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Review Article

Deep Eutectic Solvents: A Review of Syntheses, Properties, and their Applications in Different Fields

Amin Zobeid[1], **Fatemeh Abbasi**[2*], **and Sedigheh Azadi**[2]

[1]Department of Engineering, Dezful Branch Islamic Azad University, Dezful 6461645169, Iran
[2]Chemistry Department, College of Sciences, Shiraz University, Shiraz 71946-84795, Iran

Abstract

Organic solvents have been the focus of numerous restrictions and legislation recently because of their detrimental impact on the environment and toxicity to human health. In parallel, deep eutectic solvents (DESs) have emerged as more resilient and eco-friendly solvents and have a wide range of physicochemical benefits associated with their affordability and durability. On the other hand, DESs have other major advantages such as low toxicity, high availability, low flammability, high recyclability, and low volatility from an environmental and technological standpoint, which is why DESs have become a viable substitute for conventional organic solvents over the past ten years. The number of structural combinations encompassed by DESs is tremendous; thus, it is possible to design an optimal DES for each specific enzymatic reaction system. A DES can be used as a solvent or co-solvent, an extractive reagent for an enzymatic product, or a solvent for enzymatic biomass pretreatment in (bio) catalytic processes. In this review, we have attempted to first provide a simple definition of DESs. Then, we have examined the classification of DESs, their preparation techniques, and their description.

Keywords: deep eutectic solvents, green solvents, environmental impact, physicochemical advantages, traditional organic solvents, solvent/co-solvent

Corresponding Author: Fatemeh Abbasi; email: abasi.fatemeh123@gmail.com

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1. Introduction

Organic solvents are chemicals that dissolve other compounds to create a solution, which is a homogenous combination. Usually composed of carbon, they can be either polar or non-polar, which affects how well they dissolve certain kinds of solutes. From chemical synthesis and extraction procedures to cleaning and surface treatment, organic solvents are crucial in a wide range of industrial, medicinal, and laboratory applications. Acetone, ethanol, toluene, chloroform, and methanol are typical examples of organic solvents. We shall go over the benefits and drawbacks of organic solvents below [1, 2].

One of the main benefits of organic solvents is their ability to widespread dissolution of a variety of chemicals. Because of their versatility, they are essential to a wide range of industrial and chemical operations. While non-polar solvents like hexane or toluene can dissolve non-polar molecules like oils and fats, polar solvents like water or methanol are effective in dissolving ionic or polar substances. Because of their versatility, organic solvents are important in a wide range of industries, such as materials science, petrochemicals, and pharmaceuticals [3, 4]. Hence, the efficiency of chemical reactions is frequently increased by the use of organic solvents. They can increase the rate of reaction or the yield of the intended product by offering a medium for reactants to interact. Solvents, for instance, can stabilize intermediates in complex processes, reduce activation energy, or help reactants dissociate [5-7].

Many different industries frequently employ organic solvents for some purposes like pharmaceuticals, cleaning, and extraction. Dissolving active components in medication compositions is a pharmaceutical process [8, 9]. Also, to create smooth finishes, solvents aid in the dissolution of pigments and resins [10]. Some solvents such as acetone and isopropanol are good solvents for cleaning surfaces and getting rid of residues like grease [11, 12]. Oil extraction and the separation of valuable compounds from raw materials are two examples of procedures that require solvents [13, 14]. The ease of availability, portability, and storage of many organic solvents, as well as their reasonable price, have made them suitable for commercial use [15].

The potential toxicity of conventional organic solvents to both humans and animals is one of their main problems. When consumed, absorbed via the skin, or inhaled, many organic solvents are toxic. Prolonged exposure to some solvents can cause major health concerns, including cancer, respiratory disorders, and damage to the liver and kidneys. Solvents like benzene and chloroform, for instance, are recognized carcinogens; thus, using them without the right safety precautions is dangerous [16].

Organic solvents can have a detrimental effect on the environment. The formation of ground-level ozone, a major component of smog, and air pollution are both greatly influenced by organic solvents, particularly volatile ones. Solvents can release volatile organic compounds (VOCs) into the atmosphere. Furthermore, if spilled or disposed of incorrectly, many of them cause long-term environmental contamination because they are not biodegradable [17]. Also, these solvents can be flammable. Some organic solvents, such as ether, acetone, and ethanol, are extremely volatile and provide a significant risk of explosion or fire, especially in industrial settings. These solvents need to be handled and stored with extreme caution to avoid accidents [18, 19]. Another disadvantage of these solvents is that they are non-renewable. The majority of conventional organic solvents are derived from petroleum-based resources, which are non-renewable. This dependence on fossil fuels raises the carbon footprint of solvent manufacture and aids in the depletion of natural resources. Furthermore, the extraction and refinement procedures used to create petroleum-based solvents can have a big influence on the environment [20].

In summary, organic solvents have many disadvantages, such as toxicity, environmental contamination, and flammability, even if their solubility, adaptability, and capacity to speed up chemical reactions make them indispensable in a variety of applications. These difficulties have raised interest in safer, more environmentally friendly substitutes, such as Ionic Liquids (ILs), which seek to allay a number of the issues with conventional organic solvents [21, 22].

2. ILs: Definition, Properties, and Applications

An ionic liquid (IL) is a salt that is in a liquid state at or near room temperature. The term “ionic liquid” refers to a class of compounds that consist of positively charged cations (molecules with a positive charge) and negatively charged anions (molecules with a negative charge) [22].

ILs have several properties that are briefly described in the following sections. Conventional salts, such as table salt (NaCl), are solid at room temperature because of their extremely high melting temperatures (over 800 °C). In contrast, ILs are salts that have melting points that are far lower than 100 °C. The distinctive feature of certain ILs is that they are liquid at room temperature. This characteristic results from the ions’ asymmetrical and bulky composition, which inhibits the regular crystalline lattice development that is typical in conventional salts [23, 24]. Also, the vapor pressure is very low in ILs. This indicates that, in contrast to many organic solvents that can be dangerous because of their volatility, they do not evaporate readily. ILs are safer to employ in chemical processes because of their low volatility, which lowers the possibility of atmospheric pollution and lessens the need for specialized ventilation [25, 26].

Strong electrostatic interactions exist in ILs. These interactions hold the ions (cations and anions) in ILs together. ILs have a high level of thermal and chemical stability because of these interactions, which are far stronger than those in molecular liquids. Because of their resistance to decomposition and ability to maintain stability under a variety of situations, they are useful in high-temperature applications [27, 28]. Also, one of the most striking features of ILs is their wide electrochemical stability window. This implies that they can be employed across a wide variety of voltages in electrochemical applications (such as fuel cells, batteries, and capacitors) without experiencing appreciable degradation. They are therefore perfect for use in systems that need stability at high voltages, such as energy storage devices [29, 30]. One of the interesting properties of ILs is their tunability, as the chemical composition of their cation and anion components can be changed to change their viscosity, polarity, solubility, and conductivity. ILs are extremely adaptable for a wide range of applications because of their flexibility to customize their properties [31, 32].

Depending on their molecular structure, ILs can range from being viscous (thick) to having comparatively low viscosity. One significant element influencing the processing and flow of ILs in a variety of applications is their viscosity. The size and form of the ions involved can often be changed to change the viscosity of an IL [33, 34].

2.1. Structure of ILs

ILs are usually composed of two main components, cations (ions that are positively charged) and anions (ions that are negatively charged). In these liquids, the cation is a big and organic molecule. Numerous physical and chemical characteristics of the IL, such as its solubility, conductivity, and viscosity, are influenced by the structure of the cation [35, 36]. Many cations are used in ILs, the most common of which include Imidazolium-based cations (e.g., 1-butyl-3-methylimidazolium [$BMIM^+$]), Phosphonium-based cations, Pyridinium-based cations, and Ammonium-based cations. Also, the anion is a complex anion, an organic ion (like acetate), or a halide (like chloride, BF_4^-). The solubility, conductivity, and reactivity of the IL are all significantly impacted by the anion selection [37, 38]. The most common anions used in ILs include Triflate ($CF_3SO_3^-$), Chloride (Cl^-), Acetate ($C_2H_3O_2^-$), Tetrafluoroborate (BF_4^-) and Hexafluorophosphate (PF_6^-).

There is an almost limitless range of ILs that may be made by combining various cations and anions, each with unique characteristics. These characteristics can be fine-tuned by researchers to fit particular uses [39].

2.2. Synthesis of ILs

ILs can be created by combining a cation, which is typically an organic molecule, with an anion, which is frequently a salt in specific molar ratios. The process of creating ILs involves several crucial steps. [40, 41] One of them is the selection of cations and anions. The ideal characteristics for a given application determine which cation and anion are used. Imidazolium-based cations, for instance, are frequently employed for their conductivity and stability. The other is the activation and mixing of raw materials. To create an IL, the cation and anion are combined according to one of the methods mentioned above, and then, if necessary, purification steps are performed to remove impurities. The final stage is purification. To remove any remaining solvent or contaminants, ILs frequently go through procedures like distillation or filtering.

2.3. Applications of ILs

ILs have many uses in many different industries because of their special qualities. [42, 43]. One of them is green chemistry. Because of their low volatility, non-toxicity, and broad range of solubility, ILs are regarded as green solvents. They are frequently employed as solvents in ecologically friendly chemical processes, in organic synthesis, and in catalytic reactions [26, 44].

Another use of these solvents is in energy storage and conversion. ILs can be employed as electrolytes in batteries and supercapacitors because they offer excellent conductivity and stability across a wide voltage range. Their application in next-generation energy storage technologies, such as solid-state and lithium-ion batteries, is the result of this [45]. Also, these solvents are used in carbon absorption. Because ILs can dissolve gases at high concentrations, they can be utilized to capture and separate gases like CO_2.

They are therefore useful in the fight against greenhouse gas pollution [46, 47]. Biotechnology is another field that uses these solvents. ILs are utilized in the biotech and pharmaceutical sectors for enzyme catalysis, protein crystallization, and medication formulation. They are appropriate for a range of uses in drug discovery and development due to their capacity to dissolve both hydrophilic and hydrophobic substances [48, 49]. Also, because of their strong solvent power and capacity to dissolve metal salts, ILs are utilized in the extraction and purification of metals, including copper, gold, and lithium [50, 51]. ILs are perfect for use as lubricants in harsh environments because of their great thermal stability and non-volatility. Additionally, they are utilized in materials science and electronics-related specialty coatings [52, 53].

In addition to the advantages mentioned for ILs, these solvents have several disadvantages and limitations. The first limitation is their cost. Despite their many uses and advantages, ILs can be costly to produce, particularly if they are made for specialized uses. The synthesis of ILs, especially those used for carbon dioxide absorption, can be more than ten times cheaper than the cost of synthesizing conventional solvents [54-56] In fact, one of the obstacles to widespread commercial application is the high cost of synthesis and purification. Another limitation is concerns about the toxicity of these solvents. While many ILs are considered non-toxic, some of them contain dangerous anions or cations that can be damaging to health. It is essential to properly choose and evaluate the IL components to ensure safety in various applications [57, 58]. Another is the viscosity of ILs. In some applications where fluidity is critical, the high viscosity of some ILs may be problematic. For example, their use in processes that require simple flow or mixing may be limited [59, 60].

3. The Concept of DESs – Properties and Applications

Due to the limitations of ILs mentioned above, a class of green solvents called deep eutectic solvents was introduced. DESs are frequently categorized as inexpensive, less toxic, and eco-friendly analogs of ILs (ILs). The number of articles about DESs has grown rapidly since the initial publications in 2001 [61], and according to the Web of Science, there are currently over 6,000 publications on the subject.

The concept of DESs was first described by Abbott *et al*. [61]. Although DESs and ILs share many physical characteristics, their chemical structures are very different. A DES is structurally composed of at least two readily available, low-cost, non-toxic components that can self-associate at a specific molar ratio to generate a new eutectic phase.

3.1. DESs preparation methods

DESs can be prepared using a variety of techniques, including vacuum evaporation, microwave, ultrasonication, grinding, freeze drying, and heating and stirring (sometimes referred to as thermal mixing) [62]. One of the most popular techniques is heating and stirring, which involves mixing individual components with or without a specific quantity of water, then heating the mixture in a water bath or on a hot plate (between 50 and 100 °C) until a clear homogenous liquid is achieved. Although it takes more time to prepare, this method is among the easiest, least expensive, and safest. [63, 64]. The ultra-fast microwave-aided synthesis of DESs involves mixing individual components in glass bottles and exposing them to microwaves for a brief period (less than 30 s at 180 W) [65]. Similar to this, in ultrasound (US) assisted synthesis, separate ingredients are combined with a known volume of distilled water (10, 30, 75% w/w) in a screw-capped glass vial. The glass vial is then exposed to US waves (37 KHz, 30 W) at 50 ˚C until a clear, homogenous liquid is created [66]. The grinding process involves combining ingredients and pounding them in a pestle and mortar at room temperature until a clear solution is achieved. DESs made with this process are naturally pure [67]. In the freeze-drying method, aqueous solutions of DESs or the individual components are freeze-dried to sublimate the water to get the solvent in its pure form [68]. Finally, the vacuum evaporation method, in which HBA and HBD are mixed in water and evaporated at 50 ˚C using the rotator evaporator. The resulting solution is maintained in a charged desiccator until its weight stabilizes [69]. DESs based on choline chloride (ChCl) and carboxylic acids degrade at high temperatures due to the esterification reaction, regardless of the synthesis technique [70]. Furthermore, when DESs are made using three distinct techniques stirring, microwave, and ultrasonication change in their physicochemical characteristics was observed [65, 71]. The methods for preparing the DESs are summarized in Table **1**.

Table 1: Methods for preparing the DESs.

DES Preparation Techniques	**Definition**	**Reference**
Heating and stirring	The most popular technique involves mixing, stirring, and heating the components in a water bath or on a hot plate until a clear, homogeneous liquid appears.	[63, 64]
Microwave	Mixing the individual components in glass bottles and exposing them to the microwave for a short time.	[65]
Ultrasonication	The individual ingredients are combined with a known volume of distilled water and then exposed to ultrasound waves until a clear, homogeneous liquid appears.	[66]
Grinding	The natural purity of DES is made by this process, mixing the ingredients and pounding them in a mortar at room temperature until a clear solution is obtained.	[67]
Freeze drying	Aqueous solutions of DESs or the individual components are freeze-dried to sublimate the water to get the solvent in its pure form.	[68]
Vacuum evaporation	HBA and HBD are mixed in water and evaporated at 50 ˚C using a rotary evaporator.	[69]

3.2. Simulation methods for DESs

There are several methods for simulating DESs, four of which we will cover in this review.

The first method involves quantum mechanical methods. Compared to regular liquids, the interactions between molecules in DESs are more varied and challenging to comprehend. Namely, among the most important issues of interest are the characteristics of the H-bond network and charge delocalization between solvent components. The first broad idea on the physical mechanisms of choline chloride and urea in a 1:2 molar ratio (reline) was that urea and chloride anions interact by disrupting the lattice of choline chloride, which causes charge delocalization and inhibits crystallization [72]. Using electronic structural predictions, Altamash *et al*. later showed that the anion transfers the most charge to the choline cation [73]. However, it has also been demonstrated that urea and chloride interactions do not always result in a drop in melting point [74].

The next method is Density functional theory (DFT)-Derived Peculiarities of the Local DES Structure. In this step, a small but significant portion of a DES must be chosen for the simulation because electronic structure approaches are quite computationally demanding. DFT, which can describe geometry and electronic structure well and with appropriate computation times, is usually the method of choice because it strikes a fair balance between accuracy and efficiency at the quantum level. DFT functionals, for instance, are frequently used to compute the equilibrium geometries of single molecules or complexes of molecules connected by networks of different interactions in the gas phase or to mimic the effects of a solution using continuum solvation models [75-77].

The PM6 approximation [78, 79] and the SE tight-binding DFT technique GFN2-xTB are two examples of semi-empirical methods (SE) that can be used to study the conformational space of mutual arrangements of molecules in a DES [80]. This technique can also be used to quantify short-range interactions and analyze the specifics of the interactions' kind and intensity. Electrostatic potentials (ESP), reduced density gradients (RDG), and Bader's quantum theory of atoms in a molecule (QTAIM) [81] are among the techniques used to investigate these characteristics. Classification of H-bonds, the corresponding bond strengths, and covalency can be completed by examining bond critical points (BCP) in the QTAIM representation. H-bonds, van der Waals contacts, and steric effects are examples of non-covalent interactions that can be described using RDG analysis based on electron density and its derivatives [82].

DFT methods have been useful for investigating the distribution patterns of DES components as well as how DESs interact with different functional chemicals in gas separation. The development of new solutions for desulfurization of liquid fuels [83-87], capturing greenhouse gases like CO_2 or SO_2 [88-91], metronidazole extraction from plasma [92], creating effective mercury removal strategies from various gases [92], extractive detoxification of feedstocks for the production of biofuels using new hydrophobic DESs [93], capturing NH_3 [94], and phenolic compound separation from oil mixtures [95] have all made use of free energy changes and structural analyses.

In the context of nano-objects, even more complicated issues have been studied [96-98]. The interactions governing a DES made of a combination of methyltriphenylphosphonium bromide with glycerol and carbon nanotubes were described at the molecular level by Lawal *et al*. [99], who also demonstrated physisorption through hydrophobic and π–π interactions. The electronic structure of noble metal nanoparticles (Mn, M = Cu, Ag, and Au; n = 1–4) and their complexes with ChCl: Urea DES was examined by Shakourian-Fard *et al*. [100] using the M06-2X functional.

Unconventional H-bonds (C–H…Mn and N–H…Mn) and the $[Cl]^{--}$… Mn interactions were identified as the two main bonding variables that control the interactions. Shakourian-Fard *et al*. [100] characterized the adsorption of DESs on various graphene surfaces using the M06-2X/cc-pVDZ level and demonstrated that it is non-covalent and dominated by dispersion energies.

3.2.1. Relations between DFT and nuclear magnetic resonance (NMR) and fourier-transform infrared spectroscopy (FT-IR) experiments of DESs

Comparing computer predictions with experimental results, such as NMR or FT-IR, is essential. The use of NMR to probe both cations and anions through many nuclei (^{1}H, ^{13}C, ^{19}F, ^{35}Cl, ^{11}B, ^{15}N, and ^{31}P) allows for the investigation of structure-property connections and interactions in DESs [101]. Advanced investigations of interactions between cations, anions, and solutes are made possible by the use of NMR chemical shift deviations, relaxation, nuclear Overhauser effect, and diffusion experiments. As a result, the molecular design of DESs is made easier. FT-IR is a trustworthy method for examining both solid and liquid samples. It is possible to compare vibrational modes from DFT calculations with FT-IR absorption spectra. After vibration assignments, FT-IR spectra can be used to infer the presence of hydrogen bonds. The accuracy is determined by the system size and how well the range of internal vibrational frequencies inherent in the system is covered [102].

3.2.2. Periodic DFT in studies of the condensed phase of DESs

A common technique that makes it possible to investigate hundreds of atoms is periodic ab initio calculations. Although the system sizes are still constrained, this approach is a potent tool for examining the electronic structure of the DES condensed phase. Usually, periodic ab initio calculations use the DFT in the hybrid Gaussian and plane waves (GPW) approach [103].

This method was employed by Korotkevich *et al*. [104] to investigate SO_2 absorption by ChCl/glycerol DES. All atoms were subjected to the generalized gradient approximation (GGA) using the Becke Lee–Yang–Parr (BLYP) [105, 106] functional and the corresponding BLYP Goedecker–Teter–Hutter (GTH) [107] pseudopotentials for core electrons, along with the molecularly optimized double-z basis set (MOLOPT-DZVP-SR-GTH) [108]. Grimme's D3 scheme with Becke Johnson damping was used to compensate for the GGA functional's lack of dispersion interactions [109, 110]. The authors identified hydrogen bonding and other specific interactions between all components. To study the behavior of reline and its equimolar mixture with water, Fetisov *et al*. [111] employed the same methodology to perform ab initio MD (AIMD) simulations in the canonical ensemble at temperatures of 333 and 363 K.

It was shown that in hydrous reline, water competes for the anions, and the hydrogen atoms of urea have similar propensities to bond to the chloride (Cl^-) ions and the oxygen atoms of urea and water. Malik *et al*. [90] employed the same level of theory to elucidate the solvation structure surrounding CO_2 and SO_2 in ChCl-based DESs, specifically reline and ethaline.

Molecular dynamics (MD) simulations are another method. The most widely used computer method for researching material dynamics and nanostructures is MD simulations [112]. MD can explain and forecast the mechanisms underlying molecular interactions and offers direct information about molecular processes.

MD simulations are used in over half of the computational works that are devoted to studying DESs (over 150 articles in the Web of Science by the end of 2021). MD simulations are most frequently utilized to gather information regarding the nanoscale structure. Studying characteristics that are directly relevant to uses, including fuel purification and gas separation, i.e., simulations of low molecular weight molecules in DESs, is another frequent goal. Simulations of nanoscale objects and the surfaces of bigger objects are made possible by the available length scales, which are usually on the order of around 10 nm. Importantly, MD simulations can also be a helpful tool for rheological property research.

The creation of force fields (FFs) that can reproduce the dynamic and structural characteristics of DESs is the primary obstacle in MD simulations. Because of the similarities between the interactions in ILs and DESs, the history of FF development for these solvents is intimately connected. The FFs for DESs, however, have unique characteristics because of the variations in their compositions and particular interactions. Neglecting polarization may result in an overestimation of the ion-ion interactions [113] and potentially unreliable results, such as a reduction in the diffusion coefficient by several orders of magnitude. The primary issues are associated with the presence of strong ionic interactions and highly polarizable atoms and molecules [113]. Nonetheless, non-polarizable FFs are often able to correctly reproduce the structural properties of DESs [114].

Currently, two main approaches are being used to address the issue of polarization effects. The first is based on the non-covalent interactions in the current FFs that are corrected in one or more ways. This method has the benefit of using existing FFs and requiring no extra parameterization. Moreover, this approach uses no more computational resources than the usual MD. Consequently, this method has been the most widely applied in IL simulations and it is the most common approach [115]. The second method, which uses formulations that model the electronic degrees of freedom and hence demand greater computing power, involves explicitly including polarization effects in force fields.

The last method is machine learning methods. An intuitive grasp of the connection between a DES's chemical makeup and characteristics is typically necessary while designing new DESs. Because of this, molecular design may be expensive and time-consuming. The use of machine learning (ML) to address these issues is growing in popularity. To anticipate the properties of novel compounds, ML relies on statistically processing vast datasets and finding correlations between input and output data (for instance, between structure and property) [116-119]. Quantitative structure-property relationship (QSPR) prediction is one of the most common uses of this method [116, 120]. Regression analysis techniques and Artificial Neural Networks (ANNs) are the most often utilized mathematical models for this purpose [116, 117, 120].

3.3. DES property prediction

The prediction and estimation of the various DES properties using ANNs is the subject of numerous publications. For instance, Shahbaz *et al*. predicted the densities of three distinct ammonium- and phosphonium-based DESs over a variety of temperatures and compositions using an ANN with three layers (6-9-1 architecture), where the input, hidden, and output layers comprise 8, 4, and 1 neurons, respectively [121]. The temperature and the mole fraction of DES components were used as inputs. An average absolute error of 0.14% was attained. The same year, the authors published another study in which they used DESs to estimate the removal of glycerol from palm oil-based biodiesel using an ANN model with an 8-4-1 structure [122]. An absolute average deviation of 6.46% indicated that the results were in good agreement with the experimentally obtained data.

4. Main Directions of Investigations

4.1. DES structure

Deep eutectic solvents, or DESs, have grown in popularity during the last 20 years for a variety of uses [116, 123]. The number of articles about DESs has grown rapidly since the initial publications in 2001 [61]. DESs have attracted a lot of attention because of their special qualities. DESs are frequently categorized as inexpensive, non-toxic, and eco-friendly substitutes for ILs (ILs); see, e.g., Amde, Liu, and Pang [124] for a review regarding the environmental aspects of ILs. DESs have demonstrated great potential in a variety of fields, including pharmaceuticals [125, 126], biosensor development [127], membrane technology [128, 129], extraction procedures [130], biomass transformation [131], electrochemistry [132], nanotechnology [133], and additive technology [134, 135].

Determining the DES structure-property relationship is one of the primary goals. Understanding the characteristics of DESs at the nanoscopic level is crucial given the enormous number of conceivable DES combinations. DES is made up of two or more components—usually a hydrogen bond donor (HBD) and a hydrogen bond acceptor (HBA) such as quaternary ammonium salts at moderate temperature (≤80 ˚C) that form a eutectic mixture with a lower melting point than the individual components. The interactions between the compounds occur through intermolecular hydrogen bonds, which, in a certain range of compositions, cause a significant drop in the melting point (mp), resulting in a liquid-state mixture [136]. The eutectic effect is the result of the DES's melting point being lower than the melting points of its constituent parts. Because of its low cost, and environmentally friendly characteristics, this family of solvents has drawn more and more interest in the field of green chemistry. The mixing of Ch (mp = 302 ˚C) and urea (mp = 133 ˚C) in a 1:2 molar ratio, which yields a room-temperature liquid (mp = 12 ˚C), is a well-known example [137]. HDBs, which are easily accessible components found in naturally occurring primary metabolites such as amines, sugars, alcohols, sugar alcohols, polyols, and organic acids, can be employed with a broad variety of salts (choline chloride (Ch) being the most widely used) [138, 139]. DESs are referred to as natural deep eutectic solvents (NADESs) when the molecules that make them up are primary metabolites, while the words are occasionally used interchangeably in the literature to refer to the same mixtures [140]. Because of their special properties, DESs can operate as an effective solvent at lower temperatures, which makes them appropriate for a range of chemical, biological, and industrial processes.

Each component (HBD and HBA) can be changed to alter the DES's structure and characteristics. The chemical structures covered in this section are seen in Figures **1** and **2**.

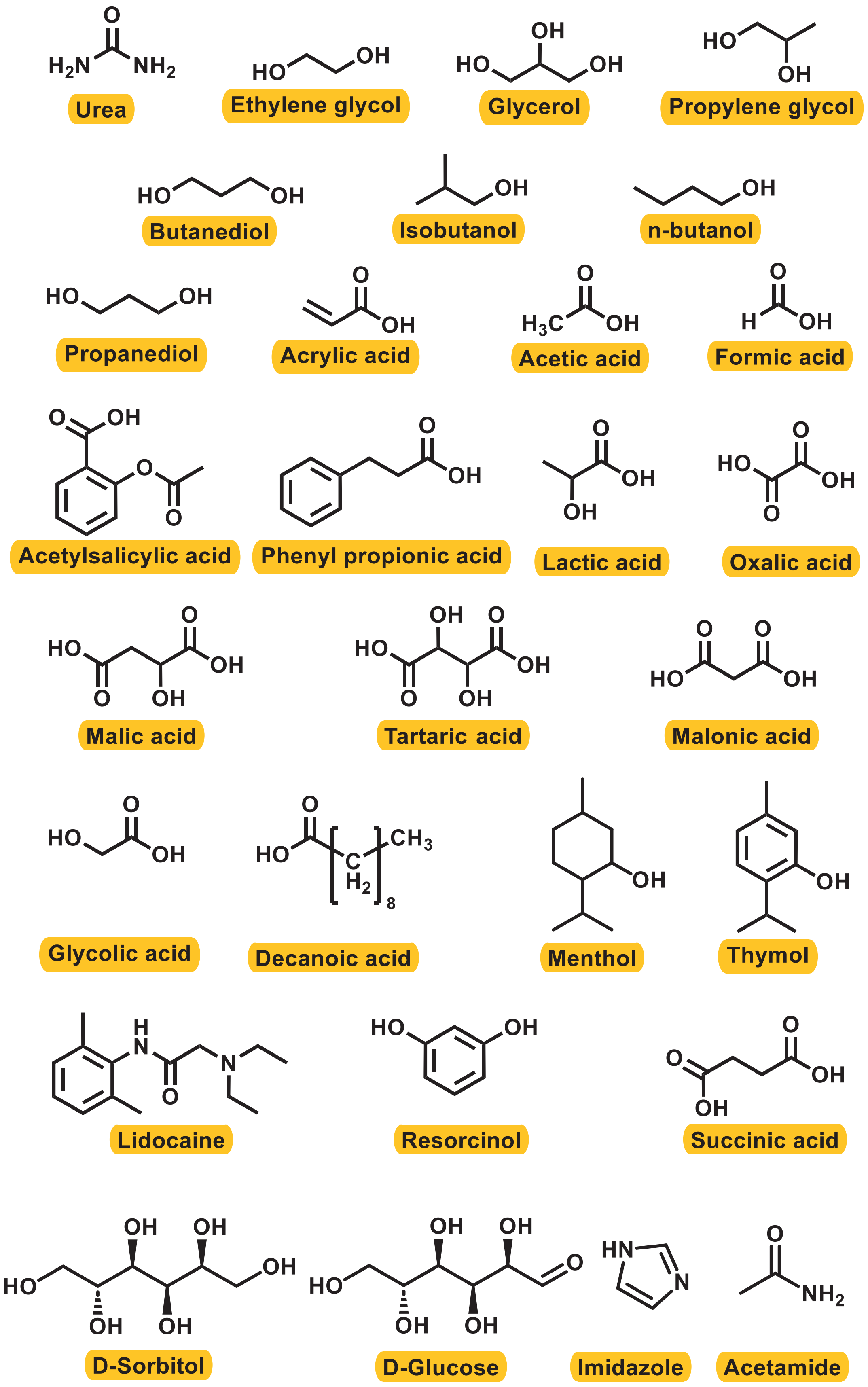

Figure 1: List of commonly used HBDs in preparation of DESs [141].

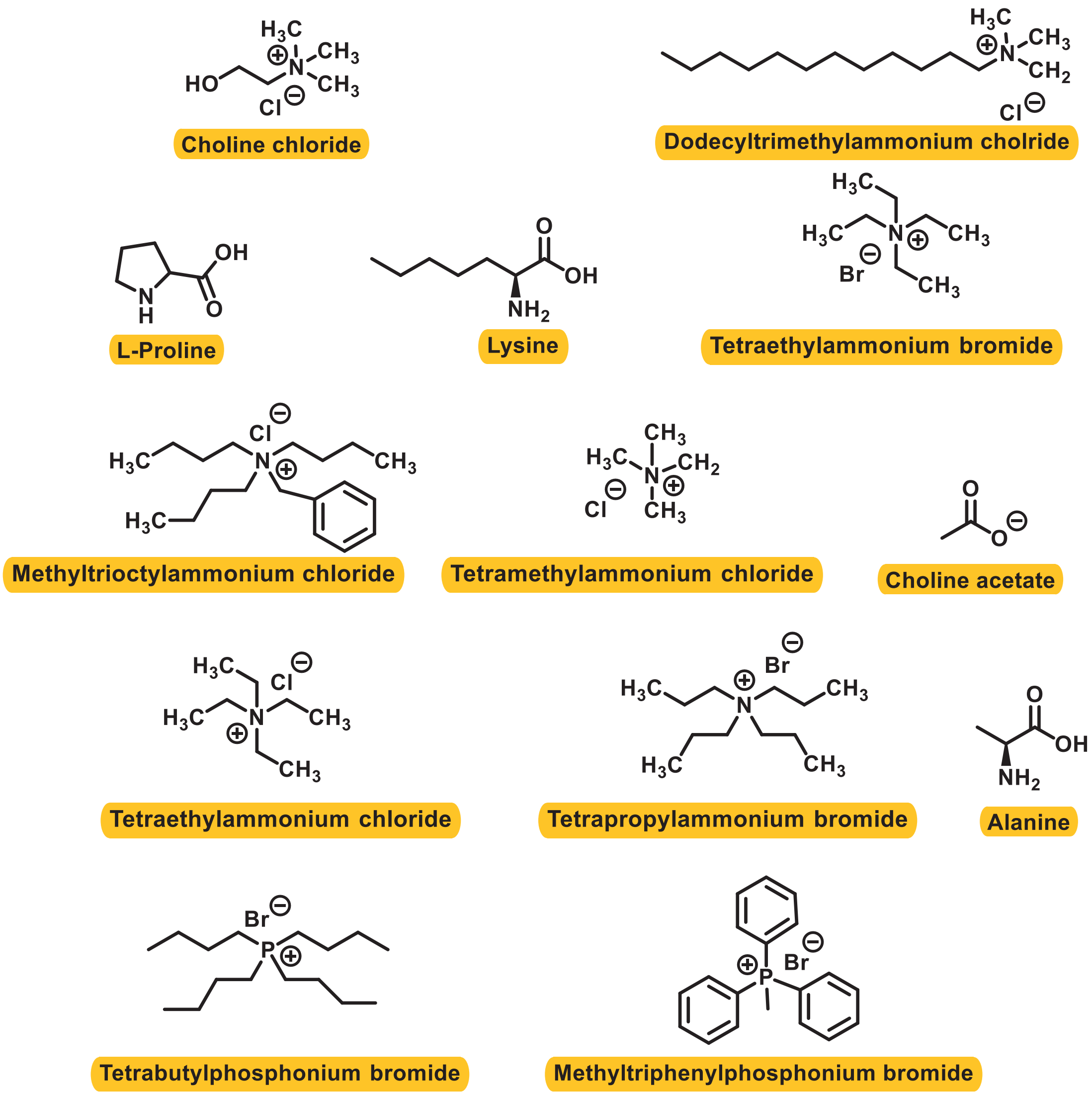

Figure 2: List of commonly used HBAs in preparation of DESs [141].

As illustrated in Figure **3**, DESs are divided into four categories based on their chemical makeup. Given that Type III comprises a subtype of natural DESs (NADESs), it merits particular consideration. Because their constituents come from natural resources, NADESs are unique. Choi *et al*. (2011) were the first to propose the phrase “natural DES” [142]. Because NADESs are biocompatible and biodegradable, their potential applications in the medical field. New DES types have emerged as a result of the significant expansion in DES development.

Therefore, a deep depression in the melting temperature of the mixture of non-ionic compounds (menthol and organic acids) was found by Verma *et al*. [143] in 2018. Later, the same effect was also found in a thymol and menthol mixture by Abranches *et al*. [144], who proposed categorizing it as a novel form of DES-non-ionic DES. Both experimental and computational researchers are particularly interested in this discovery, which broadens the characteristics and potential uses of DESs. It is crucial to note that DESs are multi-component systems made up of many kinds of molecules, such as alcohol, acids, and salts.

The adaptability of the elements creates a wide range of possibilities for developing DESs with highly adjustable characteristics.

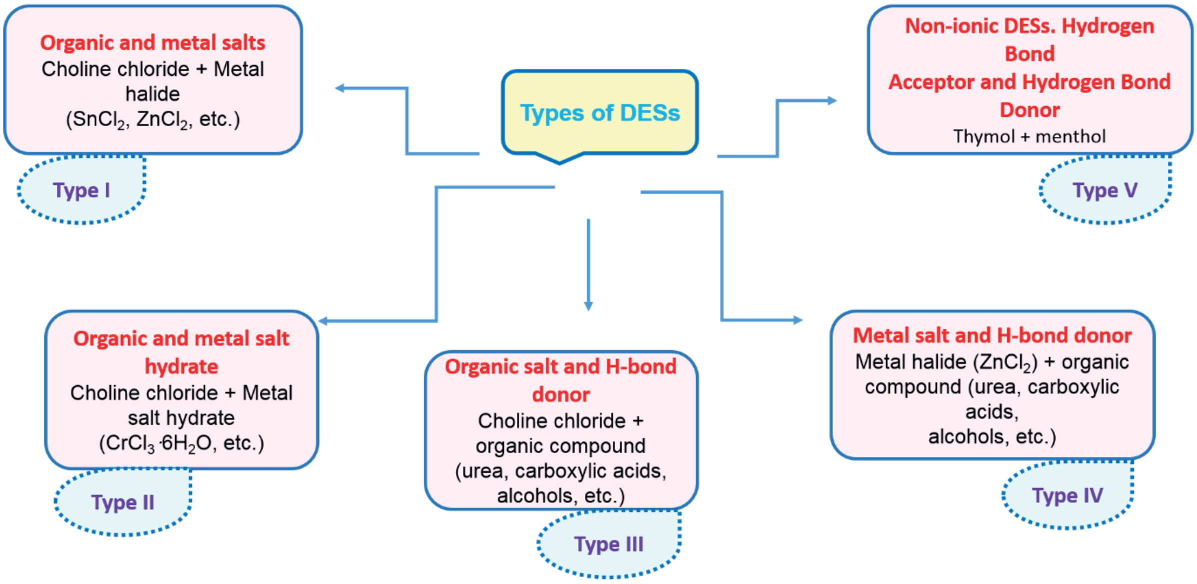

Figure 3: Types of DESs, their compositions, and examples [145].

4.2. Properties of DESs

DESs have many properties, some of which we will discuss here (Figure **4**) [146]. One of them is density (ρ). The density (ρ) of DESs is a basic and important physical property to determine the solvent selection, process design, and separation performance, as well as the large-scale application of DESs for extractions in general considering the biphasic nature of this unit operation. Also, this feature is for method design, device and container size calculations, overflow assessment, and cost prediction in large-scale applications [147]. The DES-rich phase is typically the heavier liquid at the bottom, though this isn't always the case, but the densities of DESs are often higher than those of water. For example, in liquid-liquid metal extraction from aqueous solutions, the extract-rich H(D)ESs phases are often recovered from the upper liquid phase because the H(D)ESs typically have a lower density than water (between 0.90 and 0.95 g cm^{-3}) [148].

By altering the kinds of individual components and their molar ratios, the DES density may be finely controlled [147, 149]. The densities of DESs based on bromine salts are often higher than those of their counterparts based on chlorine salts, suggesting that the anionic type would influence the DES densities [147]. Additionally, DESs made of hydroxyl HBDs showed a trend of rising densities as the number of hydroxyl groups increased (for example, glycerol (Gly)-based DESs are denser than EG-based ones).

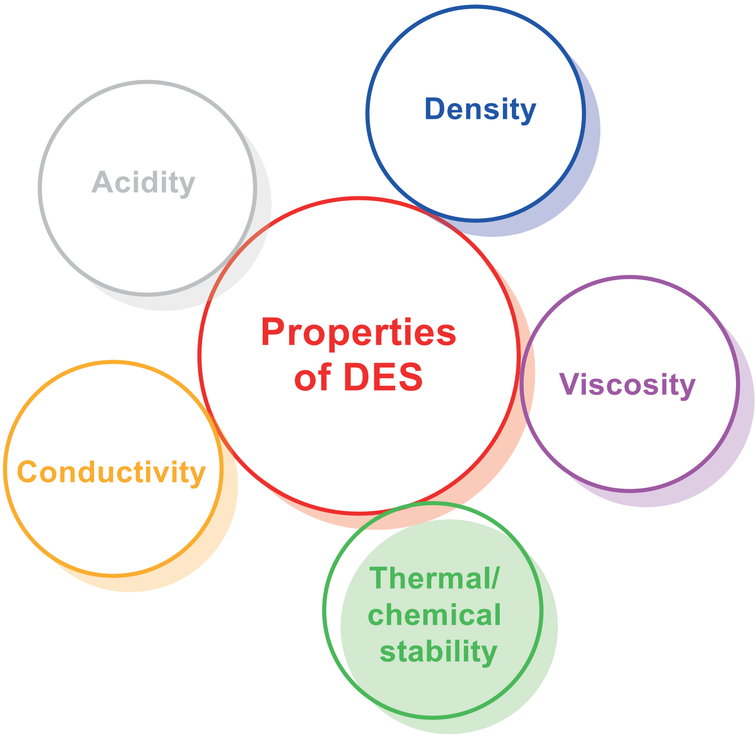

Figure 4: Important physicochemical properties of DESs for their application in metal recovery [146].

Conversely, the introduction of aromatic groups can lower the densities of DESs (for example, phenolic-based DESs have low densities) and lengthen the alkyl chain of the DESs components (for example, the densities of diacid-based DESs follow the order of oxalic acid ($C_2H_2O_4$)-based > malonic acid ($C_3H_4O_4$)-based > glutaric acid ($C_5H_8O_4$)-based) [150]. Regarding the component molar ratios, it has been widely documented that as the HBD ratios grow, the density of DES would likewise rise noticeably [151-153]. For instance, Shafie *et al*. (2019) found that when the molar ratios of HBA: HBD changed from 2: 1 to 1: 3.63, the densities of ChCl: citric acid monohydrate DESs rose sharply from 2.64 to 3.11 g cm^{-3}. The idea that larger HBD ratios would result in a stronger hydrogen bond interaction between HBA and HBD, which would reduce free space, or called the average “hole” radius, and the change of the packing efficiency and molecular arrangement of DESs, could explain this [151, 153, 154]. Furthermore, because the volumes of DESs and consequently their densities are affected by changes in molecular activity and mobility with temperature, the densities of DESs likewise fluctuate with temperature. As the temperature rises, DES densities and molar volumes typically fall and rise linearly, respectively [153, 155]. Another property of DESs is their viscosity (μ), which influences mass transfer in DES-based separation applications [156]. Similar to ILs, the large size of the ions and the comparatively high free volume of DESs are the causes of their larger μ when compared to the majority of traditional molecular solvents, such as water [123]. Low viscosities suggest that the components of DESs have weak molecular connections, which could lower their solubility and freezing point [147]. According to the above, to adjust the viscosity of DESs, a balance must be struck between other factors, including solubility characteristics and ion size, etc., which requires a greater understanding of DESs. Since the structures and molecular ratios of HBAs and HBDs in DESs have decisive impacts on DESs' viscosities, the viscosity of DESs can be modified by changing their composition. For example, the viscosities of the DESs rose as their molecular weights grew in the ammonium-based DES with Gly as HBD [157]. The viscosities of the ammonium-ChCl-based DESs would sharply fall with higher molar ratios of EG but increase with the malonic acid (MA) content when using the mixture of EG and MA as HBDs. The difference in the strength of the hydrogen bond interaction between HBA and HBD helps to explain this [153]. Furthermore, it has been reported that up to about 42 weight percent of DESs maintain their usual network of hydrogen bonds [158, 159]. Therefore, adding a small amount of water to a DES could lower its viscosity [160].

In addition to DES's composition, μ also depends on temperature. Similar to molecular solvents, models describing this dependence typically use the Arrhenius, Vogel–Tammann–Fulcher, or Andrade equations [153]. As the temperature rose, μ generally decreased. This may be explained by molecules becoming easier to flow due to a decrease in internal resistance [161].

The importance of this variable in metal separations has been documented in numerous investigations. Doche *et al*. (2017) investigated how temperature affected the rate at which Co leached from ethaline [162]. Because of the drop in ethaline's μ, the leaching rate at 80 °C was 15 times greater than that at 25 °C. Zhu and associates (2019) [163] additionally said that when the temperature was raised from 60 to 80 °C, the Zn dissolving efficiency in a ChCl-based DES rose from 48.5% to 85.2%. The conductivity (κ) of DESs is a crucial characteristic to assess their suitability as effective electrolytes, as electrodeposition is an important procedure for the recovery of metal dissolved in DESs. Both the ion migration rates in the solvents and the condition of free ions are strongly correlated with κ [147]. The "hole," or free space, idea explains why this feature changes with temperature, μ, and ion size [116, 164]. The conductivity profiles of sixteen DESs were examined by Ghareh Bagh *et al*. (2015) [165], who found a consistent trend of increasing with temperature. The reason for this is due to the increase in kinetic energy and the rate of migration of molecules at higher temperatures [153].

Ammonium-based DESs with lower viscosities tended to have greater κ than phosphonium-based DESs because κ increased dramatically as viscosity reduced due to the free mobility of ionic species as hole mobility (the ability of a hole to travel across a metal or semiconductor in the presence of an applied electric field) increased [157]. Furthermore, reducing the ion size may increase the DESs' free volume, which would lower their viscosity and enhance their conductivity [116, 166]. In electrolysis procedures, for instance, the smaller ammonium-based DESs were more suited as solvents and electrolytes due to their higher electrical conductivities than the phosphonium-based DESs [165]. The Walden rule describes the connection between temperature, viscosity, and conductivity [147]. Another characteristic of DESs is their thermal and chemical stability. The solvents' performance and reusability in large-scale applications are directly impacted by their stability [167]. According to reports, DESs have comparatively strong chemical and thermal stability, which depends on the composition's functional groups, spatial structure, and molar ratios [147].

The decomposition temperature, which is typically determined by thermogravimetric analysis (TGA), can be used to assess the thermal stability of DESs. The maximum temperature at which a DES can remain a fluid without experiencing any degradation in its physicochemical characteristics is known as the decomposition temperature [153, 155]. Thus, a higher decomposition temperature is desirable for DESs as it indicates that they can be used at a higher temperature with better thermal stability. There are two main analytical techniques for assessing the thermal stability of DESs further. One of them is the assessment of how the physicochemical properties change as a function of temperature increment, and the next one is the measurement of the mass loss as a function of time at a constant temperature [155, 168, 169]. Numerous investigations have documented that both the intrinsic thermal stability of the compositions and the intermolecular hydrogen bonds that were generated between HBAs and HBDs had a significant impact on the thermal stability of the DESs [147, 170]. For instance, the number of hydrogen bonds that the acids offer and the thermal stability of the organic acid itself determine how thermally stable the ChCl-based DESs with organic acids as HBDs are [147].

The possible compositional or property changes brought on by chemical reactions (such as oxidation, hydrolysis, etc.) are referred to as DESs' chemical stability. One of the primary issues with DESs' chemical stability is their hygroscopic nature, particularly for hydrophilic DESs like ChCl-based DESs [171]. The hydrophilicity/hydrophobicity of DESs is also very important. The nature and interactions of the HBAs and HBDs determined the DESs' affinity for water, which can be used to classify them as hydrophilic or hydrophobic [147, 168]. Due to the presence of hydrogen bonds and coulombic interactions, early established DESs produced by ChCl as HBA and alcohols, carboxylic acids, or amides (such as urea) as HBD generally exhibit hydrophilic properties, making them entirely miscible with water [147, 155]. Hydrophobic eutectics (H(D)ESs), on the other hand, are frequently made up of naturally occurring and inherently hydrophobic terpenes (like menthol and thymol), trialkylphosphine oxide, or tetra alkyl-quaternary-ammonium salt as HBAs, as well as different HBDs like long-chain carboxylic acid (like decanoic acid), which have low water solubility [147, 168, 172, 173].

One important aspect influencing DESs' use in metal extraction and separation is their hydrophilicity/hydrophobicity. Due to their excellent extraction effectiveness for non-polar analytes and water immiscibility, H(D)ESs are necessary for metal extraction and recovery from water-soluble environments [130, 147, 174, 175]. A (H)DES synthesized by decanoic acid and lidocaine was first reported by van Osch *et al*. in 2015 [176]. These solvents have been widely used as promising extractants for metals from a range of aqueous materials [168, 177, 178]. For instance, decanoic acid and lidocaine were used to create H(D)ESs, which had an extraction rate of over 99% when used to remove Co^{2+} from water [179].

Hydrophobic DESs are one of the kinds that are currently being developed. In 2015, hydrophobic DESs were introduced as liquid-liquid extraction solvents [176, 180]. The field of hydrophobic DESs has expanded significantly since then. The acidity of DESs is another important factor affecting the performance they removing metal. By using UV-Vis spectroscopy to measure the ratio of absorbances for the protonated and deprotonated forms of the indicator, one can ascertain the acidity of DESs [181]. As with other organic solvents, acidity has been measured using the Hammett function. The organic acids HBDs (such as sulfonic acid and carboxylic acid) could supply the acidity of DESs [85]. It was demonstrated that metal oxides were better dissolved by more acidic DESs. For instance, when compared to other DESs made by

Gly and EG, those made with organic acids (such as glycolic acid and L-lactic acid) were able to dissolve Nd_2O_3 more effectively [182]. Aluminates were more soluble in DESs based on malonic acid than in those based on urea and EG, while Fe_3O_4 exhibited a greater propensity to dissolve in stronger acidic DESs made of ChCl: oxalic acid as opposed to ChCl: phenylpropionic acid [183, 184].

4.3. Role of hydrogen bond acceptor (Anion)

Altering the H-bond acceptor (HBA) is another method of controlling a DES's characteristics. Both interactions (with cation and HBD) will change if HBA is replaced because it is involved in both of them. Recently, Migliorati *et al*. [185] discussed how an anion contributes to the creation of H-bond networks. Chloride, fluoride, nitrate, and acetate are the four distinct anions on which they compared the structure and characteristics of DESs. The findings demonstrated that the strength of the H-bonds between the urea and anion and the order of DES melting points do not correlate one to one; rather, a complex network of interactions is created in which the anions attempt to maximize their H-bond interactions with the other elements of the system. The nature of the anion determines how each one accomplishes this objective. It was demonstrated that polyatomic anions, such as nitrate and acetate, can bind two urea hydrogens at the same time, in contrast to monatomic anions.

4.4. Role of cation

The majority of cation research has concentrated on DESs based on ChCl. Migliorati *et al*. [186] In deep eutectic solvents (DESs), the hydrogen bond donor is crucial for driving the phase separation and making it possible for aqueous biphasic systems to form. The HBD affects the system's miscibility, regulates the partitioning of solutes, and aids in the optimization of separation or extraction procedures by forming hydrogen bonds with water molecules. Additionally, the HBD component can modify the system's characteristics, improving control over phase behavior, selectivity, and process efficiency in general.

4.5. Electrolyte-based DESs

Electrolyte-based DESs (the first and fourth types of DES) have also been found to exhibit structural heterogeneity. The direct evidence of spatial heterogeneity at the nanoscale in electrolyte-based DES Li^+/ClO_4^-: Acetamide and propionamide are alkyl amides that were reported by Kashyap *et al*. [187, 188]. The scientists showed that the segregated domains of the component electrolyte display nanoscale spatial heterogeneity; elongation of the alkylamide tail increases the strength of ion-pairing and the amount of nanoscale morphology. Additionally, they discovered that the degree of heterogeneity rises with temperature, which they attributed to the stronger correlations between the ionic species that compensate for the decline in ionic species-alkyl amide cross-correlations. For lithium-ion batteries used in low-temperature environments, new DESs based on lithium salts show promise as electrolytes. The phenomenon of metal salt-based DESs' declining melting temperature is pertinent in this regard.

Ogawa and Mori [189] used a combination of MD and DFT techniques to investigate four representative DESs based on urea or tetramethylurea as HBDs and LiCl or Lithium bis(trifluoromethanesulfonyl)imide (LiTFSI). They demonstrated the eutectic mechanism of DESs by directly comparing the coordination states between Li salts and amides with or without NH groups, such as urea (with NH) or tetramethylurea (without NH). It was established that if the cation in the DES is bulky, such as in reline, the NH group coordinated with Cl^{-} ions causes the melting point to decrease. Conversely, when the cation has a high Lewis acidity (like Li^{+}), the CO group in the amide preferentially coordinates with the cation. The presence of an NH group may not result in a drop in the electrolyte melting point for DESs based on LiTFSI and an amide. Furthermore, Li-salt:amide-based electrolytes containing NH groups are unstable on the reduction side, according to the HOMO LUMO computed from DFT to evaluate electrochemical stability. Therefore, compounds lacking any NH groups are preferred for lithium-ion batteries over ChCl-based DES.

5. Ternary DES

Recently, the possibility of the formation of ternary DESs (TDESs) has been put forth which offer lower viscosity and melting points than binary DESs.

Li *et al*. used MD to model the ternary DES choline chloride (ChCl): resorcinol (Res): glycerol (Gly) mixture [190]. It was determined that glycerol, resorcinol, and ChCl generate a large number of H-bonds that cause each component's inherent microstructure to be destroyed. Consequently, a supramolecular H-bond network binds ChCl/Res/Gly together to form a DES.

To comprehend the H-bonding interactions, Ref. [78] looked at the impact of alcohols as ternary components (*n*-butanol, isobutanol, and butanediol) as an extra HBD in a binary DES made of ChCl and malonic acid in an equimolar ratio (1:1), commonly known as maline.

The molecular orbital (MO) energy levels were the main focus of the computations. In contrast to *n*-butanol and iso-butanol, it was determined that the H-bond network that forms between maline and butanediol causes a greater melting point depression. According to the interpretation, maline and butandiol form stable and homogenous systems, unlike n-butanol and iso-butanol. These findings were further supported and significantly validated by the evaluation of the total energy. While *n*-butanol and iso-butanol with higher negative values exhibit prominent phase separation, maline: butanediol forms a homogenous mixture to generate a TDES with a less negative value.

5.1. Application of DESs

DES are a fascinating and adaptable class of solvents with many uses. Here is a summary of some of the primary uses for DES [191, 192].

These solvents are used as green solvents for chemical and biological reactions. DES is utilized in many chemical reactions and helps reduce pollution and dependency on hazardous materials because it is often less volatile and non-toxic [193-195]. Enzymes are biological catalysts. Because they can denature (lose their shape and function) when exposed to harsh solvents, they frequently perform poorly in conventional solvents. The gentler characteristics of DES enable enzymes to work more effectively. Because DES can dissolve a variety of polar and non-polar substances, it is a great choice for applications requiring enzymatic reactions, such as the creation of fine chemicals or medications [196-198]. Extraction of Bioactive Compounds using these solvents represents another application of DESs. Bioactive substances, including flavonoids, essential oils, and antioxidants, can be efficiently extracted from plants or other natural materials using DES. These substances are frequently found in food items, cosmetics, and medications. Conventional extraction techniques, such as the use of methanol or ethanol, may leave harmful residues or be less effective. However, DES can increase these compounds' solubility, which will clean and improve the extraction process [199, 200]. DEs are used in the field of electrochemistry and energy storage for fuel cells, batteries, and supercapacitors. Batteries and Supercapacitors: DES is being investigated as an electrolyte for technologies such as supercapacitors and lithium-ion batteries. In these devices, DES can offer a more ecologically acceptable and stable substitute for conventional electrolytes, such as organic solvents, which can be poisonous or combustible. Because of their broad electrochemical stability windows and strong ionic conductivity, DES can function effectively at a variety of voltages and temperatures [201, 202]. By transforming the chemical energy of fuels—typically hydrogen—into electrical energy, a fuel cell produces electricity. When compared to conventional methods, DES can be used as electrolytes in these cells, increasing performance, decreasing energy loss, and enhancing the cell's overall performance [203, 204].

DESs are used in the extraction and separation of metal ions for metal recovery and the separation of rare earth elements. When recovering valuable metals from ores or scrap metal, DES is especially useful. For example, they can aid in the recovery of metals (lithium, cobalt, and copper) that are necessary for the production of electronics, batteries, and renewable energy technologies. DES's solvent qualities enable it to dissolve metal salts create complexes, and speed up the extraction process. This can be important for recycling or obtaining rare metals that are hard to extract using conventional methods [205, 206]. Lanthanides and other rare earth elements (REEs) are crucial for renewable energy, electronics, and magnets. These metals may be effectively separated from mixtures via DES, which is a critical process for industries like electronics and clean energy. The ability to use DES to extract REEs selectively offers more economical and environmentally friendly ways to separate and purify them [207, 208]. These solvents also have pharmaceutical applications, being used in drug formulations as well as in drug delivery systems. The effectiveness of certain medications is limited by their poor solubility in water or other solvents. These medications' solubility and bioavailability may be enhanced by DES. By dissolving poorly soluble materials, DES makes it easier for the body to absorb medications, boosting their therapeutic effectiveness [209, 210].

Additionally, DES is being researched as a medication delivery system carrier. They could be used, for instance, to formulate oral drugs, gels, or lotions that deliver active ingredients in a controlled manner. They could contribute to better patient outcomes by ensuring that medications are administered more efficiently and consistently [211, 212]. DESs are used in polymerization reactions, the main use of which is in polymer synthesis and nanomaterials manufacturing.

Long chains of molecules make up polymers, and DES can be employed as a solvent to speed up polymerization operations. Polymers are widely used in the production of medical devices, biodegradable materials, and plastics. Because DESs are frequently biodegradable and less hazardous than environmental pollution, using them as solvents for these reactions can help minimize environmental pollution [213].

Materials having nanoscale structures are known as nanomaterials, and they can possess special chemical, biological, and physical characteristics. Because DES can stabilize nanoparticles and aid in their production, they are used to make these materials. Numerous industries, including electronics, medication delivery, and environmental monitoring, use these nanoparticles [214, 215]. These solvents are used as green catalysts in catalytic reactions. DES can promote the conversion of raw materials into products by acting as a medium for catalytic reactions. In this case, DES may improve the process's efficiency and selectivity by stabilizing active species or intermediates during the process. This is used in a variety of industrial chemical processes where efficiency and high precision are crucial [216-219].

The goal of green chemistry is to reduce the harm and increase the sustainability of chemical processes. In catalytic processes, DES are regarded as green solvents since they minimize waste, energy usage, and hazardous byproducts. This is crucial for creating more environmentally friendly methods of producing sustainable chemicals [194, 220]. These solvents are used in the wastewater treatment process to remove or dissolve contaminants. Heavy metals, dyes, and organic compounds are among the contaminants that DES can efficiently remove from wastewater. Traditional wastewater treatment techniques frequently involve hazardous chemicals or lead to secondary contamination. By dissolving hazardous substances to facilitate removal or converting them into less toxic forms, DES provides a more environmentally friendly alternative [221, 222]. Hydrophobic (water-insoluble) pollutants that are challenging to eliminate using conventional techniques can become more soluble with DES. DES can be used to separate and extract toxic substances, which can be particularly helpful for treating industrial effluent or cleaning up oil spills [223, 224]. These solvents are used in the food industry for food preservation and extraction of food components. DES is a viable option for food preservation because of its antibacterial properties. By preventing the growth of bacteria or fungi, for instance, they can help food products last longer on the shelf without the need for synthetic preservatives that could be harmful to human health [225, 226].

Valuable compounds from natural food sources can be extracted using DES in the food industry. This involves extracting vitamins, flavors, and other nutrients. DES are cleaner, more effective substitute for traditional food extraction techniques because they are generally safe and tailored to extract specific components [227, 228]. These solvents are used in the field of lignocellulosic biomass processing and also conversion into biofuels. Cellulose and lignin, which are difficult to decompose, are found in biomass such as wood, straw, and agricultural waste. DES can be used to pretreat these materials, breaking down the cellulose and lignin to make them more accessible for further conversion into biofuels or other chemicals. [229, 230]. It is easier to turn biomass into biofuels (such as ethanol or biodiesel) when DES is used as a pretreatment. In the drive for renewable energy sources that lessen dependency on fossil fuels, this is especially crucial [231]. These solvents are used for the separation and absorption of a number of gases, including CO_2 and SO_2. The decrease of greenhouse gas emissions is one of the world's biggest challenges. Acid gas emissions, particularly CO_2, are a major contributor to ocean acidification and climate change, and represent one of the most pressing technical challenges of this century.

Numerous sources contribute to global CO_2 emissions, including the burning of coal, oil, natural gas, or liquid gas in power plants, as well as the petrochemical and aluminum sectors. Fossil fuels are anticipated to remain a major source of energy generation for the foreseeable future, notwithstanding recent advancements in renewable energy sources. This means that CO_2 emissions will unavoidably keep rising [232-234]. Therefore, one of the biggest challenges of our day is creating more environmentally friendly and sustainable methods of capturing CO_2 from fossil fuels before, during, or after processing.

Over the past few years, a variety of CO_2 capture systems have been presented, such as sorption through membranes and solid and liquid sorbents [235]. Technology for selective membrane separation is one of the most promising approaches and is thought to be an economical way to reduce CO_2 emissions. Materials capable of efficiently separating and capturing gases on an industrial scale are necessary. Because of their unique properties and relative affordability, membranes based on DESs are highly promising [168].

The extraction of carbon dioxide from natural gas presents another challenge. In deep subterranean sources, natural gas is found as shale gas that contains non-hydrocarbon elements like CO_2. The removal of CO_2 from natural gas is one of the challenges in using it. The presence of CO_2 in natural gas is undesirable due to corrosion and low heating value [236, 237].

The intermolecular interactions of various DESs (reline, glycine, and maline) in contact with gas phases made up of pure CO_2, pure SO_2, and a model flue gas (including N_2, CO_2, O_2, and water) were investigated by García *et al*. [238] using MD simulations.

It has been established that the type of H-bonding sites present in the HBDs determines their intermolecular interactions. Using MD simulations, Kussainova *et al*. examined the mechanisms of CO_2 absorption on DESs based on monoethanol amine and methyltriphenylphosphonium (MEA) bromide [239]. The authors discovered that while interactions between CO_2 and MEA improved in the presence of the DESs, interactions between CO_2 molecules drastically diminished. Strong interactions between Br^-/CO_2 and MEA/CO_2 were also observed, indicating that these components sorb CO_2.

Alioui *et al*. [240] investigated the molecular interaction between CO_2 and various DESs using MD simulations and a theoretical approach. The energy of gas molecules' interactions with DESs was found to be correlated with their solubility: When the energy of attraction is larger, CO_2 becomes more soluble in DESs, and vice versa [240].

5.2. Water effect on DES

The way water interacts with DESs is one of the key questions. Ma *et al*. [241] have reviewed the impact of water on DESs and ILs. In practice, the presence of trace amounts of water in DESs is unavoidable in most cases [242, 243]. However, even minute amounts of water can have an impact on the H-bond network and drastically alter a DES's characteristics [241, 243]. Water molecules can be both HBDs and HBAs and can therefore drastically affect the arrangement of DES at the molecular level [242, 244].

To reduce the solvent's cost and maintain its environmental friendliness, water can also be utilized to alter several of the essential characteristics of DES, including its density, ionic conductivity, and viscosity [241, 244].

High viscosity, for instance, is regarded as one of the disadvantages of DESs that can prevent their broader use [241, 245], and DES solution viscosity decreases with increasing water content, which is frequently a favorable consequence [246, 247].

When water is added to a DES, the HBD component of the DES forms hydrogen bonds with the water molecules. The two phases—water and the organic phase—become immiscible as a result of this interaction, creating an aqueous biphasic system. The way the HBD alters the system's solvent properties is what causes this immiscibility between the phases [243, 248]. For instance, in the urea/choline chloride DES, urea forms hydrogen bonds with water molecules. This reduces the solubility of water in the DES and causes phase separation.

Much effort has been put into figuring out how water affects the micro and nanostructures of DESs [111, 249-251]. Di Pietro *et al*. examined ChCl: urea and ChCl: glycolic acid DESs with aqueous dilution using MD simulations and NMR spectroscopy [242]. When water was added, the Cl^- ions experienced asymmetric hydration, and DES components were displaced, making water the dominant ligand. Also, the impact of water on the structure of ChCl/sesamol 1:3 DES was examined by Busato *et al*. [252]. Water separates from sesamol and absorbs most of the ChCl in the aqueous region when the water/DES molar ratio is higher than 6. Weng *et al*. used MD simulations to explain the dual action of water on DESs composed of 1:2 ChCl/glycerol [245]. Both the number of H-bonds between choline and glycerol and the quantity of ChCl–glycerol supramolecular complexes in DES dramatically dropped with the addition of water. Choline can also be linked to glycerol by water rather than chloride.

5.3. Activity and stability of enzymes in DES/water mixtures

For redox biocatalysis, DESs can also be utilized as a biodegradable and non-toxic reaction medium [253]. MD simulations have been used to study the stability and activity of enzymes in DES with different water concentrations [253-255]. The structure of hen egg-white lysozyme is significantly destabilized in reline/water combinations, particularly at 50:50 reline: water content, as demonstrated by Kumari *et al*. [254]. The stability and activity of alcohol dehydrogenase in glycerine/water combinations were analyzed by Huang *et al*. [253]. The enzyme's molecular flexibility increased at 10% water concentration, which can affect the enzymatic activity.

6. Similarities of ILs and DESs

Below, the similarities between the components of DESs and ILs are presented. ILs are made up of two ions, an anion, which is negatively charged, and a cation, which is positively charged. The anion can be anything from chloride to tetrafluoroborate to hexafluorophosphate, while the cation is typically a big organic molecule such as imidazolium, pyridinium, or phosphonium [256]. But, DESs consist of a combination of a hydrogen bond acceptor (HBA) and a hydrogen bond donor (HBD) (Figure **5**). When mixed, they generate a eutectic mixture, which has a far lower melting point than any of the individual components [257]. Their commonality is that ionic species constitute both DESs and ILs. The main distinction is that DESs are made up of two components that interact through hydrogen bonds to form a eutectic mixture, whereas ILs usually have a single cation-anion pair. If we want to talk about the volatility related to these two solvents, we can say that DESs and ILs are non-volatile; that is why their vapor pressures are extremely low. Due to their volatility and ease of evaporation, traditional organic solvents (such as acetone or toluene) lead to environmental and safety concerns. Conversely, the low volatility of DESs and ILs lowers the risk of exposure to toxic fumes and solvent evaporation [258-260].

The key advantage of both types of solvents is that they are safe due to their low volatility, which eliminates the need for stringent containment or ventilation methods, especially in closed systems. They are also more environmentally friendly because they do not contribute to air pollution.

Another similarity between ILs and DESs is their use in the electrochemical field. Even at high voltages, ILs can be utilized in batteries, supercapacitors, and fuel cells due to their broad electrochemical window. For energy storage devices, where the solvent must maintain its stability under high energy conditions, this is crucial [30, 261]. Similarly, a large number of DESs have a broad electrochemical window that enables them to be used in electrochemical processes such as battery research and electroplating. However, the electrochemical characteristics of DESs are not as well studied in commercial applications as those of ILs because DESs are a newer field of study [262-264].

High solvation power is another common characteristic between ILs and DEs. ILs are very effective at dissolving hard-to-dissolve substances like metal salts, cellulose, polymers, and biomolecules like DNA or proteins. Even gases like carbon dioxide can be dissolved by them [265, 266]. Additionally, DESs are good solvents, and in industrial operations, some DESs have been employed to dissolve organics, metallic compounds, and lignocellulosic biomass. However, compared to ILs, the range of solvating power may not be as extensive or well-researched [80]. In general, the high solvating power of ILs and DESs allows them to dissolve a variety of materials, including metals, biomolecules, and both polar and non-polar chemicals. That's why they are adaptable solvents in a range of chemical reactions.

Another similarity between ILs and DEs is their environmental friendliness. Although many ILs are safer and better for the environment than conventional organic solvents, not all ILs are non-toxic. Certain ILs can be hazardous or non-biodegradable, particularly if they contain particular anions or cations. Non-toxic and biodegradable green ILs are being designed with increasing effort, and their use in eco-friendly applications is developing [267, 268]. Since many of the ingredients used to make DESs are non-toxic, biodegradable, and derived from naturally occurring materials, DESs are regarded as greener solvents. Because urea and choline chloride, for instance, are naturally occurring and non-toxic, DESs are a desirable choice for applications involving green chemistry [269, 270]. ILs and DESs have tunable properties (Figure **6**). Because different cations and anions can be used, ILs' characteristics (such as solvent strength, polarity, ionic conductivity, etc.) can be changed to meet particular requirements. Imidazolium-based ILs, for instance, typically have distinct characteristics from ammonium-based ILs [31, 271]. Altering the specific HBD or HBA used, as well as the relative proportions of the HBD and HBA, can also modify the attributes of DESs. This allows for a great deal of control over the solvent's polarity, viscosity, and dissolving power [272-274]. As a conclusion from this discussion, it can be stated that both DESs and ILs have tunable properties, which means that by changing the ratios and components in DESs or the cation/anion combination in ILs, their attributes (such as conductivity, solubility, and viscosity) can be modified.

7. Differences Between ILs and DESs

ILs and DESs differ from each other in terms of composition and structure (Figure **6**). Usually comprise a single ionic pair (an anion and a cation) that, at room temperature or close to it, forms a stable liquid phase. As mentioned above, the anions can include halides, tetrafluoroborate, and hexafluorophosphate, whereas the cations are frequently big organic molecules like imidazolium, pyridinium, or ammonium [275, 276]. DESs are combinations of two or more substances, typically an acceptor (HBA) and a donor (HBD). Together, these elements create a eutectic mixture, which is lower temperature than the melting points constituent parts [277, 278]. (The difference in the freezing point at the eutectic composition of a binary mixture of A + B compared to that of a theoretical ideal mixture, ΔTf, is related to the magnitude of the interaction between A and B. The larger the interaction; the larger will be ΔTf. (Figure **5**). A common example is the formation of a DES with a significantly lower melting point than either component alone when urea (a hydrogen bond donor) and choline chloride (a salt) are combined.

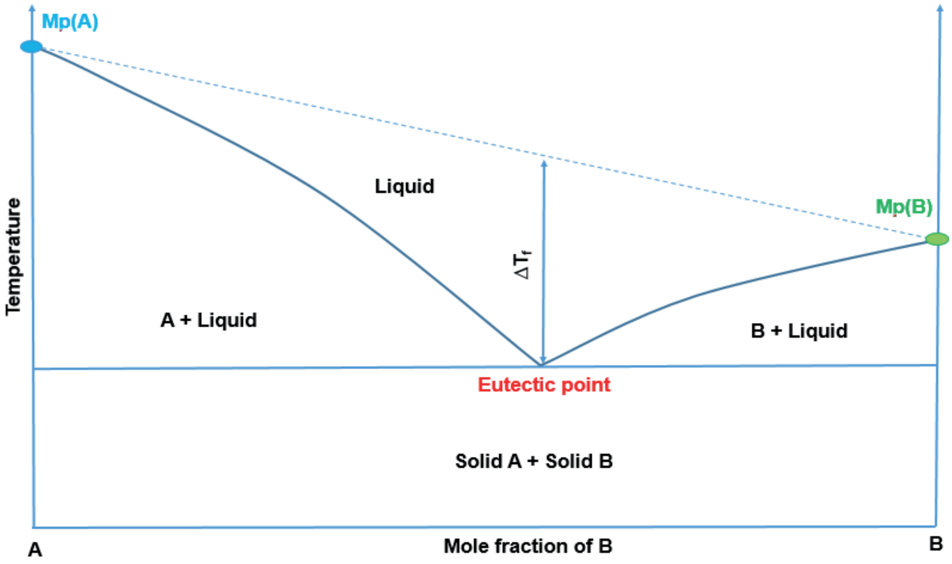

Figure 5: Schematic representation of a eutectic point on a two-component phase diagram [279].

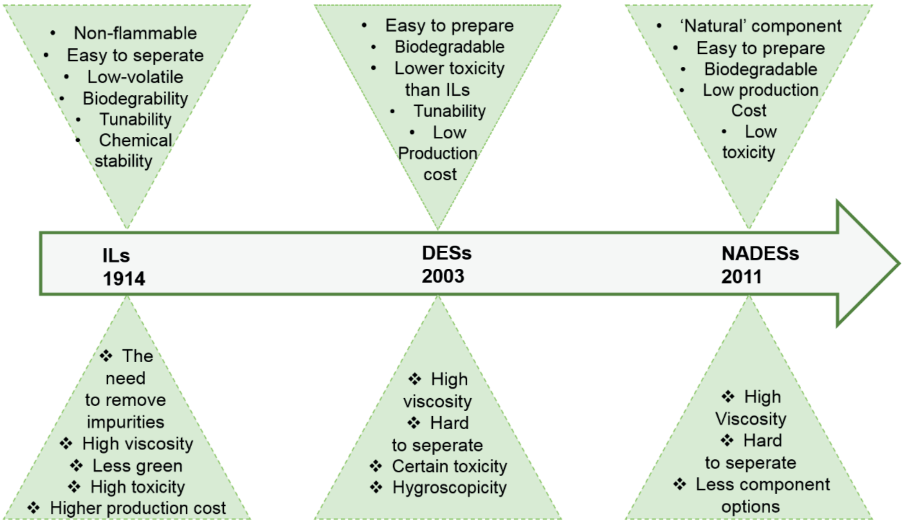

Figure 6: Similarities and differences between ILs and DESs [294].

Another difference between these two solvents is the way they are formed and synthesized. ILs are created by mixing an anion and a cation directly; the end product is usually a single, stable liquid phase. The synthesis often involves combining two pre-made ionic compounds, which calls for more accuracy and may entail costly or hazardous chemicals [280, 281]. But DESs are created when a donor and an acceptor (such as salt and urea or other tiny molecules) are mixed; the resulting eutectic mixture has a lower melting point at a particular ratio. DESs are typically easier to synthesize and frequently involve easily accessible, reasonably priced, and non-toxic substances [282].

Depending on the cation and anion combination in ILs, some of them can have a high viscosity while others have a very low one. In contrast to conventional solvents, many ILs, particularly those containing bigger cations, are renowned for having a comparatively high viscosity [283]. In the case of DES, these solvents often have lower viscosity than most ILs, which might make them easier to work with and better suited for applications requiring easy flow properties [284, 285].

The thermal stability of ILs and DESs is another difference between them. ILs are typically highly thermally stable and, depending on their makeup, can tolerate temperatures of up to 300–400°C [286]. Deep eutectic solvents have lesser thermal stability than ILs, although they are generally thermally stable. Depending on the components, their decomposition temperature is typically lower. For example, as compared to conventional ILs, urea-based DESs have a lower thermal decomposition threshold [287, 288].

The chemical structure and diversity of these solvents vary depending on the starting material used. ILs possess more structural diversity, with an enormous number of potential cation-anion pairings that can result in different properties (e.g., phosphonium-based, imidazolium-based, or pyridinium-based ILs) [281, 289]. Deep eutectic solvents, however, because they must create a eutectic mixture, their diversity is somewhat limited, and they often contain simpler components (such as salts and tiny organic molecules). Although the selection of HBD and HBA is extensive, it is typically less varied than that of ILs [290, 291].

The last difference discussed in this topic is the cost associated with ILs and DES. ILs are frequently more costly to manufacture because of the intricacy of synthesis and the need for high-purity ingredients, particularly those intended for particular uses or custom-made formulations [292]. However, deep eutectic solvents are frequently less expensive to create since they employ easily accessible and reasonably priced starting materials such as urea, choline chloride, or other small organic molecules. This makes DESs more affordable for certain applications [293].

8. Conclusion and Future Perspectives

The quest for green solvents is a rising topic contributing to the goals of green chemistry. Deep eutectic solvents constitute the most considered and investigated solvents nowadays. These solvents possess quite interesting properties, thus increasing their possibility to replace other conventional solvents in numerous academic and industrial sectors. Despite the undeniable benefits of both eutectic and deep eutectic solvents, caution must be used in their definition, design, and application, as well as in understanding their limitations. To this end, this paper included the definition of DESs, their preparation and design, their green nature, and their uses.

According to recent research, the DES-mediated biocatalytic approach is an exciting new field with a wealth of opportunities to enhance reaction sustainability and efficiency through improved substrate loading and solubility, enhanced enzyme activity and stability, and the ability to tailor biocatalyst stereoselectivity by making it more straightforward to recover products and recycle reaction participants. DESs are great options for use in environmentally friendly biocatalysis if we take into account all the environmental advantages of using them as solvents, such as low vapor pressure (less air pollution), non-flammability (process safety), and nontoxicity. The fact that DESs are made with extremely inexpensive forming components is another advantage of using them, as commercial applications require process efficiency, but there may be notable discrepancies between them, which could ultimately result in a much higher product price on an industrial scale. Additionally, steps should be taken to recover product chemicals and/or make NADES recyclable.

Despite the work developed over the last decade on understanding DESs, there are still some areas that need to be deepened. The toxicity and/or biodegradability of the DESs are still far from being fully understood in terms of what determines the synergetic effects of the DESs upon their use. The structure-activity relationship of the DESs needs to be explored to establish a more comprehensive way to choose the right DES for a certain application.

Ethical Issue

The authors are aware of and comply with, best practices in publication ethics specifically about authorship (avoidance of guest authorship), dual submission, manipulation of figures, competing interests, and compliance with policies on research ethics. Authors adhere to publication requirements that the submitted work is original and has not been published elsewhere in any language.

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