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Evolution of the ultrastructure and polysaccharide composition of flax fibres over time: when history meets science

[a]Alessia Melelli, [b]Frédéric Jamme, [c]Johnny Beaugrand, [a]Alain Bourmaud

[a]Univ. Bretagne Sud, UMR CNRS 6027, IRDL, Lorient, France

[b]Synchrotron SOLEIL, DISCO beamline, Gif-sur-Yvette, France

[c]UR1268 Biopolymères Interactions Assemblages, INRAE, Nantes, France

*Corresponding author: alain.bourmaud@univ-ubs.fr

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Abstract

Flax fibres have been used by humans for approximately 10,000 years. With time, the geographic area of production and cultivation has changed, as have the applications of flax fibres; from clothing to sails and paintings from antiquity, to automotive, fashion, and design applications in the contemporary era. The degradation process of flax fibres is the same for both ancient and modern objects made from this polysaccharidic material. This review, focusing on the cultural heritage field, after a brief description of flax plants and fibres, retraces the history of their use through Europe and the Near East, and discusses the evolution of extraction methods with human progress. Furthermore, the most important mechanisms of flax fibre degradation and the characterisation techniques currently in use are described. This study highlights the constructive interchange between engineering and cultural heritage that can be realised through a continuous comparison of antiquity and the contemporary era.

Keywords: flax fibres; cell wall; polysaccharides; cellulose; art; ageing

1. Introduction

Textiles are important inventions in human history, and the use of plant fibres, such as flax and hemp, have been used to produce not only clothes but also sails in sailing ships, flags, carpets or canvases, which are considered cultural heritage. Several of the first houses built by ancient civilisations were made with clay and straw in the form of mud bricks, which can be considered as the first biocomposite material in building construction. The use of mud bricks persists even today in several African countries as well as in India. Currently, there is renewed interest in the *know-how* and technologies of plant fibre composites for use in the material construction, automotive, sport and design industries as ecological, sustainable and cost-effective alternatives to common synthetic materials. This discovery and rediscovery of cellulosic fibres exhibits continuity in terms of technological progress. Consequently, an active exchange can be established between the engineering and cultural heritage to resolve problems in both domains and supplement the missing information, such as that of the evolution of degradation mechanisms.

Linum usitatissimum L., which is the scientific name of the common flax plant, is derived from the Latin word “*usitatus”*, which means “commonly used” (McDill et al., 2009). The Latin name of this plant highlights the importance of this plant throughout the history of civilisation. Flax is one of the oldest domesticated plants (Van Zeist & Bakker-Heeres, 1975) because its oil and fibres can be exploited, and it has been fundamental in both ancient and modern times to create clothes and objects. Despite the continuity of its plantation and application from the ancient era to the contemporary era, the cultivation and fibre extraction methods have been improved, and the plant morphology has evolved. Currently, textile flax is mainly cultivated in Western Europe; however, for thousands of years, this plant has been cultivated from the Mediterranean region to India, as well as in the Fertile Crescent area. The habitat of this plant varies from moderately cold to moderately warm regions (Yu, 2015), which is an adaptive advantage contributing to its widespread use.

Through human selection, flax plants have been optimised to obtain two main types of flax, the former suitable to produce linseed oil and the latter for fibres. In this context, several morphological characteristics of this plant have been modified. For example, the types of flax used to prepare fibres are taller with smaller capsules and fewer branches than those used to extract linseed oil (Yu, 2015).

Every year, new varieties of flax are commercialised, and the fibre yield and behaviour of each variety, as related to biotic (fungi, insects) or abiotic (wind, drought) stresses, are evaluated on an average of one to four years (ARVALIS Institut du végétal, 2019; Goudenhooft et al., 2017). Therefore, this plant undergoes substantial changes annually, which accentuates the difference between the linseed and fibre producing varieties and between the ancient and modern varieties.

To understand the evolution of this plant and its fibres in terms of morphology, cultivation and extraction methods, as well as the geographical areas involved and current application in different fields, the historical aspects must be considered.

Archaeological artefacts and historical objects can highlight the origin and use of flax and its fibres and explain the evolution of human interest in this plant, the reasons for the survival of the artefacts made from this organic material for several millennia and the type of techniques that can be used to study ancient and modern objects to compare them, describe their structure and quantify their quality.

Considering these aspects, this review summarises the history of the use, cultivation and extraction of flax, compares these aspects in the present and past contexts, highlights the ageing mechanisms that can change the initial structure, morphology and chemistry of flax fibres and clarifies the characterisation techniques available at present to study these features.

2. Flax stem organization and plant growth

Similarly to other vascular plants, flax stems have different histological tissues, as shown in **Figure 1.a,** which illustrates both young and mature steps. In this figure, it is possible to identify 1) the epidermis and cuticle, 2) cortical parenchyma, 3) sclerenchyma fibres, 4) phloem, 5) vascular cambium, 6) xylem and 7) pith.

2.1. Tissues and roles: an overview of the flax stem

Each tissue in the plant has a specific role, and the corresponding chemical composition is adapted according to this function. Xylem and phloem are vascular tissues. Xylem conducts water and raw sap and also contributes to the mechanical support of the stem (Nuez et al., 2020). Thus, the high lignin content of this tissue can help ensure the rigidity of the structure and hydrophobicity of the cells (Neutelings, 2011; Pereira, Domingues-Junior, et al., 2018). The phloem tissue has two types of cells: certain cells transport food resources such as the elaborated sap, while the primary sclerenchyma phloem fibres, which are flexible, support the structure of the stem. Flax fibres are grouped in 30–40 units and radially distributed in the whole stem. The chemical distribution examined through microspectroscopic techniques indicates that cellulose is more abundant in bast fibres than in the rest of the stem, while the content of lignin is lower but still detectable (Akin et al., 2000; Himmelsbach et al., 1999). Moreover, the lignin present in fibres has mainly G condensed epitopes. In contrast, the lignin in the xylem is mainly constituted of less condensed GS epitopes (Day et al., 2005).

The flax stem is composed of a vascular cambium (**Figure 1.a**) responsible for the generation of secondary xylem while bast fibres are generated by the procambium (Petrova et al., 2021). The cortical parenchyma, also called cortex or bark parenchyma, is richer in chlorophyll than xylem and bast fibres (Gorshkova et al., 2000), and, for this reason, it is easily distinguishable from fibres and epidermis.

The epidermis and cuticle, which are the external layers, are abundant in wax and phenolic groups, as they provide mechanical support and protect the innermost tissues of the stem from abiotic and biotic stress that may occur during plant growth (Bargel et al., 2004). Consequently, when the plant is injured, lignification occurs in the tissues involved (Paul-Victor et al., 2017) as lignin is a complex mixture of polyphenols and generally more difficult to metabolise by microorganisms than carbohydrates (Kim & Singh, 2000).

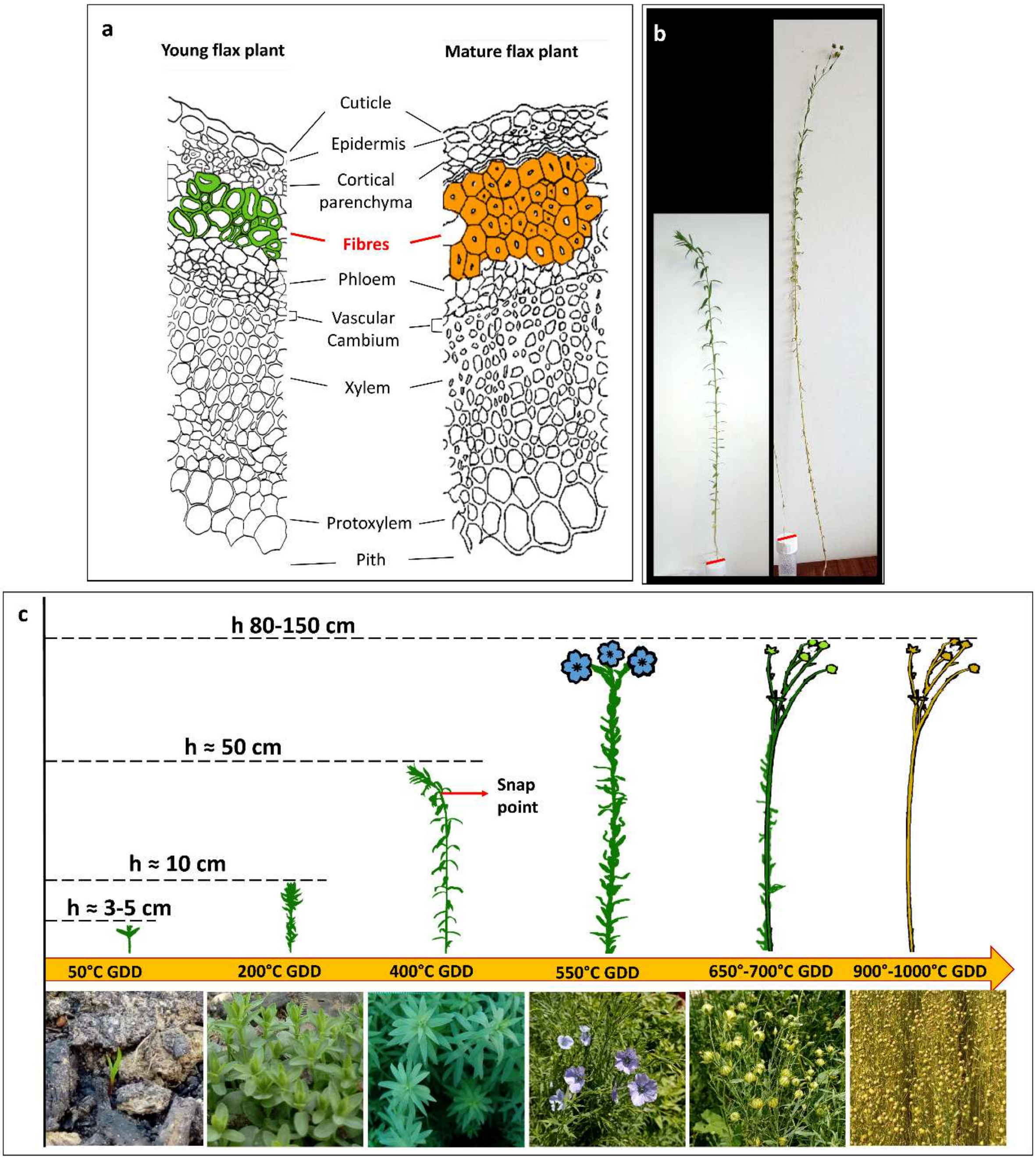

Figure 1. a) schema of a young flax stem at approximately 400 °C GDD and mature stem at 1000 °C GDD (© Erica Melelli); b) photo of two flax plants pulled out at 400 °C and 1000 °C GDD in the real scale; c) plant growth over time with flowering and browning of the capsules

The epidermis is rich in polygalacturonates, which are components of pectins, the molecules responsible for plant elongation and porosity regulation. As a plant matures, pectins become less methylated and form more cross-links with Ca^{2+}; these cross-links reduce the sensitivity to the polygalacturonase enzyme responsible for the degradation of cortical tissue during retting (Jauneau et al., 1997). The xylem is commonly referred to as woody tissue and during fibre extraction, this part of the stem is sacrificed and reduced in shives (Nuez et al., 2020).

During plant growth, the biochemistry of the different tissues of the flax plant changes, and thus, it is important to consider the maturity of the stem to determine when the flax crop should be harvested and fibres must be extracted.

2.2. Flax: a plant with a fast growth

Flax plants go through different steps of growth before becoming mature, and the cumulative growing degree day (GDD) value can be calculated to identify flowering, capsule browning and complete plant maturation. The GDD can be calculated as reported in **Eq.1**:

$$\text{GDD}=\left(\frac{T_{max}+T_{min}}{2}\right)-T_b \qquad (\textbf{Eq.1})$$

The maximum temperature is added to the minimum value and divided by two to determine the mean value, and the temperature base (Tb), which is the minimum temperature at which a specific plant can grow, is subtracted (Goudenhooft, 2018; P. Miller et al., 2018). T_b for flax plants is 5 °C. The first emergence after sowing occurs at approximately 50 °C growing degree days, flowering starts at 550 °C, followed by the growth of capsules, which are initially green, at approximately 650–700 °C. The capsules become brown at plant maturity (**Figures 1.b, c**) (ARVALIS Institut du végétal, 2016; Goudenhooft et al., 2019b). Flax plants reach fibre maturity between 850 °C and 1000 °C GDD, while the seeds are considered mature at 1000–1100 °C (ARVALIS Institut du végétal, 2016). Thus, it can be estimated that flax maturity can be achieved at approximately 110 d after sowing in Europe (Moudood et al., 2019).

During the growth of flax plants, it is possible to recognise the “snap point” as a transition point between the rigid stem, in which the fibres have already reached maximum elongation and start the thickening process, and the top of the stem where the cell elongation and intrusive growth still occur (Gorshkova et al., 2005). The top of the stem is thus flexible because fibres are still involved in the elongation process and have not yet been lignified (Gorshkova et al., 2003; Gorshkov et al., 2019). In general, the height of a flax plant may range from 80 to 150 cm at maturity (**Figures 1.b, c**), and this value also depends on the variety (ARVALIS Institut du végétal, 2019; Moudood et al., 2019). The root system is also of significance. The roots of flax cannot reach deep soils and therefore exploit the nutrients present in the first 70 cm of soil. Consequently, flax and linseed plants are sensitive to drought stress (Sertse et al., 2019; Soto-Cerda et al., 2019).

2.3. Fibre organisation and properties

2.3.1 General information on flax fibres

Flax fibres are single unit cells that originate in the phloem region from procambium (Esau, 1953) of the plant and, like jute and hemp, are among the commonly used bast fibres (Bourmaud et al., 2018). Cells are the fundamental units of living organisms. In the context of technical fibres, the cell wall is of interest because it is responsible for the rigid structure of a plant and contributes to its mechanical properties (Rogers, 2011), allowing the use of the fibres as raw materials with suitable properties, for example, as composite material reinforcement.

The diameter of flax fibres generally ranges from 8 to 25 µm (H. L. Bos & Donald, 1999), and its value changes throughout the cell (Thomason et al., 2011). The mean diameter can vary with several factors, such as the stage of maturity (Goudenhooft et al., 2018), variety, year and retting degree (Pillin et al., 2011). The diameter can vary depending on the fibre position in the stem. Fibres in the basal region have a larger diameter (approximately 25.2 ± 10.00 µm) than those on the top near the snap point (diameter of 11.4 ± 3.4 µm) (Baley et al., 2018).

Although the length changes across varieties, the fibre length is usually between 5 and 80 mm (Baley & Bourmaud, 2014; Baley et al., 2018) with an average value of approximately 20–30 mm.

2.3.2 Hierarchical structure and ultrastructure of the fibre

Flax fibres, like all cellulosic fibres, have a hierarchical structure composed of several layers (**Figure 2.a**). In engineering science, conventionally, the flax cell wall is divided into the primary layer (P), which is the outer layer and is approximately 0.2 µm thick (H. L. Bos & Donald, 1999), and the secondary layer, which is 5–15 µm thick (Morvan et al., 2003). Moreover, the secondary wall is divided into three sublayers labelled S_1, S_2 and S_3, specified for the first time by Roelofsen in 1951, who analysed flax, hemp and ramie fibres under a microscope in a polarised light environment after swelling in cuprammonium (Roelofsen, 1951).

Botanically, flax is a type of plant with gelatinous fibre cell walls and well-defined characteristics (Salnikov, 1993; Mellerowicz, 2001). The difference between lignified fibres (type S, with common distinction among S_1, S_2 and S_3 layers of the secondary cell wall) and gelatinous fibres (type G) has been described by Mellerowicz et Gorshkova (Mellerowicz & Gorshkova, 2012), and the characteristics of the G layer can be summarised as follows (Gorshkova et al., 2012, 2018): i) the lignin and xylan content of the layer is extremely low or nearly zero, ii) the layer is rich in water, and cellulose is its main component, iii) in contrast to the S layers, the cellulose microfibrils in the G layer have a small microfibril angle and are nearly parallel to the fibre axis. A recent model by Gorshkova *et al.* (Gorshkova et al., 2018), from which **Figure 2.a** is inspired and partially readapted, depicted the flax fibre structure with a very thin secondary S_1 cell wall composed of cellulose ~40%, lignin ~30%, xylan ~25%; the inner layer, also known as the tertiary cell wall (Gorshkova et al., 2018; Petrova et al., 2021) and identified as the G layer, has a cellulose content of up to 85% and the presence of rhamnogalacturonan-I (approximately 5–7%) with short galactan chains if the layer is mature and long chains if the layer is newly deposited (Gorshkova et al., 2018).

During plant growth, two mechanisms occur in flax fibre cells: intrusive growth and thickening of the cell wall layers. In the intrusive growth phenomenon, the elongation of certain flax fibres occurs faster than that of the surrounding fibres (Ageeva et al., 2005; Gorshkov et al., 2019), forcing the growing fibre to push through and grow among the other cells to find adequate space until the elongation process is complete.

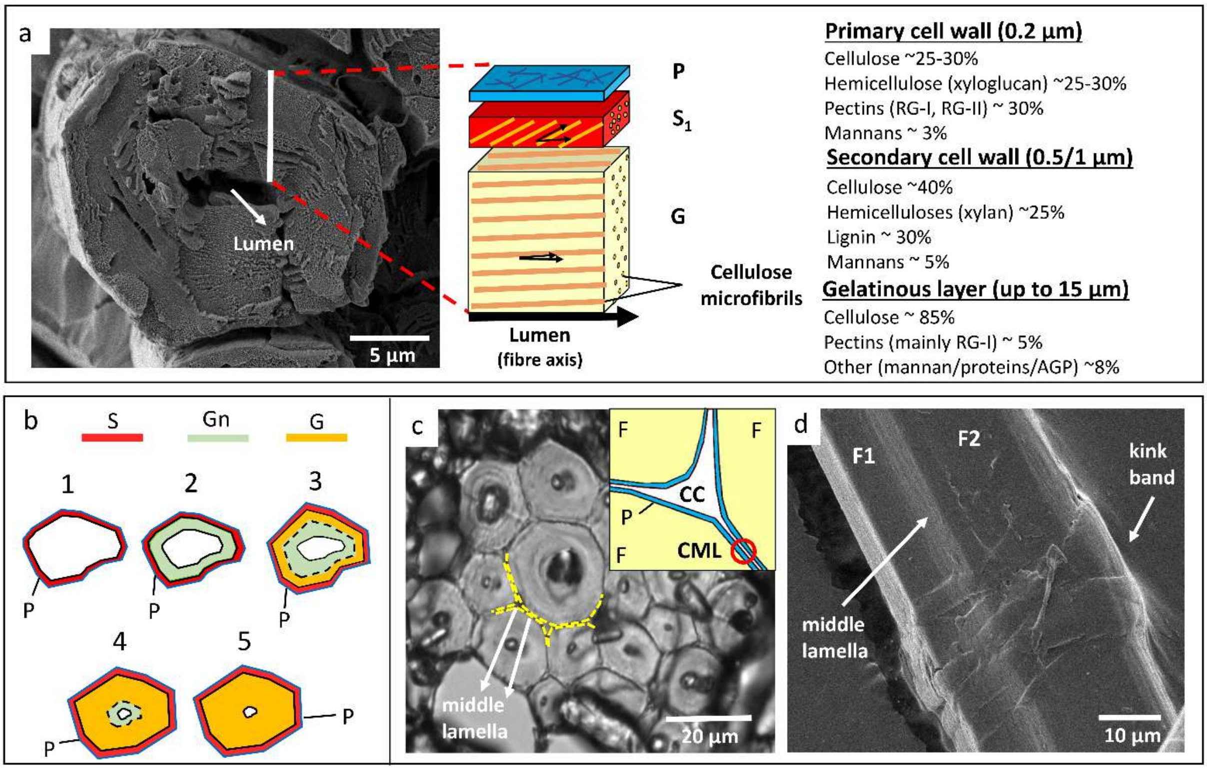

Figure 2. a) Schema of flax fibre ultrastructure and chemical composition, inspired and partially readapted from (Gorshkova et al., 2018; Manian et al., 2021) and data from (Goudenhooft et al., 2019b); b) formation of the cell wall layers, inspired by (Goudenhooft et al., 2018), and the progressive filling and thickening with the Gn layer transformed to G; c) transversal section of a flax fibre bundle, in which the middle lamella is highlighted in yellow. A schema representing the cell corner (CC) between three fibres (F) is shown, along the compound middle lamella (CML) composed of the primary wall (P) and middle lamella between two adjacent fibres; d) bundle of two fibres (F1 and F2) with a visible kink-band and outermost primary cell wall layer.

In the thickening process, the inner cell wall is divided into a mature layer, designated G, with Gn indicating the newly deposited layer (See **Figures 2.a, b**). During the development of the plant fibre, G and Gn coexist. Initially, only the Gn layer constitutes the cell wall, together with the S layer and the primary wall. However, with the maturity of the fibre, the Gn layer is subjected to a gradual thickening process that starts from the outer side and moves to the inner side of the cell, and this layer transforms to the G layer (Goudenhooft et al., 2018; Mikshina et al., 2013; Arnould et al., 2017). During this growing process, the lumen diameter is progressively reduced, in certain cases, almost replaced by the cell wall. As long as the fibre is alive, the lumen is filled with cytoplasm and vacuole, which disappears upon cell death, leaving the lumen empty (Ageeva et al., 2012; Richely et al., 2021). The different cell wall layers also have different chemistry (Gorshkova et al., 2018; Goudenhooft et al., 2019b), which also depends on the maturity of the fibre, as indicated by His *et al.* through immunogold localisation (His et al., 2001). Since it is difficult to clarify the biochemical composition for each cell wall layer separately due to their reduced thickness, in the literature, a global percentage is usually defined as the total contribution of all the layers (Jones et al., 2017; Bourmaud et al., 2018). **Table 1** lists certain percentages reported in the literature, for flax but also other plant fibres used for composite reinforcement. In addition, Figure A.1 in supplementary materials provides microscopic longitudinal and transverse views of the main fibres used for composite reinforcement.

Table 1. Literature review of the biochemical global composition of the considered bundles of fibres.

	Cellulose (%)	**Hemicellulose (%)**	**Lignin (%)**	**Reference**
***Linum usitatissimum L.* (Flax)**	60–85	14.0–20.6	1–3	(Van Hazendonk et al., 1996; Biagiotti et al., 2004; Alix et al., 2012)
***Cannabis sativa* (Hemp)**	80.2-89.4	6-12	2.6-4.6	(Marrot et al., 2013; Chernova et al. 2018)
***Corchorus capsularis* (Jute)**	58.0–71.5	13.6–24.0	11.8–16	(Khan and Ahmad, 1996; Roy et al., 2012)
Gossypium hirsutum L. (Cotton)	74-97.7	0.5-11	0.4-16	(de Morais Teixeira et al., 2010)

In addition to biochemistry, a key characteristic of the cell wall ultrastructure is the microfibril angle (MFA). Cell wall layers are composed of several cellulose chains known as microfibrils, the size of which is of the order of a few nanometres (Chinga-Carrasco, 2011), and assemblies of several microfibrils, realised through hemicellulose, pectin and variable amounts of lignin (Donaldson, 2008), form bundles known as macrofibrils. Each cell wall layer has microfibrils with different orientations against the fibre axis. This angle that cellulose microfibrils form with the fibre axis is known as the microfibril angle (MFA), illustrated in **Figure 2.a**.

In general, the primary wall is represented by a random orientation of the microfibrils (Stamboulis et al., 2001; John & Thomas, 2008; Madsen et al., 2016); however, certain recent studies indicated that mature flax fibres reorient the microfibrils inside the primary cell wall due to cell elongation (Kennedy et al., 2007; Rihouey et al., 2017).

The secondary wall has a complex structure with transition zones in which a gradual change in the angle of the microfibrils was observed (Roland et al., 1995; Baley et al., 2018), and, for this reason, in several models of flax fibres, the S_1 layer has been depicted having crisscrossed pattern of cellulose microfibrils (Wang et al., 2018; Bourmaud et al., 2019). In engineering, the S2 layer is considered the thickest main layer that should coincide with the G layer (Baley et al., 2018; Bourmaud et al., 2018), , and it is generally accepted that it has a MFA between 5° and 10° for dry flax fibres (Ansell & Mwaikambo, 2009; Bourmaud et al., 2013). On the other hand, in botany, the microfibril angle of the G layer is close to 0° and almost parallel to the fibre axis, which is one of the main characteristics of gelatinous fibres that allow them to be distinguished from other S-fibres (Mellerowicz & Gorshkova, 2012; Gorshkova et al., 2018; Gorshkov et al., 2019).

2.3.3 Middle lamella: the adhesive between fibres

Several tens of flax fibres are grouped in bundles in the phloem region of the stem. These fibres are glued through another layer, known as the middle lamella (**Figure 2.c**), which is mainly composed of pectic polysaccharides (58%), lignin (38%), and a small amount of protein (approximately 4%) (Richely, Bourmaud, et al., 2021; Zamil & Geitmann, 2017). Moreover, it is possible to distinguish the tricellular junction or cell corner (MLCC), which links three or four fibres, and the compound middle lamella (CML), which aggregates the middle lamellae and primary cell wall, thereby forming a composite system of both layers (**Figure 2.c**).

The middle lamella has often been considered the weakest layer, in which fractures can start to propagate when flax fibres are used in biocomposite materials and mechanically tested (Guessasma & Beaugrand, 2019; Monti et al., 2016). Nevertheless, certain researchers reported that fractures occur more often between the S_1 layer and G layer or between the S_1 layer and the primary wall than in the middle lamella (Day et al., 2005; Zamil & Geitmann, 2017). Based on the plant species and the specific function of the cells into the plant, the chemical composition of the middle lamellae between fibres vary, and consequently also the mechanical properties (Melelli et al., 2020), but the small thickness of this layer makes it difficult to study the composition locally without the contribution of the surrounding cell walls.

2.3.4 Kink-bands: defects along the fibres that influence the mechanical properties of modern and historical objects

An important characteristic of certain plant fibres, such as hemp and flax, is the local deformation of the cell wall along the axis. The associated defects are known as kink-bands, have a precise geometry (**Figure 2.d**) and often involve several fibres of the single bundle. These defects can be attributed to the environmental conditions during the growth of the plant and the fibre extraction process (Hughes et al., 2000). Hughes *et al.* demonstrated that hemp fibres carefully extracted from green stems have fewer kink-bands than the common fibres extracted in an automated extraction line (Hughes et al., 2000).

The geometry of kink-bands is different from that of the remaining fibre, and according to Thygesen and Gierlinger's work on *Cannabis sativa* (hemp), kink-bands have a less ordered microfibril network and higher microfibril angles (Thygesen & Gierlinger, 2013; Melelli et al. 2021a). Through FIB-SEM tomographic reconstruction, another team observed cavities in the kink-bands in flax fibres (Zhang et al., 2015), later confirmed by Melelli et al. (2021a). The abovementioned aspects are likely why kink-bands are the weakest points of plant fibres from a chemical viewpoint, although they are the preferred points at which enzymatic hydrolysis can be initiated (Thygesen et al., 2011). The density of these regions considerably influences the mechanical properties of fibres and composite materials (Davies & Bruce, 1998; H. Bos et al., 2002; Andersons et al., 2009; Sliseris et al., 2016) and has been known to limit the fibre strength in historical textiles and artworks (Herrera et al., 2010; Thygesen, 2010).

2.3.5 Small and strong: the mechanical properties of flax fibres

Despite the presence of kink-bands, flax fibres exhibit remarkable mechanical properties, which have been deeply investigated (Baley & Bourmaud, 2014; Baley, 2002). **Table 2** presents a comparison of the mechanical properties of flax fibres reported in the literature and those of other natural and synthetic fibres. The G layer is the main layer responsible for the mechanical behaviour of cellulosic fibres, especially in the longitudinal direction (Arnould et al., 2017). According to the existing studies, a small microfibril angle, small lumen and small fibre diameter lead to a high Young's modulus and superior performance (Bourmaud et al., 2013; Ramamoorthy et al., 2019). For instance, although cotton has a high cellulose content (82–98%), its MFA ranges from 20° to 30° (Ansell & Mwaikambo, 2009; Bourmaud et al., 2018), and thus, the Young's modulus and tensile strength are low; in contrast, the strain at break (in %) is high due to the increase in its MFA when subjected to tensile stress. Therefore, in ancient times, flax was used to prepare not only clothes but also sails that could resist the wind stress.

Table 2. Typical mechanical properties of E-glass compared to flax, hemp and cotton. Mean of the values modified from (Wambua et al., 2003; Hughes et al., 2007; Graupner, 2008; Lu et al., 2012; Fidelis et al., 2013; Célino et al., 2014; Bourmaud et al., 2018)

Fibre	Density (g/cm3)	Strain at break (%)	Tensile strength (MPa)	Young's modulus (GPa)	Moisture sorption (%)
E-glass	2.6	2.5–3	2000-3400	70-81	/
Flax	1.4-1.5	1.3-2.6	595-1500	40-60	8-12
Hemp	1-3-1.5	1.6-3.3	500-900	25-60	6-12
Jute	1.2-1.5	0.4-1.8	138-550	20-60	12-15
Cotton	1.5-1.6	7-12	287-800	3.3-8.5	8-25

Several studies have demonstrated that the variety [5,67] and growing conditions of the plant (Goudenhooft et al., 2019a) and the fibre extraction process (Baley et al., 2020) can affect the fibre performance. In recent years, mechanical properties have been recorded at the cell level. For flax, the indentation modulus lies between 15 and 24 GPa for a mature G layer and between 9 and 15.9 GPa for a newly deposited Gn layer (Goudenhooft et al., 2018; Arnould et al., 2017); these values also depend on the fibre position in the stem. When the flax plant is young, the thickness and local indentation moduli of flax fibres drastically change and increase from the bottom near the roots to the top near the snap point. However, the local indentation moduli and thickness become uniform when the plant reaches maturity (Goudenhooft et al., 2018). Once the stem is cut and undergoes retting, the indentation modulus of flax fibres increases with the increase in the retting degree, defined in terms of days of retting (Bourmaud et al., 2019).

3. Flax through the millennia in Europe and Near East

The interest in and use of flax plants for oil or fibres have been prevalent for several millennia. The domestication of flax has influenced the plant structure and properties as well as the area of cultivation and production. In this section, the history of flax cultivated for fibres and the evolution of the extraction methods are summarised from the origin of the use of flax through the contemporary era.

3.1. Evolution of the flax variety and geographical localization in Europe and the Near East

The first evidence of the use of *Linum usitatissimum* is dated between 9,000 and 8,000 BC in the Near East, with other instances of flax seeds observed in Turkey and Syria (Cullis, 2007). Only later did the use of these seeds spread in Egypt (Allaby et al., 2005; Helbaek, 1959; Weiss & Zohary, 2011). However, in 2009, Kvavadze *et al.* published an article on several flax fibres discovered in Dzudzuana Cave, Georgia, dated to Upper Palaeolithic period (30,000 years before present) through the radiocarbon dating of soil deposits (Kvavadze et al., 2009). The fibres were coloured, which suggests the beginning of a manufacturing process. This discovery can backdate the use of flax other than as a food source; however, the identification of these bast fibres is highly debated. The entire scientific community is not in consensus regarding the attribution to flax fibres, and this aspect must thus be further investigated (Bergfjord et al., 2010).

Although the use of flax is evident through archaeological findings, plant domestication likely occurred later in civilisations such as Mesopotamia and Egypt. These civilisations grew both domesticated and wild flax (or pale flax) as crops, as shown in **Figure 3**, which illustrates the natural distribution of pale flax in addition to the approximate areas of dated archaeological findings.

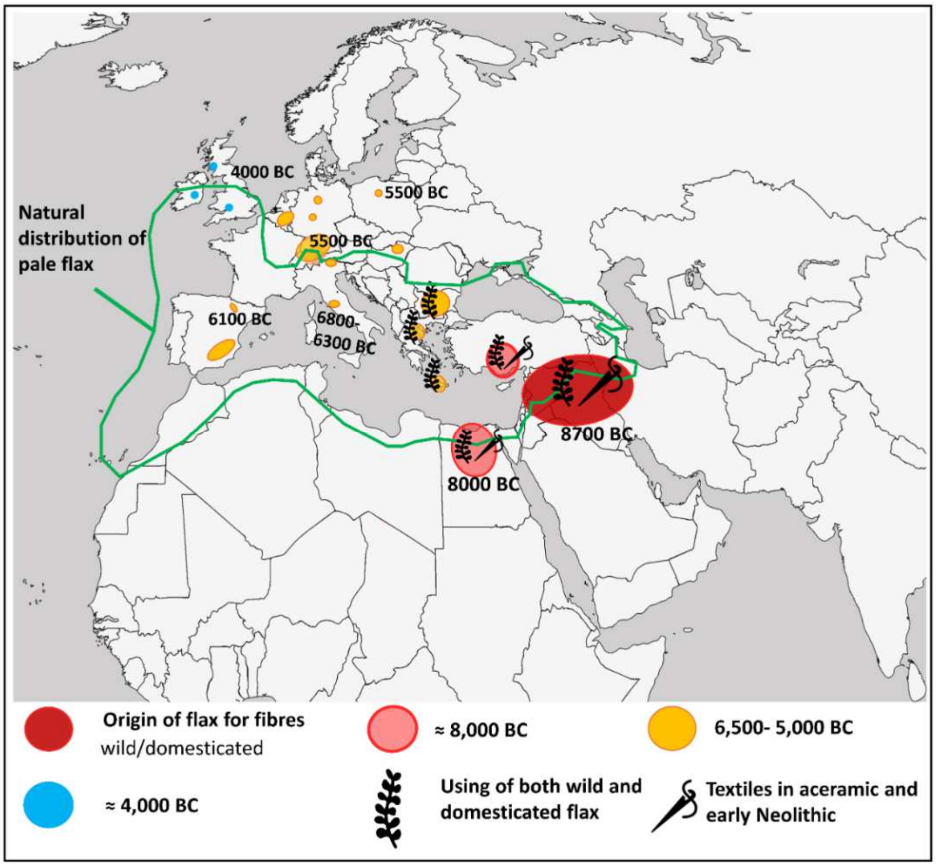

Figure 3. Map with the natural distribution of wild (pale) flax (in green) and the origin of the use of flax and its spread in Europe along with the first fabrics dated to the early Neolithic. The distribution of pale flax (green) has been obtained from (Desta, 2019; Diederichsen & Hammer, 1995; Gutaker et al., 2019). The centres of archaeological findings have been adapted from (Colledge & Conolly, 2017; Harris, 2014; Karg, 2011).

The history of the domestication and cultivation of this plant is not clear, and genetic diversity seems to suggest that independent episodes of domestication likely occurred in different geographical areas from Asia to the Mediterranean regions because of the vast geographical range of pale flax [92,97]. Before 1975, a hypothesis was presented regarding the relationship between *Linum usitatissimum*, which is domesticated flax, and *Linum bienne* as its wild progenitor, and van Zeist et Bakker-Heeres suggested that the first cultivations of flax were performed in dry soils as the natural habitat of *Linum bienne* (Van Zeist & Bakker-Heeres, 1975).

Diederichsen et Hammer compared the characteristics of the first progenitor pale flax (*L. angustifolium* Huds.) and cultivated flax *L. usitatissimum* (Diederichsen & Hammer, 1995). Morphological evaluations were performed through direct observation of the plants cultivated and analysed for three years with different accession geographic origins. The following aspects were considered: the length and width of petals, size of seeds, colour and shape of the flower, the height of plants and number of days until emergence from the soil or flowering. The authors concluded that most of the considered aspects for the two flax species were distinguishable, which highlighted the influence of the domestication of the plant on its morphology (Diederichsen & Hammer, 1995).

Muravenko *et al.* extracted the DNA of *L. angustifolium* (Huds.)*, L. bienne* and *L. usitatissimum,* and performed evaluations considering the genetic polymorphism and the comparison of the chromosome C-banding and molecular markers (Muravenko et al., 2003). The authors clarified the relationship between the three species, which are often confused and considered subspecies of one another: a dendrogram, as illustrated in **Figure 4**, indicated that *L. angustifolium* Huds. was similar to the ancient cultivated flax, and *L. bienne* was considered a subspecies of *L. usitatissimum* (Muravenko et al., 2003).

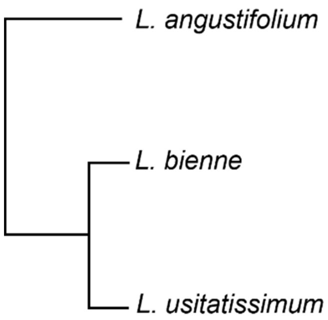

Figure 4. Dendrogram from (Muravenko et al., 2003). L. angustifoleum appeared as the progenitor of both L. bienne and L. usitatissimum.

Two years after this publication, another team of researchers used DNA extraction, PCR amplification and sequencing to analyse the cultivated and pale flax (Allaby et al., 2005). The authors suggested that a single pale flax plant (*L. angustifolium*) was likely domesticated and was the only progenitor of the cultivated flax. In Europe, the domestication of flax fibres is dated to the Neolithic period with the Linearbandkeramik Culture (5500-4500 BC) ((Kreuz, 2007), quoted in (Karg, 2011)). According to the map presented by Karg *et al.*, simplified and readapted in **Figure 3**, in contrast to that in the Middle East, only the domesticated plant was cultivated in southern and western Europe (Karg, 2011).

The use of cultivated flax increased rapidly and spread across Europe, and traces were observed in Switzerland (Leuzinger & Rast-Eicher, 2011) and Greece (Valamoti, 2011) during the Neolithic period.

Herbig et Maier measured the size of several flax seeds and evaluated capsules and shives from different archaeological sites located around Lake Constance and Upper Swabia. The authors hypothesise that different varieties of flax were used for linseed and fibres (Herbig & Maier, 2011). The variety of flax linked to the Pfyn culture was mainly used in 4,000–3,800 BC, and despite the presence of shives and linen found in the archaeological sites, the larger number of capsules and seeds and larger seeds suggests that this flax variety was principally used to produce oil. Seeds with a drastically reduced size were observed along with a higher quantity of shives and threshing traces only in a later period, during the middle and latest phases of the Late Neolithic (3,400–2,500 BC). This finding supported the hypothesis that a new variety of flax, linked with the Horogen culture and likely introduced from the Balkans, was cultivated for fibre extraction (Herbig & Maier, 2011). Even in the present era, the size of capsules and seeds, as well as their number in the tillers, are important characteristics that allow flax to be distinguished from the linseed varieties (Yu, 2015). The work performed by Herbig et Maier presented one of the earliest pieces of evidence of the use of different flax varieties for different purposes.

The extensive use of flax began with Egyptians during the dynastic period, in which flax fibres were widely used for cloth and sail production and in funerary rites. The production of these objects required considerable effort to produce linen, and Egypt emerged as the “land of linen”. The 1st dynasty (3,000 BC) corresponded to the first attempts at mummification, which was initially performed only for pharaohs and then successively extended to the remaining population (Grilletto, 2005). This practice persisted across all subsequent dynasties and for a certain period even beyond the conquering of Egypt by the Romans and its annexure into the Roman Empire. The case of the mummy of Grottarossa discovered in Rome and dated between 150 and 200 A.D. must be mentioned; an embalmed young girl was found to be treated with ancient knowledge of Egyptian mummification, and bandages of linen were identified (Ascenzi et al., 1996).

In ancient Mesopotamia and other areas of the Near East, considered the cradle of flax cultivation, the use of flax crops gradually disappeared as this plant required highly fertile soils to grow, and, consequently, aggressive expansion policies, or due to a long period of drought, which occurred in the last part of the Middle Bronze Age and the first part of the Late Bronze Age (McCorriston, 1997; Riehl, 2012). In Mesopotamia, in the 4th–3rd millennium, flax fibres were replaced with wool fibres (McCorriston, 1997), while drought during the Bronze Ages seemed to have favoured a change of crops towards drought-tolerant plants (Riehl, 2012).

Due to the flourishing trade between populations located in the Mediterranean regions, historians find it challenging to determine whether Romans or Phoenicians first introduced the use of linen in Europe; however, it is generally agreed that the expansion of the Roman Empire promoted the use of this plant for fibre extraction throughout northern Europe. In northern Europe, flax was primarily used to produce oil, and the most ancient remains of linen seem to suggest that they were originally imported from the Germans or Romans (Hald, 1950 as cited in (Ejstrud et al., 2011) and Bender Jørgensen, 1986 as cited in (Andresen & Karg, 2011)). In Denmark, there is evidence of linen fabric dated to 800 BC, and in Sweden, the first textile production is dated to approximately 200–300 AD (Andresen & Karg, 2011; Ejstrud et al., 2011; Viklund, 2011).

In the 1st century BC, Pliny the Elder wrote the *Naturalis Historia* and dedicated a part of his opera to flax (*Liber XIX)*, highlighting the importance of this plant in the production of sails that allowed commerce and contacts between populations around the Mediterranean Sea. Despite the flax cultivations in Europe, Egypt maintained primacy for centuries in the production and exportation of flax with the most advanced technology and expertise. Egyptian linen was cheaper and more abundant than other types of linen produced in the same period in Italy or by the Gallic population located in southwestern France (*Carduci*); however, Pliny the Elder also reported that the quality of this flax was inferior to that of the linen produced in Europe (NH 19, 13–14). This difference may have occurred because Egyptians sowed flax during the winter season, while to the north of the Mediterranean regions, flax was cultivated during spring (Andersson Strand, 2012), which likely facilitated the development of diversity in summer and winter varieties. Musselman reported that although flax was known to be used for both oil and fibres in the Bible, it was only associated with linen (Musselman, 2012), which suggests the importance of these plant fibres in antiquity.

Between the 9th and 10th centuries, in Zeeland and Flanders linen was generally preferred to wool for clothes because wool, although more easily produced, was considered too coarse, and Flanders became an important linen manufacturing centre (Nicholas, 1992, p. 38). As reported by Dixon in his opera dated 1854, at the end of the 13th century, Beatrice de Gaule introduced the coutil fabric in both northwestern France (Brittany, Main, Angers) and Flanders, where flax grew naturally (Dixon, 1854, p. 4). During the Middle Ages, the fustian fabric, a mix of linen and cotton that probably originated from Egypt and was successively produced in Italy and Spain, emerged as an important entity throughout Europe, especially during the 14th century (Ulrich, 1994). This fabric was manufactured also in Britain, Germany and Switzerland because European spinners could not produce cotton yarns that were strong enough to be used in the warp direction (Prance & Nesbitt, 2005, p. 290; Ulrich, 1994).

Additionally, during the 14th century, flax fibres were largely used to prepare cloths, sails, bags and fishing equipment; however, at the end of this century, a new use appeared in Europe as painting support not only for banners but also for pictures mounted on frames. The use of canvases for paintings began during the Late Middle Age, and Cennini mentioned the preparation of canvas supports in his treatise *Il Libro dell'Arte* (Chapter CLXII), written between the end of the 14th century and the beginning of the 15th century. One of the most ancient paintings on canvas surviving to this day is "*The Madonna of Humility*" by Lippo di Dalmasio, dated 1390 ca. and displayed at the National Gallery. However, the use of canvases especially coupled with the oil technique, mostly linseed oil, was popularised only during the Renaissance, with Italy being the main centre. The "*Sistine Madonna*" of Raffaello, dated 1513–1514, is an example of oil on canvas.

In the 15th and 16th centuries, other important centres of flax and linen production emerged in northern Europe, Britain and the Hanseatic area (Huang, 2014) as well as in Rus lands (Sherman, 2008).

With the Renaissance, the main cultural centre was displaced in Italy, and although the linen fabric was widely used and modest families had numerous luxurious linens and tablecloths, as reported in (Erichsen, 2020), flax was mainly produced in central Europe. In the 16th century, Poland, Bohemia and Moravia became important areas of linen manufacture (J. Miller, 2016), while Brittany, one of the main centres of flax cultivation, established major imports of flax seeds from Zeeland and Baltic countries (Pourchasse, 2019).

In approximately 1670, the Huguenots from France, Protestants who moved in large numbers to England and Ireland as well as Switzerland, Holland and Germany as religious refugees, introduced knowledge in England regarding textile production, as indicated by Louis Crommelin, a French Huguenot expert in linen manufacture, in the documents cited in (Colquhoun, 1961; Scott, 1901). During the 17th century in France, flax cultivations reached 300,000 ha, over double the area of cultivation in 2021 (140,000 ha) (Bourmaud et al., 2020). Baltic countries specialised in flaxseed production and supplied the product to the remaining European countries (Pourchasse, 2019; Bourmaud et al., 2020). Between 1660 and 1700 in England, considerable quantities of flax and hemp were imported from Norway, Denmark and the Baltic, while linen and textile yarns were principally imported from Germany, Holland, Flanders and France (Davis, 1954).

In the *Encyclopédie* dated 1751–1765, Diderot and D'Alembert reported the presence of three varieties of flax in the market: cold, medium and warm (Diderot & D'Alembert, 1751). Although the warm variety grew faster during the first phases of plant growth, at maturity, the plant was lower in height than the other two varieties and produced numerous branches with seeds; therefore, the yield of the flax fibres was low. In contrast, the 'cold' flax grew slower but was stronger and more resistant to cold environments. At maturity, the flax plant of this variety produced fewer branches and capsules and was taller than the other varieties; therefore, fibre extraction was optimal (Diderot & D'Alembert, 1751). The highest quality of seeds of the 'cold' flax variety came from Riga, but as this variety of flax plants produced a small number of seeds at maturity, so farmers in France were obliged to buy the seeds from Riga once every three or four years (Diderot & D'Alembert, 1751).

In the second half of the 19th century, Holland and Belgium specialised in the luxury market with high-quality flax fibres, while Russia became the leader of the common quality of flax fibres for all uses with a monopoly in the West European market (Ollerenshaw, 1999).

At the beginning of the 20th century, several main centres of linen manufacturing were located in Ireland, Scotland, France, Belgium, Germany and Bohemia (Ollerenshaw, 1999). Moreover, Ollerenshaw reported that with the Russian Revolution, Russian and Baltic flax production almost disappeared and re-emerged only after 1920, while the German occupation of northern Europe during the First World War affected the flax production in Belgium and France (Ollerenshaw, 1999).

In both World Wars, the production of flax was of primary importance to supply the army; further details can be found in Ollerenshaw's paper, as previously cited. In the Second World War, flax was widely used to build aeroplanes, parachutes and other military supplies (Kozłowski et al., 2020), and sustained production was observed. Kozłowski *et al.* reported that Egypt doubled its cultivation to support the flax demand of the United Kingdom (Kozłowski et al., 2020).

It must be highlighted that in Asia, India was one of the origins and/or diversification centres of flax, and production spread in the remaining continent between 5000–0 BC. In America and Australia, flax was introduced only with colonisation. The French exported flax cultivations and knowledge in Quebec approximately 400 y ago, and the English exported it in Australia less than 150 y ago (Desta, 2019).

3.2. Ancient cultivation and some extraction methods

Following the previous section, the history of flax cultivation is supplemented by a brief description of sawing practices and details of the extraction methods adopted by different populations across the centuries in Europe and the Middle East.

Today, as in the past, several steps must be implemented to obtain long fibres from flax. Once flax plants are sown and reach maturity, they are pulled out. To extract the fibres, a retting stage is implemented in which stems are laid on the soil and subjected to the action of rain, sun and microorganisms (dew retting) for several weeks or immersed in water (water retting) for several days. The two retting modes have the common objective of degrading the pectic middle lamellae in the bast fibre bundles to facilitate their extraction. After the retting step, the stem is mechanically broken (breaking), and the woody part of the stem and fibres are separated (scutching). Finally, the fibres are combed to remove the residual middle lamellae and separate the fibres (hackling or combing).

Starting from ancient Mesopotamia, flax was considered one of the most important plants and, as illustrated in the *Warka* vase (dated to c. 3200–3000 BC), it was often associated with dates as a symbol of fertility (Miller et al., 2016) because of its needs for rich soils and high moisture, which required irrigation. Apparently, in ancient Mesopotamia, only small plots of lands were dedicated to flax as a winter crop (Bedigian & Harlan, 1986; McCorriston, 1997). However, Miller *et al.* (2016) reported that, in ancient texts, flax was associated with concepts of both “field” and “garden”, while other important plants, for example, barley, were cultivated only in crop fields. Flax was associated with the goddess Inanna (Miller et al., 2016), and in general, weaving work was a prerogative of women, especially slaves and prisoners of war, as reported in (McCorriston, 1997) and its relative comments. In contrast, sowing and stem preparation, as well as fibre extraction, involved both women and men.

In the 4th–3rd millennium, Mesopotamians used both dew and water retting, and the textiles were left under the sun to bleach fibres (McCorriston, 1997). Moreover, the textiles were treated with certain plants that grow in the Middle East, such as *Salsola kali and S. soda, Salicornia europaea* and other glassworts and saltworts used to produce soda ash (Musselman, 2012; Soriga, 2017), as well as mineral clays such as Fuller’s earth (Soriga, 2017).

In ancient Egypt, flax was cultivated in mid-November after the seasonal flood of the Nile River in August–September (McCorriston, 1997; Semple, 1928a). In the beginning, only water from floods and rainfall was used in cultivations without any human intervention. However, due to the scarce rainfall that did not promote crop growth and the vital Nile flood, which decided the state of abundance or famine for the whole population (Bell, 1975), a system of basins (probably starting from the Middle Kingdom) was built to channel the water from the Nile inundation into crops (Schenkel, 1994).

The water was allowed to enter the basins for almost one month to saturate the soil, which also promoted the deposition of fertile clays and silts, and then allowed to flow out (Schenkel, 1994). Sowing occurred after this operation, and both *Linum bienne* and *usitatissimum* were sown (Vogeslang-Eastwood, 2000). At maturity, the stems were pulled out (not cut), grouped in bundles, and dried under the sun. Then the capsules were removed. As in Mesopotamia, stems were retained in current water between 10 and 14 d (water-retted) to eliminate pectin, and subsequently, the stems were successively broken with wooden mallets using stones as a work surface and a wooden knife to eliminate the rest of the bark (Andersson Strand, 2012; Vogeslang-Eastwood, 2000). Unfortunately, these work steps are not illustrated in ancient tombs or mentioned in texts, and no tools have been found in archaeological sites because of the high degradability of cellulosic objects (Andersson Strand, 2012; Vogeslang-Eastwood, 2000).

In contrast, in the Egyptian Tombs of Dagi (Middle Kingdom TT103, details shown in **Figure 5**) and Thutnefer (New Kingdom, TT104), the scutching process was illustrated: flax stems were passed through two sticks, and a wooden fan (or knife) was used as an alternative (Vogeslang-Eastwood, 2000).

Figure 5. Details of wall painting from Tombs of Dagi (TT103), where two women are shown to scutch flax stems (from the left) and probably splice flax into threads (right). Illustration from (Vogeslang-Eastwood, 2000)

Finally, the hackling step was implemented. In certain cases, this step was optional, especially when the thread was spliced rather than spun (Andersson Strand, 2012). In general, an indicator of spliced thread is if two or more threads, which are composed of twisted fibres, are plied. If the thread is spun, the yarn is generally used alone (Gleba & Harris, 2019). Ancient Egyptian threads were prepared through S-direction twisting and plying, and a wet rotation technique was used (Carroll, 1973; Vogeslang-Eastwood, 2000).

In terms of the use of flax in ancient Egypt, flax threshing and shives were used in mud brick construction, and a mix of silt, clay, sand, straw and water was used to create blocks as building materials (Cappers, 2005; Emery, 2011). Linen was employed in cloths and tunics, such as the fabric investigated in (Landi & Hall, 1979) to prepare sails (Black & Samuel, 1991; Partridge, 2012) and as bandages in the embalming process of dead bodies. As reported by Vogeslang-Eastwood, clothes were mended or retransformed and reused in other ways if they were excessively worn and torn (Vogeslang-Eastwood, 2000), and because linen production was particularly laborious, the same principle was likely also applied to other objects made of linen, as in the case of the linen shroud of a mummy that may have been previously used as a sail (Rougé, 1987).

Another important use was in cartonnage, a multi-layered painted support made of different layers, with the first layer made of linen, as shown in (Abd El Aal, 2018), followed by plaster and paint. The first condoms were likely made of linen in ancient Egypt (Smith, 2013), but the attribution of that function to small pieces of ancient linen fabric is still controversial.

As the use of flax spread from Europe through Egypt, the processing methods of flax were similar. In Upper Swabia and Lake Constance, during the Late Neolithic, the Horogen culture cultivated flax plants for fibres and oil (Herbig & Maier, 2011). Similar to Egypt and Mesopotamia, flax plants were pulled out and not cut, allowing the use of the entire length of the stem to extract fibres as long as possible. No traces of epidermis, cortex or phloem were found at the surface of the fibres, and Maier et Schlichtherle hypothesised that this could be attributed to the use of the retting process before the extraction of the fibre (Maier & Schlichtherle, 2011). The same authors also observed that certain archaeological remains of the stems were flattened and broken at different points, suggesting the use of a breaking tool to beat them (Maier & Schlichtherle, 2011). The combs for hackling and spindles found in the same sites indicate the use of a reasonably advanced method to produce linen. As reported by Maier et Schlichtherle and other authors cited in their paper, objects such as fabrics and fishing nets were preserved at the Constance Lake site.

In Italy, during the pre-Roman period and successively during the Republic, flax was mainly exported from Egypt, although Spain and Gaul exported a certain amount as well. Although a small amount of flax was also cultivated and processed in Italy, the greatest quantity was produced in the Roman colonies and used not only for clothes but also as war material to prepare tents and sail clothes (Gleba, 2002). The climate in Italy can be divided according to the country shape: northern Italy has a climate similar to central Europe, and South Italy has an arid Mediterranean climate. In ancient times, in the northern half of the country, sowing was performed during spring, while in the southern half, sowing more closely followed Egyptian practices and was performed in autumn with abundant rainfall (Gleba, 2002). Additionally, as Margarita Gleba reported, not all the soils in Italy were suitable for the cultivation of flax, and because linen production required more work than wool fibres, it was economically disadvantageous to produce it (Gleba, 2002). Furthermore, flax cultivation depletes the soil of resources, and thus, crop rotation was performed (Semple, 1928b). At present, due to the implementation of dew-retting and the release of organic matter during this stage, an opposite trend is observed, and the soil is enriched.

Similar to Egyptians, Romans used combs to remove capsules and wooden clubs to break stems after the dew or water retting process. To bleach the fibres, Margarita Gleba reported the exploitation of sun exposure, as in ancient Mesopotamia and Egypt, as well as the use of a sulfuric treatment (Gleba, 2002) and it is known in the literature that also urine was used for this purpose (Bradley, 2002).

Several spindles of different shapes were found in northern Italy (Busana & Tricomi, 2014), and in contrast to Egypt, in Europe, linen threads were mostly produced with a Z-twist (Gleba & Harris, 2019) as noted in the Etruscan textile examined by Carroll (Carroll, 1973). Moreover, the fabric mainly appeared in-plane waves (Gleba, 2002).

Tools similar to those used by the Romans to extract fibres were found in northern Europe. Because the use of flax fibres spread with the Romans, expectedly, at least initially, similar tools, such as clubs to beat the stems (**Figures 6.a**) and knives for scutching or combs for hackling (**Figures 6.b**), were adopted. An example pertains to the possible reconstruction and use of wooden clubs, as shown in **Figure 6.a** found in Sweden and dated to the Roman Iron Age, likely used to break flax stems.

Between the period of the Roman Empire and the Middle Age, several instruments and methods to extract fibres were improved. For example, in the Middle Age, Vikings likely used breakers mounted on a stand with serrated teeth, such as that illustrated in **Figure 6.a** with missing legs (Ejstrud et al., 2011). Surprisingly, not even the symbols linked with this plant changed through populations, and in Sweden and Norway and the Norse religion, flax was associated with femininity and goddesses and used in magical rituals linked with fertility and funerary rites (Viklund, 2011).

Two miniatured manuscripts created in Belgium, one dated 1515 (*Book of hours - Da Costa hours* MS M.399 fol. 12v (Pierpont Morgan Library, 1515)) and the other dated 1525–1530 ca. (*Book of hours* MS M.1175 Fol. 014r (Pierpont Morgan Library, 1525)), currently available at the Pierpont Morgan Library, show two men surrounded by a circle of flax stems, breaking fibres with a manual tool (a schematic reproduction of the *Book of hours* dated 1515 is presented in **Figure 6.c**). In both miniatures, a woman scutches the broken flax stems placed in a wooden column by hitting them with a wooden tool. This action was reported to be performed during November, and therefore, in the manuscript of Bruges MS M.1175, the miniature of the fibre extraction process has been associated with a second miniature with the sagitta zodiac sign. Another miniatured manuscript from France, *Livre d'heures* (L'Escalopier 22), dated 1555 and created for King Henri II, shows two women working on flax fibre extraction: the first woman is using a wooden breaking tool mounted on a stand to break the stems, while the other woman is scutching the fibres with a wooden knife, and the flax fibres are placed on a circular support (**Figure 6.d**). Apart from this difference, the extraction method did not seem to change across centuries.

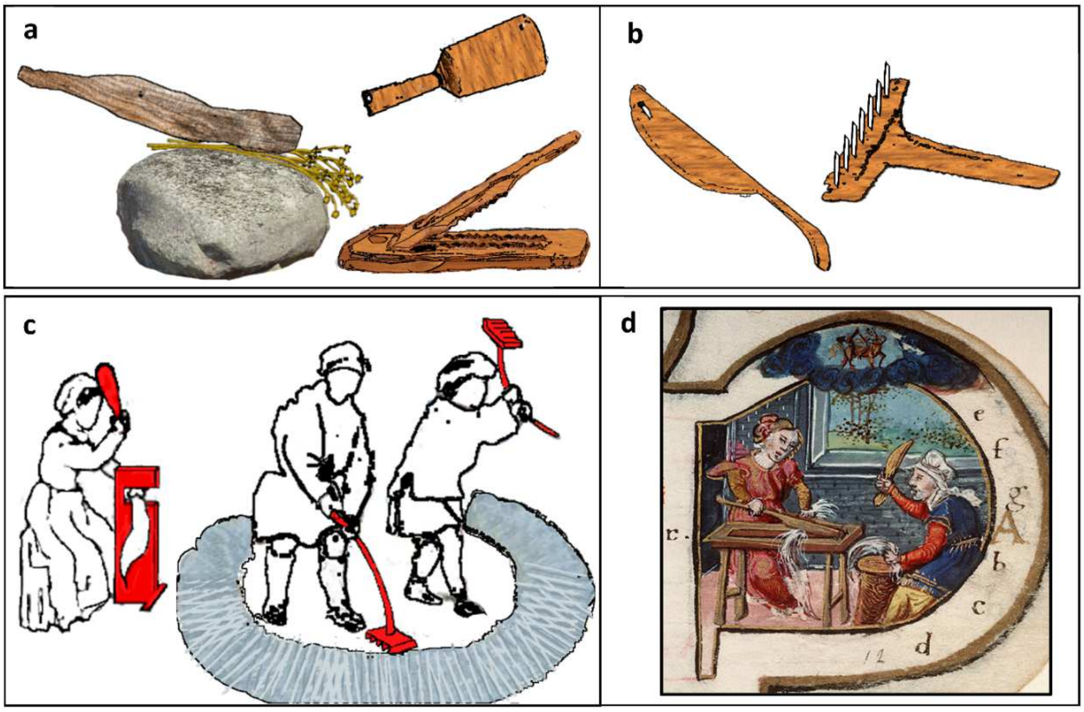

Figure 6. a) Left: possible reconstruction of the use of a wooden tool from Sweden to beat the flax stems on a plane surface (rock or wood). The tool is schematised from the archaeological tool (Item 447267. SHM 23159: K12) displayed at the Historiska Museet, original photo © Gabriel Hildebrand and right: another shape of a wooden club inspired from (Ejstrud et al., 2011; Viklund, 2012), and a schema of the breaking tool mounted on a stand found in Norway and reported in (Ejstrud et al., 2011); **b)** from the left: schema of a wooden knife found in Gloucestershire and dated to the middle Bronze Age (original photo © Cotswold Archaeology), and reconstruction of a heckle dated between 850–1000 AD found in Shetland, Scotland (Item X. HSA 318 © National Museums Scotland); c**)** schematic representation of the Miniature of the Book of Hours, MS M.399 fol. 12v, ca. 1515, Belgium (original photo © The Morgan Library & Museum, New York). On the left, a woman scutches a bundle of flax fibres with a wooden tool and a wooden column (highlighted in red) while two men beat the stems with breaking tools; **d)** Detail of the “Livre d'heures” L’Escalopier, hours in Latin with calendar, parchment of 16th century, f. 12 Ms LES 22 A, reproduced with the kind permission of © Bibliothèques d'Amiens Métropole and IRHT-CNRS.

In *L’Encyclopédie,* Diderot and D’Alembert indicate that in France, at the end of the 18th century, flax flowering occurred in June (Diderot & D’Alembert, 1751). Next, plants were pulled out based on the maturity and quality of the fibres that the farmers wished to obtain. The stems were left to dry in the field for 24 h on soil and successively grouped in bundles. The bundles were placed on the ground, in an upright position, leaning against one another to form an inverted V-shape chain of stems to enhance the airflow. The authors also reported that a shorter chain corresponded to better airflow. Dried stems were successively placed in the granary, and once the grains were dry, capsules were removed by beating the stems. For the retting process, three months were optimal: March, May and September. The retting process was performed in flowing water, in which stems were left for approximately 8 days and turned every day at the same hour of the day. The retting step was judged to be optimal by extracting three or four stems, pulling them and observing if the woody part was easily separated from the fibres. Retted stems were arranged on the ground to dry and bleached under the sun for eight days (the stems were turned after four days). Once the stems were dry, they were broken using a breaking tool mounted on a stand. A wooden knife with a dull wooden blade was used to eliminate the remaining straw, and finally, the flax fibres were combed (Diderot & D'Alembert, 1751). Diderot described another interesting method: flax was placed in a pot with boiling sea water, lime and ash in alternate layers, and the mixture was boiled for 10 h, with the addition of sea water when necessary. This method was used to prepare flax fibres to make them similar to cotton, and at the end of the process, flax fibres were ready for carding (Diderot & D'Alembert, 1751).

William Hincks performed a series of twelve lithography on linen manufacturing in Ireland for King George III of Great Britain, dated 1783. The author represented the process of flax fibre extraction. In plate II (**Figure 7.a**) two women harvest the flax crop in the background, while two other women eliminate the capsules. The man in the foreground places the stems in water. Plate III (**Figure 7.b**) illustrates the successive step when stems are extracted from the water by the man in the foreground. In the background, a woman and two men dry the retted stems near a fire and break them using breaking tools such as the tools illustrated in the miniatures shown in **Figure 6.c, d** and described by Diderot (Diderot & D'Alembert, 1751).

In plate IV, '*The Common Method of Beetling, Scutching and Hackling the Flax*' (**Figure 7.c**), a family is involved in the extraction process using the same tools as those in the Renaissance: a young girl uses a wooden mallet with stems placed in a plane support, two other women scutch the fibres on wooden columns using wooden knives and a man combs the fibres through several combs mounted on a wooden stand. In plate V, '*Interior view of a Scutch Mill*' (**Figure 7.d**), the first automated industrial process is illustrated.

Thus, even after the Industrial Revolution, manual extraction occurred. The French painter Jean-François Millet in *'Breaking Flax'* dated 1850–51 represented a woman performing the breaking process with the breaking tool mounted on a stand, although the tool appeared to be made of a larger and less elaborate wood piece. Fedot Vasilevich Sychkov, in the painting "Flax Combers"

(Мяльщицы льна) dated 1905, represented a group of women breaking fibres using the same type of breaking tool.

Figure 7. Four lithography by William Hincks, dated 1783 **a)** Plate II "View near Hillsborough in the country of Downe, representing pulling the flax when grown, hooking or putting it up to dry, rippling or saving the seed, and bogging or burying it in water". Two women remove the capsules and prepare the stems for water retting, while a man places bundles of stems in water. In the background, two other women harvest the flax plants, group them in bundles and place them upright with respect to the ground to dry before the retting step. **b)** Plate III "View in the county of Louth representing taking the flax out of the bog". After the retting process, the retted stems are moved near a campfire to be dried. In the background, two men are breaking the dried stems; **c)** Plate IV "The common method of beetling, scutching and hackling the flax". A family is represented as breaking the stems and combining the fibres; **d)** Plate V "A perspective view of a scutch mill, with the method of breaking the flax" represents one of the earliest industrial scutching mills. The four images are reproduced with the kind permission of the © British Library Board.

3.3. Actual sowing, production, extraction methods and application

The global supply of fibres has grown yearly since the end of the 20th century; however, the demand for synthetic fibres, such as polyester fibres, has decisively attracted more importance than natural fibres. In 2020, approximately 120 million tons of natural and synthetic fibres were produced, and while cotton production was about 26 million tons, flax reached slightly over 1 million tons (Engelhardt, 2020).

The global supply at present demands a high quantity of production, which requires several hectares of fertile, cultivable soil and industries that can process flax and sell fabrics and semi-finished products. According to the FAOSTAT data (www.fao.org), flax production is mainly concentrated in Europe, which alone produced 97.1% of the flax supply between 2018 and 2019, followed by Asia (1.6%), Africa (0.8%) and America (0.6%). The world leader is France, while Belgium, Belarus and Russia together cover approximately 16.5%, and the production of all the other countries completes the rest of the global supply (**Table 3**).

Table 3. Summary of the world production of flax fibres and tow in year 2019. Data from FAOStat (Crops and Livestock Products: Flax Fibre and Tow, 2021)

Country	Tonnes
France	850,350
Belgium	94,000
Belarus	46,245
Russian Federation	38,464
China, mainland	17,550
Netherlands	13,360
United Kingdom	8,199
Egypt	7,525
Chile	3,201
Argentina	2,695
Total	1,081,589

Several flax varieties are currently in use, which can be divided into winter and summer types. The choice of the variety is extremely important, with each variety having a specific fibre yield, sensitivity to pests (oïdium and fusarium), sensitivity to drought stress and wind (lodging) and precocity to maturity (ARVALIS Institut du végétal, 2020). Research teams of several laboratories have focused on genetic modifications. For example, Musialak *et al.* improved the retting process by reducing the pectin content; their transgenic flax was not only easier to ret but also more resistant to fusarium (Musialak et al., 2008), but the cultivation of GMOs is prohibited or strictly limited in Europe, making it an exception for research purposes (Directive (EU) 2015/412 of the European Parliament and of the Council of 11 March 2015 Amending Directive 2001/18/EC as Regards the Possibility for the Member States to Restrict or Prohibit the Cultivation of Genetically Modified Organisms (GMOs) in Their Territory, 2015).

After selecting the variety, seeds are sown with an optimal density between 1600 and 1800 plants/m^2. This density avoids competition between plants, which reduces the stem diameter and degrades the fibre mechanical properties and makes the plants sensitive to lodging (Bourmaud et al., 2016). For example, no remedy exists for *Fusarium oxysporum f. sp. lini,* which is a fungus that can penetrate from the soil to the root system and spread into vascular tissues (xylem and phloem) (Kroes et al., 1998). The plant dies slowly, and the fungus can affect the nearby plants. In other words, not only the mechanical properties of flax fibres are compromised, but fungal attacks can also cause the loss of the entire crop. Even if the diseased plants are eliminated, *Fusarium* can survive in the soil for several years. Therefore, crop rotation for flax is necessary, and a six-years interval between two flax cultivations is suggested (Bert, 2011).

At maturity, the plants are pulled out, and dew-retted between 3 and 6 weeks: the stems are left on the ground and turned to ensure retting in the whole stem. In Europe, the practice of water retting has been forbidden since the beginning of the 20th century due to induced water eutrophication, which refers to the pollution of freshwater with a high content of organic material generated by the action of anaerobic bacteria (Zawani et al., 2013); nevertheless, this practice is currently in use in other countries. Moreover, the cost of this process is high because fibres should be well dried once the retting is completed (Henriksson et al., 1997). Thus, dew retting is preferred, although this process is highly dependent on the weather, and warm temperatures and rainfall with alternating sunny days are essential to ensure satisfactory retting (ARVALIS Institut du végétal, 2017, 2020). Several geographical areas that produce linen, such as England or Scandinavian countries, cannot implement dew retting because of their unfavourable climate (Akin, 2013); climate change has also been responsible for inhomogeneous retting, especially in western Europe in recent years.

If the environment is excessively dry, the dew retting process is slow, and inferior retting yields stems with fibres that are difficult to extract; moreover, the well-glued cortical parenchyma is difficult to eliminate from the fibre surface, resulting in a low fibre quality (extremely coarse) and degraded mechanical properties (Martin et al., 2013; Meijer et al., 1995). In this case, the fibre bundles appear gold-coloured. In contrast, when fibres are over-retted for more time than necessary or excessive rainfall occurs, the extracted fibres are weak (Meijer et al., 1995) and appear black. In extreme cases, the cellulose can be excessively degraded and the fibre properties are inadequate to be used (Placet et al., 2017). Thus, although the retting process is the fundamental step, it cannot be controlled but can only be evaluated by an expert. Five organoleptic criteria are selected, based on the senses of sight and touch, listed in **Table 4**: nature of flax (unctuous or dry), colour, strength, fineness, and homogeneity of retting with presence/absence of woody parts. For each criterion, a value between 1 and 7 is assigned, where 7 represents the highest quality (Lefeuvre, 2014).

Table 4. Five organoleptic criteria to establish the flax fibre quality. Table readapted from (Lefeuvre, 2014)

Criteria	**Value assigned**						
Nature	**/**	**/**	**3**	**4**	**5**	**6**	**7**
			dry fibres or diseased fibres	fatigued fibres	standard	unctuous	unctuous/ silvered
Colour	**1**	**2**	**3**	**4**	**5**	**6**	**7**
	blue/black (over-retted)	bright yellow (under-retted)	diseased fibres (e.g., fusarium)	golden tonality	blue	bright blue	grey/silvered
Strength	**1**	**2**	**3**	**4**	**/**	**/**	**/**
	non-resistant	medium	satisfactory	highly resistant			
Fineness	**1**	**2**	**3**	**4**	**/**	**/**	**/**
	low	medium	satisfactory	highest fineness			
Homogeneity of retting (colour)	**1**	**2**	**3**	**4**	**/**	**/**	**/**
	large defect in colour (notable presence of straw)	a slight defect in colour (relative presence of straw)	high homogeneity	highly homogeneous			

To improve this step and render it more reproducible, enzyme retting has been studied; however, currently, this alternative is expensive (Akin, 2013; Tavares et al., 2020) and cannot be applied at the industrial scale, although the resulting fibres exhibit enhanced mechanical properties. Additionally, the use of fungal cultures was examined for the retting process. Akin *et al.* reported that among *Rhizomucor pusillus*, *Fusarium lateritium* and *Epicoccum nigrum* isolated from dew-retted flax, *R. pusillus* exhibited the highest performance because it did not attack the flax fibre cell walls and divide them from the remaining stem, in contrast to the other two fungi that led to over-retting (Akin et al., 1998). In addition to the possibility of damaging flax cell walls, certain fungi are also potential pathogens, such as *R. pusillus* for animals (Akin et al., 1998). Moreover, fungal spores are difficult to control and eliminate and can potentially grow even after the fibre extraction process. Another way to ret the stem is by chemical retting with reagents (Adamsen et al., 2002); however, this process is also expensive and cannot be used on an industrial scale (Tavares et al., 2020).

Once flax fibres are retted and well dried, they are processed in a scutching/hackling line similar to that described by Gregoire *et al.* and shown in **Figure 8.a** (Gregoire et al., 2019). Industrial extraction lines are based on ancient extraction methods transformed at a large scale and automated. The whole line is divided into different modules, as illustrated in **Figure 8.a**: a breaking module made of several rollers that crash the stems, a scutching module with turbines that scrap the broken shives from the fibres and a hackling module with combs in the last part of the line. For further explanation of the flax fibre extraction process, readers can refer to the review by Manian *et al.* (Manian et al., 2021). After their extraction, fibres can be transformed into woven or non-woven preforms. An advantage is that the same lines can be used for the extraction of other bast fibres, such as jute and hemp.

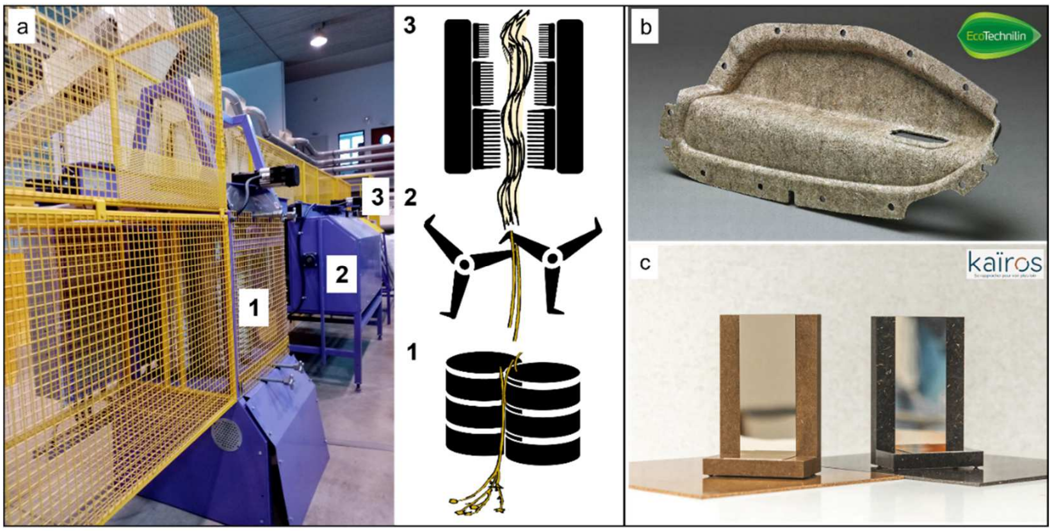

Figure 8. a) Taproot extraction line with three modules, and a schematic representation of the steps involved in general industrial extraction lines: 1. breaking module, 2. scutching module and 3. combing module; **b)** biocomposite made of flax fibres for automotive (© Ecotechnilin) and **c)** biocomposites reinforced with flax fibres employed for applicators used in cosmetic showrooms (© Kaïros).

After fibre extraction, several products are obtained: dust, shives and tow, short fibres and long fibres. Although technical fibres are the products with the highest quality and strength, the other subproducts are currently considered for use in the industry in the context of recycling (Nuez et al., 2020). At present, long flax fibres are mainly used in the manufacturing industry, household linen and design objects, although a new market of biocomposites and technical textiles has begun emerging. The definition of biocomposite is generally extended to the coupling between synthetic or mineral materials, which have a matrix function, and natural fibres, in this case flax fibres, used to enhance the mechanical properties of the matrix.

Countries worldwide are attempting to modify the practices in the industrial economy to reduce waste and carbon impacts, which are mainly responsible for climate change. This effort also involves substituting synthetic materials with plant materials when possible, such as the biodegradable plastic bags used presently for food waste (Ghosh & Jones, 2021). Consequently, biocomposites have attracted interest to promote widespread use in the future.

In contrast to blocks for building construction, such as kaolin and earth reinforced with plant fibres, the progenitors of which are ancient mud bricks (Colinart et al., 2020; Fic et al., 2013; Menasria et al., 2017; Vinceslas, 2019), biocomposites created from a mixture of polymer resins and plant fibres are materials developed in the contemporary era. These biocomposites are used for automotive and sporting goods such as bicycles but also for musical instruments and other objects used in daily life, such as those shown in **Figures 8.b, c**. Hybrid fabric with mixed carbon and plant fibres has been also developed (Shamsuyeva et al., 2019). Because such biocomposites are novel products, their mechanical properties, durability and life cycle are not well known and are being collaboratively examined in the academic and industrial domains.

Additionally, agricultural practices have been influenced by global warming and the consequent climate change. Flax is sensitive to drought, and almost all flax production is localised in France, especially north-west of France. According to the recent ARVALIS data, in 2020, almost all regions of flax production had less than 125 mm of rainfall between April and July compared to the mean calculated from the same period (April–July) of the last 20 y (ARVALIS Institut du végétal, 2020). In addition to drought, extreme weather events are uncertain, and in the future, the production of flax in France may be threatened.

4. The degradation of flax fibres: problems in modern and ancient objects

Flax fibre is an organic material with a complex structure and is naturally sensitive to changes over time. Consequently, the natural ageing process can easily modify the chemical composition, and such fibres are more susceptible to biological attacks and structural damage compared to inorganic materials.

In the context of artworks, this phenomenon can lead to problems for museums, restorers and conservation scientists focused on stabilising and preserving objects. Moreover, the engineering domain is interested in understanding the plant fibre ultrastructure responsible for their high performance and biochemical changes that occur after exposure to thermal treatments, humidity and artificial ageing processes, as well as the modifications due to coupling with materials of different natures.

This section summarises the factors that can contribute to the morphological, structural and chemical changes of flax fibres.

4.1. Two main factors of the flax fibre degradation process: water and temperature

In conservation science, water and high temperatures are the basis for almost all damage mechanisms of materials, and they mostly contribute to the ageing process. In particular, at high relative humidity and temperatures, several other degradation processes, such as biodegradation and chemical reactions, may occur.

A direct effect of high relative humidity is fibre swelling, caused by new bonds formed between water molecules with hydroxyl groups present in hemicelluloses and amorphous celluloses in cell walls (Célino et al., 2014b; Garat et al., 2020; Pejic et al., 2008) and carboxyl groups of pectins, especially in the middle lamella, that lead to mechanical and structural changes (Garat et al., 2020). Garat et al. indicated that at RH <10%, this type of absorption is prevalent, while at higher RH, between 10% and 65%, water is absorbed by pores and lumen through capillarity, leading to the abundance of free water, which are water molecules that are not chemically bonded (Garat et al., 2020).

In the context of museums and art galleries, a low relative humidity, RH 5–40%, causes the materials to become dry and brittle (Canadian Conservation Institute, 2013; Museum Galleries Scotland, 2021; Pavlogeorgatos, 2003) in addition to the structural deformation due to the loss of water molecules. The effect is accentuated if several materials of different natures are coupled, such as in oil paintings (Mecklenburg, 2007). In contrast, a relative humidity higher than 60% promotes the development of microorganisms: the first moulds appear after three months at RH 70% and after a few days at RH 90% (Canadian Conservation Institute, 2013). Furthermore, to eliminate the stress that repetitive humidity cycles can cause on the fibre structure, fluctuations of the relative humidity should be avoided in museums.

In Italy, the UNI10829 presented by Ente Italiano di Normazione suggests that ancient clothes, tapestries, natural fibres, etc. should be preserved at RH 30–50% with daily fluctuations limited to approximately 6%. Easel paintings on canvas (oils, tempera, gouache) should be preserved at RH 45–60% with fluctuations limited to approximately 6%, as cited in (Corgnati et al., 2010).

The effect of temperature is often associated with relative humidity because of its strong correlation. High temperatures can break the cellulose chain of plant fibres, which acquire a brown colour (Pertegato, 2004, p. 27). In museums, following UNI10829, ancient clothes, tapestries and other artworks made of natural fibres and easel paintings on canvas should be preserved at T 19–24 °C with daily fluctuations limited to approximately 1.5 °C (Corgnati et al., 2010). In the context of higher temperatures, Gassan and Bledzki reported that at 60 °C, amorphous cellulose forms hydrogen bonds. At 150 °C, recrystallisation occurs, and hemicelluloses are more sensitive to the degradation process than lignin or α-cellulose (Gassan & Bledzki, 2001). The authors also observed that until 170 °C, only a slight difference occurs in the tenacity and degree of polymerisation (DP). Above this limit, the degree of crystallinity increases, indicating recrystallisation after chain breakage. However, the tenacity and degree of polymerisation dramatically decrease (Gassan & Bledzki, 2001). This change in flax fibres due to extremely high temperatures was also recorded through nanoindentation and atomic force microscopy tests. At 190 °C, the indentation modulus was approximately 21 GPa, and at higher temperatures, the fibre stiffness was noted to be 16 and 14 GPa at 210 °C and 250 °C, respectively (Siniscalco et al., 2018).

4.2. Biodegradation, light and pollution: other degradation mechanisms linked with water and temperature

Biodegradation is the first of the indirect effects of water and temperature on natural cellulosic fibres and the reason why the hygrometry and temperatures must be meticulously controlled in museums.

Warm temperatures and high humidity content are ideal conditions for the development of most microorganisms and parasites that can degrade plant fibres. However, other parameters, such as pH, the presence of oxygen, light and availability of nutrients, may favour the growth of certain microorganisms and inhibit others, according to their limits of tolerance.

Two notable studies on jute fibres performed by Basu and Ghose (Basu & Ghose, 1962a, 1962b) demonstrated that different fungi can lead to different types of degradation and have different spread methods. Considering the type of spread, the authors divided the fungal species into two categories: one group can penetrate into the lumen, whereas the other group cannot (Basu & Ghose, 1962b).

Nugari *et al.* presented detailed tables of certain bacteria and fungi most frequently isolated from cellulose artworks and air of museums (Nugari et al., 2007), and other research teams summarised the methods of identification of fungi (Djemiel et al., 2020; Repeèkienë et al., 2007) and bacteria (Djemiel et al., 2020) isolated from dew-retting, water retting and standing retting flax fibres.

Table 5, presents the fungi and bacteria found in common between the retted flax and isolated from artworks. Fungi are more largely represented.

Table 5. Fungi and bacteria in common between fresh retted flax stems and cultural heritage objects made of cellulosic fibres. Table adapted from (Djemiel et al., 2020; Nugari et al., 2007; Repeèkienë et al., 2007)

FUNGI	BACTERIA
Alternaria solani	*Bacillus cereus*
Alternaria tenuissima	*Bacillus subtilis*
Aspergillus flavus	*Pseudomonas aeruginosa*
Aspergillus niger	
Botrytis cinerea	
Cladosporium cladosporioides	
Cladosporium herbarum	
Epicoccum nigrum	
Fusarium oxysporum	
Humicola grisea	
Trichoderma virens	
Verticillium nigrescens	

Water activity a_w (or free water) is essential to the growth of microorganisms and is expressed as the ratio of the partial vapour pressure of water in equilibrium with a solution (p^{equ}) to the water vapour pressure of pure water (p°). Therefore, $a_w = p^{equ}/p°$, with $a_w=1$ for pure water (Herrington & Vernier, 1995; Caneva & Ceschin, 2007; Tiebe et al., 2018). For materials such as food and other surfaces, the equilibrium relative humidity (ERH) indicates the capacity of a material to absorb and desorb water molecules in air at given temperature and under a total pressure of 1 atm. Therefore, ERH = $(p^{equ}/p^{sat})_T$, p = 1 atm, where p^{equ} is the partial pressure of water vapour in equilibrium, and p^{sat} is the saturation partial pressure of water (Herrington & Vernier, 1995).

In general, fungi need lower water activity (a_w between 0.75–0.99) than bacteria ($a_w > 0.90$) to grow, and a higher fungal activity is expected in air (Caneva & Ceschin, 2007; Kim & Singh, 2000). In contrast, if artefacts, objects or biocomposites are buried in soil or compost, bacterial activity is generally prevalent (Kim & Singh, 2000). However, in burial contexts, fungi (especially soft rot fungi) and bacteria may coexist (Kim & Singh, 2000) by attacking the same cellulosic materials and fibre cells (Singh et al., 2016). **Figures 9.a, b** show certain over-retted fibres attacked by bacteria visible on the surface of the fibre and fungi; the hyphae, in the case of flax fibres, are almost completely embedded in a biofilm.

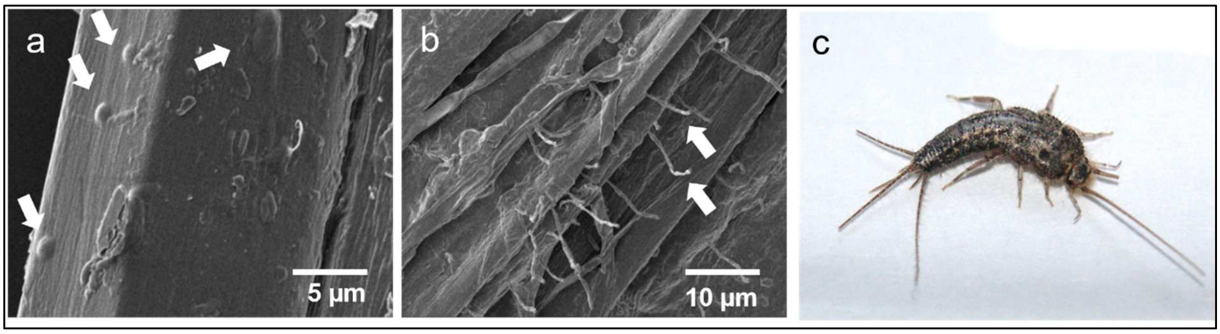

Figure 9. a) Bacterial and **b)** fungal colonisation on the surface of over-retted fibres and **c)** silverfish *Ctenolepisma lineata* (© Erica Melelli). Unpublished images.

In summary, fungi and bacteria may lead to mechanical and structural damage of the fibre through the formation of tunnels, erosion and fractures in the cell wall. Moreover, these organisms can lead to changes in the pH (generally to more acidic) due to substances secreted due to the metabolic activities and pigmented spots (Nugari, 2007; Pertegato, 2004). This change in acidity caused by enzymes can promote the acid hydrolysis of cellulosic fibres, which is one of the most destructive processes for textiles and paper.Insects may also damage linen fabric, although their action is less frequent than that of fungi and bacteria. Two of the most well-known insects that can infest linen are silverfish (*Lepisma saccharina*, *Ctenolepisma longicudata* and *Ctenolepisma lineata*), shown in **Figure 9.c**, and firebrats (*Thermobia domestica*) (Phillips & Gillett-Kaufman, 2018; Sloderbeck, 2004). The action of these insects is generally limited to hole production because they feed on cellulose fibres; however, silverfish can also produce yellowing spots (Sloderbeck, 2004).

Other indirect impacts of water and temperatures are chemical reactions, especially due to air pollution, and, in the case of seawater and aerosols, salt deposition. Air pollution is a current problem that has gained increasing attention in society. SO_2 and NO_x, generated because of oxygen in air and water molecules in moisture, can produce sulfuric and nitric acids, respectively, which react with cellulose fibres in the acid hydrolysis process (Pertegato, 2004, p. 28). Moreover, the complexity of artworks can also promote these chemical reactions because of the several layers of different natures often coupled in the same object, for example, in the form of pigments. The presence of moisture, together with the constant contact of cellulosic fibres with metals, can cause another phenomenon known as mineralisation. Metal corrosion caused by the reaction of water with metal cations creates minerals within cellulose fibres and leads to partial substitution of the organic material (Gillard et al., 1994; Peacock, 2003; Reynaud et al., 2020). Although fibre mineralisation causes structural modification of cellulose fibres such as flax, which become stiffer and brittle, metal cations, such as copper cations, can contribute to the preservation of textiles due to their biocidal activity, which limits the development of microorganisms (Gillard et al., 1994; Peacock, 2003; Reynaud et al., 2020).

In terms of salt deposition, water can contain dissolved salts, of which the most common salt is sodium chloride as in seawater. Salty water can penetrate between the cellulosic fibres, and into their pores and their lumen. The subsequent evaporation of the aqueous medium deposits salt, which crystallises and the formed crystals exert pressure, causing fractures in the cell walls and cavities between fibres (Kirker & Glaeser, 2011). This phenomenon requires alternate wetting and drying cycles, and salt, being hygroscopic, modifies the hygroscopic stability of the whole object, together with its structure, in a self-perpetuating process. Wood degraded with salt is also known as "fuzzy" wood because of its appearance (Kirker & Glaeser, 2011).

In general, the problem does not arise because these conditions do not allow the preservation of cultural artefacts made of bast fibres. For example, cotton immersed in sea water can degrade in three weeks (Dorée, 1920). Therefore, although salt growth is documented in stone (Anson Cartwright & International Scientific Committee for Stone, 2008) and wood (Mi et al., 2020) conservation, this phenomenon is not considered for bast fibres. Notably, this aspect is considered to be examined in the engineering field in the future because one of the applications of plant fibre-reinforced composites is the production of aquatic sports equipment such as surfboards (Pil et al., 2016) and sailing equipment.

The last parameter is light. Together with water and temperature, light can promote the growth of microorganisms and cause heating damage. UV radiation is particularly dangerous, especially in the presence of water molecules, because it causes photo-oxidation of cellulosic fibres, which not only alters the original colour but also produces carboxylic acid and breaks the cellulose chains (Pertegato, 2004, p. 27; Yang & Freeman, 1991). Furthermore, oxidised cellulose promotes hydrolysis (Marini Bettolo et al., 2007). Therefore, UNI10829:1999 established illumination limits similar to the temperature and humidity parameters. Textiles and tapestries can be exposed to a maximum luminance of 50 lux (luminous flux to square metre = lm/m^2), with a maximum UV radiation of 75 µW/lm and a maximum annual light dose of 0.2 Mlx • h/y. Different values are indicated for paintings on canvases, which are less sensitive to light than pure textiles (E=150 lx, UV max=75, annual light dose LO=0.2) (Corgnati et al., 2010).

5. Characterization techniques to study ancient and modern flax fibres

Several techniques are currently being used to characterise plant fibres at different scales and are briefly introduced in this section, followed by the types of information that can be obtained using these techniques.

5.1. Morphological analysis

The simplest characterisation method is optical microscopy using visible light. Compound and stereo microscopes, which are fast, inexpensive and available in every laboratory, are suitable to obtain a global view of the sample. Optical microscopy can be adopted to measure the average diameter of flax fibres and evaluate the presence/absence of kink-bands and defects along the fibres (**Figure 10.a**). In the case of fabrics, this technique can be used to evaluate the thread count and type of weaving, as indicated by Helmi *et al.* (Helmi et al., 2008). As shown in **Figure 10.b**, polarised light microscopy enables the identification of kink-bands from different bast fibres (Bergfjord et al., 2010) and their quantification through image processing (Mortensen & Madsen, 2014; Thygesen & Hoffmeyer, 2005). For further explanation of the investigation of kink-bands under polarised light, Thygesen and Hoffmeyer's paper is suggested (Thygesen & Hoffmeyer, 2005).

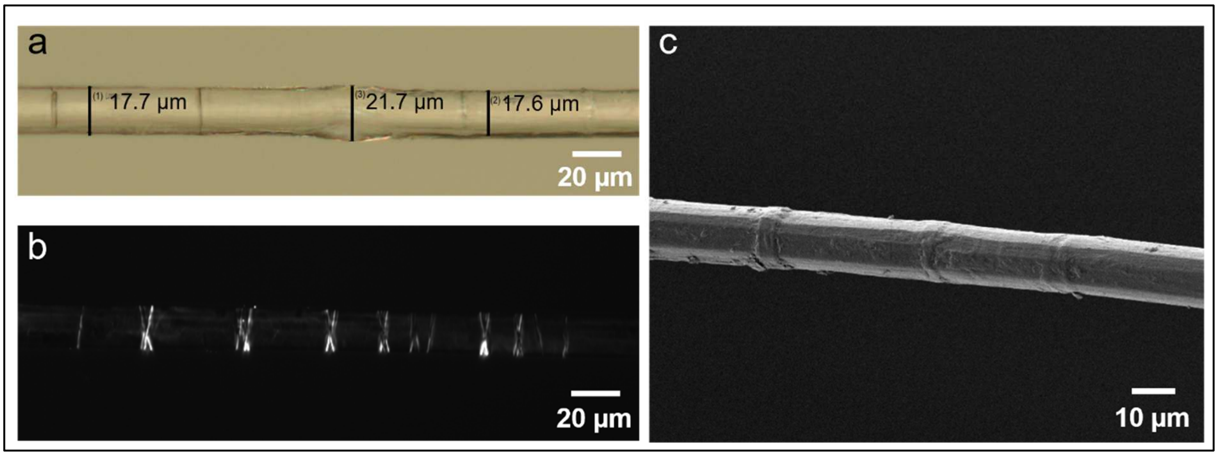

Figure 10. a) Elementary flax fibre under optical microscopy and diameter calculation **b)** elementary flax fibre under polarised light that highlights kink-bands in white (©Emmanuelle Richely) and **c)** SEM microscopy of an elementary flax fibre with three visible kink-bands.

Scanning electron microscopy (SEM) is one of the most commonly used characterisation techniques. This approach is principally used to scan the fibre surface (**Figure 10.c** and **Figure A.1**) and, in the case of artworks, it can provide information regarding the elemental composition of foreign materials if coupled with energy-dispersive X-ray spectroscopy (EDX) or wavelength-dispersive X-ray spectroscopy (WDX) systems (https://www.eesemi.com/edxwdx.htm for more details). SEM analysis can be used to clarify the fibre shape (**Figure A.1)** and kink-band shape, signs of ageing (Herrera et al., 2010) and presence of foreign materials and biological attacks (Richter, 2005). Richter reported that textile fibres can be washed in boiling water and ether at 40–60 °C to extract dust and foreign materials before SEM analysis (Richter, 2005); however, this step is not always possible and is not exempt from the risk of causing further damage to the fibres. Additionally, certain important pieces of information that can be obtained from foreign materials may be lost. Consequently, this step is rarely executed for ancient textiles and is not necessary when modern samples are considered.

Furthermore, although transmission electron microscopy (TEM) analysis is important, it is not commonly adopted because of the difficult sample preparation process; the sample must be as thin as possible (less than 100–150 nm). In contrast to SEM, this technique highlights the cell wall layers in sample cross-sections and can be used to investigate the biodegradation of the cell wall in archaeological findings (Kim et al., 1996; Kim & Singh, 2000).

5.2. Study of parietal composition

FTIR and Raman spectroscopies are vibrational spectroscopic techniques that are extensively used to analyse bast fibres, and these approaches are well known in the cultural heritage field because of their non-destructive nature. Nevertheless, in the case of plant fibre analysis, FTIR spectroscopy is generally preferred because the recorded spectrum is not influenced by the presence of fluorescence emission; in contrast, in the case of Raman analysis, fluorescence can hide signals of interest from the sample structure. Therefore, Raman spectroscopic analysis of bast fibres is generally preferred using an excitation wavelength of 785 nm in the case of micro-Raman spectroscopy (Bonizzoni et al., 2016; Kavkler & Demšar, 2011) or 1064 nm for FT-Raman spectroscopy (Edwards et al., 1997) to reduce fluorescence. Another method to decrease fluorescence, which is extremely high in ancient textiles, is the photo-bleaching method, which involves irradiating the sample with a laser for long periods, for example, 30 min, before the acquisition (Kavkler & Demšar, 2011).

Micro-Raman spectroscopy performed using an excitation wavelength of 532 nm and a 100x oil immersion objective can have a spatial resolution of approximately 240 nm (Thygesen & Gierlinger, 2013). Infrared spectroscopy can be performed at the microscale level through µ-FTIR/ATR, which is an IR spectrometer technique involving a germanium crystal that allows a contact area of 100 µm with the sample surface (PerkinElmer Inc., 2010).

With excitation wavelengths in the visible radiation or NIR (for Raman spectroscopy) range and IR range (for FTIR spectroscopy), the abovementioned techniques allow to obtain information on the sample molecular structure through the inelastic scattering of photons in the case of Raman, or absorption of the incident radiation that activate particular molecular vibrational modes of molecules present in the sample, in the case of FTIR. For further information regarding the application of vibrational spectroscopic techniques to plant cells, Gierlinger's work is suggested (Gierlinger, 2018). Vibrational spectroscopic analysis of bast fibres allows to recognize plant species (Edwards et al., 1997; Garside & Wyeth, 2003; Kavkler & Demšar, 2011) and study ageing process (Bonizzoni et al., 2016; Margariti, 2019), biodegradation (Elamin et al., 2018; Kavkler, Gunde-Cimerman, et al., 2011) and evaluate presence or absence of foreign materials, such as pigments and binders.

These two spectral techniques can also be used to calculate relative intensity ratios between peaks to evaluate the crystallinity index ($Xc_{\text{FT-Raman}}/\%= (I_{1481}/\ I_{1481}+I_{1462}) \times 10^2$ in the Schenzel method or I_{380}/I_{1096} in the Agarwal method, $Xc_{\text{FTIR}}= I_{1372}/I_{2900}$) (Agarwal et al., 2010; Kavkler, Gunde-Cimerman, et al., 2011; Schenzel et al., 2005) and lateral order index ($LOI_{\text{FTIR}}=I_{1430}/I_{897}$) (Fan et al., 2012; Nelson & O'Connor, 1964), which can reflect the state of preservation of cellulose chains in the fibres.

^{13}C cross polarisation magic angle spinning nuclear magnetic resonance spectroscopy (^{13}C CP-MAS NMR) is another technique used to evaluate cellulosic objects. NMR uses nuclei, such as ^{1}H or ^{13}C, and maps their position in the molecules by applying an external magnetic field that induces the nucleus resonance. An advantage of solid-state NMR is the possibility of studying solid samples in their native form. A recent and exhaustive review of the solid-state NMR technique can be found in (Reif et al., 2021). This technique provides complementary information to that obtained using vibrational spectroscopy, regarding the abundance or change in the molecular composition of the sample. For example, for cellulosic materials, ^{13}C NMR CP-MAS analysis was performed to evaluate naturally aged and artificially oxidised cotton and linen fibres to simulate fibres in ancient paper (Princi et al., 2005). A notable parameter that can be calculated using the ^{13}C NMR CP-MAS technique is the crystalline to amorphous ratio of cellulose. The values are calculated using the integrated areas of crystalline C4c (δ~87–93 ppm) and amorphous C4a (δ~80–85 ppm) cellulose, through the formula $R=I(C4c)/I(C4a)$ (Castro et al., 2011; Park et al., 2009). Moreover, the cellulose I_α and I_β allomorphs (Atalla & VanderHart, 1984; Larsson et al., 1997; Foston, 2014) can be evaluated, along with the effects of mechanisms such as hydrolysis (Wickholm et al., 1998).

Other complementary information regarding the fibre structure can be obtained by the calculation of the average lateral fibril dimension (Wickholm et al., 1998), which is an estimate of the ultrastructure of the cellulosic fibres and their microfibril network. NMR is micro-destructive, and at least 30 mg of sample is needed, which is a key limitation of this technique in the cultural heritage field (Capitani et al., 2012). Other techniques, such as gas chromatography/mass spectrometry (GC/MS) or pyrolysis–gas chromatography/mass spectrometry (Py-GC/MS) can be used to obtain information regarding fibre treatments (Aracri et al., 2010; Dorez et al., 2014) or pigments (Degani et al., 2015) present at the fibre surface; however, the use of these approaches is limited. In botany and engineering fields, gas chromatography is widely applied, for example to determine the polysaccharide composition, but notably, the minimum amount of sample required is 5 mg and its destructive (Lefeuvre et al., 2018; Pettolino et al., 2012).

The last technique presented here that can be used to obtain information regarding the state of preservation and ageing of cellulose fibres is the degree of polymerisation (DP) (Seves et al., 2000; Rossi, 2005; De Caro et al., 2019). This technique quantifies the monomers present in the cellulose chains through viscosity or sedimentation-diffusion measurements (Timell, 1955); thus, the technique is destructive and requires a considerably high amount of sample, i.e. between 60 and 120 mg (Rossi, 2005). In the past, there was disagreement regarding the polymerisation degree calculated using nitrate or cuprammonium and the preference for sedimentation-diffusion and viscosity. However, at present, protocols have been established through norms as the ISO 5351:2010, UNI 8282: 1994 in Italy or DIN 54270 in Germany, according to which, copper (II) ethylenediamine is used for sample dissolution and successive calculation of relative viscosity. As reported by Rossi, for raw cotton and linen, the degree of polymerisation is higher than 2000–2500 DPw, while for bleached fibres, the degree of polymerisation is higher than 1200–1300 DPw. In ancient paintings, values less than 500 DPw are considered critical and a sign that cellulose fibres have lost their strength (Rossi, 2005). An interesting example of this application can be found in (Seves et al., 2000).

Notably, in the engineering field, other destructive techniques, such as thermogravimetric analysis TGA and derivative thermogravimetry DTG, can provide indirect and approximate information regarding the chemical composition of cellulose fibres and enable the comparison of the quantity of water, cellulose components and non-cellulosic material between two samples (Jiang et al., 2019). Flax fibres, heated at a well-defined interval of 10 °C to high temperatures (600 °C) with constant monitoring of their weight, exhibit loss of water at 60 °C, and successive degradation of cellulose and hemicellulose at 300 °C, followed by lignin and pectin degradation at 400 °C (Jiang et al., 2019; Gourier et al., 2014).

5.3. Ultrastructural modifications

X-ray diffraction (XRD) is based on a collimated and monochromatic X-ray beam that hits the sample and interacts with electrons of the atoms in the sample. The X-rays are elastically scattered, and if the sample has a crystalline and well-ordered structure, the crystalline lattice generates a diffraction pattern due to the constructive and destructive interferences following Bragg's law: $2d\sin\theta=n\lambda$. XRD analysis, performed with synchrotron radiation (SR-XRD) (Herrera et al., 2010; Paris & Müller, 2003), can be used to examine the crystallinity index of cellulose fibres (Zhao et al., 2007), microfibril angle of cellulose microfibrils (Müller et al., 1998), mineralisation process (Chen et al., 1996) and presence of unknown fibres (Müller et al., 2006).

XRD can provide data in two formats: representations of the 2D diffraction pattern, and one-dimensional profiles extracted from these representations. Flax fibres yield the typical signal of cellulose I with reflections at 110, $1\bar{1}0$ and 200 (Paris & Müller, 2003). Based on these reflection peaks in the one-dimensional profile extracted from the pattern, three methods can be used to calculate the crystallinity index: i) using height ratios between the reflection peak at 200 and minimum reflection intensity between the 110 and 200 reflection peaks, ii) deconvolution of the reflection peaks and iii) amorphous subtraction. For more details regarding these three methods, readers can refer to the review presented by Rogpipi *et al.* (Rongpipi et al., 2019) or the paper written by Park *et al.* (Park et al., 2010).

Notably, the crystallinity index measured using the three methods differs, and both the deconvolution and subtraction methods yield results closer to the NMR than the height method (Park et al., 2010). To study plant fibres, small-angle (SAXS) and wide-angle (WAXS) X-ray scattering techniques are used, as indicated by (Müller et al., 2000), to obtain information regarding the fibre structure. De Caro *et al.* performed WAXS analyses to realise the ageing characterisation of historical linen threads (De Caro et al., 2019). In engineering and botany, the use of SAXS and WAXS is of significance to study the microfibril angle of cellulose microfibrils of plant fibres (Müller et al., 1998, 2000) because a lower angle corresponds to a higher tensile strength and modulus (Ioelovich, 2014).

Another less common technique to investigate the internal structure of plant fibres is focused ion beam-scanning electron microscopy (FIB-SEM), which can be used to implement tomography on single elementary fibres (Sui et al., 2015; Zhang et al., 2015).

5.4. Multiscale estimation of mechanical properties

As mentioned previously, the degree of polymerisation is a key method in the evaluation of the strength of cellulose fibres in cultural heritage fields, especially in the case of tapestries and canvases that are subjected to mechanical stress due to frames and/or their own weight due to the effect of gravity. This method has been used to evaluate the mechanical properties of Dalí's paintings by Oriola *et al.* In a set of several paintings, only one painting demonstrated DP <600 and was consequently considered at risk (Oriola et al., 2014).

An interesting and innovative method to study the strain and structural modifications of tapestries in a generalised manner is digital imaging correlation (DIC). Khennouf *et al.* (Khennouf et al., 2010), Malesa *et al.* (Malesa et al., 2011) and Malowany *et al.* (Malowany et al., 2014) mounted a system of two cameras capable of recording images and correlated the deformations of canvases and tapestries with time and environmental conditions.

Other mechanical tests are less commonly performed in the cultural heritage field but are widely used in the engineering domains. A commonly employed test is the tensile test on unitary fibres. Between 30 and 50 elementary fibres are extracted from bundles, and each fibre is glued to a plastic support or in a paper frame, which has a gauge length of 10 mm according to ASTM C1557 (*ASTM C1557-20*, 2020). The frame is successively mounted on a tensile testing machine, and a load cell of 2 N is used, which stretches the unitary fibre until rupture, allowing one to calculate its modulus and strength based on its diameter (Bourmaud et al., 2016). A larger number of elementary fibres leads to more reliable statistics and lower error; however, even few elementary fibres can indicate the fibre condition.

Tensile testing can also be performed for small pieces of fabric. Nechyporchuk *et al.* compared the mechanical behaviour of a fragment of an acrylic painting on canvas dated 15 y to a new modern canvas appositely prepared in the laboratory. The authors also considered the use of nanocellulose treatments to increase the canvas strength (Nechyporchuk et al., 2018). Recently, a combined method of digital imaging correlation and tensile testing has been applied to study historic tapestries made of silk (Rocha et al., 2018).

Nanoindentation is a characterisation technique where a tip of a hard material, often diamond, and a well defined shape penetrates into the sample surface and provide information regarding the mechanical properties of a sample at the nanoscale level. Notably, the use of this technique is increasing in the cultural heritage field (Faisal et al., 2018), for example, the nanoindentation analysis performed by Salvant *et al.* on cross-sections of *"Portrait du Docteur Paul Gachet"* and "*La Salle de danse à Arles"* of Vincent Van Gogh (Salvant et al., 2011). Additionally, Tiennot *et al.* investigated a cross-section from a canvas painting using nanoindentation, although the approach was different from that of Salvant *et al.*, as the authors mapped the mechanical properties of each tested layer and they concluded that the natural ageing process induce a stiffening of the painted layers (Tiennot et al., 2020).

5.5. Dating

The most commonly used dating method is radiocarbon dating using ^{14}C isotopes. Probably one of the most discussed and controversial works involving radiocarbon dating is that of the Turin shroud made from linen (Damon et al., 1989). It was recently concluded that a new radiocarbon analysis should be performed again in the Turin shroud only after the development of a stricter protocol

(Casabianca et al., 2019). In the already cited work of De Caro *et al.*, the use of a new dating method by WAXS was proposed with promising results (De Caro et al., 2019).

Bonizzoni *et al.* observed that more aged textile corresponded to a greater amount of fluorescence emission, and although the authors were cautious because they considered that fluorescence can also be generated from impurities, they used laser excited micro-fluorescence for dating and compared the results obtained using the μ-Raman spectra (Bonizzoni et al., 2016). To elaborate μ-Raman spectra, the authors adopted the ratio between peaks I_{1121}/I_{1196}, which correspond to the vibrational modes of symmetric and anti-symmetric stretching of C-O-C of 1,4-*β*-glycosidic bond of cellulose (Edwards et al., 2006; Jähn et al., 2002), and correlated the values obtained with the age of the textiles (Bonizzoni et al., 2016).

Another method for dating cellulosic materials is the chemical method implemented using enzymatic biosensors developed by Campanella *et al.* (Campanella et al., 2001, 2005). These biosensors can recognize methyl and carboxyl groups of cellulosic materials, the amount of which increase with ageing. Notably, the authors warned that this type of method depends on the artwork conservation conditions, and the same analysis performed on fibres extracted in different parts of a single painting may give different results (Campanella et al., 2005).

5.6. Towards cutting-edge techniques

New technologies are being constantly developed, allowing the investigation of materials already studied for centuries, such as plant fibres, with novel methodologies and combinations.

Atomic force microscopy (AFM) is a cutting-edge technique, which is based on a tip mounted on a cantilever with a laser focused on the probe reflected in a photodiode. The technique can be used in several modes, and the most commonly used application is to investigate the topography of ancient materials at the nanoscale level (Doménech-Carbó et al., 2009). AFM has also been used with a spherical probe functionalised with several consolidants, such as nanocellulose (CNF), before scanning the cotton fibres of an artificially aged canvas to test the adhesion between the treatments and cotton fibres (Bridarolli et al., 2018).

Another example pertains to Reynaud *et al.*, who coupled the AFM probe with IR spectroscopy to investigate 5,000 y old, mineralised flax fibres. The authors simultaneously obtained morphological and chemical information and clarified the mineralisation phenomenon (Reynaud et al., 2020). This particular system has already been tested on plant cell walls for botanical purposes (Pereira, Flores-Borges, et al., 2018).

In the engineering domain, another AFM mode known as the peak force quantitative mechanical property mapping (PF-QNM®) mode has been recently employed to scan cellulose fibres, as illustrated in **Figure 11.a** (Arnould et al., 2017; Goudenhooft et al., 2018). With a probe mounted on the cantilever, this technique allows to scan the sample surface and record force–distance curves that provide information on the mechanical properties of the sample at micro- and nanoscale levels. This method can distinguish the mechanical properties of thermally treated fibres used as reinforcement in composites (Siniscalco et al., 2018) and the effect of retting on the mechanical properties (Bourmaud et al., 2019) or cells with different mechanical properties at different stages of growth (Goudenhooft et al., 2018). Because this characterisation technique requires only a small amount of the sample (few millimetres of a yarn or few fibres) and can be used to investigate complex systems in which several materials are coupled, is promising and suitable to investigate samples from the cultural heritage field.

A first attempt at AFM PF-QNM analysis was recently carried out on flax yarns sampled from mortuary linen dating back to the Middle Kingdom of Ancient Egypt and from Italian paintings on canvas dated 17th-18th century, highlighting the ageing and degradation mechanisms at a new micro- and nanoscale level (Melelli et al., 2021b; Melelli et al., 2021c).

The second technique that is largely unknown in the cultural heritage and engineering fields despite its potential is the second-harmonic generation imaging microscopy (SHG). Based on a femtosecond pulsed laser in the NIR region, the non-centrosymmetric molecules in the sample can generate a second-harmonic response, a non-linear optic phenomenon, if they have a well-ordered structure. Cellulose microfibrils of plant fibres can generate SH, and several teams have already explored the ultrastructure of *Valonia* (Brown, Jr. et al., 2003; Nadiarnykh et al., 2007). Furthermore, it is possible to clarify the effect of acid hydrolysis on cellulosic materials (Peciulyte et al., 2016). For further explanations and other applications of this technique, the book of Pavone and Campagnola is suggested (Pavone & Campagnola, 2013). Reynaud *et al.* applied SHG to archaeological flax fibres that have undergone mineralisation, as illustrated in **Figure 11.b** (Reynaud et al., 2020). SHG analysis also highlighted fractures on kink-bands of ancient flax yarns (Melelli, 2021b) and clarified the presence of hydrolysis and biological attack on yarns sampled from linen canvases (Melelli et al., 2021c).

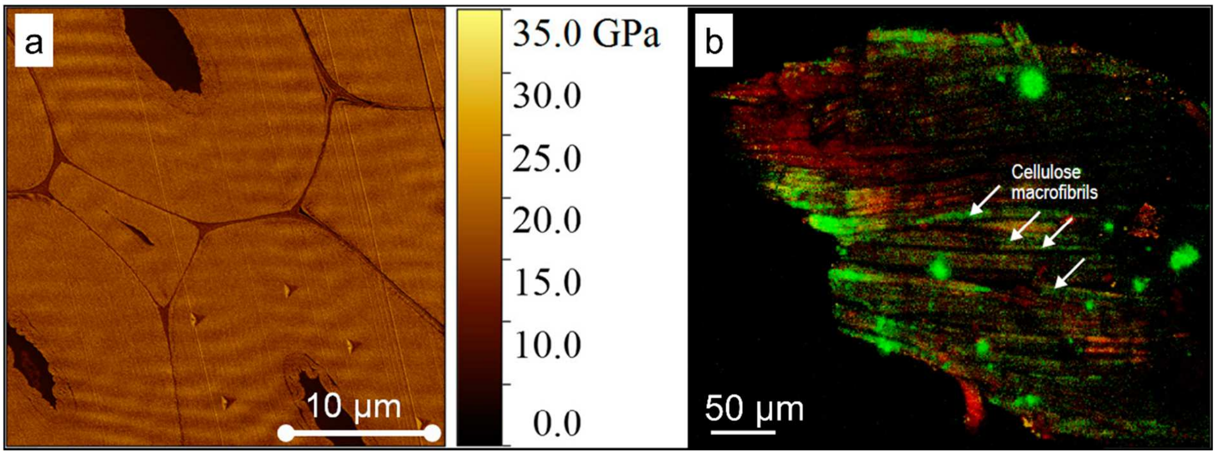

Figure 11. a) Map of indentation moduli of flax fibres obtained by atomic force microscopy in PF-QNM to study the mechanical properties at the cell wall level in (Goudenhooft et al., 2018). The mean indentation modulus is approximately 18 GPa; **b)** archaeological mineralised flax fibres investigated by second-harmonic generation microscopy from (Reynaud et al., 2020). Cellulose macrofibrils from the internal fibre structure are highlighted.

There are three advantages of using this technique: i) a single yarn or few fibres can be analysed, ii) the approach is a non-destructive method because an infrared excitation wavelength is used, and iii) the method can be used to scan a sample in-depth (Z-stack), thereby providing information regarding the inner structure of the fibres, complementary to the SEM analysis. Synchrotron radiation is a technique that is gaining increasing attention. FTIR vibrational spectroscopic analysis can be performed using synchrotron radiation to characterise ancient plant fibres. The work performed by Kavkler *et al.* on biodegraded historical textiles is a representative example: the authors compared conventional FTIR microspectroscopy and synchrotron FTIR microspectroscopy techniques (Kavkler, Šmit, et al., 2011).

In addition, at Synchrotron SOLEIL, the *Dichroism, Imaging and mass Spectrometry for Chemical and biOlogical systems* (DISCO) beamline has been specialised in the use of deep-UV (DUV) fluorescence to characterise plant cell walls. The autofluorescence present in plants due to waxes, protein, lignin and phenolics can be exploited through multi-spectral fluorescence imaging to compare cross-sections of flax stems grown in normal conditions or under gravitropism (Beaugrand et al., 2022) or to track the effects of enzymes in maize (Devaux, 2018). Maps of spectra can also be recorded by scanning fluorescence microspectroscopy and used to compare the chemical composition in different cells of the stems (Jamme et al., 2013) as well as historical artefacts, as in the investigation of the coating of ancient lutes (Echard et al., 2015).

Finally, tomography is a characterisation technique useful for understanding the internal structure of samples and virtually reconstructing such structures. Using X-rays, it is possible to obtain high-resolution tomographic images. Single flax fibres (Abbey et al., 2010; Eve et al., 2012) and bundles or fabric, such as mineralised archaeological textiles (King et al., 2019), have already been examined using this method, especially in the cultural heritage field. For details regarding the use of synchrotron radiation for plant investigation, the review written by Vijayan is suggested (Vijayan et al., 2015).

6. Summary and conclusions

This review describes the historical timeline and problems encountered in the cultural heritage and engineering fields when flax fibres are employed to create objects.

The first part of the review summarises the composition and fibre structure of flax fibres, their mechanical properties and limitations from the perspectives of cultural heritage applications. Notably, certain aspects, often interconnected, such as the microfibril angle, kink-bands and mechanical properties of the middle lamella between fibres, are known to be still not fully understood by the scientific community. The microfibril angle is one of the most important parameters that influence the mechanical properties of plant fibres; however, the real value for plant fibres and the impact of kink-bands on the microfibril network is still debated. The first part also briefly describes plant growth and all the steps from sowing until maturity.

The second section discusses the history of flax cultivation and processes from the past to the contemporary era. This section highlights how and why the centres of flax production changed over the centuries in Europe and the Middle East which, in certain cases have been caused by episodes of drought or due to the adaptability of soils. The challenges in agricultural practices, especially in the case of flax crops in France, encountered due to climate change and increasingly frequent periods of drought are of interest even today. This section also treats the evolution of flax varieties and the methods used to extract the fibres from the first flax domestication to the present day. Today it is known that the extraction method impacts on the mechanical properties of flax fibres, in particular for the generation of kink-bands, so the study of ancient extraction methods can teach us something more about the fibre ultrastructure and its response to several mechanical stress made with various tools. Moreover, some uses of flax fibres have evolved, for example from sails used in antiquity to whole boats built today with flax/resin sandwich, and others are still the same, like the linen used in fashion. Problems such as the impermeability of textiles had already been faced by ancient populations and the study of artworks can help us to rediscover forgotten techniques and to consider other alternatives to the techniques currently used.

The third part of this review summarises the main degradation process of flax fibres, which is caused by several factors, such as water,temperature and UV, with a focus on artworks. These degradation processes, which have been well studied in the cultural heritage field to preserve historical objects, are the same as those that flax fibres undergo when they are employed in the engineering field. Thus, artworks that have survived for centuries can be examined to predict the ageing process in new industrial objects.

The final section provides a brief overview of the main characterisation techniques used to investigate flax fibres and several cutting-edge techniques currently employed in engineering and botany fields, such as AFM and SHG, that can also be used to investigate historical textiles. Thus, a new combination of techniques can provide novel insights regarding the ultrastructure and degradation process of cellulosic fibres. The findings are expected to be useful to evaluate their state of preservation and possible treatments. Future research should be focused on developing and adapting these techniques to the field of cultural heritage.

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Figures captions

Figure 1. a) schema of a young flax stem at approximately 400 °C GDD and mature stem at 1000 °C GDD (© Erica Melelli); b) photo of two flax plants pulled out at 400 °C and 1000 °C GDD in the real scale; c) plant growth over time with flowering and browning of the capsules

Figure 2. a) Schema of flax fibre ultrastructure and chemical composition, inspired and partially readapted from (Gorshkova et al., 2018; Manian et al., 2021) and data from (Goudenhooft et al., 2019b); b) formation of the cell wall layers, inspired by (Goudenhooft et al., 2018), and the progressive filling and thickening with the Gn layer transformed to G; c) transversal section of a flax fibre bundle, in which the middle lamella is highlighted in yellow. A schema representing the cell corner (CC) between three fibres (F) is shown, along the compound middle lamella (CML) composed

of the primary wall (P) and middle lamella between two adjacent fibres; d) bundle of two fibres (F1 and F2) with a visible kink-band and outermost primary cell wall layer.

Figure 3. Map with the natural distribution of wild (pale) flax (in green) and the origin of the use of flax and its spread in Europe along with the first fabrics dated to the early Neolithic. The distribution of pale flax (green) has been obtained from (Desta, 2019; Diederichsen & Hammer, 1995; Gutaker et al., 2019). The centres of archaeological findings have been adapted from (Colledge & Conolly, 2017; Harris, 2014; Karg, 2011).

Figure 4. Dendrogram from (Muravenko et al., 2003). L. angustifoleum appeared as the progenitor of both L. bienne and L. usitatissimum.

Figure 5. Details of wall painting from Tombs of Dagi (TT103), where two women are shown to scutch flax stems (from the left) and probably splice flax into threads (right). Illustration from (Vogeslang-Eastwood, 2000)

Figure 6. a) Left: possible reconstruction of the use of a wooden tool from Sweden to beat the flax stems on a plane surface (rock or wood). The tool is schematised from the archaeological tool (Item 447267. SHM 23159: K12) displayed at the Historiska Museet, original photo © Gabriel Hildebrand and right: another shape of a wooden club inspired from (Ejstrud et al., 2011; Viklund, 2012), and a schema of the breaking tool mounted on a stand found in Norway and reported in (Ejstrud et al., 2011); **b)** from the left: schema of a wooden knife found in Gloucestershire and dated to the middle Bronze Age (original photo © Cotswold Archaeology), and reconstruction of a heckle dated between 850–1000 AD found in Shetland, Scotland (Item X. HSA 318 © National Museums Scotland); c**)** schematic representation of the Miniature of the Book of Hours, MS M.399 fol. 12v, ca. 1515, Belgium (original photo © The Morgan Library & Museum, New York). On the left, a woman scutches a bundle of flax fibres with a wooden tool and a wooden column (highlighted in red) while two men beat the stems with breaking tools; **d)** Detail of the “Livre d'heures” L’Escalopier, hours in Latin with calendar, parchment of 16th century, f. 12 Ms LES 22 A, reproduced with the kind permission of © Bibliothèques d'Amiens Métropole and IRHT-CNRS.

Figure 7. Four lithography by William Hincks, dated 1783 **a)** Plate II “View near Hillsborough in the country of Downe, representing pulling the flax when grown, hooking or putting it up to dry, rippling or saving the seed, and bogging or burying it in water”. Two women remove the capsules and prepare the stems for water retting, while a man places bundles of stems in water. In the background, two other women harvest the flax plants, group them in bundles and place them upright with respect to the ground to dry before the retting step. **b)** Plate III “View in the county of Louth

representing taking the flax out of the bog”. After the retting process, the retted stems are moved near a campfire to be dried. In the background, two men are breaking the dried stems; **c)** Plate IV “The common method of beetling, scutching and hackling the flax”. A family is represented as breaking the stems and combining the fibres; **d)** Plate V “A perspective view of a scutch mill, with the method of breaking the flax” represents one of the earliest industrial scutching mills. The four images are reproduced with the kind permission of the © British Library Board.

Figure 8. a) Taproot extraction line with three modules, and a schematic representation of the steps involved in general industrial extraction lines: 1. breaking module, 2. scutching module and 3. combing module; **b)** biocomposite made of flax fibres for automotive (© Ecotechnilin) and **c)** biocomposites reinforced with flax fibres employed for applicators used in cosmetic showrooms (© Kaïros).

Figure 9. a) Bacterial and **b)** fungal colonisation on the surface of over-retted fibres and **c)** silverfish *Ctenolepisma lineata* (© Erica Melelli). Unpublished images.

Figure 10. a) Elementary flax fibre under optical microscopy and diameter calculation **b)** elementary flax fibre under polarised light that highlights kink-bands in white (©Emmanuelle Richely) and **c)** SEM microscopy of an elementary flax fibre with three visible kink-bands.

Figure 11. a) Map of indentation moduli of flax fibres obtained by atomic force microscopy in PF-QNM to study the mechanical properties at the cell wall level in (Goudenhooft et al., 2018). The mean indentation modulus is approximately 18 GPa; **b)** archaeological mineralised flax fibres investigated by second-harmonic generation microscopy from (Reynaud et al., 2020). Cellulose macrofibrils from the internal fibre structure are highlighted.

Tables captions

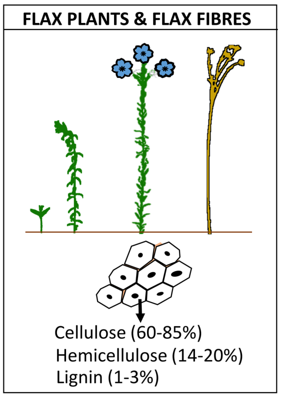
Table 1. Literature review of the biochemical global composition of the considered bundles of fibres.

Table 2. Typical mechanical properties of E-glass compared to flax, hemp and cotton. Mean of the values modified from (Wambua et al., 2003; Hughes et al., 2007; Graupner, 2008; Lu et al., 2012; Fidelis et al., 2013; Lu & Oza, 2013; Célino et al., 2014; Bourmaud et al., 2018).

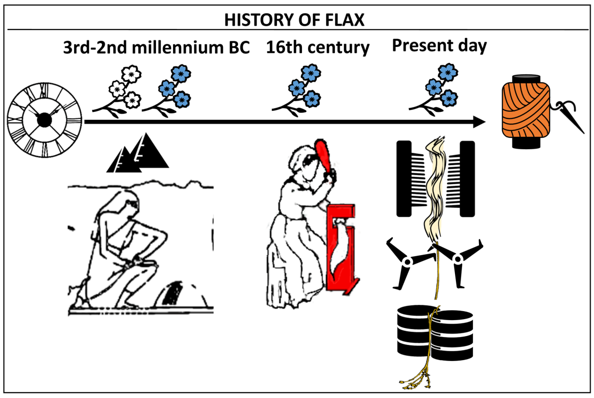
Table 3. Summary of the world production of flax fibres and tow in year 2019. Data from FAOStat (Crops and Livestock Products: Flax Fibre and Tow, 2021)

Table 4. Five organoleptic criteria to establish the flax fibre quality. Table readapted from (Lefeuvre, 2014)

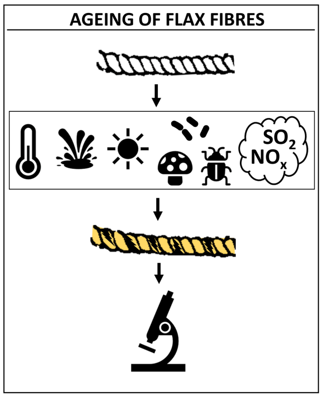
Table 5. Fungi and bacteria in common between fresh retted flax stems and cultural heritage objects made of cellulosic fibres. Table adapted from (Djemiel et al., 2020; Nugari et al., 2007; Repeèkienë et al., 2007)



FLAX PLANTS & FLAX FIBRES



HISTORY OF FLAX



AGEING OF FLAX FIBRES