Progress toward Circularity of Polyester and Cotton Textiles

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Abstract: Millions of tons of textile waste are landfilled or incinerated in the world every year due to insufficient recycle value streams and the complex composition of textile end products. The goal of this review is to highlight pathways for simplifying and separating textile wastes into valuable raw material streams that will promote their recovery and conversion to useful products. The discussion focuses on advances in sorting, separation, decolorization and conversion of polyester and cotton, the two most common textile fibers. Sorting processes are gaining automation using spectroscopic methods that detect chemical composition differences between materials to divide them into categories. Separation, through dissolving or degrading, makes it possible to deconstruct blended textiles and purify polymers, monomers and co-products. Waste cotton can produce high quality regenerated cellulose fibers, cellulose nanocrystals (CNCs) or biofuels. Waste polyester can produce colored yarns or can be chemically converted to its starting monomers for the recreation of virgin polymer as a complete closed loop. The current strategies for decolorization are presented. Life cycle assessment (LCA) studies found that recycling polyester/cotton blended fabrics for subsequent uses is more sustainable than incineration, and research on producing biomass-based poly-ester also offers feasible avenues for improving textile sustainability and promoting circular processing.

Keywords: circularity; cotton; dye removal; polyester; recycling; sustainability; textiles

1. Introduction

The textile industry has an estimated global market value of USD 1.0 trillion [1]. This major manufacturing enterprise produces millions of tons of fibers every year while at the same time generating tons of post-consumer waste. Clothing accounts for more than 73% of the textile revenue share, with technical textiles and household goods constituting the other main segments [1]. On a volume basis, global textile fiber production has almost doubled in the last 20 years, from 58 million metric tons in 2010 to 109 million metric tons in 2020 [2]. Two fiber types—polyester (52% volume share) and cotton (24% volume share) dominate the industry because of their relatively low cost, outstanding performance, and complementary characteristics [2]. Cotton fibers are primarily composed of cellulose. These fibers grow naturally as the seed hairs of plants belonging to the genus *Gossypium* [3]. Textile polyester is a synthetic fiber. According to generic definitions for manufactured fibers listed in the U.S. Code of Federal Regulations (16 C.F.R. § 303.7 (2022)) [4], the textile fiber term “polyester” refers to fibers “composed of at least 85% by weight of an ester of a substituted aromatic carboxylic acid,” which includes fibers made from poly(ethylene terephthalate) (PET). Since most polyester fiber produced today is made using PET polymer, use of the term “polyester” when referring to a textile fiber still usually implies it is made from PET, but the actual composition of specific polyester fibers could be copolymers or could be distinctly different types of polyester, such as “triexta” where the glycol used to form the ester is 1,3-propanediol rather than ethylene glycol (EG). In the present discussion, “polyester” refers to PET-based polyester fibers.

Desirable PET fiber properties include high strength, chemical and wrinkle resistance, and quick-drying properties, while cotton is favored for its high strength, color retention,
moisture absorption and comfort. Together, these two fibers alone make up > 75% of global fiber production, and hence comprise the majority of textile waste. The global fiber production number is even higher (88%) when other plant based cellulosic fibers are included. According to 2018 data from the U.S. EPA (Environmental Protection Agency), 17 million tons of textiles were disposed as municipal solid waste (MSW) in the U.S., while the recycling rate was only 14.7% [5]. Global estimates are that 65–92 million tons of textiles are wasted every year and that less than 20% of this amount is recycled [6,7]. These numbers mean that more than 80% of textile waste is landfilled or incinerated, which eventually does harm to the environment and is a loss of valuable raw materials. Growing attention paid to textile waste issues has motivated increasing research on textile recycling. A bibliometric analysis in the Web of Science database shows that the number of publications in the field of textile recycling has increased more than five times from 2011 to 2021 (Figure 1). If the keyword “circular*” is added in the search criteria “textile*” and “recycl*”, the number of publications drops by ten times, indicating that the new emerging concept of circularity for addressing textile recycling still needs more attention.

![Figure 1: Bibliometric analysis of publications about textile recycling between 2011 and 2021. The data were retrieved from the Web of Science database in August 2022, using the criteria that all fields were searched with the keyword combination “textile*” and “recycl*”.](image)

Diverting textile waste from landfills requires increased implementation of existing “open-loop” recycling approaches and demands more advanced “closed-loop” (or “circular”) strategies. Open-loop recycling means materials are collected and reused—often in a “downcycled” form with lower value than the original material—but are eventually disposed. Delaying the final disposal helps reduce total waste by decreasing the amount of virgin material produced for an intermediate application. To be successful, the quality and cost of the recycled material must match the application requirements. Among plastics, PET has the most advanced post-consumer recycling infrastructure [8]. This is because the PET polymer quality required to produce textile fibers can be obtained by mechanical recycling of PET packaging, namely PET beverage bottles. The commercial value of recycled PET (rPET), which offers sustainability benefits [9], is sufficiently high to justify the recycling costs. The term “mechanical recycling” in this context refers to physically sorting and chopping the waste materials (bottles) into chips, followed by melting the thermoplastic polymer to produce clean resin pellets that are afterwards used as raw materials in melt-spun PET fiber production. Reductions in polymer molecular weight and intrinsic viscosity occur during bottle chip melting that limit rPET from being reused for bottle-grade products.
However, the resulting resin properties are sufficient for spinning textile fibers [10]. In the United States, Unifi is a major converter of post-consumer PET bottles to rPET products, including PET resin, fibers and yarns [11].

After PET is converted to end-user textiles, the recycling options are more limited. Trade organizations, such as the Secondary Materials and Recycled Textiles (SMART) Association, promote economic opportunities for textile reuse and recycling. Business scenarios are based on extending the value, lifetime and sustainability of textiles, through industrial raw material recycling and small enterprise custom upcycling, clothing resales, or garment cutting for the industrial wipes market. These efforts focus on the recycling of pre-consumer materials and reuse of post-consumer products [12]. Industrial or post-consumer textile wastes that cannot be reused in their fabric form can be delivered to companies such as Leigh Fibers, the largest textile waste processor in North America, for mechanical shredding (also called “garnetting”) and conversion to fiber bales [13] for subsequent use in nonwoven fabrics, in composites, as fillers or for insulation [14]. Eventually, end-products from these recycle streams are disposed, leaving their fate an open loop.

“Closed-loop” recycling means that materials are reprocessed in ways that preserve their properties and value for reuse within the same product value chain, such as bottle-to-bottle or fiber-to-fiber recycling [15]. Although commercial closed-loop recycling of paper, glass and aluminum products is familiar, closed-loop recycling of post-consumer textile waste is only starting to gain traction with technological and logistical advances. One leading example is the Renewcell process [16] that claims “recycling clothes finally works” by allowing reprocessing of any cellulosic textile waste into Circulose® branded dissolving pulp that can be used to make viscose, lyocell, acetate and other types of regenerated (or “manmade”) cellulosic fibers [17]. This process uses an alkaline-oxygen treatment to disperse cellulose in a partially dissolved, partially particulate form that can be filtered to remove contaminants before precipitating the solid pulp. Similar strategies have been used to dissolve, purify and recover PET polymer using organic solvents [18]. In both these cases the polymer properties are sufficiently preserved during solution processing to extrude fibers for at least one cycle.

Repeated stresses of use and recycling will eventually cause extensive polymer degradation and loss of properties. Once this occurs, “chemical recycling” is the most promising route to closing the recycle loop because “chemical recycling can transform lower grade inputs and materials that are difficult to recover into higher grade outputs” that allow materials to “cascade up and down the quality ladder in response to market demand” [15]. This is accomplished, for example, by breaking chemical bonds and completely converting polymers back into pure monomer starting materials. After separation and purification from contaminants (e.g., other fibers, colorants, catalysts), the monomers are fed back into polymerization reactions to recreate high molecular weight polymers that are equivalent to virgin polymers and can be used in the highest value applications. Achieving competitive process economy is a persistent obstacle for recycled monomers, which have to compete with well-optimized virgin raw material supply chains. However, evolving market pressures (oil scarcity) and consumer demands (sustainability) are tilting the scale in favor of increased recycling. Lack of awareness and technical knowledge are among the obstacles that need to be overcome in achieving circular supply chains for sustainability in the textile industry [19]. Therefore, the following discussion presents strategies for open- and closed-loop recycling of cotton, polyester and their blends to enlighten and inspire further innovations that will support robust circular pathways for these important textile fibers.

2. Origin and Properties

Cotton and polyester are the two most important textile fibers (Figure 2). They have remarkably different origins and properties, yet their differences are complimentary in ways that make it desirable to use these fibers as intimate blends in clothing and other textiles, leading to end-of-life recycling challenges.
Cotton fibers are seed hairs from plants of the genus *Gossypium*. Commercial varieties produce 22–36 mm long (“staple length”) fibers, where each individual fiber is a single plant cell with a maximum cross-sectional width of about 20 µm and no two cotton fibers are exactly alike [3]. The cells grow with a thick cell wall composed primarily of cellulose (>88%) with lesser amounts of protein, wax, pectin and other trace substances [20]. Reinforcing layers of cellulose microfibrils forming the cell wall contribute to fiber strength. Dry cotton fibers have a thin ribbon-like shape that allows them to be twisted together to form fine strong yarns. As a biopolymer, cellulose is synthesized from uridine diphosphate (UDP)-activated glucose monomers in a simultaneous synthesis and translocation process across the cell membrane [21] by the cellulose synthase complex (CSC) which acts as a nanoscale-fibril spinneret to form cellulose microfibrils [22]. A common model of the CSC is a rosette structure with six subunits where each unit contains three cellulose synthase (CESA) proteins (Figure 3a). Ongoing research indicates that more complex CSC structures for cotton fibers could include a 36-mer-like cellulose synthase supercomplex (CSS) forming 72-chain microfibrils in cotton fibers [23,24]. Nascent cellulose polymer chains emerging from a CESA spontaneously assemble with adjacent chains to form microfibrils [25,26] that wrap in diagonal layers around the cell to build up the plant cell wall and ultimately form a cotton fiber that is large enough to be seen by the naked eye [3]. After harvesting, cleaning, and carding, cotton fibers are twisted together during the spinning process to form yarns that are afterwards interlaced by weaving or knitting to produce fabrics [27].
While there is a remarkable conceptual similarity between the formation of cellulose microfibrils by the CSC and the extrusion of PET fibers from a spinneret, the physical scale of the processes is vastly different. Elementary cellulose microfibrils, which self-assemble immediately as polymer synthesis proceeds, are about 3.5 nm in cross-section [31] while typical PET fibers have diameters of around 10 µm, depending on the desired fiber size.

PET is a synthetic polymer obtained by condensation polymerization of two monomers, terephthalic acid (TPA) and ethylene glycol (EG). The resulting PET resin is extruded into continuous filament fibers by melting spinning (Figure 3b), in which liquid molten polymer is pushed through small holes to form long strands that solidify when meeting cold air, followed by drawing (stretching the fiber) to increase fiber crystallinity and strength [30]. The polymer chains within each PET fiber adopt a semi-crystalline morphology, where regions of crystallinity are interspersed with amorphous regions. The crystalline regions provide strength; the amorphous regions provide flexibility and toughness. The fibers are further processed into yarns, often after cutting the long filaments into shorter staple fiber lengths. While there is a remarkable conceptual similarity between the formation of cellulose microfibrils by the CSC and the extrusion of PET fibers from a spinneret, the physical scale of the processes is vastly different. Elementary cellulose microfibrils, which self-assemble immediately as polymer synthesis proceeds, are about 3.5 nm in cross-section [31] while typical PET fibers have diameters of around 10 µm, depending on the desired fiber size.

A comparison of cotton and polyester properties (Table 1) shows similarities in strength, modulus and elongation at break that make both materials useful as textile fibers, and shows major differences in moisture regain and degradation time. Polyester is generally stronger than cotton, is wrinkle resistant (due to absence of intermolecular hydrogen bonds) and can be dyed efficiently in vibrant colors, primarily using disperse dyes, but it absorbs less water and requires a much longer time to degrade in the environment. Hydrophobicity, crystallinity and a rigid, non-natural polymer composition contribute to poor PET biodegradability [6]. Resistance to degradation is a valuable feature for in-use applications, but results in waste persistence and negative environmental impacts [7]. Gómez and Michel found that less than 1% (cumulative carbon loss) of a PET plastic was biodegraded during 660 days of soil incubation [32]. The sample used was amorphous, whereas polyester fiber possesses higher crystallinity, which means PET fiber will take an even longer time to degrade, or can be considered non-biodegradable. Li et al., studied biodegradation of polyester fabrics, finding that after a small initial degradation (~13% C converted to CO₂) in the first month, the polyester fabric stayed intact for the rest of two test months in natural soil [33]. By some estimates, it would take 16 to 48 years for PET to degrade when exposed to abiotic environmental stresses such as moisture and UV (ultraviolet) radiation [34,35]. In contrast, scoured (natural waxes removed) undyed cotton is hydrophilic and biodegrades rapidly (≤6 months) in the presence of moisture and

Figure 3. Schematic illustrations of fiber formation in natural and manufacturing processes. (a) Schematic representation of a rosette CSC producing cellulose polymer chains that self-assemble to form an 18-chain fibril, inspired by refs. [28,29]; (b) Schematic diagram of a spinneret for producing continuous PET filament fibers by melt spinning, where each fiber strand contains many PET polymer molecules.

(a) (b)
enzymes secreted by environmental microorganisms that metabolize the glucose monomers in cellulose as a food source [6]. Application of dyes, especially “reactive” dyes that form covalent bonds with cellulose, and other finishing chemicals on cotton can significantly inhibit biodegradation rates. These auxiliary chemicals also create more obstacles for recycling, such as the need to separate materials by color.

Table 1. Properties of cotton and polyester compiled from ref. [36] and other sources ∗.

| Properties          | Cotton         | Polyester      |
|---------------------|----------------|----------------|
| Fiber Cross Sectional Shape | kidney-bean   | circular (or variable) |
| Specific Gravity    | 1.54           | 1.38           |
| Moisture Regain (%) | 7.0–8.5        | 0.4            |
| Crystallinity (%)   | 53–80          | 40–55          |
| Strength (GPa)      | 0.6, 0.31–0.73 4, 0.38–0.69 5 | 0.8, 0.36(LT) 1.17(HT) 5, 0.55–0.99 6 |
| Young’s Modulus (GPa) | 8, 7.1–15.7 4 | 15, 6.9–20.7 6 |
| Elongation Break (%) | 7, 6–13.5 4   | 15, 40(LT) 7(HT) 5 |
| Degradation (yrs)   | <0.5 7         | 30–35, >100 9  |

∗ Values with a superscript are cited from the following references: 1 data cited from ref. [37]; 2 data cited from ref. [38]; 3 data cited from ref. [39]; 4 data cited from ref. [40]; 5 data cited from ref. [41] where LT means low tenacity, HT means high tenacity; 6 data cited from ref. [42]; 7 data cited from ref. [6]; 8 data cited from ref. [34], which is the estimated degradation time of PET films and bottles under wet soil; 9 data cited from ref. [32], which is the estimated degradation time of blow molded PET plastics under soil incubation.

The historical use of cotton and other natural fibers in clothing far exceeds, by millennia, the timeline of synthetic fiber use, which first became prevalent in the 1950–1960s. This could in part account for cotton’s persistent favorable reputation among consumers. According to a survey by Cotton Incorporated’s Lifestyle Monitor™, more than two in three consumers (68%) say that cotton clothing is better quality than clothing made from other manmade fibers such as polyester or rayon, and greater than 80% of consumers say cotton is the most comfortable, sustainable, trustworthy, soft, authentic and reliable compared to manmade fibers [43]. The perception of comfort is related to cotton’s superior water absorption, moisture wicking and breathability compared to polyester. However, a major economic incentive for using polyester is its low cost. In June 2022, Fiber2Fashion market data reported average trading prices of ~1.1 USD/kg polyester staple fiber, whereas cotton was ~1.9 USD/kg [44]. Cotton prices fluctuate considerably, though lower prices (average 1.29 USD/kg for US Upland cotton [45]) during the 2019–2020 season were still higher compared to polyester. Polyester/cotton blends are made as a way to get the best of both worlds—comfort, strength, easy-care and product economy. In view of their distinctly different chemo-physical properties, different strategies are needed when recycling these materials, which begins with sorting to classify the composition of textile waste and thereafter carrying out selective deconstruction and recycling processes.

3. Sorting

Sorting is the critical first step in textile waste recycling, because the presence of multiple textile components will increase the difficulty of recycling one specific material. Contaminants reduce the quality of the component targeted for recycling.

Textile sorting is often completed manually, but automated technologies are needed in regions where labor costs are high. Infrared (IRS) and near-infrared (NIRS) spectroscopic methods that detect chemical composition differences between fiber types are the most advanced. In comparison with standard chemical methods [46], they have a fast response time and can distinguish various textile types even if there are only slight differences among their spectra. Research by Riba et al., focused on developing a fast and accurate multivariate mathematical method for a direct and non-invasive sorting and classification of different textile fibers used for clothing [47]. The study included FTIR (Fourier-transform infrared) spectral data from 350 textile samples including natural, artificial and synthetic fibers, recorded in the wavenumber range 4000–650 cm⁻¹, with a resolution of 1 cm⁻¹. Results
showed that incoming unknown fiber samples were automatically classified with 100% accuracy and high speed, with no need to apply any prior analytical treatment to the textile samples. However, it was observed that dirty, wet or contaminated textiles could lower the accuracy. An NIRS technique studied by Cura et al. [48] was useful for categorizing textile waste, however, some limitations, such as sample thickness, significantly impacted the recognition process because NIRS only detects the surface of textiles. This study with NIRS-based sorting detection expanded on prior work by noting that sorting methods should be able to reject textiles that do not meet recycle purity requirements, such as rejecting textiles coated with auxiliary chemicals, such as polyurethane.

Zhou et al. [49] successfully utilized NIRS to identify fiber types including cotton, wool, PET, polylactic acid (PLA) and polypropylene (PP). They found it critical to preprocess spectra to eliminate baseline shifts which are caused by nonhomogeneous distribution of sample quality. Pattern recognition methods were used to further increase the identification efficiency. It is also feasible to measure cotton content in cotton/polyester blends via NIRS technology according to the experiments carried out by Rodgers and Beck [50]. Although some matrix differences such as dyes, finishes and fabric construction may significantly impact the intensity of NIRS spectrum results, using Deviation Math techniques or other normalization treatments can minimize the differences, making NIRS technology applicable to sorting all types of textiles. Similarly, polyester content in blended textiles can be estimated based on hyperspectral near infrared imaging [46]. A wavelength range of 1000–2500 nm was chosen as it was not significantly affected by the color of textile samples.

Digital cameras were used by Zhou et al., to detect the color of waste textile samples and establish color recognition models based on standard color databases to sort fabrics [51]. Though color sorting presents many challenges, including the wide variety of shades and level of brightness or shadow on samples, a total classification accuracy of >96% was achieved for the 466 test samples. This opens the possibility for advanced sorting systems that combine NIRS with color recognition techniques.

NIRS methods developed in research labs are now being tested in real applications. For example, Circle Economy and other partner companies carried out the Fibersort project using NIRS [52]. The technique divided textiles into 90 different categories based on their fiber composition, color composition and fabric structure. The reported production rate reached 1260 kg/h, or one sample per second, which is six times faster than a trained manual sorter [52]. However, this is still slower compared to postconsumer plastics sorting systems, such as the “FlakeSort” system developed by NRT (National Recovery Technologies Inc., Nashville, TN, 37228 USA), which processes about 2268 kg/h of polyethylene terephthalate (PET) flakes [53]. Post-consumer textile sorting also faces unique technical difficulties, such as removal of integrated attachments (e.g., buttons, zippers, labels, threads) [52].

A life cycle assessment (LCA) study by Nørup et al., provides detailed insights on the operation of a European textile waste sorting facility, and gives an appreciation for the complexity of sorting post-consumer textile waste [54]. This particular facility processed around 36,000 metric tons of waste per year, of which a remarkable 75% was identified as reusable and the average price paid (in 2017) ranged from 0.7 to 1.42 €/kg, including transportation from collection, depending on whether the waste was presorted or not. Waste that is sorted into single component streams can give good LCA outcomes. For example, in their detailed LCA study of 100% cotton shirt recycling, Kazan et al., found that using recovered cotton fibers as the raw material significantly reduced many environmental impacts by eliminating the early stages in cotton processing that are energy, water and chemical intense [55].

While companies, brands and suppliers across the textile industry need to provide technologies, logistics and leadership for recycling, the sorting stage of recycling should be supported by consumers. Increasing consumer involvement in collecting and sorting textiles for recycling can be promoted by implementing policies to: (1) raise consumer awareness, (2) provide convenient recycling locations, and (3) offer recycling rewards [56].
Since PET, cotton and PET/cotton blends are so prevalent in consumer textile products, focusing sorting and recycling efforts on these materials can help build the awareness, infrastructure and involvement needed to better utilize these major recycling streams as well as build systems capable of handling more complex wastes. The recycling options for PET, cotton and PET/cotton blends are further discussed below.

4. Cotton

Successful fiber-to-fiber cotton recycling depends on preserving a sufficiently high degree of polymerization (DP) of its naturally-produced cellulose. DP is the average number of repeating monomer residues in the polymer chains. Cotton and other plant-based fibers are agricultural products. There are no direct industrial synthetic chemical routes to polymerizing cellulose from its degradation product, glucose. Therefore, monitoring cellulose DP during recycling is important, and methods for measuring cellulose DP are available in the literature [57]. Mechanical cotton recycling involves cutting, shredding and combing waste cotton to recover intact staple length fibers and re-spinning them. During chemical cotton recycling, waste cotton is dissolved in solvents and spun into new fibers. The quality of fibers spun from mechanically recycled cotton is not as good as virgin fiber because harsh forces needed to deconstruct tightly twisted cotton yarn can cause fiber damage and cause them to break [58]. This results in shorter recycled fiber lengths, and the strength and hand (touch sensation) properties of final products decrease [59]. Lower value mechanically recycled cotton fibers of sufficiently long length (≥13 mm) can be blended (e.g., 50/50) with virgin cotton to improve the recycle yarn quality while reducing the virgin yarn raw material cost [61], though the proportion of blended virgin cotton may need to be high to achieve the quality required for garment applications [60].

Combined mechanical and chemical recycling of waste cotton can lead to better utilization of materials and more options for co-product application that take advantage of the changed fiber length and DP that occur during recycling (Figure 4). For example, short cotton fibers produced by mechanical recycling can be used as reinforcement in composites, a potential high-value application [61,62]. Additionally, low molecular weight co-products generated during chemical recycling could be used as fermentation feedstocks for energy applications, such as biofuels production (e.g., bioethanol, biogas), or to make carbon fibers, activated carbon or cellulose nanocrystals (CNCs) [60]. Cellulose nanomaterials in particular are gaining attention for their applicability in numerous advanced technology fields, such as the medical sector, environmental protection and energy harvesting/storage [63].

![Figure 4. Schematic diagram showing changes in cotton microscale properties after different stages of use and recycling. Values are compiled from [58,64–73].](image)
4.1. Cotton Cellulose Properties

The cellulose DP of virgin cotton fiber is about 9000–15,000 and the cotton fiber staple length is 25–33 mm [64,65]. After industrial laundering at 84 °C using an alkaline detergent (1% solution, pH ~12) followed by tumble drying, the molecular weight (and DP) of cellulose in cotton fabrics decreases significantly. In one study, after two to four laundering cycles, the average molecular mass decreased by more than half, along with a drop in cellulose DP from an initial 8167 to 3500 [66]. Although home laundering conditions are mild by comparison, as the number of wash cycles increases, cotton fiber ageing is inevitable. Cellulose from waste cotton that is processed by chemical recycling has a DP of around 1000, which is significantly higher than obtained in the viscose process using wood pulp (DP 300~450) [67,74]. Therefore, cotton in textile waste is a good raw material for regenerated cellulose fiber production. Alternatively, cotton waste can be converted to cellulose nanocrystals (CNCs). After alkali and bleaching pretreatment of cotton fibers, CNCs can be extracted from the resultant cellulose by acid hydrolysis [75]. Acid hydrolysis causes a significant decrease in cellulose cellulose DP toward its level-off degree of polymerization (LODP) of 250 [72]. LODP means that the DP of cellulose decreases to a relatively constant value that is governed by the maximum length of fragments that will disperse in a particular hydrolyzing medium [57]. CNCs have desirable properties, such as high modulus, excellent mechanical strength and high aspect ratio (longer length than width), which make them suitable for composites applications [73].

Table 2 summarizes the size attributes of cotton in its different recycled forms. The cellulose DP progressively decreases as cotton fibers undergo more severe treatment conditions. According to the DP of CNCs and length of a glucose unit (~5 Å) [76], the theoretical length of polymers is about 125 nm, while the length of CNCs shows a range of 28–470 nm. As discussed in a review by George and Sabapathi, CNCs are obtained from highly crystalline regions of cellulose microfibrils which show no chain folding structure [31]. Therefore, according to the length range of CNCs, there may be some folding in amorphous regions and aggregation of crystals within the structure. At the low end of CNCs length range, folding of amorphous regions may dominate the structure, or the polymer chains may be shorter. The crystallinity of CNCs isolated from waste cotton showed a value of 55.76 ± 7.82% [75].

Table 2. Summarized size attributes of cotton used in different recycling applications.

| Cotton Categories                  | DP          | Mw * (g/mol) | Length (Fiber/Crystal/Polymer) | Ref. |
|-----------------------------------|-------------|--------------|--------------------------------|------|
| Virgin cotton                     | 9000–15,000 | 1.46 × 10^6  | 25–3 mm (fiber)                | [64,65] |
| Used cotton (after laundering)    | <3500       | <5.67 × 10^5 | No data, 1.75 µm (polymer) in theory | [66] |
| Cotton waste for composites       | No data     | No data      | <10 mm (fiber)                 | [62] |
| Regenerated fibers from waste cotton | ~1000      | 1.62 × 10^5  | No data (controlled by human)  | [67] |
| CNCs                              | ~250        | 4.05 × 10^4  | 28–470 nm (crystals)           | [75] |

* Mw (molecular weight) of anhydroglucose residue in cellulose is 162.14 g/mol, length of glucose repeat unit is roughly 0.5 nm [76].

4.2. Cotton Mechanical Recycling

Both post-industry (or pre-consumer) and post-consumer cotton waste can be mechanically recycled [77,78]. Recycling pre-consumer waste is usually preferred because the waste is cleaner and the quality is under control because the detailed sequence of manufacturing treatment processes for particular batches of materials is known. The shredding, blending and carding steps that occur during waste cotton mechanical recycling produce long fiber and short fiber fractions. Fibers that are longer than one half inch (≥13 mm) can be recycled for spinning yarn [58], however, such short-recycled cotton fibers usually must be combined with longer virgin fibers to achieve the needed yarn quality [60]. The
short fiber fraction (<10 mm) can be used as reinforcement in composites. In fact, for these applications, to achieve good dispersion in the matrix (e.g., polypropylene), cotton fibers preferably have around 1 mm length [62]. The addition of coupling agents, such as maleic-grafted polypropylene, improves chemical adhesion between cotton and the matrix to increase compatibility between hydrophilic and hydrophobic phases and produce high quality composites [61]. Dyes on cotton fabric have a significant effect on composite properties, which can be beneficial in some cases, but not always. The presence of organic dyes can decrease the hydrophilicity of cotton fibers, which then increases the affinity between cotton and hydrophobic matrices, such as polypropylene. However, dyes may also interfere with the interaction between cotton and coupling agents by limiting access to reactive functional groups on cotton fibers and hindering chemical linkage formation between cotton and coupling agents, thus limiting the strength of the resulting composites. More research is needed because confounding experimental variables include not only the dyes but also the prior history of chemical (e.g., alkaline) treatments to the cotton matrix [62], which may affect interactions between cotton and the matrix. In addition to composites, waste cotton fibers can also be incorporated in nonwovens [79], acoustic insulation [80] and building materials [81]. For example, Husain et al., used a needle punching method to convert cotton fabric waste into nonwovens [79]. Hawley’s description of textile recycling systems offers additional descriptions of processes and applications for mechanically recycled textile waste, including cotton waste, such as paper pulp and wiping cloths [82].

4.3. Cotton Chemical Recycling

Chemical recycling enables closed-loop recycling of the cellulose in cotton, by converting natural cotton fibers into regenerated cellulose fibers. Researchers from Deakin University developed closed-loop recycling of cotton from denim into solvent-spun regenerated cellulose fibers using a binary solvent of ionic liquid and dimethyl sulfoxide, with a weight ratio of 20:80, for wet spinning the textile waste. The processing steps included powdering the denim, decolorization in NaOH solution, dissolution in binary solvent and wet spinning into a water coagulation bath. The regenerated cellulose fiber had properties comparable to that of viscose fiber, though due to its relatively high molecular weight, it had lower stretch and higher wet tenacity. The solvents were recovered and recycled in a closed-loop system [67]. According to the findings, a long NaOH treatment time was needed to completely remove color from cotton. Depending on the color or dye type, the treatment time ranged from 5 to 30 h, which is not amenable to fast continuous production. The researchers proposed that retaining color during reprocessing could be beneficial for saving energy and resources in subsequent dyeing process, and this would also omit the decolorization step, however, a universally applicable approach for this was not presented. For cases where it is feasible, yarns or fibers already colored with dyes could be obtained after the treatment.

Similar to the dissolving wood pulp used to make lyocell, cotton waste can be dissolved to an appropriate concentration in the cellulose solvent N-methylmorpholine N-oxide (NMMO), followed by extrusion to form regenerated fibers and removal of solvent to fix the morphology. Haule et al., found that due to the higher molecular weight of cotton fiber cellulose (DP ~15,000) compared to cellulose in wood pulp (DP ~820) [67], regenerated cotton cellulose fibers have better mechanical properties than conventional lyocell fibers, such as higher tensile strength and modulus [83]. Therefore, by blending wood pulp with cotton waste at suitable ratios, the properties of final regenerated fibers were improved compared to that of wood-based lyocell. In this case, cotton waste contributed added value.

The process of dissolving cellulose in solvents causes polymer chain cleavage and lowering of DP compared to the starting materials. Silva and Byrne carried out experiments on cotton lint waste with different levels of alkali-treatment [84]. They found an interesting relationship between tensile strength and DP of the resulting regenerated fibers. As long as the DP remained within the range 2680 to 1150, the tensile strength of the
corresponding regenerated fibers did not change significantly from the maximum value at \( \sim 180 \) MPa. However, below DP 1150 the corresponding regenerated fiber strength dropped precipitously. This means that above a certain minimum DP (1150 in this case), having longer polymer chains in the regenerated fibers did not further enhance the tensile strength, whereas decreases in DP caused significant tensile strength reduction, dropping to around 60 MPa at a DP of 495. Since higher DP cellulose polymers need a longer time to dissolve in solvents, choosing raw materials with a DP at or just above the 1150 DP minimum value can help save production time when high tensile strength is required. Such raw materials could be obtained from waste cotton fibers that are too short for yarn spinning, but have a starting DP above 1150.

4.4. Cotton Energy Recovery

Cotton wastes that do not find use in other applications can be used for energy recovery. “Recovery” is a waste management term used to describe the recovery of embedded chemical energy from waste materials, when otherwise non-recyclable waste is incinerated or converted to energy by other means [19]. Since hydrolysis of cellulose polymer ultimately produces glucose, when fiber quality or cellulose DP is too low for recycling in other ways, cotton waste can be converted to glucose as a fermentation feedstock to produce bioenergy or other bio-based products. Svetlana et al., successfully produced ethanol from mercerized cotton fabrics by enzymatic hydrolysis of cotton to glucose and then fermentation of glucose to ethanol, however, the process is time-consuming [68]. When hydrolysis and fermentation are carried out at the same time, the process is called SSF, or simultaneous saccharification and fermentation. Alkaline pretreatment of cotton fabrics increased ethanol yield by changing the crystalline structure of cotton from cellulose I to cellulose II, which is more accessible to hydrolysis [79]. Via anaerobic digestion, biogas, a mixture of methane and carbon dioxide, can be obtained from cotton waste. Ismail and Talib found that inoculum addition, alkaline pretreatment and thermophilic conditions all have significant effects on biogas production efficiency [70]. Their study used waste cotton from the medical cotton industry, hence the impact of dyes and other textile auxiliary chemicals was not considered. While this review focuses on textile wastes, the upstream agricultural production of cotton also produces wastes, such as cotton stalks, cotton seed hulls and cotton oil cake, which have good potential to generate biogas [71]. Raj et al., found that biogas methane produced from cotton textile mill wastes can be used as a dual fuel for diesel engines, saving about 60% diesel [85].

4.5. Removal of Dyes from Cotton

Although retaining dyes on recycled fabrics can save a large quantity of energy, chemicals and resources, and some special applications may be possible (e.g., using powdered dyed cotton as printing pigments [86]), in general, dyes remaining on fabrics will be a problem for subsequent applications. The removal of dyes from cotton can be classified as a reactive process or a non-reactive one. In a reactive process, the dye structure is destroyed and cannot be recovered, whereas a non-reactive process extracts the dyes intact from fabrics and the dyes retain their original form for reuse. For dyes on cotton, reactive processes are more common because the most widely used dyes on cotton, reactive dyes, must be chemically altered to remove their color or separate them from cotton [87]. Other commonly used cotton dyes include direct dyes, vat dyes (indigo dyes) and azoic dyes [88]. These dyes are physically attached to cotton, and, in principle, can be removed by non-reactive means. Figure 5 summarizes the possible approaches for removing dyes from cotton which are discussed in detail below.
Figure 5. Summary of methods for chemical dye removal from cotton (compiled from refs. [67, 89–95]).

Reactive dyes feature covalent bonds between dyes and cotton. Table 3 summarizes reactive approaches for removing reactive dyes from cotton. The dyes can be removed by hydrolysis treatment using NaOH solutions, but it takes a long time (hours) to achieve adequate decolorization [67]. Another way to remove reactive dyes is photofading. Batchelor et al., studied the photofading mechanism of commercial reactive dyes and found that after 15 h exposure under visible and UV light, the decolorization of reactive dyes on cotton ranged from 5.8 to 27% [89]. Higher dye removal was achieved with moisture on fabrics, increasing the average decolorization by 36% compared to dry conditions. However, with 15 h treatment, the maximum dye loss only reached 50% of the original dye amount. Extreme pH conditions may facilitate the fading process, but this will damage the fabric. The presence of hydrogen peroxide accelerates reactive dye degradation under UV light. Georgiou et al., found that with 1 g/L H2O2, complete decolorization of azo reactive dyes in solution was achieved in 20–30 min under UV radiation [90]. During the treatment, the dye solutions became acidic (pH 3–3.5) which may harm the cotton structure and damage operating equipment. UV light is critical in the process. Peroxide alone did not cause decolorization to occur. Oxygen promotes fading under visible light, though if fabrics are stacked together, there will be different degrees of color fading on the textiles. The proposed mechanism is that dyes are excited by visible light and electrons of excited dye molecules transfer to oxygen, forming superoxide that leads to decolorization [96].

Other dyes, such as direct dyes, can also be removed by photofading. Forster et al., found that microspectrophotometry (MSP) with visible and ultraviolet radiation sources, that are used to analyze color in textile fibers, have the potential to cause photofading of direct dyes on cotton [97]. Their MSP experiments on three types of dyed cotton (blue, red and yellow) showed photofading phenomena where the red sample experienced the most obvious effect (around 44% color loss) at the main absorbance peak after 30 min of exposure to the MSP light source and showed that photofading was more pronounced for lightly shaded fibers.

Table 3. Possible ways to remove reactive dyes from cotton [67, 89, 90].

| Dye removal Reactions | Limitations |
|-----------------------|-------------|
| D-O-Cell NaOH D-ONa + CellONa | long decolorization time |
| D-O-Cell Vis/UV (moisture) degradation of dyes | long decolorization time, low dye removal rate, different fading degree |
| D-O-Cell UV (H2O2) decolorization | acidic reaction solution |

Figure 5 (repeated text)
Instead of decolorization, waste cotton fibers or graftedcellulosic fabric waste can be used as an absorbent for direct dyes in water pollutants due to the affinity between the dyes and the absorbent. Research shows that 1 g of waste cotton fibers can absorb dozens of milligrams of direct dyes as a potential application of waste cotton in effluent treatment [98,99]. However, long times and agitation may be needed to achieve high extents of direct dye removal with cotton alone.

Certain chemical treatments on direct dyes, reactive dyes and vat dyes can achieve good color removal results [91]. Chemical treatments such as hot alkaline extraction (HCE) or its combination with other chemicals, such as sulphuric acid, ozone and hydrogen peroxide, deliver high levels of decolorization for direct dyes—over 90 ISO percent brightness—followed by reactive dyes and vat dyes. In general, vat dyes do not achieve as good dye removal as other dye types, mainly due to the high stability of vat dyes towards alkaline conditions [100]. In addition, the special dyeing process of vat dyes causes the dye molecules to become trapped inside the fiber structure which may further hinder the HCE dye removal process [101]. A notable exception is indigo, used to dye denim jeans, which can be chemically reduced under relatively mild conditions to its soluble form that can be washed out of fabric and recovered, or bleached to destroy the color [92–94]. HCE treatment of waste cotton fibers has the advantage of eliminating polyester impurities (e.g., from sewing thread) by hydrolysis, making the cotton more suitable for recycling. After HCE treatment, cellulose molecular weight decreased to around $1.0 \times 10^5$ g/mol and DP was about 616. This value is lower than DP 1150 level that gives maximum strength to regenerated cellulosic fibers [84], meaning that after chemical decolorization treatment, waste cotton fibers experience severe damage. This will weaken the performance and limit the applications.

Historically, vat dyes were removed using a reducing bath including caustic soda and sodium dithionite or sodium hydrosulphite at high temperature. Stripping assistants such as quaternary ammonium salts were combined with the removed dyes and avoid them reattaching on the fabrics [91]. In recent research, lasers are found to be capable of decolorizing vat dyes on cotton. Compared with ordinary light, laser light possesses unique characteristics: monochromaticity, directionality and coherence [102]. Štěpánková et al., utilized infrared laser light to treat cotton fabrics and studied its effect on both dyes and cotton fibers [95]. For non-dyed cotton fabrics, low laser fluency led to oxidation of samples while carbonization occurred at high laser fluency (15.6 mJ/cm²), and browning of cotton fabrics occurred. Irradiation of dyed samples resulted in lower color intensity but did not eliminate all color from the samples. Material responses during laser-material interaction are categorized into three classes: thermally activated processes, surface melting, and ablation [103]. Dye removal by laser irradiation belongs to the ablation category. In terms of the applied laser fluency (mJ/cm²), it is regarded as low fluence, where material evaporation and sublimation are the mechanism for ablation. The tensile strength of dyed and undyed yarns was measured before and after laser irradiation. As shown in Table 4, at medium laser fluency (7.8 mJ/cm²) the strength in both weft and warp directions of undyed irradiated cotton yarns decreased by more than 30% of the original tensile strength. However, the drop in strength for both yarns became smaller when there were vat dyes attached on the fibers, which indicates the dyes protect cotton yarns from infrared laser light. Notably, for both dyed and undyed irradiated yarns, warp yarns showed higher decreases in strength than weft yarns, which may be attributed to the lower density of weft yarns. Since loosely arranged yarns have larger surface area than tightly arranged ones, weft yarns tend to absorb more dye molecules than warp yarns and therefore may possess a stronger dye protective function. Kan used CO₂ lasers to remove indigo dyes from cotton denim fabric [104]. The light produced by CO₂ lasers belongs to the far infrared range of the light spectrum [105]. By controlling the laser process parameters, such as resolution and pixel time, an optimum laser power of 13 W/cm² was reached and around 87% color fading was obtained [104].
Table 4. Decrease in tensile strength in different directions of irradiated cotton yarns at 7.8 mJ/cm² laser fluency (modified from ref. [95]).

| Yarn Type   | Undyed | Dyed |
|-------------|--------|------|
| Weft yarn   | 35%    | 15%  |
| Warp yarn   | 56%    | 31%  |

Enzymatic or microbial treatment is another approach for removing vat or indigo dyes. Oxidative enzymes and microorganisms, such as laccases and ligninolytic fungi show relatively high dye removal ability [106,107]. However, these studies focused on biological treatment of effluents containing indigo or azo dyes [108] rather than dyes on waste fabrics. Azo dyes are a category of organic compounds with one or more \(-N=N-\) groups, which can include many of the dyes mentioned above such as direct dyes and reactive dyes [109]. Through an oxidative mechanism, laccase and lignin peroxidase can decolorize and remove azo dyes from textile wastewater. Variables in the treatment environment such as glucose supplementation and organic versus inorganic nitrogen source will affect enzyme production (lignin peroxidase versus laccase) and the decolorization process [110].

5. Polyester

Similar to cotton, PET has both mechanical and chemical recycling options, but the technology details differ because (1) PET is a thermoplastic that can be melt processed at temperatures above around 264 °C [111] (cotton degrades at around 375 °C [112], rather than melting) and (2) PET is a synthetic polymer that can be reproduced in an industrial reactor from its own monomer degradation products. Usually, PET mechanical recycling involves both physical deconstruction (cutting or milling) and phase change (melting) to purify the polymer and make polymer pellets. PET chemical recycling either involves dissolving the polymer in an organic solvent to separate it from contaminants, or involves hydrolyzing the ester chemical linkages along the polymer chain to produce its monomers (or useful oligomer mixtures) that can be re-polymerized after purification [113].

5.1. Polyester Mechanical Recycling

PET mechanical recycling is commercially practiced for recycling PET bottles and other pure PET packaging materials. These are made of amorphous high molecular weight polymer and bear the Resin Identification Code number 1 with a “PETE” label [8]. The polymer morphology and physical thickness and size of different PET forms (bottles, films, fibers) varies considerably. When referring to textile fibers, the term “polyester” usually means “PET polyester” fiber. PET textile fibers have high crystallinity (40–55%) [20], compared to PET bottles or films (12–23%) [114]. Lower crystallinity materials are easier to melt and are more accessible for chemical reactions, meaning that PET fibers are on the most challenging end of the recycling spectrum. Due to their high fiber strength, PET fabrics can be mechanically shredded to produce loose fiber for fillers or nonwovens, though this generally reduces the value compared to virgin fiber [15].

5.2. Polyester Chemical Recycling and Biodegradation

The chemical recycling of PET is an emerging technique for achieving closed-loop recycling or upgrading of polyester waste. Compared to mechanical recycling, chemical recycling reduces concerns about thermal degradation and the impact of this on properties of the final products, since the treatment ultimately results in the recovery of small molecule monomers, not large complex polymers. During the chemical recycling process, PET polymer is depolymerized to its original starting material monomers (EG and TPA) or oligomers (e.g., BHET, bis-hydroxyethyl terephthalate). As a reverse reaction of polymer formation, the raw materials obtained after chemical recycling can be further utilized to synthesize new products, whether they become PET again or are converted into other useful chemicals, such as azo disperse dyestuffs [115]. Via alcoholysis (sometimes called
glycolysis, not to be confused with “glycolysis” which is the metabolic conversion of glucose to energy), the depolymerization of PET by excess glycol (usually ethylene glycol) predominantly produces BHET [116]. Shukla et al., found a potential way to recycle waste PET fibers into other textile auxiliaries by converting BHET into quaternary ammonium compounds which can be used as softeners in the cotton finishing process [117].

In addition to chemical agents, microbes and enzymes also have the potential to degrade PET in an eco-friendlier way [6]. Researchers at TBI (Toulouse Biotechnology Institute) and Carbios developed a new enzyme-catalyzed degradation technology that achieved 90% depolymerization conversion of colored PET flake (~16% crystallinity) in less than 10 h [118]. This technology could potentially be applied to the recycling of PET fibers; however, it requires essential pretreatment steps (extrusion and micronization) and hydrolysis conditions of ~72 °C (near the glass transition temperature, Tg, of PET) to decrease PET crystallinity and increase the accessible area for enzyme reaction.

5.3. Biobased Polyester Monomers

An important aspect of the overall circularity concept involves using sustainable resources. This argues for changing PET production methods from using petroleum-based to bio-based raw materials [119]. Bio-based EG can be obtained from renewable resources such as ethanol, glycerol, sorbitol, sugars or (hemi)cellulose [84] (Figure 6). Using ethanol as the feedstock requires three processes to form EG: dehydration of ethanol to ethylene, oxidation of ethylene to ethylene oxide, and finally hydration of ethylene oxide to EG [120,121]. Via hydrogenolysis, intermediate raw materials such as glycerol and sorbitol can be metal catalyzed to yield ethanol, a precursor of EG [122–127]. Using sugarcane derived ethanol from India or Brazil [119,128] companies such as JBF Industries Ltd. and Greencol Taiwan Corporation are able to annually produce 500 kT and 120 kT of bio-based EG, respectively. Some 26 billion liters of ethanol made in Brazil in 2018 were almost all from domestic sugarcane [129]. In addition, the Changchun Dacheng Industrial Group Company Ltd. utilized hydrogenolysis of glucose-sorbitol to produce 1,2-propylene glycol (1,2-PG) and EG at a commercial scale of 200 kT per year [119]. While substantial, these combined amounts still only account for 2.4% of the EG needed globally to produce PET (the global EG production capacity in 2016 was around 34 million tons) [130].

![Figure 6. Simplified schematic showing several important chemical intermediates in the conversion of three different biomass raw materials to EG (compiled from refs. [119–121,123,131–133]).](image-url)
The other component of PET, TPA, has been a more elusive target for biomass-based production routes. Over the years, a number of companies have tried (and failed) to produce bio-based p-xylene, an important intermediate that is converted to TPA by catalytic oxidation. Currently, the company Origin Materials has garnered much attention and support from eventual corporate customers in their efforts to convert lignocellulosic biomass to 3-(chloromethyl)furfural (CMF), a precursor to p-xylene [134]. Figure 7a shows the chemical pathway from CMF to p-xylene. Bio-based CMF is a versatile precursor that can also be used for synthesizing products such as cyclohexanone, caprolactam, and hexamethylenediamine (HMDA). In addition to Diels-Alder (DA) reaction between 2,5-dimethylfuran (DMF) and ethylene [135], there are other chemical routes to obtain p-xylene or TPA from biomass. The raw materials can be bio-ethylene, isobutanol, isoprene and acrylic, limonene, and furfural [119]. For example, in a sequence of steps starting with a DA reaction, followed by dehydro-aromatization and oxidation, the starting materials bio-based isoprene (sugar fermentation product) and acrolein (glycerol dehydration product) [136] or acrylic acid can produce the target product, TPA monomer [137] (Figure 7b).

**Figure 7.** Chemical pathways to produce p-xylene: (a) from CMF to p-xylene and (b) from isoprene to p-xylene or TPA (complied from refs. [119,134,137]).

5.4. Removal of Dyes from Polyester

Disperse dyes are commonly used in polyester dyeing. The two most common structures in disperse dyes are monoazo and anthraquinone (Figure 8) [138].
Disperse dyes are commonly used to obtain colored yarns by mixing and melting color pigment with recycled polyester [146]. The recycled polyester is made from post-consumer water bottles or waste clothing. The solvent dimethyl formamide (DMF) has an excellent ability to extract disperse dyes from polyester, and is superior to other solvents such as chlorobenzene, methylene chloride, chloroform and acetic acid [142]. Utilizing this attribute, Wu et al., found that under appropriate experimental condition, the disperse dye removal efficiency from polyester fabrics reached 87% in DMF and increased to 99% when the fabrics were treated with saturated DMF steam reflux [143]. DMF was observed to swell the PET fibers, but not dissolve them, since there was limited weight loss (3~4%) during the treatment. Another type of dyeing polyester called “dope-colored” polyester was also investigated. Dope colored PET is obtained by two processing methods: (1) adding colorants during the pre-condensation stage of PET polymerization resulting in colored polyester chips after final polycondensation; or (2) dispersing colorants evenly in PET resins to form a pigment masterbatch which is added during melt spinning to produce colored PET fibers [144]. Dope-colored polyester has higher color fastness compared to fabrics dyed by conventional processes and eliminates the need for large amounts of water consumption during the dyeing process [145]. Experimental results showed the decolorization efficiency of dope-colored polyester only reached ~30% because the colorants were very well integrated into the PET structure which hindered their extraction by solvents [143].

Following a dope dyeing approach, the company We aRe SpinDye produces homogeneously colored yarns by mixing and melting color pigment with recycled polyester [146]. The recycled polyester is made from post-consumer water bottles or waste clothing. Color in the PET fabrics is intentionally retained so the waste polyester can be melted to generate color pigments. Unfortunately, PET fabrics contain many contaminants, such as dyes and other finishing auxiliaries, which may affect the recycling process, reduce recycled product quality, and explains the motivation for removing dyes and auxiliary agents.

Oxidative chemical agents for decolorizing PET fabrics include hydrogen peroxide or ozone. Chemical reducing agents include sodium dithionite and sodium hydrosulfite or sodium formaldelyde sulfoxylate (SFS) [147]. SFS is commonly used as a discharge printing agent that destroys the background color, leaving behind a delicate white pattern [148]. Similarly, the use of thiourea dioxide for discharge printing suggested it could increase dye removal rates [149]. Small molecule non-polar discharge accelerants, such as compounds containing benzene within the structure [150], that function by relaxing compact polyester molecular structures and increasing intermolecular voids could improve the decolorization reaction between thiourea dioxide reducing agents and disperse dyes on polyester.
While there is interest in the potential for removing disperse dyes from polyester by enzymes or microbes, such as laccase, peroxidase, or the fungus *Cunninghamella poly-\textit{morpha}* [151–153], chemical mediators may be needed to reach dyes deeply embedded in the fiber which could potentially damage the polymer. So far, studies have avoided this obstacle by focusing on disperse dye removal from aqueous solutions. However, the decolorization process takes a long time (hours or days), and efficiency varies depending on the type of disperse dye.

6. Cotton/Polyester Blends

The polyester/cotton (or “poly/cotton”) blended fabric market was valued at over USD 7 billion in 2016 [154]. This popular fiber blend is used extensively in daily wear fabrics. Polyester contributes strength, wrinkle resistance, quick drying properties and shape retention, while cotton contributes comfort, moisture management, high porosity and heat conduction. Recycling processes for these blends can be roughly divided into two categories, depending on whether the two blended components are separated or not. If the two components are not separated, recycling involves using the cotton/polyester blended waste as a whole. Aside from cutting fabric pieces into wipes or shredding to produce loose fibers, these whole blend wastes could be used in composites. For example, Zou et al., made composites by compression molding 65/35 PET/cotton fabrics [155]. Composite blends had higher modulus but decreased tensile strength compared to the PET matrix. Adding plasticizers lowered the PET melting temperature, which lowered the subsequent processing temperature, and increased the impact resistance, but significantly decreased the flexural and tensile properties of the composites. Alternatively, if the two fiber components are to be separated from each other, mechanical means alone are not sufficient to disentangle and sort intimate blends. One might imagine “melting” PET out of the blend, but high polymer viscosity makes this impractical, which leaves dissolution or degradation of one (or both) components as the more feasible separation mechanisms. The type of mechanism governs how separated components can be used afterwards. Table 5 summarizes examples of these approaches.

Table 5. Different separation and recycling approaches for polyester/cotton blends.

| Mechanism   | Treatment/Conditions | Effect on Cotton | Effect on PET | Ref.    |
|-------------|----------------------|------------------|---------------|--------|
| Dissolving  | NMNO solvent         | dissolved and regenerated | isolated | [156,157] |
|             | ionic liquid         | dissolved and regenerated | isolated | [158,159] |
|             | DMSO solvent         | dissolved and regenerated | isolated | [160]  |
| Degradation | enzymatic hydrolysis | yield glucose syrup | recovered | [161]  |
|             | enzymatic hydrolysis and yeast fermentation | yield ethanol | recovered | [156]  |
|             | anaerobic digestion | yield methane | recovered | [156]  |
|             | acid hydrolysis (high-temperature and high-pressure hydrolysis) | microcrystalline cellulose | recovered or yield TPA | [162,163] |
|             | alcoholysis          | recovered | yield BHET | [164,165] |
|             | alkaline treatment   | recovered | yield TPA and EG | [166]  |

6.1. Cellulose Separation

Selectively separating cellulose from poly/cotton blends will “clean” the polyester and either recover cellulose with a sufficiently high DP for recycling to fibers or will convert the cellulose to low molecular weight materials for other uses. Cellulose in cotton is very difficult to dissolve due to its high molecular weight, intra- and inter-molecular hydrogen bonds, and high crystallinity [57]. However, several chemical derivative and
direct dissolution recipes have been developed for the purpose of making manufactured cellulose fibers [167]. These techniques are now being investigated for textile waste separation. Cotton can be separated from blended fabrics by solvents such as NMMO, and, after enzymatic hydrolysis and yeast fermentation of the regenerated cellulose, ethanol can be produced. Jeihanipour et al., applied this recycling process to 50/50 poly/cotton blends and achieved 48 g ethanol / g regenerated cellulose, which was 85% of the theoretical yield [156]. The blend not treated with NMMO only produced 50% of the theoretical ethanol yield, indicating the importance of decrystallizing the cellulose structure for efficient hydrolysis to fermentable sugars. Likewise, NMMO-regenerated cellulose yielded more methane during anaerobic digestion compared to the untreated blend. Subramanian et al., carried out an LCA study on bio-recycling of 50/50 poly/cotton blends [161]. Via enzymatic hydrolysis, cotton was converted into glucose while the recovered polyester was re-spun into PET fibers. During the melt-spinning process, a relatively high amount (80:20) of PET bottle chips was needed to maintain a good quality for textile fiber production. In this LCA study, decreasing the fraction of added PET bottle chips did not ensure reduction in the environmental impacts because spinning fibers from recovered (textile fiber) polyester is more resource intensive than from PET bottle chips. This is because extra processing is needed to restore high molecular weight to PET from recycled polyester textile fiber.

In a fiber-to-fiber application, Haslinger et al., utilized the ionic liquid 1,5-diazabicyclo(4.3.0)non-5-enium acetate or (DBNH) (OAc) to dissolve cellulose from 50/50 poly/cotton blends, then produced fibers from the dissolved cellulose via dry-jet wet spinning. The regenerated cellulosic fibers had comparable properties (breaking tenacity 27–48 cN/tex, elongation 7–9%) to commercial lyocell fibers [158], and “cleaned” polyester was recovered as a co-product. Since milling may be used to reduce the particle size for better mixing of the raw waste material, polyester may not be recovered directly in a fiber form, but rather as a powder that would be melt-processed to form new fibers, or would be further hydrolyzed for chemical recycling. Due to its higher resistance to acid than cotton, polyester can also be separated from poly/cotton blends via acid treatment. Ling et al. [162] used phosphotungstic acid to recover polyester from (65/35) poly/cotton blend fabrics. Then, after further high-temperature and high-pressure hydrolysis, TPA was produced. Most acid used in the separation process was recovered (98%) and microcrystalline cellulose was obtained as a byproduct. Hou et al., also successfully separated polyester from a waste poly/cotton blend under hydrothermal conditions with hydrochloric acid as the catalyst [163].

6.2. Polyester Separation

Selectively separating polyester from poly/cotton blends will “clean” the cotton and either recover polyester with a sufficiently high molecular weight for recycling to fibers or will convert the polymer to monomers/oligomers for chemical recycling. A typical way to separate poly/cotton blends is using NaOH to degrade the PET component to its monomers, TPA and EG, and isolate cotton at the same time. Adding a phase transfer catalyst can shorten the PET hydrolysis time and reduce the damage to cotton [166]. Similarly, polyester within the blends can be converted into BHET via ethylene glycol alcoholysis, catalyzed by zinc acetate, and the cotton component would be recovered [164]. PET was resynthesized through polycondensation of BHET, and the results indicated that reconstructed PET met the requirements for fiber applications. To address the secondary contamination issue caused by the metal catalyst, Liu et al., first synthesized betaine-based deep eutectic solvents as a promising catalyst during the alcoholysis of PET fibers [165]. The solvents showed excellent catalytic effect with a BHET yield of 85% [165], compared to 75% recovery of BHET catalyzed by zinc acetate [168]. Notably, the reaction time of the former experiment (1 h) was also much shorter than the latter one (8 h).

Another way to isolate cotton from poly/cotton blends is to dissolve the PET. Using this strategy, Yousef et al., recovered cotton from 20/80 poly/cotton jeans using dimethyl sulfoxide (DMSO) to dissolve the polyester and other organic substances [160]. After the
leaching, dissolving and bleaching process, the total material recovery was 93 wt%, with 77 wt% as cotton and 16 wt% as polyester. High purity DMSO (99.5%) was regenerated by rotary evaporation at the same time as polyester was recovered.

6.3. Impacts of Recycling Both Components

In a case study in Sweden, researchers conducted an LCA of three recycling processes for 50/50 poly/cotton blends: (1) material reuse to a textile bag; (2) separation of cellulose from polyester by using NMMO to make regenerated cellulose and PET fibers; and, (3) recycling of polyester by manual separation and chemical recycling to resynthesize PET fibers [157]. They found that the material reuse process had the lowest global warming potential (GWP) and lowest primary energy usage by saving eight tons of carbon dioxide equivalent and 164 gigajoules (GJ) of primary energy per ton of textile waste. However, all the three recycling processes showed better LCA results than incineration of textiles with energy recovery. Therefore, it was worthwhile to recycle the poly/cotton blends and increase their additional value.

Researchers also investigated the possibility of recycling textile waste into other applications and its economic feasibility. Islam and Bhat reviewed the literature on conversion of recycled textiles to thermal and acoustic insulation materials [169]. Transformation methods included nonwovens, composite techniques and mixing with building materials. According to their assessment, some thermal insulation materials obtained from recycled textiles had even better performance than current commercial products. Athanasopoulos and Zabaniotou analyzed the thermochemical conversion of waste textiles to fuels and biocarbon [170] and indicated that torrefaction, as a low environment impact technique, can be applied for cotton and polyester mixed wastes to obtain energy-dense fuels such as biochar. In their recent review promoting circular economy, Keßler et al. [171] emphasized the importance of reducing the amount, physical size and complexity of textiles as well as applying a systems perspective that takes into account the full life cycle of textile recycling.

As mentioned above, NaOH can be used to separate poly/cotton fabrics by degrading the PET component to its monomers and isolating cotton. An LCA study compared this recycling process to single-use scenarios of mixed waste containing cotton and polyester [172]. The single-use scenarios included incineration of blends for energy recovery along with the production of virgin polyester and cotton or viscose. Results showed that the alkaline recycling process had comparable impacts to the single-use scenarios, with half of the LCA indicators favoring the single-use scenarios over the recycling scenarios. Nevertheless, alkaline recycling has potential for application in textile recycling and provides a feasible way for the implementation of the cotton/polyester recycling process.

7. Conclusions and Outlook

This review has presented the fundamental concepts and recent developments in textile recycling and discussed some possible approaches to improve the circularity of cotton, polyester and their blends as the most important textile materials for retail consumers. Before recycling, spectroscopic sorting (e.g., NIRS combined with color sorting) is on a development path toward becoming the most accurate and efficient way to automatically classify textile wastes for further processing. Properly sorted materials then proceed to mechanical and/or chemical recycling, with decolorization, when needed. Understanding the changes in DP of cotton from the virgin fiber to end products, such as CNCs, facilitates choosing the appropriate recycling process and helps prolong the use/recycling life of cotton for maximum sustainability benefits. Synthesizing regenerated fibers from waste is a prevailing choice since there are existing commercial facilities to produce regenerated cellulose fibers from wood pulp. Additionally, energy recovery from waste cotton is another valuable and feasible process. For polyester, depolymerization can achieve upcycling by reuse of purified monomers. Furthermore, synthesizing polyester from renewable resources will help close the carbon-cycle loop and provide alternatives to diminishing fossil resources. Cotton/polyester blends could either be recycled as a whole, for filling
and padding materials, or separated to recover specific components via dissolving or degrading. LCA studies indicate that recycling poly/cotton fabrics lowers the environmental impacts compared to incineration and has the potential to make the textile industry more sustainable. Removing dyes and other chemicals from textiles is a persisting challenge. Decolorization strategies explored at the lab scale include (1) chemically destroying the dye structure, by using oxidizing agents, reducing agents, alkaline treatment, photofading, or laser treatments, or (2) retaining dye structure via solvent extraction, which is usually applied to remove disperse dyes from polyester. By increasing awareness of the recycle value embedded in cotton and polyester, applying a “designed for deconstruction” philosophy during textile product development, and expanding the textile recycling infrastructure, more extensive textile recycling can already occur with proven technologies. Meanwhile, continued research on deconstructing complex textile waste streams, supported by further developments in sorting and rapid analysis, will expand the types of textile wastes that can be reprocessed toward closing the textile waste circularity loop.

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Abbreviations

| Abbreviation | Definition |
|--------------|------------|
| BHET         | Bis-hydroxyethyl terephthalate |
| CESA         | Cellulose synthase |
| CMF          | 5-(chloromethyl)furfural |
| CNCs         | Cellulose nanocrystals |
| CSC          | Cellulose synthase complex |
| CSS          | Cellulose synthase supercomplex |
| DA           | Diels-Alder |
| DMF          | 2,5-dimethylfuran; Dimethyl formamide |
| DMSO         | Dimethyl sulfoxide |
| DP           | Degree of polymerization |
| EG           | Ethylene glycol |
| EPA          | Environmental Protection Agency |
| FTIR         | Fourier-transform infrared |
| GPa          | Gigapascal (1 GPa = 1 × 10^9 Pa) |
| GWP          | Global warming potential |
| HCE          | Hot alkaline extraction |
| HMDA         | Hexamethylenediamine |
| HT           | High tenacity |
| IRS          | Infrared spectroscopy |
| LCA          | Life cycle assessment |
| LODP         | Level-off degree of polymerization |
| LT           | Low tenacity |
| MPa          | Megapascal (1 MPa = 1 × 10^6 Pa) |
| MSP          | Microspectrophotometry |
| MSW          | Municipal solid waste |
| Mw           | Molecular weight |
| NIRS         | Near-infrared spectroscopy |
Abbreviation | Definition
--- | ---
NMMO | N-methylmorpholine N-oxide
NRT | National Recovery Technologies
PET | Poly(ethylene terephthalate)
PLA | Polylactic acid
PP | Polypropylene
rPET | Recycled PET
SFS | Sodium formaldehyde sulfoxylate
SMART | Secondary Materials and Recycled Textiles
SSF | Simultaneous saccharification and fermentation
TBI | Toulouse Biotechnology Institute
TPA | Terephthalic acid
UDP | Uridine diphosphate
UV | Ultraviolet
1,2-PG | 1,2-propylene glycol

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