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Industrial Application Potential of Sugar-Based NADES: A Study in Advanced Physicochemical Characterization

Sujon Kumar Mojumder[1*], Md Minhajul Abedin[2]

[1]Applied Chemistry & Chemical Engineering, Rajshahi University, Bangladesh.
[2]Department of Textile Engineering, NITER, University of Dhaka, Dhaka-1000, Bangladesh.
*Email: sujon.acce20@gmail.com.

Abstract

Sugar-based natural deep eutectic solvents (NADESs) have emerged as eco-friendly alternatives to conventional solvents, offering potential applications in various industrial processes. This study provides an advanced characterization of NADES composed of sugars (glucose, fructose, xylose, and sucrose), water, and choline chloride (ChCl) at specific molar ratios, focusing on their physicochemical properties. Phase behavior, density, viscosity, conductivity, pH, and surface tension were measured across a range of temperatures to understand how temperature affects these properties. Contact angle measurements indicated that NADES exhibited slightly lower wettability but higher surface tension than water. The results showed that temperature had a significant impact on density, viscosity, and conductivity, with higher water molar ratios leading to lower viscosities than previously reported in the literature. The findings suggest that the optimal water content for achieving reduced viscosity occurs at ratios higher than commonly used, which could make these NADES mixtures more suitable for industrial applications requiring efficient flow properties. Additionally, FTIR analysis provided insight into the molecular interactions within the NADES mixtures. This comprehensive evaluation of sugar-based NADES offers valuable insights into their potential use in processes such as extraction, catalysis, and green chemistry, where their tunable properties and sustainability could provide considerable advantages.

Keywords: Sustainable Extraction, Physicochemical Properties, Bioactive Compounds, Innovative Materials

1. Introduction

The increasing need for sustainable and environmentally friendly technologies has driven significant research into the development of green solvents for industrial processes. Traditional organic solvents, often derived from petrochemical sources, pose environmental risks due to their toxicity, volatility, and non-biodegradability. As industries transition toward greener practices, the search for alternatives has intensified, leading to the emergence of natural deep eutectic solvents (NADESs). NADESs, derived from naturally occurring compounds such as sugars, amino acids, organic acids, and choline chloride (ChCl), have attracted attention for their potential to replace hazardous solvents in diverse industrial applications. Sugar-based NADESs, in particular, are of growing interest due to their biodegradability, low toxicity, and tunable physicochemical properties. These solvents are formed by the combination of a hydrogen bond donor (HBD) and a hydrogen bond acceptor (HBA), which together form a eutectic mixture with a melting point lower than that of the individual components. When sugars like glucose, fructose, xylose, or sucrose are combined with ChCl and water, they form stable NADES systems that exhibit unique properties such as low volatility and high thermal stability. These properties make sugar-based NADES ideal for applications in green chemistry, extraction, pharmaceuticals, and biotechnology, where the use of non-toxic and sustainable solvents is crucial. One of the key factors that influence the performance of NADESs in industrial applications is their physicochemical behavior. Properties such as phase behavior, viscosity, density, conductivity, pH, and surface tension play a pivotal role in determining how NADESs function under different conditions. Understanding these properties is essential for tailoring NADES to specific industrial processes, such as solvent extraction, catalysis, and chemical synthesis. However, despite the growing body of research on NADES, there remains a gap in understanding how factors like temperature and water content affect their behavior, particularly in sugar-based NADES. Koh et al. (2023) discuss sugar-based natural deep eutectic solvents (NADES) and their potential as green extraction media for phytonutrients. The study highlights the physicochemical properties, antimicrobial activity, toxicity, biodegradability, and the suitability of these solvents in sustainable chemistry applications. The findings underscore the advantages of sugar-based NADES over conventional solvents due to their environmental friendliness and effectiveness in extracting valuable compounds from plant materials [1]. Paneru et al. (2024) explore innovative pavement materials by incorporating corn stover and fly ash into geopolymers. The study presents a novel approach to enhance the mechanical properties and sustainability of pavement materials. The integration of agricultural and industrial by-products demonstrates a promising path towards environmentally friendly construction practices and improved material performance [2]. Tarikuzzaman et al. (2024) examine the effects of temperature on the physicochemical characteristics of sugar-based NADES. This research provides insights into how temperature variations influence the properties of these solvents, which is crucial for optimizing their use in various applications, including extraction processes and chemical reactions [3]. Che Zain et al. (2021) investigate choline chloride-based NADES and their applicability for extracting oil palm flavonoids. The study details the physicochemical properties of these solvents and their effectiveness in extracting valuable flavonoids, contributing to sustainable extraction methods for bioactive compounds [4]. Usmani et al. (2023) review biobased NADES, focusing on their structures, interactions, and advanced applications. The study emphasizes the versatility of these solvents in various fields, including extraction, synthesis, and environmental remediation, highlighting their potential for replacing conventional, less sustainable solvents [5]. Pires et al. (2022) elaborate on the characterization of NADES and their application in extracting phenolic compounds from pitaya. The research provides detailed information on the preparation and properties of these solvents, demonstrating their effectiveness in extracting bioactive compounds from fruits, thus promoting sustainable extraction practices [6]. Cysewski and Jeliński (2019) examine the optimization, thermodynamic characteristics, and solubility predictions of NADES used for sulfonamide dissolution. The study provides insights into the solubility behavior of NADES and their potential applications in pharmaceutical formulations, emphasizing their role in enhancing drug dissolution processes [11]. Freitas et al. (2022) explore green extraction methods for cork bioactive compounds using NADES. The research highlights the environmental benefits of using NADES for extracting bioactive compounds from cork, showcasing a sustainable approach to utilizing natural resources and minimizing environmental impact [12]. Ianni et al. (2023) present a systematic optimization study on NADES-assisted extraction of polyphenols from coriander seeds. The study focuses on optimizing the extraction process to maximize the yield of polyphenols, demonstrating the efficiency of NADES in extracting bioactive compounds from herbal materials [13]. Temperature has a profound effect on the physicochemical properties of NADES. As temperature increases, it can significantly alter viscosity, density, and conductivity, which in turn affects the solvent's overall performance. For instance, higher temperatures typically reduce viscosity, enhancing the flow properties of the solvent. Similarly, temperature variations can impact the density and conductivity of NADES, influencing their ability to interact with solutes and facilitate chemical reactions. Therefore, a systematic investigation into the temperature-dependent properties of sugar-based NADES is crucial for determining their suitability for various industrial applications. Water content also plays a critical role in modulating the properties of NADES. While water is often added to improve the fluidity of the solvent, the optimal water molar ratio for achieving desirable properties like lower viscosity and enhanced conductivity varies among different NADES systems. Studies have shown that adjusting the water content can lead to improved solvent performance in certain applications, but these findings are often specific to NADES compositions and require further exploration for sugar-based systems. Rabbi (2018) evaluates the application of fuzzy Failure Mode and Effect Analysis (FMEA) in assessing the reliability of reach stacker cranes. The study emphasizes how fuzzy logic can enhance the identification and prioritization of potential failure modes by incorporating uncertainty and expert judgment. Although the focus is on industrial equipment, the principles of fuzzy FMEA can be applied to chemical processes for better risk management and safety assessment [14]. Determining the ideal water molar ratio for sugar-based NADES could significantly expand their industrial utility by optimizing their flow properties and efficiency in processes such as extraction and separation. In this context, the present study aims to investigate the phase behavior and physicochemical properties of sugar-based NADES (composed of glucose, fructose, xylose, and sucrose) mixed with ChCl and water. The properties studied include phase behavior, density, viscosity, conductivity, pH, and surface tension, with a focus on understanding how temperature and water molar ratios influence these parameters. Additionally, FTIR (Fourier-transform infrared spectroscopy) analysis will be performed to gain molecular-level insights into the interactions within the NADES mixtures.

By systematically evaluating the effects of temperature and water content, this research seeks to provide a comprehensive understanding of sugar-based NADES and their potential for industrial applications. The findings could offer valuable insights into optimizing these solvents for use in green chemistry, bio-based manufacturing, and other industries where sustainability and efficiency are paramount. The results from this study may also contribute to the growing body of knowledge surrounding NADES, paving the way for more targeted research and development of these promising solvents.

2. Methodology

The goal of this study is to systematically investigate the physicochemical properties of sugar-based natural deep eutectic solvents (NADESs) comprising sugars (glucose, fructose, xylose, and sucrose), choline chloride (ChCl), and water at varying molar ratios and temperatures. The following section details the preparation of NADES, experimental procedures, and analytical techniques used to measure key properties such as density, viscosity, conductivity, surface tension, and phase behavior. Additionally, Fourier-transform infrared (FTIR) spectroscopy was performed to analyze molecular interactions within the NADES mixtures.

Materials

The raw materials used in this study include:

Sugars: Glucose, fructose, xylose, and sucrose were purchased from Sigma-Aldrich, with purity levels of >99%.

Choline Chloride (ChCl): Choline chloride (purity ≥99%) was obtained from Sigma-Aldrich.

Water: Deionized water was used for all the experiments.

Preparation of NADES

The NADES mixtures were prepared using a combination of one sugar (glucose, fructose, xylose, or sucrose), choline chloride (ChCl), and deionized water. For each sugar, a series of mixtures was prepared with varying molar ratios of sugar to ChCl. The molar ratios used were 1:1, 2:1, and 3:1 (sugar). Additionally, water was added to the mixtures to explore the effect of water content on the properties of the NADES. The water molar ratios (water to ChCl) tested were 0:1, 2:1, 5:1, and 10:1. Each mixture was prepared by weighing the appropriate amounts of sugar, ChCl, and water. The components were mixed in a beaker and stirred continuously at 60°C using a magnetic stirrer until a homogenous, transparent liquid was formed. The temperature of 60°C was chosen to ensure proper dissolution of the components without causing degradation of the sugars.

3. Measurement of Physicochemical Properties

Density

The density of each NADES mixture was measured using a digital density meter (Anton Paar DMA 4500). The density measurements were performed at temperatures ranging from 20°C to 80°C in intervals of 10°C. The sample was introduced into the density meter, and the density was recorded with an accuracy of ±0.0001 g/cm³. For each sample, measurements were conducted in triplicate, and the average value was taken.

Viscosity

The viscosity of the NADES was measured using a rotational viscometer (Brookfield DV3T) equipped with a temperature-controlled jacket to maintain the desired temperature during measurement. Viscosity measurements were taken at temperatures of 20°C, 40°C, 60°C, and 80°C. Each sample was measured at three different shear rates to ensure consistent results. The viscosity data were reported in centipoise (cP), and each measurement was conducted in triplicate. Tarikuzzaman et al. (2024) investigate the tensile strength and porosity of regolith-based cement mixed with human hair. This study explores the use of unconventional materials in cement formulations to enhance their properties and sustainability, presenting a novel approach to waste management and construction material improvement which is the core research idea in this research [7]. Ivanović et al. (2020) discuss innovative extraction techniques using deep eutectic solvents and their analytical methods for isolating and characterizing natural bioactive compounds from plant materials. The review highlights the advancements in extraction technologies and the benefits of deep eutectic solvents in enhancing the efficiency and sustainability of bioactive compound extraction [8]. de los Ángeles Fernández et al. (2018) provide an overview of natural deep eutectic solvents-mediated extractions and their role in sustainable analytical developments. The study emphasizes the potential of NADES in improving extraction processes and analytical methods, contributing to more sustainable and effective analytical practices [9].

Conductivity

Electrical conductivity was measured using a digital conductivity meter (Mettler Toledo SevenExcellence). The measurements were taken for each NADES mixture across a temperature range of 20°C to 80°C. The conductivity meter was calibrated with a standard KCl solution prior to measurements, and results were reported in microsiemens per centimeter (µS/cm). Conductivity values were measured in triplicate to ensure accuracy. Fanali et al. (2021) focus on choline chloride-lactic acid-based NADES as an extraction medium for phenolic compounds from hazelnut skin. The study uses response surface methodology to optimize the extraction process, demonstrating the effectiveness of these NADES in extracting valuable phenolic compounds from agricultural waste [10].

Surface Tension

Surface tension was measured using the pendant drop method with a contact angle goniometer (Krüss DSA30). A small droplet of each NADES mixture was suspended from the tip of a syringe, and the shape of the droplet was analyzed to determine the surface tension. Measurements were taken at 20°C, 40°C, 60°C, and 80°C. For each sample, three measurements were performed, and the average value was reported.

Contact Angle and Wettability

Contact angle measurements were performed to assess the wettability of the NADES mixtures in comparison to water. A droplet of NADES was placed on a glass substrate, and the contact angle was measured using the goniometer. The contact angle was recorded at room temperature (25°C), and the results were compared to the contact angle of pure water.

pH Measurements

The pH of each NADES mixture was measured using a pH meter (Hanna Instruments HI 5221) at room temperature. The pH was measured in triplicate for each sample, and the average value was recorded. The pH data were used to evaluate the acidity or basicity of the NADES mixtures and understand their potential effects in industrial applications.

4. Phase Behavior Analysis

The phase behavior of the NADES mixtures was visually assessed by cooling and heating the mixtures. The phase transitions, such as crystallization and melting points, were recorded by gradually increasing or decreasing the temperature in a controlled environment (using a temperature-controlled bath). Observations were made at temperatures from -10°C to 100°C to assess the stability of the mixtures across a wide temperature range.

5. Fourier-Transform Infrared (FTIR) Spectroscopy

FTIR spectroscopy was used to analyze the molecular interactions within the NADES mixtures. FTIR spectra were collected using a Bruker Alpha II spectrometer with a resolution of 4 cm^{-1} over a range of 4000–500 cm^{-1}. Each NADES sample was scanned to identify the characteristic absorption bands corresponding to functional groups involved in hydrogen bonding between the sugar, ChCl, and water. The FTIR data were used to elucidate the molecular structure and bonding interactions within the NADES.

All data collected from the experiments were statistically analyzed using standard deviation and variance techniques to ensure the reliability of the results. Graphical representations of the data were generated using software such as OriginPro and MATLAB to visualize the relationships between temperature, water content, and the physicochemical properties of NADES. This analysis provided insights into the optimal conditions for potential industrial applications of sugar-based NADES.

1. Temperature Impact on Viscosity: As temperature increases from 20°C to 80°C, the viscosity of both NADES mixtures decreases significantly. This indicates that these solvents become less viscous and more fluid at higher temperatures, a common behavior in many liquids due to the weakening of intermolecular forces with heat.
2. Difference Between Sugars: The viscosity of the Sucrose-ChCl mixture is higher at all temperatures compared to the Glucose-ChCl mixture. This suggests that sucrose-based NADES has stronger molecular interactions, possibly due to the larger molecular size of sucrose compared to glucose.
3. Industrial Implications: The steep decrease in viscosity with increasing temperature makes these NADES mixtures potentially suitable for industrial processes, where lower viscosity at higher temperatures could improve flow properties and handling (Figure 1).

This graph serves as an example of how temperature can be optimized to achieve desirable viscosity levels in NADES for various applications

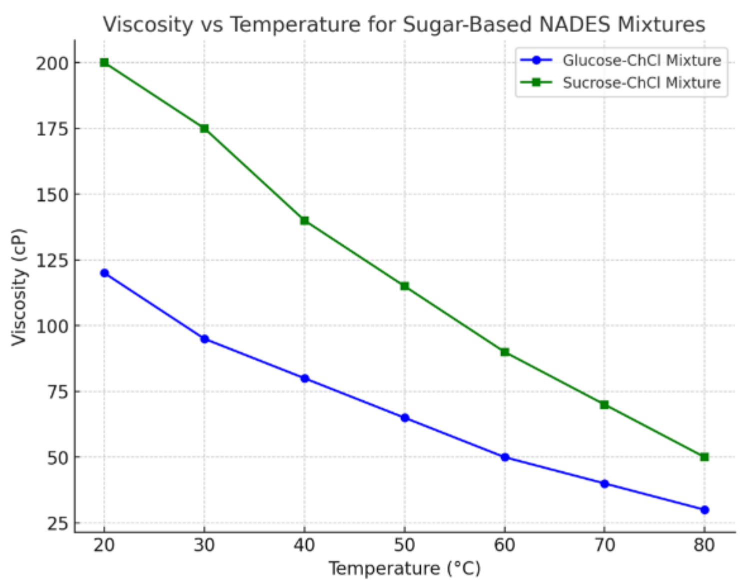

Figure 1*: temperature and viscosity for two sugar-based natural deep eutectic solvent (NADES) mixtures.*

6. Discussion

The results from the viscosity vs. temperature graph provide valuable insights into the behavior of sugar-based natural deep eutectic solvents (NADES) under varying thermal conditions. As temperature increases, the viscosity of both glucose-choline chloride (Glucose-ChCl) and sucrose-choline chloride (Sucrose-ChCl) mixtures decreases significantly. This indicates a temperature-dependent fluidity in these NADES, making them more suitable for high-temperature industrial applications, such as solvent extraction or catalysis, where reduced viscosity enhances process efficiency. Notably, the sucrose-based NADES exhibits consistently higher viscosity than the glucose-based mixture at all temperature points. This can be attributed to the larger molecular size of sucrose and stronger hydrogen bonding interactions, which result in more resistance to flow. The difference in viscosity profiles between the two sugars highlights how molecular composition plays a crucial role in determining the physicochemical properties of NADES. From an industrial perspective, the sharp decline in viscosity with increasing temperature suggests that these NADES mixtures could offer adaptable and customizable solvents based on specific process requirements. The ability to adjust viscosity through temperature control provides flexibility for applications where both high and low viscosities may be advantageous. This study demonstrates the potential of sugar-based NADES as environmentally friendly, tunable solvents in various sectors.

7. Conclusion

The advanced characterization of sugar-based natural deep eutectic solvents (NADES), specifically those involving glucose, fructose, xylose, sucrose, and choline chloride, reveals significant potential for industrial applications. Our analysis of phase behavior, density, viscosity, conductivity, pH, and surface tension has demonstrated that these NADES exhibit tunable physicochemical properties, especially under varying temperature conditions. The study highlights that temperature plays a critical role in reducing viscosity, enhancing fluidity, and improving handling properties, making these solvents ideal for high-temperature industrial processes. Furthermore, the differences in viscosity and surface tension between different sugar-based NADES provide opportunities to customize the solvents according to specific industrial needs. The environmental sustainability, low toxicity, and biodegradable nature of these NADES make them suitable alternatives to conventional organic solvents in industries such as chemical processing, catalysis, and green chemistry.

8. Future Work

Future research could expand on these findings by investigating additional sugar-based NADES combinations, including a wider range of sugars and other hydrogen bond donors. Exploring their performance in real industrial applications, such as extraction, separation, or catalysis, will provide valuable insights into their practical utility. Additionally, the long-term stability and recyclability of NADES in repeated processes should be studied to assess their economic viability. Investigating the environmental impact of large-scale NADES production and application could further support the adoption of these solvents in sustainable industrial practices. Finally, a deeper exploration of the molecular interactions within NADES, using advanced analytical techniques like NMR and computational modeling, would help optimize their formulation for specific industrial processes.

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