EXPLORING THE POTENTIAL OF DEEP EUTECTIC SOLVENTS FOR SUSTAINABLE TEXTILE TECHNOLOGIES

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Deep eutectic solvents (DESs) have garnered significant attention from researchers due to their sustainability and tunable properties, offering advantages over conventional organic solvents. DESs are considered non-toxic, less volatile, environmentally benign, and with versatile functionalities, which makes them ideal for modern textile processing. This review aims to provide a state-of-the-art overview focused on the recent advances in the implementation of DESs for different textile applications, such as green approaches for dyestuff synthesis, surface modification of natural and synthetic fibers, and waterless dyeing technologies. The first part of this review focuses on the DES components and intermolecular interactions during the synthesis of DESs. The subsequent section reported recent studies employing DESs for surface activation of natural (cellulosic, protein, and mineral fibers) to achieve special functional properties, such as better tensile strength, hydrophilicity, and dye uptake. In addition, this review also highlights some emerging applications of DESs for auxiliaries during melt spinning of synthetic fibers and subsequent modification before coloration. Some reports about DES-based green synthesis routes for dyestuffs followed by technologies to develop water-reduced dyeing processes for natural and synthetic textiles. Improved resource efficiency, darker shades, and accelerated diffusion kinetics are noted relative to conventional dyeing attributable to DES-fibre interactions for various pigment classes. This paper also provides some insights about future sustainability, economic, and regulatory recommendations toward expanding DESs implementation for responsible textile production globally.

Keywords: deep eutectic solvents (DESs), green synthesis, surface modifications, textile

INTRODUCTION

Textiles constitute one of the major manufacturing industries worldwide, with a global market size projected to reach \$1.23 trillion by 2025.[1] Asia dominates textile production, exporting nearly USD 670 billion (Table 1) and employing over 45 million workers as of 2019 (Italy Textile Industry Association, 2020). Since enormous quantities of materials are processed and dyed annually, textile production has a significant environmental footprint through extreme water consumption, elevated energy burdens of heat or pressure-using processes, and effluent release with poisonous chemicals or dye in wet processing.[2,3,4] The industry is thus incentivized to explore alternative technologies enabling greener production.

Deep eutectic solvents are a novel class of ionic solvents that have recently emerged as promising designer solvents that exhibit a unique range of environmentally benign characteristics, setting them apart from conventional volatile organic compounds (VOCs) employed in the textile sector.[3,4] First reported in 2003, DESs are typically formed by the complexation of quaternary ammonium salts, usually choline chloride, with hydrogen bond donors, such as amines, carboxylic acids, alcohols, or amides.[5] Although their precursors have high melting points, when combined in certain molar ratios, much lower melting points are attained to form liquids possessing favorable solvent properties, such as excellent dissolution capacity for a range of textile dyes, polymers, bioactive compounds, and other materials at mild temperatures.[6-8] Their strong hydrogen bonding interactions allow tuning solvent physicochemical characteristics like viscosity, conductivity, and hydrophobicity based on the DESs components to suit different functions.[9,10]

This review provides an investigation into various facets underpinning the green credentials of DESs in relation to conventional solvents. The composition and preparation methods of DESs are outlined at first. Their advantageous physicochemical properties and other factors determining their inherent eco-friendliness, like negligible volatility, non-flammability, high biodegradability, low toxicity, availability from renewable resources, and recyclability, are discussed.[3,31] These merits render DESs suitable for numerous applications across diverse domains, including organic synthesis, nanomaterial fabrication, extraction, electrochemistry, biomass processing, and more.[21,30,32,33] Recent studies reporting on the use of DESs for modifying and dying of various natural and synthetic fibers are reviewed. Finally, future research directions and recommendations to further expand the scope of DESs are suggested.

Table 1
Statistics of the global textile market (data from Statista 2024)

Category	Statistic data
Projected global textiles market size in 2025	\$1.23 trillion
Asia's textiles exports in 2019	Nearly \$670 billion
Asia's textiles industry employment in 2019	Over 45 million workers

Table 2
Classification of DESs

Type	General formula	Terms	Ref.
Type I	$Cat^+X^-zMCl_x$	M = Zn, Sn, Fe, Ga, In	11-16
Type II	$Cat^+X^-zMCl_x \cdot yH_2O$	M = Cr, Co, Cu, Ni, Fe	11
Type III	Cat^+X^-zRZ	Z = $CONH_2$, COOH, OH	11
Type IV	$MCl_x + RZ = MCl_{x-1}{}^+ \cdot RZ + MCl_{x+1}{}^-$	M = Al, Zn and Z = ONH_2, OH	17

COMPOSITION AND CLASSIFICATION OF DESs

Deep eutectic solvents often follow the compositional structure:[3] Cat^+X^-zY, where Cat^+ can be any ammonium, phosphonium, or sulfonium cation, while X represents a Lewis base, typically a halide anion. Complex anionic species form through interactions between X^- and a Lewis or Brønsted acid Y, where z indicates the number of Y molecules interacting with the anion. Most research has concentrated on quaternary ammonium and imidazolium cations, particularly concerning more practical systems involving choline chloride, [ChCl, $HOC_2H_4N^+(CH_3)_3Cl^-$].[3]

DESs comprise mixtures of inexpensive, readily available components with high hydrogen bond donor (HBD) and hydrogen bond acceptor (HBA) capacities. Typical HBAs include quaternary ammonium salts, such as choline chloride, whereas common HBDs feature metabolites, organic acids, glycols, amides, or amines. Natural amino acids, like proline, as well as their derivatives, also form DESs upon mixing with salts/acids. Metallic salts/hydrates of Zn, Sn, Fe, Al, Cu, *etc*. also act as Lewis acid HBAs. The components are mixed in appropriate molar ratios under atmospheric pressure at elevated temperatures between 60–100 °C or by microwave/ultrasound until a transparent, homogeneous liquid DES with a significantly lowered melting point compared to the starting precursors appears. Based on constituents, some DESs are hydrophobic, whereas most are hydrophilic, absorbing moisture from surroundings.[3]

DESs can be broadly categorized based on the complexing agent employed, as outlined in Table 2. Type I DESs, composed of MClx and quaternary ammonium salts, share similarities with the extensively studied metal halide/imidazolium salt combinations. Representative examples of type I eutectics include the well-known chloroaluminate/imidazolium salt mixtures and the less familiar ionic liquids comprising imidazolium salts and diverse metal halide, such as $FeCl_2$.[11-16] While the variety of non-hydrated metal halides capable of forming type I DESs within an acceptable melting point range is restricted, the spectrum of deep eutectic solvents can be broadened by employing hydrated metal halides and choline chloride, leading to type II DESs. Type III eutectics, consisting of choline chloride and hydrogen bond donors, have attracted interest due to their ability to solvate a broad spectrum of transition metal species, including chlorides and oxide.[11] Abbott *et al.* demonstrated that various transition metals can be integrated into ambient temperature eutectics and classified as type IV DESs. Typically, these metal salts would not ionize in non-aqueous environments; however, $ZnCl_2$ has been found to form eutectics with compounds such as urea, acetamide, ethylene glycol, and 1,6-hexanediol.[17]

ADVANTAGES OF DESs OVER CONVENTIONAL SOLVENTS

Unlike volatile organic solvents traditionally dominating textile chemistry, DESs offer an array of environmentally prudent, practical, and economic advantages that motivate their consideration as auxiliaries or replacements for conventional solvent-intensive dyeing, bleaching, scouring, printing, and finishing processes.[18,19]

Negligible volatility and flammability

Unlike volatile organic compounds widely used as conventional green solvents, like alcohols, ethers, or esters, most DESs exhibit no measurable vapor pressure since the strong intermolecular interactions between the HBA and HBD prevent evaporation. This drastically minimizes air pollution through fugitive emissions and exposure risks during handling/storage/transportation.[20,34-36]

The high flash points of DESs averaging over 100 °C also impart flame retardant behavior, providing safer operational parameters.[21,22] Most DESs display wide liquid temperature ranges up to 300 °C, and thermal stability limits around 200–400 °C before decomposition, being suitable for high-temperature applications. DESs composed of choline chloride and urea have been found to possess excellent oxidation stability, retaining structural integrity on exposure up to 4000 °C over 20 hours. Additionally, the high water-retaining ability in DESs renders them non-combustible. These non-flammable, non-volatile attributes coupled with thermal endurance enhance operational safety and environmental compatibility over conventional media.[36]

Waste valorization and energy/cost savings

A major motivation spurring DES research is the reliance on using inexpensive, non-toxic raw materials from renewable biosources, instead of petrochemical feedstocks for their synthesis. Common HBD include abundant metabolic intermediates like organic acids, carbohydrates, glycerol, amino acids/derivatives, and biomolecules. Similarly, the quaternary ammonium HBA employed are generally bio-based. Thus, industrial biomass wastes like sugarcane bagasse and beet pulp serve as sources for DES components, enabling valorization of biomass waste.[29] Such reliance on renewable ingredients over finite crude oil resources for fabrication lends sustainability to DESs compared to traditional volatile solvents.[30,31,45-47] Thus, DESs production/usage can promote a circular economy by waste valorization.

By virtue of being liquid at room temperature, without requiring additional energy for vaporization as needed for VOCs, DESs synthesis requires less energy input than typical solvents.[7,27,28] Their synthesis procedures are energetically more favorable than multi-step reactions necessitated for other neoteric solvents like ionic liquids. Overall, DESs possess lower embodied energy than existing solvents.[48] By selecting less energy-intensive components with available bio-replacements, the environmental footprint of DESs can be continually reduced to align with green chemistry goals.[30] The life cycle assessment of choline chloride-based DESs production evidenced a cumulative energy demand of 30.7 MJ/kg, far below that of other neoteric solvents like dimethyl sulfoxide (DMSO) and [BMIM]PF6 (263 and 885 MJ/kg), conferring energy savings.

Low toxicity

With judicious selection of GRAS status ammonium salts and biocompatible hydrogen donors like organic acids or polyols, most DESs display remarkably low toxicity levels, compared to industrial solvents,[23,24] implying they can serve as less hazardous replacements. Acute toxicity studies revealed EC50 values for typical DESs exceeded 100 g/L, while for ethanolic solutions employed in textile finishing, toxicity thresholds ranged from 0.1-30 g/L.[25]

Preliminary animal toxicity testing revealed the high LD50 oral toxicity values of >2000–5000 mg/kg body weight for choline chloride or glycerol-based DESs, consistent with their non-hazardous nature and significantly safer profiles compared to organic solvents like acetone or acetonitrile. Aquatic ecotoxicity assessments through *Daphnia magna* and *Vibrio fischeri* microbiomes also confirm low LC50 toxicity levels of >200 mg/L.[42] Furthermore, phytotoxicity analysis indicates high seed germination percentages, revealing biocompatibility with plant metabolism. Easy biodegradation of DESs prevents toxin accumulation in organisms. Taken together, these studies verify the low toxicity credentials of DESs as alternatives to hazardous conventional media.[42-44]

Assessment of model plant *Lactuca sativa* through germination efficiency, root/shoot length, and phytotoxicity index parameters indicated no significant adverse influence on metabolism at DES concentrations <2.5% v/v, consistent with suitability for sustainable agriculture applications. Sea-urchin embryo and polychaete worm species lifecycle analyses also confirmed the non-teratogenic, non-mutagenic effects, supporting the benign environmental profile of DESs as alternatives over hazardous solvents like dimethyl sulfoxide (DMSO) or dimethylformamide (DMF) traditionally used for pharmaceutical/cosmetic ingredients processing.[53-60]

Biodegradability

Since most DESs are fabricated from bio-based precursors, they display high susceptibility to biodegradation by microorganisms and enzymes. Thus, the metabolizable hydrogen bond donors, like glycerol, urea, and lactic acid, combined with choline chloride salts, enable complete biodegradability of most DESs by microorganisms.[26] For instance, Leron *et al.* reported 90% biodegradation for choline chloride: oxalic acid DESs within 5 days, monitored through carbon dioxide evolution during closed bottle tests, consistent with facile bioassimilation.[37] Comparable biodegradation of glyceline DESs was shown by Juneidi *et al*. using *Pseudomonas* bacteria.[38,39]

Ready breakdown of spent DESs enables sustainable waste discharges without persistent pollution.[21] Conversely, traditional aprotic solvents like dichloromethane, chloroform, pyridine, and DMF exhibit resistance against degradation. Easy biodegradability and the non-bioaccumulative nature of common small molecular DESs constituents make them eco-compatible.[40,41]

Various ecotoxicity studies for risk assessment have confirmed the green nature of DESs. Microtox bioassay toxicity testing on *Aliivibrio fischeri* bacteria reveals inhibition levels comparable to benign solvents like acetone/butanol, in contrast to the highly toxic impacts of traditional organics. DESs protonation enhances permeability of bacterial cell membranes, but does not lead to rupture or lysis.

Customization and recyclability

Owing to the availability of numerous hydrogen bond donors/acceptors, DESs with varied properties can be designed for targeted applications by screening alternative components, adjusting ratios, and using different mixing protocols. This versatility allows their implementation across diverse domains, as summarized in later sections. Furthermore, unreacted raw materials from initial DES preparation can be efficiently recovered and reused through ethyl acetate or acetonitrile extraction, followed by solvent evaporation. This leads to significant cost savings by minimizing waste, alongside enabling DESs recyclability.[49] Ion exchange resin treatments also offer good separation efficiency for HBA and HBD recovery. 80-90% yields are achievable for DESs regeneration from post-usage streams. Component reuse coupled with non-energy intensive, solvent-free synthesis further adds to the economic viability of DESs.[50-52]

The advantages of DES over conventional solvents are summarised in Table 3.

Applications as green solvents

The environmentally benign nature, low material costs, and tunable solvent properties have rendered DESs promising candidates for diverse applications across a myriad of domains as outlined in Figure 1 and further discussed below.

Organic synthesis medium: The excellent solubilization capacity, coupled with metal leaching ability, has enabled DESs to be used for efficient synthesis of fine chemicals, APIs, alkaloids, aroma compounds, *etc*. under benign conditions. DESs also offer stabilization for various nanoparticles.[61-64]

Biomass processing: DESs pretreatment facilitates selective fractionation of lignocellulose into cellulose fibers for bioethanol generation owing to the lignin/hemicellulose solubility. Chitin isolation from crustacean waste via DESs treatment also holds promise.[65-67]

Extraction solvents: Hydrophobic DESs variants allow efficient extraction of bioactive natural compounds from plants, residues, *etc*. DES-extracted compounds have been commercialized by companies like Ingredia®.[68-71]

Table 3
Key attributes of DESs in comparison with conventional solvents

Feature	Benefits of DESs over conventional solvents
Biodegradability and low toxicity	DESs are often made from bio-based and non-toxic components, like sugars, alcohols, organic acids, *etc*., making them much safer than conventional organic solvents.
Design flexibility	DESs can be designed and tailored to have specific physicochemical properties by selecting appropriate hydrogen bond donors and acceptors.
Simple preparation	Most DESs are easy to prepare just by mixing and heating two safe and inexpensive components together, without the need for further purification.
Non-volatility	Unlike many standard solvents, DESs have negligible vapor pressure, so they can be used safely, without exposure risks.
Thermal stability	Many DESs are liquid across a wide temperature range from below 0 °C up to close to 200 °C, allowing use under a variety of conditions.
Solvation capacity	The tunability of DESs allows optimization for dissolving a huge range of compounds from polar to non-polar molecules.
Catalytic ability	Some DESs components can act as catalysts themselves, speeding up reactions, while DESs are also useful solvents for transition metal or enzyme catalysis.

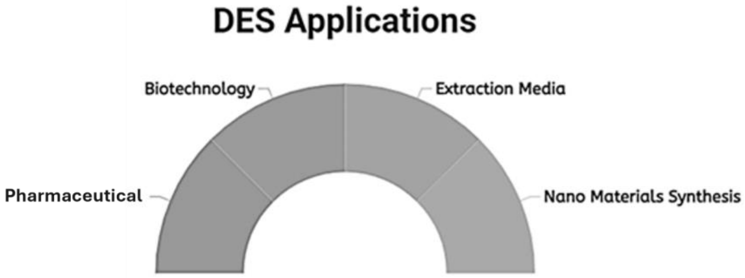

Figure 1: Applications of DESs in various fields

Absorption media: The affinity, large surface area, and recyclability have enabled DESs usage for selective capture of rare earth metals or radioactive isotopes from waste streams. CO_2 absorption for biogas upgrading has also been demonstrated.[72-74]

Electrolytes for device fabrication: Negligible volatility makes DESs useful as conductive media for the fabrication of solar cells/batteries through electrodeposition. Integration in membrane separation processes is also being explored.[3,75,76]

Enhanced oil recovery: Oil-DES emulsions allow mobilization of thicker crude deposits while bio-DESs aid microbial enhanced recovery. This provides more sustainable alternatives over surfactants/polymers.[77-79] The application of DESs has achieved advanced implementation in some applications. For instance, the viability of scaling up DES-mediated extractions to commercial levels has been established through detailed economic analysis by Prototype at Scale®, which takes into consideration all raw material/utility/operating costs. However, most applications still remain at the laboratory/bench demonstrative scale. Further techno-economic studies assessing feasibility are imperative before successful market entry of DES-based processes.

APPLICATIONS IN TEXTILE PROCESSING AND MODIFICATION

The versatile solvent capabilities and green characteristics have motivated rising research interest in employing DESs for textile applications like fiber surface modification, functional finishing of fabrics, microencapsulation of textile care actives, catalysis, and polymer processing, as well as dyeing operations.[80-82] DES treatment conferred durable wrinkle-free, antimicrobial, flame retardant characteristics and other value additions to various fabrics.[83,84] Greener options help meet stringent chemical regulations.

Facilitating dyeing, bleaching and scouring

Implementation of choline chloride-based DESs as auxiliary dyeing media for cotton, silk, wool, and polyester fabrics enabled higher color strength and penetration at lower temperatures, replacing high boiling point toxic solvents like DMF, due to the ability of DESs to solubilize diverse dyes. The infiltration of dyes into textile fibres and fabrics is complex, governed by the chemical properties of both the dye and fibre as well as external conditions. The process of dye permeation into fabrics is shown in Figure 2. For example, in cotton, DESs enhance fibre swelling, assisting dye diffusion. Pretreating wool with DESs afforded cleaner bleaching. Combining enzymes or ultrasound with DESs in a dyeing procedure has been examined to reduce the energy demands.[85-88] The impressive early-stage results motivate further translation of DESs implementation to an industrial scale.

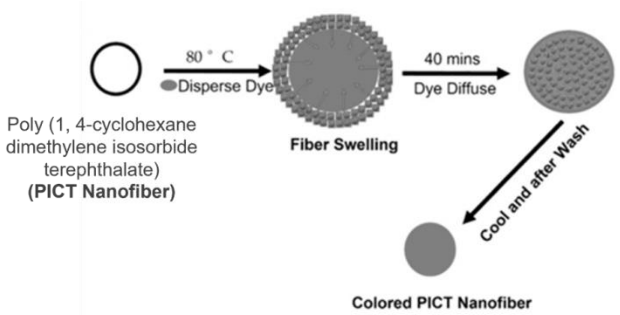

Figure 2: Process of dye permeation into fabrics[87]

Natural fiber modification by DESs

The textile industry relies extensively on the use of toxic chemicals and vast volumes of water for fabric processing, leading to negative environmental impacts. Implementing benign alternatives for textile manufacturing has thereby gained enormous attention globally. In this context, DESs have emerged as promising green media for eco-friendly modification of textile fibers and fabrics.[11,] This section provides comprehensive insights into the recent advances concerning DES-based pretreatment of various major natural textile substrates, including cellulosic, proteinous, and mineral fibers to impart useful functional attributes.[18,86]

Various treatment processes using DESs for altering natural fibers are discussed discussed in the literature, with the purpose of obtaining improved surface morphology, mechanical strength, thermal stability, hydrophilicity, dye absorbency, and other favorable properties. Pretreatment by DESs has been reported on natural fibers, such as cotton, linen, hemp, jute, bamboo rayon, as well as protein-based wool and silk, and asbestos mineral fibers.[89]

The principle of enhancing bamboo fiber properties by treatment with a reactive deep eutectic solvent is presented in Figure 3. The inherent rigidity of bamboo fibers limits their textile applications, making modification essential. To address this problem, bamboo fibers were treated with a deep eutectic solvent (ChCl-urea) to enhance spinnability and impart antibacterial properties. The treatment partially removed hemicelluloses and lignin, with dewaxed fibers treated at 105 °C showing the highest degree of substitution (DS = 0.12). The treated bamboo fibers exhibited reduced linear density (14.29 tex), lower strength (1.76 cN/dtex), and strong antibacterial activity against *E. coli* and *S. aureus*, with bacteriostatic rates of 75.5% and 72.7%, respectively. The authors concluded that such a treatment can be a sustainable route for achieving value-added bamboo textiles.[89]

Cellulose fiber modification

Recent efforts have targeted cotton fabric modification using choline chloride-based DESs to enhance hydrophilicity, adhesion, and dye uptake capacity by altering the surface morphology and chemistry of the fiber for improved wettability.[90] The influence of temperature and other parameters on the solubility of cellulose and non-cellulosic components in various DESs is presented in Tables 4 and 5. In the case of the oxalic acid/ChCl eutectic mixture, it initially forms internal hydrogen bonds, leaving abundant free ions from ChCl to subsequently bond with cellulose hydroxyl groups and facilitate dissolution. Regenerated cellulose was generated with altered crystalline properties (type II) compared to the original cellulose (type I), despite structural similarities.

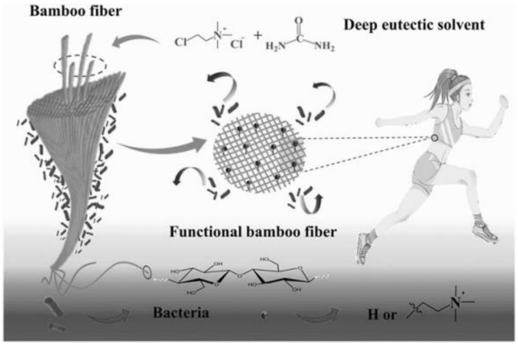

Figure 3: Enhancing bamboo fiber properties with a reactive deep eutectic solvent[89]

Table 4
Influence of temperature on cellulose solubility in oxalic acid and chlorocholine deep eutectic solvent[91]

Dissolution temperature (°C)	60	70	80	90	100
Solubility (%)	0.50	0.64	0.82	1.96	2.54

Table 5
Dissolution of lignin and cellulose in deep eutectic solvents[91]

Hydrogen bond donor (HBD)	Hydrogen bond acceptor (HBA)	Ratio	T (°C)	Lignin (wt%)	Cellulose (wt%)	Hemicelluloses (wt%)
Lactic acid	Proline	3.3:1	60	9	<1	<1
Lactic acid	Proline	2:1	60	7.56	0	-
Lactic acid	Choline chloride	2:1	60	5.38	0.00	-
Lactic acid	Choline chloride	10:1	60	13	<3	<5
Lactic acid	Choline chloride	5:1	60	7.77	0	-
Lactic acid	Choline chloride	10:1	60	11.82	0.13	-
Lactic acid	Glycine	9:1	60	8.77	0.00	-
Lactic acid	Alanine	9:1	60	8.47	0.00	-
Lactic acid	Betaine	2:1	60	9	<1	<1
Lactic acid	Betaine	2:1	60	12.03	0	-
Lactic acid	Glycine	9:1	60	11.88	0.13	-
Formic acid	Choline chloride	2:1	60	14	<1	<1
Acetic acid	Choline chloride	2:1	60	12	<1	<1
Malic acid	Proline	1:3	60	14.90	5.90	-
Oxalic acid dihydrate	Choline chloride	1:1	60	3.62	2.5	-
Urea	Choline chloride	2:1	110	-	<0.2	-
Zinc chloride	Choline chloride	2:1	110	-	<0.2	-
Urea	Choline chloride	2:1	110	-	1.43	-
Imidazole	Choline chloride	7:3	110	-	2.48	-
Ammonium thiocyanate	Choline chloride	1:1	110	-	0.85	-
Caprolactum	Choline chloride	1:1	110	-	0.16	-
Acetamide	Choline chloride	2:1	110	-	0.22	-
Oxalic acid	Allyl triethyl ammonium	1:1	110	-	6.48	-

Paparella *et al.* discussed a surface etching technique by establishing an optimal ChCl-glycerol DES and 80 °C operating temperature for achiving augmented cotton wettability.[92] The study also confirmed the conversion of crystalline cellulose I into amorphous cellulose II polymorphs, alongside disrupted inter-chain hydrogen bonding from DES interaction, facilitating accessibility for matrices like polyacrylic acid to cling, leading to around 30% enhancement in dye bath exhaustion compared to untreated samples.[93-95] In other work, El-Sayed *et al.* prepared a novel bio-based DES by complexing phytic acid with choline chloride for flame-retardant finishing of cotton fabrics using the pad-dry technique.[96] SEM imaging displayed rougher, etched surfaces with microparticles deposited from DES, contributing to improved char formation. Vertical flammability testing revealed significantly reduced afterglow and after-flame times, coupled with 30% lowered heat release rates, consistent with synergistic flame retardation between phosphorus and nitrogen-based DES constituents through catalytic charring mechanisms. Thereby, minimal strength loss was accompanied by durable flame retardancy even after repeated laundering, illustrating the promise of bio-derived DESs for eco-friendly cotton functionalization.[97-98]

Wool modification

Wool possesses skin-comforting characteristics, owing to its hygroscopic moisture buffering and regulation capacity. However, felting and shrinkage during washing arise from scales on fiber surfaces interlocking. Hence, anti-felting treatment is needed.[99] Utilizing benign DES alternatives over harsh chlorination for wool finishing can thereby produce eco-friendly anti-felt fabric.[100,101] SEM images (Fig. 4) demonstrate that DES treatment can solubilize the cuticle layer on the surface of wool fibers, providing evidence of the strong solvent capabilities of this DES towards wool.[99]

Also, Jiang *et al*. demonstrated that DES treatment imparted anti-felting behavior to wool by etching cuticle cells to reduce scales.[102] This led to the maintenance of the fabric's mechanical properties, while also causing around 30% enhancement in dye uptake. No significant strength losses occurred even after 5 wash cycles, consistent with durability requirements. The removal of scales along the fiber can be explained through the possible protonation of keratin by DES acidic components. Additionally, to impart shape memory properties, Jiang *et al*. leveraged ternary DES incorporating urea, thiourea, and choline chloride, followed by curing with citric acid crosslinker, for partial wool keratin denaturation.[102] Subsequent FTIR and DSC studies confirmed the formation of stable ester crosslinks between citric acid and hydroxyls in DES-treated wool, consistent with 92% fabric recovery from temporary shaping. Thereby, DES pretreatment can facilitate achiving desirable functional attributes, like anti-felting or shape memory, without using harsh chemicals.[103]

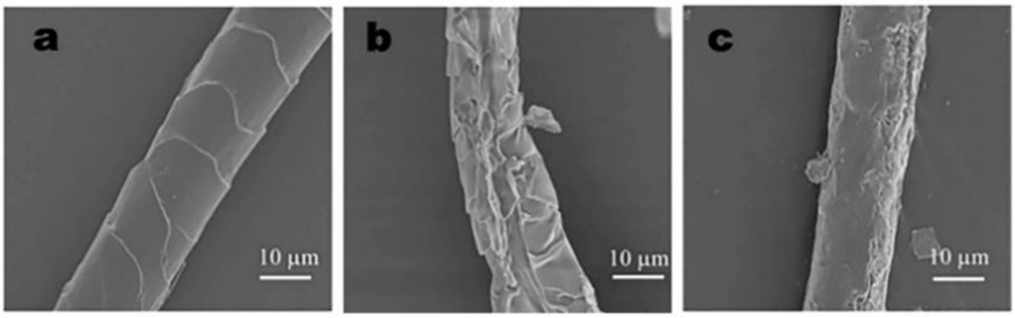

Figure 4: SEM images showing the dissolution of the surface of wool fibers by solubilizing the cuticle layer with DES treatment [99]

Silk modification

Silk fibroin derived from *Bombyx mori* silkworm cocoons also offers promising textile substrates, due to its remarkable tensile strength, moisture absorption, and oxygen permeability. Although properties vary significantly across silk varieties, treatments using DESs effects can optimize functionality.[104,105] For instance, Grala *et al*. reported that chitosan-glycerol DES treatment of muga silk for 30 minutes beneficially increased thermal stability by ~28 °C, possibly owing to molecular crosslinking.[106] Concurrently, ~3.7-fold moisture regain enhancement was achieved, attributable to glycerol plasticization. Morphological and structural analysis using SEM and FTIR also revealed progression from beta-sheet crystals toward amorphous secondary conformations. In a different study, a choline chloride-thiourea DES system was applied for the extraction and purification of fibroin from eri silk via dissolution-coagulation approaches instead of traditional degumming procedures.[107] This offered significantly higher (~96%) protein recovery yield and preservation of essential amino acids, such as glycine, alanine, and serine, facilitating subsequent material fabrication.

Deep eutectic solvents (DESs) have also been used as swelling agents to enhance the color strength of silk fabric during dyeing. In solvent-assisted dyeing, the DES causes the silk fibers to swell, enabling greater diffusion and deeper penetration of dye molecules (Fig. 5). The incorporation of a DES into the dyeing process significantly increased the color saturation of dyed silk fabrics, compared to controls. This demonstrates the potential for DESs to improve the environmental sustainability of textile dyeing by reducing water and energy consumption while brightening colors. Further confirmation in pilot and industrial systems is necessary.[108] Furthermore, the viscosity grade was retained between 3.5-4 centipoises, which can be feasible for rheology-optimized wet-spinning applications.[20,109,110] Thereby, DESs treatment provides a benign, biomimetic alternative for customized dyeing processes for silk fabrics.

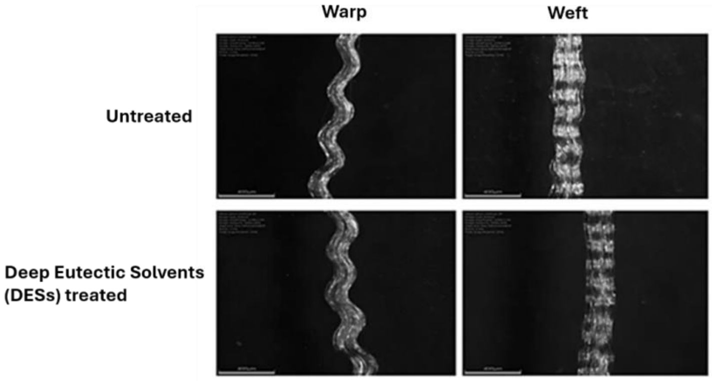

Figure 5: Swelling of silk fibers treated with deep eutectic solvents[108]

Synthetic textile fibers/fabric modification

Synthetic fibers and fabrics, such as polyesters, polyamides, acrylics, spandex, and polyolefins, dominate the global textile feedstock, owing to their versatile properties and cost-competitiveness. However, issues like heat/chemical resistance, poor dye uptake, or mechanical durability for advanced applications necessitate various modification strategies.[111,112] Conventional processing relies on toxic solvents, like dichloromethane or complex reactions. Hence, implementing green techniques is imperative. In this context, deep eutectic solvents (DESs) have attracted enormous interest as sustainable media for synthetic textile modification to improve various performance attributes through structural rearrangement, plasticization, and crosslinking.[81,113-114] Some perspectives on recent advances in DES-enabled modification of major synthetic textile materials are outlined below.

Polyester modification

Polyester and its various co-polymer derivatives, especially polyethylene terephthalate (PET), find widespread clothing applications owing to the dimensional stability, durability, and hydrophobicity of such fibers.[115] However, lack of functional groups results in poor dye uptake, therefore, fiber modification is required. Utilizing DESs for this can minimize the environmental impact of such processes, compared to conventional approaches. A complete process of PET modification is presented in Figure 6.

Poly(ethylene 2,5-furandicarboxylic acid) (PEF) is an emerging bio-based polyester polymer that can be synthesized from fructose and glucose. PEF was subjected to glycolysis by heating at 180 °C for 1 hour at atmospheric pressure. This process breaks down the PEF polymer chains, using ethylene glycol, resulting in bis(hydroxyethyl 2,5-furandicarboxylate) (BHEFDC) as the main product. With favorable thermal and mechanical properties, PEF has potential to replace conventional petroleum-derived polyethylene terephthalate (PET) for applications ranging from packaging to textiles. Transitioning to a circular economic model drives motivation for chemically recycling plastics like PEF into regenerated PEF (rPEF) after use.

Recently, S. Wu *et al*. developed an innovative closed-loop recycling approach for PEF employing a biodegradable and safe deep eutectic solvent (DES) under mild conditions. The DES enabled both depolymerization of PEF into monomers and subsequent repolymerization into regenerated PEF, with maintained performance.[116] Specifically, a eutectic mixture of urea and zinc acetate catalyzed the breakdown and reformation of the polyester bonds. This dual catalytic ability highlights the promise of DES systems for sustainable chemical recycling of bio-based polymers. Further confirmation using post-consumer PEF at pilot to industrial scales will better demonstrate feasibility. Overall, integrating green chemistry principles into recycling processes will enable continued progress toward a carbon-neutral circular economy.

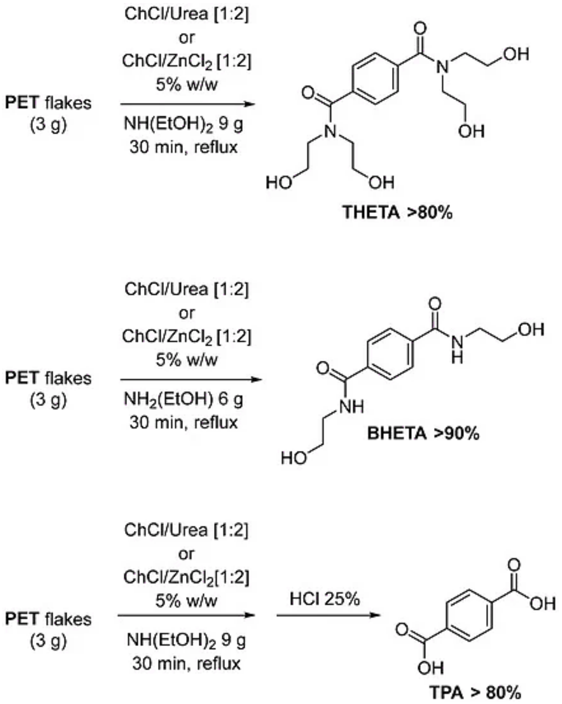

Figure 6: Aminolysis of PET into terephthalic acid (TPA), bis(2-hydroxyethyl)terephthalic acid (BHET) and tetra-hydroxyethylterephthalamide (THETA) using deep eutectic solvents[88]

Deep eutectic solvents in textile dyeing

Owing to high dye solubility potential, DESs assist the development of eco-friendly dyeing methods for textiles. The most common approach involves pre-dissolving hydrophilic dyes in choline chloride-based DESs with urea or glycerol as HBD, followed by dyebath preparation and conventional exhausting on cellulosic/protein substrates.[117,118] Other methods, such as ultrasonication, and microwave heating, have also been tested for rapid dyeing. Key interactions stabilizing dyes include electrostatic forces, π-π stacking, hydrogen bonding, and van der Waals forces with DES constituents forming organized assemblies. DES integration enhances dye wetting, swelling, and diffusion kinetics, yielding darker shades compared to conventional dyeing.[123] The low vapor pressure enables high temperature operation above 100 °C, further assisting dye diffusion into fibers.[120]

For disperse dyes applicable to synthetics, like polyester, smaller amounts of DESs are utilized to assist dispersion. Vat dyes can also be solubilized by bio-DESs, which reduce the use of harmful sodium dithionite. Research has focused on achieving optimal DES molar ratios, dyeing process parameters, and component selections for varied textiles to obtain resource efficiency alongside color fastness using eco-benign DESs as dyeing auxiliaries overcoming limitations of aqueous systems.

Also, multiple efforts have been reported for integrating DESs within existing methods for dyeing cellulosic fibers, like cotton, viscose rayon and lyocell, using direct, reactive, and natural pigments to develop sustainable water-saving alternatives.[120] Over 86% reduction in water consumption, alongside around 30% higher color strength was achieved by using 25-75% v/v ChCl:urea DES solutions containing pre-dissolved reactive dyes compared to traditional exhaust dyeing.[117,118] Benefits included accelerated dye diffusion, attributable to DES swelling causing enhanced dye uptake.[121] In other works, ultrasonic energy was adopted for intensifying DES-assisted direct dyeing on cotton, achieving 90% wetting and penetration within 5 minutes, at 20:1 liquor ratios using ChCl:glycerol DESs, consistent with eco-friendly continuous possibilities.[18,122-123]

The dyeing potential of a crude prodigiosin gel, produced by non-pathogenic *Serratia plymuthica* bacteria, has been tested on various fabrics under different temperature, pH, salt, and mordant conditions. In this context, a novel choline chloride/lactic acid deep eutectic solvent (DES) was examined as a natural dyeing medium to replace organic solvents. Nylon fabrics dyed with the bio-mordant L-cysteine at pH 8.3 showed improved color fastness to washing. The gel-based ChCl/LA DES dyebath provided enhanced K/S values (presented in Table 6). Under these conditions, nylon fabrics displayed significant antimicrobial activity against *Staphylococcus aureus* and *Pseudomonas* *aeruginosa*. The authors concluded that using the crude gel-based prodigiosin dye has advantages for eco-friendly textile dyeing. The natural reducing agent L-cysteine, untested before as a bio-mordant, along with the gel-based DES dyeing medium, improved color and antibacterial properties of nylon dyed with crude gel prodigiosin.[117]

Table 6
Multifiber fabrics dyed with prodigiosin using a gel dyeing method with DES

Gel-based deep eutectic solvent (DES)		
ChCl/LA (1:2)		
Acetate	*K/S*	2.76
	dE	0.02
Cotton	*K/S*	1.60
	dE	0.02
Nylon	*K/S*	3.33
	dE	0.01
Polyester	*K/S*	1.22
	dE	0.01
Acrylic	*K/S*	1.55
	dE	0.01
Wool	*K/S*	1.60
	dE	0.01

Furthermore, bio-based DESs fabricated from amino acid salts have been effectively applied for extraction and dyeing with natural colorants, as shown by Muhammad *et al*. Deep shades having good wash and light fastness were obtained on cotton nanofibers using DESs based on methionine and proline, owing to high solubilization capacity for hydrophobic bio-colorants like annatto, and curcumin, compared to conventional methods. Thereby, DES integration into cellulosic dyeing processes enables cleaner prospects for the dyeing procedure, with promising results.[124]

As regards wool fibers, their surface features and protein-based keratin structure make them responsive to specialty DESs for achieving customized dyeing effects.[121,124] For instance, Pawar *et al*. adopted ChCl:urea and ChCl-oxalic acid DESs to dye wool and alpaca fibers using acid-milling blue dyes.[123,125] Enhanced shades, with uniform dyeing results, arising from even diffusion into wool scales, were attained at a 98% reduction in water usage and 70% lower auxiliaries consumption whn using 30% v/v DES solutions. Stripping analysis revealed higher dye-fiber bonds and depth of shade for the DES method, consistent with possible interactions between acidic DES components and wool slowing desorption.[124,125]

CONCLUSIONS AND FUTURE RECOMMENDATIONS

The review provides an overview of the advances in DES implementation for various textile applications, ranging from structural modification of natural and synthetic fibers to clean synthesis approaches for dyes and waterless dyeing technologies.

As discussed, DESs can be employed for surface activation of cellulosic, proteinous, and mineral fibers, in order to improve their characteristics, such as tensile strength, hydrophilicity, and dye uptake, without compromising integrity and thermal stability. The use of DESs induced structural rearrangements and plasticization mechanisms, which were responsible for such customized enhancements. Emerging DES applications as auxiliaries during melt spinning of synthetic polymer fibers and subsequent modification before coloring have been pointed out. Also, DES-based clean synthesis routes for challenging dyes has been discussed, followed by an overview into recent efforts leveraging DESs as co-solvents or secondary media to develop reduced-water dyeing processes for natural and synthetic textiles. Improved resource efficiency, darker shades, and accelerated diffusion kinetics have been reported relative to conventional dyeing, attributable to DES-fibre interactions for various pigment classes.

However, it should also be pointed out that, despite the tremendous potential, commercial adoption of DESs for textile processing remains limited presently mostly to bench/lab scale demonstration studies. Significant efforts in various directions are still required before viable large scale transition.

Thus, detailed environmental impact analysis using specific textile production consumption statistics needs to be performed, covering parameters like carbon footprint, cumulative energy demands, waste disposal effects, *etc*. This would verify if anticipated sustainability benefits outweigh invested resources. Comprehensive technical and economic feasibility analysis, considering costs of DES ingredients, preparation methods, processing equipment, operational overheads, waste handling, *etc*. *versus* revenues needs to be conducted across different application scenarios to identify viable options.

In addition, a wider range of DES components, compositions, and mixing protocols need to be systematically screened to determine optimal combinations customizable for targeted textile substrates and intended property enhancements. The statistical design of experiments can help accelerate identification. The findings from preparatory lab/bench studies need to be validated on semi-commercial scales through prototype pilot trials among interested industry partners to assess replicability before successful commercialization.

Appropriate personal protection equipment usage and safe handling procedures specific to DES synthesis and usage need to be developed for risk mitigation by exposure route studies. Effective protocols also need to be established for DES disposal after usage. Regulatory incentives and environmental policies promoting non-hazardous alternatives coupled with voluntary eco-standards adoption by textile majors can accelerate sustainable DES adoption to transform the sector.

In summary, concerted efforts across various domains spanning technology, sustainability, economic and regulatory spheres are necessary to realize widespread DES implementation for textile production ultimately.

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