

Review

Opportunities and Limitations in Recycling Fossil Polymers from Textiles

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Abstract: The recovery and recycling of textile waste is becoming urgent since textiles are generating more and more waste. In one year, about 92 million tons of textile waste are produced and the fashion industry accounts for 58 million tons of plastic waste per year. Several different synthetic fibres are used in textiles, thanks to their excellent processability and mechanical properties, but on the other hand, the difficulties linked to their end of life and the release of microplastics from them during washing is currently a cause of great concern. In this context, policy actions have been aimed at promoting recycling of waste and replacing fossil-based fibres with biobased fibres. The current review, considering both scientific papers published on international journals and web sources, considers the sorting of textiles and the possible recycling of polyesters, polyamides and acrylics. Nevertheless, the contamination and presence of mixed fibres in fabrics is another issue to face for recycling. Methodologies to solve the issue linked to the presence of elastane, present in the stretch fabrics, as well as the possibility of recycling textiles in the non-woven and composite sector are investigated. Moreover, chemical recycling and enzymatic recycling of fossil polymers are also considered. Thanks to the comprehensive scheme of this review, it is possible to deduce that, while the use of biobased materials should rapidly increase in textile applications, the perspective of recycling materials obtained from waste textile into durable and/or high-performance products seems the most promising.

Keywords: textile; sorting; mechanical recycling; polyester; acrylics; fibres; polyamide; enzymes; chemical recycling; non-woven; composite

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1. Introduction: The Current Situation Regarding Textile Products

Currently, textile production is constantly expanding by catering to fashion, style, and marketing needs as well as to increasingly competitive technical challenges. The prospect of innovation and development contrasts, however, with the pollution of the environment caused by this industrial activity. Between 1975 and 2018, the production of chemical and textile fibres nearly quadrupled, from 23.94 million tons to 105.6 million tons: a more than fourfold increase in about 40 years [1]. Concerns about the environmental impact of fibre production and subsequent disposal operations have grown in prominence as demand for fabrics has increased. Several recent studies show that the textile industry endangers freshwater and atmospheric micro-systems due to the use of industrially harmful and toxic chemicals during the manufacturing process, as well as the release of these chemicals.

Looking at waste, fashion industry accounts for 58 million tons of plastic waste per year, (ranked third after packaging and construction), 20% of global wastewater, and 10%

of global/carbon emissions. It contributes to biodiversity loss, since 33% of the insecticide applied worldwide is used for tissue production. Unfortunately, only 1% of global clothes are recycled while 87% are incinerated or landfilled, due principally to the current exiguous development of recycling industrial chains [2–4]. It is necessary to transform the current linear economy, based on the taking, making and producing of waste, in a circular economy based on redesigning, reducing, reusing and recycling for zero waste [5–7]. Therefore, it is becoming urgent to organize a new productive model for making textile and fashion products based on a new eco-design production and at the same time to persuade consumers to make small and gradual behavioral changes [3,4,6].

The status of textile recycling and reuse in the world was surveyed by Lu and coworkers [8] in 2014. Their data showed that 10% of the fibre waste in China, 12–13% in Japan and 15% of the fibre waste in the US was reused or recycled. In Europe, Germany is one of the pioneer countries where as much as 66% of the projected textile fibre turnover is collected and recycled or reused according to a report from BVSE, the German federal organization for secondary raw materials and disposals [9]. More recently, a material flow analysis (MFA) was applied to quantify textile streams and recycling prospects in Europe [10]. The MFA estimated textile waste, energy and CO₂eq emission streams in Europe and determined that Europe has generated an amount of 2526–2791 kt of textile waste in 2018. In a high separate collection rates (HSCR) scenario it is possible to collect 1894–2093 kt and to reduce landfilling to 1%. Textile recycling consumes 4079815–4509260 GJ of energy in the baseline scenario. In the HSCR, CO₂eq emissions could be reduced from 2,419,261 t to 2,025,876 t (–16%). This work evidenced the essential role of textile recycling as a way to enlarge fibre and commodity lifespans and convert fast fashion into slow fashion. Similar considerations were expressed by Schumacher et al. [11], who additionally evidenced the need for harmonization of terminology, classifications, industry tools and standards to unify approaches and increase the interoperability of tools and resources. Moreover, a fundamental role can be played by broad, multidisciplinary and multi-stakeholder collaboration.

The manufacturing process of textiles consists of four primary stages: yarn production, fabric production, textile production and finishing treatment (Figure 1).

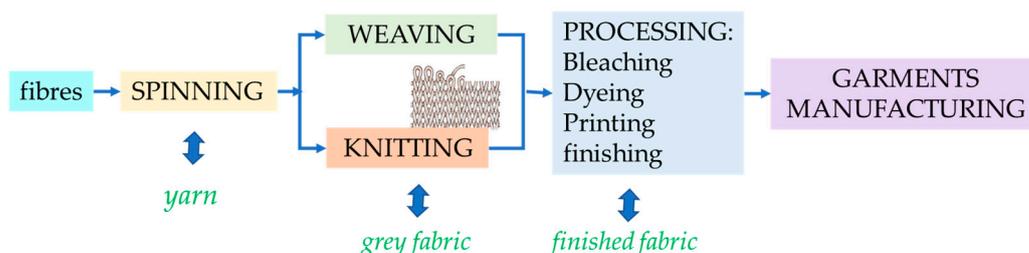


Figure 1. General scheme of textile production.

This type of industry deals with transforming raw materials (natural, artificial or synthetic fibres) into yarns and then fabrics, which are collected in reels and then commercialized. The raw material that reaches the textile industry is collected, instead, in large bales which are opened and then processed in the factory. In the textile factory, two fundamental operations are carried out: spinning and weaving. In the first phase, the fibres, first collected in bales, are washed and processed until yarns are obtained, possibly collected on a large reel. In the second phase, the textile fibre thread is then worked in other machines where it is woven with other threads to obtain the fabric [12]. In the case of natural fibres, which are short (with the exception of silk fibres that can reach 1 km of length) the fibre production is skipped. In the case of artificial and synthetic fibres, the extrusion of filaments can start from a polymer solution or polymer melt. At the end of this process, generally multifilament yarns are produced that can be used as such or

successively texturized. Cutting these yarns allows staple fibres to be produced, so that the long-extruded filaments are transformed into short fibres.

In general, spinning is the set of operations that serve to transform textile fibres into a yarn, that is, a very long and resistant thread that is collected on a reel. The other important phase in the textile industry is weaving which allows the thread to become an actual fabric, to a cloth wound on reels [13]. The selection of yarns and the design of the fabric (woven or knitted) is fundamental to obtain the desired level of lightness, comfort and softness for the final textile product.

1.1. Fibres Used in Textile Products

Textile fibres currently present in commercial products are categorized as either natural or man-made (Figure 2). On the molecular level, these are built up by polymer chains where the monomers and the polymer linkage are specific for each fibre type.

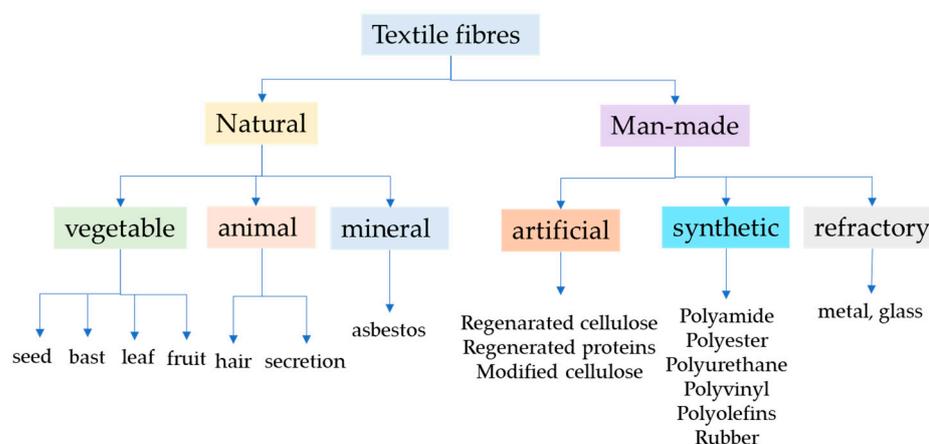


Figure 2. Scheme of fibres used in textiles.

The natural fibres fall into three different chemical classes:

- Cellulosics, which originate from various parts of plants
- Protein fibres, which are obtained from wool, hair and silk
- Mineral fibres, where asbestos is the only one naturally occurring

A natural fibre consists of thousands of polymer chains which aggregate to form complex patterns and build up very specific layered structures accounting for much of the properties of the fibre. Natural fibres range in length from a few centimeters for cotton, to around one decimeter for wool, and up to several hundred meters for silk. Shorter fibres, such as cotton and wool, are spun together to form a continuous yarn.

The man-made fibres can also be divided into three sub-groups:

- Regenerated fibres, consisting of natural polymers, mainly cellulose, that were chemically modified
- Synthetic fibres, which are produced from monomers by polymerization
- Inorganic fibres, such as glass or ceramic fibres

Man-made fibres are formed by extruding the melted or dissolved polymer through the small openings of a spinneret. With this process, both filament yarns, in which the yarn is comprised of one infinitely long fibre, or spun yarns, in which the fibres are cut to shorter pieces and spun together, can be produced [9].

Synthetic polymers mainly used in textile products are polyester, polyacrylic, polyamide, elastane and polypropylene [14,15]. Polyesters correspond to poly(ethylene terephthalate) (PET), also used for producing containers for packaging (bottles). The polyester used in textile has a lower molecular weight and a higher crystallinity than the PET used for bottles. In the past, PET for textiles was produced by polycondensation of

dimethyl terephthalate and ethylene glycol, whereas the PET for bottles was produced from terephthalic acid and ethylene glycol. In fact, the latter method results in less regular polymer chains, which is more suitable for an amorphous material. More recently, the synthesis from terephthalic acid has become the most diffused [16] because of its economical and industrial advantages. Post-consumer PET from beverage bottles is currently recycled in packaging or in textile products. PET fibres are used alone or mixed with natural fibres like cotton, for improving breathability, or with elastane, to produce stretch fabric.

Polyacrylic fibres consist of poly(acrylonitrile) (PAN) macromolecules, often copolymerized with another monomer to improve dyeability and textile processability. Typical comonomers are vinyl acetate or methyl acrylate [17]. Acrylic fibres are produced by both wet and dry spinning and are mainly used to be knitted for producing wool-like fabrics. They can be used alone or mixed with other natural or synthetic fibres.

The most diffused polyamides, used in technical fabrics or in synthetic wool, are polyamide 6,6 and polyamide 6. The former is obtained by polycondensation from adipic acid and hexamethylene diamine, the latter by anionic polymerization of ϵ -caprolactame [18]. Polyamide is used alone or mixed with cotton or other synthetic fibres.

Polypropylene, produced with an isotactic structure by updated methodologies based on Ziegler-Natta catalysts, is a polyolefin commonly used in the production of non-wovens for hygienic applications and personal protection, because of its low cost and high water repellency, which discourages the growth of bacteria [19].

Elastane is a polyurethane-based copolymer, based on flexible poly(ethyleneglycol) blocks and rigid polyurea blocks (Figure 3). It is fundamental for producing stretched fabrics based on polyester, polyamide and natural fibres and thus is often mixed with these fibres [20].

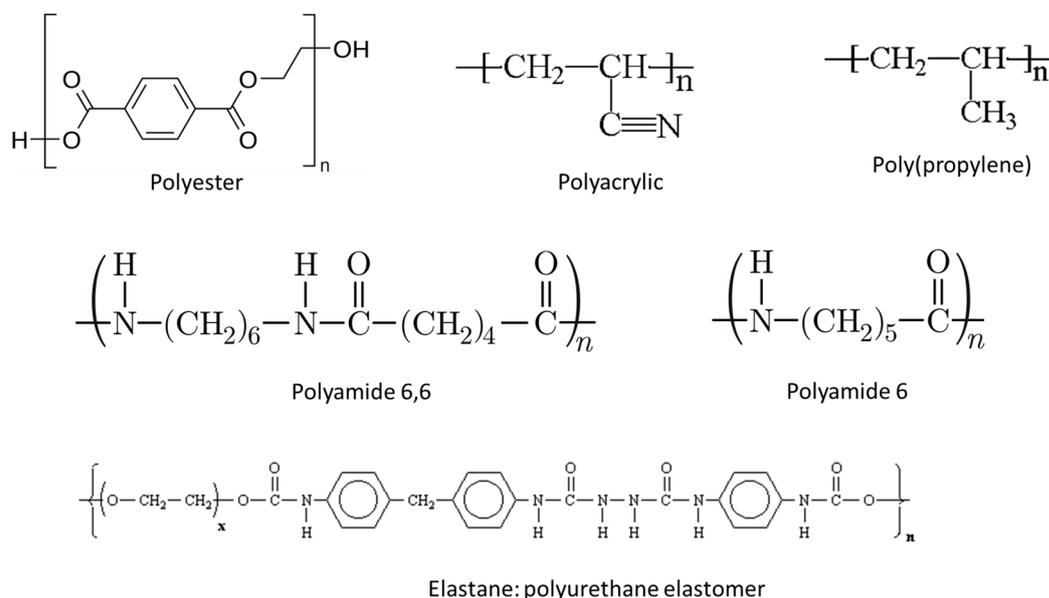


Figure 3. General structure of the main synthetic fibres widely used in the textile market.

All the described polymers are thermoplastic, so fibres can be produced by melt spinning. The orientation of polymeric macromolecules induced by the applied draw ratio favours the extensive formation of crystals that determine an increase in fibre strength. The recycling of the polymers by melt processing and re-spinning (mechanical recycling) could be advantageous. Nevertheless, the melting of mixed thermoplastics would result in a blend of immiscible polymers. Polymer blends are generally studied to achieve toughened materials, but not for the material strengthening. On the other hand, the crystallization process is often hindered by the presence of different polymeric phases. Thus,

mechanical recycling is difficult because of the impossibility of separating the different fibres that are of a micrometric thickness [21,22].

Currently, the textile sector is starting to make more and more use of biobased polymers (Figure 4) [23].

Global production capacities of bioplastics in 2022 (by market segment)

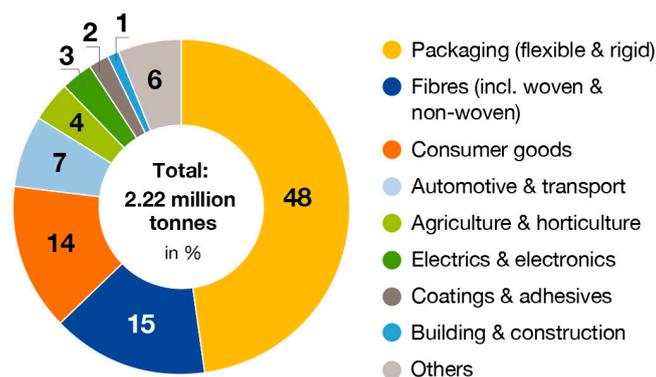


Figure 4. Global production capacities of bioplastics in 2022 [23]. Source: European Bioplastics, Nova-Institute (2022). More information: www.european-bioplastics.org/market and www.biobased.eu/markets (accessed on 12 February 2023)

In 2022, 333 ktms of bioplastics were used to produce fibres, and forecasts are predicting that in 2027 they will grow to 690 ktms [23]. Thus, in the group of man-made fibres, some bioplastics [24,25], like poly(lactic acid) (PLA), poly(butylene succinate) (PBS), bio-PET, poly(ethylene furanoate) (PEF), could also be included in the future. Replacing fossil-based products with biobased polymers is undoubtedly advantageous [26] because each fossil-based product at its end of life can generate a positive contribution to greenhouse gas (GHG) emissions. On the contrary, biobased materials of vegetal origin are carbon-neutral, because the CO₂ that they can emit at their end of life (because of incineration or degradation due to microorganisms) is balanced by the CO₂ consumed during the development of the plant. Hence, the use of man-made materials based on biobased monomers is currently promoted by institutions and adopted by companies, especially for short lifecycle applications. In this scenario, PLA, being both renewable and biodegradable in industrial composting plants, can represent an interesting perspective for replacing fossil-based polymers such as PET and PP [27,28], whereas biobased polyamides were developed to replace fossil-based polyamides in the near future [29].

Polyhydroxyalcanoates (PHA), synthesized by microorganisms, can be considered for producing natural bioplastic fibres [30], but their high cost and difficult processability is currently directing them mainly towards biomedical applications.

Natural fibres based on biopolymers such as proteins [31] and polysaccharides [32,33], obtained by waste from the agro-food chains, are also explored for applications in the textile sector, to promote the application of circular economy principles. For instance, casein from waste milk possesses high barrier properties to non-polar molecules and has been considered a candidate for fabricating paper coatings, adhesives, food packaging, textiles, and fibres [34]. Cellulose recovered from biomass [35,36] can be converted into regenerated cellulose fibres for textiles.

1.2. Microplastics Issue

Despite 30% of Europeans' wardrobes not having been used for at least one year, it is reported that a single laundry load of polyester clothes can discharge 700,000 microplastic (MP) fibres, which can end up in the animal and human food chain [2,3,5]. Microplastics, in fact, that were detected in teacups, human placenta and blood [37–39], might be the cause of toxicity or other hazardous and poorly known effects [40]. Apparel is responsible for 35% of microplastics invading oceans each year, as part of textile fibres (0.5 million tonnes of microfibrils per year). This high quantity of waste is due to the increased amount of waste apparel made of non-degradable synthetic fibres [41].

Synthetic microfibrils (MFs) are a ubiquitous class of MPs that pollute the marine and freshwater environments, thus raising an increasing global concern. These MFs mainly originate from textile industry discharges, textile releases in domestic and industrial effluents, illegal dumping and abandoned fishing gear [42–45]. Textile MFs [46,47] are the majority of MPs detected in the inflow and outflow of wastewater treatment plants.

PET and PA (mainly nylon 6 and nylon 6.6) account for the majority of synthetic MFs produced worldwide at a ratio of approximately 10:1 [48]. Consequently, the largest fraction of synthetic MFs found in municipal sewage sludge are MFs in PET [49–51]. However, because the analytical protocols for quantifying MPs in environmental matrices commonly rely on the physical separation of MPs [52,53], followed by their detection by optical microscopy and identification by FT-IR micro-spectroscopies [54] or Raman [55,56], MFs in PAs often go undetected due to their similar spectroscopic characteristics to protein materials [57,58].

Techniques combining thermal [59] or hydrolytic [60] decomposition of MPs coupled with mass spectrometry have been exploited to improve the accuracy of MP quantification. Most of the analytical protocols proposed so far involve time-consuming physical separation from the environmental matrix and particle counting, which often result in overestimation of the actual MP content due to natural fibres being mistaken for synthetic MFs, or underestimation due to a failure to separate or detect them due to their small size.

While adequate accuracy and sensitivity have recently been achieved for PET through depolymerization, chromatographic separation and quantification of its monomers by HPLC and spectroscopy or mass spectroscopy [57], the quantification of MFs in PAs is still a challenge.

Recently, the first procedure for an accurate and highly sensitive determination of total nylon 6 and nylon 6.6 content in complex environmental matrices, such as sediments, soils and sludge, was presented [61]. The salient features of the procedure include acid-catalysed hydrolytic depolymerisation followed by the attachment of a fluorescent tag to the obtained monomers and their quantitative determination by reversed-phase HPLC. This technique was adopted, together with one previously developed for PET [61], to develop a protocol for the analysis of PA and PET in sludge samples from sewage treatment plants. The actual hazard of MF depends on many factors, including the concentration and bioavailability of the fibres in the environment, their state of degradation and the way they are fed to and assimilated by consumers. In laboratory experiments, adverse effects appear to occur mainly at concentrations of MF higher than those currently measured in nature, except in situations and events of local pollution due to sedimentation, the currents and structures of aquatic landscapes, and the presence of textile production and waste disposal facilities; however, increased fibre concentrations can be expected due to continued release into the environment. There is evidence that exposure to MF can impair the fitness of individuals and can lead to defective community and population development, with consequences that are difficult to predict for the food chain [62]. Using biodegradable fibres instead of non-biodegradable can be advantageous to counterbalance this issue, but it cannot be currently considered a definitive solution. In fact, the degradation behavior of biodegradable polymers is generally slow. It depends not only on fibre composition but also on the peculiar physical-chemical and microbiological properties of the water recipe. Thus, more knowledge and research are needed regarding the best alternatives for textile materials.

1.3. Policy Initiatives

The increased consumption of clothes of the last decades is nowadays correlated with evidence that clothing has the fourth-highest impact on the environment [3]. Thus, policy initiatives have been considered, but only in the last few years (Figure 5). The current Waste Framework Directive [63] requires that Member States set up schemes for the separate collection of textiles, at the latest by 2025. Moreover, the directive sets mandatory targets for general municipal waste reuse and recycling of 55% by 2025, 60% by 2030 and 65% by 2035.

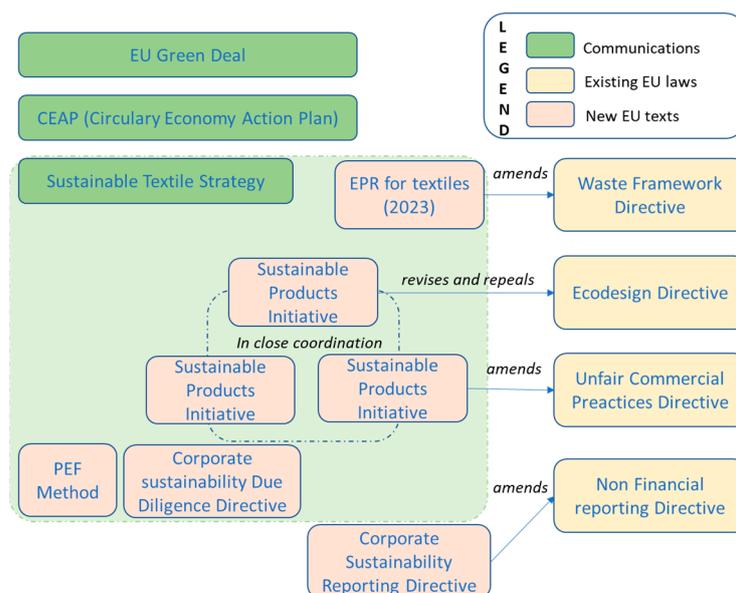


Figure 5. Scheme about integrated policy actions in Europe regarding textile recycling.

Other directives not specifically dedicated to textiles but present in the 2015 circular economy package could also mitigate some of the environmental impacts of textiles and clothes. In particular, the Packaging and Packaging Waste Directive [64] introduced targets for the recycling of 60% of all packaging by 2025 and 70% by 2030. Moreover, the Landfill Directive requires Member States to reduce the share of municipal waste land-filled to 10% by 2035.

EU legislation and initiatives focusing directly on textiles and clothes are directed towards helping consumers make more sustainable choices. For instance, the 2011 Textiles Regulation lays down rules on textile fibre names and for the labelling and marking of all textile products, including an obligation to state the full fibre composition of textile products at all stages of industrial processing and commercial distribution.

EU standards relating to textiles and clothing, developed through technical body CEN/TC 248 of the European Committee for Standardisation, are generally aimed at increasing the durability of fabrics and apparel. Some of the standards, in fact, deal with minimum performance requirements for certain types of textile products and the interactions of textile products with the environment. For instance, the EU Ecolabel for clothing and textiles, a voluntary certification program, establishes ecological criteria limiting the use of substances harmful to health and the environment, reducing water and air pollution, and criteria for extending the lifetime of clothes [65]. A voluntary instrument that Member States and public authorities can implement to the extent they wish is the EU's green public procurement (GPP) criteria for textiles, which facilitates the inclusion of green requirements in public tender documents [66].

The 2020 new circular economy action plan identified textiles as a key product value chain where urgent EU action was necessary and announced initiatives aimed at making production and consumption of textiles more sustainable. This vision was presented in

the EU strategy for sustainable and circular textiles, adopted by the Commission on 30 March 2022. According to the strategy, by 2030, textiles on the EU market should be durable and recyclable, made up largely of recycled fibres free of hazardous substances and produced in an environmentally friendly way while respecting social rights. Textiles should be collected at the end of their lifetime and their incineration and landfill reduced to a minimum thanks to innovative fibre-to-fibre recycling.

The proposed new Ecodesign Regulation would define minimum requirements for all products on the internal market regarding circularity (durability, reliability, reusability, reparability, upgradeability, recyclability, and possibility to remanufacture, recycle and recover materials). It would also include resource use and resource efficiency, the use of secondary materials, the presence of hazardous chemicals, environmental impacts, and expected generation of waste materials. Rules for specific products would be decided later, in delegated acts. The regulation would introduce a mandatory digital passport, which would, in the case of textiles, provide businesses operators and consumers with information, for instance, on harmful chemicals, repair, and fibre composition. In addition, the regulation would introduce an obligation for companies to make public information on the destruction of unsold products, while the Commission would be given the authority to ban such practices outright. Specific rules for textiles, including requirements for products, digital passports, green public procurement, disclosure of discarded products and measures banning the destruction of unsold textiles are planned for 2024.

Empowering consumers for the green transition would help in promoting products that are sustainable. Consumers would have to know if the producer offered a commercial guarantee of durability longer than the current two-year legal guarantee. They would also get information on the reparability of products, through a repair score or other specific repair information. At the same time, commercial practices that mislead consumers regarding environmental and social impacts, durability and reparability would be banned, and a number of specific commercial practices relating to false green claims would be added to the blacklist of practices that are considered misleading in all contexts. Substantiating green claims are expected to tie the information on sustainability that consumers and companies receive to a harmonised product environmental footprint (PEF) methodology. PEF methodology for apparel and footwear is also expected to be presented in 2024.

In its own-initiative resolution of 10 February 2021 on the new circular economy action plan, Parliament stated that the new EU textiles strategy should promote sustainability, circularity, traceability and transparency in the textile and clothing sector. Moreover, in its resolution of 10 July 2020 on the chemicals strategy for sustainability, Parliament stressed that a policy for sustainable chemicals requires alliances among sectors, to work on circular economy initiatives, including with the textiles industry. Parliament strongly promoted textiles recycling in the 2017 interinstitutional negotiations on the Waste Framework Directive, as part of the circular economy package.

2. Sorting of Textiles: Techniques and Facilities

2.1. Sorting Methodologies

High volumes of textile materials are often incinerated and landfilled due to the lack of specific collection of post-consumer textile waste, difficulties in separating the discarded textiles and costs associated with their sorting [67]. Nonetheless, reusing and recycling of end-of-life textiles are still considered to be better choices when compared to incineration or landfilling [68]. Regardless of the recycling process to be followed (i.e., chemical, mechanical or physical), textiles must be sorted before according to their colour, fabric type or raw material, quality and style [69].

Textile waste sorting is often carried out manually. Even though this is feasible, the high costs, low speed, and lack of automatization associated with it makes its implementation not suitable for the processing of large volumes of materials [67]. Moreover, this method is prone to misclassifications since it depends on the material content listed on the

product labels (might be faulty) and the expertise of the workers, who in turn experience high intensity and poor working conditions [68,70].

Other sorting methodologies can also be carried out, and these are divided into physical and chemical methods. Chemical methods (i.e., dissolution) are not usually considered for sorting waste textiles because they are destructive, hazardous and pollutant. On the other hand, physical methods include density and melting point measurements, among others [70]. In general terms, all the above-mentioned methods are expensive, time-consuming, and demand high training from the operator [71]. For this reason, a fast, non-destructive, and reliable method that is able to analyse large volumes of textile materials and can be implemented in-line is urgently needed for the waste textile recycling industry [68].

Some alternative physical methods have been explored during the last years to identify textile materials, such as the usage of radio frequency (RFID) tags that store information about the textile and could be read remotely. However, RFID tags being able to withstand several laundry cycles have not been developed yet. Furthermore, other methodologies based on the use of optical sensors have recently arisen. For example, colour sorting can be accomplished with the help of computer vision techniques based on an RGB camera [72–74]. RGB cameras are usually combined with infrared (IR) sensors to give further information about the textiles.

In this regard, Fourier-transform mid-infrared spectroscopy (FT-MIR) can be used to determine the material content of fabrics through the analysis of their spectral features that arise from the different molecular vibrations of the different textile components. For example, Riba et al. [67] classified textile samples based on synthetic (i.e., polyamide and polyester) and natural fibres (i.e., cotton, wool, and silk) by means of combining FT-MIR spectroscopy coupled to powerful mathematical recognition methods (such as principal component analysis, PCA). However, this technique cannot be implemented in-line and so is considered a complementary technique/extension to manual sorting [67,69].

2.2. The Advent of Near-Infrared Spectroscopy in Textile Sorting

Near-infrared spectroscopy (NIR) has gained a lot of popularity during the last decades because of its in-line and in-field capabilities (i.e., it performs real-time and non-destructive measurements). Therefore, a wide range of applications have been reported in the literature, such as in food science, agriculture, medicine, and the petrochemical industry, among others [75]. NIR spectroscopy (as FT-MIR spectroscopy) studies the energy transitions (i.e., absorptions) yielded by the interaction of the light (in this case the NIR region of 800 nm to 2500 nm) between vibrational energy states that are specific to every molecule, so a distinctive pattern can be obtained that can be thought of as a unique fingerprint (NIR spectroscopy is considered a fingerprinting technology). The NIR region displays broad bands with overlapping wavelengths that mainly provide information about NH, OH, and CH bonds, making this technique an optimal tool for the study of organic compounds [76,77].

Extensive literature can be found regarding the applications of NIR spectroscopy in textile sorting. In most of them, a portable NIR spectrometer working in diffuse reflectance mode is used and combined with powerful pattern recognition algorithms to sort and classify different textile materials (such as polyester, cotton, wool and polyamide, among others), reporting outstanding classification rates [67–71,75,78]. Such spectrometers offer the possibility of in-line implementation, but they rely on measurements performed on a single point of a sample, which could lead to misleading results when spectra are acquired from complex heterogeneous samples such as textile waste [79].

Hyperspectral imaging (HSI), which has been used for land observation remote sensing in satellites since the late 1960s, can provide both spectral and spatial information of the sample being tested; thus, heterogeneous samples are good candidates for this technology [80]. A hyperspectral image (also called hypercube) is a three-dimensional array that gathers two-dimensional spatial information ($m \times n$ pixels) and one-dimensional

spectral information (spectra of n wavelengths; Figure 6). In other words, for each pixel a full spectrum is collected (spectral/chemical information) while for each $m \times n$ -plane an intensity map of a certain wavelength is obtained (spatial information) [81].

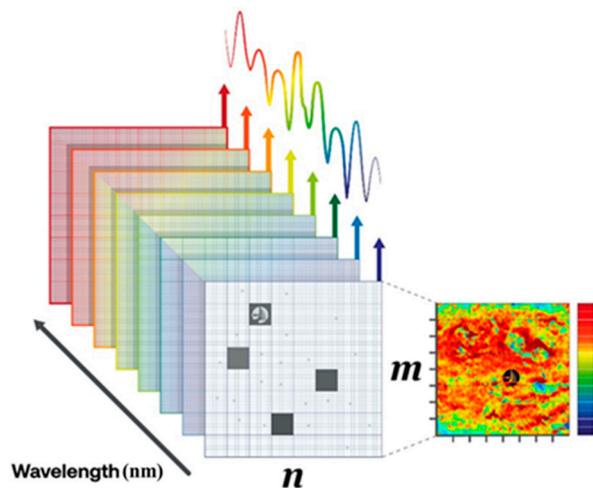


Figure 6. Graphic representation of a hyperspectral cube acquired with the industrial hyperspectral VISUM® HSI™. Each hyperplane (slide) of the cube corresponds to a spectral channel or wavelength. The 2-D image on the right displays the average intensities for all the wavelengths (heat map; see the scale).

Successful results have also been reported on the use of HSI in the NIR range for textile applications. Jiang and Li [82] obtained classification results over 90% when using a SWIR HSI system for the identification of foreign matter in lint samples. Results on the classification of synthetic fibres (such as polyamide, polypropylene, etc.) using a SWIR HSI system and multivariate classification algorithms were reported by Blanch-Perez-del-Notario et al. [83] when using a low-cost mosaic based Visible-NIR (VNIR) HSI system. The VNIR range brings some advantages, since it offers higher spatial resolution, cheaper and more compact cameras than the traditional SWIR Range. Moreover, the VNIR range enables sorting of blue denim, a relevant component of textile waste, that could otherwise be difficult to distinguish by SWIR HSI systems [83].

The workflow process for an industrial throughput for conventional NIRS and HSI technologies at an industrial level is similar. Target waste is placed onto a conveyor belt in which the monitoring systems are placed (i.e., cameras, spectrometers, lights; usually above the conveyor) so spectral acquisitions can be acquired from the waste stream. Spectral measurements are then processed with chemometrics/machine learning models implemented into the monitoring system, providing the identification of the textiles that will be physically separated using sorting equipment.

Besides all the advantages of NIRS for material textile waste sorting applications, one of the main drawbacks is that NIR measurements usually only provide chemical information from the surface of materials due to the low penetration of IR radiation. Because of that, the inner part of materials cannot be properly assessed, yielding misclassifications in thicker materials [68,84]. All in all, optical-based sorting advancements coupled to chemometrics and artificial intelligence is increasingly improving the performance of recycling textiles, which will keep more materials in recycling loops and help pave the way towards a circular economy [85].

2.3. Industrial Applications of Sorting

A novel approach to the sorting and recovery of textile cuttings and scraps may be seen in some successful initiatives at the European level, such as:

- sorting and preparing used textiles for recycling in the Amsterdam Area, “so that we become a hub for circular textiles and circular denim”, as alderman Marieke van Doorninck said [86];
- sorting the post-consumer textiles by material into various fibre classes using optical identification technology developed by Lounais-Suomen Jätehuolto Oy (LSJH), a company owned by 17 municipalities in South-West Finland [87].

Within the framework of the EU directives, the European textile and clothing employers’ association, Euratex, is already working on five textile recycling hubs in five European countries, to up-cycle textile waste and scale up industrially the collecting, sorting, processing and recycling of pre-consumer and postconsumer materials. One of these hubs is to be installed in Prato (Italy), (not far from SPIN-PET headquarters) with an annual collection capacity of 20,000 tons of wasted clothes and 14,000 tons of post-industrial textile scraps. A fibre-sorting unit for nearly 8000 tons/year capacity is included [88].

Finnish waste management company Lounais-Suomen Jätehuolto Oy (LSJH) will supply raw material to Infinited Fibre Company’s flagship plant from the full-scale textile waste refinement plant that is being prepared in the Turku region of Finland. LSJH’s plant will process all the end-of-life textiles of Finnish households in cooperation with other municipality-owned waste management companies. LSJH sorts the post-consumer textiles by material into various fibre classes using optical identification technology developed by LSJH and its partners. This ensures the quality of the raw material and the resulting fibre products [87].

3. Recycling Fossil Polymers

3.1. Polyester

Polyesters are polymers which contain the ester functional group in their main chain (Figure 3). Although there are many polyesters, in the framework of textile applications the term “polyester” as a specific material most commonly refers to polyethylene terephthalate (PET). The PET fibre dominates over 50% of the world synthetic fibre market. Woven polyester fabrics are used in apparel (textile industries) and home furnishings such as bed sheets, table sheets, and curtains. Similarly, industrial polyesters are used in tyre reinforcements, ropes, fabrics for conveyor belts, safety belts, coated fabrics and plastic reinforcements. Polyester fibres are also used to stuff pillows, comforters and cushion padding. To combine functional characteristics with greater comfort in contact with the skin, they are often woven in blends with natural fibres such as cotton. All these characteristics mean that polyester is used pure or mixed with other natural, man-made or synthetic fibres.

There are mainly three methods to recycle pure PET textile wastes. These methods can be classified into three groups, respectively: mechanical methods, thermo-mechanical and chemical methods [89], which will be discussed in a general way in Section 3.7.

Mechanical methods can be applied to all kind of waste including natural originated wastes such as wool wastes, cotton wastes, etc., and synthetic wastes. The general production steps of the mechanical method are cutting, blending and opening. Mechanical recycling machines gradually break the fabric into small pieces and make it fibrous, and these obtained fibres are reused in the production of yarn or non-wovens. The opened material, which is in web form, is fed into the spinning plants to obtain coarse yarns or fed into the non-woven plants. The mechanical method is the simplest and the one in wider use in terms of input textile materials when compared to the other methods. Mechanical methods break down the fabric, and the fibre length is reduced as an unwanted side effect, thereby affecting the spinnability and yarn strength. The main reason for this is the friction between the fibres that causes wear of fibres and melting of synthetic fibres. The fibres will have a shorter fibre length than the original fibres, and some dust will be generated. The costs are low, and generally the economic quality and value of the product is decreased with respect to the starting product.

The thermo-mechanical method can be used for synthetic fibre such as PET fibre wastes, polypropylene wastes, polyamide wastes, etc. This method is based on re-melting of wastes to obtain granules, which can then be used for different applications. This method is the most widely used for PET from beverage bottles [90,91] that is currently used in packaging [92] or textile applications [93]. Moreover, in view of an up-cycling approach, the blending of post-consumer scraps with appropriate rubbery compatibilizers allowed the production of toughened blends [94–98], not only from post-consumer bottles but also from PET/PE based packaging trays [99], suitable for injection-moulding or thermoforming parts for automotive, household appliance or protection applications [100].

In the case of textile-to textile recycling, granules coming from PET recovered from fabrics can be used in staple fibre production or in the plastic industry. Due to the PET degradation and viscosity drops with respect to bottle grade, granules are basically used in staple fibre production, since that production can also work with a higher melt fluidity. Thermo-mechanical methods are simple, and the economic value of the products is generally higher than that of the simple mechanical method. The thermo-mechanical method is mainly a re-extrusion process. This process consists of several steps: cutting, compacting/drying or drying and feeding to the extruder. Compacting makes the feeding to the extruder easier but extra steps and energy are needed and extra thermal effects occur on PET. Degradation is the most important problem in re-extrusion, particularly thermal degradation, thermo-oxidative degradation, hydrolysis and mechanical degradation [101]. Hydrolytic degradation (Figure 7a) consists of the decrease in PET molecular weight due to chain scission resulting in an increase in melt fluidity and is favoured by the presence of nucleophilic additives, such as humidity (which must be removed before extrusion through an appropriate drying step) and metal catalysts [102]. Thermal chain scission is also possible (Figure 7b).

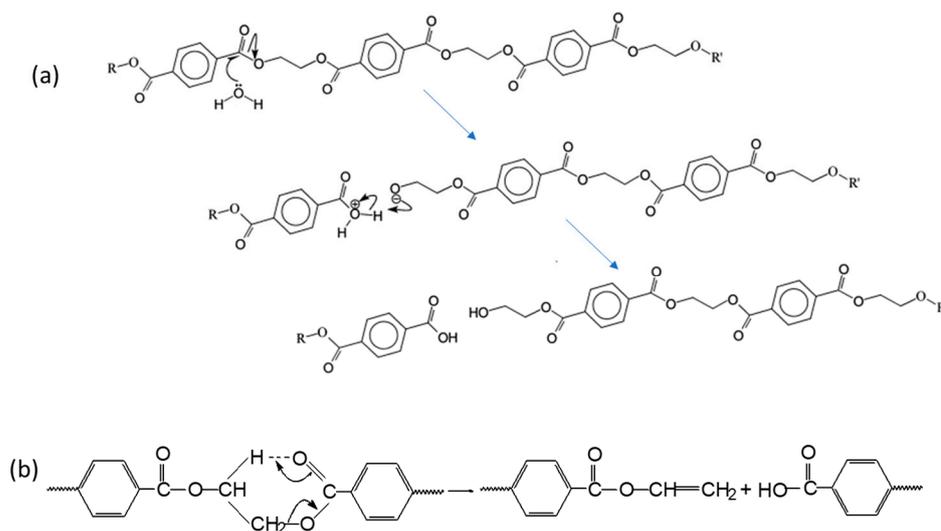


Figure 7. (a) Hydrolytic and (b) thermal chain scission of PET.

The PET waste fibres are light; therefore, they cannot flow easily from a hopper to an extruder by gravity alone, and the solid conveying rate may be insufficient due to low bulk density for supplying material to the next stages, which are plasticizing and melt conveying [103,104]. The excessive friction can cause a heat increase on the material, leading to thermal degradation. Another important feeding problem is bridging which is crossing of fibres from the hopper to the entrance of the extruder. This problem can be solved by special screw designs and force-feeders. Often, the fibre waste is also mixed with PET waste bottle scraps to limit this problem. Hydrolytic and thermo-oxidative degradation are also important in PET fibre re-extrusion. To eliminate hydrolytic degradation,

drying must be carried out before each processing operation in the melt. A special screw design to reduce stress shearing and nitrogen feeding can be recommended to prevent the thermal oxidation. To overcome the previous issues about feeding fibrous material to extruders, the work group in the ReSynt Project [105] have studied and developed a methodology for compacting recycled polyester frays and fabrics to easily feed the extruders by employing the classic feeders used for polymeric granules. In fact, the compacted material turns out to have a bulk density of 0.5 kg/L. In the Resynt project, recycled polyester-based compounds were developed by reactive extrusion to produce final injection moldable or thermoformable objects. Furthermore, the ReSynt project approached the possibility to track the recycling material and enable the consumer to know whether, and in what percentage, his product is actually recycled, by adding an appropriate tracer dye.

3.2. Polyamide

The two main polyamide fibre groups, wool (natural fibre) and PA6 (or PA6,6) (synthetic fibre), have very different recycling options, even though they both have amide bonds in their polymer backbone. Wool can be recycled mechanically, whereas for PA6, being a polycondensation polymer and a thermoplastic material, recycling options similar to those for polyesters are viable. PA6 can be melted and reshaped into new fibres of appropriate length and strength. As in the case of PET, a drying process is suggested before melting; it is revealed that the drying process prevents hydrolytic chain scission in wet materials and the intrinsic properties of PA6 polymer are preserved [106]. Su et al. [107] observed that the reprocessing up to 16 times of polyamide 6 resulted in a progressive decrease in molecular weight, but the polymeric structure, investigated by infrared analysis, was not significantly modified. Generally, thermal nonoxidative degradation of polyamide 6 and 6,6 results in the evolution of some CO₂, ammonia and formation of insaturations, nitrile and isocyanate groups [108]. However, these reactions were observed in experiments carried out for a long time (hours). On the contrary, reprocessing operations such as extrusion or injection-moulding are generally very fast (seconds).

PA6 waste is available as carpets and fishing nets, and these materials are an important feedstock for PA6 recycling [109]. The recycled polyamide shows similar characteristics compared to commercial PA6. Recycling carpets is also possible by blending various carpet components through a reactive extrusion and compatibilization process, yielding lower-quality products [89,110]. The low number of PA6 suppliers and recyclers makes recycled polyamide fibres more expensive than PET fibres.

3.3. Acrylic

Acrylic fibres are synthetic fibres made of at least 85% of acrylonitrile monomer with other comonomers, typically vinyl acetate or methyl acrylate, to obtain a final fabric with different characteristics. Acrylic fibres are used for sweaters, pants, hoodies, and a variety of other types of cold-weather clothing. Additionally, acrylic is a popular material for carpeting, upholstery, rugs, and other traditionally wool-dominated homeware product categories. It is estimated that, every year, in Europe, about 7700 tons of acrylic textile waste are disposed of by landfill or incineration. Polyacrylics can be mechanically recycled, comparable to wool. The process involves colour sorting, cleaning, unravelling and spinning again [111]. Polyacrylics cannot be melted, and although they can be dissolved, presumably no physical recycling methods are under development. Polyacrylics are formed by addition polymerisation and can therefore not be depolymerised by solvolysis methods.

Being extremely durable and able to repel water, acrylic fabrics are the most used for outdoor awnings and patio furniture. However, to have the required characteristics for such use, it is necessary to apply chemical finishes to the acrylic fabrics to increase the hydro and oil-repellency, the resistance to UV and to fire. This makes the management of acrylic textile waste tricky. The EU-funded REACT project [111] aimed to identify possible processes to treat and recycle textile waste in an ecological and economic way. In this project, a method was developed to remove unwanted substances (fluorocarbons, melamine

and acrylic resins, anti-mold agents) from the waste of acrylic fabrics with an environmentally friendly process to improve recycling, to improve sustainability and reduce the risk to the environment and to health. After elimination of the finishing chemical, the mechanical recycling and spinning permit the reuse of acrylic fabrics as raw material for other production cycles, also in combination with virgin fibres.

Pyrolysis of waste acrylic textile between 500 and 800 °C and the steam activation of the chars begin the production of activated carbon [112]. A mechanical recycling approach was attempted to prepare non-woven panels by using a needle-punching method [113] or to fill the recovered acrylic in between a double wall for thermal insulation in buildings [114].

Recently, carbon fibres were prepared in the laboratory from textile grade polyacrylonitrile (PAN) fibres thanks to a systematic investigation into the stabilization and carbonization of textile-grade PAN fibres [115]. The tensile properties of carbon fibres from textile-grade PAN fibres were found to be comparable with those of commercial PAN fibres. Thus, textile-grade PAN precursor can be an indispensable candidate for low-cost carbon fibre production, and this process would represent in the future an interesting up-cycling option. After all, the strong interest in this approach is testified to by the production of carbon fibres starting from cotton [116] and cotton/polyester mixtures [117] recently being investigated as well.

3.4. The Case of Elastan in Mixed Polymer Fabrics Recycling

Natural fabrics have good moisture absorbency and heat isolation but also have poor wrinkle resistance and elasticity. Therefore, synthetic fibres are usually mixed with natural fibres to obtain proper garment properties. Among them, elastane (also known as spandex) is a typical synthetic elastic fibre that exhibits a highly reversible extension of 400–800% [118]. This capacity has become the prerequisite for fashionable or functional apparel such as leisurewear, underwear, outer clothing, and sportswear. The worldwide consumption of Elastane has grown by 30–40% per year. It is present in fabric mixed with cotton as a component in 2–5%, but in combination with PET or PA it can reach up to 20% by weight of the fabric. Generally, it is used in knitted fabrics, in particular in plated plain knitted fabric [20]. Thus, the presence of elastane can make difficult the recycling phase of several fabrics.

The necessity of revealing and quantifying elastane in fabric was considered by different authors. Phan et al. [119] identified the thermogravimetry coupled with the infrared analysis of evolved gases as a good methodology to quantify elastane in a sample. Very recently, Boschmelier et al. [120] deepened the use of thermal methodologies for elastane quantification, considering several samples of fabric including PA, PET, cotton, wool and elastane, and proposed a methodology based on DSC analysis that they suggest as an alternative to the use of the EN ISO 1833-20:2018, based on extraction of elastane with solvent. The use of near-infrared detection is generally considered fundamental not only for quantification but also for automatic on-line sorting of fabrics scraps [68].

Regarding the recycling of fabric containing elastane, mechanical recycling was attempted by Yin et al. [121], who adopted for PA/elastane fabrics a simple process using only heat and ethanol, which involved heat treatment at 220 °C for 2 h under atmospheric pressure followed by washing with ethanol. The authors stated that the process was able to remove essentially all the spandex component from the blended fabric, resulting in a fabric consisting mainly of PA yarns.

Lv et al. [122] removed in a preliminary step the elastane by extraction with DMF solvent (solvent ratio 50:1 in mass) at 70 °C for 4 h, then carried out a thermal treatment at 220 °C followed by washing. The obtained material was successively compounded in a Brabender discontinuous mixer and characterized. Rheological measurements showed that viscosity of treated fabric samples was lower than that of untreated fabric composites, as an effect of the spandex removal after treatment. The recycled plastics from treated fabrics exhibited good mechanical properties with a tensile strength of 46.6 MPa and a

Young's modulus exceeding 2.4 GPa, but with a decreased elongation at break. However, the addition of 20% of recycled material to pure polyamide resulted in a material with a very similar mechanical behavior with respect to pure PA.

T-shirts with 95% cotton and 5% elastane were selected to be dissolved in H₂SO₄ aqueous solution, NaOH/urea aqueous solution, or LiCl/DMAc solution. Regenerated cellulose films were obtained from these three solvent systems, and their structures were studied by Fourier transform infrared spectroscopy (FT-IR) and scanning electron microscope (SEM). All three solvent systems could effectively dissolve the cellulose fibres, while resulting in different elastane components. The film obtained in NaOH/urea aqueous solution contained large undissolved elastane fibres, and its mechanical properties were weaker than the other two samples. The elastane fibres were partially hydrolyzed in H₂SO₄ solvent but were completely dissolved in LiCl/DMAc solvent, so this film had a uniform morphology, high transparency, improved tensile strength of 121.63 ± 6.16 MPa, lowest WVP of 1.03 ± 0.53 × 10⁻⁷ g m⁻¹ h⁻¹ Pa⁻¹, and a maximum decomposition temperature of 359.97 °C. Therefore, this work [123] demonstrated the possibility to directly recycle waste cotton–elastane fabrics through dissolution and regeneration, and the resultant films have potential applications as packaging materials.

3.5. Non-Woven Production Using Recycled Fibres

Non-woven fabrics are broadly defined as web structures bonded together by entangling fibres mechanically, thermally fusing the fibres or chemically bonding the fibres [124]. Typical raw materials are natural fibres (cotton, jute, linen), regenerated fibres (Bamboo fibre, viscous fibre, Tencel, modal fibre), synthetic polymer: PP, PE, PET, PA, polyester, PCL, PLA). The various methods for bonding are:

1. Adding an adhesive.
2. Thermally fusing the fibres to each other or to the other thermoplastic fibres or powders.
3. Fusing fibres by first dissolving and then re-solidifying their surfaces.
4. Creating physical tangles or tuft among the fibres.
5. Stitching the fibres or filaments in place.

The non-woven fabrics can be divided into eight types according to different manufacturing processes:

- (a) spunlace non-wovens;
- (b) Heat-bonded non-woven fabrics
- (c) pulp air-laid non-wovens
- (d) Wet-laid non-woven
- (e) Spunbond non-wovens
- (f) Acupuncture non-wovens
- (g) Stitch non-wovens

They are used in numerous applications, including baby diapers, adult incontinence products, wet wipes, surgical drapes and covers, liquid cartridge and bag filters, face masks, air-conditioning filters, soil stabilizers and roadway underlayment, erosion control, drainage systems, insulation (fibreglass batting), pillows, cushions, and upholstery padding, carpet backing, automotive headliners and upholstery, house wraps, and disposable clothing (foot coverings, coveralls).

The non-woven market size is forecast to reach US\$55.1 billion by 2026, after growing at a CAGR of 3.8% during 2021–2026 [125]. The impact of the COVID-19 pandemic was minimal on the growth of the non-woven fabric market. As governments provided economic supports for SMEs and other benefits, consumers worldwide could access essential commodities and personal protective equipment. The grow of the non-woven fabric market is mainly attributed to the increase in demand for non-woven products and solutions for hygiene, medical, construction and filtration applications.

In the contest of recycling synthetic fibres, the production of non-wovens is very important because mechanically recycled fabric usually results in fibres with a lower length that is often suitable for the production of non-wovens [126]. Thus, the non-woven market, driven by a significant growth, could partially absorb fossil polymer fibres.

3.6. Composite Production Using Waste Textile

Adding fibrous reinforcement consisting of fossil textile in composites is a potential way to increase the recycling rate and also an attempt to develop high-value applications from recycled fibres [127]. The presence of fibres affected the thermo-mechanical features and eco-compatibility of biodegradable matrices (polylactic acid, thermoplastic starch, cellulose) or non-biodegradable ones (polypropylene, polyester, polyethylene) in composite preparation. In this framework, cotton based-fabric, opened into a fibrous form by garneting, was blended with polypropylene for realizing 3D woven textiles [128]. Alternatively, it was mixed with hardwood strand core material for preparing oriented panels [129]. An insulation material was realized by a non-homogeneous medium obtained from discarded components of cotton (70%), wool (15%) and acrylic and polyester fibres (15%) recovered from the disposal of plastic bottles [130]. Denim fabric was converted into epoxy-based composites by adopting, as fabrication route, the needle-punching non-woven [131]. Cloth scraps and clippings of cotton and jute materials, together with glass fabric, were incorporated into thermosets consisting of unsaturated polyester resin for fabricating laminate composites with the stacking of fabric plies [132]. The preparation and characterization of green composites from PLA and cotton fabric waste (CFW) by considering the effect of the CFW content on the composites was considered by O-Charoen et al. [133]. CFW pellets were prepared using compression molding and die-cutting techniques and then compounded with PLA in a twin-screw extruder to form composite granules, and it was demonstrated that CFW could improve the tensile modulus, flexural strength, and Izod impact strength of PLA. However, an increased water absorption was also observed for the composite with respect to the pure matrix. The prepared composites based on PLA/CFW could be promising materials for household products such as home decorations and food trays, etc. Moreover, by improving the interfacial adhesion between PLA and CFW, other applications could be considered in the future.

In the case of recycled plastics being used as matrix, Rajendran et al. [134] has considered the environmental impact assessment by using the life cycle assessment (LCA) approach for glass fibre and flax fibre fibrous reinforcement. The resource depletion potential and global warming potential were evaluated for glass fibre- and flax fibre-filled recycled plastics and compared with their respective virgin alternatives. The resource depletion and global warming were compared at equivalent stiffnesses. The results indicated that reinforcing recycled plastics can significantly reduce the resource consumption and global warming in civil and infrastructural applications. However, the composites made from virgin plastics resulted in a greener and sustainable alternative in the automotive applications, where a lower content of fibres can be admitted. In the case of fibres that come from textile waste, the final results could certainly be different and the impact would decrease as the specific elastic modulus of the selected fibres increases.

Woven sisal fibre-reinforced green HDPE composites were prepared by a low-cost manufacturing process by reusing polyethylene bags made of sugarcane ethanol (green polyethylene) reinforced with sisal fibres [135]. The hot compression moulding (185 °C) was used to mould composite structural board, which showed higher tensile strength and a lower modulus of elasticity in comparison to pure HDPE specimens. On the other hand, randomly arranged sisal fibre-reinforced green HDPE composites had higher values for modulus of elasticity than pure HDPE, as well as composites with woven sisal fibres. These materials could be used in structural applications, such as drywall and insulated panels. The main reason to use recycled polymers and natural fibres (without chemical treatment) is due to the possibility to produce a sustainable material for thermal and acoustic insulation.

3.7. Chemical Recycling to Obtain Separated Materials, Monomers or New Products

Various chemical recycling technologies have been developed for textile waste, and for each one drawbacks, advantages, and disadvantages can be found. These processes are described below.

Pyrolysis/gasification could be utilized with unsorted and multi-material textile products even if most of the current studies used a single-type textile waste as the substrate [136]; however, it requires high temperatures and therefore is a process with a high energy consumption. Pyrolysis of textile waste typically results in pyrolytic gas (H_2O , CO_2 , CO , H_2 , CH_4 and other light hydrocarbons) and solid residues (>20% by weight). During pyrolysis, a large amount of heat must be transferred across the reactor, hence the type of reactor used for pyrolysis is crucial for obtaining a high process efficiency. Generally speaking, a continuous flow pyrolyser led to a higher yield of gas pyrolysate, while a semi-batch pyrolyser led to higher yields of liquid pyrolysate and solid residue [137]. However, almost all studies used a single set of parameters and limited types of pyrolyser (e.g., batch or fixed-bed reactor). Many pyrolysis processes have focused on the production of carbon materials for different applications, for example as adsorbents for pollutant removal. For this application, pre- and post-treatment procedures are necessary to enhance the surface area and porosity of the textile waste-derived carbon residue. Several studies have reported that polyester textile waste-derived adsorbents show better performance than other biomass-derived activated carbons [138]. Only rough economic and environmental assessments and estimations are available, and comprehensive studies on the energy and economic analysis of the pyrolysis process for textile waste recycling are necessary [139]. In general, in chemical recycling, plastics are broken down into chemical intermediates (oligomers) or their basic building blocks (monomers) by adding chemicals. Regarding chemical recycling of PET in particular, degradation, alkaline and acid hydrolysis, methanolysis, glycolysis, microwave, ionic liquid, phase-transfer catalysis, combination glycolysis–methanolysis, glycolysis–hydrolysis, methanolysis–hydrolysis and the kinetic study are reviewed by Damayanti et al. [89].

Various processes are described in the literature for chemically recycling PET textiles [140]:

-Hydrolysis; -Alcoholysis (glycolysis/methanolysis); -Aminolysis

At the end of these processes, the oligomers or monomers must be purified by filtration and/or separation processes, and dyes may be difficult to remove. After purification, the purified oligomers or monomers can be processed into new plastics or other chemical products.

The economic efficiency of chemical recycling is only given with larger mass flows. In addition, it must be considered that today's processes are usually very energy-intensive, and the economic viability depends strongly on the quality of the purification. Generally, only individual fibre types can be dissolved and processed from chemically recycled textile blends.

New technologies for chemically recycling textile fibres are emerging around the world. They have the potential to substantially reduce the textile industry's huge environmental footprint. Consumers could soon be wearing brand-new clothes made from garments that otherwise would have ended up in a landfill or incinerator. However, the technologies are far from mature [141]. Chemical recycling has been especially pursued in the case of the separation of polyester and cotton. According to Palme from Chalmers University of Technology, four approaches can be followed to deal with polycotton waste streams [142]: (1) Dissolve cellulose, maintain PET; (2) Degrade cellulose, maintain PET; (3) Maintain cellulose, dissolve PET; (4) Maintain cellulose, degrade PET.

A certain number of commercial initiatives concerning the recycling of blended textiles, mainly cotton/PET, which is highly abundant in waste, (Table 1) and several European projects (Table 2) about this topic have been reported.

The analysis of these sources (industrial initiatives and European projects) are evidence of a strong activity of companies and research centers, also involving actors of the waste management chains, regarding textile chemical recycling. Due to the presence of

mixed polymers in recovered and sorted textiles, chemical recycling methodologies often combine separation with the possibility of upcycling the materials, throughout the production of specialty chemicals that otherwise will be produced from freshly extracted oil.

Table 1. Commercial initiatives related to chemical recycling of cotton/PET.

Company and Reference	Input Stream	Technology	Product	Milestones and References
HKRITA/H&M Foundation [143]	Cotton/PET	Hydrothermal decomposition of cotton into cellulose powders and the separation of polyester fibres from blended fabrics (without compromising the polyester fibre quality)	Good-quality polyester fibres to make new textile and a cellulose powder	2021—Denim supplier ISKO gets a Green Machine from Türkiye. A consortium of actors in the textile sector, including GIZ, VF Corp and Dakota, join forces to launch a feasibility study to deploy The Green Machine in Cambodia by 2022. 2020—Kahatex, a large supplier in Indonesia, places the first order of the Green Machine. Fashion brand Monki releases the first collection made from the Green Machine technology. A pilot on cotton farming using the cellulose powder (one of the outputs from the Green Machine) is initiated in India. 2018—A pre-industrial-sized system opened in Tai Po, Hong Kong; brands and stakeholders were invited to test and implement this technology within their own operations [144].
Worn-again technology [145]	Cotton/PET from non-reusable textiles	Closed-loop solvent system able to decontaminate, separate and extract polyester and cellulose	PET and cellulosic pulp	31 May 2022—Worn Again Technologies announces plans to build a new textile recycling demo plant in Winterthur, Switzerland [146]
Sodra with Once More [147]	Cotton/PET		Cellulose pulp for textile	3 June 2022—production capacity of OnceMore® rising to 6000 tonnes
Circ, Inc (formerly, TYTON BioSciences LLC) [148]	Cotton, PET, poly-cotton	Heat and pressure-based hydrothermal technology	Cellulose and PET monomers	14 July 2022—Marubeni America Corporation, has made additional investment in Circ, Inc. (formerly, TYTON BioSciences LLC) [149], that has developed methods for recycling cotton and polyester fibres from waste textiles.
Block Texx [150]	Cotton/PET		Cellulose pulp and PET	16 August 2021—Australian start-up BlockTexx has acquired a site at Logan, Queensland, and it was working on the final design for the plant. BlockTexx says production will begin in 2022, using the company's proprietary Separation of Fibre Technology (SOFT) solution, at the plant which will recycle around 4000 tonnes of textiles in its first year [151].

Table 2. European projects dealing with chemical recycling of textiles.

Project Acronym and Reference	Input Stream	Product	Key Information
RESYNTEX [152]	Cotton, nylon, PET, wool	Monomers (glucose, TPA, EG) protein hydrolysates, polyamide oligomers	UE HORIZON 2020, from June 2015 to May 2019. It developed a new demonstration process based on a synergistic chemical and biotechnological cascading separation/transformation approach of textile basic components (proteins, cellulose, polyamide and polyester) from textile blends as basic feedstock materials for chemical and textile industries. Liquid and solid waste treatment and valorisation closed the loop
Trash2Cash [153]	Blended textile and paper waste	Regenerated cellulose	From June 2015 to November 2018. A newly developed eco-efficient cotton fibre regeneration process (Cellulosic Regeneration), a new polyester regeneration technology (PET De-Re-polymerisation) and a polyester recycling technique (Chain Extension Upgrading) were developed
RETEX [154]	Cotton, Polyester	Fibre	Interreg (V programme) EU Project, from October 2016 to October 2020. Three value chains were identified to be developed to obtain fibre and plastic by mechanical recycling: (1) Cotton/polyester blends; (2) 100% cotton; (3) 100% polyester
CISUTAC [155]	Horizon Europe project	Fibre	Horizon Europe project, co-funded by the European Union, from September 2022 to August 2026 (ongoing). Tests for fibre-to-fibre textile recycling technologies for post-consumer polyester and cotton were carried out. Pilot scale processes demonstrated the industrial application of new technologies for textile circularity
T-REX [156]	Cotton Polyester Polyamide	Fibre	Horizon Europe project, from October 2022 to August 2025. It collected and sorted household textile waste and demonstrated the full recycling process of polyester, polyamide 6, and cellulosic materials from textile waste into new garments.

3.8. Enzymatic Recycling: To Monomers or New Products

Polymer biodegradation consists in reduction of molecular weight under the transformative action of microbial enzymes. Enzymes consist of mainly peptide natural polymers with a 3D structure of folded long chains, and they can behave as catalysts of various reactions. Synthetic polymers have a high molecular weight and diffuse with difficulty across a cell wall or microbial cytoplasmic membrane. Thus, the reduction of the polymer molecular weight is necessary for the bacteria. Enzymes engaged in polymer degradation (defined as exoenzymes) show oxidative or hydrolytic functionality [157]. The enzymatic action can be described as depolymerization, and the obtained monomers and/or oligomers could be used in a subsequent polymerisation process. Chemical structural features of polymers as well as their crystallinity content can play a pivotal role in polymer biodegradation. A high crystallinity can limit biodegradation, suppressing molecular diffusion in the material. During the enzyme-catalysed reaction, the synthetic polymer's substrate binds to the highly specific enzymes' active site. Compared to the chemical recycling processes, biocatalytic recycling processes are generally eco-friendlier [158]. Moreover, the efficiency of the enzyme-catalysed biotechnological processes can be improved through enzyme engineering [159].

The past few years have witnessed the discovery of several enzymes able to catalyse the hydrolysis of amorphous PET, including several hydrolases, lipases, and esterases. After the work of Yoshida et al. [160] and the enzyme stabilization in compost investigated by Shirke et al. [161], Tournier et al. [162] studied the enzymatic degradation of PET and obtained a very good result by using leaf-branch compost cutinase (LCC), yet used to hydrolyse amorphous PET. The enzymatic step was preceded by the melting and cooling in liquid nitrogen of the sample, so that PET was in its amorphous state. When physical-chemical treatments are combined with enzymatic ones, the full process can be defined as chemoenzymatic [163]. This methodology could be applied more extensively to textiles in the future.

With a similar approach based on cutinase, Kaabel et al. [164] produced terephthalic acid from PET/cotton fabrics. Cotton was thus separated from PET. Interestingly, a comparison of terephthalic acid production from petrol to that from enzymatic PET depolymerization revealed a 69% lower energy requirement and 17% lower greenhouse gas emissions for the latter, suggesting that the development of biocatalytic recycling processes be continued [165]. On the contrary, Navone et al. [166] used the selective digestion of wool fibres from wool/polyester mixed fabrics. Complete degradation of wool fibres was achieved by using a keratinase in a two-step process with the addition of a reducing agent and undigested polyester fibres. The obtained nutrient-rich keratin hydrolysate could be used in microbial growth media or incorporated into bio-fertilisers or animal feed, contributing to the development of the circular economy. Quartinello considered a mixed fabric containing wool, cotton and PET [167] and suggested a first hydrolysis step of wool using proteases followed by a second enzymatic hydrolysis of cellulose using cellulases, and finally by poly(ethylene terephthalate) (PET) hydrolysis using cutinases. Piribauer et al. [168], after the enzymatic degradation of cotton in PET/cotton fabric, produced a recycled polyester that was fed back into the textile processing chain, and finally towels were obtained. Polyamides can be also degraded by enzymatic processes [169]. Carbios company [170] and Novozyme [171] are currently working in the field of enzymatic degradation of textiles.

4. Limitations in Textile Recycling

The collection and analysis of information regarding textile recycling have suggested that several strategies were explored by researchers in the different case studies. Much research is ongoing, and it is difficult to compare the alternatives proposed in the different studies. An analysis of limitations is summarized in Table 3.

Table 3. Limitations of the different textile recycling methodologies.

Recycling Method	Limitations
Mechanical recycling	Shorter fibres, coarse yarns and lower quality of products (mainly non-woven). Preliminary sorting can be useful for a better reliability of the process.
Thermo-mechanical recycling	Its quality depends on composition; thus, it is better for homogenous polymers or for some specific blends/composites with reliable composition. For this kind of recycling, preliminary sorting is necessary.
Chemical recycling	With the exception of pyrolysis, it requires the use of solvents or chemicals, hence generally high investments, including suitable measures for protecting health and environment, are necessary. In the case of pyrolysis, a solid absorbent and gases are obtained. Their market demand should be high in order to create a profitable supply chain.
Enzymatic recycling	It can be applied to ester, amide, ether or glycoside linkages; thus, it can be applied to polyesters, polyamide or cotton. Polyaddition polymers (acrylics, polyolefins, etc.) generally could not be decomposed by enzymatic paths. Preliminary homogeneization and reduction of dimensions is useful.

The mechanical recycling is a cheap process transforming fabric into short fibres that can be used for obtaining staple fibres for non-woven or coarse yarn for textiles having limited resistance. Thus, the limitations are above all the low quality of products.

The thermo-mechanical process consists of re-processing the material in the melt by extrusion to obtain granules that can be spun or moulded again. The spinning is more difficult with respect to virgin polymers, unless we consider staple fibres. Other processes such as extrusion, injection moulding or thermoforming can be more suitable for obtaining high-added-value products. Case-specific chains could be built in the future to promote this strategy.

The production of composites and staple fibres is linked to mechanical recycling, as the raw material is mainly obtained by this methodology. Nevertheless, composites and staple fibres can also be produced by the thermo-mechanical methodology. This technique can be suitable for specific textiles and can result in products with higher added value than the case of mechanical recycling.

Chemical recycling consists of transforming the textile in materials and chemicals that can be used as building blocks of polymers or for other industrial purposes, replacing the fossil-based ones.

Enzymatic recycling consists of using reactions occurring in microorganisms catalyzed by enzymes to decompose polymers. Sandin and Peters [172] presented evidence that textile reuse and recycling in general reduce environmental impacts compared to incineration and landfilling, and that reuse is more beneficial than recycling. Benefits mainly arise due to avoiding the production of new products. Such benefits are therefore negligible in cases of low replacement rates or if the avoided production processes are relatively clean. Additionally, for reuse, induced customer transport may cause an environmental impact that exceeds the benefits of the avoided production, unless the use phase is sufficiently extended.

5. Conclusions and Perspectives

The recycling of fossil polymers present in waste textiles could be highly beneficial to save fossil resources and so prolong the life cycle of these materials, but it is difficult because textile products are often the combination of different polymers. Despite sorting technologies being developed to support efficiently the online rapid selection of materials, the separation of single polymers is difficult. Some alternatives for recycling monomaterial textiles such as PET, PA and acrylics, such as attempting up-cycling approaches through mechanical or thermo-mechanical methodologies are possible, but these methodologies are less promising for mixed materials. Thus, separation by chemical or enzymatic methodologies could be promising to separate/eliminate some polymeric fractions (Figure 8).

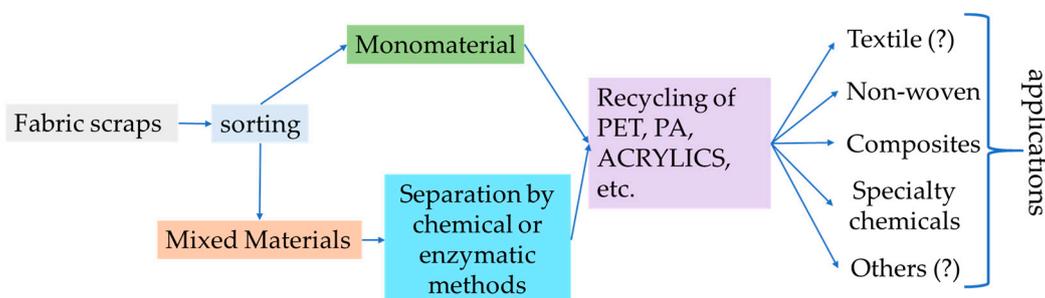


Figure 8. Integrated summary scheme about textile fabric scrap recycling.

Despite more work being necessary to explore new specific recycling chains, several possibilities can be currently considered for final applications based on the updated literature. The application in textiles seems difficult, especially due to the high thermo-mechanical performance required for re-spinning fibres. Non-wovens, used in personal protective equipment, filtration devices, packaging and furniture, allowing the use of shorter

fibres, can be considered a good alternative for mechanical recycling. Composites are another interesting field where recycled textiles can be used in the form of single fibres, fabric or non-woven, depending on the specific requirements (insulation, soundproofing, rigidity, strength, etc.), and in some cases the preliminary separation of different polymers can be skipped. It should be noticed that a high durability should be preferentially considered for final applications, so that the polymeric carbon is stored for a long time and the occurrence of the product in waste is not only slightly postponed.

Chemical modification processes resulting in specialty chemicals can also be considered. In this case the conversion of polymers into oligomers or monomers is considered. In the form of oligomers, polymers can be employed in the production of materials, (for instance, polyurethanes from waste PET oligomers used as diols) or components for glues, binders, coatings and adhesives. Other specific applications can certainly be developed in the near future, as well as new studies that will show their sustainability. Policy initiatives should promote these research activities that could contribute to converting a fossil-based industrial system into a circular one. There is a strong need to make innovative high-quality products designed, engineered and processed to meet specifications for a different economy and society, rethinking raw materials, processes and services and investing in people and technology.

It is not possible to compare the different methodologies in terms of environmental impact, as they are case-specific. A pivotal point is that for an effective recycling chain, a wide market demand should be present. The increasing attention of consumers to environmental questions is thus creating a positive contest for new recycling chains.

In general, a textile system requires a radical and creative reworking for both planetary and human health, remembering that up to 80% of a product's environmental impact is determined at its design phase. The necessity to establish more collaboration between supplier and consumer is another important element to be considered. The shift to more closed-loop systems seems underway, driven by regulatory efforts which, supporting the circular economy by innovations, new recycling solutions, efficient use of natural resources, use of by-products and creation on new jobs, will be beneficial for safeguarding the environment, ameliorating the quality of life (QoL) and reducing waste, pollution and poverty. For this purpose, the use of biobased fibres should be promoted. In particular, the use of biobased materials and sustainable biotechnologies will be useful in minimizing the environmental impact and bettering the quality of life.

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