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Mini Review

Deep Eutectic Solvents (DESs) in Waste Reduction, By-Product Valorization, and Advancing the Circular Economy

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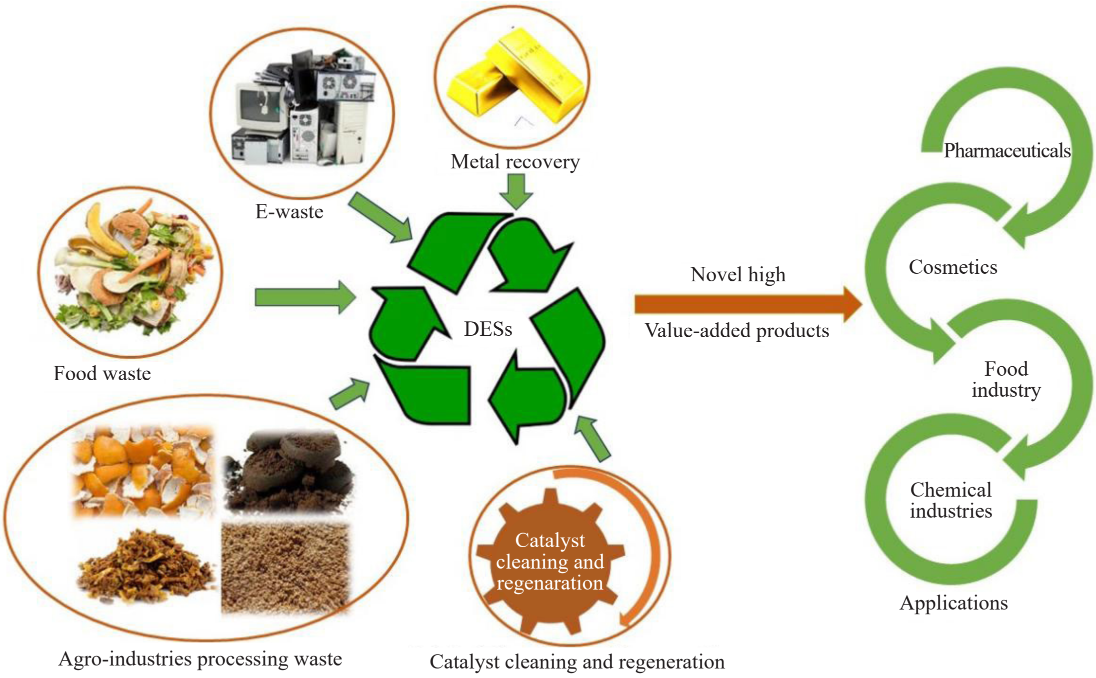

Abstract: The growing demand for sustainable solutions in industrial processes has prompted the exploration of alternative solvents that can reduce environmental impact. Deep Eutectic Solvents (DESs), characterized by their low toxicity, recyclability, and versatility, have emerged as promising candidates for waste reduction, by-product valorization, and the advancement of the circular economy. This mini-review highlights the main applications of

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DESs in various waste management strategies. Firstly, it examines DES as a green, non-toxic, and alternative solvent to conventional solvents, offering a safer and more sustainable option for industrial processes. Secondly, it examines their role in reducing food and agro-industrial waste by extracting valuable bioactive compounds from waste streams and industrial by-products. Thirdly, it discusses the recovery of valuable metals from electronic waste (e-waste), emphasizing the efficiency of DESs in metal extraction while minimizing environmental harm. Finally, it addresses the use of DESs in catalyst cleaning and regeneration, showcasing their contribution to reducing waste in industrial catalytic processes. Overall, this review provides a comprehensive and up-to-date analysis of DESs as pretreatment agents, extraction solvents, catalysts, and material recovery media, compiling the most recent advances in agro-industrial waste valorization, precious metal recovery, and catalyst regeneration. It offers comparative insights into their performance against conventional solvent-based methods and highlights emerging challenges and opportunities for large-scale implementation, emphasizing their potential to advance waste reduction and support a circular economy.

***Keywords*: Deep Eutectic Solvents (DESs), sustainable industrial processes, waste reduction, circular economy, by-product valorization

1. Introduction

Deep Eutectic Solvents (DESs) have emerged as effective green solvents that promote sustainability in industrial processes by reducing waste and optimizing the utilization of by-products.[1] Their unique physicochemical properties, combined with their eco-friendly nature, make them particularly suitable for applications in waste management and resource recovery.[2]

DESs belong to a class of green solvents known for their tunable physical and chemical properties, which allow them to be tailored for a wide range of applications in chemistry, materials science, and industrial processes.[3] They are formed by combining a Hydrogen Bond Donor (HBD) with a Hydrogen Bond Acceptor (HBA), resulting in a eutectic mixture with a significantly lower melting point significantly lower than the individual components.[4-6] A major advantage of DESs over Ionic Liquids (ILs) is their lower toxicity, biodegradability, low flammability, non-volatility, polarity, thermal stability and reduced cost of raw materials.[7] Moreover, they can be synthesized from natural primary metabolites. Recently, DESs have been developed from the combination of primary metabolites and bio-renewable starting materials, such as sugar alcohols, sugars, choline chloride, carboxylic acids, and amino acids. However, the environmental impact of DESs depends on the composition of their selected components. Their stability under various chemical and thermal conditions is crucial for applications such as electrochemistry and catalysis.[8,9]

DESs are increasingly being recognized for their ability to reduce industrial waste, offering a more sustainable alternative to conventional solvents. Traditional solvent-based processes often produce harmful by-products, require high energy input, and pose challenges in recycling. In contrast, DESs are biodegradable, less toxic, and provide significant environmental benefits.[1,10] They are particularly effective in replacing hazardous solvents such as organic chemicals and inorganic acids, which are commonly used in industries such as metal extraction, mining, and chemical production. For instance, DESs can be used to extract valuable metals from electronic waste (e-waste), eliminating the need for toxic chemicals like cyanide, thereby reducing harmful waste and lowering carbon emissions.[11]

Furthermore, DESs show great promise in by-product valorization, which involves recovering valuable compounds from industrial waste streams. Industries such as food production,[12] pharmaceuticals,[6] and chemicals often generate large amounts of by-products during manufacturing, many of which are discarded or underutilized, leading to inefficiency and increased waste.[13] DESs can recover valuable substances from these by-products, including antioxidants, vitamins, and flavors from food waste, or Active Pharmaceutical Ingredients (APIs) from pharmaceutical residues. This process transforms waste into valuable products, reducing overall waste and enhancing materials value.[5]

Additionally, the low melting points of DESs contribute to energy efficiency compared to conventional solvents, enabling more effective processing in various industrial applications, such as extraction, separation, synthesis, and catalysis.[14]

DESs are particularly valuable in waste management, with a notable application in the recovery of valuable metals from electronic and industrial waste.[15] Their ability to dissolve a wide range of compounds, combined with high solubility and selectivity, makes DESs ideal for efficient and safe metal extraction, outperforming traditional methods that use toxic chemicals. In addition, DESs can be used for treating industrial effluents and wastewater,[16,17] helping to dissolve contaminants and purify waste streams, reducing pollution, and supporting circular economy initiatives by promoting the recovery and reuse of resources.

Overall, DESs play a key role in advancing circular economy practices, helping industries shift toward more sustainable and closed-loop systems. The widespread use of DESs can significantly reduce waste generation, lower carbon footprints, and promote the sustainable use of resources, contributing to the global transition toward a more circular and environmentally-conscious economy. This paper explores how DESs contribute to waste reduction, focusing on their role in replacing hazardous solvents, recovering valuable metals from waste streams, and regenerating catalysts. The analysis is supported by multiple research studies and industrial applications.

2. Substitution for toxic solvents: A green alternative

One of the key ways DESs reduce waste is by serving as safer substitutes for hazardous organic solvents, which are often volatile, toxic, and challenging to manage post-use. Conventional solvents—frequently derived from non-renewable petroleum sources—generate large quantities of hazardous waste and pose significant environmental and health risks due to their flammability, volatility, and toxicity.[18,19] In contrast, DESs are synthesized from low-toxicity, biodegradable components, often sourced from natural metabolites, making them an eco-friendlier alternative.[19,20]

A seminal study by Abbott et al. demonstrated the potential of DESs, such as mixtures of choline chloride and carboxylic acids, to replace both ionic liquids and traditional organic solvents in various chemical processes, including extraction, synthesis, and catalysis.[21] This research highlighted DESs' ability to minimize solvent waste by enabling cleaner reaction pathways, reducing the volume of solvent needed, and facilitating solvent recycling.[22]

3. Reduction food waste and agro-industrial waste

Food loss and waste represent a significant global challenge, amounting to approximately $680 billion per year in developed countries and $310 billion in developing nations. On a global scale, around 40-50% of root crops, fruits, and vegetables, 30% of cereals, 35% of fish and 20% of oilseeds, meat, and dairy products are wasted every year. In Europe alone, nearly 88 million tonnes of food are discarded every year, leading to costs of approximately €143 billion.[23] With the global population expected to reach 9.3 billion by 2050, minimizing food waste is becoming increasingly essential. Addressing this issue offers substantial environmental and economic advantages.[24,25]

Similarly, the agricultural and forestry sectors generate considerable waste. Agricultural waste includes residues from crops such as wheat, sugarcane, rice, and olive oil, while forestry waste encompasses materials like cork granules and tree bark. On a global scale, waste production each year includes about 709 million tonnes of wheat straw, 673 million tonnes of rice straw, and 2.96 million tonnes of solid olive waste.[25,26]

Food industry waste primarily consists of organic residues derived from processed raw materials. Due to strict quality standards, the composition of this waste is generally consistent. However, managing and disposing of it presents challenges, as it often exhibits poor biological stability, potential pathogenic risks, high water content, rapid oxidation, and intense enzymatic activity.[1,27]

Despite these obstacles, food industry waste represents a valuable resource. It frequently contains natural compounds with significant potential applications in medicine, health, and nutrition. Strategies aimed at waste reduction and utilization focus on extracting these compounds, facilitating both waste minimization and its transformation into valuable resources.[24]

The increasing focus on reducing food and agro-industrial waste has highlighted the need for sustainable and efficient waste management approaches. Deep Eutectic Solvents (DESs) have emerged as a promising solution, offering innovative methods to reduce waste and convert it into high-value by-products. Due to their significant properties, DESs can be effectively applied in various processes targeting food waste and agro-industrial by-products.[28,29]

For instance, DESs can be utilized to extract a wide range of bioactive compounds such as antioxidants, polyphenols, phenolic acids, vitamins, proteins and essential oils from food waste, including fruit or vegetable peels, seeds, and pulp. These extracted compounds can then be used in products like cosmetics, dietary supplements, or functional foods. In the case of discarded grains and agricultural residues, DESs are effective in extracting sugars such as glucose and xylose, which can then be converted into biofuels or other valuable bioproducts.

Moreover, DESs serve as an environmentally friendly alternative to conventional solvents in food waste recovery. Being biodegradable, non-toxic, and can be reused multiple times, making them a sustainable choice for the valorization of food waste.

Mavai et al. reviewed various extraction methods utilizing DES systems, emphasizing applications of bioactive compounds as antimicrobials, antioxidants, flavorings, functional ingredients, nutraceuticals, additives, and preservatives. The review also discussed the potential of emerging DES technologies for food waste valorization, aligning with sustainable development goals.[30] They explored different eutectic systems for extracting bioactive compounds from food waste, including coffee husk,[31] orange peels,[32,33] Brewer's spent grain,[34] fig by-products,[29] olive waste etc.[35,36]

Mero et al. evaluated two high-performing green solvents for the fractionation of lignocellulosic biomass: Cholinium Arginate (ChArg) as a bio-based Ionic Liquid (IL) and Choline Chloride : Lactic Acid (ChCl : LA) in a 1 : 10 ratio, as a natural deep eutectic solvent, NaDES. Their study compared these green and alternative solvents in the pretreatment of apple fibers, a type of agri-food industry waste.[37]

Agro-industrial waste, including residues from agricultural processes like husks, stems, and leaves, is often underutilized or discarded. DESs offer a solution by enabling the extraction of valuable chemicals and bioactive compounds from such residues, promoting waste reduction and the creation of value-added products.

For example, agro-industrial wastes like crop residues contain cellulose, hemicellulose, and/or lignin, which can be decomposed using DESs to release fermentable sugars. These sugars can be utilized for biofuel production or converted into platform chemicals such as Hydroxymethylfurfural (HMF), furfural, and levulinic acid. Additionally, DESs can break down lignin into phenolic compounds, which have various industrial applications, including in the production of biodegradable resins, plastics, and pharmaceuticals.[1,2]

The applications of DESs in reducing food and agro-industrial waste offer significant environmental and economic benefits. Environmentally, DESs minimize the reliance on toxic solvents that harm ecosystems. They also enable the recycling and reuse of waste materials, decreasing landfill dependency and lowering methane emissions. Economically, the valorization of food and agro-industrial waste through DESs creates opportunities for green chemistry and sustainable product innovation. By transforming waste into valuable products, businesses can develop new revenue streams while reducing the need for extracting raw materials.[30,33]

Procentese et al. reported on the use of DESs to pretreat specific agro-industrial food wastes commonly produced in Europe. This research focused on apple residues, coffee silver skin, potato peels, and brewer's spent grains, which were treated with two types of DESs—choline chloride/glycerol and choline chloride/ethylene glycol with different ratios—to produce fermentable sugars. After pretreatment, the biomass underwent enzymatic digestion with commercial enzymes to convert it into fermentable sugars. The DES pretreatment was carried out under a broad range of operational conditions, providing valuable insights for further research on using DESs in lignocellulosic biomass processing.[38]

Saini et al. underscored the importance of DESs as sustainable and environmentally friendly solvents for extracting bioactive compounds such as polyphenols, carbohydrates, alkaloids, etc. from agro-industrial by-products (Figure 1). Their study examined a variety of DESs, particularly those formulated with choline chloride, for their effectiveness in isolating these valuable compounds. They also highlighted the critical role of the solid-to-solvent ratio in maintaining a homogeneous reaction system during DES-based extraction processes.[39]

Ristvojevic et al. offered an in-depth analysis of emerging trends in the use of Natural Deep Eutectic Solvents (NaDESs) for extracting bioactive compounds from food, agricultural by-products, and related residues.[11] Similarly, Molnar et al. reviewed the applications of DESs as environmentally friendly solvents for isolating organic compounds such as polyphenols, carbohydrates, proteins, and alkaloids from by-products of the food industry and agro-industrial waste. Their research focused on methods for extracting these compounds from fruit and vegetable by-products, oilseed residues, and other agricultural and food processing waste, providing a detailed overview of how DESs can be used to convert such waste into valuable products.[1]

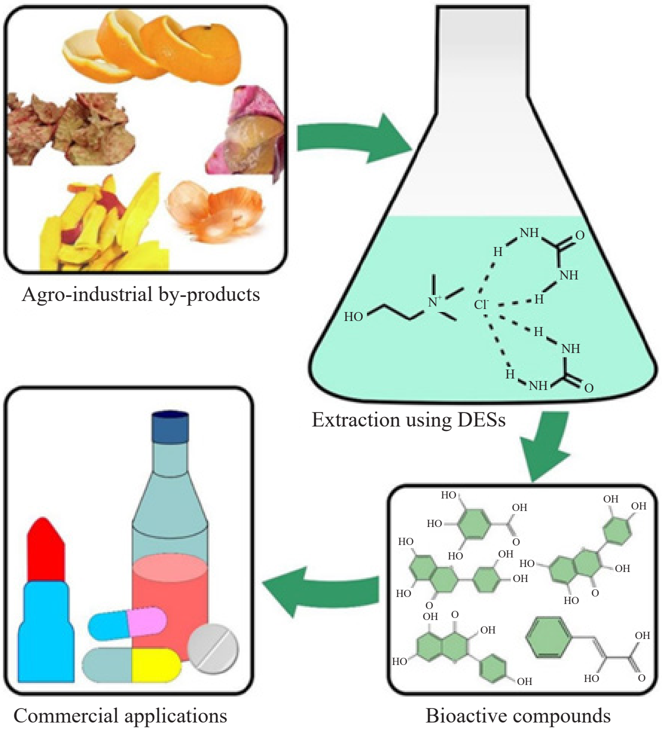

Figure 1. Deep eutectic solvents in the extraction of value-added compounds from agro-industrial by-products and their applications[39]

The effectiveness of DES-based extraction methods depends on various factors, including the solvent's structure, polarity, and physical-chemical characteristics, as well as operational conditions like extraction time, temperature, solid-to-solvent ratio, and pH. These parameters are key to optimizing the extraction process.[40] While the exact mechanisms behind DES-based extractions are not fully understood, these solvents offer significant promise due to their tunable properties and benefits compared to traditional solvents. Nevertheless, further research is necessary to address challenges such as solute recovery, toxicity, cost-effectiveness, stability, solvent recycling, and environmental impacts when scaling up DES-based methods for extracting bioactive substances.[39]

Recent studies highlight the effectiveness of DESs in extracting valuable bioactive compounds, such as polyphenols, phenolic acids, antioxidants, and polysaccharides, from organic residues. DESs offer an environmentally friendly alternative to conventional organic solvents, as they are biodegradable, non-toxic, and reusable, reducing the environmental footprint of industrial processes.[9,41] These studies also indicate that DESs can achieve higher extraction yields compared to traditional solvents while preserving the stability and bioactivity of the extracted compounds. The potential application of these extracted compounds in industries such as pharmaceuticals, nutrition, cosmetics, and biofuel production establishes DESs as a crucial tool for sustainable waste valorization and the development of a circular economy.

4. Recycling valuable metals from e-waste: A sustainable solution

DESs also play a crucial role in reducing waste by enabling the recovery of valuable metals, particularly from electronic waste (e-waste). E-waste contains precious metals such as gold, silver, and palladium, which are traditionally extracted using toxic methods like cyanide leaching. While effective, these conventional techniques result in significant environmental pollution and pose safety risks.[42]

DESs formed from specific combinations of hydrogen bond donors and acceptors, can strongly interact with metal ions and break metal-oxygen bonds. This capability allows DESs to dissolve metal oxides that are otherwise challenging to process with water-based solutions. By modifying the acidity or alkalinity, the system's dissolution capacity can be further optimized. Moreover, DESs possess intrinsic reducing properties, often eliminating the need for additional reducing agents during the leaching process. The addition of oxidants can catalytically dissolve metals in DESs, enhancing their recovery. Compared to traditional pyro- and hydrometallurgical methods, DES-based recovery processes consume less energy and produce significantly less wastewater.[43]

As mixtures of multiple components, DESs offer customizable solvent properties tailored to specific applications. For example, high viscosity, which can impede mass transfer and separation, can be mitigated by adjusting the composition and ratios of the DES components. This flexibility is particularly useful in leaching and precipitation processes. Hydrophobic DESs (HDESs), designed by modifying their composition, effectively extract and separate metal ions from aqueous solutions.[44]

Additionally, DESs overcome some of the limitations associated with conventional aqueous solutions and high-temperature molten salts. When used as electrolytes in electrochemical processes for metal and alloy recovery, DESs enable efficient extraction with minimal water involvement or interference. This unique combination of properties makes DESs an excellent choice for sustainable metal recovery solutions. These capabilities are further illustrated in Figure 2.

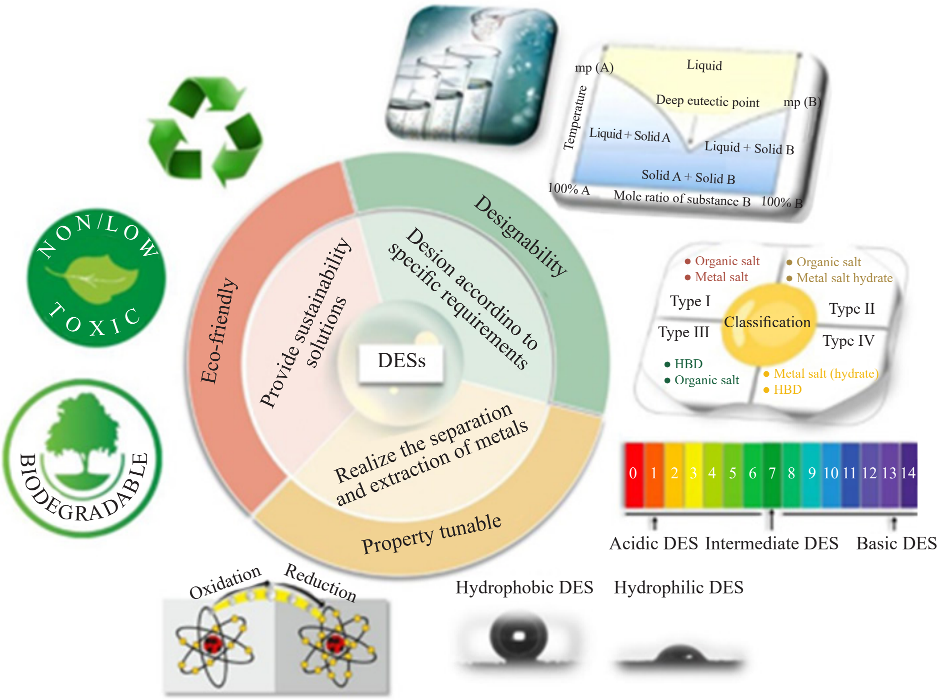

Figure 2. Properties of Deep Eutectic solvents (DESs) for metal recovery[43]

Guo et al. provided a comprehensive review of advancements in metal extraction from electronic waste using DESs. Their study examined various types of e-waste, including spent battery components, printed circuit boards, lamp phosphor residues, and discarded permanent magnets (Figure 3). The review highlighted several recovery techniques, such as redox and coordination leaching, solvent extraction, precipitation, and electrodeposition. It emphasized the distinct characteristics of DESs, their environmentally sustainable design, and the technological benefits they offer for efficient and eco-friendly metal recovery.[43]

The unique solvent characteristics of DESs make them highly effective for e-waste recovery processes that are difficult or impossible with conventional solvents. DESs are abundant in ligands, creating an optimal environment for metal ion coordination, which improves the leaching process and facilitates the selective separation of elements from electronic waste. Additionally, electrodeposition with DESs has emerged as a highly efficient and precise method for metal recovery from e-waste.[22]

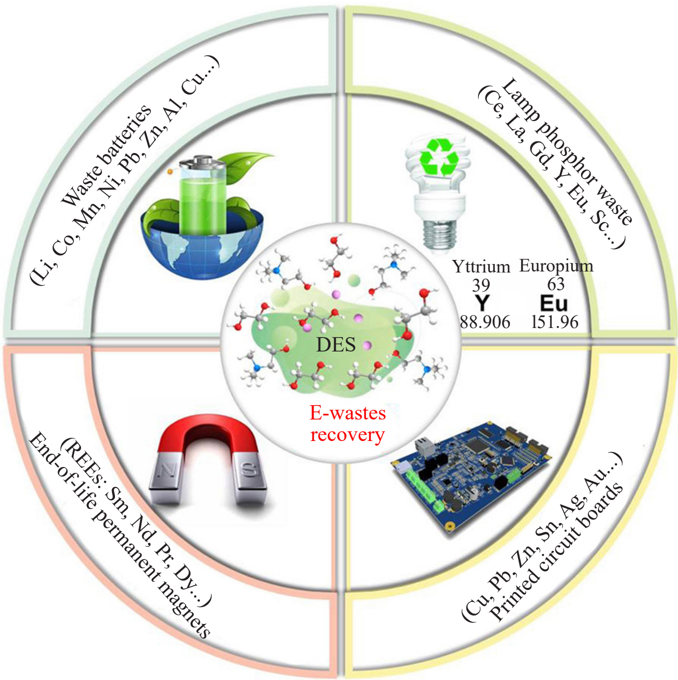

Figure 3. Recovery of metals from e-waste using DESs[43]

Despite significant advancements, several challenges remain before DES-based processes can be scaled up for pilot-level operations. The selective extraction of metals from waste is made possible by the varying solubility of metal species in DESs. However, their high viscosity often necessitates elevated temperatures and extended leaching durations compared to inorganic acids. To make these processes more efficient for industrial-scale applications, it is crucial to develop new low-viscosity DES formulations that can shorten extraction times.[45]

While DESs enable the electrochemical recovery of metals, their slow reaction rates limit practical applications, highlighting the need for research to improve reaction kinetics in these solvents.[46] Additionally, the interaction of water with DESs in the separation and extraction of metals requires further investigation. Although DESs are recognized as environmentally friendly solvents, some formulations include toxic components that are not easily biodegradable. Addressing this issue could involve recycling spent DESs and developing new formulations using non-toxic, eco-friendly components to further minimize environmental impact.[43]

5. Catalyst cleaning and regeneration: Reducing waste in industrial processes

Catalysts play a crucial role in numerous industrial chemical reactions, particularly in the petroleum and chemical sectors. Over time, catalysts can become contaminated or lose their efficiency, resulting in significant waste and requiring frequent replacement. Conventional methods for cleaning spent catalysts often rely on harsh chemicals, which create additional waste and environmental concerns.

DESs provide a sustainable alternative for cleaning and regenerating catalysts. Research has demonstrated that DESs can effectively remove contaminants and fouling agents from catalysts without damaging their active components.[47] This process reduces the amount of waste generated from spent catalysts and extends their operational lifespan, delivering both economic and environmental benefits.

Kamisono et al. introduced an environmentally friendly method for recycling Spent Automotive Catalysts (SACs), which contain small quantities of Platinum Group Metals (PGMs) along with Aluminum (Al) and Magnesium (Mg). Traditional recycling approaches often involve hydrometallurgical processes that use concentrated inorganic acids to leach SACs, dissolving unwanted metals and producing large volumes of wastewater. Instead, the researchers developed a non-aqueous direct leaching method using Hydrophobic Deep Eutectic Solvents (HDESs).[48]

The HDES, formulated from trihexyl(tetradecyl)phosphonium chloride and decanoic acid, served as an efficient and selective medium for extracting PGMs. By incorporating an organic acid and an oxidant, the HDES enabled effective leaching of PGMs while minimizing the dissolution of unwanted metals like Al and Mg. Under optimized conditions (24 hours, 80 °C, with stirring at 400 rpm), the leaching efficiencies were 90% for Pt, 100% for Pd, and 89% for Rh, while less than 5% of other metals were leached. The PGMs were subsequently recovered from the HDES through straightforward processes, such as contact with aqueous solutions. Recovery rates into the aqueous phase were approximately 80% for Rh and Pt and 90% for Pd.[48]

Additionally, the HDES demonstrated the ability to be regenerated and reused for further SAC leaching without significant loss in efficiency, achieving results comparable to fresh DES. This innovative system offers valuable insights into developing sustainable recycling techniques for PGMs from SACs, reducing waste and improving resource recovery efficiency.

Shao et al. introduced an innovative, environmentally friendly approach utilizing a closed-loop, recyclable DES-microfluidic injection system. This system, based on a choline chloride-oxalic acid DES, was used to synthesize cobalt-zinc oxide nanoblocks with a high concentration of Oxygen Vacancies (OV).[49] Ying et al. developed a DES regeneration strategy using a choline chloride/oxalic acid (1 : 1) system to produce porous Fe_3O_4 nanosheets, starting with commercially available Fe_3O_4 powder as the raw material.[50]

Similarly, Ni et al. proposed a cleaner method for the recovery of waste SmCo magnets using DESs. They developed and characterized Hydrophobic Deep Eutectic Solvents (HDESs) composed of dodecanol and tri-n-octylphosphine oxide. These HDESs were employed for the first time in extracting rare earth elements and successfully recovering Fe(III), Co(II), Cu(II), and Sm(III) from SmCo magnet leachate.[51]

Research into the use of DESs for catalyst cleaning and regeneration highlights their potential to revolutionize industrial processes. These solvents provide numerous advantages, including reduced waste production, selective metal leaching, and efficient catalyst regeneration. The studies reviewed underscore the effectiveness of DESs in recovering valuable metals and restoring catalyst performance. As further advancements are made in DES development and optimization, they are expected to become vital tools in fostering sustainable and environmentally conscious practices across various industries.

6. Conclusions

The multifaceted role of DESs in waste reduction addresses several critical challenges inherent in traditional industrial processes. By replacing toxic solvents, enabling the recovery of valuable metals from waste, and supporting catalyst regeneration, DESs provide a more sustainable and eco-friendlier alternative to conventional practices. Studies have shown that DESs not only help to decrease the generation of hazardous waste but also encourage the recycling of valuable materials, fostering a circular economy. As industries increasingly adopt DES technologies, their ability to reduce waste and promote sustainable practices is expected to grow, delivering significant environmental and economic advantages.

The future of industrial waste management and resource recovery appears bright with the broader implementation of DESs, which offer a viable route to greener and more sustainable production methods. These innovative solvents play a pivotal role in enhancing sustainability within industrial operations. Their capacity to minimize waste, transform by-products into valuable resources, and improve environmental outcomes positions them as a vital element in the shift toward circular economies. Continued research and development in DES applications are likely to unlock new opportunities for waste reduction, resource recovery, and environmentally friendly industrial processes, providing cleaner, more efficient, and cost-effective solutions for the future.

Conflict of interest

The author declares no competing financial interest.

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