



2023

Sustainable Textile Raw Materials: Review on Bioprocessing of TextileWaste via Electrospinning

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Review

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Abstract: The fashion and textile industry in its current fast-rising business model has generated a huge amount of textile waste during and after the production process. The environmental impact of this waste is well documented as it poses serious threats to lives on earth. To confront the menace of this huge pollution problem, a number of research works were carried out to examine the possible re-utilization of these waste materials without further damaging the environment; for instance, reusing, generating valuable products, or regenerating fibrous materials to form a closed loop in the cotton textile waste lifecycle. This review covers different methodologies to transform cellulosic textile materials into various products with added value, such as cellulosic glucose, cellulase, etc., and finally, to regenerate the fibrous materials for re-application in textiles and fashion. This article presents an overall picture to researchers outlining the possible value addition of textile waste materials. Furthermore, the regeneration of cellulosic fibrous materials from textile waste will be brought into the limelight.

Keywords: electrospinning; regenerating fibers; bioprocessing; cellulose; textile wastes



Citation: Suen, D.W.-S.; Chan, E.M.-H.; Lau, Y.-Y.; Lee, R.H.-P.; Tsang, P.W.-K.; Ouyang, S.; Tsang, C.-W. Sustainable Textile Raw Materials: Review on Bioprocessing of Textile Waste via Electrospinning. *Sustainability* **2023**, *15*, 11638. <https://doi.org/10.3390/su151511638>

Academic Editor: Izabela Luiza Ciesielska-Wróbel

Received: 31 May 2023

Revised: 6 July 2023

Accepted: 24 July 2023

Published: 27 July 2023



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1. Introduction and the Environmental Impact of the Textile and Apparel Industry

The textile industry is ranked the second largest polluting industry in the world because of the creation of textile waste, ecosystem pollution, and the immense use of chemicals and water [1]. The textile sector contributes to 3% of the total greenhouse gas emissions [2]. Global apparel consumption is 400% in excess of the quantity consumed 20 years earlier [3]. The reducing life cycle of textile products, the rising living standards, the expanding world population, and the increasing variety of textile materials driven by worldwide fiber purchases all contribute to a considerable volume of post-consumer and post-industrial fiber wastes [4]. Alongside the effect of globalization, the apparel industry manufactures more clothing at lower costs due to the outsourced production to low-cost countries as well as the anticipated trend of ‘fast fashion’, which commonly perceives clothing as a ‘one-use’ product. Overall, ‘fast fashion’ embodied by cheapness, agility, diversity, and mass production has prompted a rise in apparel consumption [1,5].

Consumers, to a certain extent, are still negligent of disposal approaches and sustainable consumption. The futile disposal of textiles is an emerging critical problem faced by different parts of the globe [4]. At the global level, around 75% of textile waste was dumped in landfill sites or burned; 25% was recycled or reused; and below 1% of the whole textile

was recycled as clothing. By the end of 2030, 92 million tons of textile waste will have been created annually [3]. As such, making progress in recycling and reuse technologies for transforming textile wastes from landfills is indispensable. There is a pressing need for closed-loop recycling of fabrics. Different levels of international actions incorporating various stakeholders have highlighted both environmental and economic difficulties that the textile industry is encountering. For instance, Textile Exchange promised to decrease CO₂ emissions from material production and textile fibers by 30% before 2030 and to facilitate the key role of circular economy, which are effective means for diminishing effects and combating climate change. Textile recycling and reuse are crucial to attain such innovative performance proclaimed by Textile Exchange [5].

Owing to the ever increasing amount of textile waste produced in the world, it is essential to determine some environmentally friendly ways to deal with the waste, rather than indiscriminately disposing of it in a landfill. Currently, there are two main sustainable ways to process textile waste: reuse and recycle. Reusing textile waste extends the duration of textiles to be in use, hence limiting the amount of new textiles to be produced for the market. To speed up the process of adopting recycled materials, it is imperative to investigate effective and economically viable ways to identify and classify textile materials and to reprocess them [6]. Recycling, on the other hand, is to process textile wastes for other purposes and is generally regarded as less beneficial than reuse, when suitably extending the reusing stage. At present, the textile recycling rate remains low despite the numerous recycling programs, investments, standards, and approaches that have been implemented. The textile recycling rates in the European Union, the United States, and China are 25%, 15.2%, and 15%, respectively [2].

Textile waste comprises diverse materials, pertaining to synthetic non-cellulosic fibers along with natural fibers, thus bringing the industry to a bioprocessing challenge [6]. One way to recycle textile waste is via enzymatic decomposition. In this paper, recent advancements in the bioprocesses of textile waste are first summarized and discussed, with a specific focus on physicochemical and enzymatic methods, such as the modified pretreatment methods, the production of cellulase-type enzymes from textile waste materials, and the use of cellulase to recycle textile wastes. Second, recent advances on the electrospinning technique, which is being used to produce nanofibers from a variety of materials, including polymers, ceramics, and composites, will be discussed. In particular, the use of electrospinning to respin bioprocessed textile wastes is an emerging area of research that has the potential to address the problem of textile waste and provide a sustainable alternative to traditional textile production methods. One major research gap is the need for a systematic evaluation of the properties of the electrospun nanofibers produced from bioprocessed textile wastes, which include the studies on mechanical properties, surface morphology, and the chemical composition of the nanofibers. These properties are crucial in determining the potential applications of the nanofibers and optimizing the electrospinning process. Another research gap could be the need to explore different methods for preparing bioprocessed textile waste for electrospinning. The electrospinning process typically requires a solution of the material to be electrospun, and it is unclear how different methods of bioprocessing could affect the properties of the resulting nanofibers. Challenges associated with toxic solvents and large fiber diameters also exist within traditional electrospinning. Additionally, there may be a need to investigate the scalability of the electrospinning process for producing nanofibers from bioprocessed textile waste. While electrospinning has been demonstrated at the laboratory scale, scaling up the process may require optimization of the process parameters and equipment design.

The textile and apparel industry is a major global industry, accounting for about 2% of the global GDP. Advanced production technologies have lowered costs, leading to a 'fast fashion' trend [7] where consumers buy and dispose of clothing at an increasing rate. This trend has significant environmental impacts, including depletion of non-renewable resources, greenhouse gas emissions, and excessive water and energy use, contributing to irreversible climate change [8]. The disposal rate has made the industry the second largest

polluter globally, with an estimated one garbage truck of textiles landfilled or burned every second. If not addressed, the industry could use up a quarter of the world's carbon quota by 2050 [9]. For example, according to Monitoring of Solid Waste in Hong Kong—Waste Statistics for 2018, a report from the Environmental Protection Department, Hong Kong, approximately 392 tons of textile waste materials are produced daily, of which only 4.3% are recycled locally (this percentage was comparable to the city's market share in global waste management, which is about 3% [10]), whereas the remaining textile waste ends up in the landfills [11], thus posing environmental concerns.

In recent years, significant research has been carried out on the reuse and recycling of textile materials. The reuse of textile materials requires various means to increase the service span of textile products with or without prior modification in order to be accepted by new owners [12]. On the other hand, textile recycling most often refers to the reprocessing of pre- or post-consumer textile wastes for applications in new textile or even non-textile products. Figure 1 shows the reuse and recycling of different textile materials. Post-consumer textile wastes contain yarns from processed cotton fibers, which are woven or knitted into fabrics [13] and could be woven together with other natural or synthetic fibers, such as nylon, polyester, polypropylene, and acrylic. This fiber mixture in fabrics, along with additional treatments, results in the recycling and decomposition of such materials becoming increasingly challenging.

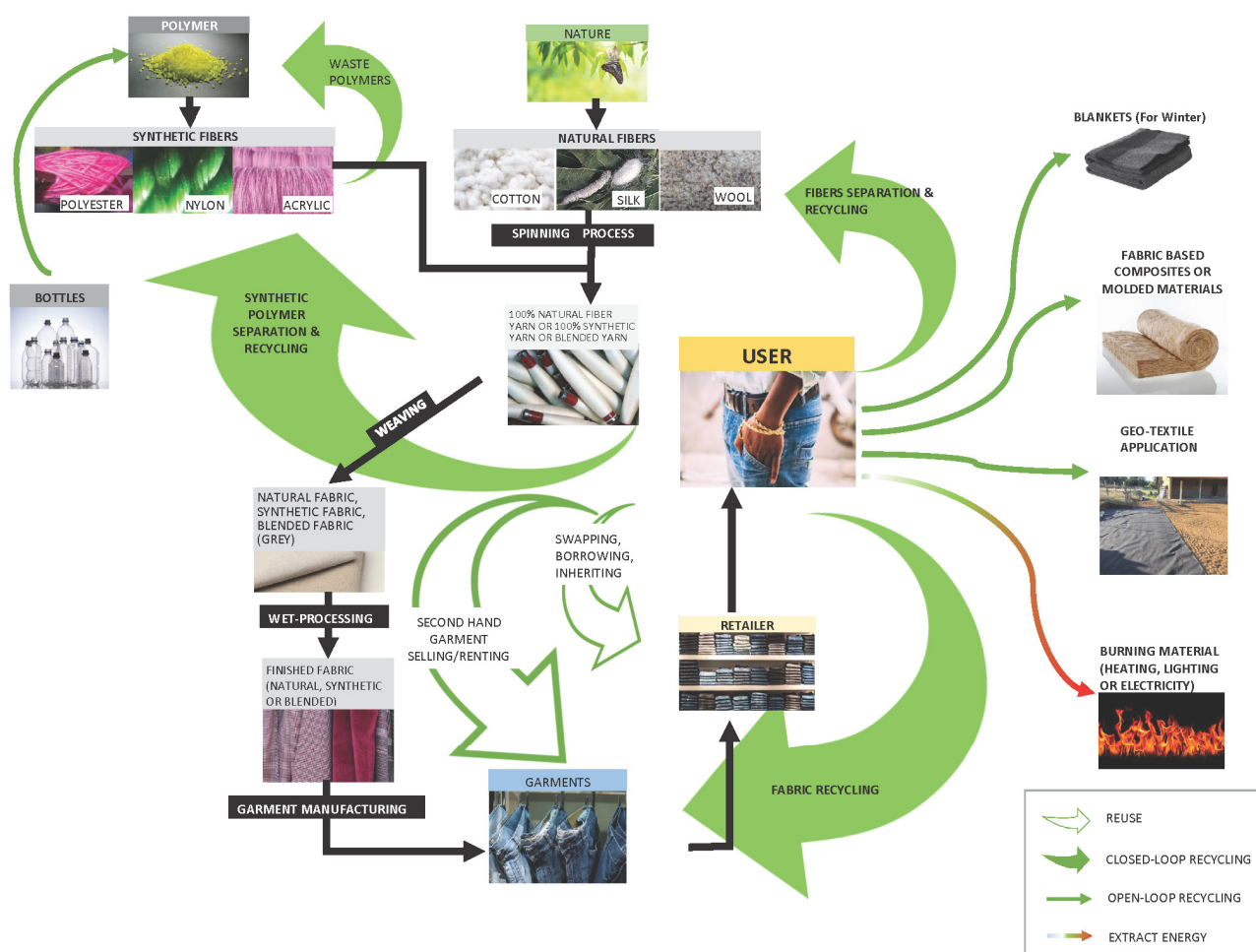


Figure 1. The reuse and recycling of various textile materials.

Several review papers covered the reuse and recycling of textile materials [14–19]. They concluded that the current waste management methods for post-consumer textile waste could be identified as waste prevention and minimization, reusing and recycling

materials, and energy recovery and disposal of waste. Amongst them, recycling is one of the best pollution control strategies. On the one hand, upcycling is defined as converting textile wastes into higher value products, say new garments. On the other hand, down-cycling is defined as converting textile wastes into raw materials of lower value. For example, textile wastes can be used as additives in thermal insulation building materials [20,21], as a binder in hydraulic lime [22], as well as in gypsum and cork composite materials [23]. Textile waste can also be synthesized into cellulose acetate [24]. Caprolactam can be extracted from textile waste and repolymerized to produce Nylon 6 [25]. Ethanol can be produced from such waste [26], while polyester and cotton raw materials can be obtained after separation from textiles [27]. The cotton components can be bioconverted into other materials with added value, such as biofuel [26], glucose syrup, and powder cellulose [28]. Due to the highly compact and crystalline nature of cotton, the challenge that remains with the bioconversion of cotton components is to ensure a high conversion rate and product yields. Chemical methods, such as first dissolving either cellulose or polyester followed by depolymerizing via hydrolysis or alcoholysis, are commonly used processes. Non-toxic and reusable solvents, such as N-methylmorpholine N-oxide, are frequently used for dissolving cotton, while dimethyl terephthalate is used for dissolving polyester. However, these recycling processes, such as methanolytic depolymerizations, are often carried out under harsh conditions, which greatly complicate the recycling process [29]. In comparison, bioprocessing can be performed to degrade textile wastes more selectively under much milder conditions and it may reduce energy consumption and carbon emissions. This process can also be more economically viable for industrial-scale production [30]. It is also a highly selective process and allows for the targeted breakdown of specific components in the textile waste and the production of high-quality products. Microbial hydrolases, such as cellulase and ligninase, are particularly useful for degrading natural polymers under enzymatic hydrolysis conditions. Recently, fungus has been used to produce cellulase from cotton textile wastes, e.g., *Aspergillus niger*, which can then hydrolyze cotton wastes [28]. Silk can be degraded by using proteinase K from a fungus called *Engyodontium album* to specifically cleave the site adjacent to the His, Phe, Trp, Ala, Ile, Leu, Pro, Val, and Met amino acids [31]. However, one should bear in mind that bioprocessing may have limited scalability, as the process may be slow and requires a large amount of space and equipment to handle large quantities of textile waste. It involves many parameters, such as enzyme type and concentration, reaction conditions, and substrate characteristics, which can be challenging to optimize for a specific application. The cost of enzymes used in bioprocessing can sometimes be very high, which may limit the economic feasibility of the process. The yield of the product from bioprocessing can sometimes be relatively low, which may not be sufficient to meet commercial demands.

The methodology of this study is divided into seven sections. In Section 1, the research background, setting, objectives, and the environmental impact of the textile and apparel industry are provided. Next, the treating of textile waste using enzymatic treatment to break down the fibers and remove any impurities or contaminants is elaborated on in Section 2. The type of enzyme and treatment conditions can be optimized based on the type of textile waste and the desired properties of the resulting nanofibers. Key textile and clothing materials, namely cellulase and cotton, are discussed in Sections 3 and 4, respectively. The various pretreatments and enzymatic hydrolysis of cellulose are explained in Section 5. Fungal cellulase production from textile waste by solid-state fermentation and the electrospinning and optimized conditions for defect-free nanofibers with desired diameter are illustrated in Section 6. The conclusions and recommendations are stated in Section 7.

2. Value Addition of Textile Waste Materials by Enzymatic Methods

Textile waste is a significant environmental problem, with large amounts of waste generated by the textile industry and discarded by consumers every year. However, textile waste can also be a valuable resource, with the potential for value addition through

various processes such as recycling, upcycling, and bioprocessing. Recycling of textile waste involves the conversion of waste textiles into new products, such as fibers, yarns, and fabrics. This process can be accomplished through mechanical, chemical, or thermal methods.

Based on the IUPAC definition, biodegradation is the degradation caused by an enzymatic process resulting from the action of cells, at least in part (IUPAC. Compendium of Chemical Terminology). Textile-based polymers can be degraded, in the presence of air and water, into smaller molecules by bacteria, fungi, and some other microorganisms that biosynthesize relevant enzymes [32–34]. This kind of degradation is known as waste recycling [35]. Most of the naturally existing and regenerated polymers (those purposefully designed to resemble natural polymers) are biodegradable, but a few exceptions are also available [36,37]. Among synthetic polymers, biodegradable aromatic polyesters, such as polybutylene adipate-co-terephthalate, can be easily degraded and researchers have designed enzymes to biodegrade polyethylene terephthalate or polyester-based textile materials at a reasonable price [38–40]. For example, Li et al. [28] investigated the enzymatic hydrolysis of cotton textile waste for the production of fermentable sugars, which can be further processed into biofuels or bioproducts. Upcycling of textile waste involves the transformation of waste textiles into higher-value products, such as fashion accessories or home decor items. This process can be achieved through various methods, such as cutting, sewing, and embroidery. For example, Gupta et al. [41] developed a method for the upcycling of denim waste into fashionable bags and accessories. Bioprocessing of textile waste involves the use of microorganisms or enzymes to break down the textile waste into valuable products, such as biopolymers or biofuels. This process is environmentally friendly and has the potential to produce high-value products from waste materials.

3. Cotton—A Cellulosic Material

The use of cotton, a type of natural cellulosic fiber, is widespread in the textile industry [42]. The fibers are harvested from the cotton ball that grows on plants belonging to the *Gossypium* family [43]. Cotton fibers are made up of long plant cells with a multi-layered cell wall structure and a natural twist. The lumen, which is the innermost part of the cell and originally the “living” part, is filled with liquid, and is protected by primary and secondary walls. The outermost layer of the cotton fiber is called the cuticle, which is made up of wax, proteins, and pectins, and is amorphous in nature. Microfibrils form the primary and secondary walls, which have varying degrees of crystallinity and molecular chain orientations [43]. Cotton fibers with well-developed cell walls are suitable for use in textiles as they possess a dried cell in which the lumen becomes a hollow space inside the collapsed cell wall [44].

Cotton fibers are composed of approximately 96% cellulose, which is the load-bearing polymer in mature fibers. Cellulose comprises almost all of the secondary cell wall components, with a high degree of polymerization (DP of 14,000), and 30% of the primary wall, with a DP between 2000 and 6000 [45]. The remaining components of cotton fibers are hemicelluloses and other trace elements [46]. The microfibrils that comprise the long cellulose chains are arranged in different orders. In the crystalline zone, cellulose chains are well-organized and linked through hydrogen bonds and van der Waals forces, while an amorphous structure contains twisted or less ordered microfibrils [47]. The amorphous cellulose is the weaker part of the cellulosic fibers and is susceptible to rapid hydrolysis by enzymes [48]. Table 1 lists the various textile production processes and the waste materials generated during each process and after use.

Table 1. Waste materials generated during the textile production processes and after use of textiles [49,50].

Material Generation	Process	Solid Waste Generation	Composition of Waste Generated	Existing Methods of Recycling Material	Further Applications
Cotton fibers	Ginning	Fiber waste	Cellulose, protein, sugar, wax	Mechanical recycling	Textile products
Yarn preparation	Spinning	Fiber lint, yarn wastes	Cellulose, protein, sugar	Re-spinning and plying	Carpet and clothes
Yarn preparation for fabrics	Weaving preparation (warping, sizing)	Fibers, yarn wastes	Cellulose, protein, starch, sugar	Re-spinning and plying	Carpet and clothes
Fabrics	Weaving/knitting	Yarn, fabrics	Cellulose, starch, sugar	Mechanical and chemical recycling	New fabrics
Fabric pretreatment	Desizing	Fiber lint, yarn	Cellulose	Mechanical and chemical recycling	New fabrics
Fabric pretreatment	Scouring and bleaching	Few or very few wastes	-	Mechanical and chemical recycling	New fabrics
Dyed and printed fabrics	Dyeing and printing	Few or very few wastes	-	Mechanical and chemical recycling	New fabrics
Finishing	Fabric finishing	Torn fabrics	Cellulose		
After use		Fabrics	Cellulose		

4. Cellulase

Cellulases are enzymes of great industrial interest due to their wide range of applications, including in the textile industry. They form a multi-enzyme system [51] that breaks down the cellulosic materials into fermentable sugars. Cellulases are produced by various types of bacteria and fungi, such as *Cellulomonas fimi* and *Thermomonospora fusca* [52], as well as filamentous fungi belonging to the genera *Trichoderma* (i.e., *T. viride*, *T. longibrachiatum*, *T. reesei*) and *Aspergillus* (*A. niger* N402, *A. niger* CKB) [53]. Among these, *Trichoderma* is the most extensively studied fungal genus for the commercial production of cellulase [54–57].

The cellulase enzyme system that performs enzymatic hydrolysis of cellulose consists of mainly three mono-enzymes, β -1,4-endoglucanases (EG), β -1,4-cellobiohydrolase (CBH), and β -glucosidase [58]. These enzymes work together to break down β -1,4-glycosidic bonds in amorphous cellulose, releasing fermentable sugars [59,60]. During this process, endoglucanase hydrolyzes chains of cellulose to form new chain ends, which are further broken down into soluble sugars (e.g., cellobiose) by exoglucanase. Cellobiose is then converted to glucose with the help of β -glucosidase. The efficiency of enzymatic cellulose hydrolysis is influenced by several factors, such as the cellulase preparation process [61], the composition of cellulosic materials [62], and the pretreatment method. The reactivity of cellulase and the properties of textile materials are also crucial factors in the hydrolysis process [63]. Furthermore, the pH and temperature of the reaction vessel significantly affect the hydrolysis process [64].

5. Pretreatment and Enzymatic Hydrolysis of Cellulose

5.1. Pretreatment

Pretreatment is the process to increase the sensitivity of textile materials, which can be prone to further treatment [61,65]. The efficiency of any pretreatment method is decided by the reactivity of cellulose and the increase in the yield of fermentable sugars from textile materials [66,67]. The basic pretreatment approaches can be classified into four categories, i.e., chemical (acid, alkali, and ionic liquid), biological, physical (milling and grinding), and physico-chemical processes. The commonly used pretreatment methods for textile waste are acid, alkali, ionic liquid, and supercritical fluid.

5.2. Acid Pretreatment

Acid pretreatment can hydrolyze the polymeric bonds in the hemicellulose to form its monomers, thus enhancing the availability of cellulose and increasing the biodegradability. Although acid pretreatment gives large glucose yields, the main problems lie with the formation of fermentation inhibitors (such as lignin-derived phenolic compounds), the high costs of acid, and the need for corrosion-resistant equipment. Researchers, including Chu et al. [68], Kuo et al. [69], Mahalakshmi et al. [70], Ouchi et al. [71], and Shen et al. [72], have used either sulfuric acid or phosphoric acid at different concentrations and hydrolysis conditions to treat the wasted cotton lint, yarns, cotton-based textiles, or blended fabrics. The acid pretreatment method involves decomposing the microstructure of cellulose fibers to expose the crystalline region with more reducing and non-reducing ends. This region can then be hydrolyzed by enzymes, which helps to release sugars by facilitating the enzymatic action. Additionally, the amorphous region of cellulose is hydrolyzed during this process [73].

5.3. Alkali Pretreatment

Alkali pretreatment involves the use of bases, such as sodium, potassium, calcium, and ammonium hydroxide, for waste textile materials [74–76]. The alkali pretreatment increases cellulose digestibility through enhancing lignin solubilization and decreasing cellulose crystallinity [77]. Another great advantage of alkali pretreatment is low inhibitor formation [66]. Normally, alkali pretreatment is carried out at room temperature but it has been tried at both lower and higher temperatures to improve the process. Jeihanipour and Taherzadeh (2008) pretreated cotton lint and jeans with 12% NaOH at 0 °C for 180 min for ethanol production. They observed that the pretreatment helped to achieve a more than 89% glucose yield after enzymatic hydrolysis by *S. cerevisiae*. They also noted that the alkali pretreatment gave a more than 30% better glucose yield than the phosphoric acid pretreatment [73]. Gholamzad et al. pretreated polyester/cotton blended fabrics with NaOH (7 wt%)/urea (12 wt%) at −20 °C, and claimed a 91% glucose yield after hydrolysis [78]. The NaOH/urea combination can be more useful in removing the hemicellulose of cotton, which enhances the surface contact of cotton cellulose during the enzymatic hydrolysis [79].

Studies have been carried out to examine when alkali pretreatment methods have been modified further to improve the efficiency of the process. Lin et al. proposed a new pretreatment method for treating cotton/PET textile waste to enhance the enzymatic hydrolysis of cellulose to glucose [80]. The conversion yield of cotton to glucose could be improved by over 98% and the residue PET fibers could be extracted for other applications.

The suggested pretreatment method is divided into three steps. Initially, the textile waste is treated with the NaOH/urea solution at 0 °C for 6 h, followed by a regeneration process in hot water, and finally, the substrate is digested by cellulase at 50 °C in acidic pH 4.8 for 72 h. The main advantage of this pioneering method is the significant enhancement in digestibility (80% cotton cellulose digested within only 72 h) of the waste textiles through the cellulose dissolution process, which is based on the cellulose dissociation theory suggested by Porro et al. [17]. Diluted NaOH treatment of cotton materials causes swelling, leading to an increase in the internal pore size and a decrease in the degree of polymerization and crystallinity. The addition of Na⁺ to the crystalline lattice leads to swelling of the native crystalline cellulose when it is in contact with a strong alkaline solution.

Increasing the digestibility of textile waste relies on the regeneration of cotton fibers. During the regeneration process, dissolved cellulose can shrink and form a new allomorph structure known as cellulose II, resulting in a morphological change. Cellulose I is characterized by parallel chains, while cellulose II is described as having an antiparallel structure that is more accessible to enzymes. NaOH is effective at changing the cellulose structure at lower temperatures due to the stronger binding of Na⁺ and OH[−] to water, which enables the breakage of hydrogen bonds within the cellulosic structure. Another feature

of this process is that PET fibers become less complex after recycling, and the remaining residue consists mainly of PET fibers that can be easily extracted. Finally, the low enzyme loading of 5FPU/g glucan, compared with other pretreatment methods, means that it is a cost-effective option.

5.4. Ionic Liquid Pretreatment

The chemistry of the anion and cation can be tuned to generate a wide variety of liquids, known as ionic liquids (IL) [81]. In recent years, pretreatment with ionic liquids has gained popularity due to the tunability of the solvent chemistry. The main advantages of this type of pretreatment are the ability to dissolve different types of biomasses, including cotton cellulose, lignin, and hemicellulose hydrolysis, and the mild processing condition. Despite these advantages, the main disadvantage of this process is the high cost of solvents. This is the reason that solvent recovery and recycling are required for this kind of pretreatment. The recovery of the IL depends on the vapor pressure; an IL with a low vapor pressure can be recovered for more than 99%, thus reducing the cost of solvent usage [82].

Zhang et al. used AMIMCl to treat un-dyed 100% cotton t-shirts [83], and after 90 min of pretreatment at 110 °C, a high sugar yield was achieved by using a reasonable amount of cellulase. Turner et al. [84] observed that the presence of this IL in cellulosic materials reduced the reactivity of cellulase enzymes during hydrolysis, which was reconfirmed by Hong et al. [85]. To overcome this problem, he suggested washing the cellulose materials with hot solvents before enzymatic treatment, which helped to achieve a very high sugar yield of 94%. De Silva et al. directly treated cotton/polyester blended yarns with AMIMCl at 120 °C for 6 h and dissolved the entire cotton portion, which was used for making regenerated cellulosic fibers [86]. The polyester portion was completely recovered in this process.

4-Methylmorpholine N-oxide (NMMO) is another solvent that can dissolve cellulose with a reasonable yield [87,88]. This process involves treating cotton materials at 80 °C with a low water content, which results in the dissolution of higher molecular weight cotton fibers. However, like AMIMCl, the presence of NMMO in cellulosic materials has a negative impact on enzymatic hydrolysis and fermentation [88,89]. Therefore, the treated materials need to be washed before enzymatic hydrolysis to improve the process. Techno-economic studies have shown that efficient recycling of NMMO is necessary to achieve an economically viable pretreatment process for cotton materials using NMMO [90].

5.5. Supercritical Fluid Pretreatment

A supercritical fluid (SCF) is any substance at a temperature and pressure above its critical point, where both gas and liquid phases coexist. Such fluids have the density of liquids but can penetrate like gas inside the cellulosic materials. This is the reason these liquids are used for pretreatment of cellulose-based biomasses [91]. The main advantages of the SCF pretreatment process are the low degradation of sugar and low costs, but high pressure is needed for this pretreatment and so special reactor vessels are required. Supercritical CO₂ (SF-CO₂) has been widely used as an extraction solvent [88]. In aqueous solution, CO₂ forms carbonic acid and can improve the hydrolysis of polymers. Liu et al. studied the performance of the enzymatic hydrolysis of cotton fibers pretreated with supercritical CO₂ [87]. Saka and Ueno investigated the impact of supercritical water (500 °C, 35 MPa) on glucose yields from various types of cellulosic fibers (such as cotton linter, ramie, rayon, etc.) and found that the performance was similar to an acid treatment with enzymatic hydrolysis [90].

6. Fungal Cellulase Production from Textile Waste by Solid-State Fermentation

At present, commercial cellulase is typically produced from soft rot fungi using solid-state fermentation (SSF) and submerged fermentation (SmF) [92]. SSF is a fermentation process that uses solid materials with low water activity as a substrate, providing a cost-effective method for producing cellulases using natural polymers derived from agro-

industrial residues [93,94]. In contrast, SmF occurs with a free-flowing suspension, is often associated with low concentrations of the end product, requires additional downstream processing steps, and consequently leads to high costs of cellulase production [95].

Cellulase contributes to approximately 40% of the total cost of the bioprocess; therefore, low-cost cellulase is highly desirable for an economically viable process. *Aspergillus*, *Trichoderma*, and *Thermoascus auranticus* are well-known producers of cellulases and are commonly used in SSF on lignocellulosic substrates such as agricultural and plant biomasses [94,96,97]. Horticultural wastes and agricultural by-products, such as rice straw, have been adopted as cost-effective substrates in cellulase production in the past decade [94,98].

Until recently, cotton-based textile waste has not been utilized as a substrate in SSF for cellulase production. However, Lin and her colleagues developed a new approach to use cotton-based textile waste for fungal cellulase production through SSF [99]. They screened six fungal strains, including *T. reesei*, *A. niger* N402, *A. niger* CKB, *Rhizomucor variabilis*, *A. oryzae*, and *T. longibrachiatum*, under different moisture conditions (65–85%) on cotton fabrics. All strains were able to grow on cotton fabrics, and their cellulase activity was evaluated after 7 days in terms of filter paper activity (FPU/g), which indicates the degradation activities of the cellulase enzymes (see Table 2). Among the six fungal strains, *A. niger* CKB yielded the highest cellulase activity (0.4 FPU/g).

Table 2. Cellulase activities of different fungal strains under various moisture conditions after 7 days [99].

Name of Strain	Optimum Moisture Condition (%)	Maximum Cellulase Activity (FPU/g)
<i>A. niger</i> CKB	70–75	0.43
<i>A. niger</i> N402	85	0.42
<i>T. reesei</i>	85	0.11
<i>R. variabilis</i>	65	0.20
<i>A. oryzae</i>	65–75	0.19
<i>T. longibrachiatum</i>	85	0.11

They applied different pretreatment methods, such as autoclaving, freezing alkali/urea soaking, and milling, on six types of cotton and cotton/polyester-based textiles to explore the effects on cellulase activity. Autoclaving was observed to be the optimal modification strategy for pretreatment due to resultant cellulase activity at the highest level. It could be attributed to the textile morphology modified by the mild hydrothermal treatment via autoclaving (121 °C, 15 psi, 15 min), which partially removed the coating of the cellulosic fibers and better exposed the cellulose to the fungus cellulase.

During the optimization process [99], it was observed that the highest cellulase activity was obtained at 28 °C. Other important factors, including moisture content, inoculum size, pH, and yeast extract concentration, were also optimized. Under the optimized SSF with a moisture content of 78%, inoculum size of 3.10×10^7 spore/g, pH 7.29, and yeast extract of 2.24%, it was noticed that the cellulase activity improved by approximately 14%. When this method was compared to other existing processes, it was observed that β -glucosidase obtained in this study had the highest activity with a significant improvement against other existing methods.

Utilization of Fungal Cellulase to Treat Textile Wastes

There have been limited studies on the use of cellulase enzymes to treat cellulosic fabrics. In one study by Lin and coworkers (2019), commercial cellulase (Celluclast 1.5 L, Novozymes) and β -glucosidase were applied to pretreated cotton/polyester blended fabrics [28,97]. The fabrics were pretreated using a mixture of 7% (*w/v*) NaOH and 12% (*w/v*) urea solution at 0 °C for 6 h and then washed thoroughly. Glucose yields were measured after 0, 9, 12, 24, 48, 72, and 96 h. The pretreatment increased the glucose yield by 30% compared to untreated textile materials. Additionally, the maximum glucose yield was over 98% after 96 h of distillation at temperatures ranging from 45–55 °C, with a cellulase dose of 40 FPU/g.

Lin et al. (2019) also compared their different textile-based fungal cellulases against commercially available cellulases (Novozyme, Cellulast 1.5 L) on a variety of textile fabrics; 100% cotton fabrics, denim fabrics, and different cotton/polyester blended fabrics [28]. The result showed that fungal cellulose extracted from cotton fabrics gave a 7% higher glucose yield compared to the commercial cellulose. The optimization of fermentation media revealed that pretreated textiles and Mandels medium were the preferred substrates for cellulase production. Using a cotton/PET (40/60 blended) based textile with 1% cellobiose addition, *T. reesei* ATCC 24449 achieved the highest fungal cellulase activity of 18.75 FPU/g. Fungal cellulase obtained from SmF resulted in similar hydrolysis yields as commercial cellulase in the hydrolysis of textile waste. Figure 2 shows the flow diagram for cellulase production from textile waste and the utilization of produced cellulase for the valorization of textile industrial waste.

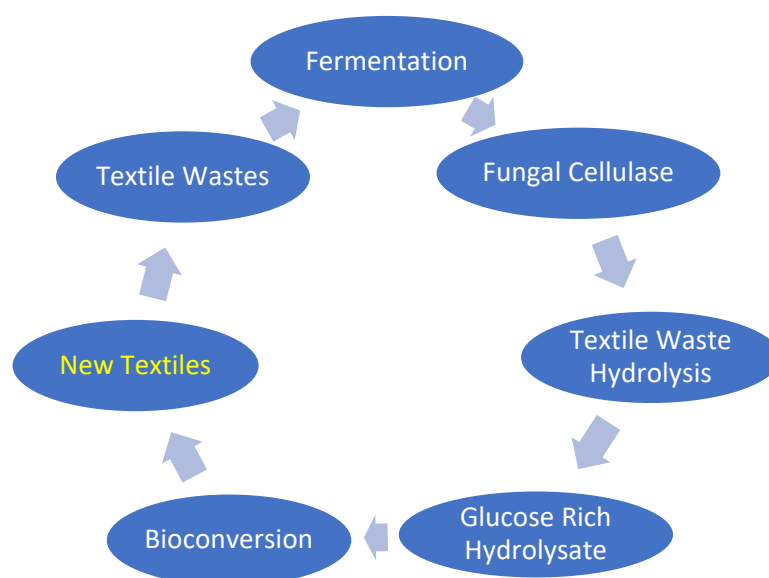


Figure 2. A flow diagram depicting the lifecycle of cotton textile waste. (The yellow highlighted the starting point of the life cycle).

The above has reviewed sustainable strategies to recover cotton from textile wastes to identify new sources of materials for future textile application. Various pretreatments of textile waste are introduced, followed by the enzymatic treatment of textile materials. In addition, the utilization of cotton textile materials for fungal cellulase production by SSF is illustrated; fungal cellulases are used to treat textile wastes for the extraction of glucose and to separate synthetic polymers, thus a biorecycling process is achieved.

7. Regenerated Fibers from Waste Materials via Electrospinning

The next step after obtaining recycled textile materials is to regenerate fibers from the waste materials, and the focus of this paper is electrospinning. There are several methods used to reprocess textile waste into fibers. Some of the most common methods include mechanical, chemical, thermal, biological, and electrospinning. Mechanical recycling involves shredding textile waste into smaller pieces and then using various mechanical processes to transform them into fibers that can be used to produce new textiles. It has the advantage of low energy consumption, minimal use of chemicals, and can be scaled up efficiently. However, the resulting fibers may not be suitable for high-quality textiles. During the process, fibers could be damaged, and their strength and durability are reduced. Chemical recycling has the advantage of producing high-quality fibers and the process is suitable for a wide range of textile waste; it can also be scaled up efficiently. However, the process requires the use of chemicals that can be harmful to the environment and results in the high consumption of energy. Thermal recycling is suitable for a wide range of

textile waste to produce high-quality fibers; however, again, high energy could be required, and it could produce harmful emissions. Now all types of textile waste could be suitable. Biological recycling has the advantage of a lower energy demand and the chemical use could be minimized. However, the process may take longer than other methods and may not be suitable for various kinds of textile waste. Electrospinning is an efficient method for synthesizing nanofibers with numerous desirable parameters, including fiber diameter, specific surface area, interconnectivity, and rigidity [100], and is suitable for a wide range of applications. Electrospun nanofibers have a high surface area-to-volume ratio, which makes them highly suitable for various applications, such as filtration, tissue engineering, and drug delivery. It allows for the production of nanofibers with a tunable diameter and morphology, which can be tailored based on the specific application requirements. In addition, it can be easily scaled up to produce large quantities of nanofibers, making it a commercially viable technique. However, one should bear in mind that electrospinning equipment can be expensive, which can limit its accessibility to some researchers and industries. The electrospinning process has a relatively low throughput, which may not be suitable for large-scale production. The process involves many parameters, such as solution properties, process conditions, and collector type, which can be challenging to optimize for a specific application and the spun fibers can sometimes have poor mechanical properties, such as low tensile strength and poor durability, which may limit their use in certain applications.

There are also different types of electrospinning, which should be carefully selected for different purposes. Solution electrospinning is the most common type of electrospinning, where a polymer solution is used as the spinning material. The solution is electrospun into nanofibers, which are collected onto a substrate [101]. In the melt electrospinning technique [102], the spinning material is a melted polymer, which is electrospun into nanofibers. This method is used for thermoplastic polymers that can be melted and solidified without undergoing chemical changes. Recently, coaxial electrospinning became an emerging technique to produce nanofibers with core-shell structures [103]. It involves the use of two or more concentrically arranged needles, where different materials are electrospun from each needle. The emulsion electrospinning technique [104] involves the use of an emulsion as the spinning material, where droplets of one material are dispersed in another material. The emulsion is electrospun to produce nanofibers with a core-shell structure. Electroblowing [105] is a modification of electrospinning, where compressed air is used to assist in the electrospinning process. This results in the production of nanofibers with a larger diameter than those produced by conventional electrospinning. Needleless electrospinning [106] eliminates the use of needles and instead uses a high-voltage electrode to generate a charged liquid jet, which is then collected onto a substrate to produce nanofibers.

The simplest electrospinning system comprises three components: a method for the influx of the polymer solution, a grounded metallic surface for the collection of ejected electrospun fibers, and a high voltage source to induce an electric field (between the spinneret and the grounded conductive collector) where these components together create a straight liquid jet of polymer material that deposits on the surface of the collector [107]. Ultra-fine fibers can be produced due to the quick evaporation of solvents together with the subsequent stretching and whipping processes [108]. Fiber parameters are closely dependent on processing variables, such as voltage, distance between the emitter and collector, humidity, and polymer solution properties, which have significant effects on the outcome of the fiber characteristics [107].

Electrospinning essentially works by the interaction between the electric charges in the polymer solution, the cellulose solution, and the external electric field. The external electric field induces a charge on the surface of the solution and the increase in electric field elongates the surface of the solution droplet and leads to the formation of threadlike fibers [107]. The advantages of jet electrospinning over other spun methods are that the small diameter of the jet enables rapid drying of the solvent and offers extreme confinement

and alignment of polymer chains as well as nanocellulose, locking them rapidly to avoid central relaxation of fibers of higher diameters to be synthesized by other methods [108].

Wet spinning is a blend of rheological and diffusional behaviors in which the orientation of fibers dictates the structural formation as well as the mechanical properties of fibers, thus being the primary method for regenerating cellulose fibers. The rheologic portion, or the viscoelastic property of the fibers, is responsible for the formation of a continuous fiber without breakage and the elastic behavior of the polymer solution during spinning. The diffusional portion consists of two parts: the diffusion of solvents from wet fibers into the solution and the coagulant from the solution into the wet fibers. Under the combined effects of the rheological and diffusional phenomena, the wet fibers precipitate out of the solution upon contact with the coagulant [109]. However, wet spinning is not the focus of this paper as the topic has already been covered by several previous literature reviews. Moreover, electrospinning presents a multitude of advantages, with the best one being its ability to cover items with nanofibers that are light in weight, hence displaying potential suitability in futuristic applications.

Cellulose is the most common naturally occurring polymer in the world and is structurally a linear polysaccharide with glucose being its monomer connected via β -1,4-glycosidic bonds. Due to its notoriously low solubility, cellulose has remained a challenge to dissolve or melt despite its abundance. Numerous strategies have been employed to increase its solubility, including the use of ionic liquids, the use of different solvent systems, and the turning of cellulose into a more soluble form. This is crucial as cellulose can be electrospun into nanofibers, but this requires cellulose to be dissolved into solution. Cellulose derivatives, such as cellulose acetate, methyl and ethyl cellulose, and carboxymethyl cellulose, have achieved better solubility than cellulose in both organic and inorganic solvents [107].

A multitude of optimized solvents for dissolving cellulose for electrospinning conditions have been developed by researchers, including a lithium chloride and N,N-dimethylacetamide (LiCl/DMAc) as well as a trifluoroacetic acid and polyvinyl alcohol (TFA/PVA) solvent system [100]. It is believed that solvent properties, such as semi-conductivity with moderate charge capacities and high volatilities, are ideal for efficient electrospinning of cellulose fibers. However, the solubility of cellulose in these solvent systems is still very dependent on the intrinsic factors of cellulose, notably its source, molecular weight, and crystallinity. On the other hand, the dissolution of cellulose by TFA takes place via the attack of the strong organofluorine acid at the glycosidic bonds, together with the low boiling point and low viscosity, which allow quick evaporation of the solvent upon deposition. In addition, experiments showed that the addition of PVA as the co-solvent was crucial for the formation of continuous nanofibers instead of beaded particles; this was attributed to the hydrogen bonding interactions between the PVA and cellulose materials. One main advantage of using the TFA/PVA over LiCl/DMAc solvent system is that the former produces fewer viscous solutions, which is important for continuous fibers via electrospinning. It also demonstrates the ability to dissolve hemicellulose as well as lignin, which is very common in both plants and recycled paper-derived materials [107].

Cellulose can be regenerated from cellulose derivatives upon electrospinning. In the case of cellulose acetate, this was performed by first dissolving the cellulose acetate in an optimized solvent system, which was then subsequently deacetylated back into the cellulose as a post-electrospinning work up. These cellulose fibers derived from cellulose acetate exhibit autonomous thread-like morphology, consistent diameters, a large surface area, and an exceptional storage capacity, thereby demonstrating the practicality of regenerating cellulose with the electrospinning method [107].

Electrospinning of cellulose is a process of much interest in recent decades and research advancement has helped to overcome the limitation of its low solubility in common solvents. This, together with dispersed instead of dissolved nanocellulose in various organic and inorganic solvents, allows the incorporation of nanocellulose into nanofibers via electrospinning while maintaining the nanostructure and properties, such as the high

mechanical strength, aspect ratio, as well as the biodegradability. The effects of various electrospinning parameters on the fiber morphology of virgin cellulose have been studied and can be summarized in Table 3 below [108]. Recycled polymeric materials may have different physical, chemical, and mechanical properties compared to virgin materials due to the presence of impurities, additives, and degradation products. These impurities and additives can affect the polymer's molecular weight, crystallinity, and thermal stability, which can influence the electrospinnability and properties of the resulting fibers. The presence of impurities in recycled polymeric materials can also affect the fiber morphology, diameter, and uniformity. For example, the presence of small particles or contaminants may cause clogging of the spinneret or result in the formation of beads instead of fibers. Therefore, to optimize the electrospinning process for recycled polymeric materials, it may be necessary to modify the process parameters, such as the solvent system, spinning conditions, and post-treatment methods, to account for the differences in the properties of the recycled materials.

Table 3. Effects of electrospinning parameters on fiber morphology.

Electrospinning Parameters	Effects on Fiber Morphology
Increased polymer concentration, i.e., increasing viscosity	Formation of longer fibers with increased fiber diameter
Increased conductivity	Formation of beaded fibers with decreased fiber diameter
Increased surface tension	Formation of beaded fibers
Increased flow rate	Formation of continuous fibers with decreased fiber diameter
Increased voltage	Formation of beaded fibers with increased fiber diameter
Increased distance between the spinneret and the collector	Decreased fiber diameter
Increased humidity	Overall decreased fiber diameter and caused uneven diameter distribution
Temperature	Had no direct effect on fiber morphology

8. Conclusions and Recommendations

The advancements in textile recycling technology and the circular economy could not only bring economic benefits, but also reduce many environmental problems caused by high energy and water consumption and a high carbon footprint. Nonetheless, there are still hurdles for these bioprocesses to be commercialized. Textile waste is always a mixture of polymers, such as cotton, polyester, nylon; each of these polymers requires specific treatment methods. Therefore, the sorting and separation of textile waste for recycling should be a critical step before the bioprocessing treatment. Manual sorting requires exhaustive labor and will lead to difficulties in identifying specific fabrics being used for either generally lower grade applications, such as rags, or for higher grade applications in which such materials could supplement the use of virgin fibers. Recent sorting technology employs near-infrared (NIR) spectroscopy for sorting fibers according to their composition and color. While this technology has been employed to the PET bottle recycling procedure [110], it is highly recommended for the textile waste treatment process. Another concern is the impurities, such as fiber-bonded dyestuff, that may be retained after the bioprocessing treatment process. Non-environmentally friendly solvents, such as dimethyl sulfoxide, are often used to decolorate the recycled fibers. Therefore, research should also be focused on exploring more environmentally friendly decolorants.

With further research and technological advancements, bioprocessing and electrospinning show promise for textile waste recycling. Adopting a circular economy approach and integrating recycling with sustainable fiber production could help reduce environmental impacts and resource depletion within the textile industry. However, government policies, industrial symbiosis, and consumer awareness will also be crucial to enable a transition towards a sustainable and circular future for the textile sector. Overall, this study has highlighted the potential of using bioprocesses and electrospinning to generate value from textile waste while regenerating sustainable cellulose fibers, as well as investigating a range

of bioprocessing and electrospinning methods for recycling cotton from textile waste in a sustainable manner. Pretreatments, including acid and alkali methods, can modify cotton fibers for enzymatic hydrolysis using cellulases, while solid-state fermentation produces fungal cellulases from textile waste that can hydrolyze cotton. Electrospinning regenerates cellulose nanofibers from waste cellulose that could be used in various applications. Further optimization and innovations building on these methods could advance the more environmentally friendly management and recycling of cotton textile waste. Finally, the recycling procedure itself, when being scaled up, consumes a large amount of water and energy and the lengthy procedural steps may involve the use of large-scale reactors, which may incur extra costs to the total capital investment. Thus, the process itself may not be as economical as the manufacturing process of the original fiber materials. This may discourage the recycling industry to consider textile recycling; therefore, concepts such as industrial symbiosis should be introduced in the process of plant design for the future development of the industry [111].

Author Contributions: Conceptualization, E.M.-H.C. and C.-W.T.; methodology, D.W.-S.S.; formal analysis, D.W.-S.S., Y.-Y.L. and R.H.-P.L.; investigation, D.W.-S.S.; resources, P.W.-K.T.; data curation, S.O.; writing—original draft preparation, E.M.-H.C. and C.-W.T.; writing—review and editing, D.W.-S.S., Y.-Y.L. and R.H.-P.L.; visualization, R.H.-P.L.; supervision, C.-W.T.; project administration, P.W.-K.T.; funding acquisition, P.W.-K.T. All authors have read and agreed to the published version of the manuscript.

Funding: This work described in this paper was fully supported by the Environment and Conservation Fund (Project No. ECF 54/2020), University Grants Committee—Research Matching Grant Scheme (RMG024), and a grant from the Research Grants Council of the Hong Kong Special Administrative Region, China (Project No. UGC/IDS(R)25/20).

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Conflicts of Interest: The authors declare no conflict of interest.

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