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Green and sustainable separation of metal ions and synthetic dyes from aqueous solutions using deep eutectic solvents. A mini review

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Abstract: The development of eco-friendly methods for removing hazardous inorganic and organic contaminants (e.g., metal ions, synthetic dyes) from water systems is of great importance for the health and life of humans and animals. Recently, there has been growing interest in the possibilities of using deep eutectic solvents (DESs) in separation processes aimed at removing various pollutants from aqueous solutions. DESs are typically non-toxic, biodegradable, and can be synthesised using simple methods. Moreover, the components used in DESs synthesis, often considered “green” solvents, can be derived from natural sources. DESs are generally recyclable and relatively cheap. This review highlights recent advancements (mainly from 2023–2024) in the application of various DESs for the removal of metal and metalloid ions, as well as synthetic dyes, from aqueous solutions using solvent extraction (SE) and membrane separation (MP). It also includes critical comments on the limitations of current methods and their potential environmental impacts.

Introduction

The systematic growth of the global population and the dynamic development of various industries are closely linked to the emission of numerous substances resulting from human activity into the environment, many of which may pose a serious threat to living organisms even in relatively low concentrations. Environmental pollution from anthropogenic contaminants is of particular importance in relation to water systems, as the availability of drinking water has been steadily declining for years, while the demand for fresh water is projected to continue increasing. It is estimated that nearly a quarter of the world's population will face drinking water shortages in the future. High water consumption and diminishing water resources are placing increasing pressure on environmental protection efforts, especially those targeting water systems. These efforts include, among others, the prevention of pollutant emissions and the development of sustainable methods for the effective removal of hazardous substances from contaminated waters (Jeong et al. 2023, Bayabil et al. 2022, Abedpour et al. 2023, Srivastav et al. 2024).

However, removing contaminants from various types of water bodies (e.g., surface water, groundwater, brine, industrial wastewater) is often challenging due to the wide variety of substances with diverse chemical properties. In

general, aqueous contaminants can be divided into three groups: organic pollutants (e.g., pharmaceuticals, synthetic dyes, pesticides, agrochemicals, phenolic compounds), many of which undergo reactions in water that can result in the formation of new, sometimes more toxic products; inorganic pollutants (e.g., metal and metalloid ions), which are typically soluble, bioavailable, and persistent in ecosystems, posing long-term threats to living organisms; microbiological contaminants (e.g., bacteria, fungi). These groups are diverse, and their properties vary widely. Nevertheless, many of these contaminants are toxic to plants and animals when released into aquatic environments and can cause serious health issues in humans, including endocrine disorders and cancers (Jeong et al. 2023, Bratovic 2023).

To address these issues, new pollutant removal methods are continually being developed, often by modifying well-established techniques. For example, the removal of synthetic dyes and metal ions can be achieved using traditional methods (e.g., adsorption, membrane separation, solvent extraction, chemical precipitation), enhanced techniques (e.g., adsorption based on novel metal-organic framework materials or nanoparticles), as well as hybrid technologies that combine conventional and advanced treatment methods (Ilame and Ghosh 2022, Sriram et al. 2022, Kaczorowska et al. 2023b, Date and Jaspal 2024, Albrektienė-Plačakė and Paliulis 2024, Białowas

et al 2024). Hybrid methods typically mitigate the drawbacks of individual techniques and enable efficient separation processes. When developing new water treatment methods, attention is not only paid to their effectiveness but also to factors such as cost, scalability (e.g., for industrial applications), and to their potential impact on the environment (Ealias et al. 2024). Efforts are being made to develop methods that are eco-friendly and fit into the trend of ‘green chemistry’ (GC).

A key goal of GC is the sustainable use of natural resources without the introduction of harmful materials. One of the main goals of GC is to maintain natural resources on earth without using harmful materials. This can be achieved in part by replacing dangerous toxic substances with safer substitutes (Abdussalam-Mohammed et al. 2020). Among the eco-friendly, bio-degradable and green materials, which significantly improve the efficiency of the ‘green’ synthesis of various materials, include, among others, nanofluids, ionic liquids (ILs) and deep eutectic solvents (Alqahtani, 2024). DESs, which fully align with the principles of GC, have been applied in various fields such as electrocoating and energy storage (Viyanni and Sethuraman, 2024). As promising alternatives to toxic organic solvents, DESs are gaining importance in analytical chemistry, particularly in solvent- and sorbent-based extraction processes (Santana-Mayor et al., 2021). They have also been successfully used as adsorbents for the removal of selected volatile organic compounds (Stupek et al., 2020).

This paper presents the latest achievements (mainly from 2023–2024) in the utilization of DESs for the separation of hazardous and valuable metal ions, as well as synthetic dyes, from aqueous solutions (e.g., model solutions, leachates, and wastewater) using solvent extraction and membrane-based methods. The main advantages and limitations of the developed techniques are discussed, with particular emphasis on their potential impact on the environment.

Brief characteristics of deep eutectic solvents

Various definitions have been used to describe DESs, which have been systematically refined as knowledge about the properties of these substances has grown. The word ‘eutectic’ originates from the ancient Greek ‘*eútēktos*’, meaning ‘easily melted’. The term ‘eutectic solvent’ refers to the liquid mixture resulting from the combination of solid components, where a liquid phase forms on establishing a solid–liquid equilibrium (i.e., a eutectic system). This phase is characterised by a decrease in the melting point compared to solid precursors. In the case of DESs, the melting point depression is significantly lower than the predicted eutectic temperature (assuming ideal thermodynamic behavior of the liquid phase formed by DESs components), thus, the term ‘deep’ is additionally used in naming these substances. It has been proposed that ‘deep’ should be used when the melting point of the resulting liquid is at least half that of the component with the lowest melting point (Prabhune and Dey 2023; Rao, 2023). Furthermore, for DESs formed from primary metabolites (e.g., organic acids, amino acids, sugars, etc.) the additional term ‘natural deep eutectic solvents’ (NADESs) has been introduced (El Achkar, 2021).

DESs usually consist of two main components: a hydrogen bond donor (HBD) and a hydrogen bond acceptor (HBA). However, they can also be formed from three different molecules, with hydrogen bonding, electrostatic interactions, or van der Waals interactions playing a key role in their formation. The properties of DESs can be ‘tailor-made’ by selecting appropriate compounds to act as HBAs and HBDs, adjusting their molar ratios, and using them in combination with a co-solvent (Prabhune and Dey 2023). Based on their composition, DESs have been classified into five basic types, formed from the following mixtures: quaternary ammonium salts and metal chlorides (Type I); quaternary ammonium salts and metal chloride hydrates (Type II); choline chloride and various HBDs (e.g., alcohols, amides, carbohydrates) (Type III); metal chlorides with neutral organic molecules as HBDs (Type IV); and neutral organic molecules acting as HBD and HBA, forming hydrogen-bonding networks (Type V). Type III and V DESs are of particular interest not only because they are considered ‘green’ (non-toxic, biodegradable, and sustainable), but also because they can be derived from natural sources, are non-flammable, non-volatile, thermally stable, recyclable and inexpensive (Prabhune and Dey 2023, Rao 2023, Majidi and Bakhshi, 2024).

Application of DESs for metal ions separation

Metal ions, especially heavy metal ions, are among the most dangerous pollutants in aquatic ecosystems, mainly due to their toxicity, high solubility in the aquatic environments, and bioavailability. They pose a threat to the health of various organisms, including humans. Depending on factors such as the type of contaminant, its concentration, and duration of exposure, heavy metal ions can damage multiple bodily systems (e.g., nervous, respiratory, digestive), and may also lead to mental dysfunction or cancer. Therefore, it is important to develop efficient and environmentally safe methods for removing heavy metal ions from aquatic environments and aqueous solutions resulting from human activities (e.g., surface water or various types of wastewater). Many novel methods for separating metal ions, developed in recent years, are based on solvent extraction and membrane processes (Majidi and Bakhshi 2024, Shrestha et al. 2021, Alguacil and Robla 2022).

Solvent extraction methods

Solvent extraction methods, based on differences in the solubility of various components in immiscible liquid phases, are among the most frequently used techniques for separating metal ions from solutions. This is largely due to their typically high efficiency, operational simplicity, and the potential use of more environmentally friendly solvents, such as DESs (Alguacil and Robla 2022). However, not all deep eutectic solvents are suitable for solvent extraction aimed at removing contaminants from aqueous solutions. For example, hydrophilic DESs, which form a homogeneous phase with water, cannot be used for this purpose. Consequently, new hydrophobic DESs, capable of forming two-phase systems with aqueous pollutant-containing solutions (e.g., metal ions), are systematically developed (Majidi and Bakhshi 2024, Shrestha et al. 2021). Recently, there has also been growing

Table 1. Examples of the application of various DESs for the separation of metal and metalloid ions from different solutions by solvent extraction (years 2023–2024, %E-extraction efficiency)

DESs composition/ reference	Separated metal ions/ type of solution	Main advantages of the method
Alkyl (hexyl, nonan) ethylenediaminium and menthol, (Wang and Hua 2024)	Cu(II), Co(II), Ni(II), model aqueous solutions	The %E of both DESs exceeded 90% in case of Cu(II) and Ni(II) ions. DESs can suitably collect trace metal ions.
Decanoic acid and lidocaine based DES dissolved in heptane, (Ola and Matsumoto 2024)	Au(III), Pt(IV), Pd(II), model aqueous solutions	Complete extraction of Au(III) (98.2%) and Pd(II) (100%), (with 500 and 300 g/L of DES, respectively).
Choline chloride and sulfamide used to form eutectic dissolution reagent with dibromohydantoin, (Feng et al. 2024)	Au(III), model solution	DES dissolution reagent allows for recycling Au(III) up to 6 times (dissolution rates >95%). In oxalic acid solution - formation of Au ⁰ .
Menthol and salicylic acid DES, used in microextraction assisted ultrasound and vortex equipment, (Mafakheri et al. 2024)	As(II), Cd(II), Hg(II), and Pb(II), water and food samples	Method can be applied to determine heavy metals in water and food samples in the ultra-trace range (recovery values of 93–100%).
Oleic acid and 1-butyl-3 methylimidazolium chloride used as solvent in extraction with tributyl phosphate (TBP), (Zhao et al. 2024)	Y(III), Sr(II), model aqueous solution with HCl	In two-stage extraction 95.06% of Y(III) was selectively separated from a model solution, Y(III) purity of 98.55% in the final product.
DESs based on 1,10-phenanthroline and natural phenolic extract thymol, (Crema et al. 2023)	Cd(II), Mn(II), Co(II), Ni(II), Cu(II), Zn(II) and Pd(II), a range of solvents; Li(I), Mn(II), Co(II) and Ni(II) in aqueous solutions	The properties of the DES depended on the concentration and nature of co-extracted polar solutes. DES allows for extraction of transition and platinum group metals and their separation from lanthanides and alkaline earth ions.
TOPO and decanoic acid, (Cruz et al. 2024)	Ce(IV), La(III), Mn(II), Ni(II), Co(II), nickel metal hydride battery leachates	In optimal experimental conditions application of DES enabled recovery of about 87% of La(III) and 98% of Ce(IV).
Choline chloride and anhydrous oxalic acid DES, with water as co-solvent and diluent, (Liu et al. 2024)	In(III), Zn(II), Fe(III), indium-bearing zinc ferrite leachates	Under specific leaching conditions, the leaching efficiencies of indium, zinc and iron were 96.27%, 95.55 % and 96.33%, respectively.
Tetrabutylammonium bromide and capric acid, or oleic acid, (Kunasekaran et al. 2024)	Cu(II), Hg(II), synthetic contaminated water streams	DESs can be promising extractants for removing Cu(II) and Hg(II) ions from the water stream. Capric acid-based DES showed %E of 98.85% for Hg(II) ions and of 54.25% for Cu(II) ions.

research into the use of various DESs for the separation of valuable rare earth elements from different media (Shuping et al. 2025). Recovering valuable elements from secondary sources (e.g., leachates from the hydrometallurgical treatment of e-waste or industrial by-products) is important not only for environmental protection (e.g., reducing natural resource extraction, minimizing landfill waste) but also for economic reasons (Martín et al. 2023). Table 1 presents recent research examples on the use of various DESs for the extraction of metal ions from different solutions (e.g., aqueous solutions, acidic leachates).

As shown in Table 1, recent studies have primarily utilized Type V DESs, composed of two neutral organic molecules acting respectively as HBD and HBA and interacting via hydrogen bonding, for the removal of hazardous metal ions from aqueous solutions and the recovery of valuable metal ions (e.g., precious metal ions, rare earth metal ions, and

lithium ions. Lithium, notably, is a key raw material in the production of lithium-ion batteries) (Crema et al., 2023). Many of these DESs are formed using relatively simple and readily available organic acids (e.g., mixtures of decanoic acid and lidocaine or salicylic acid and menthol) (Ola and Matsumoto 2024, Mafakheri et al. 2024, Kunasekaran et al. 2024). These combinations can be classified as natural deep eutectic solvents (NADESs). The growing interest in NADESs for metal ions extraction is largely attributed to their favorable properties - negligible toxicity, water insolubility, and low production costs. In general, NADESs are considered even more environmentally friendly than other types of DESs (Mafakheri et al. 2024). To improve environmental safety, naturally occurring compounds such as thymol or menthol are increasingly used in NADES synthesis (Wang and Hua, 2024; Mafakheri et al., 2024; Crema et al., 2023). However, not all NADESs are suitable for the separation of metal ions because of poor extraction performance

(e.g., DES composed of choline chloride and betaine). As a result, there is ongoing development of new NADESs aimed at improving extraction efficiency. One promising approach involves incorporating well-known chelating agents, such as various alkyl derivatives of ethylenediamine, into DESs, often alongside other components like menthol. These DESs have demonstrated high efficiency (over 90%) in the extraction of Cu(II) and Ni(II) ions and are also effective for separating trace metals (Wang and Hua 2024). Another strategy is to use chemical compounds that have been successfully used as carriers in membrane processes or as extractants in solvent extraction (SE) (Kaczorowska, 2022). Recent examples of such components used to create effective DESs include TOPO and TBP (Zhao et al. 2024; Cruz et al. 2024). However, before these novel DESs can be widely applied in industrial processes (e.g., for wastewater treatment), it is necessary to understand their mechanisms of action – particularly how they bind specific metal ions – and to thoroughly characterise their physical properties. One current research direction focuses on quantum-chemical methods for this purpose (Kumar et al. 2024, Patel et al. 2024). Developing simple and robust models for predicting specific DES properties (e.g., density) could significantly accelerate the formulation of optimal solvents for targeted separation process. Finally, the efficiency of metal ion extraction using DESs also depends heavily on experimental conditions, such as the type of metal ions, the solution composition, and other system parameters (Zhao et al. 2024). Determining both the optimal DES composition and the optimal extraction conditions (the influence of individual parameters, their mutual dependencies) can be time-consuming and complex. This remains one of the reasons why, despite many advantages, deep eutectic solvents are still primarily used for metal ion extraction at the laboratory scale.

Membrane separation

Membrane processes based on the application of membranes (i.e., thin layers constituting permeable or semipermeable phases, acting as barriers between two adjacent liquid phases and controlling the transfer of specific substances between them), have often been successfully used for the separation of metal ions from aqueous solutions. Among the numerous types of membranes, polymer membranes (PMs) with a polymer matrix, such as polymer inclusion membranes (PIMs, containing a liquid phase held within the polymeric network), are becoming increasingly popular because their properties can be easily modified by changing membranes components (i.e., quantitatively and qualitatively, the content of polymer, ion carrier, plasticiser), which, in consequence, enables, among other benefits, control of the separation processes efficiencies. Furthermore, the advantages of PIMs include the possibility of using environmentally safe components for membrane formation, performing simultaneous extraction and back-extraction, and the potential for membrane reuse. As a result, PIM-based separation methods are considered eco-friendly and economical, and may potentially be used on a larger industrial scale in the future (Kaczorowska 2022, Baoying et al. 2023, Wang et al. 2023).

Recently, attempts have been made to use various DESs as carriers in PMs intended both for the recovery of valuable metal ions (e.g., Au) and for the removal of hazardous metal

ions (e.g., Cu) from different solutions. For example, a novel, homogenous, and flexible PIM containing a hydrophobic DES based on tetraoctylphosphonium bromide ($P_{8888}Br$) and D2EHPA as a carrier, poly(vinylidene fluoride) (PVDF) as a polymer matrix, and 2-nitrophenyl ether (2-NPOE) as a plasticiser, was applied for the recovery of Au(III) ions from aqueous mixed-metals hydrochloric acid solutions and showed excellent selectivity towards gold ions (about 99% of Au(III) ions were selectively transported to the receiving phase) (Liu et al. 2021).

Polymer membranes consisting of a DES based on diacetamide and Aliquat 336 as a metal-binding agent, poly(vinyl chloride) as a polymer matrix, and bis(2-ethylhexyl) adipate (ADO) as a plasticiser have been used for the removal (via adsorption) of copper(II) and zinc(II) ions from aqueous solutions obtained by leaching computer pins with concentrated nitric acid, achieving removal of about 97% of Cu(II) and 96% of Zn(II) ions. Moreover, after comparing the efficiency of membranes containing either DES or the ionic liquid Aliquat 336 as carriers, it was shown that the use of the deep eutectic solvent carrier allowed for a significant reduction in the duration of the separation process, which could have a significant impact on process costs, especially for large-scale applications (Kaczorowska et al. 2023a). So far, deep eutectic solvents have been used much less frequently in polymer-membranes-based processes than in solvent extraction. However, the reported results, indicating high efficiency and selectivity (which is crucial in the separation of specific ions from multicomponent solutions, such as various leachates) of PMs containing DESs in metal ion separation, suggests that further research will be conducted in this direction. In terms of environmental protection, it is also important that separation methods based on PMs (inclusion or adsorption) containing DESs are eco-friendly and inexpensive, and membranes that can be reused after regeneration.

Application of DESs for synthetic dyes separation

Dyes are unsaturated synthetic organic compounds that absorb electromagnetic radiation in the visible wavelength range and are usually soluble in water and oil (Nithya et al. 2021). The classification of dyes based on their ionic nature includes non-ionic dyes (disperse and vat dyes), anionic dyes (direct, acid, and reactive dyes) and cationic dyes (basic dyes) (Benkhaya et al. 2020, Sriram et al. 2022). Disperse dyes are sparingly soluble in water and must be dispersed with the aid of surfactant. They are small, polar molecules that do not possess charged anionic or cationic groups in their structure and usually contain anthraquinone or azo groups. Vat dyes, on the other hand, are water-insoluble, polycyclic aromatic molecules based on quinone structure (keto forms) (Moody et al. 2004, Zahid et al. 2021, Khodabandeloo et al. 2023). Direct and reactive dyes exist as ions in water and are highly water-soluble (Zhang et al. 2024). Acid dyes, which are based on azo, anthraquinone, triphenylmethane, and Cu-phthalocyanin chromophoric systems, are also soluble in water due to the presence of acidic groups (Chavan et al. 2011). Cationic dyes can dissociate into positively charged ions and negative counterions in aqueous solutions (Chakraborty et al. 2022). These dyes do not degrade

easily and can accumulate throughout the food chain, making them more toxic than anionic and non-ionic dyes (Hussain et al. 2022). Due to the complex structure of various dyes and their toxic and carcinogenic properties, discharging such pollutants into the environment can be very harmful. Therefore, there has recently been an intensive search for new methods that enable the effective separation of different types of dyes from aqueous solutions, while also being safe for the environment (Kuśmirek et al. 2023). Among the well-performing and eco-friendly methods are solvent extraction and membrane processes based on the utilisation of various DESs.

Deep eutectic solvents as synthetic dyes extractants

Different DESs can be highly effective in the removal of selected synthetic dyes (non-ionic, anionic, cationic) from aqueous solutions via solvent extraction processes. For example, DESs prepared with quaternary ammonium salts and various hydrogen bond donors (HBD) show high selectivity and efficiency in removing cationic and anionic dyes. DESs composed of tetrapropylammonium bromide (TPAB) or tetraethylammonium iodide (TEAI) with 1-nonanol (1N) as the HBD have been successfully used for the extraction of methyl orange (MO) from the aqueous solutions. The obtained DESs, 1N:TPAB and 1N:TEAI, enabled much faster extraction (15 minutes) compared to pure HBD alone, where removal of MO was 80% after 5 hours of the process (Omar et al., 2022). Moreover, non-toxic DESs have also shown high extraction efficiency for Alizarin Red S (ARS). For example, Yasir et al. used a DES composed of triocylamine (TOA) and salicylic acid (SA) for the removal of alizarin from high-concentration aqueous solutions, finding that under optimal conditions about 98% of the dye was removed in just 5 minutes of the process. Additionally, they examined the influence of synthesized DES on human cells and reported that it showed minimal cytotoxicity, depending on the DES concentration (Yasir et al. 2022). A DES consisting of choline chloride and decanoic acid was also successfully used to remove methyl violet (MV) from river water using ultrasound-assisted liquid-liquid microextraction (UALLME). The results showed that the developed method was characterised by high robustness, a shortened extraction time, and the use of green DES that did not negatively affect the environment (Haq et al. 2023).

Martínez-Rico et al. (2024) synthesized a deep eutectic solvent based on quaternary ammonium compound and decanoic acid using a method that did not lead to atmospheric emissions. They used this DES to remove selected anionic dyes (reactive red 29 dyes (RR29), direct black 19 (DBk19), acid blue 80 (AB80)) from aqueous solutions and found that the purified water could be reused in another extraction process (Martínez-Rico et al. 2024). Vat dyes, such as indigo carmine (IC), can also be efficiently removed from various matrices by adsorption or solvent extraction processes. Kizil et al. applied ultrasonic-assisted hydrophobic deep eutectic solvent-based dispersive liquid-liquid microextraction to determine indigo carmine in food samples, using a DES composed of tetrabutylammonium bromide as HBA and decanoic acid as HBD (Kizil et al. 2024). Chen et al. synthesised DESs of types III and V and applied them in vortex-assisted liquid-liquid

microextraction (VALLME) to remove anionic (tartrazine, TA) and cationic (phenosafranin, PF) dyes. They found that both DESs (one based on trioctylmethylammonium chloride and thymol, and the other based on decanoic acid and thymol), enabled effective and selective separation of dyes from aqueous solutions (Chen et al. 2024).

DESs of type V, consisting of non-ionic compounds, show great potential for removing contaminants from aqueous solutions. They do not contain chlorides and can have various degrees of hydrophobicity (Duque et al. 2023). Blanco et al. investigated the performance of a type V DES composed of thymol and decanoic acid as an extractant for Congo red (CR) removal. The extraction efficiency depended on the molar ratio of HBA to HBD, with a 2:1 ratio showing very high efficiency (above 96%) over three consecutive extraction cycles (Blanco et al. 2022). Hassan et al. synthesised DESs consisting of trioctylphosphine as HBA and salicylic acid (SA) as HBD to remove methylene blue (MB) from aqueous solutions. The extraction efficiency of MB decreased with increasing temperature and the mixing time was crucial in the solvent micro-extraction processes, both at the laboratory and industrial scales. Additionally, the DES exhibited minimal cytotoxicity according to ISO10993-5 standards (Hassan et al. 2022).

The review shows the high potential of DESs for the separation of selected dyes from aqueous solutions via solvent extraction. Although SE is one of the separation methods that can contribute to the sustainable removal of pollutants from wastewater, it also has several disadvantages, such as the need to use solvents (often toxic), as well as high financial costs and energy consumption. In recent years, DESs have been successfully used to eliminate or reduce the use of toxic reagents typically required during solvent extraction processes (Chemat et al. 2020).

Membrane separation

As with metal contaminants in aqueous solutions, deep eutectic solvents have also been used in membrane processes to remove synthetic dyes. Santhosh et al. converted waste polysulfone (W-PFS) into multifunctional carbon materials using a DES composed of choline chloride and FeCl_3 , for water treatment applications, specifically for the removal of malachite green (MG). The resulting membranes showed recyclability for up to 10 cycles, maintaining a yield of 92.1% for MG removal (Santhosh et al. 2024). A DES composed of *N,N*-diethylethanolammonium chloride and ethylene glycol was used to enhance the polymer membranes prepared with poly(vinylidene fluoride-co-hexafluoropropylene) and carbon nanospheres, which were next utilized for the separation of methyl orange. The prepared PVDF-co-HFP/CNS membrane showed higher anti-biofouling properties than the pure PVDF-HFP membrane (Aljumaily et al. 2022). Moreover, natural polymers modified with DESs have also been used to remove selected dyes from aqueous solutions. Chitosan modified with a eutectic solvent significantly improved the removal of acid blue 80 (AB80) compared to pure chitosan (Blanco et al. 2023). Furthermore, type V DESs have been used to prepare nanocomposites with graphitic carbon nitride, which were utilised to achieve high-yield removal of malachite green from aqueous solutions (Kurtulbaş et al. 2024). Table 2

Table 2. Examples of various composition DESs used to remove selected dyes (years 2023–2024)

Dye class	Name and structure of the dye	Composition of DES / reference	Removal efficiency/ methods
Cationic dyes	MV 	Choline chloride and decanoic acid (Haq et al. 2023)	~ 99%, UALLME
	PF 	Decanoic acid and thymol (Chen et al. 2024)	99.9%, VALLME
	MG 	Choline chloride and FeCl ₃ (Santhosh et al. 2024) Citric acid and lactic acid (Kurtulbaş et al. 2024)	515 mg/g, MP 59.52 mg/g, MP
Direct dyes	DBk19 	Tetrabutylammonium chloride and decanoic acid (Martínez-Rico et al. 2024)	67.85%, SE
Acid dyes	TA 	Triethylmethylammonium chloride and thymol (Chen et al. 2024)	99.3%, VALLME
	AB80 	Tetrabutylammonium chloride and decanoic acid (Martínez-Rico et al. 2024)	60.86%, SE
		Choline chloride and urea (Blanco et al. 2023)	>98%, MP
Reactive dyes	RR29 	Tetrabutylammonium chloride and decanoic acid (Martínez-Rico et al. 2024)	83.27%, SE

shows examples of various DES compositions used to separate selected dyes by SE and PMs based methods.

As shown by the data presented in Table 2, the application of DESs as extractants in SE and as carriers in PMs intended for the removal of synthetic dyes from various solutions has yielded excellent results. Considering other important advantages of DESs, such as non-toxicity and biodegradability, their utilization in larger-scale separation processes may represent a promising new direction for sustainable synthetic dye removal in wastewater treatment processes.

Conclusions

Based on the presented examples of the application of different types of DESs in separation processes (solvent extraction and membrane processes) for the removal of metal ions and synthetic dyes from various aqueous solutions, it can be concluded that most of the developed methods exhibited high efficiency under optimal experimental conditions (Ola and Matsumoto 2024, Mafakheri et al. 2024, Liu et al. 2024).

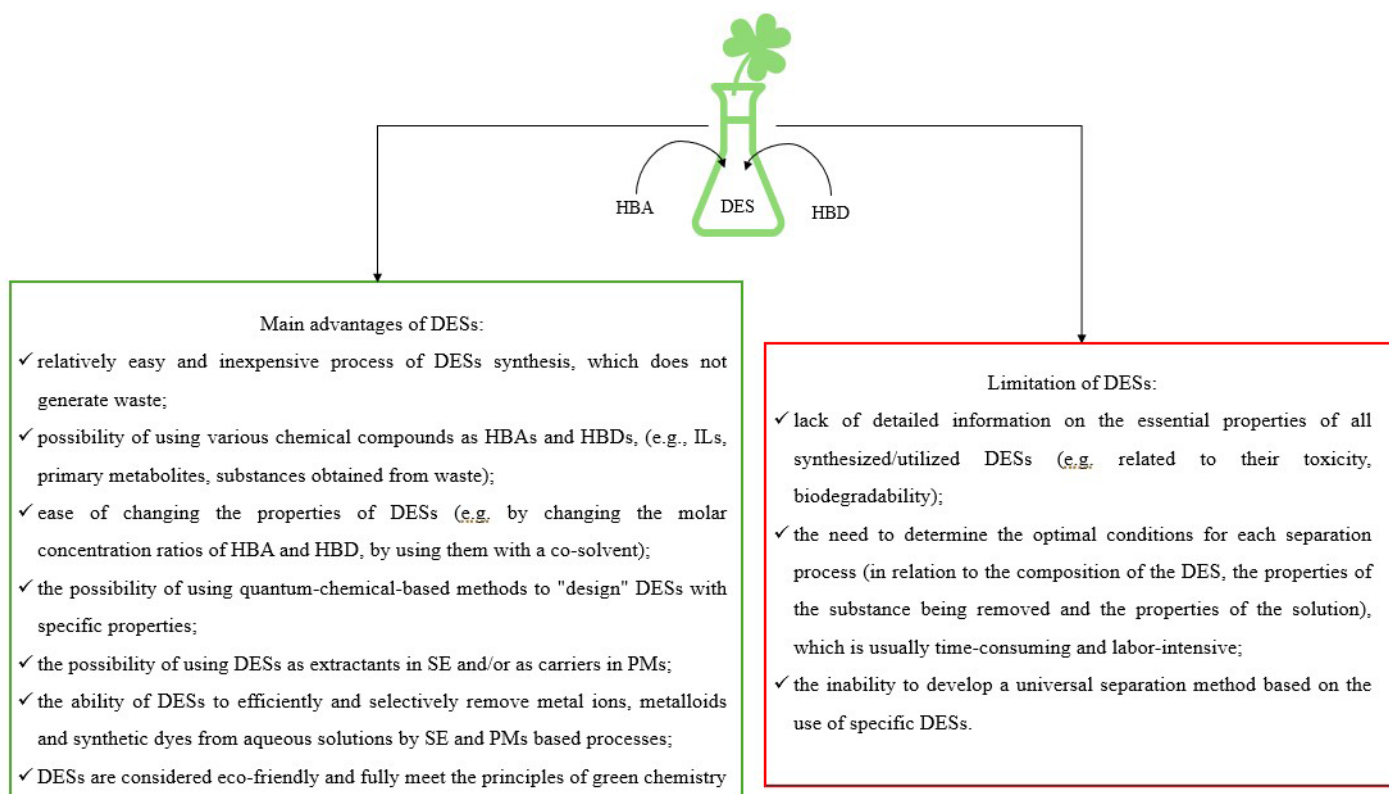


Figure 1. Basic advantages and limitations of DESs in relation to the possibility of separating metal ions and synthetic dyes from aqueous solutions

The high efficiency and selectivity of the separation processes are particularly important, especially considering the potential for applying these methods on a wider, industrial scale in the future, where polluted waters or industrial effluents typically have complex compositions. However, it should be emphasised that determining the optimal conditions for conducting separation processes remains time-consuming and labor-intensive due to the numerous influencing factors (including the qualitative and quantitative composition of DESs) and their mutual interdependence. The inability to develop a universal separation method based on the utilization of specific DES – requiring the optimization of solvent extraction or membrane processes for each specific case depending on the properties of the target substance and solution - remains one of the major limitations. Although efforts have been made to shorten the time needed to determine optimal separation conditions, such as through the application of computational methods to predict the properties of newly designed deep eutectic solvents (Kumar et al. 2024, Patel et al. 2024), most reported studies still focus on the application of DESs for the separation of pollutants from relatively simple model solutions.

It is also important to consider the characteristics of DES used in separation methods and their environment impact. While DESs are generally regarded as non-toxic, biodegradable, obtainable from renewable sources, and easy to synthesise, not all DESs possess these advantages to the same degree. Some DESs are significantly 'greener' than others. It has been reported, that even among widely used DESs containing choline chloride as a hydrogen bond acceptor and various common hydrogen bond donors (e.g., glycerol, ethylene glycol, urea, glucose, lactic acid), differences in

key sustainability characteristics were significant. Moreover, comprehensive data on crucial properties such as toxicity or biodegradability are not always available (Nejrotti et al. 2022).

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Zielona i zrównoważona separacja jonów metali i barwników syntetycznych z roztworów wodnych przy użyciu cieczy głęboko eutektycznych. Mini przegląd.

Streszczenie. Opracowanie przyjaznych dla środowiska metod usuwania niebezpiecznych zanieczyszczeń nieorganicznych i organicznych (np. jonów metali, barwników syntetycznych) z systemów wodnych ma ogromne znaczenie dla zdrowia i życia ludzi i zwierząt. Ostatnio obserwuje się wzrost zainteresowania możliwościami wykorzystania cieczy głęboko eutektycznych (DES), w procesach separacji przeznaczonych do usuwania różnych zanieczyszczeń z roztworów wodnych, ponieważ DES są zazwyczaj nietoksyczne i biodegradowalne i można je otrzymać za pomocą prostych metod syntezy. Ponadto, składniki do syntezy DES, uważanych za „zielone” rozpuszczalniki, mogą być pozyskiwane ze źródeł naturalnych, a ciecze głęboko eutektyczne można zazwyczaj poddawać recyklingowi i są one stosunkowo niedrogie. W niniejszym przeglądzie omówiono najnowsze osiągnięcia (głównie z lat 2023–2024) w zakresie stosowania różnych DES do usuwania jonów metali i metaloidów oraz barwników syntetycznych z roztworów wodnych przy użyciu ekstrakcji rozpuszczalnikowej (SE) i procesów membranowych (MP). W pracy zawarto również krytyczne uwagi na temat ograniczeń opracowanych metod oraz ich potencjalnego wpływu na środowisko.