Chapter

An Evaluation of Recycled Polymeric Materials Usage in Denim with Lifecycle Assesment Methodology

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Abstract

Today, World economy is only 8.6% circular, which creates a huge potential in materials reuse. To close the Emission Gap by 2032, this percentage needs to be doubled. The circular economy ensures that with less virgin material input and fewer emissions. With the help of effective recycling technologies, virgin material use can be decreased and especially petroleum based materials impact can fall within planetary boundaries. This book chapter analyzes different chemical and biological recycling technologies, their advantages and challenges in denim production. Moreover, Life Cycle Assessment (LCA) analysis will be used to evaluate the environmental impact of recycled polymeric materials usage in denim fabrics. Finally, it concludes by challenges and the future of chemically recycled materials in denim production and opportunities to evaluate waste as a raw material to design circular systems.

Keywords: denim, recycled polymeric materials, life cycle assessment, design for sustainability, circularity

1. Introduction

Recycling has been placed on the political agenda in the early 1990s with the rapid increase in the production and consumption of plastics and the increasing concerns over limited resources and plastics waste. As a result, a new industry, the plastics recycling industry, emerged. The main concerns behind the emergence of this industry were the environmental impact of plastics production, its waste management and the fact that almost all synthetic polymers are non-degradable. In many countries, used plastics end up in landfills and the landfill space, as well known, is limited. Two approaches, as a solution to these challenges were discussed to achieve the goal of sustainable development for material consumption [1]: dematerialisation and transmaterialisation.

Dematerilization, increasing the rate of recycling, is highligted as one of the three solutions in a recent report published by Ellen Mac Arthur Foundation [2]. Global Commitment Report stated these three solutions as: to eliminate the plastic

items we do not need; innovate so all plastics we do need are designed to be safely reused, recycled, or composted; and circulate everything we use to keep it in the economy and out of the environment. Transmaterilization, a shift to bio-based raw materials, on the other hand, have attracted much attention due to the public concerns over limited fossil fuels and climate change recently [3, 4]. Global bioplastics production capacity is set to increase from around 2.1 million tonnes in 2020 to 2.8 million tonnes in 2025 [5].

Denim industry has started to vastly use recycled polymeric materials in the last 5 years. Recycled PET and recycled elastane as an example of synthetic polmers and recycled modified cellulose as a bio-based polymer are the main materials in use. Many novel bio-based plastics, such as PLA and stratch plastics are still at their early stage in use for denim fabrics.

A research gap exists on the environmental impact of denim fabrics from recycled polymeric fibers using the LCA methodology in the context of Turkey. For this reason, this chapter was conducted such that it firstly addresses sources of polymeric recycled materials as well as their chemical and biological recycling process. Finally, Life Cycle Assessment (LCA) methodology is adopted to quantify and compare the environmental impact of denim fabrics from recycled synthetic/man-made fibers.

2. Sources of polymeric recycled materials

Being part of the European Green Deal, the new Circular Economy Action Plan points out textiles and plastics as two of the key value chains that will be addressed as a matter of priority [6]. The textiles' system is characterized by significant greenhouse gas emissions and a high use of resources: water, land and a variety of chemicals [7, 8]. Textile industry is the third consumer of plastics after Packaging and Building & Construction Industries, representing a quarter of the world carbon dioxide budget [9–11]. Every year, 80 billion new garments are produced for fast-growing fashion industry [12], and apparel business utilizes over 97% natural and synthetic based (principally plastic) virgin materials, only 12% of which is recycled into another item after disposal [13–15]. Moreover, apparel industry represents up to 2% of worldwide oil request, and in this manner a portion of the 300 million tons of plastic is created consistently. The production of fibers, their finishing processes and the chemicals for the required functional properties add up to about 20% of worldwide modern water contamination credited to the sector [13–15]. There is no need to say that effective recycling of such wastes will bring ample environmental and economic benefits such that about 7.5 million cubic yards of landfill space, 17 million tons of CO2, and 4.2 trillion gallons of water can, for example, be saved [12, 16, 17]. In addition to these, it should also be noted that the world population of 7.6 billion people is anticipated to reach 8.6 billion in 2030, 9.7 billion in 2050 and almost 11 billion in 2100, thus resulting in a further increased consumption of textiles and apparel, and in turn that of synthetic (polymeric) fibers [18].

The cause of the plastic pollution is mostly due to the fact that plastic, being generally landfilled or incinerated, does not decompose in the environment, continually accumulating in the waterways, agriculture soils, rivers and oceans, massively contributing to global warming [10, 11], causing damages to biodiversity and ecosystem services, and leading to social and economic drawbacks [19, 20]. Plastics have tended to replace traditional materials such as wood, glass, etc. because of their lower weight, flexibility, and simple processing. They can be made from a single polymer, or multiple layers of different polymers, or other materials. Generally, plastics can be classified into petrochemical-based or bio-based, depending on the material from which they are made. Petrochemical-based plastics can be separated into thermoplastics and thermosets.

Thermoset plastics are permanently cross-linked together and therefore difficult to be reformed whereas thermoplastics can be remelted and reformed, and thus are the most commonly used plastics in the economy [21]. Bio-based plastics are, however, derived from biomass (e.g., starch, sugar, and vegetable oils) excluding materials from geological formations or fossilized, as defined by the European Standard EN 16575 [22–24]. Bio-based plastics can be classified into two groups, namely polymers made entirely from biomass and polymers made partly from biomass [23]. Furthermore, all plastics, regardless of whether they are petrochemical or bio-based, can be designed to behave in two distinct ways: biodegradable and non-biodegradable. Biodegradable plastics can be decomposed in the environment by the activity of microorganisms (bacteria or fungi) into water, carbon dioxide (CO2), methane (CH4), and biomass (e.g., growth of the microbial population), though their biodegradability can vary based on the plastic's inherent and designed properties [25, 26], climatic and process specific conditions, and degradation speed [21, 27, 28]. Biodegradable plastics may also be compostable, which are capable of undergoing biological decomposition in a compost site as part of an available program, and the resultant breakdown fragments are completely used by the microorganisms under the certain conditions certified by the international standards ISO 17088, EN 13432 (Europe), ASTM D400, and D6868 (United States) [28, 29]. It should, though, be noted that not all biodegradable plastics are compostable, but all compostable plastics are biodegradable [30].

A slower development within the field of recycling of plastics in terms of the methods employed, created added value, properties of recycled polymers, etc., causes some problems regarding the inclusion of such polymeric materials in the economic cycle [30, 31]. Among typical examples of waste stream products, are there short-life packaging materials (bags, bottles, etc.), used goods (computers, cell phones, etc.), demolition materials from buildings, and disposables. About 1 million plastic bottles, for example, are wasted every minute and are estimated to double in the next 20 years [32].

With about 13% of the market share, textiles are an important source of man-made polymers. Nearly 63% of the textile fibers are made from petrochemical materials such as nylon, acrylic, polyester, and polypropylene, and these fibers' production and fate give rise to significant carbon dioxide (CO2) emissions [33]. From the perspective of plastics, the scope of this section is, therefore, limited to textiles made of synthetic fibers (**Figure 1**). Polyester is one of the most popular fiber in the textile industry [34], which is followed by polyamide (nylon).

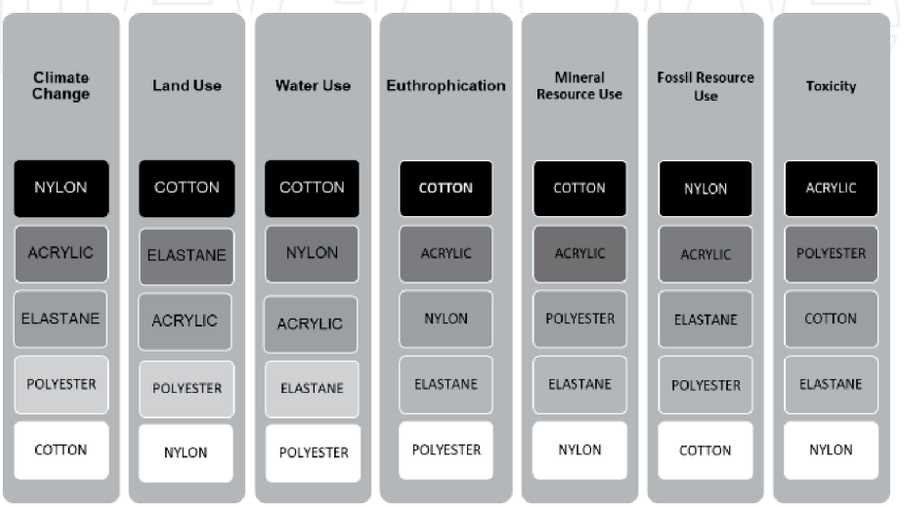

Figure 1.
Comparison of the environmental impacts of the manufacturing of 1 kilogram of dyed, woven fabric (black = worst, white = best) (adapted from [35]).

Polyester alone had a market share of around 52% of total global fiber production, and approximately 58 million mt of polyester was produced in 2019. Polyamide, on the other hand, accounted for 5.6 million mt and approximately 5 percent of the global fiber market in 2019 [36]. In the last years, recycled PET (rPET) production has enhanced dramatically, but only 30% of PET bottles were recycled [37, 38]. In 2019, the estimated rPET share of polyester staple fiber was as high as around 30 percent whereas that of polyester filament was at around 6 to 7% [36]. Recycled PET fibers have the potential to replace virgin PET (v-PET) fibers, and these fibers can be blended with other polymers to create the required properties for each relevant application. But more research appears to be needed to uncover the further potential of rPET fiber based applications [31, 39–42].

Polyamides (PAs), also known as nylons, are other polymeric materials which are widely used in many engineering applications including textile fibers. This is due to their excellent mechanical properties, chemical resistance, wear resistance, dimensional stability, low friction, etc. and ease of processing. Unfortunately, these useful properties are also the ones causing significant environmental consequences. In fact, nylon accounts for about 10% of the debris, mostly in the form of fishing nets in oceans. Contamination is also another issue as far as nylon is concerned. This is mainly because of the fact that nylon is melted at lower temperatures, meaning some contaminants, i.e. non-recyclable materials and microbes or bacteria, can survive. Therefore, all nylon waste must be cleaned thoroughly before a recycling process [41].

As a final note, there is a number of textile companies that have successfully applied the various recycling technologies to produce commercially available raw polymeric materials as is presented in **Table 1** [43, 44]. Sportswear brands are in particular increasingly using recycled synthetic fibers. Most use rPET made from PET bottles [45, 46], but some brands work with recovered ocean plastics, recycled nylon made from discarded fishing nets, and/or with recycled elastane. Also, there are several brands looking for plant-based fibers such as lyocell, Tencel to replace polyester [47, 48].

2.1 Sources of biologically recycled materials

Due to the public interest in the environment, climate change and the limited resources of fossil fuel, bio-based plastics which could be divided into three principal groups, have been investigated for some time [49]:

Manufacturer	Product	Material	Source
Unifi	Repreve®	Recycled polyester	Plastic bottles, industrial waste, fabric scraps
Tenjin	Ecocircle®	Recycled polyester	Consumer and industrial waste
Aquafil	Econyl®	Recycled nylon	Post-consumer waste, fishing nets, old carpets, etc.
Speedo	Powerflex Eco®	Recycled nylon	Fabric scraps, offcuts, fishing nets, old carpets
Jeplan	BRING Material™	Recycled polyester	post- and pre-consumer textiles
Polygenta	perPETual	Recycled polyester	recycled plastic bottles and pre-consumer textiles

Table 1.
Samples of recycled polymer based commercial products [43, 36].

1. bio-plastics that are biodegradable, like starch plastic, cellulose polymers, proteins, lignin and chitosan plastics, polylactic acid (PLA), polyhydroxy alkanoates (PHAs), but also polyhydroxybutyrates (PHBs), polyhydroxyvalerate (PHV) and their copolymers in different percentages (PHBV). This class includes polymers such as PVC, PE, PP, PET, nylon and polyamides (PA), named as bio-plastics because the starting monomers could be obtained from biological resources;

2. bio-plastics based on petroleum resources, which are 100% biodegradable, like polycaprolactone (PCL), polybutylene succinate (PBS), polybutylene adipate (PBA) and its copolymers with synthetic polyesters like polybutylene adipate-terephthalate (PBAT) and polyvinyl alcohol (PVOH);

3. bio-plastics obtained by using monomers from both biological and petroleum resources like polyesters obtained with petroleum-derived terephthalic acid and biologically derived ethanol, 1,4-butanediol and 1,3-propanediol, such as polybutylene terephthalate (PBT), polytrimethylene terephthalate (PTT), polyethylene-co-isorbite terephthalate (PEIT), polyurethane (PUR) and epoxy resins (thermoset plastic).

The actual development stage of emerging bio-based polymers is given in **Figure 2**. While the main application of bio-based plastics is in packaging, around 11% of global production (240.000 tones), are used in textiles, mainly polytrimethylene terephthalate (PTT) and PLA [20, 35, 50, 51].

Bio-based synthetic fibers are often mentioned as environmentally friendly alternatives to traditional, virgin fossil-based ones. Mostly bio-based synthetic fibers, such as bio-PET, are developed to have the same properties and therefore chemical composition as their fossil-based counterparts [35]. It is important to note

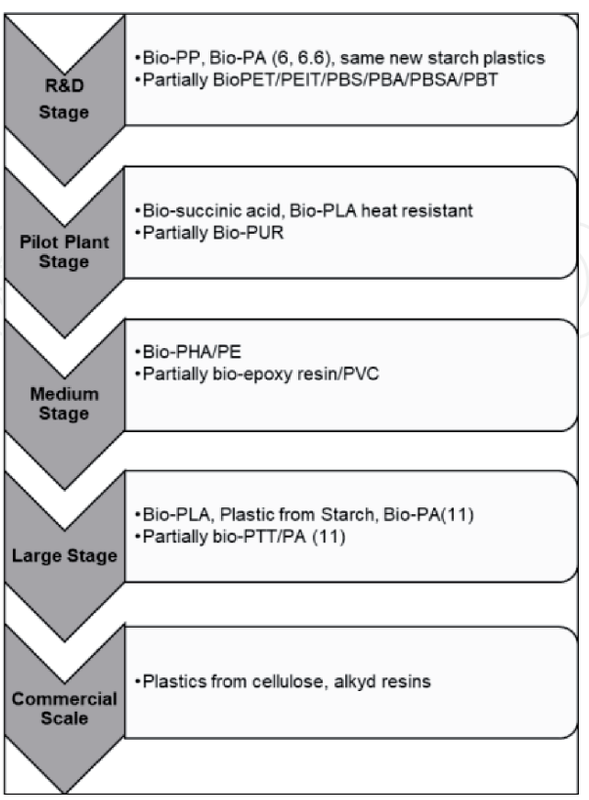

Figure 2.
Development stage of emerging bio-based polymers (adapted from [49]).

that bio-based origin does not imply that the fibers are bio-degradable [13–15]. The key to bio-based synthetics lies in bio-based feedstocks, and the production of these feedstocks has some sustainability issues such as the use of land. This is due to the fact that the cultivation of biomass for bioplastic production can compete with food production for arable lands [35, 52].

As is well known, there are differences in terms of environmental impact between crop based-and waste-based feedstocks. Crop-based corresponds generally to commodity crops such as corn or sugar cane whereas biomass or waste-based raw materials employ agricultural residues and organic waste. There are commercially available bio-based fibers and yarns and some of them are given in **Table 2** [36]. Obviously, feedstocks from waste will most likely be more environmentally preferred and more efficient, as they generally do not require new production of crops and they reutilize residues that would otherwise end up discarded [52]. Additionally, although there is growing interest in textile and apparel industry for bio-based fibers, some barriers such as production cost and low process efficiencies, hinder their further commercialization. Also, some new bio-based fibers, having different structures and properties than conventional ones, cannot yet be handled in current textile manufacturing processes.

Addressing climate change is one of the most urgent action areas for the textile industry. If apparel industry continues on its current path, by 2050, it could use

Bio-based polyester
Far Eastern: TopGreen®, bPET filament made with 30% bio-based feedstock from sugarcane.
Far Eastern: bio-based PTT and bio-based PLA made with NatureWorks Ingeo™, which is made from corn.
INVISTA: LYCRA® T400® EcoMade fiber. More than 65% of the overall fiber content comes from a combination of chemically recycled plastics (PET bottles) and renewable plant-based resources (corn).
Palmetto Synthetics: bio-based PLA staple fiber made with NatureWorks Ingeo®, which is derived from corn.
Radici: CornLeaf filament yarn based on Ingeo™ PLA biopolymer, which is made from corn. And a 30% bio-based polyester filament yarn produced from bio-PET resins made with plant-based bio-MEG.
Toray: Ecodear® PET, a 30% plant-based polyester fiber derived from sugarcane and Ecodear® PTT, a 30% plant-based and a 100% bio-based PLA filament.
Trevira: bio-based PLA fibers and filaments made with Nature Works LLC Ingeo™ which is made from grain (corn).
Bio-based polyamide
Cathay: TERRYL®, a bio-based polyamide line offering PA56, PA510, PA512, PA514 and co-polymers chips and filament with 31 to 100% renewable shares.
Fulgar: EVO®, a 100% bio-based polyamide yarn made from castor oil.
RadiciGroup: Biofeel®, a 64 to 100% bio-based polyamide filament yarn derived from castor oil and agricultural waste and Dorix® 6.10 is 64% bio-based polyamide staple fiber/polyamide yarn
Toray: ECODEAR® PA 6.10, a bio-based polyamide filament derived from the castor bean.
Recycled/bio-based elastane
Asahi Kasei: Roica™ EF, The Global Recycled Standard (GRS) certified recycled elastane, polyurethane filament
Invista: LYCRA® EcoMade fiber, elastane made with recycled content (20% pre-consumer content, diverting waste, and keeping materials in use) and LYCRA® 162 R fiber, an elastane fiber with 70% bio-based content derived from corn.

Table 2.
Bio-based fibers and yarns (adapted from [36]).

more than 26% of the carbon budget associated with a 2 °C pathway. Therefore, in addition to academic institutions several companies have been exploring innovative approaches to recycle carbon and directly use it as feedstock for textiles. Covestro is working with university partners and various textile manufacturers to develop the production process on an industrial scale and aim to make the innovative fibers ready for the market. The company announced in 2019 that they have succeeded in making elastic textile fibers based on CO2 and so partly replacing crude oil as a raw material [53]. Fairbrics, being another example, has developed a novel process to create the components of polyester from waste CO_2, and with Airwear, became a Global Change Award winner in 2020 [36, 54]. The significant amount of textile waste generated is also a potential feedstock for bio-based products [55]. A detailed discussion for this very resource and its conversion to feedstock is given in Section 4.

3. Chemical recycling methods

Heaps of polymer wastes are generated due to the extensive use of polymers in many applications and generally life time of the product is exceeded by that of the polymers that the product is made of. The recycling strategies allow production of new polymeric materials from waste. There are existing and emerging approaches for recycling polymer waste which may contain thermoplastics or crosslinked polymers. The categories of recycling technologies that include primary, secondary (mechanical), tertiary (chemical), and quaternary approaches may be summarized as follows [17, 31, 44, 56–66]:

Primary recycling, also termed as closed-loop recycling, refers to reprocessing of industrial byproducts and pre-consumer scrap materials to give a product that will be used for the same purpose as the original one without loss of properties. Primary textile waste may be single or complex polymers that are usually easy to recycle.

Secondary recycling involves mechanical applications such as grinding, melting, and reforming, for processing post-consumer products into new ones with different physical/chemical properties. In contrast to primary recycling, extraction/dissolution and purification of materials is needed for secondary recycling since materials with unknown composition and purity are treated.

Tertiary recycling approach covers chemical processes such as pyrolysis and hydrolysis, in which chemical structure of waste is broken and converted to basic chemical constituents, monomers, or fuels. It is also known as feedstock recycling.

Quaternary recycling is waste-to-energy conversion process where the energy of fuel value of waste is recovered via incineration or pyrolysis.

Polymers, either orinating from natural or synthetic sources, can be recycled commercially by using one of the recycling processes aforementioned. **Figure 3** presents the polymer recycling approaches.

Other than the recycling approaches presented, biodegradable recycling is an emerging approach especially performed for natural based textile materials. Special microorganisms, enzymes, diverse bacteria and fungi are utilized for degradation of biological polymers (cellulose, chitin, wood, hemp) and organic compounds (PET, polylactic acid-PLA, 1,4butanediol, etc.) [59].

To recycle polymeric waste, chemical recycling technologies offer complementary solutions to mechanical recycling. Chemical recycling, in other words feedstock recycling, breaks down the synthetic fibers for repolymerization and yields the monomers of the polymers (or partially depolymerises to oligomers) by performing processes such as hydrolysis, pyrolysis, gasification, condensation, glycolysis, hydrocracking, dissolution etc [57, 59].

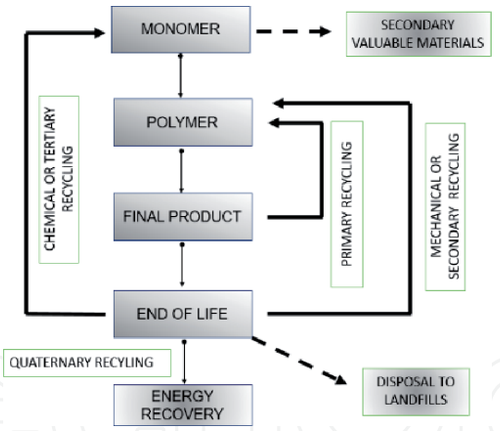

Figure 3.
Polymer recycling approaches (adapted from [56]).

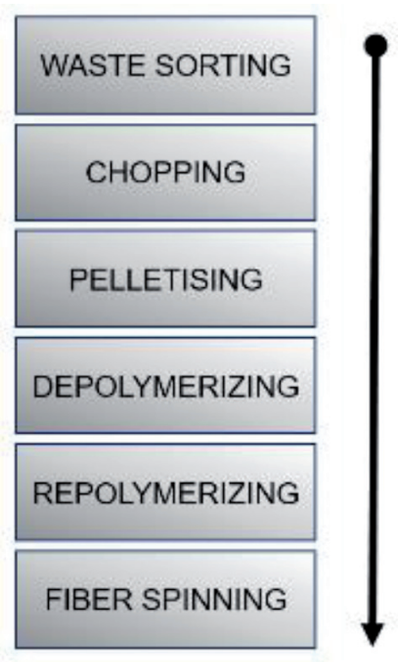

Figure 4.
Simplified process diagram of chemical recycling (adapted from [60, 67].

For chemical recycling the synthetic fibers are chopped, pelletized, depolymerised by chemicals and repolymerised for fiber formation. An overview of the process is shown in **Figure 4**.

Before chemical recycling, the feedstock has to be carefully sorted and all non-textile elements and contaminants (buttons, zips, etc.) need to be removed. Missing or washed-out care labels in garments make manual sorting inaccurate and automated near-infrared identification techniques are required for successful sorting. After sorting, the feedstock is shredded into small fragments to facilitate the dissolving process. The recovery of synthetic fibers involves depolymerisation followed by the production of polymer pellets. A cellulosic pulp is obtained from the cotton. Solvents used are typically recovered to minimize waste and reduce processing costs [35].

Pyrolysis, sometimes referred to as thermolysis, is a thermo-chemical process that polymers are subjected to various temperatures and pressure levels in the presence of catalysts or reactive gases and, decomposed. Pyrolysis processes are economically viable for polyamide 6 polymers. For depolymerisation of polyester, frequently studied approaches are glycolysis, hydrolysis and metanolysis that use glycol, water and methanol, respectively [57].

Solvolysis is depolymerization of cross-linked polymers by using solvents to break covalent bonds in the tridimensional network. The process can be conducted in a wide range of temperature and pressure by employing different solvents. The need for high temperature, pressure and harsh chemicals limit the industrial adoption of solvolysis [17].

Chemical processes allow successful fiber-to-fiber recycling since undesired non polimeric constituents such as colorants, catalysts, surface treatments, backing materials, and other auxiliary chemicals used in textile production are removed. On the other hand, contamination rates as high as 20–30% by weight is economically feasible for chemical recycling. As a result of degradation or contamination of the physical quality during mechanical recycling, chemically recycled polymers offer better inherent quality properties. Another benefit for chemical recycling is that form of the polymer -bottle, jacket, industrial scrap, automobile component, etc.-to be recycled does not matter [58].

Despite its advantages, there are also limitations for large scale applications of chemical recycling processes due to the requirement of considerable amounts of energy inputs for the present methods and due to many uncertainties about their environmental impacts. Even though processes for chemical recycling are technically viable, the ecological and economical impacts need to be questioned. Technologies for chemically recycling of polyester and some other polymers have been existing for some time however, building and operating costs of chemical recycling facilities are higher than those of mechanical recycling facilities. Presence of additives and chemicals used during polymerization might also complicate the processing and affect the purity and quality of the monomers obtained after chemical recycling [35, 58, 59, 65].

Existing chemical processes for recycling polyester are expected to increase and new chemical recycling approaches for polyester in laboratory scale are expected to be developed until 2030. Chemical recycling of cotton and other cellulose based fibers are expected to be developed in full scale by 2030 [68].

3.1 Chemical recycling approaches for commonly consumed polymers

3.1.1 Chemical recycling of polyester (PET)

Commercially available chemical recycling options for polyester waste are [31, 56, 66, 69]:

- Glycolysis in which Bis(2-Hydroxyethyl) terephthalate (BHET) and oligomers are derived using glycol. Virgin PET cannot be produced by glycolysis since waste is partially depolymerized to BHET, and colorants or dyes are not removed. It is a simple, economical and flexible process that can be applied to conventional PET production plants.

- Methanolysis in which polymer is depolymerized to dimethyl terephthalate (DMT) and ethylene glycol (EG) by using methanol. DMT is then purified by crystallization and distillation. PET feedstocks with lower quality are acceptable in the methanolysis process as compared with the glycolysis because purification of DMT is easier compared to BHET obtained by glycolysis.

- Hydrolysis in which polymer is depolymerized to Terephthalic acid (TPA) and ethylene glycol (EG) by using water. High-pressure (1.4–2 MPa), high-temperature (200–250°C) and a long depolymerization time are needed for the process. The hydrolysis of PET can be performed as acid, alkaline, and neutral hydrolysis.

Condition of the input material is important for chemical recycling of PET. When the input material is dyed, for example, decolorization and the removal of dyes require an additional step in the process. Plastic bottles from PET with high molecular weights can be successfully recycled by chemical processes. Even though recycling reduces the chain lengths, recycled materials have an average molecular weight high enough for fiber production [64].

3.1.2 Chemical recycling of nylon

The most widely used nylons are Nylon 6,6 (derived by the polymerization of adipic acid and hexamethylenediamine) and Nylon 6 (obtained from caprolactam). Chemical recycling of nylon includes a depolymerization process followed by distillation to obtain and recover their monomeric constituents: caprolactam (for Nylon-6), and HMDA and adipic acid (for Nylon-6,6). Nylon can be efficiently depolymerized to monomer using chemical and thermal approaches [56, 61, 64, 66].

While chemical recycling, the molecular structure of the polymer is broken down using chemical reactions. After the reaction the products obtained can be purified and used to produce either the same or a related polymer.

During thermal recycling, breaking down of the polymer chemical structure relies on a reaction triggered by heat.

Various chemical processes demonstrated and developed for recycling Nylon 6 and Nylon 6,6 are:

- Hydrolysis of Nylon 6, patented by AlliedSignal, Inc., in which the scrap is dissolved using high pressure steam (963–997 kPa) at 175–180°C for 0.5 hour in a batch process. Then it is continuously hydrolyzed using super-heated steam (350°C) and 790 kPa to form caprolactam. This process needs no additional purification for the recovered monomer.

- Hydrolysis of Nylon 6,6, in which polymer is depolymerized to adipic acid and hexamethylene diamine (HMDA) by the hydrolysis of the polymer in concentrated sulfuric acid. The adipic acid is purified by recrystallization and the HMDA is recovered by distillation after neutralizing the acid.

- Ammonolysis is a process preferred by the DuPont Company for the depolymerization of nylon 6,6 carpet waste. The reaction of nylon 6,6 and nylon 6,6/nylon 6 mixtures with ammonia under 300 and 350°C temperature and 68 atmospheres in the presence of an ammonium phosphate catalyst to gives a mixture of monomers from both nylon 6 and nylon 6,6 polymers.

High costs, challenging materials issues, multiple processing steps requiring high operational knowledge are among the barriers to widespread adoption of chemical recyling methods for nylons.

3.1.3 Chemical recycling of cotton

For fiber to fiber recycling in textiles, the chemical recycling of synthetic materials is not new but studies on natural fiber chemical recycling is relatively new. Although energy products, bio-based products, bacterial cellulose, glucose, aerogels etc. are among the potential applications of chemically recycled cotton, chemical recycling of cotton waste is widely used to produce new fibers that can be used in a variety of products.

The two main routes for chemical recycling of cotton is based on the dissolution of cellulose [61, 70, 71]. Accordingly, either glucose monomers are depolymerized for use in other applications or a polymer dissolution route is followed where cellulosic fibers separate and regenerate by use of solvents. Via the latter process, chemically modified or pure cellulosic fiber products, which can be used as feedstock for regenerated man-made cellulosic fibers (MMCF), may be recovered. A simplified process diagram for chemical recycling of cotton is given in **Figure 5**.

In the Lyocell method, cotton fibers are dissolved (dissoving pulp) using N-methylmorpholine N-oxide (NNMO). The regenerated MMCF are produced by processing the dissolved pulp and and blending with other plant-derived pulp products such as wood, flax, hemp, etc.

Ionic Liquids (ILs) are organic salts in liquid state with low melting point, have chemical and thermal stability, and are non-flammable. They are less aggressive to the environment and considered to substitute organic solvents. The ionic liquid process, yet not commercially available, can be applied to all cotton and also to blends of polyester-cotton.

Another checical recycling approach for cotton is the Ioncell-F cellulose spinning process which uses ionic liquid DBNHOAc without co-solvents or stabilizers [62, 71].

3.1.4 Chemical recycling of blends

The shortcoming of the depolymerization of waste from blended fibers depend on the differences in the fiber regeneration processes. There are ongoing efforts for successfully chemical recycling of blended materials.

In the products from cotton and polyester, chemical recycling has proven to be successful as selective degradation method is used by which the fibers can be artificially isolated and transformed into new ones. When a process using n-methylmorpholine-N-Oxide is applied, cellulose is dissolved. The dissolved cellulose and polyester are then separated by filtration and the captured polyester is respun into a fiber, filament, or yarn while the broken down cellulose can be utilized for the formation of MMCF [44, 60].

Financially viability of chemical fiber-to-fiber recycling of polycotton blends depends on the price and availability of sufficient volumes of well- textile waste and the market value of the resulting cellulosic pulp and polyester pellets [35].

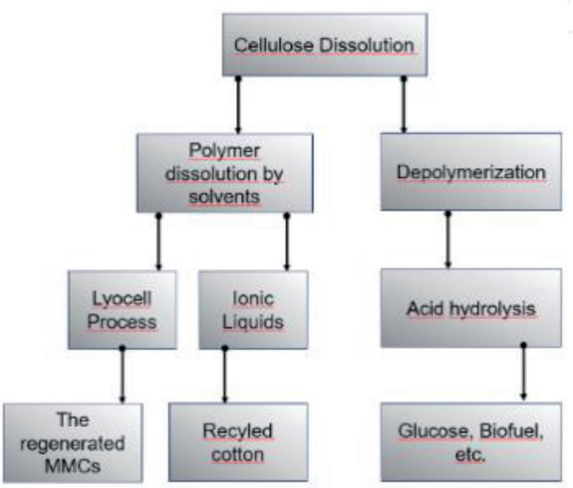

Figure 5.
A simplified process diagram for chemical recycling of cotton (adapted from [61]).

The Japanese Teijin process, for example, is able to handle mixes of 80 percent polyester and 20 percent cotton. However, the process is very sensitive, and only uses Teijin products as input [68].

Nylon and spandex are also two fibers that are commonly used together especially in sportswear and usually, the percentage of nylon is considerably higher than that of spandex. Spandex can be expelled from the blend by dissolving it in solvents, for example, N,N-dimethyl formamide. However, this is a costly process with adverse environmental issues. There has been another successful approach for removing spandex by first treating the blend with heat to degrade the spandex and then washing by ethanol. At the end of the process spandex is successfully evacuated leaving just the nylon [44, 60].

4. Biological recycling methods

Polymers can be degraded by a synergetic combination of different degradation mechanisms present in nature. Enzymes or by products produced by microorganisms such as bacteria, yeasts, fungi, enzyemes, are responsible for the occurance of microbiological degradation. Also, mechanical, chemical or enzymic aging of polymers can be caused by macro-organisms that eat and, sometimes, digest polymers. The main steps of biological degradation are depolymerisation, down to oligomeric or monomeric fragments, and mineralization. Enzymes are the biological catalysts, which can cause huge increases in reaction rates in an environment that is not favorable for chemical reactions [72].

Biological recycling, considered as an improved form of chemical recycling by some researchers, is an emerging approach and expected to be relatively cleaner than conventional textile recycling approaches [31, 73]. During biological recycling, certain polymers can be converted into compost or other substances under specific process parameters (pressure, pH, etc) within the presence of microorganisms [74]. There are several bio-recycling methods developed to recover cotton and PET fiber from textile waste. Research on pretreatment and hydrolysis of cellulose (biodegradable part in textiles) has been carried out to convert it into fermentable glucose [75]. However, polyester in the cotton-based textiles restricts bioconversion of textile waste, as it obstructs the enzymes from reaching the cotton during hydrolysis. Hence, current researches mainly focus on optimizing the conditions for pretreatment and enzymatic hydrolysis, for effective and efficient bioconversion of cotton–polyester blends [76]. According to one such biological recycling method PET fibers could be recovered from polyester-cotton blended wastes by hydrolyzing cotton via enzymatic methods to obtain glucose. The remaining non-biodegradable component-polyester can be re-spun into fibers. The carbon components of most man-made polymers cannot be broken down by the enzymes of microorganisms and that is why such polymers are resistant to biological degradation. Despite this, a commercial approach has been succesfully developed by CARBIOS for enzymatic recycling of PET in various plastics or textiles [58, 77]. Accordingly, the process biologically recycles PET by using an enzyme capable of specifically depolymerizing the polymer to its monomers.

A few organizations, in collaboration with several clothing brands, are also developing sustainable solutions to effectively manage textile waste for obtaining bio-based raw materials. The Hong Kong Research Institute of Textiles and Apparel (HKRITA), for instance, has joined hand with the Hong Kong Polytechnic University to develop new bio-based textiles that combine the properties of Polyactic (PLA) and polyhydroxybutyrate-co-hydroxyvalerate (PHBV) to function as a green alternative to non-biodegradable synthetic polymers in the market. PLA is derived from renewable sources such as cassava roots, corn, and sugar cane,

whereas PHBV is naturally produced by fermentation process of bacteria [78, 79]. Worn Again Technologies has, however, focused on converting polyester and polycotton blended textiles, and PET plastic back into circular raw materials using their special recycling technology which is stated to be able to separate, decontaminate and extract polyester and cellulose from textile waste, polyester bottles and packaging to produce dual PET and cellulose outputs [80].

Clearly, textile waste with high cellulose content, mainly from cotton, can also be used as an alternative feedstock for man-made cellulosic fibers' (MMCF) production by recycling of post-production, pre-consumer (e.g., samples or stock that cannot be sold) or post-consumer textiles. Promising natural fibers with high α-cellulose content and low hemi-cellulose content are also found in the fibers of banana, pineapple, and abaca leaves. Man-made cellulosic fibers (MMCF) producers are, therefore, intensifying their research and development (R&D) activities to focus on alternative feedstock for cellulose production [81]. Regarding that, Circulose® is one of the few commercially available products. It is "dissolving pulp" from 100% textile waste, such as worn-out jeans and production scraps, used to manufacture viscose, lyocell, modal, acetate other types of regenerated fibers [82]. SaXcell, an abbreviation of Saxion cellulose, is another regenerated virgin textile fiber made from chemical recycled domestic cotton waste. The end product is SaXcell, a regenerated virgin cellulose fiber that can be spun into yarns and turned into fabrics [83]. As may be seen from these few examples, the use of alternative fibers could become an interesting option for the production of man-made cellulosic fibers (MMCF), but they still face some economic, technological and social barriers to scalability.

5. Usage of recycled polymeric materials in denim

Denim itself is very well-known for its durability and long-lasting properties. Although there is no certain lifetime for a jean, the common belief is that it lasts longer than any other garment. This belief relies on the fact that Jeans used to be the uniform of the miners during Gold Rush in the United States.

The fabric parameter that indicates the durability is the strength value of the fabric. Strength value is derived from both yarn strength and the construction. Since the construction remains the same for denim fabric, yarn strength should be analyzed to better understand the contribution of recycled fiber to the strength of the yarn. Fiber fineness (dtex), staple length (mm) and tenacity of the fiber (cN/Tex) are the most important parameters that contributes to the yarn strength. Fiber fineness directly determines the number of fibers in the cross-section of the yarn. The finer the fiber, the number of fibers contributing to the strength will increase and the yarn strength will be higher. Staple length refers to the average length of individual fibers. The shorter the staple length, the more difficult it can be to spin. More twist is needed if the staple length is short. Therefore, this parameter plays an important role on spinnability and strength of the yarn. Tenacity of the fiber directly relates to the yarn strength. The stronger the fiber, the stronger the yarn will be.

Mechanical recycling of natural fibers, mainly cotton, shortens the fiber length drastically. Therefore, a perfect mix of pre & post consumer recycled materials and alternative natural materials is essential. Chemical recycling overcomes this challenge since the required fiber length can be set in the process and the tenacity of recycled fibers are almost the same with the virgin versions in most of the cases. Hence, this ability creates a longer lifetime for a recycled denim with recycled polymeric materials. As seen in **Table 3**, PET and Recycled PET have the highest tenacity values in comparison to the Cotton, Tencel, Recycled Tencel and PLA. This means that using PET in denim increases its durability hence lifetime. On the

Physical Properties	Unit	Cotton*	Recycled Cotton*	Lenzing Tencel	Lenzing Tencel Refibra	Pla-Trevira	Recycled Pet-Unifi Repreve	Regular Pet
Fiber Fineness	dtex	1,77	1,65	1,3	1,7	1,7	1,56	1,39
Staple Length	mm	29	24	38	38	38	38	38
Tenacity	cN/Tex	30	< 30	36	33	33	56	57,5

**Cotton & Recycled Cotton-Company Specific Data.*

Table 3.
A comparison of physical properties of selected fibers [84–87].

other hand, both Tencel/Refibra and PLA have higher tenacity values than Cotton. In theory, using chemically recycled polymeric material and biologically produced PLA in denim production increases the lifetime of the jean.

However, the environmental impact of petrolueum based fibers are fairly high. Synthetic polymers, PET in this study, are produced from fossil fuels. The environmental concerns regarding the use of synthetic polymers bring the main discussion in this study: The environmental impact of recycled polymeric materials and bio-based materials in denim production.

In the previous study of the authors, the effect of using regular, organic and recycled cotton in a denim fabric on environment was analyzed [88]. As a continuum, this chapter analyzes the environmental impact of using polymeric and recycled polymeric fibers in denim fabrics. The past 5 years witnessed the commercialisation of recycled versions of elastane, Tencel®, T400® and many novel bio-based plastics, such as PLA, starch plastics. Today, main recycled polymeric materials that are used in denim production are as follows:

- Recycled Polyester
- Recycled Elastane
- Recycled Tencel- Tencel® x Refibra®
- Recycled Bio-based Polyester- LYCRA® T400® EcoMade fiber

In order to eliminate the effect of finishing processes on the lifecycle assesments of the fabrics, Recycled Elastane and Recycled Bio-based Polyester are excluded in this study. Instead, recycled bio-based polymer, Poly Lactic Acid (PLA) fiber is included. In this study, six fiber compositions for the same denim construction (**Table 4**) are determined to calculate and analyze the effect of different recycled polymeric materials in denim.

Article Code	Composition
Article 01	100% Cotton
Article 02	80% Cotton- 20% PET
Article 03	80% Cotton-20% Recycled PET
Article 04	80% Cotton 20% PLA
Article 05	80% Cotton-20% Tencel
Article 06	80% Cotton- 20% Refibra

Table 4.
Article specification.

Fiber	Source	Global Warming Potential (kg CO^2e/ kg fiber)	Water Use (l/kg fiber)	Land Use (m^2a / kg fiber)	Eutrophication Potential (kg PO_4^{3-} e /kg fiber)	Abiotic Depletion Potential (kg Sb e / kg fiber)
Cotton	Ecoinvent 3 Company specific blend	3,07	1861	7,91	0,013	0,018
PET	Ecoinvent 3	5,26	24,0	0,13	0,012	0,054
Recycled PET	Ecoinvent 3	1,84	1,78	0,07	0,004	0,012
PLA	Confidential Source	3,1	15	0,085	0,0052	0,040
Tencel	[89, 90]	2,32	20	2,4	0,0019	0,007
Refibra	Confidential Source	2,00	18,4	1,68	0,0019	0,007

Table 5.
Summary of raw material data used in this Study.

PET, recycled PET and PLA fibers are supplied from Reliance, Unifi and Trevira, respectively. Tencel & Refibra are lyocell fibers produced by Lenzing AG. More than 99% of wood and dissolving wood pulp used by the Lenzing Group is either certified by FSC® and PEFC™ or inspected in line with these standards. The trees grow quickly without the use of pesticides, fertilizers, irrigation or GMO. Tencel with Refibra Technology contains 30% recycled material as pre-consumer cotton scraps and post-consumer garments from the textile value chain as raw materials and produced with Tencel technology [89].

Table 5 below presents the raw material data set used in the calculation of lifecycle assesment values of the articles listed above.

6. Lifecycle assesment methodology and selected environmental impact categories

The life cycle assessment methodology takes into account all the impacts originated from the inputs and outputs of a system starting from the fiber cultivation/ production till the end of life of the jean, cradle to grave. An experimental study was conducted to represent the environmental impact of using recycled polymeric materials in a denim fabric in comparison to using virgin materials. In doing so, the methodology given in the authors' previous study was employed [88]. All of the calculations were performed from cradle to denim factory gate. Furthermore, the inventory was based on the 2020 denim production figures of a denim company in Turkey. As the assessment tool, SimaPro software, developed by the Pré Sustainability, was used. SimaPro is one of the leading Life cycle assessment (LCA) softwares that has been used for more than 25 years by the industry and academics in more than 80 countries. SimaPro uses two types of data: primary and secondary [91]. Primary data involves the basic specifications of a denim fabric, for example Article X, 150 cm in width and 14.89 oz./yd^2 in weight.

This data is exclusive to the fabric production practices of the factory. Secondary data, however, comes from the database and it includes the impacts originated from producing that much raw material and all other inputs such as chemicals at

each stage. For the secondary data, Ecoinvent database that is embedded into the software and is the most common Life Cycle Inventory (LCI) database worldwide, was utilized [92]. To be able to perform life cycle assessment of a specific good or service, one needs to have inventory data for the complete supply chain. Due to the huge amount of data needed for this purpose, it is practically impossible to collect and organize the data of the complete background system. In that respect, the Ecoinvent database provides this very system fulfilling the data required for the assessment. The Ecoinvent v3 database contains Life Cycle Inventory (LCI) data taken from various sectors such as energy production, transport, building materials, production of chemicals, metal production and fruit and vegetables. The entire database consists of over 10,000 interlinked datasets, each of which describes a life cycle inventory on a process level [93]. SimaPro software provides six libraries that each contain all the processes that are found in the Ecoinvent database, but use different system models and contain either unit or system processes [94]. The three Ecoinvent system models are as follows: "allocations at point of substitution", "cut-off by classification" and "consequential". For life cycle assessment (LCA) of a product, the production of an item (e.g. denim fabric) is simulated, using both consumption/production (primary) data of a factory and the corresponding secondary data from the Ecoinvent database. The next step is to choose the environmental impacts to be calculated. The whole process is given in **Figure 6**.

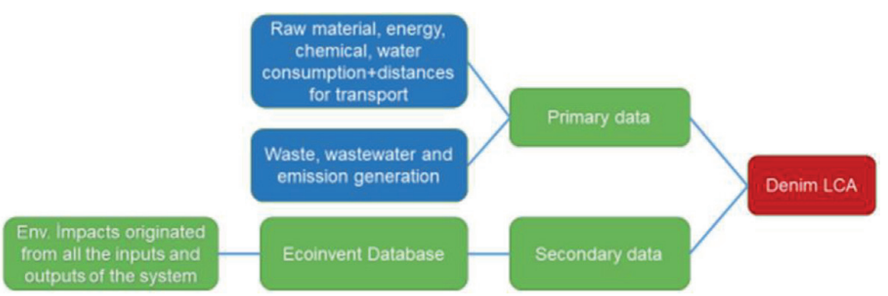

Figure 6.
LCA calculation process.

Indicator	Unit	Description	Example impact	Methodology [92–101]
Global warning potential	kg CO_2 eq (kilogram carbon dioxide equivalent)	Emission of greenhouse gases (GHGs)	Climate change	IPCC 2013 GWP 100a [95]
Freshwater use	lt (liters)	Excessive freshwater taken from the environment	Water scarcity	Life cycle inventory
Land use	m^2 a (meter square per annum)	The amount of agricultural area uccupied	Deforestation	ReCiPe 2016 Midpoint (H) [96–97]
Eutrophication potential (EP)	kg PO_4^{3-} eq (kilogram phosphate equivalent)	Emission of substances to water contributing to oxygen depletion	Nutrient loading to water stream-water pollution	CML 2 baseline 2000 [98, 99]
Abiotic resource depletion	kg Sb eq (kilogram antimony equivalent)	Measure of mineral, metal, and fossil fuel resources used to produce a product	Mineral scarcity	CML 2 baseline 2000 [98, 99]

Table 6.
Selected environmental impact categories.

6.1 Selected impact categories

One of the most important parts of life cycle assessment (LCA) is the outputs, in other words the environmental impacts of the product. With SimaPro software, it is possible to calculate over 100 environmental impact categories. For this study, five impact categories were selected [88]. These impact categories, their definitions and calculation methodologies within the SimaPro software are presented in **Table 6**.

7. A comparison of recycled polymeric materials using LCA methodology

The results of the life cycle assessment (LCA) calculations are presented in **Figure 7**. The process flow in the calculations is as follows: Fiber production, transportation, spinning, warp preparation, dyeing and sizing, weaving, finishing, quality control & packaging. It should also be noted that the main differentiator in the calculation of impact categories is fiber mix.

7.1 Global warming potential

The highest Global Warming Potential was calculated for Article 02–80% cotton & 20% PET. The particularly high result for this article is primarly related to its fossil fuel source and high energy requirement of fiber production stage. Once the polyester is recycled as in Article 03, the global warming potential decreases to the lowest value since depletion of fossil fuels is eliminated. On the other hand, the impact value decreases by 13% when the bio-based polymer, PLA is used (Article 04). The fiber production stage is still an important contributor to the global warming potential value when Article 04 is considered.

Although production of pulp and wood-based fibers is also energy needed to process Tencel® and Refibra® does not only rely on fossil fuels. Lenzing site in Austria uses around 80% renewable energy whereas the pulp plant in Paskov employs 100% renewable fuels. Dissolving wood pulp production in Lenzing's biorefineries is not only self-sufficient in terms of meeting its own energy needs, it actually produces surplus energy. This surplus energy (steam and electricity) is used on-site for purposes such as fiber production or export to the local grid, which in turn lowers Lenzing products carbon footprint [84, 89].

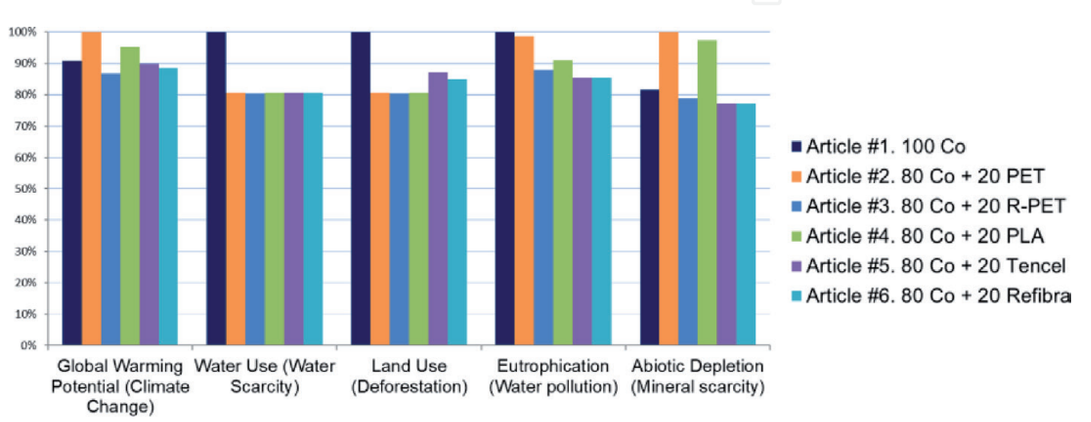

Figure 7.
The results of comparative LCA for different polymeric materials in denim fabric.

7.2 Water use

Water use is dominated by Article 1 with 100% cotton fiber content. This effect originates from the water use in cotton cultivation stage of the overall lifecycle of the denim fabric. Once 20% of cotton is replaced with polymeric materials, the level decreases nearly the same amount, by 20% for the rest of the articles.

Water usage of man-made polymeric fibers has a minor effect in this impact category. Man-made cellulosic fibers, on the other hand, are made from trees such as eucalyptus and beech that can grow with rain water and do not require irrigation which in turns reduces the freshwater use for fiber production hence the fabric [90]. Besides, TENCEL technology; manufactured in closed loop production which recycles water and reuses the solvents at >99% recovery rate. This avoids waste, ensures high resource utilization and results in less water consumption as well as fewer emissions.

7.3 Land use

When the land use for the production of bio-based fibers, cotton, Tencel®, Refibra® and PLA is taken into account, cotton requires a drastically high amount of land compared to the rest of the fibers. PLA land use, here has the lowest value. Tencel and Refibra are slightly higher than PLA. Data shows that cotton requires 65% more land than Tencel and 95% more land than PLA.

The other fibers are syntethised from petroleum, and therefore, Article 02 an 03 have the lowest impact in the impact category under discussion.

7.4 Eutrophication

Eutrophication is the indicator of water emission of substances, mainly nutrients contributing to oxygen depletion and is an indicator of water pollution. The eutrophication potential is mainly originated by the emission of phosphate and nitrate from fertilizers that are used in cotton cultivation.

Although 20% of cotton is replaced by PET in Article 02, the eutrophication level almost stays the same since fiber production requires high energy. Article 04 with PLA and Article 03 with recycled PET score third and fourth with regard to eutrophication impact. Articles with Tencel ® and Refibra® have the lowest impact because of the energy resources that used for their production. Besides, these fibers are compostable and biodegradable in freshwater, marine and soil conditions.

7.5 Abiotic depletion

Abiotic depletion reflects the environmental impacts from using non-renewable energy and material resources. As regular PET fiber is made from crude oil, it has higher abiotic depletion impact compared to other fibers (**Table 4**). This fact increases this impact for Article 2 compared to Article 1–100% cotton version. PLA follows PET for the abiotic depletion impact due to high energy consumption in the lactic acid and lactide production [102]. The differences between manufacturers must be identified and the same calculation should be conducted when renewable energy sources and heat recovery systems are used as in Lenzing® manufacturing system. Therefore, abiotic depletion values for Article 05 and 06 are recorded as the lowest in the study.

8. Discussion

This study mapped and discussed the environmental impact of recycled and bio-based polymeric fibers in a denim fabric. LCA was used as a framework since it is mostly recognized as a tool to quantify the overall impact from a systems perspective.

The data suggested that based on generic data, categorizing fibers as good or bad for the environment is incomplete. The production methodologies used for fibers and their influence on the subsequent product lifecycle are as equally important as fibers' direct environmental impact. A doubled life span of a garment decreases the average garment's climate impact by 52% considering the consumer will buy less garments in a given duration [103, 104]. Therefore, PET, and Recycled PET fibers in this study provided a longer lifetime to a product considering their fiber properties as stated in **Table 3**. On the other hand, fossil fuel and energy use related environmental impact categories such as Global Warming Potential and Abiotic Depletion were calculated as the highest for Article 02 where virgin PET was used. It should be noted that the energy system with which the fibers are produced here may change this data in either positive or negative direction. Once virgin PET is replaced with recycled PET, as in Article 03, these impacts reduce since fossil fuel usage is eliminated. The addition of Recycled PET content reflected the least amount of global warming potential impact. Both PET and Recycled PET presented the best impact for water use and land use categories. Since non renewable resources are already consumed for the virgin material production and post consumer PET remains as a waste for thousands of years in the system if not recycled, one should consider post consumer PET recycled materials (e.g. plastics from oceans) as a solution. On the other hand, the issue with microplastics shedding from synthetic/man-made based textiles which appears to be an important environmental problem, is not discussed in the LCA methodology.

Bio-based man-made polymers, such as Tencel, Refibra and PLA can be stated as the second group to increase the life span of a garment. Especially, Tencel and Refibra scored the lowest in every impact category analyzed in the study, except for the land use. In land use, the value is still lower than cotton. PLA, however, appears to have better values in every environmental impact category, when compared to PET. Although, the study shows that for Global Warming Potential, Eutrophication and Abiotic Depletion impacts, recycled PET is recorded better than PLA, it should be stated here that this result may change with the energy system used for the production of fibers. Besides, PLA is compostable and biodegradable in freshwater, marine and soil conditions.

To conclude, it is unlikely to state that one fiber can be the leader of sustainability alone as there is not one metric that is sufficiently broad enough to incorporate all the fields of sustainability. However, the key point for decreasing footprint of a denim fabric is to use both renewable resources and energy. Therefore, recycled and bio-based materials appear to be an effective solution for a lower environmental footprint in the life of a denim garment. Nevertheless, by bearing the fact that today's waste can be tomorrow's raw material in mind, sources of recycled PET (post or pre consumer) as well as recyclability of garments should be discussed in more detail, which are the topics of further studies.

Conflict of interest

"The authors declare no conflict of interest."

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