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A REVIEW OF DEEP EUTECTIC SOLVENTS IN GREEN EXTRACTION OF CHITOSAN: COMPOSITION, EFFICIENCY, AND RECYCLABILITY

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Abstract

Chitosan, a biopolymer derived from chitin-rich biomass such as crustacean shells, has garnered attention for its biodegradability, biocompatibility, and wide-ranging applications. However, conventional chemical extraction methods relying on strong acids and bases pose significant environmental and safety concerns, often leading to molecular degradation and low product quality. This study explores the use of deep eutectic solvents (DESs) as a green alternative for chitosan extraction. DESs, formed from combinations of hydrogen bond donors and acceptors, offer tunable properties, lower toxicity, and recyclability. The article highlights the structural advantages, extraction efficiency, and environmental benefits of DESs over conventional methods. It also examines the integration of process intensification technologies, such as microwave and ultrasound-assisted extraction, to enhance yield and reduce energy consumption. The findings underscore DESs' potential to produce high-purity chitosan while supporting sustainability goals and industrial scalability, offering a viable pathway toward eco-friendly biopolymer processing.

Keywords: Biopolymer; Chitosan; Deep eutectic solvents; Green extraction; Sustainable processing

Abstrak

Kitosan, biopolimer yang berasal dari biomassa kaya kitin seperti cangkang krustasea, telah menarik perhatian karena biodegradabilitas, biokompatibilitas, dan aplikasinya yang luas. Namun, metode ekstraksi kimia konvensional yang mengandalkan asam dan basa kuat menimbulkan masalah lingkungan dan keamanan yang signifikan, seringkali menyebabkan degradasi molekuler dan kualitas produk yang rendah. Penelitian ini mengeksplorasi penggunaan pelarut eutektik dalam (DESs) sebagai alternatif hijau untuk ekstraksi kitosan. DESs, yang terbentuk dari kombinasi donor dan akseptor ikatan hidrogen, menawarkan sifat yang dapat diatur, toksisitas yang lebih rendah, dan dapat didaur ulang. Artikel ini menyoroti keunggulan struktural, efisiensi ekstraksi, dan manfaat lingkungan DES dibandingkan metode konvensional. Artikel ini juga mengkaji integrasi teknologi intensifikasi proses, seperti gelombang mikro dan ekstraksi berbantukan ultrasonik, untuk meningkatkan hasil dan mengurangi konsumsi energi. Temuan ini menggarisbawahi potensi DES untuk menghasilkan kitosan dengan kemurnian tinggi sambil mendukung tujuan keberlanjutan dan skalabilitas industri, menawarkan jalur yang layak menuju pemrosesan biopolimer ramah lingkungan.

Kata kunci: *Biopolimer; Ekstraksi hijau; Kitosan; Pelarut eutektik dalam; Pemrosesan berkelanjutan*

1. INTRODUCTION

Chitosan extraction from crustacean biomass, such as shrimp shells and squid pens, predominantly uses conventional chemical methods involving strong acids and bases. These traditional protocols typically utilize sodium hydroxide (NaOH) for deproteinization and hydrochloric acid (HCl) for demineralization. While such treatments effectively enhance extraction yields,

Table 1. Production yields, purity levels, and environmental benefits of various chitosan sources

Sources	Yields (%)	Purity (%)	sustainability potential
Crustaceans	~30–40	~85–95	Moderate (by-product, seasonal) (Huang et al. 2018; Vicente et al., 2021)
Insects	~15–28	~70–90	High (fast reproduction, less resource use) (Yi et al., 2024)
Fungal	~4–9	~77–90	High (Scalable, controlled cultivation) (Cao et al., 2012).

they raise substantial environmental and occupational health concerns due to the generation of alkaline and acidic effluents. Improper disposal of these chemical residues can result in significant ecological damage, including water and soil contamination, as well as bioaccumulation and endocrine disruption in ecosystems (Dong et al., 2023; Egorov et al., 2023; Kim et al., 2023). Workers involved in these processes are also at risk due to the corrosive nature of the chemicals, which can cause burns and respiratory complications (Abbas and Al-Shammari, 2022; Hegde and Selvaraj, 2024). Furthermore, these aggressive extraction conditions may compromise the structural integrity of chitosan, especially its molecular weight and degree of deacetylation (Ibrahim et al., 2019). In response to these drawbacks, alternative extraction techniques have been explored. These include milder organic acid treatments and biological methods such as enzymatic hydrolysis and microbial fermentation (Beaney et al., 2005; Gohi et al., 2016). Although these methods are environmentally friendlier, they often suffer from low extraction efficiencies and extended processing durations, limiting their scalability and industrial applicability (Hegde and Selvaraj, 2024). The variability in raw material composition, particularly in protein, mineral, and chitin content, further complicates process standardization and affects the consistency of the final chitosan product (Ibrahim et al., 2019; Ooi et al., 2021).

Amidst these challenges, deep eutectic solvents (DES) have emerged as promising green alternatives for biopolymer extraction. Composed of a hydrogen bond acceptor (HBA) and donor (HBD), DESs possess unique physicochemical properties such as tunable polarity, low toxicity, and biodegradability (Bisht et al., 2021; Morais et al., 2020; Paiva et al., 2014). These solvents are effective under mild conditions, reducing energy consumption and chemical waste. For instance, Triunfo et al. (2022) achieved a 98% demineralization efficiency using natural DESs on *Hermetia illucens*, outperforming conventional acid-based approaches. Bradić et al. (2019) further demonstrated that DES can selectively extract chitin from crustacean shells while preserving its structural integrity. Additionally, DESs support deacetylation reactions, as shown by (Vicente et al., 2020), enabling the production of high-quality chitosan with tailored physicochemical properties.

DES-based extractions still face technical challenges such as high viscosity, inconsistent performance, and limited scalability. Recent studies propose solutions including the use of co-solvents or ternary DESs to improve mass transfer, and response surface methodology for optimizing solvent composition and extraction parameters (Szopa et al., 2024). Efforts to enhance solvent recyclability through closed-loop regeneration systems are also progressing. Furthermore, molecular modelling and spectroscopic tools have helped clarify how DESs interact with various chitinous substrates, revealing that solvent performance is highly dependent on biomass type (Li et al., 2022). For example, crustacean-based substrates rich in minerals require acidic DESs for demineralization, while fungal biomass with high glucan content may benefit from milder, protein-targeting DESs (Bradić et al., 2019). Understanding and matching DES systems to substrate characteristics is thus crucial for process refinement and broader applicability.

Furthermore, emerging studies have begun to demonstrate the application potential of DES-extracted chitosan in domains such as biomedicine and environmental engineering. Chitosan obtained via natural DESs has shown superior physicochemical properties, such as higher degree of deacetylation (up to 91%) and preserved molecular weight (~481 kDa), which are crucial for biomedical functionalities like drug delivery, wound healing, and antimicrobial coatings (Kyriakidou et al., 2021; Zhou et al., 2014). In environmental contexts, DES-derived chitosan exhibits enhanced solubility and surface activity, improving its performance in water treatment and heavy metal adsorption (Renault et al., 2009). These advantages, often unattainable through conventional acid–alkaline processes due to polymer degradation and residual chemical contamination, underscore the transformative potential of DESs in producing high-functionality, application-ready biopolymers. This study provides a holistic view that bridges process development with application-oriented outcomes by presenting DES-derived chitosan's extraction characteristics and functional performance.

2. CHITOSAN: STRUCTURE, SOURCES, AND EXTRACTION METHODS

2.1 Structure and Physicochemical Properties of Chitosan

Chitosan is a linear polysaccharide primarily composed of β-(1→4)-linked d-glucosamine units derived from the partial or complete alkaline deacetylation of chitin. This transformation removes acetyl groups from N-acetyl-d-glucosamine residues, increasing the number of free amino groups available along the polymer chain (Hemmami et al., 2024; Sinardi et al., 2018; Suneeta et al., 2016). The degree of

deacetylation (DDA) is a critical structural parameter, reflecting the proportion of deacetylated units in the chitosan chain and significantly influencing the biopolymer's physicochemical properties and application potential (Sarofa et al., 2025; Synowiecki and Nadia Ali Abdul Quawi Al-Khateeb, 2003).

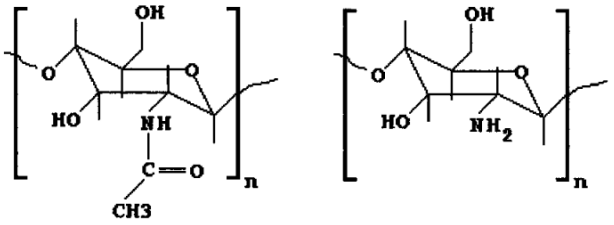

Figure 1. Chemical structure of (a) Chitin, (b) Chitosan

The DDA directly determines the cationic nature of chitosan. In acidic environments (pH < 6.5), the amino groups become protonated, imparting a net positive charge to the polymer (Kim, 2018). This protonation facilitates electrostatic interactions with negatively charged entities, such as microbial membranes, proteins, and contaminants in aqueous systems. Consequently, chitosan exhibits notable bioadhesiveness, antimicrobial properties, and flocculating capacity, supporting its widespread use in biomedical applications and environmental remediation (Hemmami et al., 2024; Kim, 2018).

Chitosan's solubility is also closely linked to its DDA and molecular weight. High DDA values increase solubility in dilute acidic solutions by promoting chain repulsion and hydration due to the abundance of ionizable amino groups (Ahing and Wid, 2016; Suneeta et al., 2016). In contrast, chitosan with lower DDA or higher molecular weight often demonstrates limited solubility due to reduced protonation and increased chain entanglement. These attributes can influence film-forming behavior and rheological properties, which are essential for material processing and end-use performance (Potivas and Laokuldilok, 2014).

The deacetylation process influences more than just functional group availability—it also affects molecular weight distribution, chain flexibility, and the distribution of deacetylated regions. Harsh alkaline treatment can result in polymer chain scission, reducing molecular weight and altering viscosity, mechanical strength, and thermal stability (Potivas and Laokuldilok, 2014; Synowiecki and Nadia Ali Abdul Quawi Al-Khateeb, 2003). Studies using molecular simulations suggest that non-uniform deacetylation can affect self-assembly, aggregation, and solubility behavior (Cambiaso, 2025).

Dacetylation parameters**,** such as temperature, alkali concentration, and exposure time, enable tailoring of chitosan's solubility, cationic properties, and reactivity, which must be controlled to meet specific functional requirements. Chitosan with a high DDA is particularly suited for applications requiring high chemical reactivity and solubility, such as drug delivery or water purification, whereas lower DDA chitosan may be preferred for applications requiring enhanced mechanical strength (Ahing and Wid, 2016; Potivas and Laokuldilok, 2014; Sarofa et al., 2025). Thus, understanding and optimizing the structural determinants of chitosan remains a pivotal step in maximizing its application versatility.

2.2 Conventional Extraction Techniques

The conventional extraction of chitosan from raw chitin-containing biomass, such as crustacean shells, primarily involves a two-step chemical process: demineralization and deproteinization. In the demineralization phase, hydrochloric acid (HCl) is typically used to dissolve inorganic components, mainly calcium carbonate. Protocols commonly employ dilute HCl solutions with concentrations ranging from 1% to 3% (w/v), which are sufficient to remove mineral content while minimizing structural damage to chitin (Abidin et al., 2020; Hossain and Iqbal, 2014). Gîjiu et al. (2022) and Hossain and Iqbal (2014) demonstrated that treating raw material with 3% HCl at approximately 25°C for 1 hour yields efficient mineral removal without excessive depolymerization. However, extended exposure or harsher acid conditions can lead to degradation of the chitin polymer, adversely affecting the molecular weight and acetylation profile of the final product (Abidin et al., 2020; Percot et al., 2002).

Subsequent deproteinization uses sodium hydroxide (NaOH), which disrupts and solubilizes proteinaceous materials. Treatment conditions vary considerably based on the biomass source and targeted purity level. For instance, milder conditions may involve 4% NaOH at ambient temperature (≈28°C), whereas more aggressive protocols use 50% NaOH at 110°C for up to 3 hours to achieve complete deproteinization (Gîjiu et al., 2022; Hossain and Iqbal, 2014; Nardo et al., 2019). While higher concentrations and temperatures enhance protein removal, they can also degrade the chitin matrix, resulting in low-molecular-weight chitosan with an altered degree of deacetylation (Gîjiu et al., 2022; Pachapur et al., 2015).

To optimize these steps, experimental designs such as response surface methodology and Box–Behnken models have been employed, allowing for systematic assessment of variables like reagent concentration, temperature, and exposure time (Arpi et al., 2022; Gîjiu et al., 2022). These models facilitate the identification of parameter combinations that balance effective purification with minimal biopolymer degradation, tailored to specific source materials (Abidin et al., 2020; Hossain and Iqbal, 2014; Percot et al., 2002).

Despite their effectiveness, acid-base extraction methods pose several environmental and technical challenges. Substantial quantities of hazardous chemical waste are generated, necessitating costly and resource-intensive neutralization and disposal procedures (Kaur and Dhillon, 2013; Zulkarnain et al., 2024). These include HCl and NaOH effluents, which, if mismanaged, contribute to environmental pollution (Maddaloni et al., 2020; Tan et al., 2022). Additionally, processes involving high temperatures significantly

Tabel. 2. Comparative analysis of conventional and des-based chitosan extraction

Aspect	Conventional Extraction	DES-based Extraction
Process Description	Chemical demineralization (HCl) and deproteinization (NaOH) under harsh conditions (high temperature, strong acids/bases) (Rissouli et al., 2024).	Utilizes deep eutectic solvents (DESs) composed of hydrogen bond donors and acceptors, operating under milder, environmentally friendly conditions (Wang, 2023)
Extraction Efficiency	High demineralization (up to 97–98%) and deproteinization (~90%), but requires multiple chemical treatments (Rissouli et al., 2024).	Demineralization ~97.78±0.73%, deproteinization ~81.33±0.91% using lactic-NADES (Sunton et al., 2024); similar efficiencies (up to 99%) also achieved with novel ternary DESs.
Yield	Yields typically range from 15% to 40%, depending on the raw material and process optimization (Belwal et al., 2020).	Chitosan yield using DES methods reported at ~21–25%, depending on solvent type and extraction conditions (Sunton et al., 2024; Wang, 2023).
Molecular Weight	Chemical methods often cause degradation, resulting in variable molecular weights (200–500 kDa) (Rissouli et al., 2024).	Molecular weight tunable depending on DES composition: 264–541 kDa with lactic acid/glycerol DES; malic acid systems yield lower MW (~<300 kDa) (Wang, 2023).
Purity	High purity (>90%) achievable, but risk of residual chemical contamination (Belwal et al., 2020).	High purity (>91–92%) maintained even after multiple recycling cycles of DES (Sunton et al., 2024; Wang, 2023).
Recyclability	Solvents (acids/bases) are not recyclable; significant chemical waste is generated (Calvo-Flores et al., 2018).	DESs are recyclable for multiple cycles without significant efficiency loss (Isci & Kaltschmitt, 2022).
Environmental Impact	Significant due to the use of strong acids/bases, large water consumption, and generation of hazardous waste (Mohan et al., 2022).	Environmentally friendly; reduced chemical waste, lower toxicity, and alignment with green chemistry principles (Sunton et al., 2024; Wang, 2023).
Energy Consumption	High, due to heating and intensive washing steps (Rissouli et al., 2024).	Lower, especially when combined with microwave or ultrasound-assisted extraction (Coscarella et al., 2023; Durante-Salmerón et al., 2024).

increase energy consumption and further elevate the risk of polymer degradation (Maddaloni et al., 2020).

Maintaining the delicate balance between efficient protein removal and the preservation of chitosan's structural and functional integrity remains a technical bottleneck. Harsh alkali conditions, although effective, may cause over-deacetylation, negatively affecting the solubility and biological activity of the chitosan (Maddaloni et al., 2020). In response, alternative approaches such as microwave-assisted extraction have been explored for their ability to reduce processing time and energy demands, although issues related to consistency and scalability persist (Apriyanti et al., 2018).

2.3 Green Chemistry in Biopolymer Extraction

Green chemistry in biopolymer extraction encompasses environmentally sustainable approaches prioritizing energy efficiency, low toxicity, and resource recycling. These methods align with the twelve principles of green chemistry, aiming to reduce the ecological footprint and the operational burden associated with traditional chemical processes (Chemat et al., 2012). Green extraction focuses on minimizing waste and using safer solvents and technologies to achieve high extraction efficiency with reduced environmental and health risks.

Efficiency in green extraction is achieved through innovative process intensification techniques. Approaches such as one-pot extraction, which combine multiple stages into a single operational step, significantly reduce processing time and resource use (Mukherjee et al., 2023). Technologies like subcritical water extraction (Ferreira et al., 2023), microwave-assisted extraction, and ultrasound-assisted extraction have demonstrated capabilities to accelerate mass transfer, enhance solubilization, and reduce energy consumption while maintaining or improving product yield and quality (Boggia et al., 2016; Pal and Jadeja, 2019). These methods streamline workflows and support high process efficiency with lower operational inputs.

Toxicity minimization is another cornerstone of green extraction. Traditional methods often rely on volatile organic compounds (VOCs) or corrosive mineral acids and bases that are hazardous to both human health and the environment (Mahmood and Moniruzzaman, 2019). In contrast, green extraction

employs non-toxic and biodegradable solvents, such as ethanol, water under subcritical conditions, and deep eutectic solvents (DESs), which are characterized by low volatility and high recyclability (Pal and Jadeja, 2019). Techniques like electroporation, which enable solvent-free extraction, further reduce the chemical load of biopolymer recovery processes (Eleršek et al., 2020).

Resource recycling and circular economy integration are fundamental to green extraction. Closed-loop processes that enable solvent recovery and by-product valorization reduce waste and enhance the operation's sustainability. Studies have demonstrated that solvent systems in green extraction methods can be effectively recovered and reused without significant performance loss, contributing to reduced material demand and environmental burden (Makris & Lalas, 2020; Mukherjee et al., 2023). Moreover, lifecycle assessment studies support that these methods lower energy use and mitigate greenhouse gas emissions (Güler et al., 2024).

3. DEEP EUTECTIC SOLVENTS: DESIGN, MECHANISM, AND APPLICATION IN CHITOSAN EXTRACTION

Deep eutectic solvents (DES) are a class of "green" solvents formed by mixing two or three components to create a eutectic system with a melting point significantly lower than that of the pure individual components (Hong et al., 2019; Kianpour et al., 2022; Velez and Acevedo, 2022). Their formation relies heavily on strong hydrogen bonding interactions between the hydrogen bond donor (HBD) and the hydrogen bond acceptor (HBA), disrupting the crystalline structures of the original compounds (Abbott et al., 2007; Doherty and Acevedo, 2018; Hammond et al., 2016). Consequently, DES mixtures remain liquid at relatively low temperatures, facilitating their use in various green extraction applications.

DES are classified based on the number of constituents: binary systems consist of one HBA and one HBD mixed at a specific molar ratio to reach a eutectic point, such as the classic mixture of choline chloride with urea or glycerol (Hong et al., 2019; Kianpour et al., 2022; Velez and Acevedo, 2022). Ternary systems involve three components, allowing for the fine-tuning of properties like viscosity, polarity, and solubility, thereby broadening the applicability of DES in fields like bioactive compound extraction and nanomaterial synthesis (Abranches et al., 2019; Kumar and Banerjee, 2021; Pour et al., 2023).

Key components of DESs are quaternary ammonium salts, notably choline chloride as the HBA, and small molecular HBDs such as urea, glycerol, and organic acids like citric or succinic acid (Hammond et al., 2016; Hong et al., 2019; Paiva et al., 2014). The hydrogen bonding interaction between HBA and HBD not only depresses the melting point but also imparts unique properties such as biodegradability, low toxicity, and economic viability (Doherty and Acevedo, 2018; Paiva et al., 2014; Velez and Acevedo, 2022).

Compared to ionic liquids (ILs), DESs differ significantly in formation, properties, and environmental impact. DESs are prepared by simply mixing HBAs and HBDs, while ILs require complex synthesis to match specific cations and anions (Mahaindran et al., 2023; Suriyanarayanan et al., 2019; Vahidi et al., 2023). DESs typically exhibit moderate viscosity and polarity, which is excellent for biopolymer extraction, while ILs offer high electrochemical stability but face concerns regarding environmental toxicity and cost (Aboshatta and Magueijo, 2021; Espino et al., 2016; Mahaindran et al., 2023). DESs, derived from natural, biodegradable materials, present a greener alternative with simpler preparation and lower environmental burden (Halder and M. Natália D. S. Cordeiro, 2019; Morais et al., 2020).

The formulation of DESs for chitosan extraction commonly involves choline chloride as HBA paired with HBDs like lactic acid, glycerol, acetic acid, or malic acid (Paiva et al., 2014; Vicente et al., 2020). Lactic acid contributes not only to the hydrogen bond network but also to biomass demineralization and deproteinization (Fraige et al., 2018; Strižincová et al., 2024). Glycerol's polyhydroxyl groups enhance solvation and viscosity control (Wang, 2023). Acetic acid, often added in small amounts (~7.5%), provides additional acidity to aid mineral removal without causing significant polymer degradation (Wang, 2023; Zhang and Neau, 2001). Malic acid, although less commonly used, brings a dicarboxylic structure that strengthens the hydrogen bonding network and improves extraction performance (Paiva et al., 2014; Vicente et al., 2020).

Choline chloride's quaternary ammonium structure promotes efficient hydrogen bonding, crucial for disrupting the crystalline structure of chitin during extraction (Doherty and Acevedo, 2018; Paiva et al.,

Table 3. Comparison of conventional vs DES methods for chitosan extraction

Parameter	Conventional Method	DES Method
Yield (%)	20-30 % (Depending on source) (Sunton et al., 2024)	30-45% (higher, depending on DES and source) (Wang, 2024)
Molecular Weight (kDa)	High variability, often degraded (Zhang & Neau, 2001)	264-655 kDa (controlled, higher stability) (Wang, 2023)
Degree of acetylation (%)	79-85 (Sánchez et al., 2021)	85-95 (higher, purer product) (Sunton et al., 2024)
Environmental impact	High chemical use; significant waste (Ben Aoun et al., 2024)	Low chemical usage, eco-friendly, and recyclable solvents (Khandelwal et al., 2016)

2014; Pandey et al., 2017). Organic acids, acting as HBDs, help dissolve proteins and minerals while maintaining chitosan integrity by balancing acidity and hydrogen bonding strength (Pandey et al., 2017). Importantly, solvent viscosity must be optimized, too high viscosity can hinder solute diffusion, while controlled addition of water or adjusting HBA/HBD ratios can mitigate this issue and enhance mass transfer (AlYammahi et al., 2023).

Mechanistically, DESs disrupt the hydrogen-bonded networks binding proteins, minerals, and chitin in crustacean biomass. DESs form new hydrogen bonds with functional groups in proteins and minerals, destabilizing and solubilizing these components (Bradić et al., 2019; Roda et al., 2019). Specific interaction between acidic DES components and calcium carbonate promotes solubilization into calcium salts, with minimal chitin degradation (Vigier et al., 2015; Zhang & Neau, 2001). The optimized hydrogen bonding network ensures selective dissolution of non-chitinous material, preserving chitin's molecular integrity (Vicente et al., 2020; Zhao, 2019).

Experimental findings corroborate the efficiency of DESs. Bradić et al. (2019) extracted chitin from lobster and shrimp shells using choline chloride-based DESs with various donors, achieving mild processing and high biopolymer integrity. Rodrigues et al. (2021) demonstrated the efficacy of low-phytotoxic DESs in recovering chitin from brown crab shells, while Bisht et al. (2021) showcased aDESs for effective dissolution without degradation. Zhao (2019) combined citric acid and DES with microwave assistance, optimizing demineralization and deproteinization. Huang et al. (2018) employed naturally derived DESs for high-purity chitin extraction, while Saravana et al. (2018) extended DES applications to fabricate chitin-based films, supporting downstream valorization.

Optimizing parameters such as temperature, solvent-to-solid ratio, and microwave irradiation time is crucial. Moderate temperatures reduce DES viscosity and enhance extraction kinetics without polymer degradation (Arpi et al., 2022; Segaran et al., 2022). The solvent-to-solid ratio must balance solvent efficiency and economic usage (Palai et al., 2025; Yusof, 2023). Microwave irradiation time must be tuned to maximize extraction yield without thermal damage (Arpi et al., 2022; Palai et al., 2025).

4. PERFORMANCE EVALUATION AND INDUSTRIAL POTENTIAL OF DES IN CHITOSAN EXTRACTION

The extraction of chitin and chitosan using deep eutectic solvents (DESs) is strongly influenced by the solvent's composition, viscosity, and pH, critically impacting extraction yield, purity, and molecular weight. DES composition is pivotal, as the selection of hydrogen bond acceptor (HBA) and hydrogen bond donor (HBD) components defines the polarity and solvation capacity of the solvent. Systems based on choline chloride with glycerol, malic acid, or urea have been successfully applied to dissolve α-chitin and extract chitin from crustacean shells (Huang et al., 2018; Vicente et al., 2020). Tailoring DESs with milder acid functionalities allows simultaneous demineralization and deproteinization while preserving the chitin structure (Rodrigues et al., 2021) and ternary DES systems have been developed to enable room-temperature extraction while preventing polymer degradation (Vicente et al., 2021).

Viscosity also plays a major role, as highly viscous DESs can hinder mass transfer and diffusion, leading to reduced extraction yields despite their ability to stabilize dissolved biopolymers (Ling and Hadinoto, 2022). Lower viscosity enhances kinetics but risks molecular degradation if not carefully managed. Similarly, pH is crucial: acidic DESs promote demineralization and deproteinization but may induce chain hydrolysis (Vicente et al., 2021), whereas near-neutral DESs better preserve molecular weight and chain integrity (Rodrigues et al., 2021). Thus, optimal DES design involves a careful balance between composition, viscosity, and pH to achieve high-purity chitosan with minimal degradation.

Variations in DES formulation further impact chitosan's deacetylation degree and chain length compared to conventional processes. Natural DESs composed of choline chloride and organic acids have produced chitosan with a high degree of deacetylation (91%) and molecular weight (~481 kDa) (Kimi and Hamdi, 2023), while conventional alkaline treatments often cause substantial chain scission (Hossain and Iqbal, 2014). DES viscosity also influences uniformity; high-viscosity solvents impede diffusion and cause heterogeneous extraction, whereas optimized DESs enhance solute penetration and control deacetylation without extensive degradation (Nouri et al., 2016). Moreover, the mild pH conditions in DES systems minimize glycosidic bond cleavage, yielding chitosan with superior macromolecular structure for biomedical and materials applications.

Beyond extraction performance, the recyclability and stability of DESs are essential for sustainable use. Repeated recycling without proper regeneration leads to impurity accumulation, disruption of hydrogen bonding, increased viscosity, and pH drift factors that compromise extraction efficiency (Durante-Salmerón et al., 2024). Studies have shown that DESs can typically be reused for up to 4–6 cycles without significant loss of performance, particularly when low volatility or organic acid-based systems are used (Isci and Kaltschmitt, 2022). However, solvent degradation accelerates after multiple cycles if no regeneration steps are applied. Regeneration techniques include simple filtration, vacuum distillation, or liquid–liquid phase separation to remove residual biomass or degradation products (Huang et al., 2008). Adjusting water content or rebalancing the HBA:HBD ratio is also effective in restoring viscosity and pH (Ijardar et al., 2022).Therefore, to support industrial feasibility, robust, energy-efficient regeneration protocols must be developed alongside solvent screening and process optimization strategies (Ijardar et al., 2022).

When comparing DES-based extraction with conventional acid–base (HCl/NaOH) methods, clear advantages emerge regarding product quality and

Table 4. DES recycling and regeneration strategies

Study / Reference	Regeneration Technique	Cycles Without Efficiency Loss	Notes
Jeong et al. (2015)	Filtration, dilution with water	Up to 5	Effective for particulate-rich DES, a low-cost method
Isci and Kaltschmitt (2022)	Vacuum distillation, phase separation	4–6 cycles	Suitable for low-volatility DES; energy-intensive
Nakasu et al. (2025)	Rebalancing HBA:HBD ratio, pH adjustment	3–5 with correction steps	Requires initial composition data; stabilizes physicochemical profile
Siddiqui et al. (2025)	Removal of degradation products via liquid-liquid extraction	Up to 6 cycles post-regeneration	Targeted for protein/polysaccharide-rich systems
Ijardar et al. (2022)	Integrated monitoring and viscosity control	Varies (dependent on formulation)	Recommended for industrial scaling with real-time feedback systems

environmental sustainability. DES processes produce chitosan with high deacetylation degrees and molecular weights while preserving crystallinity (Kimi and Hamdi, 2023; McReynolds et al., 2022). In contrast, harsh acid–alkali protocols typically disrupt crystalline structures and lower molecular weights through severe hydrolysis (Verardi et al., 2023). Although conventional steps such as demineralization and deproteinization may proceed faster, the sequential and aggressive treatments increase the risk of polymer degradation. DES extractions, while kinetically slower, benefit from integrated, one-pot operations that simplify processing and minimize solvent handling (Zhao, 2019).

Furthermore, DES systems yield chitosan with higher purity due to selective dissolution and minimal side reactions (Kimi and Hamdi, 2023; McReynolds et al., 2022), unlike HCl/NaOH methods, which can leave residual contaminants or induce unwanted reactions. Technological improvements, such as microwave-assisted DES extraction, promise to further reduce processing times without sacrificing the environmental benefits intrinsic to DES systems.

However, the scale-up of DES-based extraction faces thermal and mass transfer challenges at the pilot level. High DES viscosity impairs convective heat transfer and solute diffusion, requiring sophisticated thermal management and mass transfer enhancements (Vicente et al., 2021; Алексеев et al., 2022). Elevated operating temperatures can improve diffusion (Abdulbari and Basheer, 2019), but risk polymer degradation. Reactor design modifications and the integration of sonochemical or microchannel technologies have shown potential for improving thermal profiles and mass transfer in pilot-scale

Table 5. DES vs conventional cost comparison

Parameter	Conventional Method (HCl/NaOH)	DES-Based Method	References
Solvent Cost	Low (bulk acid/base); corrosive	Moderate (choline chloride and organic acids)	Paiva et al. (2014), Wang (2023)
Equipment	Requires corrosion-resistant materials (e.g., stainless steel)	Mild reaction conditions; compatible with glass/ceramic setups	Morais et al. (2020)
Energy Consumption	High (thermal treatment, intensive washing)	Lower (due to mild temperatures and fewer steps)	Coscarella et al. (2023)
Waste Management	High cost (toxic effluent, neutralization required)	Low (biodegradable, less hazardous, recyclable)	Abd El-Ghany et al. (2025), Vinci et al. (2023)
Solvent Recovery Feasibility	Not recyclable; high disposal cost	Up to 4–6 cycles with regeneration (e.g., filtration, rebalancing)	Isci and Kaltschmitt (2022), Jeong et al. (2015)
Estimated Operational Cost	Medium–High (due to waste handling)	Medium (depending on recovery and scale-up)	Nikolić et al. (2019), Siddiqui et al. (2025)
Scalability	Established but environmentally burdensome	Emerging, promising with optimization	Vicente et al. (2020), Vinci et al. (2023)

systems (Amaral et al., 2021; Liu et al., 2020; Prawang et al., 2019).

From an economic and environmental standpoint, life cycle assessments (LCA) integrating life cycle costing (LCC) consistently show that DES-based extraction systems offer reduced environmental impacts compared to conventional acid-base methods. DES extraction systems reduce emissions, toxic waste, and energy consumption due to milder reaction conditions (Serna-Vázquez et al., 2021; Vinci et al., 2023). While initial costs for DES preparation and solvent recovery infrastructure can be higher, estimated at 10–20% above traditional systems, these are frequently offset by significant savings in waste treatment, chemical handling, and energy input over time (Isci and Kaltschmitt, 2022; Nikolić et al., 2019). Comparative cost analyses indicate that conventional methods often incur higher downstream remediation costs due to corrosive waste management, whereas DES processes enable closed-loop reuse and safer handling. To enhance practical insight, Table X qualitatively compares economic parameters between DES-based and traditional extraction routes.

Nonetheless, economic competitiveness depends on factors such as solvent recyclability, process optimization, and capital investment in specialized equipment (Vinci et al., 2023). Future developments should focus on enhancing solvent recovery efficiency and refining scale-up designs to ensure that DES-based chitosan extraction can achieve both technical and economic viability.

5. CONCLUSION

Deep eutectic solvents (DESs) have emerged as a transformative medium for chitosan extraction, providing high extraction efficiency, superior product purity, and opportunities for solvent recycling. A natural DES composed of choline chloride and acetogenin demonstrated the extraction of chitosan from snail shells with a high degree of deacetylation (91%) and molecular weight (481 kDa), reflecting the selective and gentle nature of DES extraction processes compared to conventional chemical methods (Kimi and Hamdi, 2023; Li and Row, 2016). The strong hydrogen-bonding capabilities and tunable physicochemical properties of DESs facilitate disruption of chitin networks and enable efficient deacetylation and solubilization, thus preserving the structural and functional qualities of chitosan (Tang et al., 2015).

DES-based systems also exhibit low toxicity, biodegradability, and stability, enabling their reuse across multiple cycles with minimal performance loss (Li and Row, 2016). Their ability to be regenerated economically supports sustainable extraction processes with lower environmental footprints than traditional volatile organic solvents. Together, these findings position DESs as superior alternatives for producing high-purity chitosan applicable to biomedical, pharmaceutical, and industrial fields (Kimi and Hamdi, 2023; Tang et al., 2015).

Future perspectives in DES-based chitosan extraction increasingly involve integrating DESs with intensified extraction technologies such as ultrasound-assisted extraction (UAE) and microwave-assisted extraction (MAE). UAE enhances mass transfer via cavitational forces, creating localized high-pressure zones that disrupt chitin matrices and promote DES penetration (Coscarella et al., 2023; Durante-Salmerón et al., 2024; Wang, 2023). MAE, by contrast, utilizes rapid, uniform heating through dipolar rotation and ionic conduction mechanisms, improving solvent–matrix interactions and boosting extraction rates while maintaining chitosan's structural integrity (Hao et al., 2021). These synergies reduce processing time, solvent consumption, and energy input, fully aligning with the principles of green chemistry (Durante-Salmerón et al., 2024; Hao et al., 2021).

Process optimization approaches such as response surface methodology have demonstrated the feasibility of fine-tuning extraction parameters, including solvent composition, ultrasound/microwave intensity, and extraction duration, to maximize recovery efficiency and minimize environmental impact (Coscarella et al., 2023; Wang, 2023).

Development of biomass-derived DESs represents another critical advancement. Derived from renewable raw materials such as lignin, carbohydrates, and agricultural residues, these DESs embody circular economy principles by valorizing waste streams (Kim et al., 2020; Onwucha et al., 2023). Their low vapor pressure, tunability, and favorable environmental characteristics, combined with their ability to close the production loop, render them highly attractive for sustainable industrial applications (Satlewal et al., 2018; Tang et al., 2017; Wang, 2023). Strategic molecular design of biomass-based DESs can significantly enhance pretreatment and solubilization efficiencies (Ryu et al., 2024), while their application in recycling spent materials, such as cathode active components from lithium batteries, further highlights their potential (Morina et al., 2021).

Ecotoxicity and biodegradability studies affirm that biomass-derived DESs generally outperform traditional organic solvents in terms of environmental safety, supporting the transition to regenerative and closed-loop production models (García et al., 2015; Srivastava, 2020). Comprehensive life cycle and toxicity assessments are vital to ensure that the benefits of DESs are fully realized in sustainable industrial contexts (Hariyanto et al., 2023).

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