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From textile waste to sustainable biomaterials: the role of ionic liquids and deep eutectic solvents in protein-based textile waste valorization

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The 2030 Agenda aims to create strategies based on circular economy while addressing economic, environmental, and social issues. In this context, implementing a sustainable bioeconomy for waste valorization is indispensable for the appropriate development of society. Although vital to global socioeconomic development, the textile industry is one of the most environmentally harmful sectors due to its reliance on non-renewable resources, high energy consumption, and significant waste generation. In response to these challenges, this critical review addresses sustainable strategies for the valorization of protein-based textile waste, specifically silk and wool, emphasizing the potential of ionic liquids (ILs) and deep eutectic solvents (DESs) as alternative and efficient solvents for protein recovery. The proteins from silk and wool can be efficiently recovered and reused through upcycling strategies in various value-added applications. This approach strengthens a sustainable bioeconomy focused on waste recovery and valorization, which is essential for balanced and environmentally responsible socioeconomic development. Based on their relevance, this review discusses the chemical and structural properties of the proteins present in silk and wool, namely fibroin, sericin, and keratin, as well as conventional and advanced recycling methods. It further discusses the mechanisms of protein dissolution in ILs and DESs, highlighting their efficacy in selectively dissolving these target biomacromolecules while preserving structural integrity and functionality. This review also presents recent advances in upcycling these proteins into high-value materials, including biomedical scaffolds, hydrogels, films, and nanostructures. Furthermore, a SWOT analysis is shown to critically assess the current potential and limitations of IL and DES-based approaches to recycle protein-rich textile waste, identifying strengths such as process selectivity and structural preservation, as well as challenges related to cost, scalability, and environmental impact.

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1. This review discusses recent advances in green strategies for valorizing protein-based textile waste (silk and wool) using ionic liquids and deep eutectic solvents, covering selective dissolution, degumming, protein recovery, and upcycling into high-value biomaterials within circular textile systems.
2. The topic is of broad interest because it sits at the interface of green solvents, waste valorization, and biomaterials, directly addressing key UN SDGs and the urgent need to decarbonize and detoxify the textile value chain while enabling new bio-based products from underused protein-rich waste streams.
3. This review emphasizes how IL- and DES-based recovery of silk- and wool-derived proteins enables the fabrication of advanced biomaterials, including robust films and membranes, porous scaffolds, injectable and self-healing hydrogels, nanofibers, and functional coatings for wound healing, tissue engineering, controlled release, cosmetic formulations, and separation or adsorption systems. By linking solvent selection, protein structure, and processing routes to end-use performance, it shows how textile protein waste can be repositioned as a versatile feedstock for high-value, low-toxicity biomaterials within a circular textile framework.

1. Sustainability, circular economy, and waste valorization

The textile industry is a fundamental pillar of the global economy, playing a crucial role in clothing production, which is considered the second most basic human need. However, it is also one of the most environmentally damaging industries,

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second only to the oil sector.[1] Textile production is responsible for approximately 1.2 billion tons of greenhouse gas emissions annually, and projections suggest that by 2050, the fashion industry could consume up to 25% of the global carbon budget. This alarming scenario underscores the urgent need to adopt sustainable practices and circular economic models to mitigate environmental impacts and promote balanced development.[2,3]

The current clothing system, encompassing the manufacture, distribution, and use of garments, operates predominantly linearly.[4] More than 68% of today's fibers are derived from non-renewable resources, such as fossil fuels, to produce clothing through highly polluting processes. Additionally, synthetic fibers are non-biodegradable, and due to their microscopic size, synthetic fibers pose an even greater environmental risk than larger plastic waste.[5]

Fast fashion accelerates turnover and waste generation; however, sector unsustainability is fundamentally determined by life-cycle burdens and design choices.[6] While fast fashion undeniably increases product throughput and shortens use-phase duration, a rigorous sustainability approach should encompass the full textile life cycle and, critically, the design stage, which sets the boundary conditions for circularity.[7] Moreover, textile fibres are seldom composed of chemically pure polymers; instead, textile materials contain deliberately incorporated additives and applied finishing chemistries, such as stabilizers, pigments, and processing auxiliaries, that modulate service-life performance, while increasing chemical heterogeneity and thereby complicating sorting, recycling-process compatibility, and robust environmental assessment.[8] Accordingly, sustainable transition strategies must integrate life-cycle thinking with design for recyclability, including the conception of fibre systems and formulations that remain compatible with circular processing rather than being optimized solely for short-term cost and functionality.[2] Decisions on fibre selection, material complexity (mono-material *versus* blends), and design for fractionation largely determine whether garments can be effectively sorted and routed to high-value recovery pathways rather than disposal. In this context, fibre selection should be assessed not only by performance metrics but also by criteria such as environmental impact profiles, production volumes, and end-of-life compatibility.[6]

Within this system-level perspective, fast fashion business models intensify environmental burdens by promoting excessive consumption and accelerating the generation of heterogeneous waste streams. Disposable garments frequently incorporate synthetic fibres and blended constructions, which are comparatively more challenging to recover and therefore are often diverted to landfill or incineration.[3,8] Mixed-fibre waste streams, in particular, present significant technical and economic barriers because end-of-life management typically requires multiple, tightly coupled operations, which can translate into higher process complexity, environmental footprint, and costs. As a result, it is crucial to explore alternative recycling methods to convert textile waste into valuable resources. Developing innovative fiber recovery and repurposing solutions is essential, particularly when conventional recycling routes prove economically and environmentally unsustainable.[9,10]

Sustainability is a broad concept that refers to meeting present needs while ensuring that future generations can meet their own without compromise.[11] Within the textile industry, sustainability involves implementing practices that reduce environmental harm, promote social equity, and ensure economic viability.[12] The UN's 2030 Agenda, with its 17 Sustainable Development Goals (SDGs), provides a global framework to guide actions toward sustainability. Several SDGs are directly relevant to the textile sector, including Goal 8 (Decent Work and Economic Growth), which promotes fair and safe working conditions within the textile supply chain; Goal 9 (Industry, Innovation, and Infrastructure), which encourages the application of circular economy principles and the implementation of clean and efficient processes to increase resource efficiency and reduce dependence on virgin materials; Goal 12 (Responsible Consumption and Production), which aims to establish sustainable production and consumption patterns, reducing waste and resource depletion; and Goal 13 (Climate Action), which focuses on combating climate change, encouraging industries to reduce carbon emissions (Fig. 1). The textile industry can directly contribute to achieving these goals by adopting sustainable practices. For instance, reducing material waste, utilizing renewable energy sources, and implementing more efficient production processes may align the textile industry with the principles of the 2030 Agenda.[13,14]

In this context, replacing fossil-based textiles (*e.g.*, polyester, nylon and acrylic fibers) with protein-based alternatives (such as wool, silk and emerging regenerated protein fibers) represents an important lever for reducing the dependence on non-renewable resources and potentially lowering the carbon footprint of textile value chains. Protein-based textiles are derived from renewable feedstocks and can be designed for

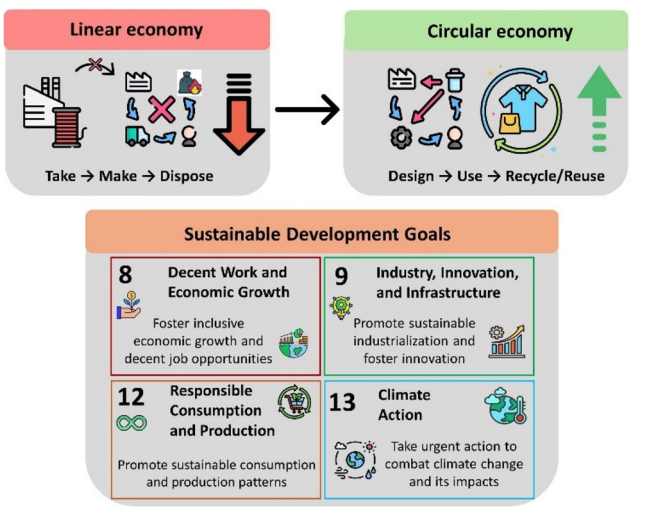

Fig. 1 Transition from linear to circular economy and the sustainable development goals (SDGs) related with the textile industry.

enhanced biodegradability and lower persistence in the environment, which aligns with the objectives of SDGs. However, this transition alone is not sufficient to ensure sustainability: protein-based materials must be integrated into circular strategies that prioritize durability, repair, reuse and high-value recycling routes, thereby closing the loop and preventing the emergence of a new linear bio-based model.[13,15]

The circular economy has emerged as a must to the traditional linear economic model, which follows the "take-make-dispose" pattern. The linear model is inherently unsustainable, as it relies on the continuous processing of natural resources and generates significant waste. In contrast, the circular economy proposes a closed-loop system where materials are continuously reused, recycled, and reintegrated into the production cycle, thereby minimizing waste and reducing the need for new resource extraction (Fig. 1).[16] Currently, the global economy is less than 10% circular, leading to a large amount of valuable waste.[17] Adopting a circular economy in the textile industry has been extensively promoted as an effective solution to mitigate the negative environmental impacts associated with fashion production and consumption.[18] Although this concept continues to evolve, it emphasizes optimizing resource utilization to avoid obsolescence. The key objective is implementing innovative and sustainable strategies for managing natural resources and waste, fostering a culture of reuse, sharing, repair, refurbishment, and recycling.[19]

Based on the exposed, this review adopts a focused scope on the valorization of protein-containing textile waste into biomaterials while addressing sustainable strategies centered on the recovery and transformation of silk- and wool-derived proteins using ionic liquids (ILs) and deep eutectic solvents (DESs). The rationale for this focus is that fibroin-, sericin-, and keratin-based materials offer high value-added potential with stringent technical requirements in biomedical, cosmetic, and advanced material applications. In response to the environmental burden of conventional textile-waste disposal, the review discusses selective dissolution, regeneration, and functionalization pathways designed to reduce reliance on hazardous reagents while improving process circularity. Recent advances in enzymatic hydrolysis, solvent-mediated fractionation, and biopolymer modification are critically examined, with route assessment framed not only by recovery feasibility but also by product qualification criteria, contaminant control, and compatibility with circular manufacturing schemes. Finally, this review provides valuable insights for researchers and industrial professionals on recent advancements in the upcycling and sustainable valorization of textile industry proteins.

2. Waste from the textile industry

The textile industry is one of the world's most polluting and resource-intensive sectors. Its linear model, featured by excessive raw material consumption, large-scale waste production, and inadequate disposal methods, has led to significant environmental challenges.[20] As a response, the concept of value-added waste has emerged as a crucial factor in advancing sustainable textile practices. Value-added waste is transforming textile waste into products with greater economic, environmental, or functional value. By leveraging circular economy principles, innovative recycling technologies, and sustainable material alternatives, textile waste can be reintegrated into the production cycle, thereby mitigating pollution and promoting economic benefits.[3,21] Textile waste can be classified into two main categories: pre-consumer and post-consumer. Pre-consumer waste includes materials discarded during manufacturing, such as fabric scraps, defective textiles, and surplus production, which are generally easier to recycle due to their controlled composition. In contrast, post-consumer waste arises from changing consumer preferences, fashion trends, and the natural wear and tear of clothing. Post-consumer waste represents the largest volume among these categories and poses the highest disposal challenges due to its heterogeneous composition. Managing this waste largely depends on consumers, further complicating its effective recovery.[8,22,23]

Given this scenario, two fundamental concepts have been developed for textile waste recovery: upcycling and downcycling. Upcycling refers to transforming textile waste into new products of higher quality and added value while preserving as much of the material's original characteristics as possible. This method reduces the need for new raw materials, extends product life cycles, and has been widely adopted in sustainable fashion and innovative textile design. In contrast, downcycling involves converting textile waste into lower-value products, often degrading the quality of the original material. While downcycling does not maintain the same standard as the original product, it still plays a role in waste reduction. It can be applied in industries such as construction, where textile waste is repurposed into thermal and acoustic insulation materials.[24,25] Considering the need for efficient textile waste recovery strategies, it is essential to account for the specific characteristics of each fiber type, as their properties directly influence recycling feasibility, reuse potential, and overall environmental impact.[26,27] Consequently, different types of fibers present distinct challenges and opportunities in textile waste management. According to a report by Textile Exchange, approximately 124 million tons of textile fibers were produced in 2023. Of this total, polyester accounted for 52%, followed by cotton with approximately 24%, while wool and silk together represented 12%.[28,29] In addition to protein-based waste, the first two classes are briefly discussed below due to their relevance.

2.1 Cotton waste

Cotton is one of the textile industry's most widely used natural fibers due to its softness, breathability, and high moisture absorption capacity. Additionally, its biodegradability makes it a more sustainable option compared to synthetic fibers derived from fossil fuels.[30] While mechanical recycling of

cotton is feasible, it results in fiber degradation due to length reduction, which affects its strength and durability for new textile applications. Consequently, recycled cotton fibers are generally used to produce lower-value products such as thermal insulation, upholstery, and non-woven materials.[31] Textile waste composed of cotton fibers has a high cellulose content, ranging from 88% to 96%, which allows it to be a valuable source of raw material for various industrial applications.[32] Cellulose, a renewable and biodegradable biopolymer, has gained increasing interest in producing biofuels, bio-based chemicals, and biomaterials.[33] Compared to other biomass sources, such as agricultural and wood waste, cotton-based textile waste offers significant advantages for biorefineries, as it contains a high proportion of relatively pure cellulose with low amounts of hemicellulose and lignin. Traditional lignocellulosic biomass, such as sugarcane bagasse and wood waste, typically consists of 35–50% cellulose, 20–35% hemicellulose, and 10–25% lignin.[34] The presence of lignin and hemicellulose in these materials requires an efficient process to separate the components before converting cellulose into value-added products. In contrast, cotton textile waste requires less intensive cellulose extraction and conversion pretreatment, which has become a promising alternative for biorefinery applications.[35]

2.2 Polyester waste

Polyester is one of the most widely used synthetic fibers in the textile industry due to its exceptional mechanical properties, cost-effectiveness, and versatility. Its popularity stems from its high tensile strength, durability, wrinkle resistance, and dimensional stability, contributing to its wide application, such as apparel, home textiles, and industrial fabrics. Polyester's hydrophobic nature is attributed to moisture-wicking properties, meaning it is preferred for sportswear and outdoor gear. Furthermore, its resistance to chemical degradation, UV radiation, and abrasion extends the lifespan of products, reducing the need for frequent replacement.[31]

Polyester's structural integrity and thermal stability are also unique features that render suitable for high-performance textiles, such as fire-resistant fabrics, medical textiles, and geotextiles used in infrastructure projects.[36] However, its extensive use comes with significant environmental concerns. Polyester production relies heavily on non-renewable fossil fuels, mainly crude oil, contributing to high carbon emissions and resource depletion. Moreover, the chemical structure of polyester, composed of highly stable ester bonds, exhibits an extreme resistance to microbial degradation, resulting in its persistence in terrestrial and aquatic environments.[37]

One of the most pressing issues associated with polyester textile waste is its low biodegradability, which leads to long-term accumulation in landfills and natural ecosystems. Studies indicate that polyester can take hundreds of years to degrade, releasing microplastics and toxic by-products into the environment. In addition to microplastic pollution, the incineration of polyester waste, which is often employed as a disposal method, generates hazardous emissions, including volatile organic compounds and greenhouse gases. These emissions exacerbate climate change and air pollution, further intensifying the environmental burden of polyester waste.[25,31] Polyester's versatility, resistance, and adaptability establish it as a fundamental pillar of modern textiles. As industry continues to evolve, innovations in polyester fiber engineering, recycling technologies, and alternative production methods have been studied to strengthen further its role as a high-performance and sustainable material in the global textile market.[31]

2.3 Polycotton blend waste

Blended textiles have become a key material class in modern apparel and home textiles, emerging from a design paradigm in which functionality is engineered by combining polymers with complementary physical and chemical behaviours.[25] This relevance is enhanced by the global profile of textile fiber production, in which polyester and cotton dominate market participation, resulting in a large global stock of textile products in which blends are structurally viable and economically attractive.[38] In this context, cotton and polyester blends (polycotton) represent one of the most widely used combinations, combining the intrinsic comfort attributes of cellulose fibers with the durability and shape retention properties typical of synthetic polyesters.[39]

Polycotton performance is not a simple additive outcome but rather the result of interactions between fiber chemistry, microstructure, and fabric structure. Cotton is valued for its softness, breathability, and high moisture absorption capacity, features that are derived from its hydrophilic cellulose structure rich in hydrogen bonds.[40] In contrast, polyester has high strength, durability, wrinkle resistance, and dimensional stability, as well as a predominantly hydrophobic nature associated with moisture absorption capacity and higher resistance to chemical and environmental agents.[34] Blending these fibers enables manufacturers to tune hand feel, comfort, dimensional stability, drying behavior, and service life, while maintaining processability and cost-effectiveness across broad product categories. However, the multi-material design strategy that enables polycotton's functional advantages also introduces intrinsic physicochemical heterogeneities, resulting in systematic trade-offs and vulnerabilities.[41]

By combining polymers with markedly different surface energies, swelling behaviours, and thermal/chemical stabilities, the blend can engender: (i) heterogeneous wetting and dye uptake profiles across fiber domains; (ii) increased sensitivity to finishing chemistries and auxiliaries that may preferentially adsorb onto one phase; and (iii) more complex aging pathways under use conditions.[42] At the waste stage, these intrinsic heterogeneities are compounded by post-consumer variability (finishes, dyes, and co-blends), resulting a feedstock that is chemically and structurally more resistant to selective separation than mono-material textiles. This complexity becomes especially critical when polycotton is represented in post-consumer waste streams. The textile system currently operates largely under a linear model with substantial dependence on fossil-derived fibers and significant environmental

burdens, which intensifies the urgency to manage end-of-life textiles as recoverable resources rather than disposable residues.[43]

Mixed-fiber waste has been recognized as a major bottleneck because treatment often requires multiple, complex steps and can be associated with a high environmental footprint and high costs, undermining both feasibility and scalability when compared to more straightforward mono-material streams.[25] In addition, synthetic components such as polyester are characterized by high persistence and low biodegradability, which contributes to long-term accumulation and broader environmental risks when disposal pathways dominate over recovery. Accordingly, polycotton constitutes a waste stream that is both abundant and intrinsically difficult to manage at end-of-life, reflecting its widespread use and multi-material design.[32,44]

3. Recycling textile waste

Within the context of the circular economy, textile waste recycling plays a crucial role by enabling the transformation of manufacturing and post-consumer waste into value-added products. For example, fabric waste can be recycled to produce new yarn or repurposed in building materials, packaging, and other applications. Additionally, the circular economy promotes sustainable design principles, emphasizing creating durable, repairable, and recyclable products.[45] Implementing circular economy strategies reduces environmental impact and generates economic benefits. The reuse of textile waste not only reduces the reliance on virgin raw materials but also tap into new market opportunities driven by growing consumer demand for environmentally responsible products. Furthermore, the circular economy can stimulate the creation of employment opportunities, particularly in sectors such as recycling and waste management.[7,46]

Despite its potential, the recovery of textile waste faces significant challenges. A major obstacle is the complexity of the textile industry's supply chain, which spans multiple sectors, from fiber production to post-consumer disposal. The lack of adequate infrastructure for collection, sorting, recycling, and management remains a significant barrier, particularly in developing countries.[47,48] Furthermore, the treatment process typically involves multiple complex steps, often associated with a high environmental footprint and substantial costs. As a result, it is crucial to explore alternative recycling methods to convert textile waste into valuable resources. Developing innovative fiber recovery and repurposing solutions is essential, particularly when conventional recycling routes prove economically and environmentally unsustainable.[47,48] According to Textile Exchange, a global organization monitoring the industry's environmental impact, post-consumer textile waste is becoming a valuable global commodity. In 2022, the European Environmental Agency reported that each person disposes of approximately 11 kilograms of post-consumer textile waste annually.[49] This issue is further exacerbated by findings from A New Textiles Economy, which reported that every minute, the equivalent of one truckload of clothing is sent to landfill or incineration, while less than 1% of post-consumer textile waste is recycled.[50] Overcoming existing barriers, such as the complexity of the supply chain and inadequate recycling infrastructure, requires a joint effort from everyone involved in the textile sector on a global scale. These challenges, however, also open space for innovation and collaborations. An integrated approach involving governments, industries, startups, non-governmental organizations, and consumers can facilitate the implementation of policies that encourage recycling and increased investment in infrastructure. These actions are essential for transforming the textiles industry into a circular and sustainable economy.

To effectively address the growing accumulation of post-consumer textile waste and promote a circular economy, developing advanced recycling strategies that enable the recovery and transformation of discarded materials into valuable resources is essential.[19] In this context, pretreatment methods are crucial in turning waste into value-added bioproducts. Pretreatment methods, such as enzymatic and chemical hydrolysis, are widely employed to break down cellulose from textile waste, facilitating its conversion into fermentable sugars that serve as precursors to produce bioethanol, bioplastics, organic acids, biomedical materials, and other value-added bioproducts.[30,44] Polyester waste can also be repurposed into bioplastics, such as polylactic acid, a biodegradable alternative to petroleum-based plastics.[34] Another innovative approach is upcycling, which involves transforming waste into high-value products.[29] These processes are essential for integrating textile waste into biorefinery systems, contributing to a more sustainable and circular economy (Fig. 2).[51]

Enzymatic hydrolysis is widely employed for the valorization of cellulosic fractions in textile waste since cellulolytic enzyme systems provide high reaction selectivity and enable saccharification under aqueous, near-neutral conditions, typically minimizing uncontrolled side reactions relative to strongly acidic or alkaline media.[52,53] Cellulase cocktails commonly comprise endoglucanases, exoglucanases (cellobiohy-

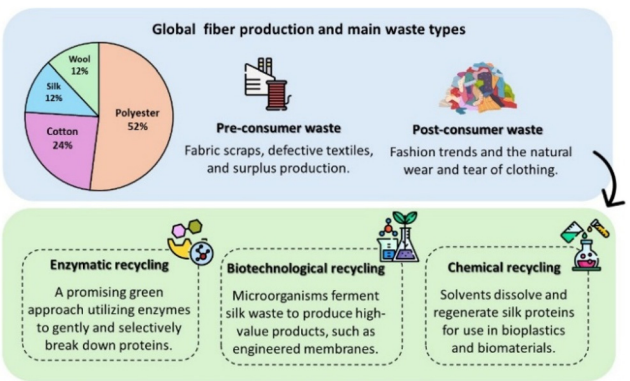

Fig. 2 Main global textile waste types and recycling routes to valorize textile-based waste.

drolases), and β-glucosidases acting synergistically; endoglucanases cleave internal β-1,4-glycosidic bonds (preferentially in amorphous regions), exoglucanases release cellobiose units from chain ends, and β-glucosidases convert cellobiose into glucose, completing the saccharification sequence. Thus, conversion is frequently constrained by cellulose crystallinity and limited substrate accessibility, while surface area and hornification also significantly influence the process; consequently, enzymatic implementations often require upstream conditioning to increase surface area and porosity and to facilitate enzyme penetration. In this context, elevated operating temperatures and/or solvent-mediated swelling treatments are commonly employed to improve chain mobility and enzyme access to less accessible domains, thereby increasing overall saccharification efficiency; however, these requirements can increase energy demand and additional solvent recovery requirements, which can undermine the definition of enzymatic routes as inherently environmentally benign. Therefore, the process intensity of enzymatic hydrolysis is determined not only by the biocatalytic step itself but also by pretreatment severity, residence time, and enzyme loading, which collectively govern energy demand and overall resource use. Furthermore, enzymatic hydrolysis often requires long processing times, frequently on the order of days, and its economic viability is further challenged by the high cost of enzyme formulations; moreover, hydrolysis efficiency is strongly dependent on feedstock purity and on substrate physicochemical properties, which directly affect enzyme accessibility and catalytic performance.[40,54,55]

Chemical hydrolysis offers an alternative depolymerization route that can achieve rapid cleavage of glycosidic bonds, but reported operating windows span a broad range of reagent concentrations and thermal conditions and typically impose additional downstream requirements.[56] Acid hydrolysis commonly relies on strong mineral acids and can be conducted either under dilute-acid, high-temperature conditions or under concentrated-acid, lower-temperature regimes, potentially resulting in the formation of secondary by-products; the latter may increase sugar yields but introduces acid recovery and neutralization demands. Alkaline hydrolysis using bases such as NaOH, $Ca(OH)_2$, or ammonia can disrupt lignocellulosic structures and enhance cellulose digestibility, and alkaline pretreatment is frequently integrated to reduce crystallinity and improve subsequent enzymatic saccharification. However, chemical hydrolysis can generate inhibitory side products, including furfural, hydroxymethylfurfural and phenolic derivatives, which may interfere with downstream bioconversion and often require neutralization, detoxification, and separation steps to improve sugar purity and process compatibility.[40,57]

It can also be repurposed into high-performance materials through advanced processing techniques to further enhance the value of recovered cellulose beyond its conversion into fermentable sugars. The extracted cellulose can produce nanocellulose, a material with high technological potential for applications in biodegradable films, high-strength composites, and biomedical devices.[58,59] Recently, cellulose residues have been used to manufacture artificial cellulose fibers, such as viscose, modal, and lyocell. Haslinger *et al.*[25] demonstrated that chemical recycling of pre- and post-consumer dyed cotton waste into new artificial cellulosic fibers, can retain color and exhibit superior properties to commercial viscose and lyocell. In addition, high-quality activated carbon, suitable for various adsorbent applications, has been successfully produced from cotton biomass waste materials.[33]

Regarding the recycling of synthetic materials, distinct strategies are used to ensure their reintegration into the production cycle. Currently, there are two primary methods for recycling polyester: mechanical and chemical. Mechanical recycling involves crushing, melting, and re-extruding polyester, converting it into new fibers. This method is extensively used for recycling polyethylene terephthalate (PET) bottles into textiles; however, it has limitations when applied to textile waste, as thermal degradation reduces material quality, limiting its reuse in high-performance applications.[60–64] In contrast, chemical depolymerization has historically been adopted as a rapid and effective route for PET recycling because it cleaves polyester chains into reusable building blocks, most notably terephthalic acid (TPA) and ethylene glycol (EG), while also generating process-dependent intermediates such as bis(2-hydroxyethyl) terephthalate (BHET) and dimethyl terephthalate (DMT). However, these processes are frequently conducted under relatively severe operating windows (elevated temperature/pressure and/or strongly acidic or basic media), which can impose environmental and economic burdens; accordingly, alternative catalyst/solvent systems, including deep eutectic solvents (DESs) and ionic liquids (ILs), have been explored as more sustainable catalytic platforms capable of delivering monomer streams suitable for repolymerization into fibers with properties comparable to virgin material.[65–67]

The major chemical depolymerization routes for polyester comprise hydrolysis, glycolysis, methanolysis (alcoholysis), and aminolysis. Hydrolysis can be conducted under acidic, alkaline, neutral, or enzyme-assisted conditions and, when appropriately tuned, may enable selective polyester degradation within mixed-fibre matrices. More specifically, methanolysis typically yields DMT and EG, glycolysis produces BHET, and hydrolysis cleaves ester bonds to regenerate TPA and EG.[68,69]

Within chemical PET recycling, recent catalyst system design has also enabled lower-severity depolymerization with high monomer recovery. Recently, Zhou *et al.*[70] reported the use of an alkaline ethylene glycol route for selective PET depolymerization under relatively mild conditions, achieving high PET conversion and TPA yield with short reaction times. The process offers practical advantages *versus* conventional alkaline hydrolysis, including easier product separation, lower acid demand in downstream neutralization, and reduced effluent burden. The catalytic medium was reused across multiple cycles, and larger-scale tests with real dyed textiles maintained high performance, high-purity TPA recovery, and selectivity in mixed-fiber systems. Overall, the route is promising for textile PET circularity, with remaining scale-up priorities in water-

balance control, high-solids handling, and continuous colorant management.

Biological depolymerization has emerged as a promising, lower-severity route for polyester recycling, relying on microbial, and more specifically enzymatic, ester-bond hydrolysis catalyzed by PET-active biocatalysts such as cutinases, esterases, lipases, and PETases, which can convert PET into recoverable monomers and intermediates. In many cases, enzymatic performance is enhanced through pretreatments intended to increase fibre-surface hydrophilicity and improve enzyme access; nevertheless, biocatalytic action is often insufficient to disrupt the macroscopic fabric structure without additional process intensification.[71–74] A notable advance was the identification of *Ideonella sakaiensis* 201-F6 by Yoshida *et al.*,[75] which can grow on PET at moderate temperature (30 °C) *via* a coordinated PETase/MHETase system, which PETase hydrolyzes PET to release MHET and EG, and MHETase subsequently converts MHET to TPA and EG. Taniguchi *et al.*[76] highlighted that the structural analysis of PETase revealed unique characteristics that may contribute to its superior catalytic performance. These features include an additional disulfide bond between C203 and C239 and a canonical α/β hydrolase fold containing a conserved catalytic triad composed of S160, H237, and D206. Importantly, enzymatic PET depolymerization is strongly governed by polymer morphology, with higher rates generally observed near or above PET's glass transition temperature in water (T_g > 60 °C), where increased chain mobility enhances ester-linkage accessibility; however, crystallinity remains a dominant kinetic barrier because enzymes preferentially hydrolyse amorphous domains.[77]

A critical issue in enzymatic recycling is the crystallinity of the polymer. It is well known that enzymes perform more efficiently in the amorphous regions of a material, as these areas exhibit higher chain mobility compared to crystalline structures. Consequently, enzymatic depolymerization is generally more effective at temperatures above PET's glass transition temperature (T_g), which is >60 °C in water.[55,69] Under these conditions, the degree of crystallinity decreases due to the greater flexibility of the polymer chains, exposing ester bonds to enzymatic attack. Nevertheless, several challenges remain, such as the low catalytic efficiency of natural enzymes and the necessity of enzyme engineering to enhance degradation rates.[36,78]

In addition to fiber recycling, PET can be valorized through integrated depolymerization-upcycling schemes in which PET provides a monomer stream that can be routed to bio-upcycling processes yielding value-added products, including polyhydroxyalkanoates (PHAs) and a range of aromatic and platform molecules such as vanillin/vanillic acid, catechol, and muonic acid, among others, thereby shifting the objective from polymer circularity alone to broader chemical circularity.[79] Furthermore, thermochemical conversion routes such as pyrolysis can generate aromatic-rich fractions containing TPA and oligomers, which may be further processed to enable downstream biological or chemical upgrading; remarkably, hydrolytic pyrolysis has been reported to yield a solid fraction enriched in TPA alongside oligomeric species.[80]

Another major challenge in textile waste recycling is the presence of mixed fabrics, particularly cotton-polyester blends. These mixtures are widely used in the textile industry due to the combined advantages of both fibers: cotton provides comfort and high moisture absorption, while polyester enhances durability, wrinkle resistance, and dimensional stability. However, conventional recycling methods do not allow selective recovery of individual fibers without significant degradation. Given this challenge, researchers and industry experts have conducted numerous studies to develop innovative technologies that enable the recovery of mixed textile waste through upcycling and more efficient strategies for the sustainable management and reintegration of these materials into the production chain. For instance, Yousef *et al.*[42] demonstrated that recovering cotton and polyester fibers from used jeans could result in economic returns of up to US$1629 per ton while reducing the carbon footprint by 1440 kg CO_2-eq per t of waste. Additionally, Maciel *et al.*[59] investigated the potential of recovering cotton fiber waste to produce cellulose nanocrystals, which are promising for biomedical applications.

4. Valorization of protein-based textile waste

In addition to the other types of textile waste, recycling protein-based textile waste represents a strategic approach to promoting sustainability in the textile industry and reducing the environmental impact associated with the inappropriate disposal of these materials. Among the most relevant protein-based textile streams are silk- and wool-derived materials, alongside other keratin-rich animal hair fibers, whose intrinsic properties and biochemical compositions give them considerable potential for generating new products with high value. The following will detail the main characteristics, recovery methods, and applications associated with recovering silk and wool.

Protein-based textiles comprise fibres whose load-bearing components are structural proteins, and they originate primarily from animal and insect sources. In the apparel and technical-textile landscape, the most relevant proteinaceous fibres comprise sheep wool and other keratin-rich animal hair fibres (cashmere, mohair, alpaca, and angora) sourced from mammalian fleece or hair, as well as silk fibres produced by insects, predominantly silkworms, and, more rarely, by arachnids.[81,82] Within this class, wool-based streams dominate in terms of mass throughput, with global wool generation estimated at approximately 2 million tons per year, providing abundant keratin-containing feedstock for recovery and upgrading.[83] In contrast, silk occupies a smaller-volume niche but remains highly value-dense, with global raw silk production on the order of 92 000 tons. Along the production chain, silk generates distinct waste fractions (cocoon waste, yarn/fabric scraps, and degumming residues), which differ

markedly in protein composition and downstream processing requirements.[84]

4.1 Silk recycling

Silk is highly valued for its mechanical properties, typical shine, softness, and excellent biocompatibility. Produced primarily by the silkworm *Bombyx mori*, silk consists of two structural proteins: fibroin and sericin.[85,86] Fibroin accounts for 70–80% of the total weight of silk and is responsible for its mechanical strength and structural stability, while sericin comprises 20–30%, acting as a protective adhesive layer around the fibroin. This composition gives unique attributes to silk, including high tensile strength, flexibility, and thermoregulatory properties, which explain its widespread use in the textile industry.[87] Silk waste can be classified into different categories based on its origin in the production chain: cocoon waste, which includes defective cocoons or those unsuitable for spinning; yarn and fabric waste, which consists of cutting scraps, broken yarns, and fabrics discarded due to defects or excess production; and degumming waste, in which sericin removed during the silk degumming process is often discarded by the textile industry, despite its potential for various industrial applications.[84,88]

Fibroin is a highly organized fibrous protein, with a molecular weight ranging from 200 to 350 kDa.[89] This protein has an ordered structure composed of antiparallel β-sheets stabilized by hydrogen bonds, which impart water insolubility and resistance to conventional organic solvents. However, this highly crystalline structure can be easily denatured by physical and chemical factors, such as high temperature, humidity, exposure to air, UV radiation, and pH variations, which may compromise its functional properties.[90,91] Fibroin comprises heterodimeric peptides connected by disulfide bonds and complexed with the small glycoprotein P25. In silk fibroin, there are twelve crystalline hydrophobic domains, with amino acids arranged in a repetitive sequence, predominantly consisting of approximately 43% glycine (Gly), 30% alanine (Ala), 12.2% serine (Ser), 4.8% tyrosine (Tyr), and 2.5% valine (Val).[92] Also, eleven non-crystalline hydrophilic regions contain glutamic acid, aspartic acid, arginine, and lysine in non-repetitive sequences. This protein exhibits two main structural conformations: (i) an amorphous and water-soluble secondary structure, primarily composed of random coils and α-helices; (ii) a crystalline structure, characterized by at least 55% antiparallel β-sheets, which confer high crystallinity, mechanical strength, and structural stability.[93,94]

Converting silk fibroin into a dry or crystalline powder facilitates storage, as the solid form is more stable than the liquid form. Consequently, fibroin is often processed into a solid state for later application in various fields, including biotechnology, biomedicine, and material science.[95] This structural protein is particularly notable for its biocompatibility, adjustable biodegradability, and mechanical strength, establishing it as a highly versatile functional biomaterial. Its broad range of applications includes tissue engineering, wound healing, controlled drug release, gene therapy, bone regeneration, and the production of biodegradable biofilms. Additionally, when properly purified and sterilized, fibroin demonstrates cytocompatibility comparable to conventional biomaterials, such as poly(lactic acid) and collagen. Complete removal of sericin is crucial to ensure that fibroin remains immunologically inert and maintains a low inflammatory potential, thereby expanding its suitability for biomedical applications.[96,97]

On the other hand, sericin is a hydrophilic globular protein highly soluble in water and primarily composed of polar amino acids, such as serine, aspartic acid, and threonine. Its molecular mass ranges from 20 to 400 kDa, depending on environmental conditions, the degree of hydrolysis, and the extraction method used. This broad variation in molecular weight endows sericin with versatile biochemical properties, making it a highly promising biopolymer for various industrial and biomedical applications.[85] Furthermore, its high solubility enables its use as a stabilizing agent, film-former, and surface modifier in different formulations, making it widely utilized in cosmetics, pharmaceuticals, biomedicine, and the food industry. Studies indicate that sericin exhibits antioxidant, anti-inflammatory, moisturizing, antimicrobial, and wound-healing properties, in addition to UV resistance, qualifying it as a valuable ingredient in skin care formulations, tissue regeneration, and advanced therapeutic systems.[98] Despite its beneficial properties, a significant portion of sericin is removed and discarded during the silk degumming process in the textile industry. This waste represents a considerable loss of a functional biopolymer that could be reutilized in high-value bioproducts.[97]

Silk recycling aims to recover and reuse its primary components, fibroin and sericin, through mechanical, chemical, biotechnological, and enzymatic approaches. Mechanical recycling involves re-spinning silk to produce new yarns and fabrics. This process allows silk to be reintroduced into the production chain, reducing the need for virgin biomass. However, fiber degradation during the process may compromise the final material's quality.[99,100] Another mechanical approach includes converting silk waste into short fibers for composite materials, structural reinforcements, and thermal and acoustic insulation products. Koçak *et al.*[101] employed the extrusion molding to prepare a composite using regenerated fibroin fibers and high-density polyethylene. Similarly, Govindaraju and Jagannathan[102] prepared composites from waste fibroin fibers and polypropylene in a 50 : 50 ratio, in which six fiber webs were hot-pressed into composites using a compression molding machine, achieving a tensile strength of up to 37.16 MPa.

Chemical recycling of silk employs solvents to dissolve fibroin and regenerate silk in various formats. Due to numerous hydrogen bonds in silk, its physical and chemical properties are highly stable, requiring effective dissolution strategies for processing. Controlled hydrolysis using acids or bases breaks fibroin into smaller peptides, which can be reused in biomaterials and bioplastics. Selective precipitation processes allow the formation of regenerated fibers with pro-

perties similar to natural silk.[103,104] Mollahosseini *et al.*[99] successfully recycled regenerated fibroin fibers using 9.3M LiBr aqueous solutions. Complete removal of Li^+ and Br^- ions from the fiber structure was ensured through bathing in Na_2SO_4/$(NH_4)_2SO_2$ (50/50 w/w) solutions, followed by treatment with a methanol/water solution (80/20 w/w). Furthermore, ultrafiltration and liquid-phase extraction techniques can be applied to purify sericin, enabling its use as a functional biopolymer in protective coatings, cosmetic formulations, and pharmaceutical products.[105]

Biotechnological recycling utilizes genetically modified microorganisms to produce biomaterials derived from fibroin and sericin. Microbial fermentation converts protein waste into substrates for biomaterial production, yielding high-value products such as membranes for tissue engineering and biodegradable nanocomposites. Sericin, rich in functional amino acids, can produce bio-based paints, bioactive adhesives, and controlled drug-release systems.[91,106]

Enzymatic recycling has emerged as an alternative approach, utilizing enzymes such as proteases, keratinases, and hydrolases to selectively degrade fibroin and sericin. Genetically modified enzymes have been explored to enhance silk degradation efficiency and optimize their conversion into bioactive peptides.[90,107] These treatments are typically implemented in aqueous media at mildly alkaline to near-neutral pH (pH 8–9) and moderate temperatures (50 °C). However, their performance should be discussed by jointly considering residence time and yield. Enzymatic degumming studies reported sericin removals of 18–24 wt% within 1 hour, increasing to nearly 28 wt% upon prolonged treatment, indicating that time-to-yield can be a dominant driver of overall process intensity.[108] In addition, enzyme loading strongly modulates outcome: in a comparative study, the sericin extraction rate enhanced from 4.3% to 19.7% as protease concentration increased from 0.1% to 2.0%, with no additional gains beyond this loading, indicative of enzyme saturation.[109] These quantitative trends are mechanistically coherent with silk ultrastructure, which fibroin contains highly ordered β-sheet-rich crystalline domains that restrict enzyme diffusion and limit access to susceptible peptide bonds, so effective conversion frequently depends on accessibility enhancing steps (controlled physical intensification, swelling/conditioning strategies, or assisted hydrolysis) and/or enzyme engineering to improve catalytic efficiency and stability, including genetically modified enzymes aimed at boosting peptide yields and selectivity toward bioactive sequences.[110] Emerging approaches such as nanotechnology-assisted hydrolysis and immobilized-enzyme systems have likewise been explored to improve operational stability and accelerate time-to-yield, enabling post-consumer silk waste and production scraps to be upgraded into new products without reliance on strongly aggressive chemical treatments.[111,112]

Silk proteins impose intrinsic recalcitrance to biocatalytic conversion because enzyme performance is governed not only by catalytic specificity but also by substrate ultrastructure and mass-transfer constraints. *Bombyx mori* silk fibroin exhibits a hierarchical organization in which β-sheet-rich crystalline nanodomains are embedded within less ordered regions; these tightly packed crystallites confer mechanical strength and chemical stability, but simultaneously reduce chain mobility, restrict diffusion, and limit exposure of hydrolysable peptide bonds to the enzyme active site.[87,95] Consequently, effective enzymatic processing frequently requires accessibility-enhancing conditioning before, or concomitant with, hydrolysis such as controlled swelling/partial disordering of hydrogen-bond networks using concentrated salt/chaotropic media and related dissolution, regeneration strategies, as well as physical intensification approaches that increase surface area and porosity.[103] Method comparison requires reporting operational windows and performance metrics that jointly include temperature, pH/medium, residence time, and yield/conversion, together with enzyme loading and any pretreatment and solvent-management requirements, since cumulative energy demand and resource intensity are functions of both operating severity and time-to-yield.[113]

4.2 Silk degumming

A comprehensive understanding of silk recycling processes also requires an in-depth examination of **degumming** techniques, as the removal of sericin plays a crucial role in determining the quality and functionality of regenerated fibroin (Fig. 3).

Chemical degumming methods are widely employed in the industry due to their high efficiency. Using alkaline agents, such as sodium carbonate and sodium bicarbonate, facilitates the effective removal of sericin in hot aqueous media, allowing for the purification of fibroin and sericin. Dou and Zuo[114] investigated the effect of Na_2CO_3 concentration on the structure of regenerated silk fibers and observed that a 5% sodium bicarbonate treatment completely removed the sericin. However, the degummed silk obtained was fragile and brittle, compromising its mechanical integrity. In addition, surfactants such as alkyl polyglycoside (APG) exhibited superior sericin removal efficiency compared to conventional degumming methods, minimizing fibroin degradation. Wang *et al.*[115] found that complete separation of sericin from fibroin could be achieved using a 0.25% APG aqueous solution for 30 min at 100 °C, with the solution boiled three times. This degumming process did not cause any breakage of the fibroin peptide chains. However, differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA) revealed that the recovered fibers showed reduced thermal stability, indicating potential alterations in the structural integrity of the fibroin.

Another chemical approach involves alkaline oxidation, utilizing oxidizing agents such as hydrogen peroxide to degrade sericin macromolecules into smaller units, thereby facilitating their dispersion in aqueous solutions. Although alkaline oxidation is highly effective in sericin removal, the severity of these treatments can adversely affect the mechanical properties of structural proteins, leading to fibroin brittleness and reduced elasticity. Furthermore, organic acids have also been investigated for silk degumming. Acetic acid and citric acid are

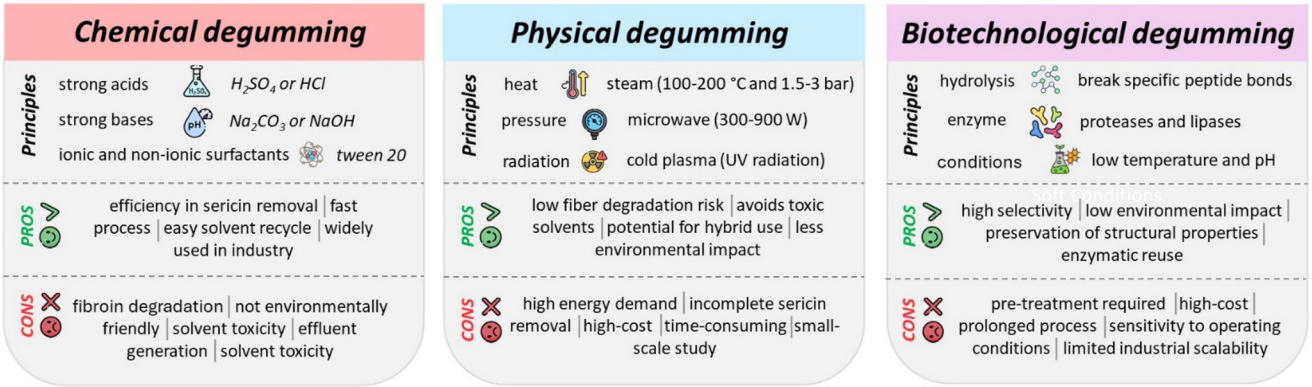

Fig. 3 Chemical, physical, and biotechnological silk degumming: each method's principles, advantages, and limitations.

widely used due to their capacity to selectively solubilize sericin, while preserving the structural integrity of fibroin. These mild acids reduce the proteins' electrostatic charge, facilitating their efficient removal without causing excessive fiber degradation. A comprehensive study conducted by Das *et al.*[116] evaluated the effectiveness of various organic acids, including citric, oxalic, malic, and succinic, classified into chloroacetic, dibasic, and hydroxycarboxylic acids.

The findings suggested that succinic acid exhibited one of the best performances in sericin removal while maintaining the mechanical strength of the fibers, followed closely by tartaric acid and monochloroacetic acid. Conversely, trichloroacetic acid emerged as the most potent compound among those analyzed, although it caused less weight loss in the fibers, suggesting a more controlled effect on sericin degradation.[116]

Physical degumming encompasses innovative techniques, such as pressurized steam treatment, which removes sericin by applying heat and humidity under controlled pressure, thereby reducing the excessive use of chemical reagents. High-temperature heat treatment has also been investigated as an alternative method, enabling the efficient removal of sericin without compromising the crystalline structure of fibroin. However, physical methods alone often require the addition of chemical or enzymatic agents to achieve satisfactory degumming efficiency. Moreover, these techniques are not well-suited for large-scale applications due to high energy consumption and potential damage to fibroin fibers caused by prolonged exposure to extreme conditions.[99,117] Emerging methods, such as microwave irradiation and low-temperature plasma, have demonstrated potential for the selective removal of sericin by modifying the chemical structure of the silk surface, thereby enhancing its affinity for dyes and functional coatings. Haggag *et al.*[118] conducted a comparative study on the degumming of virgin silk threads, utilizing both traditional and microwave radiation methods combined with various extraction agents, including mineral acid compounds, alkaline solutions, household soap, and commercial protease enzymes. The study revealed that protease enzymes were the most effective agents in both degumming processes. Experimental results indicated that the sericin extraction rate increased from 4.3% to 19.7% as the protease enzyme concentration increased from 0.1% to 2.0%. Furthermore, no additional improvement in sericin removal efficiency was observed beyond this enzyme concentration, suggesting a saturation effect in the enzymatic degumming process.

In **biotechnology degumming**, using of specific enzymes has emerged as an auspicious approach for the selective and environmentally sustainable degumming of silk. Among the main classes of enzymes utilized, proteases and hydrolases play a crucial role in the targeted degradation of sericin, ensuring the preservation of the crystalline structure of fibroin and maintaining the mechanical integrity of silk fibers. Additionally, the application of enzyme cocktails, combining proteases with cellulases or lipases, has been extensively studied to enhance process efficiency, reduce processing time, and minimize the generation of toxic waste. The incorporation of cellulases can improve sericin dispersion, facilitating its removal. At the same time, lipases assist in breaking down hydrophobic residues, resulting in a more homogeneous fiber that is better suited for subsequent processing stages.[108,119] Several studies have explored protease and lipase enzymes for silk degumming, offering an alternative to conventional chemical methods. Suwannaphan *et al.*[119] focused on a serine protease from a newly isolated *Bacillus* sp. for efficient silk degumming. The enzyme demonstrated high specificity towards sericin, ensuring complete removal while preserving fibroin integrity. The study also highlighted its ability to degrade sericin and bleach silk fibers simultaneously, making it a multifunctional alternative to traditional degumming processes.[119] Another work by Sumana *et al.*[109] suggested that alkaline proteases perform better than acid or neutral proteases, as they provide uniform sericin removal and improve silk quality. However, enzymatic degumming has some limitations, such as lower softness and increased bending stiffness compared to conventional chemical methods. Despite these challenges, enzymatic treatments significantly reduce water, energy, and chemical consumption and could become an attractive and sustainable alternative.

4.3 Wool

Wool is renowned for its unique properties, including moisture-wicking abilities and resistance. Primarily derived from the sheep's fleece, Merino wool stands out as one of the most popular breeds.[120] Annually, around 2 million tons of wool waste are produced worldwide,[121] with up to 95 wt% consisting of keratin, a fibrous structural protein. The remaining components are lipids, water, and ash.[122]

Keratin is the third most abundant biopolymer in nature, following cellulose and chitin, and can also be obtained from other sources, such as human hair and chicken feathers.[123–125] The protein's primary structure comprises around 19 amino acids, including cysteine, arginine, leucine, aspartic acid, glutamine acid, glycine, methionine, valine, and serine.[126] Keratin's binding motifs, such as arginine-glycine-aspartic acid (RGD) and leucine-aspartic acid-valine (LDV), confer attractive biological properties for keratin, promoting cellular adhesion and binding with other proteins.[127] The secondary structure of keratin is defined by the organization of ordered structures,[128] including α-helix and β-sheets.[129] α-keratin, which is found in mammals (*e.g.* wool and human hair), has a molecular weight of around 65 kDa,[130,131] while β-keratin, commonly found in birds and reptiles (*e.g.* chicken feathers), has a molecular weight of 10 kDa.[123,132] Keratin has high biocompatibility, low toxicity, antioxidant and anti-inflammatory effects, and excellent water-holding capacity.[133] These properties have inspired diverse applications, from biomedical applications[134,135] to food packaging,[136,137] and water treatment.[138,139]

Despite the increasing interest in recycling wool waste, the recovery and processing of keratin remains a challenge due to its low solubility in water and most organic solvents, which is attributed to the robust intermolecular bonds between polar and nonpolar amino acids, as well as its disulfide bonds (–S–S).[123] Several methods have been proposed for keratin recovery, such as acid/alkali hydrolysis, reduction, microwave, and enzymatic treatments.[140,141] However, most methods involve hazardous and toxic solvents, prolonged times, high temperatures, and/or high costs for industrial applications. Additionally, most of these techniques may have a negative environmental impact, are not cost-effective, and lead to potential protein degradation. Thus, developing more sustainable and cost-effective methods for keratin recovery from wool waste is crucial.[140,142]

Keratin recycling from wool waste involves various dissolution techniques, which can be divided into conventional and advanced approaches. Traditional methods for keratin recovery include alkaline/acid hydrolysis, oxidation, and reduction. In **alkaline hydrolysis**, strong basic chemicals (*e.g.*, sodium hydroxide) are used at high temperatures (120–170 °C) for prolonged times (up to 20 h). Subsequently, acids are added to neutralize the hydrolysate and precipitate keratin.[140,143] This process dissociates hydrogen from sulphate and carboxylic groups, yielding a soluble keratin fraction.[140] **Acid hydrolysis**, on the other hand, uses chemicals like hydrochloric acid, which break down disulfide and partial peptide bonds in keratin.[144] Compared to alkaline hydrolysis, acid hydrolysis can be more economically viable due to the lower cost of solvents.[140] However, acid hydrolysis often results in less effective biomass dissolution and lower protein yield.[140] For example, Zhang *et al.*[144] hydrolyzed wool using HCl (4 M) at 95 °C, achieving a hydrolysis yield of 33 wt%.[144] **Oxidizing** agents, such as peracetic acid ($C_2H_4O_3$), are often applied to dissolve keratin-rich biomass, oxidizing the disulfide linkages in keratin and converting them into hydrophilic pendant sulfonic groups on the cysteine amino acids.[145] However, this method also raises concerns about the toxicity of oxidizing agents and the low keratin yields. For instance, Shavandi *et al.*[146] oxidized wool using $C_2H_4O_3$ (2 w/v %) for 12 h and obtained only 5 wt% of keratin yield.[146] The **reduction method** involves agents such as L-cysteine, urea, and 2-mercaptoethanol,[140] breaking the disulfide bonds of keratin and allowing the solubilization of keratin molecules.[143] These conventional methods typically involve long dissolution times, high temperatures, and hazardous chemicals, with environmental concerns due to inadequate solvent disposal and recovery. Furthermore, they often result in low protein yields and potential protein degradation.[144,145]

Advanced techniques, such as superheated water, steam explosion, microwave-assisted, ultrasound-assisted, and enzyme-assisted dissolutions, offer potential solutions for higher keratin recovery yields in shorter times. In **superheated water hydrolysis**, high temperatures (around 170 °C) are commonly used for wool valorization. Bhavsar *et al.*[147] investigated the hydrolysis of wool keratin at 170 °C (7 bar pressure) for 1 h and achieved a keratin yield of 31 wt%.[147] This method offers an environmentally friendly alternative, using water as the solvent. However, the elevated temperatures can lead to protein degradation. **Steam explosion** technology involves applying high-temperature steam to biomass, forcing the steam into biomass tissues, followed by explosive decompression. This technique typically occurs at 140 °C to 230 °C for short intervals (1 to 10 min).[148,149] Xu *et al.*[150] treated wool using steam explosion technology and observed that wool's hydrophilicity, dissolving ability, and mechanical properties decreased as the steam explosion pressure increased, being attributed to the breakage of disulfide bridges and the destruction of the crystalline structure.[150] **Microwave-assisted technology** uses microwave energy to directly interact with biomass through molecular interactions with the electromagnetic field. The heating rate and the power absorbed by the sample depend on several factors, such as microwave power and sample geometry.[151] Dias *et al.*[126] used high-pressure microwave technology combined with food-grade acids to recover keratin from wool, achieving 45 wt% of yield.[126] **Ultrasound technology** enhances mass transfer through acoustic streaming and mechanical vibration, improving the keratin hydrolysis.[152] **Enzyme-assisted techniques**, such as using proteolytic enzymes (*e.g.* keratinases) or keratinolytic microorganisms, involve sulphitolysis (breakdown of disulphide bonds to cysteine and S-sulfocysteine), proteolysis (cleavage of peptide

bonds), and deamination (attacking insoluble keratinous materials, leading to proteolysis and the release of free ammonium). The efficiency of enzymatic hydrolysis depends upon substrate concentration, enzyme type and dosage, reaction time, and temperature.[140] Compared to thermal or chemical methods, enzymatic hydrolysis is considered more environmentally friendly. However, it requires long processing times and is expensive, posing a scale-up challenge.[140,153] In summary, advanced techniques offer advantages by minimizing the use of harmful chemicals, thus reducing the environmental impact. However, these methods have not been cost-effective enough, and further optimization is needed to maximize keratin recovery yields while preserving keratin's structural integrity.[152,154]

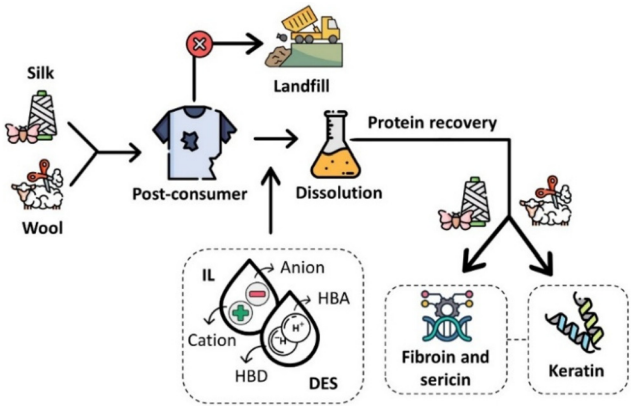

Fig. 4 Flowchart of protein dissolution and recovery from silk and wool textile waste using alternative solvents (ILs and DESs).

5. Protein dissolution in ionic liquids (ILs) and deep eutectic solvents (DESs)

Proteins are a group of biopolymers with a wide range of molecular, physicochemical, functional, and nutritional properties, resulting from differences in the number, type, and sequence of amino acids in their structure. Intra- and intermolecular interactions, such as hydrogen bonds, disulfide bonds, hydrophobic forces, and electrostatic interactions, control their secondary (α-helices, β-sheets), tertiary, and quaternary structures. A crucial feature of proteins is their solubility, which is strongly influenced by surface charge, polarity, hydrophobicity, isoelectric point, and environmental conditions such as pH, ionic strength, temperature, and especially the type of solvent.[155,156]

The permittivity of the solvent (ε), which refers to its ability to mediate electrostatic interactions, constitutes a determining parameter in the protein dissolution. Solvents with a permittivity lower than water's intensify electrostatic interactions (repulsive and attractive), which can induce the conformational unfolding of proteins. Such unfolding favors the exposure of hydrophobic domains, facilitating intermolecular interactions and protein precipitation. Therefore, water-miscible solvents favor the stabilization of water-soluble proteins through the formation of solvation layers; however, they can also increase the solubility of hydrophobic proteins.[156,157] Furthermore, additives such as salts (following the Hofmeister series) can promote either "salting-in" or "salting-out" of proteins, and surfactants frequently compromise protein structure and function, necessitating the development of more efficient alternatives. Thus, the appropriate choice of solvent is decisive for preserving or modulating the structure and functionality of proteins. In this context, ionic liquids (ILs) and deep eutectic solvents (DESs) have been highlighted as highly versatile solvents, capable of interacting in a controlled manner with proteins, providing advantages such as low volatility, high thermal stability, high solvation selectivity, and solvent recovery and reuse (Fig. 4).[158,159]

ILs are salts with an organic cation and organic or inorganic anions, thus having low lattice energies and low melting points compared to inorganic salts. These compounds are different from conventional salts due to their relatively low melting point, which is explained by the presence of ions with a large volume and an asymmetrical charge distribution, reducing the electrostatic forces between them and favoring the liquid state. As a result of their particular structure, ILs have remarkable properties, such as high thermal stability, extremely low vapor pressure, and excellent solvation capacity. One of their most relevant properties is their designer solvent capacity, enabled by the possibility of choosing suitable cation-anion combinations. This design flexibility has contributed to the continuous growth observed in IL applications, including protein dissolution, stabilization, extraction, and purification in ILs and their mixtures with molecular solvents.[160–162]

The process of protein dissolution in ILs occurs through multiple molecular interactions, mainly involving: (i) small anions, such as chloride and acetate, which promote the disruption of interactions between polypeptide chains due to their high nucleophilicity and ability to establish strong hydrogen bonds with protein functional groups, favoring the partial or total unfolding of the protein structure and exposing regions that become more accessible to solvation by ions; (ii) hydrophobic and large anions, such as $[NTf_2]^-$, tend to interact preferentially with hydrophobic regions of the protein structure; (iii) aromatic cations derived from imidazolium, such as $[C_2C_1im]^+$ and $[C_4C_1im]^+$, contribute to the stabilization of polar domains and control solvation dynamics through directional interactions; and (iv) modulation of the protein's surface charge through electrostatic forces established between cations and carboxylate groups of the amino acid side chains, reorganizing the solute-solvent interface.[163,164]

The interaction of ILs with proteins involves a combination of electrostatic forces, hydrogen bonds, and van der Waals interactions, which allow non-covalent (and, occasionally, covalent) interactions in the protein structure to be easily broken. IL cations are mostly chaotropic, while hydrophobic anions, such as $[OTf]^-$, $[NTf_2]^-$, and $[PF_6]^-$, play a relevant role

in protein stability *via* local hydrophobic interactions. Furthermore, increasing the hydrophobicity of ILs by extending their alkyl chains strongly influences their interactions with proteins. Thus, the role of hydrogen bonds in protein solubilization and stabilization emerges as a common denominator in the action of ILs.[156,165]

Another critical parameter is the system's temperature. Moderate temperatures, typically below 100 °C, are ideal for reducing the viscosity of ILs, enhancing molecular diffusion, and facilitating ion penetration into protein aggregates. While slightly higher temperatures can be controlled, prolonged exposure to elevated temperatures, especially above 120 °C, can compromise the thermal stability of ILs and induce peptide degradation, leading to irreversible denaturation and loss of protein functionality.[166] The concentration of IL also influences the efficiency of the dissolution process. Optimized solubility is observed in systems composed of pure IL or IL-water mixtures from 10% to 40%. However, very high IL concentrations can favor protein aggregation due to unbalanced electrostatic forces compromising conformational stability.[156,163]

Besides ILs, DESs have also been explored as sustainable alternative solvents due to their low toxicity and intrinsic biodegradability. DESs are homogeneous mixtures that combine two or more compounds, usually consisting of a hydrogen bond acceptor (HBA) and a donor (HBD). These components form intermolecular associations through hydrogen bonds, reducing the melting point of the system compared to its pure constituents due to charge delocalization facilitated by hydrogen bonding.[158,167,168] Moreover, the wide range of possible combinations between different HBDs and HBAs allows for the creation of diverse eutectic mixtures. This versatility is often referred to as solvent design, as the physicochemical properties of these solvents can be finely tuned by modifying their chemical structures. This tuning can be applied to the extraction process to modulate/design specific solvents to optimize the target product's separation efficiency. Parameters such as the molar ratio between the components, the water content (controlled hydration), and the operating temperature also directly impact the solvation efficiency, selectivity, and stability of the biomolecules during the process.[169–172]

In protein dissolution, DESs are believed to act similarly to osmolytes, organic solutes that stabilize the native conformation of proteins under stressful conditions. These compounds interact with the surface of proteins to preserve their structural integrity. The stabilization promoted by osmolytes occurs through a preferential exclusion mechanism, in which solutes are excluded from the immediate vicinity of the protein in solution, favoring its hydration and native structure. The DESs' interaction with proteins is mainly mediated by the preferential exclusion mechanism, in which the eutectic components are excluded from the immediate hydration layer of the protein, favoring its native structure and preventing conformational collapse. This mechanism is supported by solvophilicity and solvophobicity effects, as well as factors such as excluded volume and modulation of the surface tension of the medium. Solutes that increase the surface tension of the solution stabilize proteins, and *vice versa*, due to the preferential exclusion of solutes from the protein's surface.[158,173–175]

In the case of acidic DESs, applied to dissolve structural proteins such as fibroin and keratin, the process favors the controlled deconstruction of the main protein matrix, promoting the selective breaking of disulfide bonds between polypeptide chains. It is important to note that even with this destabilizing action of the protein network, the integrity of the secondary structures, such as β-sheets and α-helices, is often preserved, as demonstrated by spectroscopic (X-ray and circular dichroism) and thermal (DSC) analyses.[175,176] Additionally, parameters such as the DES acidity, the density of hydrogen bonds, the viscosity of the system, and the ability to selectively interact with polar or hydrophobic residues of the proteins are key to optimizing both the dissolution efficiency and the functional preservation of the proteins.[177,178] Therefore, the rational choice of DES composition and precise control of operating conditions are fundamental strategies for the success of the protein dissolution process in eutectic environments.

In the context of dissolving silk proteins, such as fibroin and sericin, and wool protein (keratin), ILs and DESs have demonstrated high efficiency. ILs such as 1-butyl-3-methylimidazolium acetate ($[C_4C_1im][C_1CO_2]$) and 1-ethyl-3-methylimidazolium chloride ($[C_2C_1im]Cl$) promote the selective degumming of sericin while preserving the β-lamellar regions of fibroin. Similarly, DESs based on cholinium : urea or cholinium : lactic acid facilitate sericin extraction with minimal structural degradation, maintaining the mechanical and functional integrity of the fibers. Additionally, the DES composed of L-cysteine : lactic acid promotes the controlled cleavage of disulfide bridges, enabling the extraction of soluble keratin without compromising its amino acid composition.[89,96]

Finally, the main advantage of protein extraction *via* ILs and DESs is preserving the biopolymers' microstructural properties during the dissolution process. Nevertheless, when dissolved in these alternative solvents, proteins can perform differently than in aqueous solutions. It is observed that incorrect folding and aggregation can occur due to the rearrangement of disulfide bonds and the exposure of hydrophobic surfaces during protein-IL and protein-DES interaction.[158]

6. Protein recovery from waste textiles using ILs and DESs

6.1 Fibroin and sericin from silk

The recovery of protein textile fibers using ILs and DESs represents an innovative and sustainable approach, which has been highlighted as a promising solution for the selective dissolution of silk and its reintegration into high-value applications.[179,180] Unlike conventional methods, which often involve harsh chemicals and lead to fiber degradation, ILs and DESs provide effective and selective dissolution, preserving protein structures while enabling efficient separation and puri-

fication. Both ILs and DESs interact with hydrogen bonds and hydrophobic domains of proteins, disrupting intermolecular forces while maintaining peptide chain integrity (Fig. 5).[181,182]

Recent studies have shown that ILs, such as $[C_4C_1im][C_1CO_2]$ and $[C_2C_1im]Cl$, along with cholinium chloride-based DESs, exhibit high efficiency in selectively dissolving silk sericin while preserving fibroin integrity.[176,183] The ability of these solvents to fine-tune dissolution conditions (pH, temperature, and ionic strength) enables ideal conditions for controlled protein recovery, reducing structural damage and ensuring better downstream processing. This process allows the regeneration of silk fibers for applications in biomedical materials, advanced composites, and biodegradable textiles.[184,185]

After IL/DES-assisted dissolution, protein recovery is typically implemented by phase inversion and selective separation. The most common workflow combines: (i) anti-solvent-induced precipitation/coagulation of fibroin or keratin (usually by controlled addition of water or alcohols), (ii) solid-liquid separation (filtration/centrifugation), and (iii) polishing of the protein-rich phase by dialysis or membrane processing (ultrafiltration/diafiltration) to reduce residual ionic species and low-molar-mass impurities before regeneration into films, gels, fibers, or scaffolds. In parallel, the IL/DES-rich mother liquor is conditioned for reuse by water/anti-solvent management, concentration adjustment, and solvent recovery; in several reported systems, solvent recovery and reuse are necessary process steps for technical and economic feasibility.[106,177,186,187]

In post-consumer streams, recovered protein phases may co-carry textile-origin impurities, including colorants, finishing residues, processing auxiliaries, and low-molar-mass extractables, which can compromise biocompatibility and batch reproducibility if not adequately controlled. Therefore, downstream qualification requires contaminant-oriented purification beyond bulk precipitation, combining staged washing, membrana polishing, precipitation with antisolvents, adsorption, and solvent-phase conditioning to reduce residual ionic species and non-protein contaminants to application-compatible levels.[85,188] For biomedical-grade positioning, regenerated fibroin/keratin materials should be discussed under a quality-by-design framework in which molecular, structural, biological, and process-chemistry attributes are jointly controlled. At the molecular-structural level, dissolution/regeneration in ILs or DESs can preserve amino-acid composition while inducing condition-dependent redistribution of secondary/supramolecular organization.[88,189] Biological qualification should include at least *in vitro* cytocompatibility and blood-contact screening. Recent studies[189] on IL-processed silk scaffolds have reported robust fibroblast/stem-cell adhesion and proliferation together with hemolysis rates below 2%, supporting their preliminary biomedical relevance. Nevertheless, comprehensive *in vivo* safety and performance assessments remain essential to meet regulatory requirements standards. Process-related quality control is equally critical; residual-solvent levels and solvent-recovery performance must be explicitly quantified, since ILs or DESs carryover into the recovered protein phase can occur and directly affect material purity.[189,190]

The degumming mechanism by ILs involves specific molecular interactions: the $[C_4C_1im]^+$ and $[C_2C_1im]^+$ cations interact with the oxygen atom of the carbonyl group (C═O) present in peptide bonds, while the hydroxyl anion (OH^-) attacks the carbon molecule of the peptide group, leading to the formation of a tetrahedral intermediate (Fig. 4). Then, the electrons of the oxygen bonded to the imidazolium group are rearranged, restoring the carbonyl (C═O) structure and promoting the cleavage of the C–N peptide bond. This bond cleavage occurs through the abstraction of a proton from HCl, forming an amine. During this process, the ILs can be efficiently recovered by abstracting protons from HCl, enabling their reuse within the system, thus enhancing the sustainability and cost-effectiveness of the degumming process.[185,191]

Chloride and acetate anions are important H-bond acceptors capable of breaking hydrogen bonds between backbone polypeptide chains, which play a fundamental role in the dissolution of silk fibroin. This mechanism facilitates the controlled breakdown of the protein structure while preserving its functional properties. Moreover, the application of ILs in protein dissolution holds promise, not only for improving the solubility but also for minimizing cytotoxicity and partially preserving the mechanical and structural integrity of proteins.[192,193]

In silk processing, DESs have demonstrated their efficiency in dissolving silk sericin while preserving the structural integrity of fibroin, thereby enabling the controlled extraction of valuable proteins with minimal degradation.[194] Recent studies have shown that cholinium chloride-based DESs, particularly those combined with organic acids or urea, facilitate the effective degumming of silk by disrupting hydrogen bonds within the sericin structure. DES micro-etching alters the fiber surface, exposing amino acid residues and increasing affinity for sericin dissolution. Additionally, the DES-treated silk demonstrates improved breathability, hydrophilicity, and biocompatibility, which expands its usability in advanced biomaterials.[187,195,196]

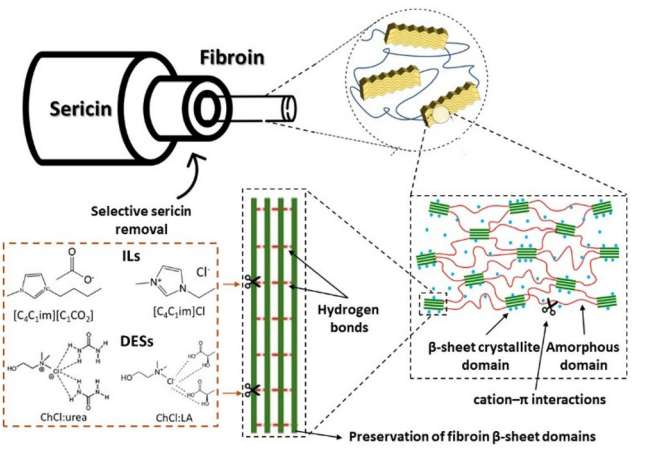

Fig. 5 Illustration of the application of alternative solvents for the selective sericin removal and efficient silk fibroin dissolution.

The degumming mechanism of DESs primarily involves the selective interaction of the solvent with peptide bonds and hydrogen bonds within the sericin matrix, allowing for the breakdown of sericin's polymeric structure without causing significant damage to fibroin. Deng *et al.*[176] evaluated the cholinium chloride (ChCl) and urea DES as an alternative degumming method. The ChCl : urea DES was able to remove sericin while selectively preserving the structural integrity of fibroin. The effectiveness of this DES is attributed to its ability to solubilize sericin by forming strong hydrogen bonds with its polar amino acid residues, thereby destabilizing the protein network and facilitating its selective extraction. Furthermore, the degumming efficiency remained stable after seven cycles, indicating the possibility of recycling and reusing the solvent. Compared to traditional alkaline treatments that lead to hydrolysis damage, DES degumming maintained the crystallinity and mechanical integrity of the fibroin, and the silk exhibited improved moisture absorption and dye absorption, which is crucial for textile applications.

Other studies, such as the one developed by Yang *et al.*,[197] demonstrated that the DES composed of cholinium chloride : acetate (ChCl : OA, 1 : 1 molar ratio) disrupts the hydrogen bonding networks within sericin, breaking its polymeric structure while keeping fibroin intact. The HBD and HBA species contributed to the selective dissolution by interacting with sericin's hydrophilic groups (*e.g.*, –OH, $–NH_2$, –COOH), leading to its removal from silk fibers. At the same time, fibroin's β-sheet structure remains stable due to its stronger intermolecular interactions, allowing for its efficient recovery.[197] Tan *et al.*[198] observed that DESs formed by cholinium chloride : lactic acid (ChCl : LA) and cholinium chloride : malic acid (ChCl : MA) disrupt internal hydrogen bonds and hydrophobic interactions within silk fibers, leading to selective swelling and loosening of the structure. This fact enables the protein's recovery without requiring harsh chemical treatments. Furthermore, the interactions between DES components and silk proteins prevent excessive degradation while facilitating selective separation of fibroin and sericin. Hu *et al.*[199] found that ChCl : LA showed high efficiency in selectively fragmenting intermolecular interactions, weakening amorphous regions without destroying the crystalline domains of antiparallel β-sheets. Lactic acid in the DES provides a slightly acidic environment, capable of protonating functional groups such as amine and carboxyl in fibroin, inducing electrostatic repulsion and reducing cohesion between polypeptide chains.

6.2 Keratin from wool

In response to the limitations of conventional and advanced methods for wool dissolution and keratin recovery, ILs and DESs have emerged as promising and effective alternatives. When properly designed, these solvents can effectively dissolve keratin-rich biomass, enable higher keratin recovery yields while contribute to the sustainability of the process, and maintaining the primary structure of keratin and its functional properties (Fig. 6).[151]

Polesca *et al.*[26] reported a study on keratin recovery from feathers using aqueous IL systems, where protein recovery was optimized by adding a coagulant (water, ethanol, or acetone), followed by centrifugation and washing of the solid phase to reduce IL residues. Aqueous IL $[C_4C_1im][C_1CO_2]$ (80 wt%) obtained the highest dissolution results at 100 °C for 4 h, and optimal precipitation occurred with ethanol, solution : coagulant ratio 1 : 2 (m/m), 5 °C, and 1 h, with keratin recovery of 90 wt%; IL recovery and reuse in four successive cycles with 95% solvent recovery was also demonstrated. In another study[200] with an aqueous solution of $[N_{111(2OH)}][C_1CO_2]$ at 80 wt%, dissolution was also carried out at 100 °C for 4 h, and the recovery step was optimized by response surface methodology, achieving 93 wt% keratin recovery under 20.25 wt% ethanol in water, 5 h, and a solution : coagulant ratio of 1 : 1.45 (m/m), with reuse of the IL for four cycles without significant loss of performance.

Molecular dynamics simulations by Zhang *et al.*[201] revealed that ILs interact with cysteine residues, facilitating the cleavage of disulfide bonds, a crucial step for keratin dissolution. Studies have shown that at least 65% of disulfide bonds must be cleaved to fully dissolve keratin, although lower cleavage percentages can compromise the mechanical properties of the

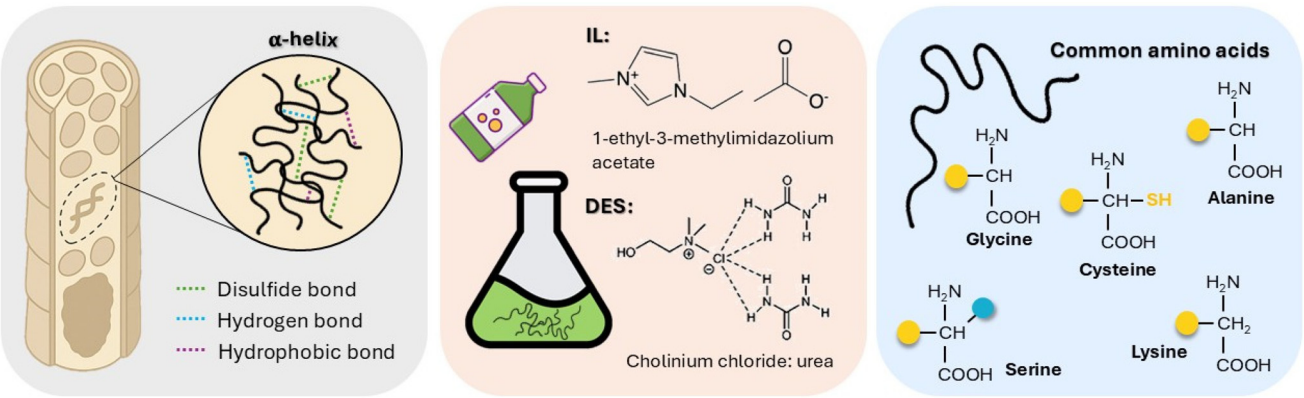

Fig. 6 Illustration of the application of alternative solvents for efficient wool keratin dissolution.

final product.[201] To investigate the effectiveness of different ILs, Liu *et al.*[202] evaluated 621 ILs for dissolving three keratin models using the COSMO-RS method and validated their predictions experimentally with wool. Among the ILs tested, 1-ethyl-3-methylimidazolium acetate ($[C_2C_1im][C_1CO_2]$) was identified as one of the most efficient to dissolve wool keratin, based on the logarithmic activity coefficient ($\ln y$) and the strong hydrogen bond acceptor ability from the acetate anion, a characteristic also confirmed experimentally at 120 °C.[203] Similarly, Qin *et al.*[204] evaluated 143 ILs for keratin dissolution, emphasizing the role of anion-protein interactions, with chloride, acetate, and diethyl phosphate (DEP) emerging as the most efficient anions.[204] These findings were supported by Zheng *et al.*,[205] who demonstrated that the nature of the anion can modulate the dissolution time in ILs. According to the authors' results, acetate, dimethylphosphate, and chloride provide faster and more effective keratin breakdown, since they are strong H-bond acceptors capable of breaking hydrogen bonds between peptide chains.[123,204,206]

While anions have the primary role in keratin dissolution, recent studies have highlighted the cooperative role of both cations and anions.[179] Imidazolium-based cations have demonstrated significant efficacy in keratin dissolution mechanisms compared to other cationic structures, such as tetrabutylammonium and tetrabutylphosphonium, due to their ability to form intramolecular hydrogen bonds with keratin molecules.[207] Zheng *et al.*[205] studied the effects of IL anion-cation combinations in wool keratin dissolution, revealing that the dissolution efficiency follows the order: imidazolium < phosphonium < ammonium < pyridinium. Interestingly, while the chemical structure of cations significantly affects the dissolution mechanism, the length of the side chains has only a minor impact. Temperature also plays a crucial role in modulating the cleavage of disulfide bonds of keratin. Ghosh *et al.*[208] demonstrated that increasing the temperature from 120 °C to 180 °C significantly reduced keratin yield (from 57% to 18%), since the excessive heating promotes the formation of water-soluble amino acids and low molecular weight peptides, eventually affecting protein properties. Thus, these studies highlight the need for controlled conditions to preserve the protein's integrity during processing.[205,208]

The use of DESs for wool dissolution has also gained attention in recent years due to their biodegradability, low cost, and low toxicity when properly designed. Cholinium chloride-based DESs have demonstrated efficacy in cleaving the disulfide bonds in keratin. For instance, Moore *et al.*[209] used cholinium chloride : urea (2 : 1 molar ratio) at 170 °C to dissolve wool, exploiting the well-established denaturant effect of urea and the hydrogen bond accepting properties of the DES.[209] Jiang *et al.*[206] dissolved wool using cholinium chloride : urea (1 : 2 mol mol^{-1}) at a temperature range of 110–130 °C for 5 h, demonstrating that increasing the temperature enhanced disulfide bond cleavage and promoted changes in the secondary structure of the protein, with increased β-sheet and reduced α-helix contents.[206]

Mouro *et al.*[210] evaluated the effectiveness of cholinium chloride : urea and L-cysteine : lactic acid for wool dissolution at 130 °C for 3 h. Their results indicate that L-cysteine : lactic acid has higher wool dissolution efficiency, achieving 68.83% wool dissolution, compared to cholinium chloride : urea, which achieved 42.88%. Such efficiency was attributed to the hydrogen bonding ability of the DES, leading to higher wool solubilization.[210] Further advancements by Okoro *et al.*[211] reported a keratin recovery of 93.77 wt% using L-cysteine : lactic acid under optimal conditions. The authors also observed changes in the secondary structure of keratin, with a decrease in the α-helix content as temperature increased.[211] Wang and Tang,[212] who used cholinium chloride : oxalic acid for wool dissolution, also observed, by XRD analysis and SDS-PAGE, that the α-helix structure was destroyed during the dissolution, and not regenerated again, resulting in small protein fragments. Nevertheless, despite the secondary structural changes, the amino acid composition was unaffected, ensuring the protein's properties and potential applications.[212]

7. Applications for recovered proteins

7.1 Fibroin and sericin from silk

The use of ILs (Table 1) and DESs (Table 2) in processing silk fibroin and sericin waste has significantly expanded the potential applications of these wastes, transforming them into a valuable resource for biomedical, electronic, packaging, and composite material industries (Fig. 7).[113] Processing based on ILs and DESs is used as a selective separation strategy; however, the native structure of the target proteins is not completely preserved. Dissolution proceeds through disruption and reorganization of non-covalent interactions, particularly hydrogen-bonding networks, which enables sericin extraction and fibroin recovery but does not maintain the original native three-dimensional structure after regeneration.[179] Under controlled solvent composition and operating conditions, fibroin recovery may occur with limited degradation in the main chain; however, regenerated materials typically exhibit condition-dependent structural redistribution, including changes in secondary-structure balance and supramolecular order. Across the available studies, reported preservation is hierarchy-dependent and supported by method-specific analytical evidence at the primary, secondary, or supramolecular structural level.[175]

One of the most prominent applications of green solvent-processed fibroin and sericin is in tissue engineering and regenerative medicine, where it serves as a biocompatible and biodegradable material for creating scaffolds, hydrogels, and bioinks. These applications have received increasing attention due to their cytocompatibility and biodegradability.[179,199]

Fibroin scaffolds prepared with ILs provide porous structures that emulate the extracellular matrix (ECM), allowing for efficient cell adhesion, proliferation, and differentiation. It is

Table 1 Innovative applications of biomaterials based on silk proteins and ionic liquids

Raw material	Biomaterial	Ionic liquid	Application	Ref.
Fibroin	Hydrogel	$[C_2C_1im][C_1CO_2]$	Biomedical materials	193
Fibroin	Hydrogel	$[C_2C_1im][C_1CO_2]$	Bioelectronics	213
Fibroin	Gel	$[C_2C_1im][C_1CO_2]$	Conductive gel, biomedical materials	214
Fibroin	Gel	$[C_2C_1im][C_1CO_2]$	Gel protective layer	215
Fibroin	Composite	$[C_2C_1im][C_1CO_2]$	Biomedical materials	183
Fibroin	Nanocarrier	$[C_2C_1im][C_1CO_2]$	Anticancer therapy	216
Fibroin	Hydrogel	$[C_3C_1im]Cl$	Water electrolysis	217
Fibroin	Scaffold	$[C_3C_1im]Cl$	Tissue engineering materials	218
Fibroin	Hydrogel	$[C_4C_1im][C_1CO_2]$	Tissue engineering materials	219
Fibroin	Scaffold	$[C_4C_1im][C_1CO_2]$	Tissue engineering scaffold	220
Fibroin	Composite	$[C_4C_1im][C_1CO_2]$	Engineering and biomedical materials	221
Fibroin	Film	$[C_4C_1im][C_1CO_2]$	Tissue engineering materials	222
Fibroin	Sponge	$[C_4C_1im][C_1CO_2]$	Cartilage-related biomedical	223
Fibroin	Scaffold	$[C_4C_1im]Cl$	Bone tissue engineering	93
Fibroin	Film	$[C_4C_1im]Cl$	Artificial skin coating	224
Fibroin	Microsphere	$[C_4C_1im]Cl$	Drug delivery	225
Fibroin	Film	$[C_4C_1im]Cl$	Coagulation material	213
Sericin	Scaffold	$[C_4C_1im]Cl$ and $[C_4C_1im][BF_4]$	Tissue engineering materials	226
Fibroin	Scaffold	$[C_4C_1im]Br$	Tissue engineering materials	227
Fibroin	Film	$[C_4C_1im][PF_6]$	Electrolyte film, energy materials	192
Fibroin	Ionogel	$[C_nC_1im]Cl$ (n = 2, 6, and 10)	Bioelectronics	96
Fibroin	Nanoparticle	$[C_nC_1im]Cl$ (n = 1, 2, 4, and 6)	Biomedical materials	228
Fibroin	Hydrogel	[im][SH]	Wound healing	229
Sericin	Hydrogel	[im][PNB]	Wound healing	230
Fibroin	Ionogel	[Ch] ($[C_1CO_2]$, [DHP], Cl)	Drug delivery	231
Fibroin	Film	[THEA][Lac]	Tissue engineering materials	186

[im] – imidazolium; [C2C_1im] – 1-ethyl-3-methylimidazolium; [C_3C_1im] – allyl-3-methylimidazolium; [C_4C_1im] – 1-buthyl-3-methylimidazolium; [Ch] – cholinium; [THEA] – triethylammonium; [C_1CO_2] – acetate; Cl – chloride; [SH] – sulfhydryl; [PNB] – phenylboronic; [DHP] – dihydrogen phosphate; Br – bromate; [BF_4] – tetrafluoroborate; [PF_6] – hexafluorophosphate; [Lac] – lactate.

Table 2 Innovative applications of biomaterials based on silk proteins and deep eutectic solvents

Raw material	Biomaterial	Deep eutectic solvent (molar ratio)	Application	Ref.
Sericin	Hydrogel	ChCl : urea (1 : 1)	Bacterial inhibition	176
Fibroin	Membrane	ChCl : urea (1 : 1)	Wastewater treatment	176
Fibroin	Membrane	ChCl : urea (1 : 1)	Bioelectronics	196
Sericin	Nanosheets	ChCl : $[C_1CO_2]$ (1 : 1)	Thermal management	197
Sericin	Nanostructure	ChCl : $[C_1CO_2]$ (1 : 1)	Biomedical electronic	232
Sericin	Gel	ChCl : Gly (1 : 2)	Biomaterials electronic	233
Fibroin	Scaffold	ChCl : LA (1 : 1)	Wound dressing	199
Fibroin	Membrane	ChCl : LA and ChCl : MA (1 : 1)	Biomaterials electronic	178
Fibroin	Membrane	GH : urea (1 : 2)	Energy storage	198
Fibroin	Aerogel	GH : urea (1 : 2)	Air purification	234
Fibroin	Scaffold	D-Sorbitol : L-proline (2 : 1)	Cooling materials	187
Fibroin	Film	LiBr : urea (1 : 1)	Anticoagulation materials	177

ChCl – cholinium chloride; GH – guanidine hydrochloride; LiBr – lithium bromide; Gly – glycerol; LA – lactic acid; MA – malonic acid; $[C_1CO_2]$ – acetate.

crucial for applications in bone, cartilage, and skin regeneration. These fibroin scaffolds have been widely used in wound healing because they retain moisture, promote tissue repair, and offer antibacterial properties.[85,107] Gonçalves *et al.*[220] developed biocompatible scaffolds for tissue engineering applications from silk fibroin using $[C_4C_1im][C_1CO_2]$. The produced scaffolds exhibited a high porosity of approximately 87.9%, an average pore size of 135.3 μm, and interconnectivity of 95.4%. Mechanical and swelling studies showed that the scaffolds maintained stable viscoelastic properties over a wide frequency range and demonstrated high swelling capacity, which is beneficial for tissue regeneration. Cytocompatibility tests using L929 fibroblast cells and human adipose-derived stem cells indicated excellent cell adhesion, proliferation, and viability. Furthermore, hemolysis assays confirmed that the scaffolds had hemolytic rates below 2%, rendering them suitable for biomedical applications.

Recently, some studies have evaluated the production of micro and nanofibrous scaffolds by dissolving fibroin or sericin in ILs or DESs. In the presence of solvents with polyelectrolyte behavior, it has been found that these parameters tend to influence the electrospinning process negatively since

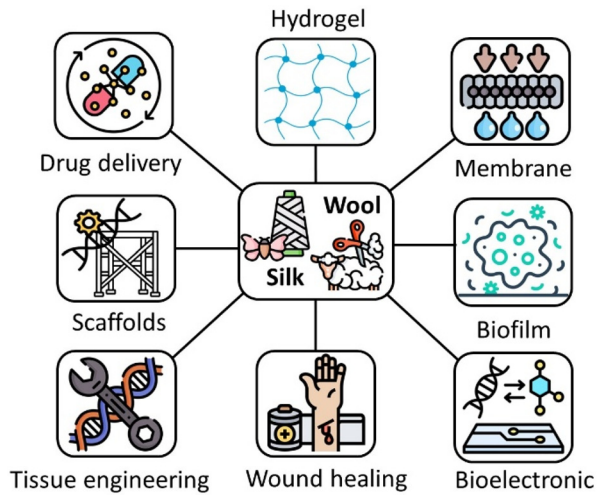

Fig. 7 Main applications for proteins recovered from silk and wool.

the solution's viscosity and the polymer's concentration become relevant in the movement of the charge. Furthermore, it was reported that conductivity and surface tension were essential factors in triggering mass transport by electrical forces to control fiber formation.[220,235] An interesting study by Rivera-Galletti *et al.*[183] presented the design of a film composed of silk fibroin and cellulose acetate, dissolved in $[C_2C_1im][C_1CO_2]$. The alteration in the protein's secondary structure induced a fluctuating conformational shift in cellulose at the microscopic level, leading to modifications in its crystalline structure, lower glass transition temperatures, and enhanced thermal stability compared to pure cellulose acetate. Silva *et al.*[219] prepared chitosan-silk fibroin hydrogels for tissue engineering using $[C_4C_1im][C_1CO_2]$. The *in vitro* results demonstrated that the hydrogels support the adhesion and proliferation of human dermal fibroblasts. Moreover, the hydrogels presented a microporous, lamellar structure and viscoelastic behavior.[219] One of the key applications of silk fibroin is regenerated materials, where IL-based processing procedures have been explored for regenerating animal proteins. For example, Goujon *et al.*[186] demonstrated that protic ILs (triethylammonium lactate) effectively coagulate silk fibroin, although some challenges remain in preserving its final chemical composition.

Further studies, such as the one developed by Li *et al.*[222] revealed that the silk protein-IL complex retained the α-helix secondary structure in the regenerated films, contributing to their enhanced tensile strength and superior resistance to ultraviolet radiation compared to films produced *via* conventional methods. This structural stability further expands their applicability in biomedical coatings, flexible electronic devices, and protective materials. Jia *et al.*[236] reported that, using cholinium-based IL, the random coil conformation rapidly transforms into a highly ordered β-sheet structure. This self-assembly behavior enables the development of structures with tailored physicochemical properties.

Seoud *et al.*[237] explored the dissolution of fibroin in IL, highlighting the importance of temperature in maintaining protein integrity during processing. This fact results from the ILs' ability to disrupt the naturally H-bonded fibroin network, which is sensitive to temperature. Additionally, Pereira *et al.*[238] used $[C_4C_1im]Cl$ to create mesoporous fibroin/silica hybrids *via* sol–gel techniques, demonstrating their potential for pre-osteoblast cell adhesion and viability. ILs can dissolve degummed fibroin, forming a regenerated protein fiber with a Gly–Ala–Gly–Ala–Gly–Ser conformation, which assembles antiparallel β-sheets. This arrangement is sustained by packing Hort Gly and Ala amino acid side chains in hydrophobic crystalline regions.[214] Wang *et al.*[187] demonstrated an environmentally friendly and scalable method for silk nanofibrils scaffold using the DES composed of D-sorbitol : L-proline (2 : 1, molar ratio). The nanofibrils maintain their natural hierarchical structures and amphiphilic properties, enabling efficient dispersion and exfoliation of 2D materials. This membrane exhibits superior heat-dissipating efficiency compared to commercial silicone pads, displaying promising results for thermal management applications.

Green solvent-assisted silk fibroin and sericin hydrogels are gaining traction in bioprinting applications, where they serve as bioinks for fabricating customized tissue implants and organoid models. Their ability to be 3D-printed into complex structures while maintaining high mechanical stability has been considered suitable in artificial skin grafts, vascularized tissues, and osteochondral implants. In wound healing, silk protein-based hydrogels provide a moist environment, excellent gas exchange, and antimicrobial properties, promoting faster tissue regeneration while preventing bacterial infections. Incorporating bioactive molecules, such as growth factors and peptides, further enhances their healing efficiency.[230] These hydrogels can be combined with living cells, growth factors, and extracellular matrix components, enabling the fabrication of vascularized tissues, functional skin grafts, and organoid models. Fibroin-sericin hydrogels have also been investigated for applications in ophthalmology, dental regeneration, and soft tissue engineering. Their transparency and non-toxic nature make them promising materials for corneal repair and intraocular lens coatings. At the same time, their ability to integrate with surrounding tissues supports their use in periodontal regeneration and gingival repair.[100,229] Sheng *et al.*[229] studied the development of a novel protic IL, imidazolium sulfhydryl-containing (imidazole-SH), to create hydrogels with antibacterial and angiogenic properties. The *in vitro* assays indicated that these hydrogels effectively inhibited the growth of *Escherichia coli* and *Staphylococcus aureus*, thus exhibiting strong antibacterial properties. Moreover, the hydrogels played a significant role in reducing inflammatory responses triggered by lipopolysaccharides. The angiogenesis and transwell migration assays with human umbilical vein endothelial cells further confirmed that hydrogels actively promoted vascular repair and cell migration, which are crucial for accelerating wound healing. Yang *et al.*[234] reported the development of silk fibroin-based aerogels using the DES formed by guanidine

hydrochloride : urea (1 : 2, molar ratio), where these aerogels exhibited a lightweight structure, high porosity, and exceptional superelasticity. The aerogels demonstrate multifunctional properties, including high mechanical resilience, effective air filtration, and thermal insulation. Moreover, their structural integrity and performance remain stable under extreme conditions, such as repeated compression and exposure to high and low temperatures.

Green solvent-processed fibroin has been used in drug delivery to develop nanoparticles, microspheres, and hydrogels for controlled drug release. Its ability to encapsulate bioactive compounds ensures sustained release, enhancing the bioavailability of therapeutic agents such as antibiotics, anti-inflammatory drugs, and anticancer treatments, allowing for prolonged drug activity while minimizing systemic toxicity. The versatility of IL-treated fibroin extends to hydrogel-based drug carriers, where its hydrophilic nature and tunable degradation rate enable controlled drug diffusion and localized therapy. These fibroin-based drug carriers are being explored for wound dressings, transdermal patches, and injectable gel formulations, demonstrating high potential for personalized medicine and bioresponsive drug delivery systems.[106,239] Deng *et al.*[225] investigated [C_4C_1im]Cl in the self-organization of fibroin-based nanoporous microspheres for the controlled release of doxorubicin (DOX), an anticancer drug. The results showed that the release of the drug was faster in the composite microsphere (53.5% in the first 4 hours), while the pure protein showed a more controlled and gradual release. The *in vitro* and *in vivo* tests confirmed the anticancer efficacy of the DOX-loaded microspheres. Furthermore, it was observed that the microspheres protected the drug at high temperatures (200 °C), ensuring a higher stability. The controlled release was attributed to electrostatic and hydrophobic interactions between the drug, protein matrix, and IL. Another promising application is in flexible and biodegradable electronics, where IL or DES-modified fibroin has been successfully integrated into wearable biosensors, bioelectrodes, and biodegradable circuits. Fibroin-based ionogels synthesized with imidazolium-based ILs exhibit excellent ion-conductivity and mechanical flexibility and become suitable for biosensors, bioelectronic interfaces, and wearable bioelectronics.[96] The electroconductivity of the system can be optimized by influencing ion mobility, which depends on the specific size of cation and anion units. Notably, since cations are generally larger than anions, they significantly affect charge mobility and the system's overall conductivity.[240] Meanwhile, intermolecular forces that influence the viscosity of ILs also play a relevant role in ion mobility, thus affecting electroconductive properties.[241]

Fibroin or sericin-derived biodegradable electronic skins and flexible sensors also offer high sensitivity and environmental sustainability, providing a viable alternative for next-generation medical and wearable devices. Moreover, IL and DES-based silk fibroin membranes have been used to develop bioresorbable capacitors and biodegradable batteries, paving the way for dissolvable electronics that reduce electronic waste. Silk protein's optoelectronic properties, enhanced through IL-assisted processing, enable its application in bio-opto-electronic interfaces, nerve stimulation devices, and energy-harvesting materials.[192] Liu *et al.*[242] reported the development of ionic conductive poly(vinyl alcohol) (PVA) hydrogels using a binary solvent system of water/IL, specifically [C_2C_1im][C_1CO_2]. It was demonstrated that incorporating this IL into the hydrogel matrix improves its physical crosslinking, enhancing mechanical strength, high transparency, and excellent anti-freezing properties. The hydrogel remains flexible and conductive even at −50 °C and retains over 90% of its weight after two weeks of exposure to open-air conditions. The study also explores the application of these hydrogels in flexible sensing devices. This sensor exhibited long-term stability and a wide working temperature range, allowing a new route for advances in bioelectronics. Moreira *et al.*[96] investigate silk fibroin and ILs' use to create stable ionogels. The results showed that the choice of IL affects the secondary structure of fibroin and its mechanical properties. The ionogels based on 1-decyl-3-methylimidazolium chloride ([$C_{10}C_1$im]Cl) exhibited greater thermal, mechanical, and air stability. Additionally, due to their high ionic conductivity, these ionogels were tested as gas sensors, responding to different volatile compounds through changes in electrical conductivity. Sun *et al.*[196] evaluated the development of a silk sericin and DES-based gel (ChCl : urea, 1 : 1, molar ratio), which presented remarkable results in antibacterial properties with a 99% bactericidal rate against *Staphylococcus aureus* and *Escherichia coli*. *In vitro* cytotoxicity assays demonstrated excellent biocompatibility, confirming the material's potential for biomedical applications. Furthermore, the gel was integrated into a strain sensor to monitor macroscopic and microscopic human motions. Yang *et al.*[232] developed a novel approach to fabricating biomedical electronics from fibroin and ChCl : OA (1 : 1, molar ratio). The nanostructures exhibit outstanding electrical conductivity, high electromagnetic interference shielding, excellent electrical heating performance, and a sensitive humidity response. They also demonstrate efficient degradability in alkaline conditions, addressing environmental concerns related to e-waste.

7.2 Keratin from wool

Due to its high abundance, high biocompatibility, and low toxicity, keratin has been investigated to develop several biomaterials with diverse geometries (*e.g.*, films and fibers) for applications in the biomedical field, food packaging, and water treatment.[123,142,243] Recent studies exploring the dissolution of wool using ILs and DESs for keratin recovery and subsequent development of biomaterials are summarized in Table 3.

As presented in Table 3, several studies have demonstrated the successful dissolution of wool keratin using ILs and DESs to produce biomaterials. For instance, Li and Wang[244] developed keratin-based films by dissolving wool in [C_1C_1im]Cl and [C_4C_1im]Cl at 130 °C. The authors gradually incorporated wool at 1 wt% intervals until complete dissolution. The resulting wool keratin/IL solutions were cast onto glass substrates and immersed in various coagulant solvents (water, methanol, and

Table 3 Recent studies using ILs and DESs for the development of keratin-based materials

Biomaterial	Solvent	Relevant outcomes	Ref.
Film	$[C_1C_1im]Cl$ $[C_4C_1im]Cl$	Wool was dissolved in ILs and cast into films using a coagulant agent Resulting films exhibited a homogeneous surface morphology	244
Film	$[C_2C_1im][C_1CO_2]$	Biocomposites films with protein : polysaccharide ratios of 25 : 75 and 75 : 25 w/w were produced using various coagulant solvents Ethanol-treated films with lower protein content presented a smoother surface The final composition and coagulant type significantly influenced film structure, thermal stability, and morphology	245
Film	$[C_4C_1im]Cl$	Increasing dissolution temperature resulted in stress decrease by 43% and strain increase by 190% Glycerol addition enhanced chain mobility, enabled better thermal processing and reduced brittleness	246
Nanofiber	$[C_4C_1im]Cl$	The composite nanofibrous showed high antibacterial activity (89.21% against *E. coli*, 60.70% against *S. aureus*) Keratin incorporation promoted thinner, more uniform nanofibers with improved hydrophilic property and moisture permeability	247
Fiber	$[C_4C_1im]Cl$ $[MC_1im][DMP]$	Keratin : cellulose composite fibers were regenerated using water as coagulant Cellulose addition improved the mechanical properties of wool fibers Compared with commercial cotton and wool fibers, the regenerated fibers showed better properties, indicating good comfortability for wearing	248
Film	$[N_{111}(_2OH)][DHP]$ $[N_{111}(_2OH)][Seri]$	Wool keratin and cholinium-based ILs were incorporated into collagen-glycerol formulations to produce films Resulting films exhibited enhanced compactness, mechanical strength, electrical conductivity and dielectric response	249
Fiber	$[mTBDH][C_1CO_2]$ [mTBDH][Lev]	Integrating high-molecular-weight cellulose into wool keratin formulations improved the regenerated fibers' rheological behaviour and tensile properties, while reducing fibrillation and enhancing soft texture	250
Nanofiber	ChCl : urea L-Cys : LA	L-Cys : LA enabled the production of more uniform nanofibers The resulting materials exhibited significant bioactivity and biodegradability	210

[mTBDH] – 7-methyl-1,5,7-triazabicyclo(4.4.0)dec-5-ene; [Lev] – levulinic acid; $[N_{111}(_2OH)]$ – cholinium acetate; [DHP] – dihydrogen phosphate; [DMP] – dimethyl phosphate.

ethanol) for 12 h at room temperature. Scanning electron microscopy (SEM) confirmed the homogeneous surface of the films.[244] In a similar approach, Ghosh *et al.*[246] employed $[C_4C_1im]Cl$ to dissolve wool keratin at temperatures from 120 to 180 °C. The regenerated keratin was plasticized with glycerol at a weight ratio of 2 : 1 and compression molded into films at 120 °C under 10 MPa. Mechanical testing revealed a tensile strength of 10.3 MPa and 18% of strain when keratin was dissolved at 120 °C. However, increasing the dissolution temperature to 180 °C reduced the tensile strength by 43% while enhancing the strain by 190%. The addition of glycerol improved thermal processability and reduced brittleness.[246] Nevertheless, despite promising material properties, the studies focused primarily on film development and characterization, without addressing specific applications.

Aiming to expand the scope of keratin-based materials, Zhong *et al.*[247] dissolved wool fabrics in $[C_4C_1im]Cl$ at 130 °C (wool : IL ratio of 0.124 : 1), followed by the addition of 12.5 wt% polyacrylonitrile (PAN) in DMF to produce a homogeneous electrospinning solution. The resulting nanofibrous membrane showed enhanced hydrophilicity and notable antibacterial activity, with inhibition rates of 89.2% against *E. coli* and 60.7% against *S. aureus*.[247]

In addition to keratin, biocomposites by blending keratin with other biopolymers, particularly cellulose, have emerged as a strategy to improve the material's properties. Sun *et al.*[248] prepared wool keratin : cellulose (50 : 50%) composite fibers from textile waste using $[C_4C_1im]Cl$ and 1,3-dimethylimidazolium dimethyl phosphate ($[MC_1im][DMP]$). Mechanical tests indicated that cellulose enhances the mechanical strength of the fibers. At the same time moisture analysis demonstrated that the developed fibers present better properties than commercial cotton and wool fibers, suggesting good comfort for wearing and potential for textile applications.[248] In a different study, Rybacki *et al.*[245] investigated wool keratin: cellulose biocomposite films using $[C_2C_1im][C_1CO_2]$ at a total polymer content of 10 wt% (protein : polysaccharide ratios of 25 : 75 and 75 : 25 w/w). After dissolution, the solutions were molded and coagulated in various solvents (water, ethanol, and hydrogen peroxide solutions) for 48 h. SEM revealed that films with higher cellulose content and coagulated in ethanol display smoother surfaces and more homogeneous surfaces. In contrast, higher keratin content and lower hydrogen peroxide concentration led to more prominent areas of roughness.[245] Fang *et al.*[250] dissolved wool keratin in 7-methyl-1,5,7-triazabicyclo[4.4.0]dec-5-enium acetate ($[mTBDH][C_1CO_2]$) and 7-methyl-1,5,7-triazabicyclo[4.4.0]dec-5-enium levulinate ([mTBDH][Lev]) at 85 °C, and incorporated high-molecular-weight cellulose to enhance the rheological behaviour of the solution. This approach enabled fiber formation with improved tensile properties, reduced fibrillation, and a soft texture.[250] These findings highlight the influence of polymer composition and regeneration conditions on the final material properties.

Andonegi *et al.*[249] incorporated cholinium-based ILs and wool keratin into native collagen formulations (with 20 wt% glycerol and 0.5 M acetic acid, 1 : 2, collagen : acetic acid ratio). Compression-molded films exhibited reduced water uptake (325%) compared to control films (375%), indicating that the incorporation of both ILs and wool promoted the interactions with the polar groups of collagen and glycerol, thereby reducing the availability of these groups for water binding. SEM revealed a more compact microstructure in the modified films, further contributing to the reduced water uptake. Mechanically, the modified films demonstrated enhanced tensile strength, electrical conductivity, and dielectric response, indicating the potential of keratin-IL-collagen systems in functional materials.[249]

In addition to ILs, DESs have emerged as a promising alternative solvent for keratin dissolution and subsequent development of high-performance biomaterials. Mouro *et al.*[210] used cholinium chloride (ChCl) : urea and L-cysteine (L-Cys) : lactic acid (LA) to dissolve wool keratin, followed by direct incorporation into polyvinyl alcohol (PVA) matrices at varying ratios to fabricate electrospun nanofibers. L-Cys : LA showed superior performance, yielding uniform fibers. A comprehensive physicochemical characterization was conducted, including pH, conductivity, and viscosity. Notably, the nanofibers exhibited significant antioxidant and antimicrobial activity, reinforcing the viability of DESs as alternative solvents for producing functional, biodegradable, and sustainable nanofibers.

These studies illustrate the growing interest in wool valorization for developing high-performance biomaterials through integrated green processing strategies. The use of ILs and DESs facilitates efficient keratin recovery while maintaining its functional properties and enables the design of sustainable biomaterials with potential in diverse fields. Most of the presented studies highlight the influence of processing parameters, coagulant solvent, and polymer blends to produce biomaterials with improved properties. However, despite promising results, further research is needed to evaluate performance for desired applications and address scalability challenges.

8. Market growth potential

The global market for natural textile fibers has historically been important for various industrial sectors, from fashion to technical applications. Nevertheless, in recent decades, scientific advances and the growing demand for sustainability have increased the value of the structural proteins present in these fibers. After obtaining the protein fibers, regardless of the process used (whether from virgin sources or post-consumer textiles), these compounds can serve as raw materials to produce higher-value-added materials.[251]

Virgin silk has a prominent position in the luxury market, with an estimated value of US$16 billion in 2024. Its production involves labor-intensive processes, including silkworm rearing, cocoon harvesting, and degumming to obtain raw fibroin. Despite its high unit value (50–150 US$ per kg), the sector faces challenges related to scalability and environmental impact, particularly concerning water consumption and chemical use during processing. Similarly, virgin wool commands a global market estimated at US$38 billion, with a lower price per kilogram (5–20 US$ per kg) but a significantly higher production volume. Its supply chain, based on sheep farming, is subject to pressures related to animal welfare and competition with low-cost synthetic fibers. Both markets exhibit modest growth rates (Compound Annual Growth Rate –CAGR– of 3.8% for silk and 2.5% for wool), indicating market maturity and expansion limitations.[251,252]

Proteins extracted from these fibers have gained prominence due to their technological applications and economic value. Fig. 8 shows that each structural protein differs significantly in market size and selling price. Keratin leads the market at US$2.5 billion, followed by fibroin (US$1.8 billion) and sericin (US$900 million). This hierarchy reflects keratin's versatility, which is widely used in biomedical and cosmetic industries, along with the abundant raw material supply from the wool industry waste. Complex extraction processes and high demand for high-purity grades justify its price (US$500 per kg).[253] In contrast, despite having a smaller market, fibroin commands a high price (1000 US$ per kg) due to its applications in high-value sectors that leverage its unique mechanical properties.[82] Sericin (240 US$ per kg), with a lower market value, remains underutilized, but recent studies highlight its potential as a controlled drug release vehicle, a growing sector with a CAGR of 14.2%.[254]

Given this scenario, recovering protein fibers from post-consumer textiles emerges as a promising strategy to mitigate environmental impacts, overcome the expansion limitations of virgin raw material markets, preserve animal welfare, and reduce the intensive use of natural resources. The valorization of post-consumer textile waste minimizes environmental

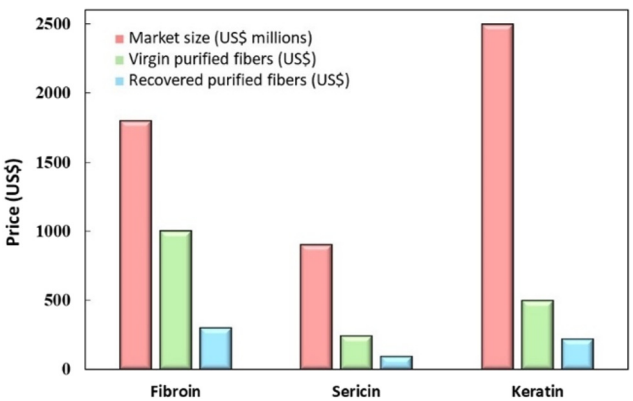

Fig. 8 Price comparison between market size (US$ million per kg, in light pink), virgin purified fibers (US$ per kg, light green), and recovered purified fibers (US$ per kg, light blue) according to the 2024 survey. The average selling price of virgin and recovered fibers was considered.

impact. It represents an economic opportunity, as these fibers have high market value and could be reused rather than discarded in landfills, preventing significant losses.[81]

The structural textile protein market presents a dynamic and increasingly attractive economic landscape. Studies indicate that fibers such as fibroin, sericin, and keratin, extracted from discarded clothing, currently achieve 60–80% of the market value of virgin proteins, with a trend toward accelerated appreciation. Recovered fibroin ranges between 240–360 US$ per kg, depending on purity level and application.[255] The biomedical segment, which requires purity above 94% and a molecular weight exceeding 70 kDa, exhibits a 35–40% increase in value compared to textile-grade fibroin. Sericin varies between 65–120 US$ per kg, with pharmaceutical applications valued nearly twice as much as cosmetic ones when low immunogenicity is demonstrated. Recovered keratin ranges from 175–260 US$ per kg, with its highest value linked to high levels of free sulfhydryl groups (>60%) and biocompatibility certifications.[256]

Recovery costs vary significantly: fibroin (48–72 US$ per kg), sericin (32–55 US$ per kg), and keratin (25–65 US$ per kg). Gross margins range from 45–72%, with sericin having the shortest payback period (2.8 years) due to lower purity requirements for cosmetic applications. Despite its high processing cost, fibroin offers the highest margins (58–72%) in the medical sector. The European market stands out with 22% higher prices, driven by government subsidies covering 15–20% of costs, reflecting strong demand in medical applications (72%) and a well-established reverse logistics infrastructure. Asia accounts for 54% of global volume, with 30% lower operational costs and a focus on textile reuse. In North America, medical-grade keratin is growing at an annual rate of 14%. At the same time, South America shows moderate growth, driven by raw material availability and sustainable policies, with Brazil and Argentina leading in natural fiber production and recovery.[257,258]

The data presented in Table 4 reveals a complex and diversified outlook for the protein biomaterials market, where each application exhibits unique market value, pricing, and growth potential. A detailed category analysis helps identify strategic opportunities and determine the most attractive upcycling route.[259] For biomaterial-oriented deployment, market attractiveness must be interpreted jointly with regulatory-relevant quality constraints. Application readiness depends on whether recovered proteins meet purity, residual-contaminant, and structural-consistency thresholds required for the intended end use. Consequently, economic ranking alone is insufficient to classify route feasibility, since two routes with similar price signals may differ substantially in qualification burden, analytical control requirements, and downstream compliance effort.[104,189] Scaffolds dominate the market at US$2.1 billion, driven by demand for personalized regenerative medicine solutions and advancements in 3D bioprinting, enabling structures with controlled porosity. Meanwhile, drug delivery systems have emerged as a strategic segment, reaching US$1.8 billion, distinguished by high-precision drug-loading capacity (50–180 mg of drug per gram of material). Fibroin has proven advantageous in these applications, offering superior biocompatibility with rejection rates below 5%. Although representing a smaller market (US$1.2 billion), hydrogels boast attractive gross margins (70–80%) due to cost-effective crosslinking processes and cosmetic applications.

Environmental interpretation should be based on process-level, which comparative assessment should account for pre-treatment demand, dissolution conditions, anti-solvent use, washing/polishing intensity, solvent-recycle efficiency, and wastewater management per unit of qualified product. This is particularly relevant for protein biomaterials, in which additional purification and quality-control requirements can became key drivers of cumulative resource use.[260]

In addition to market size and selling price, the interpretation of application feasibility should incorporate protein-throughput descriptors, distinguishing the quantity of protein treated upstream from the quantity embedded in final products. For fibroin- and keratin-based value chains, the treated-protein demand is governed by extraction/recovery performance, including recovery yield, purification losses, and recycle efficiency, whereas product-level demand is governed by application-specific protein loading.

Incorporating quantitative context, global raw silk production is reported at approximately 91 221 t in 2022 and around 91 415 t in 2024, indicating a relatively small but high-value protein-fibre stream compared with major textile fibres.[252] For wool, global production is substantially larger, with 1977.3 million kg greasy wool and 1051.2 million kg clean wool reported for 2022, confirming the higher mass-throughput relevance of keratin-rich feedstocks at the sector level. However, literature and industry reports remain structurally fragmented for downstream circular metrics, whose most studies report laboratory or pilot recovery yields under specific conditions, while audited annual totals of fibroin or keratin recovered and application-resolved allocations are rarely disclosed in harmonized units.[258,261] This data gap arises from methodological non-uniformity across studies and reports, whose outputs are expressed in different units (mass, area, or

Table 4 Market size, selling-price basis, and throughput comparability across fibroin- and keratin-derived applications, according to the 2024 research[a]

Applications	Market size (US$ millions)	Selling price	Produced quantity[b]
Scaffolds	2100	300–900 un^{-1}	2–7 million units
Drug delivery	1800	50–180 mg^{-1}	10–36 kg
Hydrogel	1200	250–800 g^{-1}	1.5–4.8 t
Wound healing	950	70–200 cm^{-2}	475–1357 m^2
Membranes	780	400–1500 m^{-2}	0.5–1.9 million m^2
Bioelectronic	650	20–80 un^{-1}	8–32 million units
Biofilm	550	60–150 m^{-2}	3.7–9.2 million m^2

[a] Only products presenting actual industrial-scale production were considered. [b] Per year.

piece count), process boundaries are defined differently (extraction step only *versus* extraction plus purification/regeneration), and industrial datasets frequently omit material-throughput and formulation details due to confidentiality. In addition, many sources do not report key mass-balance descriptors, such as recovery losses across separation steps, solvent-recycle performance, and protein loading in the final product. Consequently, market-value metrics should be interpreted together with explicitly reported process variables, and quantitative comparisons across applications should be limited to studies using consistent units and equivalent system boundaries.[200]

Wound healing applications, valued at US$950 million, have also shown impressive advancements. Modern dressings based on protein fibers exhibit an exudate absorption capacity of up to 300% and competitive pricing. Regenerative membranes, with a market of US$780 million, represent another area of significant growth. These membranes, developed from protein fibers, allow for precise thickness control and adjustable porosity, which makes them suitable for various clinical needs. Despite holding a market value of US$650 million, Bioelectronics stands out with the highest CAGR (18.7%), driven by breakthroughs in implantable medical devices, particularly biodegradable sensors for continuous monitoring and temporary epidermal electronics. Data analysis reveals a significant challenge in directly comparing applications due to heterogeneity in pricing units (US$ per g, US$ per m^2, US$ per unit). This diversity reflects the specific nature of each application but complicates comparative assessments.[81,83,252,253,257]

9. SWOT analysis and future perspectives

The recovery of protein-based textile waste using ILs and DESs has great potential for aligning the textile industry with circular economy principles and sustainability. However, this approach also faces significant challenges. A SWOT (Strengths, Weaknesses, Opportunities, and Threats) analysis is shown in Fig. 9, allowing a comprehensive assessment of the technology's positive and negative aspects.

Among the main advantages provided by ILs and DESs are environmental sustainability, such as low volatility, non-flammability, and thermal stability, which significantly reduce the environmental impact compared to conventional solvents. Furthermore, when properly designed, these alternative solvents can be finely tailored for specific applications, such as extracting protein from complex matrices, for which they have already demonstrated high efficiency. Another strong point is their efficiency in dissolving and preserving proteins. ILs such as $[C_4C_1im][C_1CO_2]$ and $[C_2C_1im]Cl$, as well as cholinium chloride-based DESs, selectively dissolve proteins without degrading their structure, maintaining mechanical and biological properties essential for advanced applications. Processes involving extractions and separations using ILs and DESs have been reported to be highly efficient in atom economy, producing less waste and undesirable by-products. Moreover, these processes have other sustainable credentials, such as lower toxicity and biodegradability than conventional methods. Using new bio-based solvents can provide new

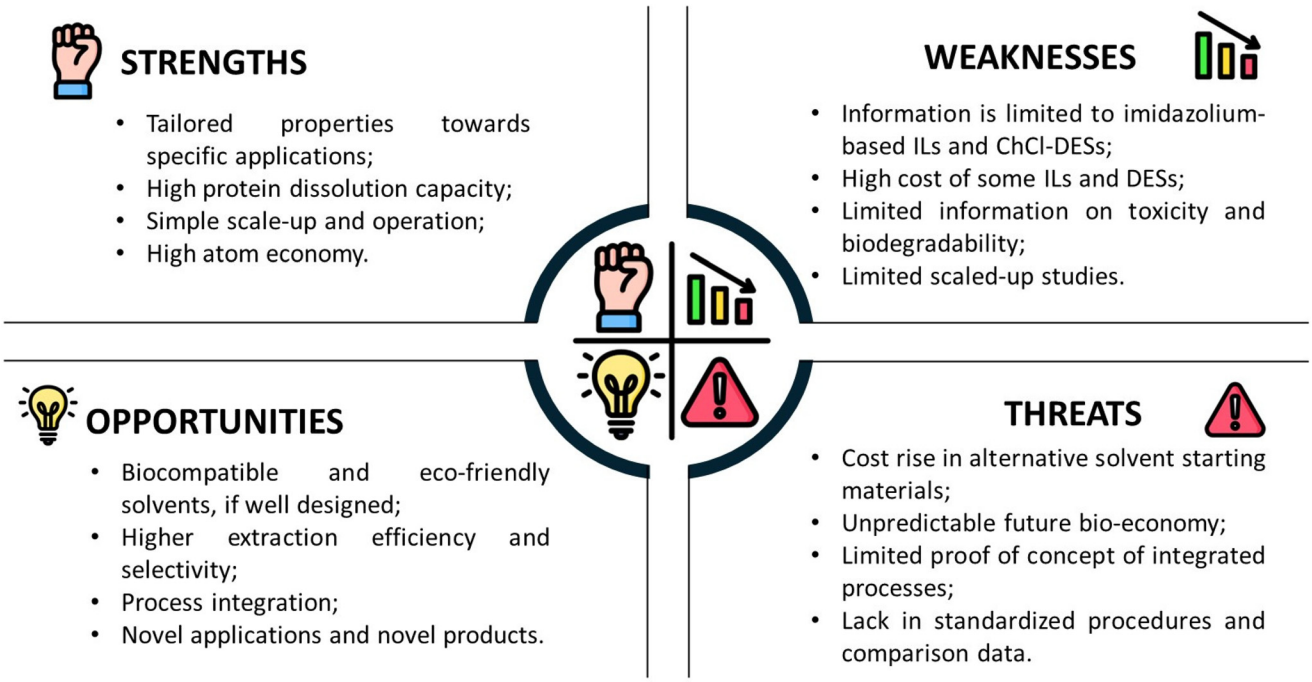

Fig. 9 SWOT analysis of the protein recovery process from silk and wool clothing waste using ILs and DESs.

opportunities in separation processes, as they are less toxic and biodegradable, allowing for developing more sustainable processes. Finally, technological innovation, such as the strategic combination of ILs and DESs with advanced extraction methods (microwave-assisted – MAE, ultrasound-assisted – UAE) and optimized enzymatic processes, can significantly increase the efficiency and selectivity of protein recovery, expanding the potential for recycling textile waste.

The large-scale adoption of this technology faces significant challenges, starting with the high production costs of ILs and DESs, which represent an essential economic barrier. The transition from laboratory processes to mass production also presents technical difficulties that could delay its industrial implementation. The efficiency of the process still varies considerably depending on the type of fiber processed (silk or wool) and the presence of contaminants, often requiring additional pre-treatment steps. Complementing these obstacles, market uncertainties and the political scenario's instability can impact the development of a sustainable bioeconomy.

Despite the challenges, the biomaterials market is growing, with significant global demand. This scenario is driven by the search for sustainable products in fashion and health, creating a fertile environment for strategic investments and cross-sector partnerships. Meanwhile, advances in green chemistry, such as developing new ILs and DESs that are less toxic and more efficient, can reduce operating costs and significantly improve process performance. Integrating these solvents with emerging technologies, such as 3D bioprinting for personalized medicine, opens a path for disruptive innovations in the sector. However, one of the most promising opportunities resides in extracting structural proteins from post-consumer waste. In addition to transforming materials that would usually be destined for incineration and, therefore, have no added value into products with high commercial value, this approach directly reduces the pressure on the demand for virgin raw materials constituting a bilateral economic benefit: it simultaneously creates revenue streams and mitigates environmental impacts.

Nevertheless, some risks could hinder the consolidation of this technology. Competition with synthetic fibers such as polyester, which still dominate the market due to their low cost, is a significant challenge. Rigorous environmental regulations, especially for medical and food applications, could delay commercialization due to the need for complex certifications. Finally, inefficient reverse logistics, especially in developing countries, limit the collection and sorting of textile waste, which is essential for feeding the recycling process.

Conclusions

The growing concern about the environmental impacts of the textile industry has driven the search for innovative and sustainable solutions for the valorization of the respective waste, especially those of protein origin, such as silk and wool. This review has shown that ILs and DESs represent promising alternatives to conventional recycling methods, due to their ability to selectively solubilize fibroin, sericin, and keratin, preserving their structural and functional properties. By modulating parameters such as temperature and properly designing the solvent, it is possible to optimize the efficiency of the process and recover biomacromolecules with high value. Applying these regenerated biopolymers in areas such as tissue engineering, controlled drug release, biodegradable electronic devices and composite materials highlights the transformative potential of approaches based on ecological solvents. Thus, incorporating these technologies into the textile production chain represents a significant step towards a more circular, innovative, and environmentally responsible model, directly meeting the UN's Sustainable Development Goals and building a greener textile industry.

Author contributions

Conceptualization: F. S. B. and M. F. G.; data collection: F. S. B., C. P., and B. D. R.; writing and editing the original draft: F. S. B., C. P., and B. D. R.; review, editing and manuscript checking: M. A. Z. C. and M. G. F.; funding acquisition: B. D. R., M. A. Z. C., and M. G. F.

Conflicts of interest

There are no conflicts to declare.

Data availability

No new data were created or analysed in this study. Data sharing is therefore not applicable to this article.

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