# **Textiles for Circular Fashion**

Part 2: From Renewable Carbon to Fibres

PAULIEN HARMSEN, WOUTER POST, HARRIËTTE BOS



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### **Preface**

My search for Circular Fashion started a couple of years ago, from a professional point of view but soon also on personal level, with the contents of my wardrobe. In the spirit of 'practise what you preach' and inspired by many others, I decided to create and wear my own fossil-free wardrobe, made from sustainable materials only. This implies elimination of all fossil-based materials, starting with polyester. From experience I know now that it is quite hard to do this, as fossil-based materials are everywhere, from fabrics to zippers and buttons, so I am not there yet. It's all about feeling, touching, creating, wearing, and evaluating.

It inspired me to write this publication, where chemistry, creativity and fashion comes together. Like part 1 of this series, it is a combination of general information on origin of materials, production methods, lots of (polymer)chemistry and material properties. And again, I tried to simplify the chemistry as much as possible, but sometimes it was inevitable to include some complex parts. To enlighten it a bit, I illustrated boring parts with pictures of self-made garments and electron microscopy pictures of the corresponding fabrics. I was especially delighted with the nice jacket of herringbone fabric (composed of recycled yarns), as it was a complex pattern, and it took me many hours to make.

This booklet is the second piece of the big puzzle called circular fashion. Harriëtte and I (and many other colleagues) are fully committed to making sustainable textiles part of WUR, as it fits very well in our mission "To explore the potential of nature to improve the quality of life". I hope that this publication contributes to a more sustainable production of fashion and more appreciation of your own clothes, as now you have a glimpse of how they are made. Happy reading!

Paulien Harmsen

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### 1 Introduction

### 1.1 Why this study?

Clothing forms one of the elementary needs for humankind. The present production of clothing and other textiles is a world-wide business of significant size. The global fibre production amounts to over 100 million tons per year, and it is becoming clearer every day that the textiles industry has a serious environmental impact. The industry applies hazardous substances and polluting processes. Next to that, it uses significant amounts of non-renewable resources, many of which are based on fossil feedstock, mostly fossil oil. Climate change forces us to phase out the use of fossil feedstock for energy and materials, and it is inevitable that this transition will have a serious effect on the way in which textiles and clothing are produced.

The two questions (1) how and where we are going to source the feedstock, and (2) what we need to change in the production (and use) of clothing and textiles to make them intrinsically circular\*, are the main focus of this study. By describing the present situation and highlighting possible future options and scenarios, we aim to contribute to a substantial and urgently needed change in the production of our clothing.

### Circularity in textiles and fashion

Circularity is a concept that originates from the field of industrial ecology, combined with circular design concepts such as cradle to cradle. The underlying concerns are the ever-increasing depletion of non-renewable feedstock. Circular solutions thus aim at fulfilling societal demand while minimising the input of virgin resources. Circularity concepts therefore go much further than just recycling of materials into new products.\*

When taking a closer look at all the fibres used in clothing and textiles, what stands out is that they all have carbon as their main constituent: C, the sixth element in the periodic system, and one of the most common elements of all known life. But carbon, and more specific carbon dioxide ( $CO_2$ ), also has a negative connotation, as it is related to rising of global temperatures. If we want to limit the global temperature rise to 1,5 °C, we need to reduce  $CO_2$  emissions as soon as possible. Therefore, we urgently need to convert to an economic system that sustainably reduces and compensates the emissions of  $CO_2$ , with the long-term goal to create a  $CO_2$ -neutral economy. This implies that we will have to base our future production on *renewable* carbon rather than on *fossil* carbon.

<sup>\*</sup> See also the previous "Groene Grondstoffen" booklet: Textiles for Circular Fashion; part 1: Fibre Resources and Recycling Options

What exactly does this conversion to renewable carbon mean for the production of clothing and textiles? What options doe we have? We'll dive a bit deeper into this question in paragraph 1.4, but first let us explore the physical structure and composition of textiles in more detail in the next paragraph, and take a closer look at the feedstock presently used to produce textile materials in paragraph 1.3.

### 1.2 Textiles are complex materials

Of all materials we know, textiles fall in the category of the most complex materials, both in terms of the way they are produced and in terms of their final composition and structure. Textile production is a global affair, and the chain of industries, from raw materials to garments, is complex, scattered, and interwoven. A wide range of techniques are applied to turn raw materials into textiles and finally into garments. This is illustrated in more detail by Figure 1, where the grey blocks in the figure each represent a different processing step and therefore often a different type of industry. Before even a yarn is produced (depicted in the centre of the figure), many steps have already been taken to turn a raw material into a fibre that can be spun into a yarn. Textile fibres most often consist of carbon-based polymers, long-chain molecules, that form the basis of many materials and living tissue. The polymers are made by linking small molecules (monomers) together, one by one, through a chemical synthesis reaction (polymerisation).

This distinction between the intermediate structures <u>fibres</u>, <u>polymers</u> and <u>monomers</u> is important, and we refer to it in this booklet as the 'level of assembly'. All textile materials start at monomer level, but polymerisation and fibre formation is either done by men or by nature, depending on the resource being used:

### Fibres

Natural fibres come from <u>biobased resources</u>. If we take a plant fibre, such as cotton, the steps from monomer (glucose) to polymer (cellulose) to fibres are already performed in the plant. In this case, yarn production starts at the fibre level.

### Polymers

Fibres from <u>biobased resources</u> can also be based on natural polymers such as cellulose. Cellulose enters the textile production chain then at the level of polymer, as the monomer to polymer step was done in the plant from which the polymer is extracted. Extrusion spinning into a fibre still needs to be done in the textile production process. In this case, yarn production starts at the polymer level.

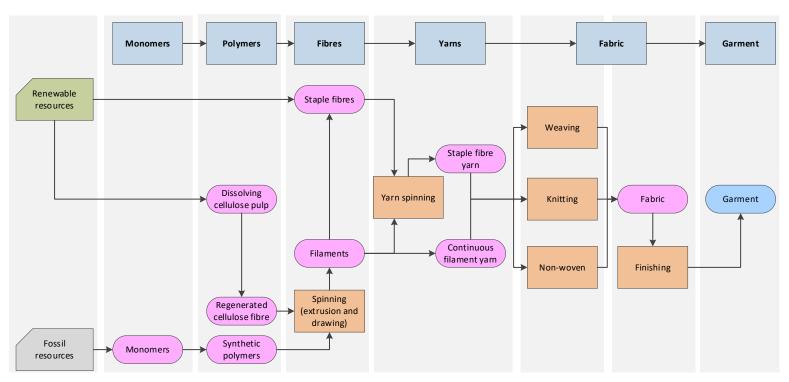


Figure 1: Textile production chain. From resources to monomers, polymers, fibres, yarns, fabric and garments; a simplified scheme.

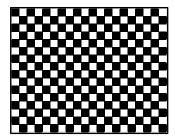
### Monomers

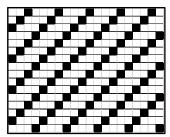
Producing a textile fibre from <u>fossil feedstock</u> always starts with the production of a monomer (with a large variety available), followed by polymerisation into a polymer, and finally extrusion spinning into a fibre. Monomers can also be produced from <u>biobased resources</u>, and after extraction, polymerisation into a polymer and extrusion spinning can be done by man. In these cases, yarn production starts at the monomer level.

This distinction in the starting level of assembly, and thus the steps that need to be taken before a yarn is obtained, is especially important when we discuss the development of alternative feedstock for textiles, as we will see in Chapter 3.

Once a yarn is constructed by yarn spinning of the fibres<sup>†</sup>, conventional processes like weaving, knitting, and felting are essential processes in textile production. Although new processes like 3D-printing are gaining attention, the conventional processes are expected to remain dominant in textile production. Therefore, it is crucial that alternative renewable resources (based on renewable carbon) fit in this existing infrastructure of textile production.

To make the complexity of textiles a bit more tangible, the theoretical background of textile materials in this publication is accompanied by garments made of the corresponding (mostly woven) fabrics. In addition, Scanning Electron Microscopy (SEM) pictures of the fabrics are included. These pictures show more details of the fibre and the fabric structures.





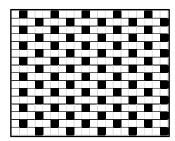


Figure 2: The three basic weave structures. From left to right: plain weave, twill weave and satin weave. The dark squares represent the warp threads, the white squares the weft threads.

<sup>&</sup>lt;sup>†</sup> It is slightly confusing that both the (extrusion) spinning process from a polymer into a fibre and the yarn spinning process of (a bundle of) fibres into a yarn are both generally called spinning.

Fibres come in a range of shapes and properties. For natural fibres this is determined by nature, while man-made fibres can be manufactured in any shape and property required. For example, cotton has a characteristic twist, wool fibres show scales on the surface, viscose has striations along its length, and polyester is often a very smooth fibre. The SEM pictures show the fibres longitudinally (lengthways), and just by looking at the fibre structure and surface, the material can be identified. But also the weave structure is illustrated by the SEM pictures. Weaving is one of the techniques to create a fabric from a yarn. All kinds of different weave structures can be produced by varying the number of warp yarns (yarns that run vertically through the length of the fabric) crossed by weft yarns (yarns that run horizontally). There are three basic weave structures: plain weave, twill weave and satin weave (see Figure 2). When the weave structure of the textile is clearly visible in the SEM-picture, it is mentioned in the captions.

### 1.3 Contemporary textile fibres and their carbon sources

Over the last decades, the textile industry has experienced a transition from using the traditional renewable, biobased feedstocks (plant fibres (cotton, flax, hemp), animal fibres (wool, silk)) towards using fossil-based alternatives (mainly polyester). As a result, the current fibre production is dominated by synthetic fibres from fossil resources. This is illustrated by the Sankey diagram in Figure 3, showing the relation between the origin of presently used resources (at the bottom) and the resulting textile fibres (on top) on a world-wide scale. The thickness of the arrows represents the amount of the various materials, and the world-wide numbers are also given in the graph.

This diagram is based on data from the Market Report 2021 of Textile Exchange (Textile Exchange 2021). This report provides specific data on the global fibre production volume (for 2020 this was 109.000 kton/y), and covers all fibre applications (apparel, home textiles and 'others'). A rough estimation of the fibre volume per application presented in the report indicates that apparel, with more than 80 % of the total fibre volume, forms by far the largest group.

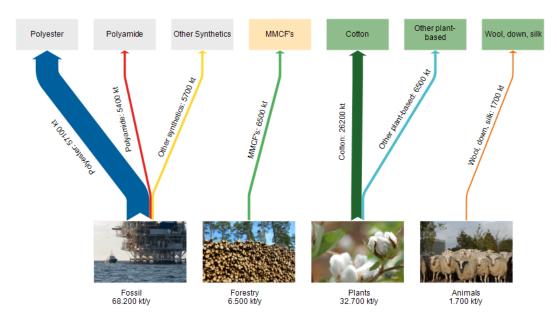


Figure 3: Global fibre production in 2020 (109.000 kton/year) and relation between resources (bottom) and fibre products (top). The thickness of the arrows represents the amount of materials. Data from Textile Exchange (Textile Exchange 2021). MMCFs: man-made cellulosic fibres.

Figure 3 shows that of the total fibre volume, fossil-based synthetic fibres form the largest group (63 wt%), followed by vegetable fibres from fibre crops (30 wt%) and regenerated fibres (man-made cellulose fibres, MMCFs) (6 wt%). Animal fibres only make up 1 wt% of the total fibre mix. Figure 3 clearly illustrates the major task for the textiles industry to source sustainably when fossil resources are phased out by 2050, and all fossil-based polyesters, polyamides and other synthetics will need to be replaced by either i) other renewable fibres or ii) based on another source of renewable carbon. In the next paragraph we will focus especially on these alternative sources of renewable carbon.

### 1.4 Sources of renewable carbon

Fossil carbon-based feedstock (coal, oil and natural gas) forms an important base of our present society. It provides most of the energy that we use nowadays, and forms an important feedstock for products like textiles, packaging materials, tires, and many others we use every day.

Climate change forces us to phase out the use of fossil feedstock. The introduction of renewable energy technologies is taking up speed, and this energy transition focuses mostly on sun, water, and wind as new energy sources. In contrast to fossil feedstock, these new energy sources are not based on (the incineration of) carbon, and the energy transition is therefore often referred to as the decarbonisation of the energy system. However, as we discussed above, many materials are based on carbon, and for these we need to find a renewable form of carbon to replace fossil carbon.

Nova Institute was one of the first institutes that realised and propagated that there are basically only three sources of renewable carbon available for materials (Carus et al. 2020):

- Biomass, which is the source for food, feed and many materials
- Recycled materials, which can be reconverted into new materials
- CO<sub>2</sub>, which can be converted into all kinds of monomers, and subsequently into all kinds of polymers and materials

The relation between the transition towards renewable energy and the transition towards renewable materials is shown in Figure 4. In this scheme the demand for materials, food, and heavy transport fuel (which cannot be easily electrified) are presented as target applications for the renewable carbon transition. Hydrogen is in this vision seen as the most promising option as energy carrier.

Textiles are not mentioned explicitly in this graph, but form part of the categories of plastics (polyesters, polyamides, and other synthetic fibres) and materials (MMCFs, plant-based and animal-based fibres). For the sourcing of feedstock of our future textiles the materials transition thus gives us options for biomass, recycled materials, and CO<sub>2</sub>. In this booklet we present our vision on the potential contribution of all these new routes to produce sustainable future textiles. Important other aspects towards sustainable future circular textiles, such as using garments longer and buying less clothing are not discussed, but nevertheless form an important aspect of the materials transition that lies ahead of us.

# Renewable Carbon Recycling Biomass CO2 Water Wind Wind Solar WAGENINGEN

# **Renewable Carbon for a Fossil Free society**

Figure 4: Relation between the transition pathways towards renewable carbon and renewable energy

### 1.5 Methodology

For textiles, the three basic types of fibre categories include i) natural fibres, ii) regenerated fibres, and iii) synthetic fibres, where regenerated and synthetic fibres are collectively known as man-made fibres. As shown in Figure 3, synthetic fibres currently are mainly made from fossil resources (non-renewable carbon), while regenerated and natural fibres are made from biomass (renewable carbon). In the future, these three basic fibre groups have three renewable carbon sources from which they can be produced: not only biomass, but also recycled materials and CO<sub>2</sub>.

Each source of renewable carbon has its own potential to generate the three basic textile fibres, and this publication presents these options. Our methodology is based on the analysis of five main components shown in Table 1. From left to right it describes the relation between basic inputs required for feedstock production (like sun, water,

energy), carbon sources, intermediate structures (relevant for level of assembly), polymer categories and finally the textile fibre categories. The relation between carbon sources and textile fibre categories is described for contemporary textile fibres in chapter 2, and for future circular textile fibres in chapter 4.

Table 1: Components in our methodology to describe the routes from carbon to textile fibres

Basic input	Carbon source	Intermediate structure	Polymer category	Textile fibre category
Illustrates where the carbon comes from, in what shape or form, and other process requirements like	Shows the origin of the carbon source: fossil, biomass (and what type of biomass), CO <sub>2</sub> , recycled content.	Monomers: Small molecules (building blocks), starting point of a carbon-based material	Natural polymers: Polymers of biological origin formed by living organisms	Natural fibres: Natural fibres of biological origin formed by living organisms
energy (sun, green electricity) and water.		Polymers: Larger molecular structures of large number of monomers linked together	Condensation polymers: Man-made polymers, formed by reaction of two monomers, while eliminating small molecules (water)	Regenerated fibres: Man-made fibres of natural polymers (like cellulose)
		Fibres: Structures of polymers that form a material with specific material properties	Addition polymers: Man-made polymers, formed mainly by one type of monomer; polymerisation by chain reactions	Synthetic fibres: Man-made fibres of condensation polymers or addition polymers

The collection of fibres (so called fibre mix) for contemporary textile fibres is dominated by one type of polyester (polyethylene terephthalate (PET)) and one type of plant-based fibre (cotton). However, for future circular textile fibres this can be expanded with a whole range of other (new) polymers or fibres, as there are many more undiscovered sources to make textiles from. A start for this discussion is given in chapter 3, where for each relevant class of polymers (polysaccharide (cellulose), polyester, polyamide, and polyurethane) the renewable options are described.

### 1.6 Reader's guide

This booklet presents the options we have to produce future circular textiles from renewable carbon sources and the challenges ahead. All the information provided in this publication is based on public sources.

In Chapter 2 the currently used textile fibres are described based on the resources they are made from today (fossil, forestry, plants, animals). This chapter provides the basic information to support our vision on the required transition from contemporary to future circular textiles. Chapter 3 provides an overview of the various options for circular textile fibres, considering the fibre mix we aim for to produce clothing with a range of properties (soft, strong, elastic, breathable, water repellent etc). This chapter is aligned along the family of polymers that are relevant for textiles (polysaccharides, polyesters, polyamides, polyurethanes). Chapter 4 describes then the coupling between the desired polymers and the three sources of renewable carbon available (biomass, recycled materials, and CO<sub>2</sub>). Finally, this publication presents some recommendations for further actions in chapter 5.

# 2 Carbon sources for contemporary textile fibres

### 2.1 Introduction

This chapter describes the origin and resources of our current textile fibres. It is structured along the lines of the four main resources used for textiles, i.e. fossil, forestry, plants and animals. In this chapter only those fibres are described that are produced on commercial scale.

### 2.2 Fossil

### 2.2.1 Introduction

Most of the synthetic textile fibres are made from fossil resources. We describe here the production of polyesters, polyamides, polyurethanes, polyolefins and polyacrylics.

### 2.2.2 Polyesters: PET

The term 'polyester' is a generic name for all polymers containing ester-linkages (-O-CO-) in their polymeric chain. In the apparel sector polyester stands for one of these types of polymers with the acronym PES. The systematic chemical name of this polymer is polyethylene terephthalate or PET. Besides PET, there are also other polyesters used in apparel, and these are being evaluated in Chapter 3.

### General description

PET is nowadays by far the most applied fibre in textiles. There is a global overcapacity of PET production, leading to low prices. Production volume in 2020 was 57.100 kton, largely produced in China (Textile Exchange 2021). The combination of a low price and desirable properties contributes to the success of PET fibres for textiles.

PET comes in various types of fabrics (knitted, woven and non-woven), among which the well-known fleece fabrics. Figure 5 is an illustration of a garment made of 100% PET fabric called 'washable flausch', combined with SEM pictures of the fabric.



Figure 5: Example of a polyester fabric composed of 100% PET. Garment and SEM pictures of the fabric.

### Production route

The production route of PET is shown in Figure 6 and further explained in the text below. It is based on a polycondensation reaction of two different monomers, a diacid and a diol, both shown in Figure 7.

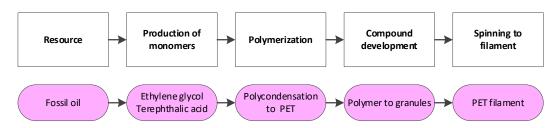


Figure 6: Production route of PET

<u>Resource and production of monomers</u>: PET is produced from two monomers, terephthalic acid (TPA) and 1,2 ethanediol or ethylene glycol (EG) (Figure 7). The monomers are produced from naphtha, a side-fraction from the production of diesel and gasoline. Alternative routes from renewable feedstocks are under development (see Chapter 3).

Figure 7: Monomers terephthalic acid (left) and ethylene glycol (right).

<u>Polymerization and compound development</u>: In a polymerization reactor the building blocks are converted to a polymer which in essence is a very long molecule that consists of repeating monomer units. The polymer PET (Figure 8) is produced by a polycondensation reaction, where the acid- and alcohol-monomers are joined together one by one into a very long chain, and one water molecule is split off with each step. After polymerising, the polymer is processed (often together with additives to deliver additional properties such as colour or water resistance) via extrusion into a granulate that serves as feedstock for the fibre spinning process.

Figure 8: Chemical structure of polyethylene terephthalate (PET)

Spinning to filament: PET fibres are produced using a spinneret, a plate with very small holes to extrude a polymer solution or melt to form fibres. The granules are fed into an extruder and heated and compressed, causing them to melt. The melted polymer is spun at a temperature of 280-290 °C by pushing it through the spinneret. The spinneret holes (dies) are generally 0.2-0.4 mm in diameter. After the formed fibrils leave the spinneret they are drawn, i.e. extended in length direction which aligns the individual polymer chains, thereby increasing their strength. Drawing is usually carried out at a temperature of 60-90 °C (Mather and Wardman 2015). Fibres with a higher strength can be produced by applying a higher draw ratio, i.e. stretching the fibres and aligning the polymer chains further. The spinning process in theory produces filament fibres of infinite length, but the filament can also be cut into staple fibres of a required length, for instance to ease blending with other fibres<sup>‡</sup>.

### 2.2.3 Polyamides: nylon 6 and nylon 6,6

The term 'polyamide' is a name for all polymers containing amide linkages (-CO-N-) in their polymeric chain. Synthetic (man-made) polyamides are known by the generic name nylons, of which nylon 6 and nylon 6,6 are best known. In the past, polyamides dominated the synthetic fibres market. Nowadays, this market position is overtaken by PET, but nylons are still an important textile polymer with a production volume of 5.450 kton/y in 2020 (Textile Exchange 2021).

### Production route of nylon 6,6

Nylon 6,6 are synthetic fibres built up by the polycondensation of two monomers, i.e. a diacid and a diamine. The production route of nylon 6,6 is shown in Figure 9 and further explained in the text below.

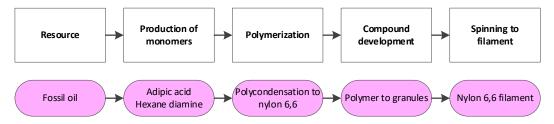


Figure 9: Production route of nylon 6,6

<sup>&</sup>lt;sup>‡</sup> For more information see also the previous "Groene Grondstoffen" booklet: Textiles for Circular Fashion; part 1: Fibre Resources and Recycling Options

Resource and production of monomers: Nylon 6,6 is produced from two monomers, adipic acid and hexane diamine (Figure 10). The monomers are produced from naphtha, a side-fraction from the production of diesel and gasoline. Alternative routes from renewable feedstocks are under development (see Chapter 3).

$$HO \longrightarrow OH$$
  $H_2N \longrightarrow NH_2$ 

Figure 10: Monomers adipic acid (left) and hexane diamine (right)

<u>Polymerization and compound development</u>: For nylon 6,6, hexane diamine and adipic acid are reacted in methanol at high temperature to form hexamethylene diadipate, so called nylon salt. This salt is precipitated from the methanol solution and dissolved in water to a concentration of 60% and further concentrated to >80% by evaporation. The material is then heated to around 250 °C for polycondensation after which nylon 6,6 is formed (Mather and Wardman 2015).

$$\begin{bmatrix} 0 & H & N \\ N & N \end{bmatrix}_{n}$$

Figure 11: Chemical structure of nylon 6,6.

<u>Spinning to filament:</u> Just as with the polyester fibres, the nylon is fed into an extruder where it is melted and subsequently spun into filaments. The filaments are subjected to a drawing process to improve the fibre properties such as tensile strength and stiffness.

# Production route of nylon 6

Nylon 6 fibres are synthetic fibres produced by ring-opening polymerization of one monomer called caprolactam. The production route of nylon 6 is shown in Figure 12 and further explained in the text below.

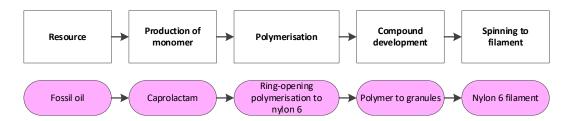


Figure 12: Production route of nylon 6

Resource and production of monomers: Nylon 6 is produced from one monomer called caprolactam, a cyclic compound with an amide group (Figure 13). Caprolactam is produced from naphtha, a side-fraction from the production of diesel and gasoline. Alternative routes from renewable feedstocks are under development (see Chapter 3). Caprolactam can be synthesized from a mixture of cyclohexanol and cyclohexanone (with starting compounds benzene, toluene, and butadiene) (Mather and Wardman 2015).

Figure 13: Monomer caprolactam

<u>Polymerization and compound development</u>: Nylon 6 is produced by a ring-opening polymerization. A catalyst (often water) is required to convert a small amount of caprolactam to  $\epsilon$ -aminocaproic acid, which in turn initiates the polymerisation process. The polymerisation reaction that takes place at temperatures between 250 and 270 °C at atmospheric pressure (Mather and Wardman 2015).

Figure 14: Chemical structure of nylon 6.

<u>Spinning to filament</u>: Just as with the polyester and nylon 6,6 fibres, the nylon is fed into an extruder where it is melted and subsequently spun into filaments. The filaments are subjected to a drawing process to improve the fibre properties such as tensile strength and stiffness.

### 2.2.4 Other synthetics

Polyurethane: elastane

Polyurethanes are a family of polymers with a wide range of characteristics and applications. In textiles, specific polyurethane structures with an extremely high elasticity are applied, known as elastomers. Elastane is an example of an elastic polyurethane, also known by the trade names Spandex or Lycra. Elastane fibres exhibit a very high degree of stretch under stress and will return to their original shape when the stress is relieved. Only a small amount of elastane (2-5%) is enough to give a fabric its desired stretch property. Global elastane fibre production is around 1.080 kton/y (Textile Exchange 2021), mainly for apparel purposes. This volume seems small, but it represents an amount of blended fibres containing a small portion of elastane of 20.000-50.000 kton, which is substantial.

Elastane fibres are made up of two main building blocks, a (polyester or polyether) polyol and an aromatic di-isocyanate. Furthermore, a bifunctional chain extender, such as ethylene diamine, is used to form the final polymer. Generally, all these building blocks are produced from fossil oil. Elastane fibres can be produced by melt spinning, wet spinning and dry spinning, where dry spinning is applied most (Mather and Wardman 2015). In dry spinning, elastane is dissolved in an organic solvent and this polymer solution is forced through the spinneret. The solvent is then evaporated by hot air after the solution leaves the spinneret, causing the filaments to be formed.

### Polyolefins: PE and PP

Polyolefins form the largest group of synthetic polymers in production volume, with polyethylene (PE) and polypropylene (PP) as the most abundant examples. They are produced by polymerising an alkene monomer (-C=C-) into a chain by addition polymerization.

PP is used in textiles mainly in sportswear such as walking socks, swim wear and lightweight wear for climbers. PP fibre production volume was 2.900 kton in 2020 (Textile Exchange 2021). PP is produced from propene which is a by-product of the production of transport fuels from fossil oil. Propene is polymerised into polypropylene and turned into granules that can subsequently be used in the spinning process. PP fibres are produced by melt spinning. The melting temperature of PP is relatively low, around 160–170 °C, so the spinning process is performed at lower temperatures than for example PET spinning.

PE used in textiles is a special grade with very high molecular weight (very long polymer chains). This type of PE can be spun in a special process called gel spinning which

produces fibres of exceptional strength. The fibres possess good cut and puncture resistance and are therefore applied in items such as protective clothing, cut-resistance gloves, and fencing suits.

### Polyacrylics

Polyacrylic polymers have acrylonitrile as their main monomeric building block. For textile fibres, next to acrylonitrile, other co-monomers are incorporated in the acrylic polymer chains. Acrylic fibres are bulky and warm and have been used a lot for replacing (expensive) wool, for instance in sweaters and scarves, and as linings in boots and gloves. Production volume was 1.710 kton in 2020 (Textile Exchange 2021).

The raw material for acrylonitrile is crude oil. Polyacrylics are produced by polymerising acrylonitrile mostly together with other co-monomers by addition polymerisation. The polymer is formed by reacting the main monomer and co-monomers in the same step. Typical co-monomers are vinyl acetate and methacrylate, also produced from crude oil. Acrylic fibres will decompose below their melting temperature. Therefore, they cannot be melt spun but are produced by solution spinning. The solvent used is partly dependent on the types of co-monomers that are present in the polymer. After spinning, the filaments are generally cut into staple fibres for further yarn spinning.

### 2.3 Forestry

### 2.3.1 Introduction

Forestry is the main supplier of wood to produce cellulose pulp for cellulose-based fibres called MMCFs (man-made cellulosic fibres). Cellulose, a natural polymer made up of glucose units, is used to produce paper, textiles, and all other kinds of materials. In 2020 the production volume of MMCFs was 6.500 kton (Textile Exchange 2021). The production route of cellulose textile fibres is shown in Figure 15 and each step is further explained in the next paragraphs.

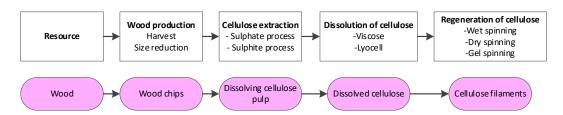


Figure 15: Production route of man-made cellulosic fibres (MMCFs) from forestry

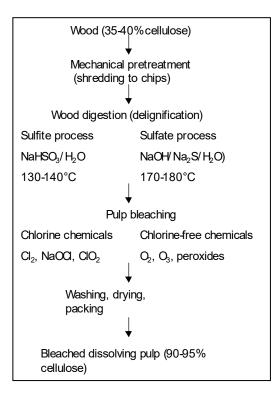
### 2.3.2 Resources and wood production

Trees are specifically grown to produce cellulose pulp, and reforestation is practised in most areas, making trees a renewable resource capturing large amounts of CO<sub>2</sub>. Both softwoods and hardwoods are being used to produce cellulose pulp. The trees are debarked and processed to wood chips.

### 2.3.3 Cellulose extraction

Wood is also called 'lignocellulosic biomass', referring to a material that is build-up of natural polymers cellulose, hemicellulose and lignin in a 'lignocellulosic matrix'. To be able to use the cellulose polymer for textile fibres, it needs to be extracted from this matrix by cellulose pulping processes. For textile applications, a highly pure cellulose is required called dissolving cellulose pulp.

Dissolving cellulose pulping processes are designed to remove as much lignin, hemicellulose, and extractable materials as possible (relevant for the purity of the pulp), whilst avoiding degradation of the cellulose polymer (relevant for the degree of polymerisation (DP) of cellulose). The conditions of these processes need to be adapted to the specific type of biomass (hard wood, soft wood, tree species). Pulping processes to produce dissolving cellulose pulp from wood include the sulphite process or the prehydrolysis-kraft process. Both pulping processes are briefly described in Figure 16.



Sulphite process: Raw materials suitable for the sulphite process are for example hemlock, spruce, fir, pine in softwoods, and Eucalyptus and Acacia in hardwoods.

Sulphate process: the sulphate process (or prehydrolysis-kraft pulping process) is suitable to utilize various woods, even if these are not suitable for the sulphite process. So, the prehydrolysis-kraft process has the potential to use more wood varieties and presently, oak, beech and gum wood are used.

Figure 16: Production of dissolving pulp from wood

The total world production of dissolving pulp is 8.6 Mton/year§. To produce this volume, more than 20 Mton of wood is required, as wood contains 35-40 wt% cellulose. Production of dissolving cellulose pulp accordingly generates large side streams of lignin and hemicellulose amongst others.

### 2.3.4 Dissolution and regeneration of cellulose

### 2.3.4.1 Introduction

Cellulose is a natural, linear molecule with a high degree of crystallinity and intermolecular hydrogen bonding. As a result, it is not easy to produce a cellulose fibre from a cellulose pulp, as cellulose does not melt upon heating and is not easy to dissolve. To produce cellulosic fibres for textiles, the cellulose in the dissolving cellulose pulp needs to turn into a soluble form and subsequently regenerated (reformed) into a certain

<sup>§</sup> https://www.globenewswire.com/fr/news-release/2020/02/05/1980374/0/en/Global-Dissolving-Wood-Pulp-Industry-Outlook-2020-2025-China-Still-Dominates-the-Global-Market-Despite-a-Trade-War.html

shape, in this case a fibre. As the processes of dissolution and regeneration of cellulose are closely connected, they are combined in this paragraph.

Cellulose dissolution and fibre production can be done in many ways. Here the main commercially available cellulosic fibres are described, including their production processes: viscose (and its variants modal and cupro), lyocell, and acetate. Overall production volume of MMCFs was 6.500 kton in 2020, with viscose (5.200 kton) as main type (Textile Exchange 2021).

### 2.3.4.2 <u>Viscose</u>

For the viscose process, the dissolving cellulose pulp is treated with carbon disulphide to form cellulose xanthate, which is subsequently dissolved in sodium hydroxide. The fibres are then produced by wet spinning: the solution of cellulose xanthate (DP around 270) is extruded and forced through a spinneret, a plate with thousands of small holes, into a bath containing sulfuric acid, sodium sulphate and zinc sulphate. In this bath, the cellulose xanthate reacts back (is regenerated) to cellulose, forming the viscose fibre. During the regeneration process the core of the fibre shrinks, which causes the outer layer to wrinkle. This gives viscose fibres their typical jagged cross-sectional shape and striations along their length (Mather and Wardman 2015). After extrusion, thousands of filaments lie parallel to each other and form a tow. The tow is drawn to increase its strength and is then usually cut into staple fibres of approximately 40 mm length. The staple fibres are formed into a yarn.

Figure 17 is an illustration of a garment made of viscose in a so-called satin weave structure, clearly shown by the left SEM-picture. Also, the characteristic striated fibre surface of viscose is shown by the right SEM-picture.

Viscose variant: Modal

Modal is a type of 'High Wet Modulus' viscose, a stronger variant of viscose, with a very precisely defined strength. The fibre is spun in a slightly different way, into a bath with a different chemical composition, to obtain a stronger fibre which is soft to the touch.

### 2.3.4.3 Cupro

When cellulose (in this case usually from cotton linters and not from wood, as a higher DP is required) is dissolved in a solvent containing cuprammonium ions, the hydroxyl groups of the cellulose form a complex with these cuprammonium ions.

The resulting solution with 9% cellulose has a blue colour and the DP is in the range of 600-800, giving the spinning solution a very high viscosity. Fibres are formed by wet

spinning in a bath containing dilute sulphuric acid. The resulting yarns are extensively washed in water to remove the toxic copper sulphates and ammonium sulphates. Cupro is often applied as lining in garments, but also as a silk like fabric for blouses as shown in Figure 18.



Figure 17: Example of a viscose fabric (satin weave) composed of 100% viscose. Garment and SEM pictures of the fabric.



Figure 18: Example of a cupro fabric (plain weave) composed of 100% cupro.

Garment and SEM pictures of the fabric.

# 2.3.4.4 Lyocell

Much effort has been devoted to find other solvents for cellulose and developing other processes for cellulose-based fibres than viscose or cupro. A successful result of this is the development of lyocell fibres. These fibres also produced from dissolving pulp, but the process is chemically less complicated because the cellulose is dissolved directly in a solvent called N-methyl-morpholine-N-oxide (NMMO). NMMO is an amine oxide, a

highly polar and water-soluble compound, in which cellulose can be dissolved at 100 °C. The fibres are spun by dry-jet wet spinning process: after leaving the spinneret and before entering the water bath, the fibres pass through an air gap, which causes the filaments to stretch and results in a strong orientation of the polymeric molecules. Lyocell fibres are therefore more crystalline and more oriented than viscose fibres, and as a result they possess a higher strength than viscose fibres, both under wet and dry conditions. Figure 19 is an illustration of a garment made of 100% lyocell.

### 2.3.4.5 Acetate

Cellulose acetate and triacetate are cellulose derivatives. Triacetate is cellulose of which all the hydroxyl groups (3 on each glucose) are converted into an acetate ester, resulting in a degree of substitution of 3. Cellulose, with a substitution degree of 2.4 on each glucose, is commonly known as cellulose acetate. Cellulose acetate is mainly used for linings in jackets and in cigarette filters.

### 2.3.4.6 Production of MMCFs summarized

The production of regenerated fibres includes two essential processes: dissolution of the cellulose from the pulp, and regeneration of cellulose through a spinning process. This production approach does not alter the molecular content of the cellulose but does result in some changes in polymeric structure compared to raw cellulose (Chen 2015). These changes alter/modify the fibre properties in terms of strength, absorbency,

drapability, dyeability etc. The dissolution of cellulose and the subsequent regeneration of cellulose are closely connected but there are differences, summarized in Table 2. Viscose, Modal and Cupro are regarded as the first generation regenerated cellulose fibres, whereas Lyocell is seen as the next generation with a less complicated process.

Table 2: Characteristics of production processes of regenerated cellulose fibres

Regenerated cellulose fibre	Spinning solution	Spinning technique and spinning bath
Viscose	Sodium cellulose xanthate in weak caustic soda	Wet spinning in sulfuric acid (10%), sodium sulphate (18%) and zinc sulphate (1%)
Modal	Sodium cellulose xanthate in weak caustic soda	Wet spinning in dilute sulfuric acid and sodium sulphate
Cupro	Cuprammonium cellulose complex	Wet spinning in dilute sulfuric acid
Lyocell	Cellulose in NMMO	Dry-jet wet spinning



Figure 19: Example of a lyocell fabric (satin weave) composed of 100% lyocell. Garment and SEM pictures of the fabric.

### 2.4 Plants

### 2.4.1 Introduction

Plant-based fibres are fibres that are produced by plants like cotton and flax. When these fibres are extracted from the plants, the natural fibre structure is kept intact as much as possible. Whereas the properties of man-made fibres can be tailor-made for specific applications, this is not possible for natural plant-based fibres, and only specific plant species produce fibres suitable for textiles.

Plant-based fibres are applied in many applications like ropes, mats, curtains, carpets, and packaging material. For application in apparel, the plant-based fibres need to have the following additional important requirements:

- Soft to the skin. This is mainly determined by the fineness (diameter) of the fibre and the composition. High-cellulose content fibres without lignin are preferred, whereas lignin-containing fibres are less suitable for apparel.
- Can be spun into a fine yarn by spinning techniques. This is determined by fibre strength, length, but also structure. For example, cotton fibres are relatively short compared to flax, but can be spun into a yarn due to the characteristic twists (convolutions).

Plant fibres are obtained from various parts of the plants, i.e. seed, bast, leaf or fruit. The function of the fibre in the plant largely determines their composition and properties. In this chapter the different plant fibres are being discussed, including their processing steps, properties, and applications.

### 2.4.2 Overview of plant species

An overview of plants that can produce fibres or fibre bundles is listed in Table 3. Only seed fibres (cotton) are single cells and are referred to as 'fibres', whereas fibres originating form bast, leaf, fruit or grass are naturally organised into bundles and are therefore called 'fibre bundles' (Mwaikambo 2006).

In Table 3 some taxonomy is included, as *species name*, *family* and *clade* of the plants is included. Taxonomy is the science of biology that studies organisation of living organisms, and organisms are grouped based on shared characteristics. Taxonomy can be relevant here, to arrange various plants species in families, and to search for new plants with similar preferential characteristics (see also Chapter 3). A short description of the plant families can be found on page 95 of this booklet.

Table 3: Overview of plant fibres from fibre crops. Adapted from (Mwaikambo 2006, Keijsers et al. 2013, Sfiligoj Smole et al. 2013, Textile Exchange 2021)

Plant	Species name	Family	Clade	Part of the plant	Used for apparel	Volume (Mton)
		Se	ed fibres			
Cotton	Gossypium species	Malvaceae	Dicot	Seed	Yes	26.2
		Ва	st fibres			
Flax	Linum usitatissimum	Linaceae	Dicot	Stem	Yes	1.0
Hemp	Cannabis sativa	Cannabaceae	Dicot	Stem	Yes, small scale	0.17
Jute	Corchorus olitorius	Malvaceae	Dicot	Stem	Yes, small scale	3.3
Ramie	Boehmeria nivea	Urticaceaea	Dicot	Stem	Yes, small scale	0.1
		Le	eaf fibres			
Sisal	Agave sisalana	Asparagaceae	Monocot	Leaf	No	0.2
Raffia	Raphia species	Arecaceae	Monocot	Leaf	No	Unknown
Fruit fibre						
Coconut	Cocus nucifera	Arecaceae	Monocot	Coconut husk	No	1.2
Grass fibres						
Bamboo	Bamboo species	Poaceae	Monocot	Stem	Yes, small scale	Unknown

Flowering plants are divided in two major groups, the monocotyledons and the dicotyledons (see Figure 20). The seeds of monocotyledons typically contain one embryonic leaf or cotyledon, where the seed of dicotyledons has two embryonic leaves or cotyledons. The relevance for textile fibres lies in the structural differences between monocots and dicot plants, how the leaves are formed, and how vascular bundles (i.e. the fibres) are organised. Most of the plant-fibres in Table 3 applied in apparel (seed fibre and bast fibres) are dicots, whereas the other plant-fibres (leaf, fruit and grass fibres) are monocots and are applied in other applications.

In the next paragraphs the seed, bast, leaf, fruit and grass fibres are explained in more detail.

MONOCOT		DICOT	
Single Cotyledon		Two Cotyledon	
Long Narrow Leaf Parallel Veins		Broad Leaf Network of Veins	
Vascular Bundles Scattered		Vascular Bundles in a Ring	
Floral Parts in Multiples of 3		Floral Parts in Multiples of 4 or 5	

Figure 20: Differences between monocot and dicot. By Flowerpower207-Own work, CC BY-SA 3.0,

https://commons.wikimedia.org/w/index.php?curid=26233760

### 2.4.3 Seed fibres

Seed fibres include cotton and kapok. Seed fibres are in general high in cellulose and very low in lignin content, as the fibres do not give strength to the plant, in contrast to bast or leaf fibres. Cotton is by far the most commercially important fibre from all fibre crops; kapok might be an interesting fibre and is further explained in chapter 3.

### Cotton

### General description

Cotton is the most popular plant-based fibre for apparel. For consumers it is attractive as it is soft, breathable, strong (also under wet conditions) and machine washable. Special feature of the cotton fibre is the 3D-structure (twists) that enables spinning of these relatively short fibres into yarn or threads by yarn spinning. Figure 21 is an illustration of a garment made of a high-quality cotton fabric, and the corresponding SEM-pictures show the typical 3D-structure of cotton.



Figure 21: Example of a cotton fabric (plain weave), composed of 100% cotton. Garment and SEM pictures of the fabric.

### Production route

The production of cotton is shown in Figure 22 and further explained in the text below.

<u>Resource</u>: Cotton is a seed hair coming from the perennial cotton shrub (e.g. *Gossypium hirsutum*, see Figure 23) (Seagull and Alspaugh 2001), but for commercial purposes cotton is grown as an annual crop. The shrub grows in many parts of the world, but main producers of cotton are China and India (Yu 2015).

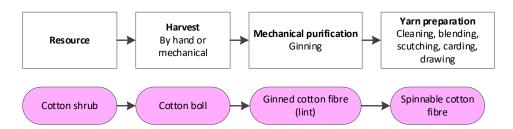


Figure 22: Production route of cotton fibres



Figure 23: Gossypium hirsutum (cotton)

<u>Harvest:</u> The mature cotton bolls are removed from the plant by hand or by mechanical harvesters, which can be cotton strippers or cotton pickers. Cotton strippers remove the complete boll from the plant, resulting in a higher trash content of harvested seed cotton (Seagull and Alspaugh 2001).

Mechanical purification: After harvesting, the seed cotton is transported to the ginning factory, where the long cotton fibres are separated from the seed, other plant matter (twigs, leaves etc.), and shorter cotton fibres. Only the long lint fibres are packed into bales and transported to spinning mills (Yu 2015). The shorter fibres, called 'cotton linters', are unsuitable for spinning but are an excellent source of pure cellulose to produce value paper or regenerated cellulose fibres.

<u>Yarn preparation</u>: After ginning, the cotton is transported to the mills where the cotton is further purified and blended with other cotton batches. The cotton is then further processed by several techniques to create rovings (long and narrow bundle of fibres) that are used in the spinning process.

#### 2.4.4 Bast fibres

Bast fibres are obtained from stems or stalks of dicot plants like flax, hemp, jute or ramie, and are often referred to as "soft" fibres. The fibres occur in bundles, consisting of long elementary fibres (i.e. the vascular bundles in a ring in Figure 20). They are responsible for transport of soluble compounds throughout the plant.

The fibres are embedded in substances (like pectin, waxes) which must be removed during the processing of fibre extraction. The fibres are arranged around a woody core, which is separated from the fibres in a later stage of the production process.

#### Flax

## General description

In the past, flax was widely used throughout Europe, but nowadays flax is considered a luxury fibre (Mather and Wardman 2015). Flax is a non-lignified, soft, flexible, and lustrous fibre. Fabric made from flax fibres is called linen. It is a good conductor of heat, and linen garments are excellent for warm weather. Linen tends to wrinkle and blending with other cellulose-based fibres such as viscose or cotton can make it feel softer and less prone to wrinkling. An example of a garment of linen is shown in Figure 25.

#### Production route

The production route of flax fibres is shown in Figure 24 and further explained in the text below.

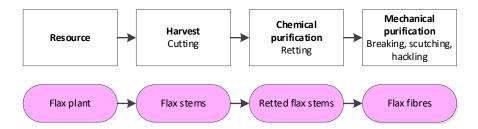


Figure 24: Production route of flax fibres



Figure 25: Example of a linen fabric (plain weave), composed of 100% linen.

Garment and SEM pictures of the fabric.

<u>Resource</u>: Flax (*Linum usitatissimum*, see Figure 26) requires a temperate climate and can be grown in many parts of Europe. The best fibres are obtained from the coastal regions of Northern France, Belgium, and the Netherlands (Mather and Wardman 2015).

<u>Harvest</u>: Flax can be harvested by pulling the plants from the soil.



Figure 26: Linum usitatissimum (flax)

<u>Chemical purification</u>: After harvest, the fibres need to be loosened from the woody stem. This is done by retting, where micro-organisms break down the cellular tissue surrounding the fibres, waxes and pectins, which bind them to the stem. Often this is done by dew retting, which means just spreading the harvested plants on the soil for several weeks, until the fibres come loose.

<u>Mechanical purification</u>: After retting the flax stems are led through the fluted rollers of a brake to break the woody core in small pieces, the shives. The shives are removed through a process called scutching, and the resulting fibre bundles are refined to thinner technical fibres by hackling, which is a combing process, and formed into a tow. The tow can then be spun into a yarn.

### Hemp

Hemp fibres can be spun into yarns that can be woven or knitted to produce a fabric. Hemp fibres are coarser than flax, making them less suitable for apparel, and rather difficult to bleach (Sfiligoj Smole et al. 2013). For this reason, hemp is often blended with other fibres to produce clothes. Hemp fibres are strong, making them suitable to produce jeans or work clothes, possibly combined with a softer fibre like viscose. An example of a hemp fabric and garment is shown in Figure 27. Compared to the linen example, the hemp jacket is less flexible and has a worse drape and pliability.



Figure 27: Example of a hemp fabric (plain weave), composed of 100% hemp. Garment SEM pictures of the fabric

Hemp (*Cannabis sativa*, see Figure 28) grows in various climates and on various soils. It grows easily to a height of 4 m without agrochemicals (Sfiligoj Smole et al. 2013). Hemp fibre (from industrial hemp) and marijuana are produced from the same species of *Cannabis sativa* but from different strains. Industrial hemp is planted only centimetres apart with most of its leaves concentrated at the top. In contrast, the marijuana plant is quite dens, leafier, shorter, bushier (Mwaikambo 2006). The hemp fibres are extracted using the same retting process as for flax.



Figure 28: Cannabis sativa (hemp)

## Jute

Jute (*Corchorus olitorius*, see Figure 29) is a long and shiny fibre that can be spun into coarse, strong threads, less applicable for apparel. It is one of the cheapest natural fibres, but together with bast fibre the largest in volume. Jute is mainly used to produce lower-value products, such as ropes and bags. It is also widely used for carpet manufacture as backing material. The jute plant grows in warm, humid climates (India, Bangladesh); processing is similar to flax.



Figure 29: Corchorus olitorius (jute)

#### Ramie

Ramie fibres are characterised by coarse, thick cell walls, they lack twists and have striated surfaces. Ramie is the strongest of the bast fibres and has a good wet strength (strength under wet conditions). Disadvantage is the low elasticity, making ramie fibres stiff and brittle. Ramie has had limited acceptance for textile use, as spinning of the fibre is difficult, and weaving is complicated by the hairy surface of the yarn produced, resulting from lack of cohesion between the fibres (Mather and Wardman 2015). Ramie is used to make products as industrial sewing thread, packing materials, fishing nets, and filter cloths. It is also made into fabrics for household furnishings (upholstery, canvas) and clothing, frequently in blends with other textile fibres.

Ramie (*Boehmeria nivea*, see Figure 30) is an herbaceous perennial plant in the nettle family *Urticaceae*, native to eastern Asia. The plant is grown, in Brazil, India, Japan, South-East Asia and Southern Europe.



Figure 30: Boehmeria nivea (ramie)

The retting process used for flax and hemp is not sufficient for the extraction of ramie fibres. Ramie fibres are extracted from the green plant by decorticating processes (Mwaikambo 2006). A decorticator is a machine used to strip fibre bundles from the stem of the plant. Decorticated fibres are washed before drying and combed and sorted into various grades.

### 2.4.5 Leaf fibres

Leaf fibres are often referred to as "hard" fibres, and have limited commercial value, mainly because they are generally stiffer and have a coarser texture than bast fibres. They are obtained from the leaves or leaf stalks of various monocotyledonous plants. Most widely applied leaf fibres are raffia and sisal, but not for apparel applications.

#### Raffia

Raffia fibre is widely used throughout the world. It is used in twines, ropes, baskets, placemats, hats, shoes, and textiles.

Raffia is obtained from the raffia palm (see Figure 31) native to tropical regions of Africa, and especially Madagascar. The fibre is obtained from the membrane on the underside of frond leaves. The membrane is taken off to create a long thin fibre which can be dyed and woven as a textile.



Figure 31: Raffia palm

## Sisal

Sisal fibre from *Agave sisalana* (see Figure 32) is coarse and inflexible. Lower grades are processed by the paper industry because of its high cellulose and hemicellulose and low lignin content. Medium grade fibre is used in the cordage industry for making ropes and twines. Higher grade fibres (after treatment) are converted into yarns and used by the carpet industry (Sfiligoj Smole et al. 2013).

Major sisal fibre growing countries are Brazil, Tanzania, and Kenya, and sisal is native to Mexico. Leaves are removed from the plant, and the fibres are obtained by a decortication process, in which the leaf is crushed between rollers and then mechanically scraped (Sfiligoj Smole et al. 2013).



Figure 32: Agave sisalana (sisal)

### 2.4.6 Fruit fibre

In the category fruit fibre there is only one fibre suitable for textile production, i.e. coir fibre from coconuts. Coir has the third largest market share of plant-based fibres, after cotton and flax.

## Coconut

The coconut palm tree (*Cocus nucifera*, see Figure 33) grows in tropical areas (Indo-Malaysian region, East and West Africa, and Central and South America). The fibres are originating from the bast of the coconut. The fibres are a residue stream from the food industry that produces coconut milk and butter from the coconut.

The retting process is traditionally employed to extract coir fibre bundles where the husk is immersed in water. The decorticating process can also be used to separate the fibre bundles (Mwaikambo 2006).

Coir is mainly used in home textiles, like mats and carpets, and ropes.



Figure 33: Cocus nucifera (cocus)

## 2.4.7 Grass fibres

The family of grasses (Poaceae) is one of the most economically important plant families, providing us with food and feed. Relevant crops include maize, wheat, straw, barley, and rice. Besides cereal grasses, it also includes plants like bamboo and miscanthus, and the grasses of natural grassland and species cultivated in lawns and pasture. Normally grasses are not considered a fibre crop, but bamboo is an exception and is further explained here.

### Bamboo

Bamboo is a versatile crop with a broad range of applications. For apparel, bamboo products are usually based on regenerated cellulose fibres like viscose or lyocell. A small portion of the cellulose pulp production is made from non-wood sources like bamboo (van Dam et al. 2018). In addition, it is also possible to produce yarn from bamboo fibres, but this is done on a limited scale.

Bamboo (Figure 34) is grown in Asia, where it is of significant economic importance, but is also cultivated in Europe. It belongs to the fastest growing plants in the world. In bamboo (like in other grasses), the internodal regions of the stem are usually hollow and the vascular bundles in the cross-section are scattered throughout the stem. Fibres can be removed from the strands by e.g. boiling in an alkali solution, or cellulose is extracted by cellulose-extraction processes as described in section 2.3.3. There is also

an alternative process that produces 'bamboo linen', although these fibres contain much more lignin than bast fibres. The woody stalks are then crushed mechanically and naturally retted, releasing the bamboo fibres (Mather and Wardman 2015).



Figure 34: Green bamboo leaves

#### 2.5 Animals

### 2.5.1 Introduction

Animal fibres are protein-based fibres from mammals, insects, or birds. Mammals provide wool (sheep), angora (rabbits), mohair and cashmere (goats), or alpaca (alpacas), all containing the natural polymer keratin. Certain insects can produce silk (natural polymer fibroin), and birds provide down (keratin). Wool and silk are the two animal fibres further explained here. Production volumes are small, 1000 kton for wool from sheep, and 110 kton for silk (Textile Exchange 2021).

#### 2.5.2 Wool

Wool, the hair of sheep, is one of the most complex structures in textile fibres. This complexity leads to a specific range of mechanical, physical, and chemical properties. Wool is one of the oldest fibres known to humans, it was one of the first fibres to be spun into a yarn and woven into a fabric. Wool is a highly crimped protein fibre mainly composed of keratin (Babu 2015). Certain qualities of wool are very suitable for apparel applications, and an example of a woollen fabric is shown in Figure 35. The SEM pictures clearly show the scaled surface of the wool fibres.

The wool is removed from the sheep by shearing. The sheared wool is called raw wool and is cleaned before further processing. Raw wool can contain impurities up to 50% by weight. These impurities include waxes, suint (perspiration salts), vegetable matter, and dirt. Raw wool is typically cleaned (scoured) at mild temperatures with mild alkali and detergent (Hauser 2015). The cleaned wool is carded to orient the fibres, which eventually leads to stronger yarns. The carded fibres are gently scraped into strands called roving, and the roving is then spun into a yarn.



Figure 35: Example of a woollen fabric, composed of 100% wool. Garment and SEM pictures of the fabric.

# 2.5.3 Silk

Silk is an animal fibre produced by some insects to build their cocoons or webs. Most silk that is used for textile applications is produced by the caterpillar of the mulberry silk moth *Bombyx Mori*. Silk is widely valued for its strength, lustre, and bright colours. An example of a silk fabric is shown in Figure 36.

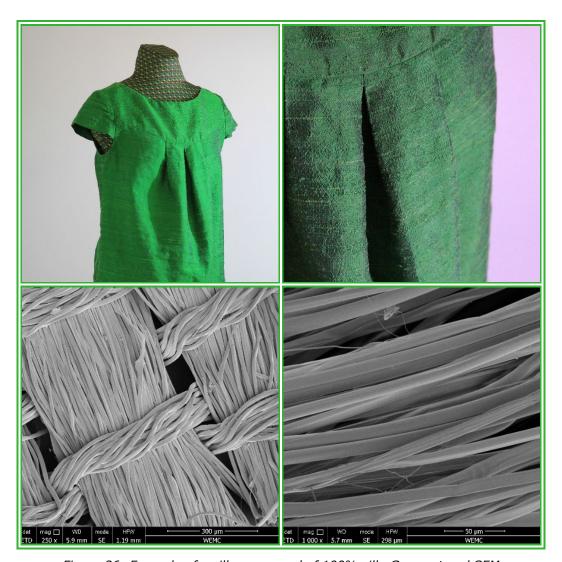


Figure 36: Example of a silk, composed of 100% silk. Garment and SEM pictures of the fabric.

The caterpillar of the silkworm feeds on the leaves of the white mulberry and forms a cocoon. These cocoons are harvested and immersed in heated water, enabling the protein filaments to be unwound as yarns. Typically, the single threads from 4 to 20 cocoons are combined while being unwound onto a reel, the threads are slightly twisted together to form a single raw silk yarn (Babu 2015).

## 2.6 From carbon to contemporary textile fibres

In this chapter, the carbon sources for our contemporary textile fibres have been explained. Figure 37 shows schematically the relation between basic input, these carbon sources, the subsequent intermediate structures (monomer, polymer, fibre), polymer categories and textile fibre categories.

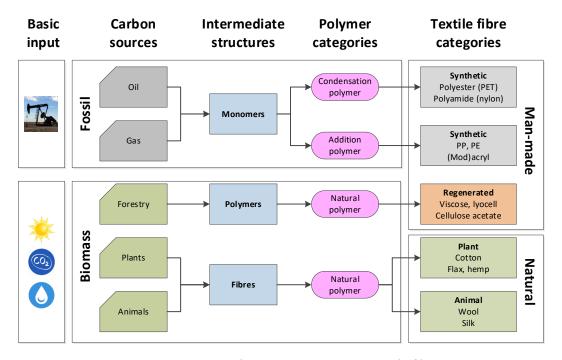


Figure 37: From carbon to contemporary textile fibres

For contemporary fibres, this scheme is rather straightforward; for future circular fibres it will be much more complex, as will be explained in the next chapters.

## 3 Future circular textile fibres

#### 3.1 Introduction

The previous chapter describes the production routes of presently used textile fibres from non-renewable fossil resources and renewable ones from biomass (forestry, plants, and animals). For future circular fibres this ordering of resources will be different. As we explained in section 1.4, we will have in the future no more non-renewable carbon sources but three different renewable carbon sources for textile fibres, i.e. biomass, recycled content and CO<sub>2</sub>.

This chapter describes in more detail the circular options that will be available (now and in the future), ordered along the lines of level of assembly introduced in Chapter 1 (fibres, polymers, monomers). The next paragraphs explain in more detail the ongoing and/or desired developments for the most important polymer types for textiles: polysaccharides (cellulose), polyesters, polyamides, and polyurethanes.

### 3.2 Polysaccharides: cellulose

#### Focus on natural and condensation polymers

In analogy of our previous publication on textiles for circular fashion (Part 1: Fibre resources and recycling options), we classify the various types of textile fibres based on the main chemical bonds in the polymeric structure.

For <u>contemporary textile fibres</u>, the relevant polymers include polysaccharides (natural polymers), polyesters (condensation polymers), polyamides (condensation polymers), polyurethanes (condensation polymers), polyolefins (addition polymers) and polyacrylics (addition polymers) (see also Table 1).

For <u>circular textile fibres</u> we focus on natural and condensation polymers and exclude addition polymers. Based on resource use efficiency and recyclability, addition polymers are poor candidates for circular fibres.

#### 3.2.1 Introduction

Cellulose-based fibres already form a major fraction of the textile fibre mix, and it is foreseen that this fraction will increase the coming years. Cellulose is an abundantly available natural polymer and is, due to its molecular structure, highly suitable for textile applications. Cellulose is used as a natural fibre or as a resource of cellulose pulp for man-made fibres.

The current sources of cellulose for textiles are described in chapter 2 (wood, plants), but there are other, less familiar or forgotten biomass types or biomass residues that might also be suitable for textiles. To aid this search for alternative cellulose sources

and their applicability, we constructed a decision tree for the three categories of textile fibres we know: natural (fibre), regenerated (polymer) and synthetic (monomer). In Figure 38 the questions are presented that need to be answered when dealing with the various sources of feedstocks and residues for textile applications.

The fastest route to a textile is the use of biomass that supplies natural fibres that can be spun directly into a yarn (fibre); when this is not possible the second-best option is to use the biomass as a source of cellulose pulp for regenerated cellulose fibres (polymer); and the last option is to use it as a source of sugars for synthetic fibres other than cellulose (monomer).

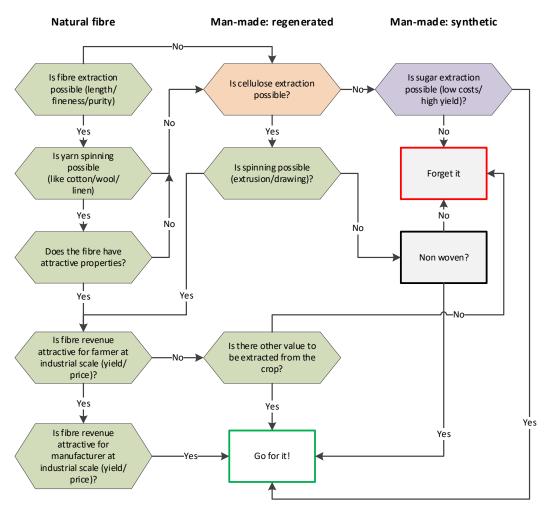


Figure 38: Decision tree for routes from biomass to fibres

The next paragraphs elaborate on less common or alternative plants that produce natural cellulosic fibres, developments in the field of regenerated cellulose, and production of cellulose not by plants but by bacteria.

## 3.2.2 Alternative sources of cellulosic natural fibres (fibres)

In this paragraph, alternative sources of cellulosic natural fibres are described for seed, bast, leaf and grass fibres.

### 3.2.2.1 Seed fibres

In the category seed fibres, the alternatives are limited. One example is kapok, another example of a plant producing seed fibres is cattail.

## Kapok

The kapok tree (*Ceiba pentandra*, see Figure 39) grows in tropical climates (South America, Central America, West Africa). It belongs to the Malvaceae family, together with the cotton tree (*Bombax ceiba*, an Asian tropical tree) that produces white, cotton-like fibres.



Figure 39: Ceiba pentandra (kapok)

Kapok is a highly lignified seed fibre compared to cotton. The kapok fibres are separated from the seeds by shaking, as they are loosely held to the seed. Kapok is a smooth, twist-less fibre. Its cell wall is thin and covered with a thick layer of wax. A wide lumen is filled with air, so the fibres are hollow. It has excellent thermal and acoustic insulating

properties, and an exceptional capability of absorbing non-polar liquids like oil. It is mainly used in the form of stuffing (insulation) and nonwoven products, and rarely used in yarns (Sfiligoj Smole et al. 2013). Kapok cannot be spun as the fibres are too thick and short, and the surface is too smooth.

#### Cattail

Cattail (genus Typha, see Figure 40) are aquatic or semi-aquatic, herbaceous perennial plants belonging to the Typhaceae family. The genus Typha is largely distributed in the Northern Hemisphere, where it is found in a variety of wetland habitats. Applications of cattail are diverse, ranging from food, feed to paper or building material.



Figure 40: Cattail

For textiles the plant might be interesting. Seed hairs may be used as insulation material (comparable to kapok), but also fibres in the stem might be useful. Cattail belongs to a small family of monocot plants and resembles grasses like reed. It seems possible to obtain a technical fibre from the stem through an alkaline degumming process (resembling fibre extraction from bamboo), meeting basic spinning conditions like fibre length, thickness, and strength (Liu et al. 2011). Chakma (Chakma et al. 2017) states that "the textile and sub microscopic characteristics of cattail fibre suggests that the fibre may be used for apparel and non-apparel applications (insulation and biomedical)".

### 3.2.2.2 Bast fibres

Bast fibres are "soft" fibres obtained from the stems or stalks of dicot plants by retting. In Europe, the main plants grown for fibre production in the past were hemp, flax, nettle, and hop (Skoglund et al. 2020). Flax and hemp are described in chapter 2 as conventional textile fibres; hop and nettle are described here as alternative (or forgotten) sources of fibres for textiles.

#### Hop

Hop (*Humulus lupus, see* Figure 41) belongs (like hemp) to the Cannabaceae family. They have the natural organisation of fibre bundles in the bast and are therefore interesting as fibre crops for textiles.



Figure 41: Humulus lupulus (hop)

Hop is native to the Northern Hemisphere (Europe, western Asia and North America), and the fruits are used to produce beer. It is a perennial herbaceous plant up to 10 meters tall, living up to 20 years. Hop fibres from the stems have been used in the past for textiles and paper. Beer production leads to a large amount of residual material, since only the female flowers are being used. Lukesova (Lukešová et al. 2019) reported that in the past, people tried to use this leftover material in different ways. Still, hop fibres were most likely not a very commonly used material. One of the reasons was that the retting process of the long, branched climbing plant was difficult compared to the straight stems of flax, hemp and nettle, which were easily bundled into sheaves.

Nonetheless, there are records that mention the use of hop fibres for textile production (Skoglund et al. 2020).

#### Nettle

Stinging nettle (*Urtica dioica*, see Figure 42) belongs to the family of Urticaceae, like ramie (see chapter 2). It is a perennial herbaceous plant spread in Europe, Asia, and America. The crop has gained interest because it is a source of many added-value natural products exploiting all plant parts (stem, leaf, root, and seed) (Di Virgilio et al. 2015).



Figure 42: Urtica dioica (nettle)

Nettle production in Europe began in the 19<sup>th</sup> century, and during WO I and WO II nettle was promoted as a substitute for scarce cotton. Nettle production stopped when processing facilities were destroyed during the wars, and cheaper fibres became more readily available (Vogl and Hartl 2003). So technical feasibility of nettle textile is proven, and now there are small volumes of nettle textiles on the market again.

In nettle, the fibres are located between the outer bark and the central woody core, arranged in bundles held together with pectins. Fibre extraction is challenging according to Di Virgillo (Di Virgillo et al. 2015). Hemp and flax processing methods could be adapted to stinging nettle, keeping in mind that stinging nettle is prone to over-retting.

### 3.2.2.3 Leaf fibres

Leaf fibres are regarded as "hard" fibres, being stiffer and coarser in structure than bast fibres, and have so far limited commercial value for apparel applications. Alternatives to sisal and raffia include abaca, banana and pineapple fibres.

#### Abaca

Abaca or manila hemp is extracted from the abaca plant *Musa textilis* (see Figure 43). The plant belongs to Musaceae family, resembling species that bear edible banana fruit, and grows mainly in the Philippines.



Figure 43: Musa textilis (abaca)

The fibres are obtained from the leaf sheaths in bundles of individual cells. They are removed from the strands by boiling in an alkali solution. Abaca fibres are used in the paper industry and textile industry to make ropes, strings, threads and fabric for clothing or curtains. New developments are abaca fibres for the development of nanomaterials, biodegradable materials and composite materials that are utilized by the automotive industry, aeronautics, shipping, construction, and furniture (Simbaña et al. 2020).

## Banana

Musa acuminata (see Figure 44) is a species of banana native to Southeast Asia. To obtain the best fibre quality, the plants are cut when they are almost at the flowering stage, so before any fruit has formed. Processing is done manually and includes pressing, drying, and combing. The fibre that is being produced is usually of low quality

because separation of the fibre bundles is done either after the fruits have just developed, or when they have ripened ready for food purposes (Mwaikambo 2006).



Figure 44: Musa acuminata (banana)

# Pineapple

The pineapple (*Ananas comosus*, see Figure 45) is a tropical plant with an edible fruit and is the most economically significant plant in the family Bromeliaceae.



Figure 45: Ananas comosus (pineapple)

The pineapple is native to South America, where it has been cultivated for many centuries. It is also a significant fruit in India. The plant grows as a small shrub. It is possible to extract fibres from the leaves for textiles, and pineapple leaf fibres might become more important to the textile industry (Kannojiya et al. 2013) than they are now.

#### 3.2.2.4 Grass fibres

Grasses have lignocellulosic stems that are hollow, except at the nodes. They have in common that they have a typical lignocellulosic composition of cellulose (35-40 wt%), hemicellulose (20-25 wt%) and lignin (15-20 wt%), and they lack the natural organisation of fibre bundles in the bast. Fibres that are present in the stems are in general short and stiff, and less suitable to produce fibres for apparel.

Grasses that provide us with food (rice, wheat, maize, sugar cane) generate residues in the form of straw, trash and/or husk. Often these residues have competing uses like feed, as energy source, or as nutrients for the soil. For applications in apparel, these sources can possibly serve as a source of cellulose (Adhia et al. 2021). However, they have in common that the ash content is higher than in wood, and this may pose problems in the cellulose extraction process.

## 3.2.2.5 Summary and outlook

In this chapter the main alternative sources for cellulose fibres were described. Alternatives can be found in plant residues, or plants that were used in the past to make textiles. It can be of interest to search for other plants in families that are known for their fibre production like Malvaceae (cotton, kapok), Cannabaceae (hemp, hop), and Urticaceae (ramie, nettle). Nevertheless, the number of alternatives is limited.

Table 4 shows an overview of plants and their suitability for clothing as insulation material, natural fibre, cellulose pulp for regenerated fibres, or as sugar source for synthetic fibres. To produce natural fibres for yarns, the options lie mainly within bast fibres from the stems of dicot plants, or leaf fibres from leaves of monocot plants. One of the challenges is to isolate the fibres in a controlled manner by retting or other types of enzymatic processes. For cellulose pulp a high cellulose content is required, and this is found in bast or leaf fibres. Grasses suffer from high ash content, making them less suitable. Almost all plants addressed in this work can be used as a source of sugar. Finally, insulation textiles is also of interest, as nowadays for insulation mostly PET is used. For this application, seed fibres such as kapok or the seeds from cattail are highly interesting.

Table 4: Overview of alternative sources for cellulose-based textiles and suitability as insulation material, natural fibre, cellulose pulp or as source of sugars for biobased synthetic fibres. Most achievable (green), less achievable (orange), not achievable (red)

	Species name	Family	Clade	Part of the plant	Application in apparel					
Plant					Insu- lation	Natural fibre	Regene -rated	Syn- thetic		
Seed fibres										
Kapok	Ceiba pentandra	Malvaceae	Dicot	Seed						
Cattail	Typha	Typhaceae	Mono- cot	Seed						
Cuttun	species			Stem						
Bast fibres										
Нор	Humulus Iupulus	Cannaba- ceae	Dicot	Stem						
Nettle	Urtica dioica	Urtica- ceaea	Dicot	Stem						
Leaf fibres										
Abaca	Musa textilis	Musaceae	Mono- cot	Leaf						
Banana	Musa acuminata	Musaceae	Mono- cot	Leaf						
Pineapple	Ananas comosus	Bromelia- ceae	Mono- cot	Leaf						
Grass fibres										
Wheat	Triticum aestivum	Poaceae	Mono- cot	Stem						
Maize	Zea mays	Poaceae	Mono- cot	Stem						
Rice	Oryza sativa	Poaceae	Mono- cot	Stem						
Sugar cane	Saccharum officinarum	Poaceae	Mono- cot	Bagasse trash						
Miscan- thus	Miscanthus	Poaceae	Mono- cot	Whole plant						

Besides fibre crops, we also looked at the possibilities to use agri-food residues such as cereal grasses. Fibres from cereal grasses are in general not suitable for yarn production as they are too thick and short. However, the world's primary crop production comes not only from cereals like wheat, rice, maize, barley (58%), but also from legumes (belonging to the Fabaceae family) like beans, lentils, lupus, peas, peanut (27%) and

starch crops like potatoes and cassava (15 %)(Graham and Vance 2003). When the human diet is expected to shift more and more towards vegetable protein sources instead of animal ones, the production of legumes might increase. It is of interest to study these 'new' agri-food residues from legumes as a possible resource for textiles.

## 3.2.3 Regenerated cellulose (polymers)

In this paragraph, developments regarding regenerated cellulose fibres are described. Research topics include more sustainable processes for cellulose dissolution and regeneration. Some of these developments are mentioned here: ionic liquids, cellulose carbamate, and a cellulose dissolution system without solvents.

## Ionic liquids

Ionic liquids (ILs) are liquids made-up of salts (cations and anions). Certain ILs can dissolve cellulose without prior derivatization and offer a potentially more environmentally friendly alternative to traditional processes. The properties of ILs can be modified, tailored towards their application by selection of the specific cations and anions. For large scale applications, criteria like ease of production and recyclability are very important (Felgueiras et al. 2021).

Although this is a promising class of materials for textiles and other product categories, the processing, reuse and in some instances toxicity of the solvents and the high associated costs are often not comparable to that of the current state of the art (Mendes et al. 2021). However, a new class of regenerated fibres called Ioncell has been developed based on ILs that displays better mechanical properties than other regenerated cellulose fibres. Several garments have been produced, and proof-of-concept on pilot scale is foreseen in 2023-2024 (Ioncell 2022).

#### Cellulose carbamate

This technology is based on the formation of a cellulose derivative called cellulose carbamate by transformation of cellulose with urea. Cellulose carbamate is soluble and shapable in sodium hydroxide solution (Harlin 2019). It is patented under the name CarbaCell and is based on the synthesis of cellulose carbamate in xylene as transfer medium. The technical sequence from cellulose to fibre is similar to the viscose method (Klemm et al. 2005).

Advantage of the carbamate process is that cellulose carbamate is relatively stable at room temperature, permitting long time storage without quality loss; a disadvantage might be the use of xylene as organic solvent. Industrial tests have shown that cellulose

carbamate can be processed without any problems on viscose spinning machines (Klemm et al. 2005).

#### Solvent-free

A totally new approach is solvent-free processing of cellulose. Wood pulp can be transformed into a spinning-ready fibre suspension by mechanically refining without harmful chemistry (no dissolving, no regeneration). Upon spinning into filament, the suspension flows through a unique nozzle at a high pressure. The right kind of extrusion causes the fibrils to rotate and align with the flow to create a natural textile fibre. The fibre is then dried and collected, ready for spinning into yarn (Spinnova 2022).

#### 3.2.4 Bacterial cellulose (polymers)

Apart from plants, certain bacteria, algae and fungi produce cellulose as well. By variation of criteria like substrate, cultivation conditions, additives, and bacterial strain, it is possible to control the degree of polymerisation (DP), molar mass, molar mass distribution, and supramolecular structure of cellulose. Amongst the cellulose-forming bacteria, bacteria from the *Acetobacter* strains are very suitable to produce cellulose. *Acetobacter xylinus* species produce extracellular cellulose that is easily isolated as fibre material (Klemm et al. 2005).

Bacterial cellulose is different from plant-based cellulose, in terms of high DP (2000-8000), crystallinity (60-90%) and high purity (no association with hemicellulose, lignin or pectin). Another difference is the extremely high-water content; bacterial cellulose can store over 90% of its own weight of water. This high moisture content results from the microstructure of bacterial cellulose and affects both physical and mechanical properties.

Applicability of bacterial cellulose in the textile and apparel industry is gaining more attention (Fernandes et al. 2019, da Silva et al. 2021). However, its hydrophilicity and porous structure may pose problems for application in textiles (Provin et al. 2021). Bacterial cellulose has the disadvantage that it is time consuming and costly to produce. Pure media component costs, coupled with the time necessary to incubate the materials, likely mean that bacterial cellulose will continue to be used for high value products only (Harmon et al. 2020). Significant developments will need to be made for bacterial cellulose to challenge traditional cellulosic sources like cotton and wood.

### 3.2.5 Cellulose summary

Cellulose, being the most abundant natural polymer on earth, is biodegradable in most natural environments, renewable, affordable, and is applied in many applications. Plants are the main producers, but also microorganisms like bacteria can make cellulose. The chain length of cellulose (DP) is an important structural parameter and varies with origin and treatment of the material. For textiles, cellulose presents unique structural features and is the most important and widely available renewable resource.

Alternative and additional resources are required to alleviate the pressure on wood and cotton as main sources of cellulose pulp and cellulose fibres. These can originate from bast fibres, grasses (bamboo), or lignocellulosic agro-food residues. Technology needs to be adapted to extract cellulose from these non-wood sources/agrifood residues. Once a pure cellulose pulp is obtained, it can feed into existing cellulose dissolution processes like viscose, lyocell, or other new processes under development (ionic liquids, cellulose carbamate). In addition, recently developed technologies allow the production of filaments with good mechanical performance without dissolution or any complex chemical processes (Felgueiras et al. 2021). These fibres produced without solvents might be promising for cellulose-based textiles.

## 3.3 Polyesters

#### 3.3.1 Introduction

In many textile products, polyethylene terephthalate (PET) is the main polyester type that is referred to. However, the polyester family is much larger and more versatile, and offers more options for use in textiles.

This paragraph gives an overview of the various types of polyesters and alternatives for more circular polyesters for textile applications. It is divided in three parts: biobased aromatic polyesters (monomers), biobased aliphatic polyesters (monomers) and natural polyesters produced by bacteria (polymers).

### 3.3.2 Biobased aromatic polyesters (monomers)

PET is an aromatic polyester, i.e. a polyester with an aromatic group in the chain originating from the diacid terephthalic acid. This aromatic group gives rise to a higher thermal stability and enhanced mechanical performance compared to aliphatic polyesters (without an aromatic group), which is highly beneficial for textile products. In this section we explain the differences between partially biobased and fully biobased aromatic polyesters.

#### 3.3.2.1 Partially biobased aromatic polyesters: PET, PTT, PBT and PBAT

The combination of terephthalic acid and various diols results in polymers with different characteristics. Three examples include polytrimethylene terephthalate (PTT), polybutylene terephthalate (PBT) and polybutylene terephthalate adipate (PBAT) (see Figure 46). PTT is used as textile fibre in carpets and apparel, while PBT is more a technical polymer for specialty applications and less relevant for textile. PBAT is a more flexible and elastic aromatic polymer and is biodegradable in various environments such as soil. As a result, PBAT is often used as a biodegradable alternative for non-degradable PE films.

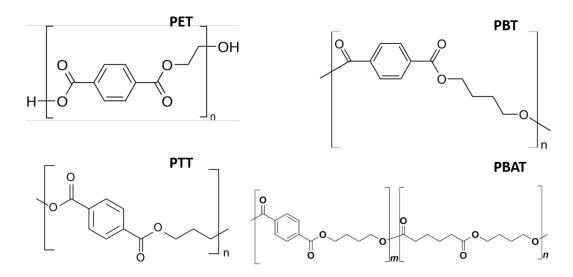


Figure 46: The aromatic polyester family: PET, PTT, PBT and PBAT

Biobased developments mainly focus on the production of the diol building blocks, as these monomers can readily be produced from biomass via e.g. fermentation of sugars:

- PET is partly biobased when the diol (1,2 ethanediol or ethylene glycol (EG)) comes from renewable resources. Global bioPET production capacity is estimated at 164 kton (European Bioplastics 2021).
- PTT is partly biobased when the diol (1,3 propanediol or PDO) is from renewable resources. Global bioPTT production capacity is estimated at 194 kton (European Bioplastics 2021).
- PBT is partly biobased when the diol (1,4 butanediol or BDO) is from renewable resources. BDO can be produced from renewable resources via various routes (Harmsen et al. 2014). BioPBT is not on a commercial scale to our knowledge.
- PBAT is partly biobased when the diol (1,4 butanediol) and adipic acid (1,4-butanedicarboxlyic acid) are made from renewable resources. Global biobased PBAT production capacity is estimated at 120 kton (Chinthapalli et al. 2019).

The biobased production of terephthalic acid is unfortunately less straightforward. Proposed routes to make biobased terephthalic acid are via chemical conversion of isobutanol, muconic acid, limonene or furfural, or thermochemical conversion of lignocellulosic biomass (Nakajima et al. 2017).

### 3.3.2.2 Fully biobased aromatic polyesters: PEF and PBF

Since it is complex to produce the terephthalate group via a biobased route, much efforts have been put in the development of alternative aromatic monomers. The most prominent example in this respect is 2,5-furandicarboxylic acid (2,5-FDCA), which has successfully been employed to produce PEF (polyethylene furandicarboxylate) and PBF (polybutylene furandicarboxylate).

PEF is the fully biobased alternative polymer for PET, where terephthalic acid is being replaced by 2,5-FDCA (see Figure 47). PEF is 100% biobased and is not called bioPEF as it has no fossil counterpart. PEF has good thermal and gas barrier properties, making it an interesting material for bottles. However, concerns were recently raised on its UV stability, which might impact the functional use in clothing (Maaskant and van Es 2021). It is expected that PEF will enter the market in 2023 (EuropeanBioplastics 2021). Replacing terephthalic acid by 2,5-FDCA as well as 1,2 ethanediol by (bio) 1,4-butanediol results in the polymer PBF, which is the fully biobased variant of PBT. Another variation is the use of 1,3-butandiol which yields PTF, the fully biobased variant of PTT.

Figure 47: The monomer 2,5- furandicarboxylic acid (2,5-FDCA) (left structure) and polymer PEF (polyethylene furandicarboxylate) (right structure).

#### 3.3.3 Aliphatic polyesters: PLA, PGA, and PBS (monomers)

Contrary to aromatic polyesters, aliphatic polyesters do not contain a carbon-based ring structure (e.g. benzene or furan ring). Aliphatic polyesters are often considered to be more susceptible to biodegradation than aromatic polyesters, as the aromatic structures may hinder biodegradation (Szostak-Kotowa 2004). In this section three aliphatic polyesters are being discussed: PLA, PGA and PBS (Figure 48).

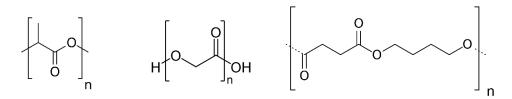


Figure 48: Aliphatic polyesters PLA (left), PGA (middle) and PBS (right)

## PLA (polylactic acid)

PLA is a biobased and biodegradable polyester with a production volume of 400 kt/year. This is partly driven by the fact that PLA can be produced very efficiently from various types of biobased feedstock. Main industrial PLA production facilities are located in the United States and Thailand, where the polymer is made from corn starch and sugar cane feedstocks respectively. This feedstock is processed into lactide (cyclic diester of lactic acid) which is subsequently synthesized into high molecular polymers using ring-opening polymerization.

Benefits of PLA compared to other biobased polymers are its relatively low costs, commercial availability of various grades, transparency, and a high level of gloss and stiffness. Disadvantages of PLA are its moderate heat resistance and toughness. PLA is mainly used in packaging materials (foils, bottles) and to a lesser extent in textiles (apparel and home). A distinct feature of PLA is that it is biodegradable under industrial composting conditions and to a much lesser extent in the open environment. This could be an advantage for the use of biobased and biodegradable aliphatic polyesters in durable textile applications.

## PGA (polyglycolic acid)

PGA is produced from glycolic acid by means of polycondensation, or by ring-opening polymerization comparable to PLA. The industrial process to produce high molecular weight PGA cost-effectively has only matured in the past decade. As a result, this material is now approaching the biodegradable plastic market. At this point in time, glycolic acid monomer is not yet produced biobased, but it is anticipated that this will change in the upcoming decades.

PGA is a very rigid and high strength polymer with outstanding gas and moisture barrier properties, and is reported to biodegrade in most natural environments (e.g. soil and sea). PGA can be processed into fibrous structures, but applicability is limited due to its hydrolytic instability. By combining it with other polymers such as PLA into a copolymer, the properties can be modified and thereby its applicability improved. Its fast

biodegradation rate in various environments makes that PGA-based fibres are interesting for medical applications, but less interesting for (durable) textile applications like apparel (Montes de Oca and Ward 2006, Jem and Tan 2020).

### PBS (polybutylene succinate) and PBSA (polybutylene succinate adipate)

PBS is a non-transparent polyester that is produced by condensation polymerisation of succinic acid (1,4-butanediacid) and BDO (1,4-butanediol). PBS is biodegradable in (industrial) composting environments and soil. PBS is an interesting biobased and biodegradable alternative for polyolefins as its mechanical and thermal properties are in the same range as that of polyethylene (PE) and polypropylene (PP). PBS is currently being produced mainly from fossil sources, but both monomers in PBS can be produced by means of fermentation of sugars from biomass to create a 100% biobased material.

Polybutylene succinate adipate (PBSA) is a co-polymer that is produced in a similar way as PBS, by adding adipic acid to the polymerization process. Upon increasing adipate content, the polymer becomes more flexible and tough, and the biodegradation rate in (industrial) composting and soil environments is accelerated.

Both PBS and PBSA can be processed into fibrous structures via melt spinning, as they possess sufficient melt strength at elevated temperatures.

## 3.3.4 Natural polyesters from bacteria (polymers)

Asides from synthetic polyesters based on biobased monomers, there is another class of polyesters which are produced within bacteria and thus from natural polymers. The industrially most relevant family of these natural polymers are the poly(hydroxyalkanoates) (PHAs).

PHAs are produced by specific bacterial cultures and, based on the microorganism type and the feedstock (typically fatty acids), different types of polymers with a wide range of thermal and mechanical properties can be produced. Chain length of the PHAs determines the flexibility of the polymer; short chains provide rigid polymers, while long chains result in more flexible polymers (Nakajima et al. 2017). The PHAs are produced in granular form as an energy reserve within the bacterial cells. An extraction process is required to isolate these natural polymers, making the overall production process cost-intensive.

PHAs are unique in their biodegradation behaviour in the open environment as they are currently the only biobased polymers that are certified to biodegrade under both soil and marine conditions. For textile applications this could be a disadvantage as it may result in less durable clothing (Nakajima et al. 2017). Another anticipated disadvantage for use in textiles is the low melt strength that PHAs typically possess, although PHA

polymers with a higher melt strength are expected to reach the market in the upcoming decade (Molenveld et al. 2022).

### 3.3.5 Polyester summary

Polyesters are among the most promising families of polymers based on renewable resources, as many of the biobased diacids and diols are relatively easy to produce by fermentation of sugars, and a variety of properties can be created. One possible production route of polyesters based on glucose is shown in Figure 49.

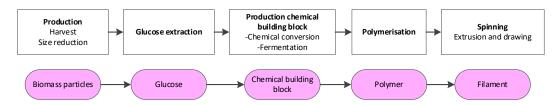


Figure 49: Production route of biobased monomers for synthetic fibres like polyesters

Aromatic polyesters based on terephthalic acid have properties that are excellent for textile applications (strong, thermally stable). PET and PTT are examples of aromatic polyesters that are applied in textiles on large scale. Unfortunately, these polyesters can up till now only be partially biobased as biobased terephthalic acid production is quite challenging. In addition, most of these aromatic polyesters are resistant to biodegradation. This may pose problems in the form of microfibres that accumulate in nature. PBAT is an example of an aromatic polyester that is biodegradable in various environments (composting and soil) but the presence of a high amount of aliphatic monomers yields mechanical properties that are highly different than that of PET and PTT. Therefore, PBAT is not considered to be a direct alternative for the current state of the art polyester fibres in textiles. This might be different for PEF as its molecular structure and thereby its properties are a much better match with PET and PTT. This new aromatic polyester is expected to enter the market in 2023. It is regarded as a highly relevant material for the packaging of drinks, food and non-food products, but its applicability for textiles is as yet unknown. Initial results show that the biodegradation rate of PEF in soil is expected to be higher than that of PET. However, as these studies are performed on weathered materials and high temperatures it cannot be stated that this polymer can contribute to the prevention of microplastic accumulation in the environment (de Jong et al. 2022).

Aliphatic polyesters are less strong and less thermally stable than aromatic ones. On the other hand, they are more susceptible to biodegradation (under the proper conditions) and fully biobased production is less complex. PLA and PBS are examples of aliphatic polyesters that are applied in textiles, but on a much smaller scale than PET or PTT. Another example are PHAs, a class of naturally produced polymers by a variety of microorganisms. It is estimated that production capacities will increase tenfold in the next years. However, applicability in textiles is uncertain due to less favourable properties of PHAs in relation to textile processing and applications.

Table 5 presents an overview of all polyesters discussed in this paragraph, and their main characteristics. Most of the polyesters applied in textiles are drop-ins\*\* based on terephthalic acid. The only exception forms aliphatic PLA, fully biobased and biodegradable, but with less attractive properties for textiles.

Table 5: Overview of polyesters relevant to textiles

Polymer	Fully biobased	Aromatic	Aliphatic	Applied in textile*
PET	In development	X		Yes
PTT	In development	X		Yes
PBT	In development	X		No
PBAT	In development	X		Unknown
PEF	Yes	X		No
PLA	Yes		X	Yes
PBS	In development		X	Yes
PHAs	Yes		X	No

<sup>\*(</sup>European Bioplastics 2021)

\*\* Biobased versions of existing petrochemicals which have established markets.

## 3.4 Polyamides

### 3.4.1 Introduction

Polyamides are the only class of polymers that are produced as synthetic polymers (nylons), natural polymers (proteins), as well as natural fibres (wool, silk). This makes polyamides the most versatile one of all polymers discussed in this chapter.

This paragraph describes some of the initiatives on biobased alternatives for synthetic polyamides nylon 6 and nylon 6,6. On the topic of natural polymers within the group of polyamides, one example of regenerated proteins is described (in analogy to regenerated cellulose). Developments on natural fibres (from animals) are very limited and this topic is not further discussed.

### 3.4.2 Biobased synthetic polyamides (monomers)

Synthetic polyamides are produced via the polycondensation of a diacid and a diamine, or ring-opening polymerization of a lactam. Biobased alternatives for the diamines are more challenging than the diacids.

Part of the diacids is already biobased by using vegetable castor oil from the tropical plant *Ricinus communis*. Castor oil consists for a large part (85%) of ricinoleic acid, an unsaturated fatty acid (C18) with one hydroxyl group. Ricinoleic acid is the raw material for the diacids required to produce the polyamides PA4,10, PA 6,10, PA10,10 and PA11<sup>+†</sup>.

Global production volume of all biobased polyamide fibres is estimated at 20 kton/year, around 0.4% of the polyamide fibre market (Textile Exchange 2021). This volume is mainly due to the use of castor oil. For textiles especially nylon 6 (PA6) and nylon 6,6 (PA6,6) are relevant. It is less likely that these polyamides are produced from castor oil and other biobased routes are being developed.

### Nylon 6,6

Nylon 6,6 is a polyamide that is produced by the polymerisation of adipic acid and hexanediamine. Several routes are being developed to make these building blocks from renewable materials (Harmsen et al. 2014):

 Adipic acid is an important bulk chemical with applications in food and non-food products. Biobased processes are possible via fermentation of sugars or chemical conversions. A yeast has been developed that can produce adipic acid from

 $<sup>^{\</sup>dagger\dagger}$  Polyamides are named after the number of carbon atoms of each of the monomers, in which the first number corresponds to the diamine and the second with the diacid.

- vegetable oils and sugars in a single step, but also chemical-catalytic conversion routes are under development.
- Hexanediamine can be produced by chemical-catalytic conversion of adipic acid from glucose. In view of the developments for biobased propylene and butadiene, there are also opportunities to produce biobased hexanediamine by fermentative routes.

## Nylon 6

Nylon 6 is a polyamide produced by ring-opening polymerization of caprolactam. Biobased processes to produce caprolactam via fermentation of sugars are being developed (Genomatica 2020).

## 3.4.3 Regenerated proteins (polymers)

Comparable to regenerated man-made cellulose fibres (MMCFs), it is also possible to make man-made regenerated protein fibres (RPFs). Development of these types of fibres was accelerated during World War II, due to limited availability of resources for uniforms like wool. However, RPFs were quickly forgotten when the war ended, and fossil-based fibres took over.

Research for RPFs is gaining attention again, to make better use of protein-rich waste streams from the food or feed industry. In a review by Stenton et al., the potential for RPFs within a circular economy is described (Stenton et al. 2021). Table 6 shows an overview of RPFs on the market in the past, and the resources that were used to make the fibres.

Table 6: Overview of Regenerated Protein Fibre production (Stenton et al. 2021)

Product name	Company	Resource	year
Lanital	Snia Viscosa	Milk casein	1936
Fibrolan	Courtaulds	Milk casein	Unknown
Merinova	Snia Viscosa	Milk casein	1945
Ardil	ICI	Oilseed Peanut	1960
Vicara	American Virginia Carolina Corp	Corn zein	1960

For the manufacturing of casein fibres, a range of undesirable and potentially harmful chemicals were used throughout production. Formaldehyde was used as a crosslinking agent to improve the tensile strength, and high concentrations of sulphuric acid were used in coagulation of the RPFs. Still, the RPFs suffered from mechanical shortcomings, particularly the lack of tensile strength, especially when wet. To improve the tensile strength of RPFs, a water-soluble polymer such as polyvinyl alcohol (PVA) can be added.

This innovation led to the first industrially produced fibres from soybean proteins and PVA (Rijavec and Zupin 2011).

It has been demonstrated that globular (food) proteins like casein, soy and zein can be used to make RPFs. Dilute alkali solutions are applied to swell and dissolve the protein, followed by fibre extrusion into an acid solution. However, when other protein sources like feather keratin and wheat gluten are used, the situation is more complex (Poole et al. 2009). Thus, when production of fibres from protein waste sources is desired, new research in this field is required.

## 3.4.4 Polyamides summary

Polyamides are important polymers for textile applications. It is the combination of biobased options and attractive properties that make polyamides very relevant for future textiles. All intermediate structures (fibres, polymers, monomers) can in theory be provided by biobased options with the following remarks:

- Animals supply natural polyamide fibres in the form of wool or silk. Properties of animal fibres are excellent for apparel but not attractive for vegetarian/vegan people, or people allergic for wool.
- Natural polyamide polymers can be found in food, but also in residual waste streams like keratins from chicken feathers or animal hairs. More research is required to allow for the adequate processing of these proteins into textile fibres and to obtain fibre properties (especially wet strength) that meet the requirements of commercial textiles.
- Monomers for polyamides for textiles can be produced from vegetable oil or sugars.
   Although many synthetic polyamides are already partly biobased, the polyamides relevant for textiles are often still made from fossil resources as biobased alternatives are more expensive. For the diacids there are various options, but the diamine monomers are more challenging.

# 3.5 Polyurethanes

### 3.5.1 Introduction

An increasing amount of textile products contain elastane fibres to improve the elasticity of the fabric. These fossil-based, synthetic fibres are typically classified as polyurethanes. (Biobased) alternatives for elastane are described in this section.

#### 3.5.2 Biobased alternatives for elastane

Elastomers or elastic polymers are highly complex polymers, and some background knowledge of the production route is required to understand the approach towards biobased alternatives. Here it is described for one of these elastomers, i.e. elastane.

Elastane is different from other polyurethanes in that they are 'segmented' polyurethanes. They are block copolymers with a molecular structure with alternating 'soft' and 'hard' segments (-S-H-S-H-), giving elastane its characteristic properties. The soft blocks are coiled in their unstressed balance state. When force is applied, the soft blocks uncoil, become stretched and will be aligned approximately along the fibre's axis. When the stress is released, the soft blocks relax back into their balance state. The hard blocks, on the other hand, can form very strong inter-chain bonds with other hard blocks from a neighbouring chain. These strong bonds work as crosslinks in a netted structure that prevent slipping of the molecular structure in the stretched state, and make sure that the fibre relaxes back into its original shape. The bonds between the hard segments of neighbouring chains are formed mainly by non-covalent hydrogen bonds. This implies that, contrary to covalently cross-linked rubbers, elastane fibres can be dissolved and/or melted (Mather and Wardman 2015), which can be advantageous in recycling.

Production of block copolymers, the soft and hard segments in elastane, is based on isocyanate chemistry. This synthesis is based on three main stages (Mather and Wardman 2015):

- 1. Formation of a macroglycol prepolymer (soft segment), which is a polyether or polyester
- 2. Reaction of this macroglycol with aromatic diisocyanate, to result in a macrodiisocyanate prepolymer
- 3. Formation of segmented polyurethane by reaction of the prepolymer with a diamine or a diol

A large variety of chemicals can be used to produce the soft and hard segments of elastane. Especially the soft segments can be made from biobased alternatives, while the options for the hard segments are limited. This makes that elastane can only be partially biobased, and not fully biobased.

#### 3.5.3 Other alternatives for elastane

Natural elastic polymers are natural rubber from the rubber tree (*Hevea brasiliensis*). Synthetic elastic fibres were developed during World War/WWII as alternative to natural and synthetic rubbers. Much of the early work on the development of elastomers was based on new types of polyamides. However, despite intensive research efforts, products based on polyamides never achieved commercial success because they would not withstand repeated stretching cycles required for elastane applications (Mather and Wardman 2015). Many thermoplastic elastane alternatives have been developed since, but most are unsatisfactory in terms of elastic properties.

Of all the alternatives available, natural rubber is still the best candidate, although the properties of polyurethane fibres remain superior. Polyurethane fibres are approximately twice as strong as rubber fibres. As a result, less material is required in a fabric blend to give the desired elastic performance (Mather and Wardman 2015). Natural rubber can still provide elastic properties but will never reach the excellent properties synthetic polyurethane fibres can provide.

#### 3.6 Discussion

In this chapter we reviewed the options available to produce textile fibres from nonfossil resources. This chapter was aligned along the four major polymer types relevant for textile applications, i.e. cellulose, polyesters, polyamides, and polyurethanes. Per polymer type the following can be concluded:

- Cellulose forms already a major part of the textile fibre mix, and this is expected to
  increase. Cellulose is abundantly available in nature, is not a food source for humans,
  and is very suitable for textiles. For cellulose <u>fibres</u>, alternatives are needed to
  alleviate the pressure on cotton. For cellulose <u>polymers</u>, alternatives are needed to
  alleviate the pressure on wood. Cellulose pulp production then would need to be
  adjusted to non-wood resources.
- Polyester forms the bulk of the textile fibres, relying heavily on the aromatic polyester PET. Aromatic structures give attractive properties to textile fibres, but are more complex to produce from biobased feedstock. There are fully biobased aliphatic (PLA and PBS) alternatives available, but they all have in common that their mechanical and thermal properties required for textile applications are inferior to aromatic polyesters like PET and PTT.
- Polyamides are the only class of polymers that can be found in synthetic polymers, in natural polymers, as well as in natural fibres. Of the synthetic polymers, only nylon 6 and nylon 6,6 have relevant properties for apparel. Monomers of nylon can be made biobased, but the diamines and lactams required are challenging to produce. Proteins can also be regenerated into textile fibres (like cellulose) but many issues need to be solved, like the (wet) strength. Natural fibres like wool and silk belong to the upper class of textile fibres due to their excellent properties. They will remain a niche market.
- Elastomeric polyurethanes are relevant in textiles because of their elastic properties. These elastomers like elastane are highly complex (co)polymers containing aromatic structures, where only the soft segments have biobased options.

Now that we have a complete overview of the fibres and polymers that can play a significant role in future circular textiles, the next step is to connect these textile fibres to the renewable carbon sources in the most effective way, which is further described in the next chapter. To facilitate this process, a table was constructed to aggregate all the information gathered in this chapter. For each type of polymer, the corresponding textile fibre, intermediate structure, and biobased options are presented (Table 7). The colours help to pinpoint the main issues, where green is obviously better than red.

Table 7 shows that biobased options are not always feasible, especially when the polymers are fossil-based by origin and/or contain aromatic chemicals. For these cases, other renewable carbon sources such as  $CO_2$  or recycled content may be better alternatives than biomass. This discussion is further explained in the next chapter.

Table 7: Compilation of main polymer types: biobased options and applicability for textiles.

Textile fibre	Intermediate	Examples	Fossil	Biobased	Applicable for
category	structure		based	options	apparel
	Poly	saccharides (Ce	llulose)		
<i>Natural</i> Plant-based	Fibre	Cotton, linen	No	Yes	Yes
<i>Man-made</i> Regenerated	Polymer	Viscose, Iyocel	No	Yes	Yes
<i>Man-made</i> Bacterial	Polymer	Bacterial cellulose	No	Yes	Unclear
		Polyesters			
Man-made Synthetic aromatic drop-in	Monomer	PET, PTT	Yes	Partial	Yes
Man-made Synthetic aromatic new polymer	Monomer	PEF, PBF	No	Yes	Unclear
Man-made Synthetic aliphatic	Monomer	PLA, PBS	No	Yes	Unclear
<i>Man-made</i> Bacterial	Polymer	PHAs	No	Yes	Unclear
Polyamides					
Man-made Synthetic aliphatic	Monomer	Nylons	Yes	Partial	Yes
<i>Man-made</i> Regenerated	Polymer	RPFs	No	Yes	Unclear
<i>Natural</i> Animal-based	Fibre	Wool, silk	No	Yes	Yes
Polyurethanes					
Man-made Synthetic aromatic	Monomer	Elastane	Yes	Partial	Yes

# 4 Carbon sources for circular textile fibres

#### 4.1 Introduction

The previous chapters described the rationale of renewable feedstock for fibres in view of the upcoming phasing out of fossil feedstock, and the main consequences for the transition from contemporary to future circular textiles. The available circular options to produce the required intermediate structures (monomers, polymers, fibres) for textiles, based on the most important polymers for textiles (polysaccharides (cellulose), polyesters, polyamides, polyurethanes) are described. This chapter combines these intermediate structures to the only three available carbon sources from which these structures can be produced: biomass, recycled content and CO<sub>2</sub>.

#### 4.2 Biomass

Biomass is the most versatile carbon source for carbon-based materials, as it includes all kinds of nature-based materials we can use in our fossil-free society. This publication has shown that for contemporary textiles we heavily rely on fossil resources, and when renewable resources are used, it is limited to a small number of specific plants (cotton, trees) or animals (sheep). However, to achieve the goal of fossil-free textiles it is expected that we will need a much broader selection of resources provided by nature, i.e. other plants, animals but also other totally different organisms.

One approach could be to search for organisms that are grouped based on shared characteristics. For plants, we applied taxonomy in Table 3 and Table 4 to arrange various plant species in families and to search for new plants with similar preferential characteristics. But besides the well-known plants and animals, there are much more living organisms less familiar to us but potentially interesting as providers of carbon-based materials.

The 'Tree of living organisms' shown in Figure 50 might be an inspiration for this. Animals and plants we know for textiles, but less familiar are fungi. Fungi are an example of organisms that are highly interesting as producers of a new type of material called mycelium. Recent developments around fungal materials (Haneef et al. 2017, Karana et al. 2018, Appels et al. 2019) illustrate how fast things can evolve, and maybe also towards textiles.

In the context of this publication, we define biomass as 'all carbon-based fibres, polymers and monomers created by living organisms', so much broader than the well-known examples discussed in chapter 2 on contemporary textile fibres.

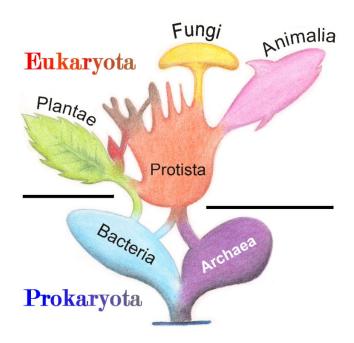


Figure 50: Tree of living organisms (created by Maulucioni y Doridí, 2013). Eukaryotes are organisms whose cells have a nucleus; prokaryotes are single celled organisms without a nucleus.

For future circular textiles we see the following opportunities for fibre, polymer and monomer production by biomass:

• Natural <u>fibres</u> are mainly produced by animals and plants, but production volume and number of plant species applicable for textiles can possibly be enlarged, with some examples shown in paragraph 3.2. Regarding animal fibres, it is not foreseen that volumes will grow much given the environmental burden of animals and the trend for plant-based products. However, we could make better use of wool when it is a residue stream from the meat production. With steep rising gas prices, heating and insulation of buildings is getting more expensive, and wool in the form of insulation material, blanket or sweater can help to stay warm when it is colder. New developments have shown that besides animals and plants, also fungi can produce fibres (mycelium) that might be applicable in textile.

- Natural <u>polymers</u> for regenerated fibres can be produced by plants, animals, fungi, but also by bacteria. In the previous chapter, examples are shown for polysaccharides and polyamides. Cellulose (polysaccharide) is and will remain one of the most important natural polymers for textiles. Alternative sources of cellulose (for example agri-food residues) might be used to alleviate the pressure on forestry as main supplier of cellulose pulp. Another interesting source are proteins (polyamide) for regenerated protein fibres. These protein polymers can originate from plant residues or animal residues.
- Monomers for synthetic fibres (acids, alcohols, amines) can be produced by animals, plants, fungi, and bacteria, but maybe also by other living organisms and possibilities are endless. For now, mainly sugar or starch-rich food resources are used to feed bacteria that produce monomers, but also all kinds of agri-food residues might be used.

# 4.3 Recycled content

For the future feedstock that comes from recycled content, resources will come from garments (post-consumer) or residues from the production in the factories in the form of yarns or fabrics (post-industrial). These residues are not yet optimised for recycling, and there are distinct differences between them. The post-consumer stream forms the most challenging one of the two since it is typically not well-defined, contaminated and contains fibres of lower quality due to washing and wearing. Textile recycling can be performed toward each 'level of level of assembly': fibres, polymers, or monomers. Whether a garment is suitable for fibre, polymer or monomer recycling is determined in large part by the fibre composition and the chemical structure of the polymers that make up the fibres (Harmsen et al. 2021).

The overall aim of the recycling process is to keep the structure of the materials intact as much as possible and to minimise processing: recycling towards fibres is most preferable, followed by polymers and subsequently monomers:

Fibre recycling implies the preservation of the fibres after the disintegration of the fabric by mechanical means (cutting, tearing, shredding, carding). Usually this is done with natural staple fibres like cotton and wool. Fibre length is in this process an important parameter, as the fibre length generally is reduced as an unwanted side effect, causing quality loss. This problem can be overcome by blending with virgin renewable resources. An example of a fabric composed of recycled content is shown in Figure 51.



Figure 51: Example of a herringbone fabric, composed of recycled cotton, viscose, and wool. Garment and SEM-pictures of fabric.

Polymer recycling includes the disassembly of the fibrous structure while the
polymers remain intact. This is mostly done by physical means (melting or dissolving
of the polymers) using comparable processes as for the recycling of plastic
packaging. Also here, virgin renewable resources are required to overcome quality
loss (for example due to lower degree of polymerisation). Recycling of polymers is
very suitable for cellulose-based textile fibres (cotton, linen, viscose, lyocell) to yield

- regenerated cellulose fibres (viscose, lyocell) and for synthetic thermoplastic fibres (PET, PTT, nylons, PLA, PBS).
- Monomer recycling implies that the polymers are broken down to their (original) chemical building blocks. This is mostly done by chemical methods, for example (enzymatic) hydrolysis of polycondensation polymers like polyesters and polyamides. In contrast to polymer and fibre recycling, for recycling to monomers there is no need for addition of virgin resources, as (re)starting from monomer level will always result in virgin quality fibres. However, the requirement of extra process steps and the need for effective separation and reclaiming the recycled monomers from pollutants are clear disadvantage for this route. Recycling to monomers is highly suitable for man-made synthetic fibres (condensation polymers) and less achievable for natural polymers.

#### 4.4 CO<sub>2</sub>

 $CO_2$  has become synonymous with climate change, as it is one of the greenhouse gases that induces a global temperature rise. But  $CO_2$  is also a commodity which is extensively used in industry, especially in the food sector. It is presently produced, for instance, as a by-product of the production of ammonia by the Haber-Bosch process. In this process, atmospheric nitrogen ( $N_2$ ) is converted to ammonia ( $N_3$ ) by a reaction with hydrogen ( $N_2$ ) under high pressure and temperature. The major source of hydrogen is methane ( $CH_4$ ) from natural gas (a fossil resource), and the carbon is released in the form of  $CO_2$ . In a fossil-free society, this source of  $CO_2$  is only available when biogas is used, and also other means of  $CO_2$  capturing are required.

 $CO_2$  can be distilled from the air, but that method is expensive and highly inefficient as the concentration of  $CO_2$  in the atmosphere is very low. Other more promising technologies focus on removing and storing  $CO_2$  from sources with higher concentrations, so-called point sources. These can be found at the exhaust of incineration plants, at biomass fired power stations, at certain food producing plants using fermentation, or at small scale manure digesters. In fact, all materials containing carbon are a potential source of  $CO_2$ , including residues coming from biomass or recycle streams that cannot be used through any other means. This makes the  $CO_2$  route the most robust one, as requirements to input streams (residues) are rather limited. Disadvantage of this route is the large amount of energy required for this type of processes.

Once the  $CO_2$  is concentrated it can be used as feedstock for thermochemical or electrochemical processes, or biochemical processes where bacteria are applied to produce small molecules. These small molecules can be converted into monomers to produce the polymers and in the end synthetic fibres, of which some possible routes are

illustrated schematically in Figure 52. These monomers, like ethanol or methanol, can then be further converted into synthetic textile fibres based on condensation polymers like polyesters and polyamides.

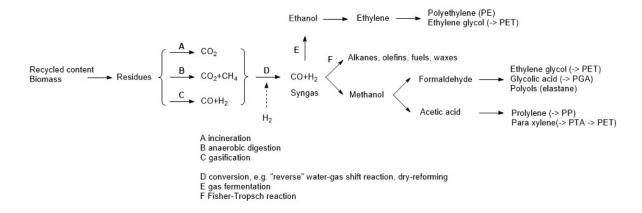


Figure 52: Schematic illustration of the routes from  $CO_2$  to synthetic polymers (by Daan van Es)

### 4.5 Discussion

All the information from the previous paragraphs is compiled in a schematic figure. Figure 53 shows, for future circular textile fibres, the relation between required basic input, carbon sources, the intermediate structures, polymer categories and textile fibre categories. This scheme is much more complex than the scheme for contemporary textiles shown in Figure 37, as now we expect to have multiple renewable resources at our disposal. For the three basic categories of textile fibres (i.e. natural, regenerated and synthetic) we have the following circular options:

- For <u>natural</u> fibres, biomass will be the main carbon source, supplemented with recycled content. Sustainable production of natural fibres will form a major challenge, in combination with land use. Production of natural fibres will have to compete with other applications that also need biomass like food, feed and building materials
- For <u>regenerated</u> fibres, natural polymers (e.g. cellulose, proteins) are required and these can only be produced by living organisms. Main question here is, which organism produces the desired polymer in the most optimal way. Looking for example to cellulose, there are multiple sources, as many organisms produce cellulose. For textile applications, plants will remain the largest producers of

- cellulosic biomass, but which plant on which location is a relevant question. In addition, natural polymers for regenerated fibres can also be sourced from recycled content, for example recycled cotton for viscose.
- For <u>synthetic</u> fibres, all three carbon sources (biomass, recycled content and CO<sub>2</sub>) can be utilized, and fossil resources can be phased out. Main question here is, which source and related production route is the most optimal in terms of technical feasibility, economic feasibility, and sustainability. This needs to be evaluated for each type of monomer required. For example, terephthalic acid, one of the two monomers to produce PET, is difficult to produce from biomass, so recycled content or CO<sub>2</sub> might be a better option. Another example is lactic acid for PLA, this can be produced very efficiently from sugars produced by plants, making other (less developed) routes less attractive to pursue.

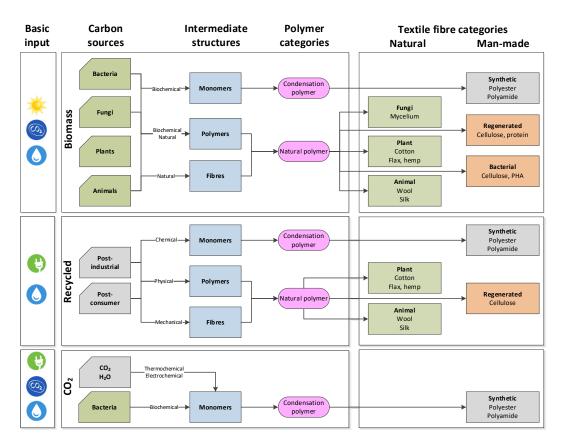


Figure 53: From renewable carbon to future circular textile fibres.

# 5 Concluding remarks and recommendations

## 5.1 Concluding remarks

The purpose of this publication is to describe the options we have to produce circular textiles from renewable carbon sources, and the challenges that lie ahead to reach this goal. Over the last decades, the textile industry has experienced a transition from using renewable, biobased feedstocks from plants and animals to fossil-based alternatives with the rise of polyester (PET or PES) and elastane as the two most prominent examples. As a result, the current fibre production is dominated by synthetic fibres from fossil resources, and the task to change back from fossil to renewable is huge. All kinds of new functionalities are introduced over the last decades, making a switch back without compromising on product performance nearly impossible.

But maybe this is not necessary, as we have shown that we have several options to produce circular textiles from renewable carbon sources. However, this is not a free pass to keep on producing the volumes we are used to. Circular fashion starts with reducing and reusing, and with quality over quantity. The true environmental impact of a garment is not only determined by the production phase, but also by the use phase and the end-of-life phase, and lifespan of a garment becomes an important parameter.

Finally, this last chapter explains very briefly why functionality ('a quality of being suited to serve a purpose well') of a textile fibre is so important for the transition from contemporary to circular textiles. This chapter is finalised with some recommendations for further research.

### 5.2 Functionality versus new carbon sources

There are two ways of addressing this transition from fossil to circular fibres: starting from a desired functionality (at the end of the value chain) or starting from (new) carbon resources (at the beginning of the value chain). Below there are some examples to illustrate this:

### Functionality

The rise of PET and elastane in apparel is above all due to their attractive properties like strength, thermal stability, easy care and stretch. Without this appealing functionality, combined with low prices and ability to produce large volumes, their growth would have been much less.

#### New carbon sources

- Some companies are striving to make their fossil-based synthetic polymers (partly) from biobased content (thereby maintaining the same material properties), but these developments are very slow. Of all synthetic polymers applied in textiles, in 2020 only 241 kton is biobased (European Bioplastics 2021), which is 0.35 wt% of the global production volume (68.200 kton).
- Newly developed polymers include fully biobased PLA (aliphatic polyester) and PEF (aromatic polyester). PLA is already on the market for many years with a current production volume of 400 kton (European Bioplastics 2021), but its applicability in apparel is limited due to less favourable properties (strength, heat resistance). Another new polymer is PEF that is expected to enter the market in 2023. Since PEF is said to have comparable properties to PET, it might be an interesting fibre for textiles.
- Bast fibres other than flax are gaining attention as well, with hemp as a good example. Although hemp has excellent credentials in sustainable production and can be used to make textile, its properties may be less attractive for apparel (strong, stiff, coarse fibre, fabric wrinkles badly). In combination with high prices (production volumes are very small), hemp is attractive to a small group of people but not (yet) for a large public. On the other hand, for some apparel applications like jeans, strong and stiff can be very attractive.

These examples illustrate how important functionality is. For the transition from fossil to renewable textile fibres, (new) fibres should be developed based on the sustainable carbon resources that are available. These new fibres should be designed with a set of key properties in mind. For apparel, these are for example softness, strength (under dry and wet conditions), thermal stability (ironing), limited wrinkling (after washing). In addition, fibres and fabrics should be recyclable and not release harmful chemicals or microplastics into the environment.

### **5.3** Emerging questions for further research

From this study three emerging research questions were formulated for the three basic categories of textile fibres:

1. Which plant, animal or fungus is suitable for sustainable production of <u>natural</u> <u>fibres</u>?

Natural fibres that have proven their suitability as textile fibres, likely will remain highly important for the textile industry. There are no alternatives for wool and cotton that display the same properties and improving the sustainability of the production of these

fibres is an important theme for the future. A better use of lower wool qualities (unfit for apparel) should be encouraged, for example as insulation material. Alternative plant-based fibres from bast, leaf or grass are required to alleviate the pressure on cotton as main source of plant-based fibres. For many of these fibres, production volumes must be increased, and production must become more economically attractive while keeping low environmental impact. Completely new natural materials/fibres like mycelium should be investigated for potential use in the textile industry.

To improve applicability in apparel, fibres with less good qualities/functionalities could be blended with other fibres, thereby still complying with design-for-recycling principles. Otherwise, applicability in home or technical applications, as a biobased alternative for fossil-based materials could be assessed.

### 2. How do we source cellulose for <u>regenerated fibres</u>?

Forestry is the main supplier of wood to produce cellulose pulp, the raw material that is used to make regenerated textile fibres. This will continue in the future and production volume might even grow, but also other cellulose sources could be used to supply the cellulose pulp. Two possible streams are rated for their suitability in Table 8, i.e. agrifood residues and recycled content. This exercise can be expanded with other sources to highlight the weak spots that need more research.

Table 8: Suitability of agri-food residues and recycled content as alternative sources of cellulose for textile applications

	A	gri-food residues	Recycled content			
Aspect	Rating	Explanation	Rating	Explanation		
Technical feasibility	Moderate to low	Purity and/or DP is too low	Good	Provided cleaning is sufficient and degradation during reprocessing is limited to a minimum.		
Economic feasibility	Moderate	Cellulose extraction is more complicated and thus more expensive	Moderate	Processing is costly, especially purification		
Sustainability	Excellent	Underutilized biomass is used, alleviates pressure on forestry	Excellent	Utilization of recycle streams		
Security of supply	Moderate	Competing uses (soil quality, feed, energy)	Low	Especially for post-consumer streams		
End-of-life options	Excellent	Recycling options are good and degradable by nature	Good	Recycling options are good and degradable by nature		

# 3. Which <u>synthetic polymers</u> are future proof?

Synthetic fibres provided new functionalities to apparel when they were introduced into the market many years ago. This publication has shown the possibilities to make biobased alternatives, or the design of completely new polymers with similar characteristics.

For the main synthetic textile fibres used today, the most obvious renewable carbon sources and other relevant aspects are listed in Table 9. Largest concerns are with PET, with limited biobased options, present high volume, and low costs. PET can very well be recycled but is presently mainly recycled from packaging materials like bottles by physical recycling methods, while post-consumer textiles are more fit for chemical recycling to monomers. Other concerns are with elastic polymers like elastane. They are added in small amounts to fabric and are considered a contaminant in recycling. We foresee that circular synthetic fibres will remain a major part of the textile fibre mix. Main question is which type of polyester, polyamide or polyurethane is future proof?

Table 9: Renewable carbon options for main synthetic textile fibres

Aspect	PET, PTT		Nylon 6	Nylon 6,6		Elastane	
Building block	Terephthalic acid	Ethylene diol, propane diol	Caprolac- tam	Adipic acid	Hexane diamine	Soft blocks	Hard blocks
Preferred carbon source	1 Recycled 2 CO <sub>2</sub>	1 Biomass 2 Recycled	1 Recycled 2 Biomass	Biomass	Biomass	1 Biomass 2 Recycled	Un- known
Technical feasibility	Moderate	High	High	Mode- rate	Low	High	Very low
Economic feasibility	Moderate	Moderate	Moderate	Low	Low	Moderate	Very low
Security of supply	High	High	Moderate	Mode- rate	Low	High	Very low
Biodegrada- ble	No		No	No		No	
End-of-life options	Recyclable		Recyclable	Unknown		Is considered contaminant in textile recycling	
Main issues	Security of supply for TPA		Security of supply	Unknown		Lack of circular options for hard blocks	
	Microplastics						

We hope that this publication contributes to a fruitful discussion on the major underexposed challenges we face concerning textiles, but that for sure will impact the way we produce and wear our clothes.

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# List of plant families

Source Wikipedia, on alphabetical order.

#### **Bromeliaceae**

The Bromeliaceae (the bromeliads) is a family of monocot flowering plants of 75 genera and around 3590 known species native mainly to the tropical Americas, with a few species found in the American subtropics and one in tropical west Africa. The family includes both epiphytes and terrestrial species, such as the pineapple (Ananas comosus). Many bromeliads can store water in a structure formed by their tightly overlapping leaf bases.

### Cannabaceae

Cannabaceae is a small family of flowering plants. Members of this family can be trees (e.g. Celtis), erect herbs (e.g. Cannabis), or twining herbs (e.g. Humulus). The family includes about 170 species grouped in about 11 genera, including Cannabis (hemp, marijuana), Humulus (hops) and Celtis (hackberries).

Cannabaceae is of the rose order (Rosales). Members of the family are erect or climbing plants with petal less flowers and dry, one-seeded fruits. Hemp (Cannabis) and hop (Humulus) are the most economically important species.

# **Fabaceae**

The Fabaceae or Leguminosae, commonly known as the legume, pea, or bean family, are a large and economically important family of flowering plants. It includes trees, shrubs, and perennial or annual herbaceous plants, which are easily recognized by their fruit (legume) and their compound, stipulate leaves. Many legumes have characteristic flowers and fruits. The family is widely distributed and is the third-largest land plant family in number of species.

Along with the cereals, some fruits and tropical roots, several Leguminosae have been a staple human food for millennia and their use is closely related to human evolution. The family Fabaceae includes several important agricultural and food plants, including Glycine max (soybean), Phaseolus (beans), Pisum sativum (pea), Cicer arietinum (chickpeas), Medicago sativa (alfalfa), Arachis hypogaea (peanut), Ceratonia siliqua (carob), and Glycyrrhiza glabra (liquorice). Several species are also weedy pests in different parts of the world, including: Cytisus scoparius (broom), Robinia pseudoacacia (black locust), Ulex europaeus (gorse), Pueraria montana (kudzu), and a number of Lupinus species.

#### Malvaceae

Malvaceae is a family of flowering plants estimated to contain 244 genera with 4225 known species. Well-known members of economic importance include okra, cotton, cacao, and durian. Several species are pests in agriculture, and others are garden escapees. Cotton (four species of Gossypium), kenaf (Hibiscus cannabinus), cacao (Theobroma cacao), kola nut (Cola spp.), and okra (Abelmoschus esculentus) are important agricultural crops.

#### Musaceae

Musaceae is a family of flowering plants composed of three genera with about 91 known species. The family is native to the tropics of Africa and Asia. The plants have a large herbaceous growth habit with leaves with overlapping basal sheaths that form a pseudostem, causing some members appear to be woody trees. Cultivated bananas are commercially important members of the family, and many others are grown as ornamental plants.

#### Poaceae

Poaceae or Gramineae is a large and nearly ubiquitous family of monocotyledonous flowering plants known as grasses. It includes the cereal grasses, bamboos and the grasses of natural grassland and species cultivated in lawns and pasture. With around 780 genera and around 12,000 species, the Poaceae is the fifth-largest plant family, following the Asteraceae, Orchidaceae, Fabaceae and Rubiaceae. The Poaceae are the most economically important plant family, providing staple foods from domesticated cereal crops such as maize, wheat, rice, barley, and millet as well as feed for meat-producing animals.

#### Typhaceae

The Typhaceae are a family of flowering plants, sometimes called the cattail family. Typha is a genus of about 30 species of monocotyledonous flowering plants in the family Typhaceae. The genus is largely distributed in the Northern Hemisphere, where it is found in a variety of wetland habitats.

#### Urticaceae

The Urticaceae are a family of flowering plants. The family name comes from the genus Urtica. The Urticaceae include several well-known and useful plants, including nettles in the genus Urtica, ramie (Boehmeria nivea), māmaki (Pipturus albidus), and ajlai (Debregeasia saeneb).

Urticaceae species can be shrubs (e.g. Pilea), lianas, herbs (e.g. Urtica, Parietaria), or, rarely, trees (Dendrocnide, Cecropia). Their leaves are usually entire and bear stipules. Urticating (stinging) hairs are often present. Urticaceae species can be found worldwide, apart from the polar regions.

# **Publication details**

# **Textiles for Circular Fashion**

Part 2: From renewable carbon to fibres

Paulien Harmsen, Wouter Post and Harriëtte Bos

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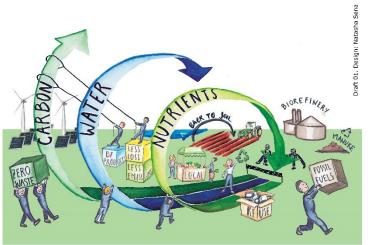
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