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Deep Eutectic Solvents: A Review Of Molecular Design, Interfaces, And Sustainable Nanomaterials

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Abstract

The increasing demand for advanced functional materials for environmental monitoring, remediation, energy conversion, catalysis and healthcare has intensified the search for synthetic methodologies that simultaneously deliver structural precision, high functional performance and reduced environmental burden. Conventional materials-processing routes frequently rely on volatile organic solvents, corrosive reagents, high temperatures and energy-intensive operations, thereby introducing significant environmental, occupational and economic concerns. In this context, deep eutectic solvents (DESs) have emerged as a versatile class of designer liquids capable of integrating solvent, reagent, templating, stabilizing and structure-directing functions within a single reaction environment. Since the pioneering report of choline chloride/urea eutectics by Abbott et al., DESs have evolved from alternative ionic-liquid-like media into multifunctional platforms for materials synthesis. Their tunable composition, low volatility, high solvating ability, broad electrochemical stability, strong hydrogen-bonding interactions and compatibility with diverse inorganic precursors provide opportunities for manipulating nucleation, crystal growth, particle aggregation and surface chemistry.

This review critically examines the role of DESs in the sustainable synthesis and engineering of functional nanomaterials, with particular emphasis on ternary metal oxides, spinel ferrites, scheelite-type molybdates, layered double hydroxides (LDHs) and carbon-based heterostructures. The fundamental chemistry of DES formation, classification, physicochemical properties and structure–property relationships is first discussed. Particular attention is given to the multifunctionality of DESs as reaction media, complexing agents, reducing environments, templates and growth-directing species. The influence of hydrogen-bond donors, component ratios, water content and precursor coordination on material nucleation and morphology is subsequently examined. The electrochemical relevance of DES-derived materials is discussed in relation to surface area, defect chemistry, mixed valence, charge-transfer kinetics, adsorption, catalytic activity and interfacial electron transport. Recent advances in the detection of nitroaromatic compounds, antibiotics, pharmaceutical contaminants, heavy-metal ions and other environmentally relevant analytes are critically evaluated. Finally, the review identifies unresolved challenges involving viscosity, mass transfer, reproducibility, residual DES, toxicity, life-cycle impacts, mechanistic understanding, solvent recovery and scale-up. The emerging direction is therefore not merely the substitution of conventional solvents with DESs, but the rational integration of solvent molecular design with materials engineering and sustainability assessment.

Keywords: ***Deep eutectic solvents,green chemistry,functional nanomaterials,spinel ferrites,scheelite oxides,layered double hydroxides,electrochemical sensing, environmental remediation, sustainable synthesis, structure–property relationships.***

1. Introduction

The rapid development of modern technologies has created an increasing demand for functional materials with precisely engineered structural, chemical, optical, magnetic, catalytic, and electrochemical properties. Such materials underpin diverse technological sectors, including energy conversion and storage, catalysis, environmental remediation, healthcare, sensing, electronics, and advanced manufacturing [1–5]. The performance of a functional material, however, is determined not only by its intrinsic composition but also by the synthetic route through which its structure, morphology, crystallinity, surface chemistry, and defect characteristics are established. Consequently,

contemporary materials research is progressively shifting from a sole emphasis on material performance toward an integrated consideration of material function, synthetic efficiency, resource utilization, energy demand, and environmental compatibility [6–8].

Conventional materials-synthesis protocols frequently depend on volatile organic solvents, corrosive reagents, high reaction temperatures, prolonged processing times, or multistep purification procedures. Although such methodologies can provide excellent control over reaction kinetics and product formation, their environmental burden may arise from solvent consumption, atmospheric emissions, hazardous waste generation, and energy-intensive processing [9–11]. The solvent component is particularly important because solvents can constitute a substantial fraction of the mass used in chemical processes and may simultaneously function as reaction media, heat-transfer media, extraction agents, dispersants, or crystallization environments [12]. The redesign of solvent systems therefore represents one of the most direct approaches to improving the sustainability of chemical and materials manufacturing.

The emergence of green chemistry has fundamentally changed the philosophy of chemical synthesis by shifting the focus from pollution treatment toward pollution prevention at the molecular and process levels [13]. The twelve principles articulated by Anastas and Warner established a framework for designing chemical processes that minimize hazardous substances, waste generation, energy consumption, and environmental impact while retaining the desired functionality of the resulting products [14]. Within this framework, the selection and design of safer solvents and auxiliaries have received particular attention because solvent choice influences reaction efficiency, safety, toxicity, energy requirements, separation, recycling, and ultimately the overall environmental profile of a process [15,16].

Water, supercritical fluids, bio-based solvents, ionic liquids (ILs), and other alternative solvent systems have consequently been investigated as replacements for conventional volatile organic solvents [17–21]. Among these alternatives, deep eutectic solvents (DESs) have emerged as particularly versatile media because their composition can be readily tailored by combining suitable components, frequently without the need for complex synthetic procedures or extensive purification [22–25]. Beyond their use as reaction media, DESs can participate directly in materials formation by functioning as solvents, coordinating media, reducing environments, templates, dispersants, stabilizing agents, and sources of specific chemical functionalities [26–29].

The significance of DESs in materials chemistry has expanded substantially since their introduction. Abbott et al. demonstrated in 2003 that mixtures of choline chloride and urea exhibit an unexpectedly large depression of the melting point, producing a liquid system at temperatures far below those of the individual constituents [30]. This discovery established a simple and conceptually powerful route toward low-melting eutectic media and subsequently stimulated extensive research into their physicochemical properties and applications. Modern investigations have extended DES chemistry from extraction and separation processes to electrochemistry, catalysis, electrodeposition, biomass processing, nanotechnology, and the synthesis of advanced functional materials [22,20–33].

The attraction of DESs for materials synthesis arises from their unusual combination of low volatility, high solvating capacity, ionic character, tunable polarity, relatively straightforward preparation, and strong interactions with inorganic and organic precursors [23,25,33]. More importantly, their composition can influence the nucleation and growth of inorganic phases, allowing the solvent itself to become an active component of materials engineering rather than merely an inert reaction medium. Recent reviews have highlighted the ability of DESs to act simultaneously as solvents, templates, capping agents, reducing environments, metal-ion coordinating media, and functionalization agents during nanomaterial synthesis [26,28,36].

Nevertheless, the increasing use of DESs should not be interpreted as evidence that every DES is intrinsically ―green.‖ Their environmental performance depends strongly on the chemical identity and concentration of their constituents, preparation method, toxicity, biodegradability, water content, recyclability, energy demand, and downstream processing requirements [24,37–39]. Thus, the contemporary understanding of DES-based sustainability has evolved from a simplistic comparison with volatile organic solvents toward a more

comprehensive assessment encompassing life-cycle considerations and the complete materials-synthesis process.

In this context, DESs provide an important bridge between green solvent engineering and functional-material design. Particularly attractive are DES-mediated routes to ternary metal oxides, spinel ferrites, scheelite-type oxides, layered double hydroxides (LDHs), carbon-based heterostructures, and related nanostructured materials. These materials possess tunable electronic structures, high surface-to-volume ratios, abundant surface-active sites, redox properties, and favorable charge-transfer characteristics that make them valuable for environmental catalysis and electrochemical sensing [40–45]. The combination of DES-enabled synthesis with such functional materials therefore provides an opportunity to address two interconnected challenges: reducing the environmental footprint of material fabrication while simultaneously producing materials capable of detecting or degrading environmental contaminants.

1.1 Green chemistry and sustainable materials synthesis

The development of sustainable chemical technologies requires a transition from end-of-pipe pollution control toward the prevention of environmental burdens at their source. Green chemistry provides such a framework by incorporating environmental considerations into the molecular design of chemicals and processes [13,14]. Rather than treating waste, toxicity, and energy consumption as secondary consequences of chemical production, green chemistry considers them fundamental design parameters.

Among the twelve principles of green chemistry, the prevention of waste, reduction of hazardous synthesis, design for energy efficiency, use of renewable feedstocks, catalysis, and the use of safer solvents are particularly relevant to materials synthesis [14]. Conventional nanomaterial preparation may involve organic solvents, surfactants, reducing agents, stabilizers, mineralizing agents, and other additives that can generate secondary waste streams. Moreover, thermal treatment, calcination, hydrothermal processing, or prolonged reaction periods can significantly increase the energy requirement of synthesis [41,46].

A sustainable materials-synthesis strategy therefore requires more than simply obtaining the desired phase. Reaction yield, atom utilization, solvent toxicity, energy consumption, process temperature, reaction time, waste generation, recyclability, and post-synthetic purification must be considered collectively. In this regard, solvent engineering has become an important

component of sustainable materials chemistry because the solvent frequently determines the physical environment in which precursor dissolution, complexation, hydrolysis, nucleation, crystal growth, aggregation, and particle stabilization occur [15,16].

The replacement of conventional solvents with alternative media has consequently developed into a major research direction. Water remains the most desirable solvent from the standpoint of availability, cost, and inherent safety, but its limited ability to dissolve many nonpolar compounds and its influence on hydrolysis, solubility, and crystal growth restrict its universality [17]. Supercritical fluids offer distinctive transport and solvation properties but generally require high pressures and specialized equipment [18]. Ionic liquids provide extremely low vapor pressure and substantial structural tunability, yet their synthesis, purification, cost, toxicity, and environmental persistence can vary considerably depending on their constituent ions [19,20].

DESs have emerged within this landscape as a complementary class of tunable liquids. Their preparation commonly involves simply combining two or more components at an appropriate composition and temperature, often without a separate solvent-exchange or purification step [22,30]. This apparent simplicity, combined with their physicochemical tunability, has encouraged their investigation as reaction media for sustainable synthesis.

Importantly, the sustainability of a solvent cannot be assigned solely from a single property such as vapor pressure. A solvent with negligible volatility may still possess undesirable toxicity, poor biodegradability, or high embodied energy. Accordingly, recent DES research increasingly emphasizes a multidimensional definition of greenness, incorporating toxicity, biodegradation, renewable sourcing, recyclability, water consumption, energy demand, and life-cycle impacts [37–39]. This critical perspective is essential when evaluating DES-mediated materials synthesis.

1.2 Green solvent engineering

Solvent selection traditionally relies on parameters such as polarity, viscosity, dielectric constant, boiling point, and chemical compatibility. In green solvent engineering, these criteria are supplemented by environmental, occupational, and process-safety considerations [15,16]. An ideal solvent would combine high process performance with low toxicity, negligible environmental persistence, minimal volatility, efficient recyclability, low energy demand, and ready availability.

A number of alternative solvent classes have therefore been explored, including water, supercritical carbon dioxide, bio-derived solvents, ILs, polymeric solvents, and DESs [17–21,20]. However, each class presents its own balance of advantages and limitations. The challenge is consequently not to identify a universally superior solvent but to design a solvent environment appropriate to a specific chemical transformation or materials-synthesis pathway.

This concept is particularly relevant to nanomaterial synthesis. Particle nucleation and growth are governed by complex interactions among precursor concentration, supersaturation, temperature, reaction kinetics, interfacial energy, diffusion, surface adsorption, and ligand or solvent interactions [47,48]. A solvent can therefore influence not only whether a material forms, but also its crystallographic phase, particle size, morphology, aggregation state, porosity, surface composition, and defect population.

DESs are especially interesting in this regard because their molecular and ionic components can interact directly with precursor species. Carboxylic acids, polyols, amides, sugars, and other hydrogen-bond donors can modify metal-ion coordination and local chemical environments, while quaternary ammonium salts can contribute ionic character and strong hydrogen-bond interactions [22,25]. Changes in HBA/HBD identity or molar ratio can consequently alter viscosity, conductivity, polarity, acidity/basicity, water affinity, metal-ion speciation, and mass transport.

Recent work has further demonstrated that DESs can serve as multifunctional environments for nanomaterial formation. Siegel et al. emphasized their potential as ―all-in-one‖ media capable of simultaneously fulfilling several synthetic functions, including solvent, metal precursor environment, reducing medium, amphiphile, and capping or stabilizing agent [28]. Similarly, Sugiarto et al. highlighted the expanding role of DESs in the synthesis of metals, metal oxides, carbon materials, polymers, and other functional nanostructures [26].Thus, DES-based synthesis represents a transition from the conventional concept of a solvent as a passive reaction medium toward a more integrated concept in which the solvent contributes actively to the formation and functionalization of the final material.

1.3 Deep eutectic solvents: evolution, chemistry and classification

The term deep eutectic solvent was introduced by Abbott et al. in 2003 in their pioneering study of mixtures containing choline chloride and urea [30]. The authors reported that a mixture of choline chloride and urea in a 1:2 molar ratio produced a liquid with a melting point dramatically lower than those of its individual components. The phenomenon was attributed to strong interactions between the chloride ion and urea, which substantially destabilize the crystal lattices of the parent components [30].

This discovery provided an alternative route to low-melting liquid media that shared several characteristics with ILs while offering a remarkably simple preparation strategy. Subsequent investigations demonstrated that DESs could be generated from a broad range of hydrogen-bond acceptors (HBAs) and hydrogen-bond donors (HBDs), expanding the chemical space available for solvent design [22,23,20].A conventional DES generally contains at least two components whose interaction produces a melting point significantly below that predicted for an ideal mixture. In many widely investigated systems, the HBA is a quaternary ammonium salt such as choline chloride (ChCl), whereas the HBD may be urea, glycerol, ethylene glycol, malonic acid, oxalic acid, lactic acid, sugars, or other hydrogen-bonding molecules [22,25,20]. The frequently investigated ChCl-based systems are attractive because choline-derived components are comparatively inexpensive and readily available, while their combination with diverse HBDs allows substantial control over solvent properties.

The deep eutectic phenomenon is fundamentally associated with non-ideal interactions between the components. In a ChCl–urea system, for example, hydrogen bonding between chloride and the N–H groups of urea alters the distribution and mobility of charge and lowers the effective lattice stability of the mixture [30,20]. More generally, hydrogen bonding, electrostatic interactions, Lewis acid–base interactions, van der Waals forces, and metal coordination can contribute to the non-ideal behavior of DESs [22,25,32].The magnitude of melting-point depression depends on both the chemical identities of the components and their molar ratio. Consequently, the HBA:HBD ratio is not merely a formulation parameter but an important materials-design variable. Changing the ratio can modify viscosity, conductivity, polarity, water activity, hydrogen-bond density, metal-ion coordination, and transport properties, thereby altering the environment in which nucleation and crystal growth occur [25,32,49].

DESs were initially classified into several categories according to the nature of their constituents. The classical classification includes Type I systems composed of quaternary ammonium salts and anhydrous metal chlorides, Type II systems containing hydrated metal chlorides, Type III systems generally involving quaternary ammonium salts and molecular HBDs, and Type IV systems consisting principally of metal chloride species and HBDs [22,20]. Type III DESs have become particularly important for materials synthesis because they can be prepared from metal-free HBAs and a wide range of molecular donorMore recently, additional classifications have been proposed to accommodate systems that do not fit neatly into the original framework. In particular, Type V DESs have been proposed for hydrophobic eutectic systems in which non-ionic molecular components participate predominantly through hydrogen-bond interactions [50]. This expansion reflects the increasing diversity of DES chemistry beyond the original ChCl-based systems.

Another important development was the introduction of natural deep eutectic solvents (NADESs). Choi et al. proposed that naturally occurring primary metabolites such as sugars, amino acids, organic acids, and choline derivatives can form eutectic mixtures capable of dissolving and stabilizing natural compounds [51]. NADESs subsequently attracted considerable attention because they potentially combine the functional advantages of DESs with renewable biological feedstocks. However, as with conventional DESs, their actual environmental performance must be assessed on the basis of composition and complete life-cycle behavior rather than assumed solely from the —natural‖ origin of their components.

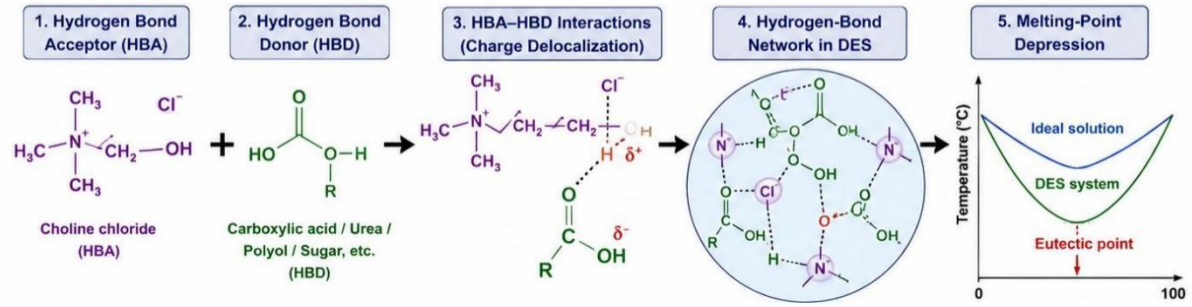


Figure 1. Schematic representation of DES formation showing HBA–HBD interactions, charge delocalization, hydrogen-bond-network formation, melting-point depression

The physicochemical diversity of DESs is one of their greatest advantages. Their viscosity, conductivity, density, polarity, thermal stability, acidity/basicity, and water affinity can be manipulated through component selection and composition [22,25,32]. Such tunability is particularly valuable for materials synthesis because the solvent can be designed around the requirements of a specific precursor or target material.

1.4 Physicochemical characteristics governing DES-enabled materials synthesis

The properties of DESs are closely interconnected, and their collective influence determines their performance during materials synthesis. Viscosity, for example, can strongly influence precursor diffusion and mass transport. Although high viscosity may suppress molecular mobility, it can also inhibit uncontrolled aggregation and provide a stabilizing

environment for growing nanoparticles [26,49]. Ionic conductivity influences charge transport and becomes especially important in electrochemical synthesis and electrodeposition [28]. Polarity and hydrogen-bonding ability determine the solubility and coordination environment of precursor species, while acidity or basicity can influence hydrolysis, condensation, dissolution, and redox reactions [25,32].

Water represents another particularly important variable. Small quantities of water can substantially reduce DES viscosity and increase ionic mobility, thereby facilitating mass transport. However, increasing water content may progressively disrupt the hydrogen-bond network responsible for the eutectic behavior and eventually transform the system from a DES-rich medium into an aqueous solution containing DES components [54]. Therefore, water content must be considered an experimental parameter rather than an incidental impurity.These properties provide DESs with several potential functions during the synthesis of functional materials. Depending on the system, a DES may simultaneously act as:

1. a reaction solvent that dissolves inorganic or organic precursors;
2. a complexing medium that modifies metal-ion speciation;
3. a template or structure-directing medium that influences nucleation and crystal growth;
4. a capping or dispersing environment that suppresses uncontrolled aggregation;
5. a reducing medium when chemically active components are present;
6. a precursor reservoir supplying particular elements or functional groups;
7. an electrolytic medium for electrochemical deposition or synthesis;
8. and a surface-functionalization environment that influences the properties of the resulting nanomaterial [26,28,36,55].

This multifunctionality distinguishes DES-mediated synthesis from simple solvent substitution. Instead of replacing one liquid with another while leaving the underlying synthesis unchanged, DES chemistry allows the reaction environment itself to become an engineering parameter.

1.5 DESs as multifunctional platforms for functional-material synthesis

The use of DESs in materials synthesis has expanded rapidly from relatively simple electrodeposition and metal processing to the preparation of complex nanostructured oxides, sulfides, phosphides, carbon materials, polymers, LDHs, and hybrid architectures [26,28,36]. Tomé et al. emphasized that DESs can provide distinctive opportunities for the preparation of polymers, electrodeposited materials, nanomaterials, and sensing platforms [34].

The importance of this approach lies in the possibility of controlling composition and structure simultaneously. In conventional synthesis, precursor selection, solvent selection, surfactant selection, and morphology-control additives are frequently treated as separate variables. In a DES, several of these functions may be integrated into a single chemically tunable medium. The resulting reduction in the number of auxiliary reagents can potentially simplify processing and reduce waste.

However, the extent to which such processes are genuinely sustainable depends on the complete synthesis. A DES-mediated process that requires prolonged heating, large excesses of precursor, extensive washing, or energy-intensive calcination cannot automatically be regarded as environmentally superior merely because the reaction medium has low vapor pressure. Therefore, DES-based materials research should increasingly combine structural characterization with quantitative sustainability metrics, including solvent recovery, material yield, energy consumption, waste generation, component toxicity, and life-cycle assessment [37–39].

This distinction is particularly important for metal-oxide nanomaterials. Many DES-mediated routes ultimately require calcination or thermal treatment to achieve the desired crystallinity. While the DES may facilitate precursor organization and reduce the complexity of the wet-chemical stage, the overall environmental advantage must be evaluated across both the solution-phase synthesis and subsequent thermal processing.

Nevertheless, the potential is substantial. Recent literature demonstrates that DESs can facilitate the formation of nanomaterials with controlled morphology and enhanced functional properties, while providing opportunities for simplified synthesis and multifunctional reaction environments [26,28,36]. These characteristics make DESs particularly attractive for designing electroactive functional materials, where subtle differences in crystallinity, surface area, defect chemistry, and exposed active sites can determine analytical and catalytic performance.

2. DES-Mediated Synthesis of Functional Materials: Mechanistic Principles and Design Strategies

The growing application of deep eutectic solvents in materials chemistry is not simply a consequence of their low volatility or facile preparation. Their greater significance lies in the ability of DESs to modify the chemical and physical environment in which nucleation, crystal growth, aggregation, phase transformation, and surface functionalization occur. In conventional synthesis, solvent, ligand, template, reducing agent, and stabilizer are frequently introduced as independent components. DESs can integrate several of these functions within a single reaction medium, thereby providing an opportunity to manipulate material formation through solvent–precursor interactions and controlled supramolecular organization [22,26,28,36].

The synthesis of functional nanomaterials generally involves a sequence of interconnected events, including precursor dissolution, complex formation, hydrolysis or condensation, nucleation, crystal growth, aggregation, and, where applicable, phase transformation during subsequent thermal treatment. Each of these stages can be influenced by the physicochemical characteristics of the reaction medium. DESs are particularly suitable for such control because their constituent ions and molecules interact strongly with metal ions and developing inorganic surfaces [25,32,36].

2.1 Influence of DES composition on precursor chemistry and nucleation

The chemical identity of the HBA and HBD is one of the principal determinants of DES behavior. Choline chloride, for example, provides a chloride-rich ionic environment, while HBDs such as urea, ethylene glycol, malonic acid, oxalic acid, and sugars introduce different combinations of hydrogen-bonding, coordinating, acidic, or reducing functionalities. Consequently, changing the HBD can modify metal-ion complexation and precursor reactivity even when the HBA remains unchanged [22,25].

Metal ions introduced into a DES do not necessarily retain the same coordination environment that they possess in water or conventional organic solvents. Chloride ions, oxygen-containing functional groups, hydroxyl groups, carboxylates, and amide functionalities may interact with metal centers, changing their effective activity and mobility.

Such interactions can influence the rate at which precursor species undergo hydrolysis, condensation, precipitation, or conversion into crystalline phases [28,36].This phenomenon becomes particularly important in multicomponent oxides. The simultaneous incorporation of different metal cations requires appropriate control over their relative reaction rates. If one precursor hydrolyzes or precipitates substantially faster than another, compositional segregation or secondary-phase formation may occur. A strongly coordinating DES can moderate such differences by temporarily stabilizing precursor species, thereby providing a more homogeneous environment for subsequent nucleation [36,55].

The HBA:HBD molar ratio provides an additional degree of control. Changes in composition modify hydrogen-bond density, viscosity, polarity, conductivity, and metal-ion coordination. Consequently, the same precursor system can potentially generate materials with different particle sizes or morphologies when synthesized in DESs of different composition [25,32].

For example, acidic HBDs such as malonic acid and oxalic acid can provide environments substantially different from those generated by neutral HBDs such as urea or ethylene glycol. The resulting differences in proton activity, complexation, and precursor dissolution can affect oxide formation and crystal growth. Similarly, polyols may interact with metal centers through multiple oxygen-containing sites, potentially modifying nucleation and particle aggregation.

2.2 DESs as structure-directing and templating media

The ability of DESs to influence morphology represents one of their most valuable characteristics in nanomaterial synthesis. Nanomaterial properties are strongly dependent on particle size, shape, exposed facets, porosity, aggregation state, and hierarchical organization. Even when two materials possess identical chemical compositions and crystal structures, differences in morphology can result in markedly different catalytic and electrochemical behavior [47,48].

DESs can affect morphology through several mechanisms. Strong interactions between DES components and specific crystal surfaces can selectively retard the growth of particular facets. At the same time, the relatively high viscosity of many DESs can restrict diffusion and reduce uncontrolled particle collision and aggregation [26,36]. These effects can favor the formation of smaller particles, nanosheets, nanorods, porous structures, or interconnected hierarchical architectures depending on the chemical system.The concept of DES-mediated structural direction is especially relevant for materials employed in heterogeneous catalysis and electrochemical sensing. High surface area and accessible active sites increase the number of locations available for analyte adsorption or catalytic transformation. Shorter diffusion pathways can additionally facilitate charge and mass transport. Therefore, a DES-induced change in morphology can translate directly into enhanced functional performance [40,43].

The structure-directing effect should nevertheless be distinguished from conventional templating. In some systems, the DES does not create a permanent physical template. Instead, its constituent molecules modify precursor speciation, interfacial energies, nucleation rates, or surface adsorption, indirectly determining the final morphology [28,36]. This distinction is important because it emphasizes that DES-mediated morphology control can originate from dynamic chemical interactions rather than from a rigid template.

2.3 Viscosity, mass transport and particle growth

Viscosity is among the most distinctive physicochemical characteristics of DESs and has a pronounced influence on materials synthesis. Many DESs exhibit substantially higher viscosities than water or conventional organic solvents [22,32]. High viscosity can reduce diffusion coefficients and consequently slow the movement of precursor species through the reaction medium.At first glance, reduced diffusion might appear disadvantageous. However, controlled mass transport can be beneficial for nanomaterial synthesis because excessively rapid precursor movement can promote uncontrolled nucleation and aggregation. A viscous medium can moderate these processes and provide a more controlled growth environment [26,49].The effect is strongly dependent on temperature. Increasing temperature generally decreases DES viscosity and increases ionic mobility, thereby accelerating mass transport and chemical reactions [32]. Thus, temperature and DES composition must be considered together rather than independently.

Water addition provides another route for controlling viscosity. Small amounts of water can dramatically lower viscosity and enhance transport, potentially accelerating nucleation and crystal growth [54]. However, excessive water can disrupt the extensive hydrogen-bond network that characterizes the eutectic system. Therefore, controlled water incorporation can be viewed as a means of tuning the reaction environment, whereas uncontrolled water content can compromise reproducibility.

2.4 DES-mediated control of crystallinity, defects and surface chemistry

Beyond particle size and morphology, DESs can influence the crystallinity and defect chemistry of functional materials. The coordination environment established during precursor dissolution can affect the rate of phase

formation, while the chemical atmosphere during thermal conversion can influence oxidation state and surface composition [36,55].

Defects such as oxygen vacancies, cation vacancies, antisite defects, and lattice distortions are increasingly recognized as important determinants of catalytic and electrochemical activity. Such defects can modify adsorption energies, electronic conductivity, charge-transfer kinetics, and the density of electrochemically active sites [56–58]. Consequently, DES-mediated control over precursor chemistry may provide an indirect route for tuning defect populations.This is particularly important for transition-metal oxides. Variations in the DES environment may therefore influence not only crystal growth but also the chemical state of surface species. Such effects can be particularly beneficial for electrochemical sensing, where electron-transfer kinetics at the electrode–electrolyte interface are directly related to surface composition and electronic structure.

2.5 DES-assisted energy-efficient synthesis

One of the frequently proposed advantages of DES-mediated synthesis is the possibility of reducing reaction severity. The high solvating capacity and strong interaction with precursor species can facilitate material formation under conditions different from those required in conventional synthesis [26,36]. In hydrothermal and solvothermal approaches, DESs may modify precursor reactivity and crystal growth, potentially allowing the formation of particular morphologies at comparatively moderate temperatures.Many oxide nanomaterials ultimately require calcination at elevated temperatures to obtain the required crystallinity. Thus, replacing a conventional solvent with a DES does not automatically guarantee a lower total energy footprint. A meaningful comparison should account for the entire process, including DES preparation, reaction heating, washing, drying, calcination, and solvent recovery [37–39].This consideration is particularly important when evaluating the scalability of DES-mediated materials synthesis. Laboratory-scale experiments often emphasize material properties while giving limited attention to solvent recovery, process intensification, mass transfer, and energy consumption. Future development therefore requires coupling DES chemistry with process engineering and life-cycle assessment.

3. Functional Ternary Metal Oxides for Environmental Applications

Ternary metal oxides constitute one of the most chemically versatile families of functional materials. Their general compositions can be represented by formulas such as AB_2O_4, ABO_4, A_2BO_4, and related stoichiometries, encompassing diverse crystal structures and combinations of metal cations. Their properties can be tuned through elemental substitution, cation ordering, oxygen non-stoichiometry, particle-size control, and morphology engineering [40,59].

The combination of variable oxidation states, semiconducting behavior, magnetic properties, catalytic activity, and electrochemical response has made ternary oxides attractive for applications including photocatalysis, electrocatalysis, batteries, supercapacitors, water treatment, sensing, and environmental monitoring [40–45].For the DES-mediated materials considered here, two structural families are particularly relevant: spinel-type AB_2O_4 oxides and scheelite-type ABO_4 oxides. Although structurally distinct, both offer extensive opportunities for compositional modification and morphology-controlled synthesis.

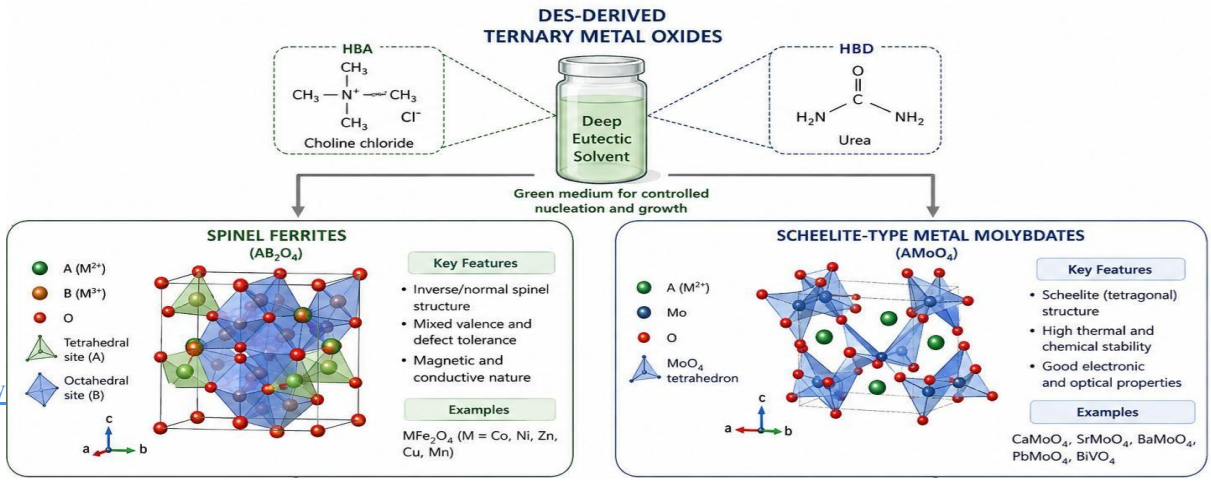


Figure 2.Structural relationship and functional diversity of DES-derived ternary metal oxides, highlighting spinel ferrites and scheelite-type metal molybdates and their roles in photocatalysis, electrocatalysis, and environmental sensing.

3.1 Spinel-type AB_2O_4 oxides

Spinel oxides possess the general formula AB_2O_4, where A and B commonly represent divalent and trivalent metal cations, respectively. The ideal spinel structure consists of a cubic close-packed oxygen framework containing tetrahedral and octahedral cation sites, with the conventional structure described by the $Fd\bar{3}m$ space group [33,60].

The distribution of cations between tetrahedral and octahedral sites gives rise to different spinel configurations. In an ideal normal spinel, divalent A^{2+} ions occupy tetrahedral sites while trivalent B^{3+} ions occupy octahedral sites:

$$(A^{2+})_{tet}[B^{3+}{}_2]_oC_tO_4$$

In an inverse spinel, the divalent cation occupies octahedral sites while half of the trivalent cations occupy tetrahedral positions:

$$(B^{3+})_{tet}[A^{2+}B^{3+}]_oC_tO_4$$

Real spinels frequently exhibit partial inversion rather than perfectly normal or inverse distributions. The inversion parameter depends on ionic radius, temperature, synthesis conditions, cation electronic configuration, and defect chemistry [60,61].This structural flexibility produces considerable variation in magnetic, optical, catalytic, and electrochemical behavior. Ferrites containing Mg, Zn, Mn, Co, Ni, and other transition-metal ions have therefore been extensively investigated for catalysis, magnetic separation, energy-related applications, and sensing [62–65].

An especially important advantage of ferrite nanoparticles in environmental applications is their potential magnetic separability. Following photocatalytic or adsorption-based treatment, magnetic particles can potentially be recovered from aqueous systems using an external magnetic field, reducing the difficulty associated with separating nanoscale catalysts from treated water [63,64].

3.2 DES-Mediated Synthesis, Photocatalysis and Electrochemical Applications of Spinel Ferrites

Deep eutectic solvents provide an attractive synthetic environment for controlling the formation of spinel ferrites because their components can interact strongly with metal precursors through hydrogen bonding, coordination and ionic interactions. Choline chloride-based DESs containing carboxylic acids, urea, polyols or sugars can consequently influence precursor solubility, hydrolysis, nucleation and crystal growth before thermal conversion into the final oxide [22,26,28]. The chemical nature and molar ratio of the HBA and HBD can therefore contribute to differences in crystallinity, particle size, morphology, aggregation behaviour and surface chemistry.

The use of ChCl–malonic acid, ChCl–oxalic acid, ChCl–urea, ChCl–ethylene glycol and ChCl–fructose systems represents a systematic investigation of chemically different DES environments rather than merely a replacement of one conventional solvent with another. Differences in acidity, hydrogen-bonding ability, viscosity, polarity and coordination behaviour among these systems can modify the chemical environment surrounding the metal precursors and consequently influence the formation of $MgFe_2O_4$ nanoparticles. Establishing such relationships is important for understanding how DES composition can be translated into specific structural and functional characteristics.

$MgFe_2O_4$ is particularly attractive for environmental applications because of its chemical stability, magnetic properties, redox activity and electrochemical behaviour [62,65]. When synthesized as nanoparticles, these characteristics can be complemented by increased surface accessibility and shortened diffusion pathways. Such features make $MgFe_2O_4$ a promising electrode-modifying material for the electrochemical determination of nitroaromatic pollutants and pharmaceutical residues.The photocatalytic activity of ferrites is similarly governed by their electronic structure, surface properties, particle dimensions and defect chemistry. Under appropriate irradiation,

semiconductor ferrites can generate photogenerated charge carriers that participate in oxidation and reduction reactions at the material–solution interface. Iron-containing ferrites can additionally contribute to Fenton and photo-Fenton-like pathways through Fe^{2+}/Fe^{3+} redox cycling and the generation of reactive oxygen species [66–68]. These highly reactive species can attack and decompose a broad range of organic contaminants.

Among $MgFe_2O_4$, $ZnFe_2O_4$ and $MnFe_2O_4$, differences in cation distribution and electronic structure can result in substantially different photoresponses. $MnFe_2O_4$, for example, can exhibit favourable visible-light activity because of its electronic structure and redox characteristics [63,64]. Such compositional effects demonstrate the importance of systematic cation substitution when designing ferrite catalysts for pollutant degradation.The same structure–property relationships are relevant to electrochemical sensing.

Electrochemical sensing is particularly attractive for environmental monitoring because it can provide rapid analysis with relatively simple instrumentation, low sample requirements and potential for portable or on-site operation [62–66]. DES-derived ferrites, therefore, provide an opportunity to connect sustainable synthesis directly with analytical functionality.The significance of this approach becomes even greater when considering simultaneous detection. Environmental samples frequently contain several contaminants rather than a single target species. The development of a multifunctional electrode capable of resolving the electrochemical responses of multiple analytes can reduce analysis time, sample consumption and instrumental complexity [62–66]. DES-derived $MgFe_2O_4$, AFe_2O_4 and related spinel systems therefore represent an important class of materials for integrating sustainable synthesis with multifunctional environmental sensing.

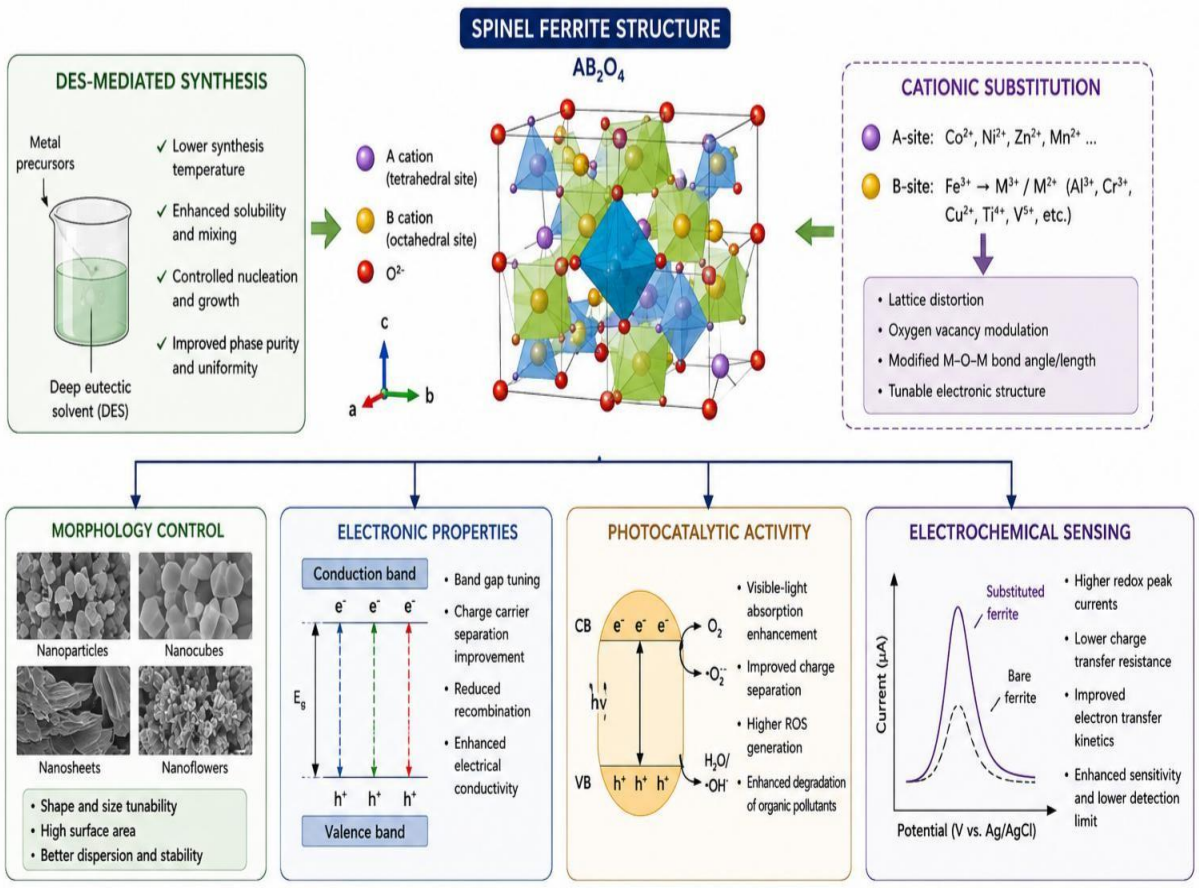


Figure 3. Structural characteristics of spinel ferrites and the influence of DES-mediated synthesis and cationic substitution on morphology, electronic properties, photocatalytic activity and electrochemical sensing performance.

Spinel ferrite	Composition	Tunable properties	Environmental application
$MgFe_2O_4$	Mg^{2+}/Fe^{3+}	Morphology, particle size, surface area	Electrochemical pollutant sensing
$ZnFe_2O_4$	Zn^{2+}/Fe^{3+}	Band gap, defects, morphology	Photocatalysis, pollutant sensing
$MnFe_2O_4$	Mn^{2+}/Fe^{3+}	Magnetic properties, particle size	Pollutant removal, sensing
$CuFe_2O_4$	Cu^{2+}/Fe^{3+}	Band structure, redox activity	Photocatalytic degradation
$CoFe_2O_4$	Co^{2+}/Fe^{3+}	Magnetism, morphology, surface activity	Environmental remediation
$FeCo_2O_4$	Fe^{2+}/Fe^{3+}–Co^{2+}/Co^{3+}	Conductivity, redox activity	Electrocatalysis, sensing
$CuCo_2O_4$	$Cu^{2+}/Co^{2+,3+}$	Surface activity, morphology	Pollutant degradation
$NiFe_2O_4$	Ni^{2+}/Fe^{3+}	Magnetic response, surface functionality	Extraction of water pollutants

Table 1. Representative DES-derived spinel ferrites, their compositional characteristics, tunable physicochemical properties and environmental applications.

4. Scheelite-Type Metal Molybdates for Environmental Applications

Scheelite-type metal oxides represent an important class of ternary functional materials with the general composition ABO_4. Among them, metal molybdates containing Mo^{6+} as the B-site cation have attracted considerable attention because of their structural diversity, chemical stability, optical properties, catalytic activity and electrochemical characteristics [37–39,73]. Depending on the nature and oxidation state of the constituent cations, metal molybdates can adopt different crystallographic arrangements, with their structural stability being strongly influenced by ionic size, coordination environment and cationic interactions.In a typical scheelite-type structure, the Mo^{6+} centres are surrounded by oxygen atoms to form MoO_4 tetrahedral units, while the A-site metal ions occupy larger coordination environments within the oxide framework [37–39]. The arrangement and distortion of these polyhedral units can influence the electronic structure, defect chemistry and surface reactivity of the material. Consequently, variation of the A-site cation provides an effective strategy for modifying the physicochemical and electrochemical behaviour of molybdates.

Metal molybdates such as $MgMoO_4$, $MnMoO_4$, $CaMoO_4$ and related compounds have been investigated for

applications in photocatalysis, sensing, energy storage, dielectric devices and heterogeneous catalysis [38,39,73]. Their functional performance is strongly associated with crystallinity, particle size, morphology, specific surface area and defect concentration. At the nanoscale, these characteristics can be manipulated to increase the number of accessible active sites and improve interfacial charge-transfer processes.

The synthesis methodology plays an important role in determining these structural characteristics. Conventional preparation routes for metal molybdates include precipitation, sol–gel processing, hydrothermal synthesis, combustion and solid-state reactions. Although these methods can produce crystalline materials, they may require high temperatures, prolonged reaction times or organic additives. The incorporation of DESs into hydrothermal synthesis provides an alternative strategy in which the reaction medium can simultaneously influence precursor chemistry and material growth.

Choline chloride–urea is particularly relevant as a reaction medium because the components of the DES can interact with metal ions and precursor species through hydrogen bonding and coordination interactions. The resulting chemical environment can modify precursor dissolution, nucleation and crystal-growth kinetics. In addition, the relatively high viscosity of DES media can influence diffusion and aggregation processes, thereby contributing to the development of nanoscale and hierarchical architectures [22,26,28].

The ability to obtain hierarchical structures is particularly advantageous for electrochemical applications. Hierarchical materials contain structural features across multiple length scales and can combine a high density of active sites with interconnected pathways for mass and electron transport. Such architectures can facilitate the diffusion of analytes from the bulk solution towards electrochemically active regions while maintaining a large electrode–electrolyte interface.The comparison of $MgMoO_4$, $Fe_2(MoO_4)_3$ and $MnMoO_4$ provides an opportunity to examine how the identity and oxidation state of the metal component influence the structural and functional properties of molybdate materials. Differences in metal–oxygen bonding, electronic configuration, defect chemistry and surface characteristics can substantially alter catalytic and electrochemical behaviour. Thus, compositional variation represents an important component of rational molybdate design rather than simply a change in chemical composition.

Among these materials, $MnMoO_4$ is particularly attractive for electrochemical sensing because manganese can participate in redox processes while the molybdate framework provides a structurally stable oxide matrix. The combination of nanoscale dimensions, hierarchical morphology and redox-active centres can enhance the interaction between the electrode and target analytes.The environmental significance of such materials is further demonstrated through their application to the simultaneous determination of chemically diverse contaminants. Hydroquinone, mercury ions and nitrite represent important environmental pollutants originating from industrial, agricultural and other anthropogenic sources. Their simultaneous detection is analytically challenging because the target species differ substantially in their chemical nature and electrochemical behaviour.

A well-designed molybdate-modified electrode can facilitate the detection of multiple species by providing catalytic sites and favourable electron-transfer pathways. The resulting analytical response depends on the adsorption of target molecules or ions, interfacial charge transfer and the intrinsic electrocatalytic properties of the modifier [62–66]. Therefore, the development of DES-derived metal molybdates provides an important connection between sustainable material synthesis, structural engineering and environmental analysis.

5. Layered Double Hydroxides and Carbon Nitride-Based Heterostructures

Layered double hydroxides (LDHs) constitute an important family of anionic clay-like materials that have received considerable attention because of their compositional versatility, layered architecture, ion-exchange capacity and surface reactivity [40–43]. Structurally, LDHs are derived from brucite-like metal hydroxide layers in which a fraction of divalent metal cations is replaced by trivalent cations. This substitution generates positively charged hydroxide sheets, with the excess charge being compensated by anions and water molecules located within the interlayer galleries.

The general composition of LDHs can be expressed as $[M^{2+}_{1-x}M^{3+}_{x}(OH)_2]^{x+}(A^{n-})_{x/n}\cdot zH_2O$, where M^{2+} and M^{3+} represent divalent and trivalent metal cations, respectively, A^{n-} represents the charge-balancing interlayer anion, x corresponds to the fraction of trivalent cations and z represents the amount of interlayer water [40–43]. The ability to vary the identity and ratio of the constituent metal ions, together with the possibility of exchanging interlayer anions, provides extensive opportunities for tailoring the structural and chemical properties of LDHs.

The positively charged hydroxide layers and accessible interlayer regions provide LDHs with distinctive anion-exchange and intercalation capabilities. These properties, together with their relatively high surface area, tunable composition and abundance of surface hydroxyl groups, have resulted in their investigation in catalysis, adsorption, environmental remediation, drug delivery, energy storage and electrochemical applications [41,42]. The chemical flexibility of LDHs also permits the incorporation of transition-metal ions, allowing their electronic and redox properties to be adjusted according to the intended application.

Transition-metal-based LDHs are particularly interesting for electrocatalysis because their metal centres can undergo reversible oxidation-state changes during electrochemical processes. Among these materials, Ni–Fe LDHs have attracted extensive attention because of the complementary redox characteristics of nickel and iron and their compositional tunability [43,74]. The interaction between Ni and Fe centres can modify the electronic structure of the hydroxide layers and provide catalytically active sites for interfacial reactions.Despite these advantages, the relatively low intrinsic electrical conductivity of LDHs can restrict their performance in applications that require efficient charge transport. Aggregation of nanosheets can also decrease the accessible surface area and reduce the number of electrochemically active sites. These limitations have stimulated the development of LDH-based composites and heterostructures in which the hydroxide layers are coupled with electrically active carbonaceous materials [74,75].

Graphitic carbon nitride (g-C_3N_4) is a nitrogen-rich, metal-free semiconductor that has attracted significant interest because of its chemical stability, suitable electronic structure, thermal resistance and comparatively facile preparation [76,77]. Its framework consists predominantly of carbon and nitrogen arranged into conjugated structural units, providing an extended π-electron system capable of participating in charge-transfer processes. The electronic properties of g-C_3N_4 can be further modified through elemental doping, defect engineering and heterostructure formation.

Sulfur doping is one such strategy for modifying the electronic characteristics of g-C_3N_4. Because sulfur possesses a different atomic size and electronic configuration from nitrogen,its incorporation can introduce structural distortions, defect states and changes in charge distribution within the carbon nitride framework. These modifications can influence charge-carrier transport and the availability of surface-active sites [76–78].The combination of LDHs with doped carbon nitride provides an effective approach for overcoming the limitations associated with the individual components. In particular, coupling Ni–Fe LDH with sulfur-doped g-C_3N_4 can produce a two-dimensional/two-dimensional heterostructure in which the two components establish intimate interfacial contact. Such an architecture can improve the distribution of LDH nanosheets, reduce aggregation and provide additional pathways for charge transport [75–78].

The interfacial interaction between Ni–Fe LDH and sulfur-doped g-C_3N_4 can generate synergistic effects that are not readily achievable using either component independently. The LDH contributes redox-active Ni and Fe centres, while the carbon nitride framework can facilitate charge transport and provide an extended surface for analyte interaction. Sulfur incorporation can further modify the electronic environment and introduce additional active sites. The resulting heterostructure therefore combines compositional tunability, two-dimensional morphology and interfacial electronic interactions.

The use of a DES during the synthesis of such heterostructures provides an additional dimension of control. Choline chloride–urea can act as a structured reaction medium in which metal precursors and carbon-nitride-related components interact under controlled conditions. The hydrogen-bonding environment and viscosity of the DES can influence precursor dispersion, nucleation and growth, potentially promoting the formation of well-integrated

nanoscale architectures. Thus, the DES is not simply a solvent replacement but can contribute to the development of the final material structure [22,26,28].The resulting Ni–Fe LDH/sulfur-doped g-C_3N_4 heterostructure is particularly relevant to electrochemical detection of nitroimidazole pharmaceuticals such as dimetridazole. Nitroimidazole compounds contain electrochemically reducible nitro functionalities, making them suitable targets for voltammetric determination [62–66]. However, achieving sensitive detection in complex matrices requires an electrode interface with efficient electron transfer, sufficient active-site density and favourable interactions with the analyte.

The two-dimensional heterostructure can provide a large accessible interface between the electrode, electrolyte and target molecules. Redox-active nickel and iron centres may facilitate electron-transfer reactions, while the carbon nitride component can improve interfacial conductivity and structural stability. The combined effects can lead to enhanced peak currents, reduced charge-transfer resistance and improved analytical characteristics.The performance of such a sensor is generally evaluated using sensitivity, linear response range, detection limit, selectivity, repeatability, reproducibility and long-term stability. These parameters are particularly important for pharmaceutical contaminants because their concentrations in environmental matrices can be low and their electrochemical responses may be influenced by coexisting species [62–66].

More broadly, LDH/carbon nitride heterostructures illustrate an important principle in modern functional-material design: combining components with complementary physicochemical characteristics can generate interfacial properties that exceed those of the individual constituents. When this heterostructure engineering is coupled with DES-mediated synthesis, the resulting strategy simultaneously addresses material functionality and synthetic sustainability.

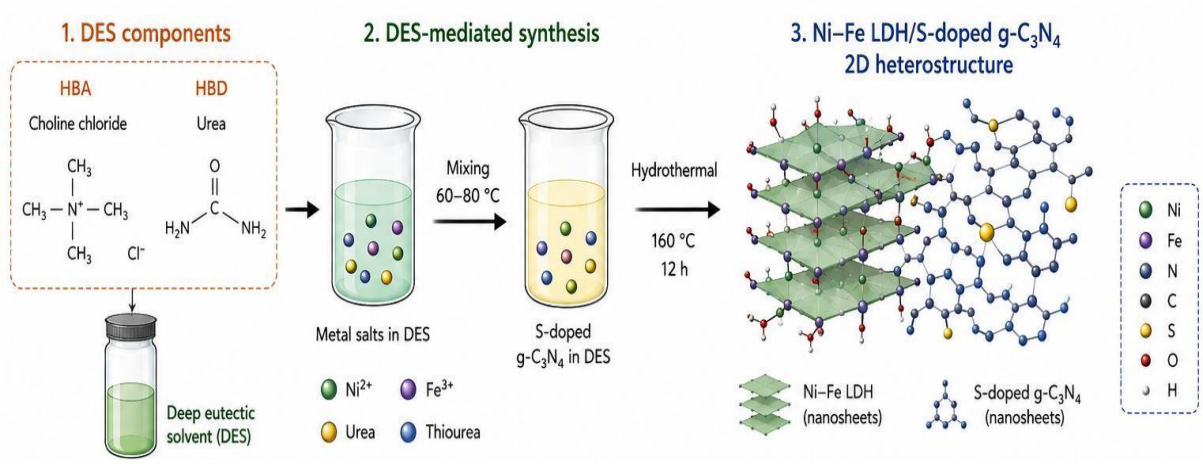


Figure 4. Schematic representation of DES-mediated formation of Ni–Fe LDH/sulfur-doped g-C_3N_4 two-dimensional heterostructures and their proposed role in interfacial charge transfer during electrochemical sensing.

6. DES-Derived Functional Materials for Electrochemical Detection of Emerging Contaminants

The increasing occurrence of chemical contaminants in water, food, and biological systems has created a growing demand for analytical methods capable of rapid, sensitive, and reliable detection. In addition to conventional pollutants such as heavy metals, phenolic compounds, and synthetic dyes, emerging contaminants including antibiotics, pharmaceutical residues, and antimicrobial agents have received considerable attention because of their persistence, biological activity, and potential adverse effects on ecosystems and human health [69–72,79]. Their detection is particularly challenging because these compounds may occur at trace concentrations and often coexist with chemically diverse species in complex environmental matrices.

Electrochemical sensing has emerged as an attractive strategy for environmental analysis because of its inherent

advantages, including rapid response, relatively simple instrumentation, low sample consumption, high sensitivity, and potential for miniaturization and on-site monitoring [62–66]. In contrast to conventional chromatographic and spectroscopic methods, electrochemical techniques can provide direct information about the redox behavior of target species while requiring comparatively simple electrode configurations. Techniques such as cyclic voltammetry (CV), differential pulse voltammetry (DPV), square-wave voltammetry (SWV), and electrochemical impedance spectroscopy (EIS) are therefore widely employed for evaluating the interaction between analytes and modified electrode surfaces [62,63].The performance of an electrochemical sensor is strongly governed by the physicochemical characteristics of the electrode interface. Conventional bare electrodes may exhibit limited electrocatalytic activity, sluggish electron-transfer kinetics, insufficient selectivity, or inadequate sensitivity toward trace contaminants. The incorporation of nanostructured functional materials can overcome several of these limitations by increasing the electrochemically active surface area, providing abundant adsorption and catalytic sites, facilitating interfacial electron transfer, and modifying the local chemical environment of the electrode [62–66]. Consequently, the rational design of electrode modifiers has become a central aspect of contemporary electrochemical sensor development.

Within this context, DES-derived materials offer an additional level of control because the solvent employed during synthesis can influence nucleation, crystal growth, particle aggregation, surface chemistry, defect formation, and final morphology. Thus, the role of a DES extends beyond that of a conventional reaction medium. Depending on its composition, a DES may simultaneously function as a solvent, coordinating medium, template, stabilizing environment, or reaction-promoting agent [22,26,28]. Such multifunctionality is particularly valuable for obtaining nanoscale and hierarchical materials with properties tailored toward electrochemical applications.

The contaminants investigated using DES-derived materials encompass several environmentally relevant classes, including nitroaromatic compounds, antibiotics, pharmaceutical residues, phenolic compounds, heavy-metal ions, inorganic ions, and synthetic dyes. Nitroaromatic compounds such as 4-nitrophenol are important industrial pollutants, whereas nitrofuran and nitroimidazole pharmaceuticals can enter environmental systems through pharmaceutical and agricultural activities [79–81]. Hydroquinone represents an important phenolic contaminant associated with industrial processes, while mercury ions are among the most concerning toxic metal species because of their persistence and bioaccumulation [82,83]. Nitrite is another environmentally relevant inorganic contaminant whose excessive occurrence in water can pose risks to ecological and human health [84].

A notable advantage of DES-derived nanomaterials is their suitability for simultaneous electrochemical determination. Environmental samples commonly contain multiple contaminants rather than a single target species. Therefore, a sensor capable of distinguishing and quantifying several analytes within a single measurement can reduce analytical time, sample consumption, and experimental complexity [62–66]. The successful simultaneous determination of nitrofurantoin and 4-nitrophenol using $MgFe_2O_4$-based electrodes illustrates the potential of spinel ferrites for multi-analyte sensing. Similarly, the investigation of ZnB_2O_4 spinels with systematic B-site substitution provides an opportunity to understand how compositional changes influence the simultaneous electrochemical response toward nitrofurazone and dimetridazole.

The electrochemical behavior of these materials can be interpreted in terms of several interconnected parameters. Particle size directly affects the available surface area and the number of accessible active sites, while morphology determines analyte diffusion and accessibility of electroactive regions. Crystal structure and cation distribution influence the electronic properties and redox activity of the material, whereas surface defects and oxygen vacancies can provide additional adsorption and reaction sites [60–65]. The overall electrochemical response therefore results from the combined influence of structural, morphological, compositional, and interfacial characteristics rather than from a single material parameter.

For ferrite-based electrodes, the presence of transition-metal centers can facilitate electron-transfer reactions involving electroactive analytes. The substitution of the A-site or B-site cations in spinel structures provides a means of modifying the electronic environment and redox characteristics of the material. Such compositional engineering can consequently alter the peak potential, peak current, charge-transfer resistance, and catalytic kinetics observed

during electrochemical measurements [60–65].Scheelite-type metal molybdates provide another useful platform for multi-analyte detection. The comparison of $MgMoO_4$, $Fe_2(MoO_4)_3$, and $MnMoO_4$ demonstrates how changes in metal composition can influence morphology, surface activity, and electron-transfer behavior. In particular, hierarchical $MnMoO_4$ architectures can provide increased surface accessibility and favorable electrochemical characteristics, making them suitable for the simultaneous determination of hydroquinone, Hg^{2+}, and NO_2^-. The simultaneous detection of chemically different analytes is particularly significant because it demonstrates the ability of a single functional interface to interact with multiple contaminant classes.

Heterostructure formation represents a further strategy for improving electrochemical performance. Combining two materials with complementary electronic and surface properties can generate interfacial charge redistribution and additional pathways for electron transport. The DES-mediated Ni–Fe LDH/sulfur-doped graphitic carbon nitride system exemplifies this approach. The Ni–Fe LDH component provides redox-active metal centers, while sulfur-doped graphitic carbon nitride contributes a nitrogen-rich conductive framework and additional surface-active sites. Their intimate 2D/2D contact can facilitate charge transfer while minimizing aggregation of individual components [74–78].

Such heterostructural design is particularly relevant to the electrochemical detection of dimetridazole, a nitroimidazole pharmaceutical containing an electrochemically reducible nitro group. The improved interfacial characteristics of the composite can promote electron transfer between the analyte and electrode, thereby enhancing the analytical signal. Similar principles can be applied to other pharmaceutical and antibiotic contaminants in which the electrochemical response is governed by the accessibility of redox-active functional groups at the electrode interface [62–66].The practical performance of a sensor cannot, however, be established solely from its current response. Important analytical parameters include linear dynamic range, limit of detection, sensitivity, selectivity, repeatability, reproducibility, stability, response time, and recovery in real samples [62–66]. A material that produces a high current under optimized laboratory conditions may not necessarily provide reliable environmental analysis if it suffers from poor selectivity or instability. Accordingly, evaluation using complex real matrices such as water, food, pharmaceutical, or biological samples is essential for establishing practical applicability.

DES system	**Material**	**Target analyte**	**Electrode / Technique**	**Principal analytical characteristic**
ChCl:Urea	NiFe-LDH/Mo_2C	Nitrite	GCE / DPV	High sensitivity
ChCl:Urea	NiFe-LDH/S-g-C_3N_4	Dimetridazole	GCE / DPV	Enhanced charge transfer
ChCl:Urea	$PrVO_4$ nanoparticles	Furaltadone	GCE / DPV	Low detection limit
ChCl:Malonic acid	$MgFe_2O_4$	4-Nitrophenol	GCE / DPV	Good sensing response
ChCl-based DES	$MnFe_2O_4$	Methylene blue	GCE / DPV	Improved electrocatalytic activity

ChCl-based DES	$BaMoO_4$/rGO	Bisphenol A	GCE / DPV	Enhanced peak current

Table 2. Representative DES-derived functional materials investigated for electrochemical detection of environmental and pharmaceutical contaminants, highlighting the DES system, material composition, target analytes, electrode configuration, electrochemical technique, and principal analytical characteristics.

An important feature connecting the different systems discussed in this review is the relationship between DES composition, material structure, and analytical performance. Variation in the HBA–HBD combination can alter precursor coordination and reaction kinetics, which subsequently affects crystallization and morphology. These structural differences influence surface area, charge-transfer properties, adsorption behavior, and catalytic activity, ultimately determining the electrochemical response toward a particular analyte. Therefore, the development of DES-derived sensors should move beyond empirical optimization toward a mechanistic understanding of how solvent chemistry governs material formation and electrochemical function.

From a broader environmental perspective, DES-derived electrochemical sensors can contribute to decentralized and real-time monitoring strategies. Their compatibility with miniaturized electrodes and portable electrochemical instrumentation creates opportunities for field-based analysis where conventional laboratory techniques may be impractical [62,63]. When combined with selective recognition strategies, microfabricated electrodes, wireless readout, and data-processing platforms, these materials could contribute to next-generation monitoring systems capable of detecting contaminants at their source or directly within affected environments.

Thus, the integration of DES-mediated synthesis with nanostructured electrode engineering represents a promising pathway toward sensitive and multifunctional environmental sensors. The ability to systematically modify composition, morphology, crystal structure, and heterointerfacial interactions provides a versatile materials platform for addressing the increasingly complex requirements of contemporary environmental monitoring.

7. Critical Perspective: Sustainability, Challenges and Future Opportunities

Despite the rapid expansion of DES chemistry, several challenges must be addressed before DES-mediated materials synthesis can be considered universally sustainable. The first is the tendency to equate low volatility with environmental benignity. DESs may have negligible vapor pressure, but this does not necessarily imply low toxicity or complete biodegradability [37–39]. The environmental behavior of a DES depends on the identities of its constituents and their interactions.The second challenge concerns viscosity and mass transfer. High viscosity can be advantageous for morphology control but disadvantageous for large-scale processing because it may reduce mixing efficiency and mass transport. Industrial implementation will therefore require careful optimization of temperature, water content, reactor design, and solvent composition [32,49].

A third consideration is DES recovery and reuse. Although DESs are frequently described as recyclable, practical recovery can be complicated when the solvent becomes contaminated with metal ions, reaction by-products, or nanoparticles. Efficient separation and purification strategies are consequently required to establish genuine process sustainability [37–39].The fourth challenge is energy consumption during post-treatment. Many DES-derived metal oxides require drying and calcination to develop the desired phase and crystallinity. The energy requirements associated with these stages should be incorporated into sustainability assessments rather than omitted from the analysis.

Finally, the field would benefit from greater use of quantitative green metrics and life-cycle assessment. Material yield, E-factor, process mass intensity, solvent recovery, energy consumption, toxicity, and carbon footprint should increasingly accompany conventional characterization data. Such assessments would allow meaningful comparison between DES-mediated synthesis and established aqueous, hydrothermal, sol–gel, combustion, and solid-state

approaches [37–39].Future research should therefore move toward rational DES design, in which computational chemistry, spectroscopy, thermodynamics, and machine-learning approaches are combined to predict HBA/HBD interactions and their influence on precursor chemistry. Such strategies could reduce empirical trial-and-error optimization and facilitate the development of application-specific DESs.

Another promising direction is the integration of DES synthesis with hierarchical materials engineering. Combining controlled nanoscale crystallization with microscale or mesoscale architectures could provide materials with simultaneously high active-site density, rapid mass transport, and enhanced mechanical stability. The integration of ferrites, molybdates, LDHs, and carbon-based materials into multifunctional heterostructures is particularly promising for environmental remediation and sensing.The ultimate objective should therefore not be simply to replace a conventional solvent with a DES. Instead, the field should pursue a more comprehensive concept of sustainable materials-by-design, where solvent composition, precursor chemistry, crystal structure, morphology, surface functionality, energy consumption, recyclability, and final application are optimized as an interconnected system.

Conclusions and Outlook

Deep eutectic solvents have evolved from an alternative class of low-melting liquids into versatile platforms for the synthesis and engineering of functional materials. Their principal scientific value lies in their chemical tunability and multifunctionality, enabling them to participate actively in precursor dissolution, coordination, nucleation, crystal growth, stabilization, templating, and surface modification [22,26,28,36].The combination of DES chemistry with ternary metal oxides and layered materials offers a particularly powerful strategy for developing materials with controlled structural and electrochemical properties. Spinel ferrites provide compositional flexibility, magnetic separability, redox activity, and photocatalytic functionality; scheelite-type metal molybdates offer tunable structural and electrochemical characteristics; while Ni–Fe LDH/carbon nitride heterostructures provide complementary redox and charge-transport properties.

Across these material families, a common design principle emerges: DES composition can influence material structure, structure determines physicochemical properties, and these properties ultimately govern sensing and catalytic performance.The environmental relevance of these systems is equally important. Electrochemical detection of nitroaromatic compounds, pharmaceuticals, phenolic pollutants, heavy-metal ions, inorganic contaminants, and dyes demonstrates the potential of DES-derived materials for rapid environmental monitoring. Meanwhile, ferrite-based photo-Fenton and photocatalytic systems extend their utility from detection toward pollutant degradation and remediation.

Toxicity, biodegradability, renewable sourcing, solvent recovery, water consumption, energy demand, post-treatment, and life-cycle impacts must be considered collectively. A DES-based synthesis should therefore be regarded as genuinely sustainable only when the complete process, rather than the solvent alone, demonstrates an environmental advantage.Taken together, the convergence of green solvent engineering, DES chemistry, hierarchical materials design, compositional tuning, and electrochemical technology establishes a promising foundation for next-generation environmental sensing and remediation. Continued integration of mechanistic understanding with quantitative sustainability assessment will be essential for translating DES-mediated materials synthesis from laboratory-scale demonstrations into robust, scalable, and genuinely sustainable technologies.

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