technology. The figures presented illustrate the optimized structures of DES, hydronium ion, water, GO, and the GO+DES complex in the presence of hydronium ion and water. It is observed that hydronium ions and water molecules are strategically positioned between GO and DES molecules, particularly near oxygen or carboxy group sides. This arrangement suggests strong interactions between the components, potentially facilitating proton transport and enhancing membrane conductivity.

Table 2: Energies of DES, Hydronium ion, water, GO, and GO+DES complex in the

	DES	Water	Hydronium	GO	GO+DES+Hydronium+Water
Energy (kJ/mol)	-791.73	-226.86	-268.61	-823.76	-1491.91

The optimized energies obtained from computational simulations offer profound insights into the stability and energetics of the molecular systems under investigation, as illustrated in Table 2. One of the key findings is that DES (Deep Eutectic Solvent) demonstrates a notably favorable energy of -791.73 kJ/mol, suggesting a configuration that is highly stable. This insight is crucial for understanding the behavior of DES in various applications where stability is a critical factor. Similarly, the energies associated with water and the hydronium ion are determined to be -226.86 kJ/mol and -268.61 kJ/mol, respectively. These values indicate the stable presence of both water and the hydronium ion within the molecular system, providing essential information for studies related to aqueous environments and electrolyte solutions.

On the other hand, graphene oxide (GO) exhibits a relatively high energy of -823.76 kJ/mol. This higher energy level is indicative of the complex structure and intricate interactions within GO. Understanding the energetics of GO is crucial due to its wide-ranging applications in fields such as electronics, energy storage, and biomedicine. The relatively high energy of GO underscores the need for careful consideration of its behavior and interactions in various environments.

Of particular interest is the energy of the complex formed by GO, DES, water, and the hydronium ion, which is significantly lower at -1491.91 kJ/mol compared to the sum of the energies of the individual components. This observation highlights the presence of strong synergistic effects within the complex, resulting in enhanced stability. The substantial decrease in energy suggests that the interactions between GO, DES, water, and the hydronium ion are highly favorable, leading to a more stable configuration than what would be expected from the mere sum of individual energies.

Conclusion

Our study provides valuable insights into the role of task-specific Deep Eutectic Solvents (DES) in enhancing hydronium ion mobility within Graphene Oxide (GO)-based Proton Exchange Membranes (PEMs). The optimized structures and energetic landscape elucidate the strategic positioning of hydronium ions and water molecules between GO and DES, indicating strong molecular interactions conducive to proton

transport. Moreover, the favorable energetics of DES and the significant decrease in energy observed in the GO+DES+water+hydronium ion complex underscore the synergistic effects and enhanced stability imparted by the incorporation of DES into PEMs.

These findings hold promising implications for the advancement of PEM technology, particularly in applications such as fuel cells and water electrolysis, where proton conductivity and membrane stability are critical. By leveraging task-specific DES as additives, researchers can effectively tailor PEM properties to meet specific performance requirements, thus accelerating the development of next-generation proton exchange membranes.

Moving forward, further experimental validations and performance assessments are warranted to corroborate the computational predictions and elucidate the practical implications of DES-enhanced PEMs. Additionally, continued research into the design and optimization of task-specific solvents and membrane materials will be instrumental in realizing the full potential of DES-based approaches for PEM technology. Overall, this study contributes to the ongoing efforts in advancing proton exchange membrane systems for sustainable energy and environmental applications.

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