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Mechanistic Insights Into Cellulose Dissolution in Solvents for Advanced Industrial Applications: A Systematic and Bibliometric Review

Chigozie Charity Okwuwa[1] | Fatmawati Adam[1,2] | Michael E. Ries[3] | Nor Hanuni Ramli[1] | Samuel Olugbenga Olunusi[1]

[1]Faculty of Chemical and Process Engineering Technology, Universiti Malaysia Pahang Al-Sultan Abdullah, Kuantan, Pahang, Malaysia | [2]Centre of Excellence for Advanced Research in Fluid Flow, Universiti Malaysia Pahang Al-Sultan Abdullah, Kuantan, Pahang, Malaysia | [3]School of Physics and Astronomy, University of Leeds, Leeds, UK

Correspondence: Fatmawati Adam (fatmawati@umpsa.edu.my)

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ABSTRACT

Cellulose dissolution is important for various industries, including textiles, bioplastics, foods and pharmaceuticals, yet achieving efficient dissolution remains challenging. Deep eutectic solvents (DES) have emerged as promising alternatives to traditional solvents due to their low toxicity, biodegradability and sustainability. This review critically examines recent mechanistic interactions between cellulose and different solvents, focusing on the green solvent known as DES, aiming to enhance industrial applications. It begins by discussing the possible conversion of cellulose nanocrystals from their different sources and the limitations of conventional solvents in dissolving cellulose. It then explores the interactions between cellulose and DES components, explaining the mechanisms that facilitate cellulose dissolution. This study focuses on the trends of DES and their role in dissolving cellulose. Bibliometric methods were employed to analyze these trends, along with identifying current research gaps, challenges and their dissolution in DES, thereby contributing to the creation of cost-effective industrial processes. Additionally, it outlines opportunities for further research and innovation in this field.

1 | Introduction

Cellulose, the most abundant and widely utilized natural polymer on Earth, is an important resource for textiles, pharmaceuticals, packaging, biofuels, bioplastics and renewable energy applications [1]. It is essential to different industries due to its structural properties and adaptability [2]. Cellulose can be derived from three main sources: plants, microbes and animals [3]. The paper and pulp industries primarily utilize wood-derived cellulose, which constitutes 40%–50% of the lignocellulosic biomass composition [4]. Plant-based cellulose is found in cotton, hemp, jute and other plant fibres. Cotton, for instance, contains nearly pure cellulose, making it an ideal material for textile production, while hemp and jute are utilized in the production of ropes, carpets and biodegradable plastics [5]. Its unique properties, such as biodegradability, renewability, mechanical stability and biocompatibility, have given rise to immense scientific interest in developing advanced methodologies for its sustainable and efficient utilization [6]. Structurally, cellulose is a linear polysaccharide of β-1,4-linked glucose units held together by extensive intra- and intermolecular hydrogen bonds [7], forming a rigid, extended polymer network whose remarkable tensile strength and insolubility in water and common organic solvents are dictated by this complex hydrogen-bonding network [8]. While the crystalline regions of cellulose impart mechanical strength, the amorphous domains cover reactivity and solubility [9]. These various characteristics make cellulose resistant to dissolution, thereby hindering its conversion into value-added materials and biofuels. Conventional dissolution techniques, such as the use of concentrated sulphuric acid and *N*-methylmorpholine-*N*-oxide (NMMO) [10], though effective, are environmentally damaging, energy-intensive and toxic; generate hazardous waste; and impose significant economic and ecological costs. As industrial sectors worldwide seek renewable alternatives to petrochemical feedstocks, the efficient, clean and scalable dissolution of cellulose has become an immediate priority and technological imperative with far-reaching implications for climate change mitigation, sustainable manufacturing and energy security [11–13]. Biodegradability, renewability, stability and biocompatibility are remarkable features that have increased considerable interest in exploring novel methodologies for efficient utilization [14–16]. Table 1 provides a summary of the extraction methodologies for obtaining cellulose nanocrystals (CNCs) from a variety of sources. It compares the processes by stating the raw materials, chemicals used for purification and hydrolysis, fundamental methods employed (acid hydrolysis and enzymatic treatment) and the reaction conditions. This information in the table allows for a direct comparison of how different experimental parameters influence the success of the extraction process and the final yield of CNCs obtained.

Due to its linear configuration and complex hydrogen-bonding structure, cellulose dissolution in conventional solvents is limited, which has caused some modification, such as esterification, etherification, and silylation, as shown in Figure 1 [47]. The essential characteristics of cellulose, such as its high tensile strength and resistance to water dissolution and other common organic solvents, are caused by the hydrogen bond network present [48]. Traditional nonaqueous solvents, like NMMO, are effective but can be energy-intensive [49]. Pretreatment methods like ultrasonication can enhance this process by physically modifying the cellulose, reducing its crystallinity and molecular weight while increasing its specific surface area and pore volume, thereby facilitating more uniform solvent penetration and faster dissolution [50]. The dissolution mechanism in NaOH is temperature-dependent, where low temperatures increase pH and reduce hydrophobic interactions, promoting ionization of cellulose hydroxyl groups [51]. All-cellulose composites (ACCs) use a NaOH solvent system combined with a freezing step, showing that a concentration of 8%–12% NaOH enhances dissolution through fibre welding; the efficacy of NaOH can be hindered by factors like high-pressure CO2 (HPCD), combined with acid, which can disrupt hydrogen-bonding networks, making cellulose more amenable to solvents like NaOH/urea/water; thereby improving dissolution [52].

The dissolution mechanism in ILs, such as 1-ethyl-3-methylimidazolium acetate ([EMIm][OAc]) mixed with dimethyl sulfoxide (DMSO), involves complex solute–solvent interactions. Molecular dynamics simulations reveal that dissolution rates are influenced by external factors like pressure, with high pressure decreasing the solvent's diffusion rate and strengthening cellulose–cellulose interactions, leading to dissolution [53]. Studies on amino acid-based ILs (AAILs) indicate that cellulose solubility is primarily governed by the solvent's Lewis acidity (SA) and basicity (SB), with higher basicity (strong hydrogen bond acceptance) promoting dissolution, except when hindered by the anion's large molecular volume or intramolecular hydrogen bonding [54]. ILs, novel phosphonate-based ILs have been synthesized and shown to dissolve cellulose at moderate temperatures, with their regeneration behaviour and hydrogen-bonding strength, calculated by density functional theory, providing a roadmap for their application in spinning processes [55]. Similarly, new green solvent systems combining tetraethylammonium hydroxide with imidazole compounds demonstrate high solubility for cellulose of varying degrees of polymerization, where the imidazole acts by reducing crystallinity and weakening intersheet forces without destroying hydrogen bonds, thus facilitating the action of the hydroxide ions [56].

In the quaternary ammonium/imidazole system, high temperatures required for dissolving high-degree-of-polymerization cellulose cause significant depolymerization [57]. Similarly, in phosphonate-based ILs, molecular weight analysis shows that degradation is a function of both time and temperature, necessitating a careful balance to achieve dissolution without compromising the material properties of the regenerated cellulose [58]. Laboratory studies have demonstrated promising dissolution efficiencies, and moving to industrial processes requires overcoming challenges in solvent recovery, recyclability, mass transfer limitations and compatibility with downstream applications such as fibre spinning, composite fabrication and biofuel production. Addressing these gaps is necessary, given the increasing global demand for sustainable polymers and the high environmental pressures to phase out toxic, resource-intensive technologies.

This review addresses this by presenting a comprehensive study of cellulose dissolution in solvents and integrating perspectives from chemistry, materials science and process engineering. This was done by (1) studying the sources, modifications, dissolutions and applications of cellulose in industries; (2) analysing the bibliometric data of current findings of cellulose dissolution in solvents; (3) the fundamental principles governing deep eutectic solvents (DES) as solvents for cellulose dissolution were examined, focusing on their unique solvent–solute interactions and their ability to disrupt the hydrogen-bonding network within cellulose; and (4) the current challenges and prospects for DES and cellulose dissolved in DES were proposed. In light of this, the present review not only synthesizes state-of-the-art knowledge but also proposes a way to bridge fundamental gaps, design scalable and green processes and align cellulose dissolution science with the global mandate for sustainable development.

2 | Bibliometrics on Cellulose Dissolution and Modification in General Solvents

Recently, bibliometrics has been recognized as a valuable method for predicting and analyzing research needs and future trends [59]. Keyword analysis uses co-occurring keywords in the literature set to ascertain the connections among topics in the represented research field. The change and frequency of keywords over time can reflect the hotspots and transitions of the research field and capture the research's main points. The keyword search analysis ('cellulose dissolution' OR 'cellulose modification' OR 'cellulose solubility' OR 'cellulose applications

TABLE 1 | Cellulose nanocrystal obtained from different sources.

Materials	Chemicals	Methods	Conditions	CNC obtained	Ref
Coconut agrowaste	NaOH, H_2O_2, H_2SO_4, PVA	Jaw crusher, ball milling	8 h at 50°C	3.42 nm	[17]
Lemon (*Citrus limon*) seeds	H_2SO_4, APS, TEMPO, sodium bromide, HCl and NaOH	Ultrasonication	1.5 h at 45°C	10–20 nm	[18]
Brewer's spent grain	NaOH, CH_3COOH and NaCl	Milling	2 h at 80°C	90–440 nm	[19]
Rice husk	$NaClO_2$, NaOH, CH_3COOH	Alkali treatment, bleached, acid hydrolysis	4 h at 100°C–130°C	10–15 nm	[20]
Rice straw (RS), wheat straw (WS) and barley straw (BS)	KOH, $NaClO_2$, CH_3COOH, C_2H_6O, $C_6H_5CH_3$, H_2SO_4	Acid hydrolysis and ultrasonication	75 min at 50°C	60–150, 40–110 and 15–90 $\mathcal{M}$m	[21]
Softwood forestry logging residues	NaOH, CH_3I, DMSO, TFA, $NaBH_4$, H_2SO_4, $NaClO_2$, C_5H_5N, $C_4H_6O_3$, $CH_3CO_2CH_2CH_3$, CH_3COCH_3, CH_3COOH, CH_3COONa	Ultrasonication, alkaline, bleaching, hydrolysis	20 min at 10°C	2.8–3.4 nm	[22]
Soy hulls	$NaClO_2$, NaOH, CH_3COOH,	Bleaching, ultrasonication	30 min at 40°C	4.36 nm	[23]
Agave tequilana Weber var. azul bagasse	H_2SO_4, HCl	Acid hydrolysis	45 min at 4°C	8.6 nm	[24]
Gelidium elegans red algae marine biomass	NaOH, H_2O_2	Alkalization treatment	2 h at 80°C	21.8 nm	[25]
Date palm waste	$NaClO_2$, NaOH, CH_3COOH, H_2SO_4	Ultrasonication, hydrolysis	45 min at 45°C	2.6 to 2.7 nm	[26]
Sugarcane bagasse fibres	H_2SO_4, $C_4H_8O_6$	Hydrolysis	7 h at 32°C	5 nm	[27]
Recycled Tetra Pak food packaging	NaOH, H_2SO_4, H_2O_2	Dialysis	2 h at 30°C	11.4–14 nm	[28]
Hardwood-bleached kraft pulp, softwood-bleached kraft pulp, cotton linters, cattail and red algae fibres	C_4H_9NO	Hydrolysis	50 min at 50°C	171–432 nm	[29]
Beer industrial residues	CH_3COOH, NaOH, CH_3COCH_3, HCl, KOH, $NaClO_2$,	Hydrolysis	1 h at 75°C	73–145 nm	[30]
Waste paper	NaOH, DI water,	Dialysis	30 min at—2°C	50 nm	[31]
Sweet lime pulp waste	*Komagataeibacter europaeus* SGP37 under static, intermittent fed-batch cultivation, H_2SO_4, NaOH	Hydrolysis, microorganism culture	1 h at 90°C	38 gL^{-1}	[32]
Discarded cigarette filters	NaOH, NaOCl, C_2H_5OH,	Hydrolysis, dialysis	45 min at 45°C	8.28 nm	[33]

(Continues)

TABLE 1 | (Continued)

Materials	Chemicals	Methods	Conditions	CNC obtained	Ref
Jute fibres	NaClO2, ZnO, KOH, Oxalic acid, sulphur, zinc dithiocarbamate, zinc mercaptobenzothiazole (ZMBT),	Hydrolysis, Alkalization	2 h at 70 °C	50 nm	[34]
Cotton, rice straw and grape skin	NaOH, H_2O_2, H_2SO_4, $NaClO_2$/KOH	Hydrolysis	2 h at 55°C	10 nm	[35]
Garlic skin	H_2SO_4	Hydrolysis, dialysis	2 h at 60°C	58–96 nm	[36]
Ground nut	$NaClO_2$, NaOH, H_2SO_4	Ultrasonication, dialysis	75 min at 45°C	18 nm	[37]
Coffee grounds	KOH, NaOH, H_2SO_4, HCl, $NaClO_2$, CH_3COOH,	Hydrolysis, dialysis	2 h at 90°C	120 nm	[38]
Ferula gummosis (Fg)	NaOH, H_2SO_4, $NaCLO_2$, CH_3COOH	Acid hydrolysis, bleaching	2 h at 80°C	22.11 nm	[39]
Sweet potato residue	NaOH, H_2O_2,	Hydrolysis, bleaching	45 min at 60°C	20 to 40 nm	[40]
Chardonnay grape skins	$C_6H_5CH_3$, CH_3CH_2OH, H_2SO_4, $NaClO_2$, H_2O_2, CH_3COOH, NaOH, N_2	Hydrolysis, bleaching	5 h at 70°C	5 nm	[41]
Onion skin	$C_3H_8O_3$, H_2SO_4, N_2	Bleaching, hydrolysis	3 h at 60°C	20–35 nm	[42]
Corncob	NaOH, KOH, H_2SO_4, $NaClO_2$, CH_3COOH, CH_3CH_2OH	Hydrolysis, Dialysis	30 min at 45°C	4.15 nm	[43]
Grape pomace	C_2H_5OH, H_2SO_4, NaOH, H_2O_2	Hydrolysis, bleaching	1 h at 30°C	10–20 nm	[43]
Vine shoots waste	NaOH, H_2SO_4, CH_3COOH, $NaClO_2$	Hydrolysis, dialysis, bleaching	20 min at t 15°C	14 nm	[44]
Red algae	NaOH, $NaClO_2$, CH_3COOH	Hydrolysis, bleaching	2 h at 80°C	5.2–9.1 nm	[45]
Forest residues	NaOH, H_2SO_4	Bleaching, dialysis	40 min at 45°C	2.8–3.4 nm	[22]
Sago seed shell	H_2SO_4	Hydrolysis, dialysis	40 min at 45°C	9.4 nm	[46]

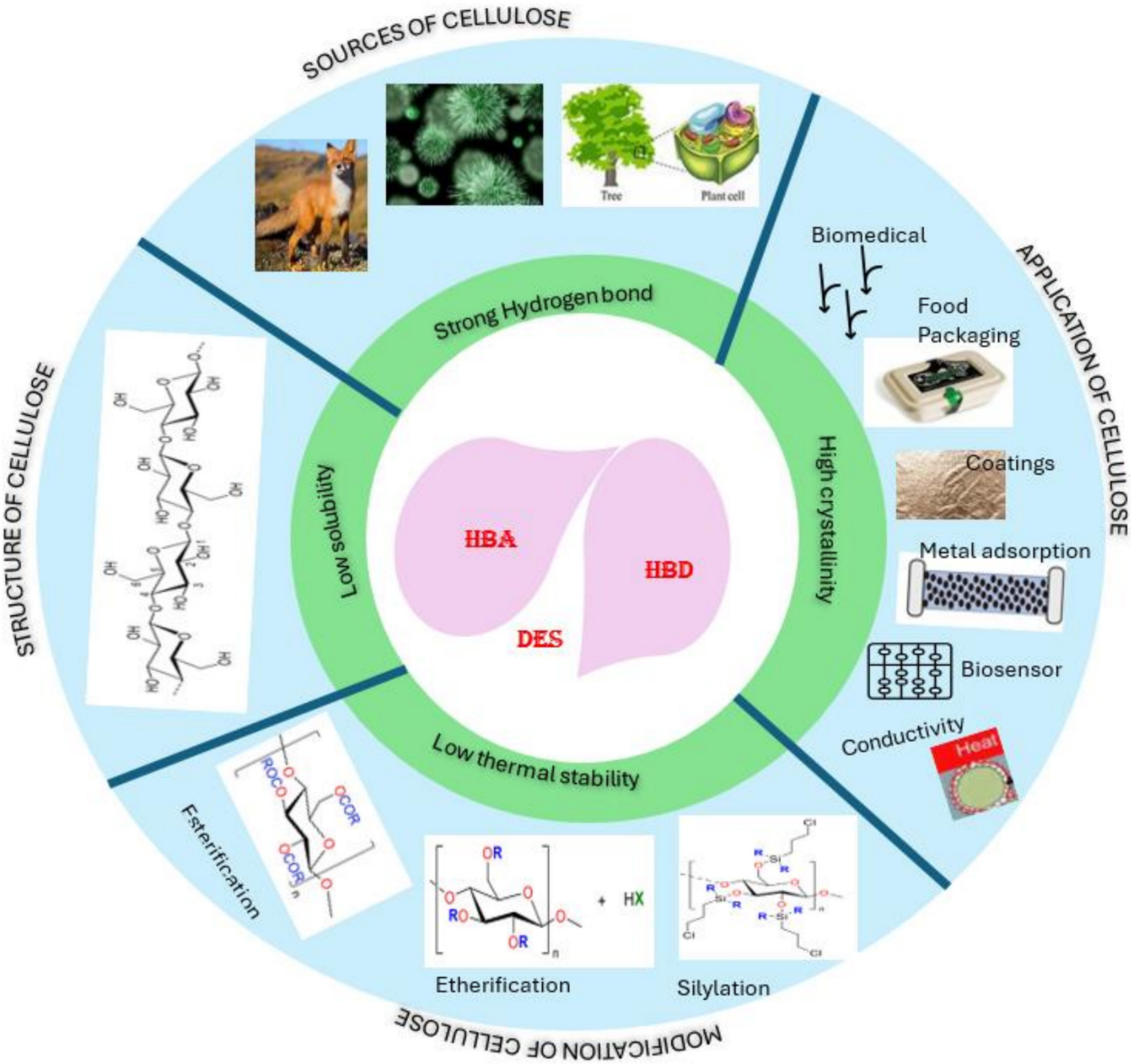

FIGURE 1 | Sources, structure, modification and application of cellulose.

in industries') was used in Scopus from 2000 to 2024, showing 950 published articles.

The keyword co-occurrence analysis generated by VOSViewer, with a threshold of five keywords for a minimum number of occurrences of a particular keyword, shows that 718 keywords meet this threshold. The pie chart and the histogram graph show different subject areas and territories that were obtained from Scopus. According to Figure 2a, based on keyword co-occurrence analysis, the global geographic distribution and co-operative network relationship of research results from 2000 to 2024 were further grouped in a pie chart (Figure 2b) according to the subject areas where these words are frequently applied; material science, chemistry, chemical engineering, environmental science and engineering were seen to use these words frequently. The top five countries in terms of publications were China, the United States, Germany, Finland and Sweden. The National Natural Science Foundation of China was the largest funding agency with 161 studies in the Scopus core collection. Figure 2c shows the documents per area and territories.

These keywords highlight crucial trends in cellulose dissolution research, which have been categorized into seven distinct clusters as shown in Table 2. Cluster 1 emphasizes cellulose, a naturally abundant, renewable and biodegradable polymer, making it a promising candidate for environmentally friendly packaging materials [60]. Also concerning plastic pollution, cellulose-based packaging offers a sustainable alternative due to its biodegradability and capacity to be sourced from renewable biomass [61]. Cellulose can be processed into films, coatings and composites with strong mechanical properties, flexibility and oxygen barriers, and recent modifications have improved its hydrophobicity and strength for food packaging [62]. Moreover, cellulose-based packaging can be combined with antimicrobial

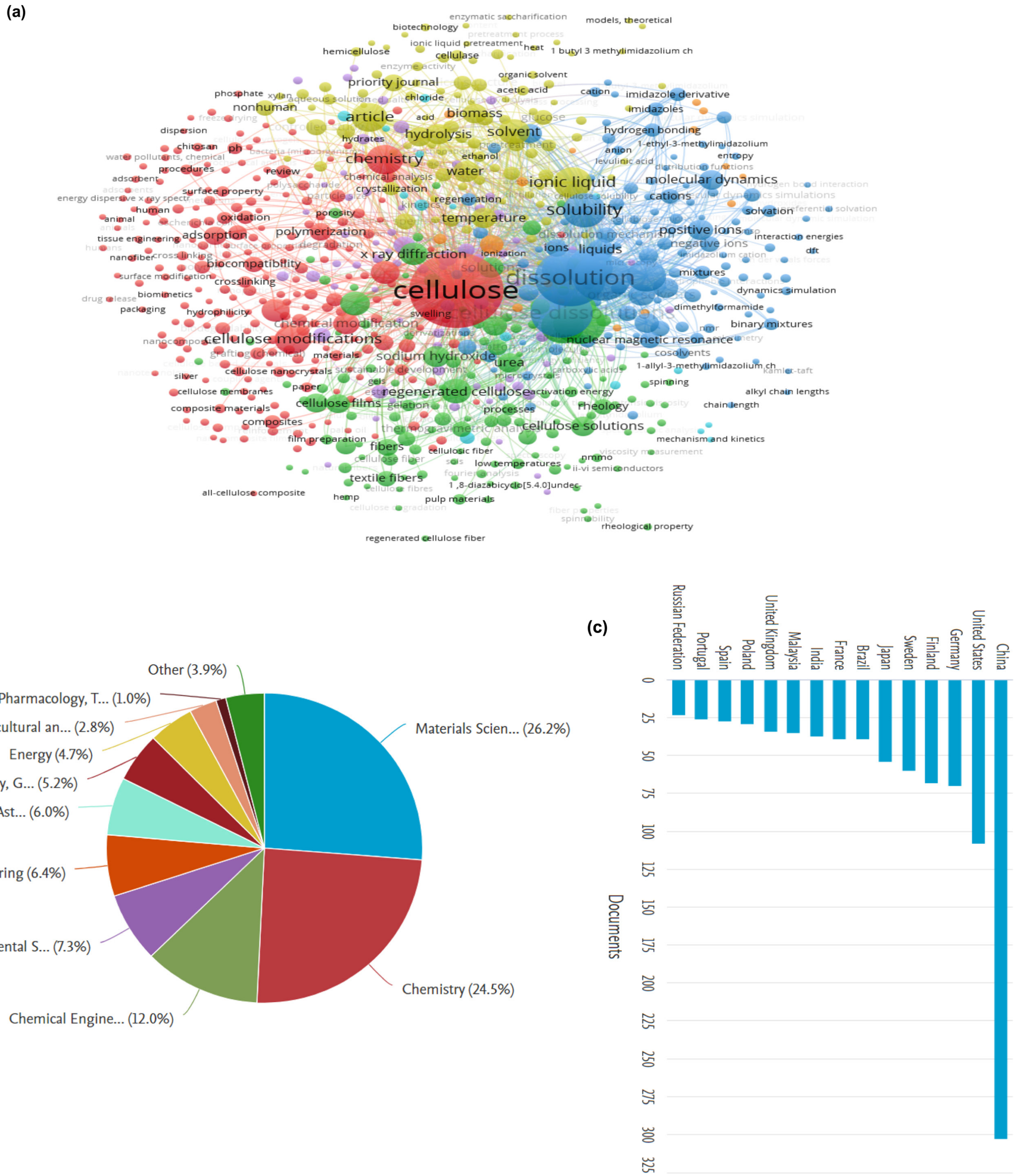

FIGURE 2 | Bibliometric analysis on the topics of cellulose dissolution/modification (a) co-occurrence analysis from frequent keywords from VOSViewer (b) documents by subject areas (c) documents per area/territories.

agents to prolong shelf life [63], further expanding its use in the food industry. Using cellulose in packaging not only reduces dependency on petroleum-based plastics but also helps to minimize environmental impacts due to its ability to degrade in natural environments without leaving harmful residues.

At Cluster 2, the chemical modification of cellulose was seen to enhance its properties and enable its use in advanced applications, such as biomedicine, textiles and environmental sustainability [64]. Through processes like etherification, esterification and oxidation, the surface properties, solubility and mechanical strength of cellulose can be improved [65]. Modified cellulose can be tailored for specific uses, such as in drug delivery systems, where functional groups are introduced to control drug release [66]. In textile engineering, cellulose modifications enhance dyeability, water resistance and antimicrobial properties, while in environmental applications, modified cellulose is applied in pollutant adsorption and water purification [67]. Moreover, cellulose derivatives are increasingly used in electronics, where their conductivity and flexibility make them suitable for flexible

TABLE 2 | Summary of coword analysis on cellulose modification/dissolution.

Cluster color	Number of keywords	Cluster labels	Representative keywords
1– Red	246	Cellulose as a biodegradable packaging material	Absorption, biodegradable, biomaterial, cellulose modification, etherification and packaging materials
2– Green	134	Modified cellulose for advanced applications	Bleaching, cellulose dissolution, crystalline materials, dissolution process and regenerated cellulose
3– Blue	128	Molecular simulations of hydrogen bonds in cellulose	Hydrogen bonds, molecular simulations, nuclear magnetic resonance, solubility and solvents
4– Indigo	107	Optimization of cellulose pretreatment in DES	Cellulose pretreatment, chemical compositions, chemical reaction, chemical structure, deep eutectic solvent, optimization, toxicity and sustainable chemistry
5– Purple	68	Stability of the synthesized process in cellulose dissolution	Catalyst, carbohydrate, cellulose hydrolysis, green solvents, synthesized, stability, substitution reaction and valorization
6– Light blue	19	Mechanism and kinetics of cellulose chain disentanglement and decrystallization	Cellulose chain, cellulose fibres, cellulose solubility, cellulosic fibres, chain disentanglement, decrystallization, mechanism and kinetics
7– Orange	16	Flow kinetics and agglomeration in the chemical industry	Agglomeration, chemical industry, flow kinetics, light scattering, x-ray scattering and x-ray diffraction

screens and sensors [68]. By modifying its molecular structure, cellulose becomes a versatile material that aligns with the principles of green chemistry, offering eco-friendly solutions across various industries.

Cluster 3, the molecular simulations provide valuable insights into the behaviour of hydrogen bonds within cellulose, particularly their role in its solubility and mechanical properties [69]. Hydrogen bonds between cellulose chains contribute to the material's high crystallinity and stability, making dissolution difficult. Simulating hydrogen bond interactions in different solvents allows researchers to predict how cellulose interacts with various chemical environments [70]. These simulations can help identify optimal solvents that break hydrogen bonds more effectively, facilitating cellulose dissolution without compromising its structure. Understanding the dynamics of hydrogen bonding through molecular simulations can also explain the mechanisms behind cellulose chain disentanglement and decrystallization [71]. Such insights are critical for improving cellulose processing techniques, enabling more efficient production of regenerated cellulose fibres, films and composites for industrial applications. These simulations aid in designing better solvents, thereby enhancing cellulose's usability in sustainable, high-performance materials.

Cluster 4 shows that the use of DES in cellulose pretreatment has emerged as a sustainable alternative to conventional methods, which have high potential in biomass processing [72], offering lower toxicity and improved environmental profiles [73]. Optimization of this pretreatment process involves adjusting solvent compositions and conditions to maximize cellulose dissolution, decrystallization and further utilization in applications such as biofuels, biocomposites and biodegradable materials [74, 75]. DES, formed by combining a hydrogen bond donor (HBD) and a hydrogen bond acceptor (HBA), can disrupt the hydrogen bonds in cellulose, enhancing its solubility and reactivity. By fine-tuning the DES components, such as choline chloride and glycerol, researchers can improve the efficiency of cellulose dissolution while maintaining the solvent's low environmental impact [76]. Optimization studies also explore the influence of temperature, reaction time and solvent-to-cellulose ratio to achieve the best performance [77, 78]. The goal is to develop an eco-friendly and efficient method for processing cellulose into valuable materials while minimizing the solvent's toxicity and its environmental footprint.

Cluster 5 emphasized the importance of the stability of synthesized catalysts or solvents during cellulose dissolution, which is seen as crucial for improving the efficiency of cellulose processing for various industrial applications [79]. Green solvents such as ionic liquids and DES have gained attention recently due to their ability to dissolve cellulose under mild conditions. However, maintaining the stability of these solvents and preventing degradation during the dissolution process is essential to ensure reproducibility and cost-effectiveness [80]. Studies on catalyst stability in cellulose dissolution focus on understanding the chemical interactions between cellulose and the solvent, as well as the role of temperature and reaction time in preserving solvent efficiency [81, 82]. The goal is to achieve a sustainable and stable dissolution process that can be applied to produce regenerated cellulose products, such as fibres and films. Ensuring the stability of these systems not only enhances the dissolution process but also extends the potential applications of cellulose in various industries.

Cluster 6 shows the disentanglement and decrystallization of cellulose chains, which are critical steps in the dissolution process, directly influencing the production of regenerated cellulose materials [83]. The mechanism of cellulose dissolution involves the disruption of intermolecular hydrogen bonds, leading to the unraveling of tightly packed crystalline regions [84]. By understanding the kinetics of this process, researchers can identify the conditions that optimize cellulose dissolution in solvents like DES or ionic liquids [85]. The rate at which cellulose chains disentangle depends on factors such as solvent concentration, temperature and time [86]. Decrystallization is essential for transforming highly ordered cellulose into an amorphous form, which is more easily processed into fibres, films or biocomposites [87, 88]. Investigating the mechanism and kinetics of these phenomena provides valuable information for improving the efficiency and scalability of cellulose processing techniques for various industrial applications, including textiles, packaging and biomedical products [89].

Cluster 7 brought the chemical industry into understanding that the flow of kinetics and the aggregation of particles during processes such as crystallization, drying or mixing is essential for optimizing production efficiency and product quality. Agglomeration refers to the clustering of fine particles, which can affect the flow properties of materials and the uniformity of chemical reactions [90]. Techniques like light scattering and XRD provide valuable data on particle size distribution and structural properties, helping to monitor and control agglomeration during processing [91]. Light scattering allows real-time observation of particle behaviour in suspension [92], while XRD provides insights into crystalline structure and phase changes during agglomeration. By analyzing flow kinetics and agglomeration, researchers can develop strategies to improve material handling, reduce processing costs and enhance the performance of products in industries such as pharmaceuticals, food processing and materials engineering. Understanding these dynamics is critical for designing more efficient and reliable chemical processes [93]. Hence, the results of the bibliometric analysis provided a foundation for better predicting future research and understanding the current state of research on cellulose dissolution, as it tends towards utilizing DESs and will be useful for future researchers in related research areas.

3 | Cellulose Dissolution in Conventional Solvents and the DES

The dissolution of cellulose in different solvents is an area with significant implications for various industrial applications, ranging from producing textiles to developing biodegradable materials [94]. Cellulose has an extensive hydrogen-bonding network, making it insoluble in water and most common organic solvents, necessitating specialized solvents for its dissolution [95]. Traditional solvents for dissolving cellulose include phosphoric acid-based solvents, LiCl-based solvents, *N*-methylmorpholine *N*-oxide/water, ionic liquids and NaOH–water solutions. These have been extensively used due to their ability to disrupt the hydrogen-bonding network in cellulose, facilitating its dissolution [48]. DMAc/LiCl, for instance, is highly effective in dissolving cellulose, allowing for subsequent chemical modifications and processing into fibres and films [96] as shown in Figure 3. However, these solvents pose significant environmental and safety concerns, including toxicity, flammability, solvent recovery and recycling difficulties. In recent years, the development of ionic liquids (ILs) has provided a promising alternative for cellulose dissolution. Ionic liquids, salts in the liquid state at relatively low temperatures, exhibit unique properties such as low vapour pressure, high thermal stability and the ability to dissolve a wide range of substances, including cellulose [97]. ILs such as 1-butyl-3-methylimidazolium chloride ([Bmim]Cl) have shown remarkable efficacy in dissolving cellulose, enabling homogeneous chemical modifications and the production of high-performance materials [98].

Cellulose exhibits both crystalline and amorphous regions, a property that significantly influences its physical and chemical behaviour. The crystalline regions are highly ordered, resulting from the regular arrangement of cellulose chains and extensive hydrogen bonding [99]. These regions contribute to cellulose mechanical strength and resistance to enzymatic and chemical degradation, while the amorphous regions are less ordered, allowing for some flexibility and accessibility to chemical reagents [100]. The degree of crystallinity can vary depending on the source of cellulose and its processing methods, affecting properties such as solubility, reactivity and mechanical strength [101]. Techniques like XRD are often used to analyze the crystalline structure of cellulose, which provides insights into the arrangement and proportion of crystalline versus amorphous regions [102]. One of the major challenges in processing cellulose is its resistance to solubilization due to the strong hydrogen bonding in its crystalline regions. This resistance has led to a growing interest in innovative solvents, particularly DESs, which offer a promising solution for cellulose dissolution. DESs are emerging as environmentally friendly, tunable solvents capable of breaking down the hydrogen bonds in cellulose, thereby facilitating its dissolution without the need for harsh chemicals or extreme conditions. The use of DESs enables a more efficient and sustainable approach to processing cellulose, as they are often biodegradable, non-toxic and derived from renewable resources [103].

The dissolution of cellulose in DES follows a mechanism driven by the unique hydrogen-bonding network of the solvent. The process initiates with the penetration of the DES into the amorphous regions of the cellulose fibril, where the less-ordered polymer chains are more accessible. The dissolution is primarily facilitated by the synergistic action of the two components of the DES, the HBA, typically a quaternary ammonium salt like choline chloride, and the HBD, such as urea, glycerol or a carboxylic acid [104]. The HBA (the chloride anion, Cl^-) is the nucleophile, attacking and breaking the extensive intra- and intermolecular hydrogen bonds between the hydroxyl groups (-OH) of adjacent cellulose chains. Also, the HBD donates protons to form new

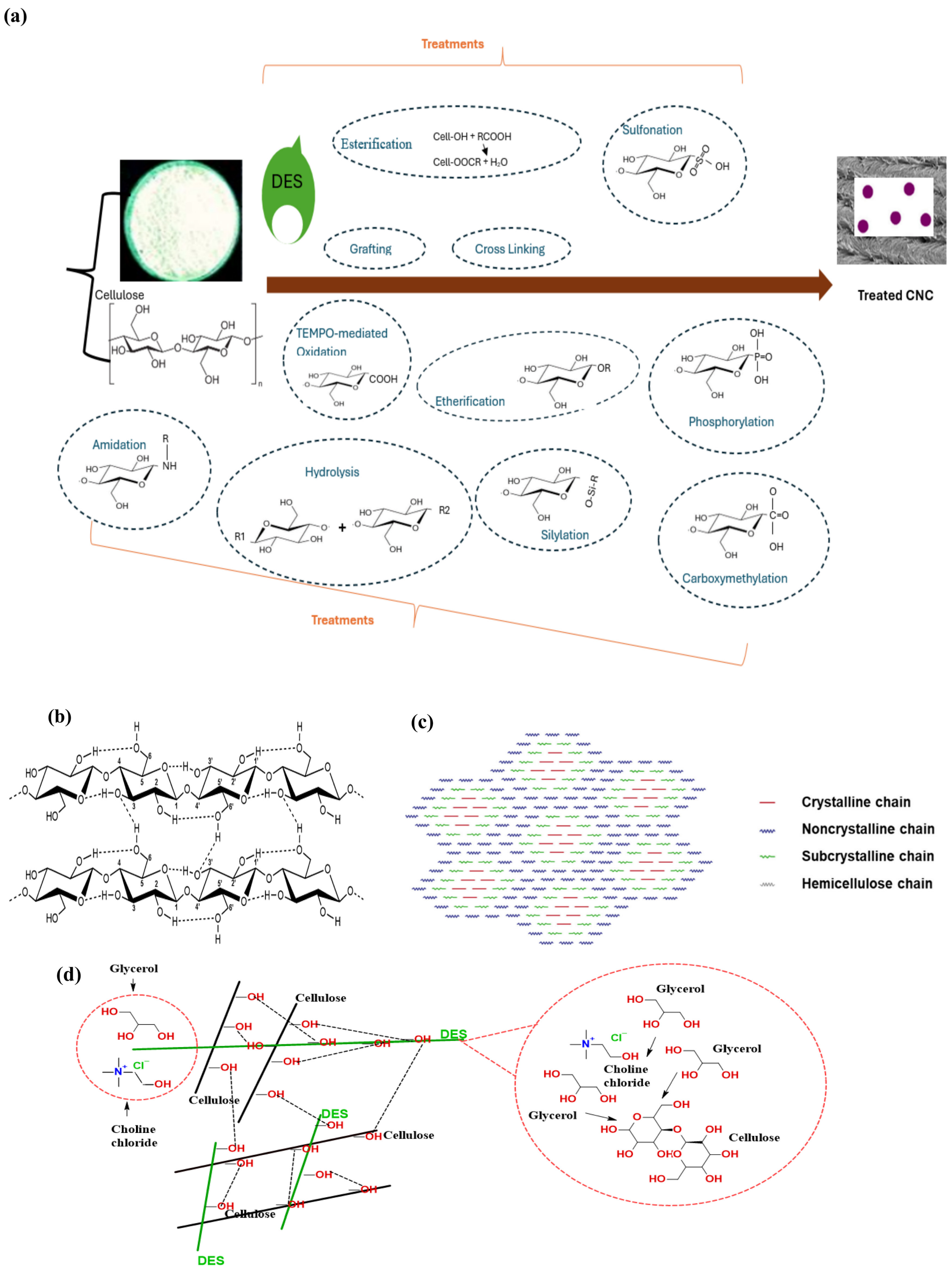

FIGURE 3 | (a) Modifications of cellulose in solvents, (b) molecular structure of cellulose highlighting intermolecular hydrogen bonding, (c) cellulose intermolecular chains and (d) interaction mechanism between cellulose and DES.

hydrogen bonds with the oxygen atoms of the cellulose glucopyranose rings. This dual action effectively helps the hydroxyl groups of cellulose, preventing the reformation of the native hydrogen-bonding network and stabilizing the individual cellulose chains in solution [105], leading to the disruption of the rigid crystalline structure resulting in the swelling of the fibril and the eventual separation of the polymer chains into a homogeneous solution, where the cellulose is molecularly dispersed and shielded by the DES components.

The functional groups in cellulose, particularly the hydroxyl groups, play a pivotal role in its reactivity and potential for chemical modification. These hydroxyl groups can contribute to various chemical reactions, which produce cellulose derivatives with tailored properties. For instance, cellulose acetate, made by acetylation of the hydroxyl groups, is used to manufacture textiles and photographic films [106]. Carboxymethyl cellulose, another derivative, is widely used as a thickening agent in food and pharmaceuticals [107]. However, the effectiveness of these modifications often depends on the accessibility of the hydroxyl groups, which is greatly enhanced by the dissolution of cellulose in a green solvent known as DESs [108], which is mild, environmentally friendly and highly biodegradable. Dissolving cellulose in DESs, as shown in Okwuwa et al. [82, 146] study, not only increases the availability of these functional groups for chemical modification but also enables the development of novel cellulose-based materials [82]. Biosynthetically, cellulose is produced by cellulose synthase enzymes located in the plasma membrane of plant cells. These enzymes polymerize glucose units into long cellulose chains, which are then extruded outside the cell membrane, where they aggregate to form microfibrils [109]. While this natural process is critical for plant growth and structural integrity, it also presents an opportunity for biotechnological applications. Dissolving cellulose in DESs provides a pathway to change its structure for specific uses, such as producing cellulose with enhanced mechanical or thermal properties for high-performance materials. Table 3 shows the comparison between the conventional solvents and the DES.

Advanced analytical techniques play a crucial role in elucidating the molecular structure of cellulose before and after dissolution in DES. FTIR and NMR spectroscopy are commonly used to identify the functional groups and understand the chemical environment within cellulose [121]. FTIR provides information on the vibrational modes of chemical bonds, revealing details about the hydroxyl groups and other functional groups present in cellulose. NMR spectroscopy offers insights into cellulose's molecular dynamics and interactions, allowing for detailed structural analysis [122]. Additionally, techniques like XRD and electron microscopy provide information on cellulose fibres' crystalline structure and morphological characteristics, further enhancing our understanding of their molecular architecture [123]. Computational modelling and molecular dynamics simulations have also become invaluable tools for studying cellulose at the molecular level. These methods allow researchers to predict the behaviour of cellulose molecules under different conditions, providing insights into their interactions, mechanical properties and reactivity [124]. By combining experimental data with computational models, scientists can better understand cellulose's molecular structure and its implications for various applications [125]. The molecular structure of cellulose is also significant in its role in sustainable and green chemistry. As a renewable and biodegradable polymer, cellulose offers an environmentally friendly alternative to synthetic polymers derived from fossil fuels [126]. Its chemical versatility allows for the development of a wide range of materials and products, from biodegradable plastics to biofuels and pharmaceuticals [127]. By leveraging our understanding of cellulose's molecular structure, researchers can design processes and products that minimize environmental impact and contribute to a more sustainable future [128]. The tunability of ILs, achieved by modifying the cation and anion components, allows for the optimization of their solubility and reactivity with cellulose [129]. Despite these advantages, the high cost of ILs and the challenges associated with their recovery and recycling have limited their widespread adoption in industrial processes [130].

This has led to another innovative class of solvents that has gained attention for cellulose dissolution, which is known as the DES, and has demonstrated excellent capability in dissolving cellulose under mild conditions, offering an environmentally friendly and cost-effective alternative to traditional solvents and ILs [131]. The biodegradability, low toxicity and potential for recycling of DES further enhance their appeal as sustainable solvents for cellulose processing.

Water-based solvent systems, such as aqueous sodium hydroxide (NaOH) and cuprammonium solutions, have also been explored for cellulose dissolution [132]. These systems capitalize on the ability of certain alkali or complexation agents to disrupt the hydrogen-bonding network in cellulose, rendering it soluble [133]. Aqueous NaOH, often combined with additives like urea or thiourea, can dissolve cellulose at low temperatures, although the resulting solutions are typically limited to low cellulose concentrations [134]. Cuprammonium solutions, used in the production of cupro fibres, form soluble complexes with cellulose, but environmental and economic considerations restrict their use due to the involvement of heavy metals and the complexity of the solvent recovery process [135]. The choice of solvent for cellulose dissolution significantly impacts the final cellulose products' efficiency, sustainability and properties.

Cellulose dissolution in DES represents a groundbreaking advancement in green chemistry, providing an eco-friendly alternative to traditional solvents [136]. DES is a class of solvents formed by mixing two or more components, typically a HBD and a HBA, which interact to create a eutectic mixture with a melting point significantly lower than that of the individual components [137]. This unique property of DES allows for the effective dissolution of cellulose under mild conditions, which is a significant improvement over traditional solvents that often require harsh conditions and pose environmental concerns [138]. The interaction between cellulose and DES is primarily driven by hydrogen bonding, where the hydroxyl groups of cellulose interact with the HBDs and acceptors in the DES, leading to the disruption of the extensive hydrogen-bonding network within the cellulose [139]. This disruption facilitates the dissolution process, making cellulose more accessible for subsequent chemical modifications and applications.

One of the key advantages of using DES for cellulose dissolution is its environmental benignity [140]. Many DES are composed

TABLE 3 | Comparison of deep eutectic solvents (DES) and traditional solvents for cellulose processing.

Feature	Traditional solvents (viscose process, NMMO and ionic liquids)	Deep eutectic solvents (DES)	Advancement of DES	Ref.
Environmental impact and green chemistry	The viscose process (CS_2/NaOH) is highly toxic and generates hazardous waste (H_2S). Ionic liquids are poorly biodegradable. NMMO is prone to exothermic decomposition [110]	Components are often natural, biodegradable and low toxicity (choline chloride + urea).	DES aligns with green chemistry principles, offering a safer, more sustainable and environmentally benign alternative that generates minimal hazardous waste.	[111]
Toxicity and safety	CS_2 is neurotoxic and flammable. Some Ionic Liquids can be toxic or have unknown ecotoxicity. NMMO requires careful control to avoid runaway reactions [112].	DES are composed of compounds such as choline chloride and organic acids. They are non-flammable and have low vapour pressure.	DES reduces workplace hazards, eliminates the need for complex containment systems for volatile toxins, and simplifies safety protocols.	[113]
Cost and synthesis	Ionic liquids are often expensive to synthesize and purify. NMMO is a commercial chemical with increased cost. The viscose process has high costs associated with solvent recovery and pollution control [114]	Materials are cheap, abundant and commercially available. Synthesis is simple: mixing components with mild heating, requiring no purification, resulting in 100% atom economy.	The low cost of components and simple preparation make DES highly attractive for scalable industrial applications, reducing production costs.	[115]
Tunability/designer solvents	Ionic liquids are tunable by changing cation/anion pairs. However, the synthesis of new ILs can be complex. Traditional solvents have fixed properties [116].	Properties as viscosity, polarity and solubility can be easily tailored by selecting different HBD and HBA.	DES allows for fine-tuning to dissolve specific types of cellulose or to integrate functional properties directly into the material during processing.	[117]
Biodegradability and recycling	Recycling of CS_2 in the viscose process is energy-intensive. Many ionic liquids are persistent in the environment. Solvent recovery is often complex [118].	Many DES are biodegradable. While recovery via antisolvent addition is similar to ILs.	The biodegradable nature of many DES components mitigates long-term environmental impact. The nature of the system facilitates recycling efforts.	[119]
Dissolution mechanism	NMMO and ILs primarily break inter- and intramolecular hydrogen bonds in cellulose. The viscose process is a derivatization (formation of cellulose xanthate) [55].	DES functions by breaking the extensive hydrogen-bond network of cellulose through the interaction of the HBA and HBD.	DES provides an effective dissolution pathway similar to advanced ILs.	[119]
Viscosity	Many pure Ionic Liquids and cellulose solutions in NMMO can have high viscosity, which leads to difficulty in processing [120]	DES has high viscosity, which hinders cellulose dissolution and mass transfer.	High viscosity is a limitation when compared to the traditional systems. However, the tunability of DES offers a pathway to optimize this property.	[117]

of naturally derived, biodegradable and non-toxic components, such as choline chloride combined with urea, glycerol1 or various organic acids [141]. These DES not only dissolve cellulose efficiently but also align with the principles of green chemistry by minimizing the use of hazardous substances and reducing the overall environmental footprint of the dissolution process [142]. Additionally, DES often exhibit low volatility and high thermal stability, which further enhances their suitability for industrial applications [143]. The ability to recover and recycle DES after cellulose dissolution adds another layer of sustainability to this approach, making it economically viable as well [144]. The recyclability of DES reduces waste and lowers the cost associated with solvent use, thus promoting more sustainable industrial practices.

The versatility of DES in dissolving cellulose is another critical factor contributing to its widespread adoption. By carefully selecting and tailoring the components of DES, the solvent system can be optimized for specific cellulose sources and desired outcomes. For example, DES, composed of choline chloride and urea, has been shown to dissolve microcrystalline cellulose effectively [145, 146], whereas mixtures involving choline chloride and glycerol can dissolve more recalcitrant forms of cellulose, such as those found in lignocellulosic biomass [147]. This tunability allows for precise control over the dissolution process, enabling the production of cellulose derivatives with tailored properties for various applications. Furthermore, the mild conditions under which DES operates help preserve the integrity of the cellulose, resulting in high-quality products suitable for high-value applications [148].

Dissolving cellulose in DES also opens new avenues to produce cellulose nanomaterials [149]. By leveraging the unique properties of DES, cellulose nanofibers and nanocrystals with controlled size and morphology can be produced [150]. These nanomaterials exhibit exceptional mechanical strength, high surface area and unique optical properties, making them ideal for applications in nanocomposites, reinforcing agents and even electronic devices [151]. The ability to produce high-quality cellulose nanomaterials in an environmentally friendly manner represents a significant advancement in material science, offering sustainable alternatives to conventional nanomaterials derived from non-renewable sources [152]. The laboratory-scale studies have demonstrated the effectiveness of various DES; scaling up these processes requires careful consideration of factors such as solvent cost, recovery and recycling efficiency [153]. The long-term stability, reusability and industrial application of DES need to be thoroughly investigated to ensure their practicality for continuous industrial operations [154]. Advances in process engineering and solvent recovery technologies will play a crucial role in overcoming these challenges and establishing DES as a mainstream solution for cellulose dissolution.

4 | Applications of Cellulose Dissolved in the DES

Cellulose, an abundant and renewable natural polymer, has drawn substantial interest for its broad range of applications when dissolved in DESs [155]. DESs, composed of HBDs and HBAs, present a greener alternative to traditional solvents due to their non-toxic, biodegradable and tunable properties [156]. These solvents have enabled significant advancements in the utilization of cellulose, which is notoriously difficult to dissolve because of its extensive hydrogen-bonding network and crystalline structure [157]. When dissolved in DESs, cellulose can be harnessed for a variety of industrial and scientific applications, driving innovation in several fields [158]. In the textile industry, the dissolution of cellulose in DESs facilitates the production of regenerated cellulose fibres, such as lyocell and viscose, which serve as sustainable alternatives to synthetic fibres like polyester and nylon [159, 160]. These regenerated fibres exhibit superior properties, including high tensile strength, breathability and biodegradability [161]. The use of DESs in this process minimizes the environmental impact compared to conventional methods that rely on harsh chemicals. The fibres produced are increasingly sought after in the fashion industry, which is pressured to reduce its environmental footprint and adopt more sustainable practices [162]. Beyond textiles, the potential of cellulose-DES systems extends to the realm of bioplastics. Dissolving cellulose in DESs makes it possible to create films and composites that serve as renewable substitutes for traditional petroleum-based plastics [163]. These bioplastics are utilized in packaging, agricultural films and disposable items, where biodegradability is crucial [88, 164]. The cellulose-DES-derived bioplastics not only reduce dependency on fossil fuels but also mitigate plastic pollution, aligning with global efforts to tackle environmental issues associated with plastic waste [165]. Some of the applications of cellulose dissolved in DES are shown in Table 4.

In the biomedical field, cellulose dissolved in DESs is used to develop advanced functional materials such as hydrogels and aerogels [168]. These materials are prized for their high porosity, tunable mechanical properties and biocompatibility. Hydrogels, for instance, find applications in wound dressings, drug delivery systems and tissue engineering scaffolds [169]. Aerogels, with their ultralightweight and high surface area, are employed in insulation, adsorption of pollutants and as carriers for controlled drug release [170]. The ability to tailor the properties of DESs enables the customization of these materials for specific medical applications, enhancing their efficacy and utility [171]. Another significant application of cellulose-DES systems is in the production of nanocellulose [172]. Nanocellulose, derived from

TABLE 4 | Deep eutectic solvent systems for cellulose dissolution and their industrial applications.

DES system	Improved properties	Industrial applications	Ref
ChCl: Gly (1:2)	EB: 85.3%, capsule loop strength: 39.9 N	Hard capsules	[166]
ChCl: formic acid (1:2)	Lignin solubility: 14	Biofuels	[11]
ChCl: oxalic acid (2:1)	Dissolution rate: 2.5%	Electrical conductivity	[18]
ChCl: lactic acid (1:1)	Enhanced microbial activity	Biomedical application	[140]
ChCl: acetic (1:2)	Improved biodegradability	Packaging industry	[167]

cellulose fibres, exhibits exceptional mechanical properties, a high aspect ratio and a large surface area, making it an ideal reinforcing agent in composites. These composites are used in automotive parts, construction materials and high-strength papers [173]. The solubilization of cellulose in DESs also has profound implications for the bioenergy sector [174]. The dissolution process enhances the enzymatic hydrolysis of cellulose, which is a critical step in the production of biofuels and biochemicals from lignocellulosic biomass [175]. This advancement is particularly important for the efficient conversion of agricultural and forestry residues into valuable bioenergy resources, thereby contributing to the sustainable energy landscape [176]. The use of DESs in this context supports the principles of green chemistry, promoting cleaner production processes and reducing environmental pollution [177].

Furthermore, the dissolution of cellulose in DESs paves the way for the development of novel functionalized materials with specific chemical properties. For example, cellulose can be modified to introduce functional groups that impart antimicrobial, flame-retardant, or conductive properties [112]. These functionalized cellulose materials are applicable in various industries, including healthcare, electronics and construction. The versatility of DESs allows for the fine-tuning of solvent properties to achieve optimal dissolution and functionalization of cellulose, thereby expanding its application scope [178]. The sustainability aspect of using DESs in cellulose processing is noteworthy. Additionally, DESs can be easily recovered and reused in the process, further enhancing the sustainability of the cellulose dissolution and application cycles [179]. This aligns with the growing emphasis on circular economy principles, where the focus is on reducing waste, reusing materials and recycling resources to minimize environmental impact. Scientists are exploring the customization of DESs by varying the types and ratios of HBDs and HBAs to tailor solvent properties for specific applications [180]. This customization is key to optimizing the efficiency of cellulose dissolution and improving the performance of the resulting materials. Advanced analytical techniques are being employed to understand the interactions between cellulose and DESs at the molecular level, providing insights that drive further innovation [181].

5 | Safety Measures in Cellulose Dissolution

While DESs are less toxic than conventional solvents, the chemical materials involved in the cellulose dissolution process still need safety protocols to ensure the protection of workers, operational equipment and the environment [174]. Proper safety protocols should be followed, starting with the use of personal protective equipment (PPE) such as gloves, safety goggles, lab coats and respiratory protection when necessary. All chemicals should be clearly labelled and stored in suitable containers, away from incompatible substances, with updated material safety data sheets (MSDS) readily available and familiar to all personnel [182]. Ventilation systems, including fume hoods and local exhaust ventilation, are essential to prevent the buildup of harmful vapours, while temperature control within the dissolution area is crucial to mitigate the volatility of DES components [183]. Spill kits with absorbent materials, neutralizing agents and protective equipment should be easily accessible, and personnel must be trained in spill response procedures. Regular inspection and maintenance of dissolution equipment are necessary to prevent leaks or malfunctions, with pressure and temperature sensors in place to monitor operating conditions. Training programs covering chemical handling, equipment operation, emergency response and waste disposal are vital, supported by detailed standard operating procedures (SOPs) for all process aspects [184]. An emergency response plan, including evacuation routes, first-aid procedures and emergency services contact information, must be established and communicated to all staff, with well-stocked first-aid stations available. Proper waste disposal in line with regulatory guidelines is essential, also avoiding the release of chemicals into drains or regular trash, and recycling and reuse practices should be implemented wherever possible [185]. Continuous monitoring systems for air quality, chemical concentrations and environmental conditions help maintain a safe working environment, and regular risk assessments identify potential hazards, guiding mitigation efforts [186]. Environmental protection measures include effluent treatment to remove harmful chemicals before discharge and regular monitoring of environmental parameters to ensure compliance with regulations. Adhering to local, national and international regulations regarding chemical safety, workplace safety and environmental protection is non-negotiable, with comprehensive documentation of safety protocols, training records, risk assessments and regulatory compliance activities maintained [187]. By implementing these rigorous safety measures, the risks associated with cellulose dissolution using DESs can be minimized, ensuring the protection of personnel and the environment while enabling the continued use of this innovative and sustainable method. Continuous improvement and strict adherence to regulatory standards are essential for maintaining a safe and efficient operation, promoting the broader adoption of cellulose-DES systems across various industries [188].

6 | Conclusion

The dissolution of cellulose using DESs represents an approach with significant potential to drive sustainable innovation across various industries. This method addresses the long-standing challenge of breaking down cellulose's robust hydrogen-bonding network and crystalline structure, which is traditionally difficult with conventional solvents that pose environmental and health risks. DESs, characterized by their low toxicity, biodegradability and tunable properties, offer a greener alternative, facilitating efficient cellulose dissolution and enabling the creation of a wide range of high-performance materials. The application spectrum is vast, encompassing the textile industry, where regenerated cellulose fibres provide sustainable alternatives to synthetic ones, and the bioplastics sector, where cellulose-derived films and composites reduce reliance on petroleum-based plastics and mitigate environmental pollution. In biomedical fields, cellulose-DES systems enable the development of advanced functional materials like hydrogels and aerogels, which are essential for wound dressings, drug delivery systems and tissue engineering due to their high porosity and biocompatibility. The industrial perspectives for cellulose dissolution using DESs are promising, driven by the global demand for sustainable and biodegradable materials. By addressing the technical, economic and regulatory barriers, cellulose-DES systems can significantly contribute to the development of eco-friendly materials and processes, paving the way for a more sustainable future. The advancement of this technology aligns with the principles of green chemistry, promoting cleaner production methods and reducing environmental impact, thereby supporting global efforts to create a circular economy and mitigate climate change. The dissolution of cellulose in DESs offers a compelling solution to current environmental and industrial challenges, providing a pathway to sustainable material innovation and driving progress towards a greener and more resilient future.

Author Contributions

Chigozie Charity Okwuwa: conceptualization, writing – original draft, writing – review and editing, and visualization. **Fatmawati Adam:** writing – review and editing and supervision. **Michael E. Ries:** writing – review and editing and validation. **Samuel Olugbenga Olunusi:** writing – review and editing. **Nor Hanuni Ramli:** writing – review and editing.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

Data sharing not applicable to this article as no datasets were generated or analysed during the current study.

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