Sustainable Tree Bark Objects by Combining Science and Design

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Abstract

Bark, the boundary between trees and their environment, comprises about 10–20% of their total volume. Approximately 4 million m^3 of bark is collected annually in German sawmills. Only a small part of this bark is used as an end product; the majority is burned wet in sawmills due to reduced economic interest, a lack of production facilities and a lack of knowledge about other uses, as compared to wood. Reasons for this are related to the high variability of bark properties between and among tree species and even within one tree. The aim of this thesis is to understand the material properties of selected types of bark through scientific and design methods and to explore processing possibilities that lead to longer lifecycle applications and cascade uses.

In the beginning of this thesis, bark from species endemic to Brandenburg (northeast Germany), Scots pine (*Pinus sylvestris,* L.), English oak (*Quercus Robur,* L.), European larch (*Larix decidua*, Mill.) and European white birch (*Betula pendula,* Roth), were manually peeled to obtain large pieces of bark, which were afterwards air dried and modified with a variety of processing techniques.

In the first procedure, two or more pieces of peeled bark were placed crosswise, the rhytidome (outer bark) facing each other, and hot pressed into panels. First, mechanical tests showed that bending stiffness, bending strength and transverse tensile strength are similar to wood-based panels such as particle board. A strong effect of the fiber orientation on the bending properties was detected. Experiments on raw bark with specifically adjusted water content during the pressing process showed effects on swelling behavior and mechanical properties. In the transverse direction, large swelling was observed with increasing relative humidity. This swelling remained permanent, even upon drying, for some bark. However, bark panels appeared smooth after the pressing process, with surface roughness similar to solid sanded wood. This surface quality and their machinability with conventional processing methods, such as sawing and computerized numerical control milling gives them potential furniture and paneling applications. Additionally, densification into more complex forms were realized with oak bark.

In a second approach, a procedure was developed to preserve flexibility and to keep the texture and color of freshly peeled bark. The presented experiments show that it is possible to flexibilize mirror pine bark and the phloem of larch bark, but oak and birch could not be flexibilized. The successful flexibilization of mirror pine bark is presumed to be based on the high

proportion of conductive phloem. The effect of glycerol water solutions on the mechanical properties of bark were quantified by performing tensile tests. Interestingly, bark samples treated with glycerol showed improved flexibility and stress at failure compared to wet bark. To obtain more information about the influence of glycerol water treatment on mechanical properties of pine bark, tensile tests were performed with adult spruce wood and compression wood. A remarkable difference between bark and wood is that the glycerol treatment of wood does not lead to larger tensile strain compared to water immersion. This finding suggests that pine mirror bark has a high cellulose microfibril angle (like compressed wood). Comparative tensile tests with two different sets of cowhide leather samples showed that flexible pine bark is more than five times stiffer than leather when stressed along the grain – even though the leather and bark samples showed similar haptic feel, flexibility and processing possibilities.

The third procedure explores possibilities of using bark ashes – a final waste product of the wood processing industry. Tree bark is known for its different chemical composition compared to wood. Selected bark ashes were analyzed by inductively coupled plasma (ICP) mass spectrometry and then mixed with transparent porcelain glazes and fired. The bark ash glazes gave colored glazed bowls, with colors characteristic of the elements potassium and manganese: beige, yellow, rose and deep dark brown. Calcium acted as a flux and produced net-like surface patterns in the spruce ash glaze.

Finally, the processes developed during the thesis were successfully transferred into design prototypes. By using glycerol, it was possible to flexibilize pine bark, allowing for different textile design objects. Flexibilized pine bark – based on its protective function for trees – was applied to fashion in the form of a jacket, and further optimized through weaving techniques. In addition, shoes with robinia bark soles were manufactured, highlighting further applications for tree bark. Optimized processing techniques open up the possibility of larger-scale flexibilized bark applications in architecture. To reflect the potential of bark in architectural applications, an installation which people could stand inside and experience being completely surrounded by bark was built. In form of a hollow walk-in sphere, three-dimensional fabrication – largely free of the geometric limitations of the starting material (bark) – was demonstrated. The abstract form of a sphere leaves enough space for free associations and encourages communication and exchange about bark as a possible future design material.

Zusammenfassung

Rinde ist die Schutzschicht des Baumes. Sie bewahrt das für den Baum lebenswichtige Kambium vor Umwelteinflüssen (Hitze, Kälte, Strahlung) und Fraßfeinden. 10- 20% des Gesamtvolumens eines Stammes sind Rinde. Wird ein Baum für industrielle Zwecke gefällt, wird aus Rinde meist Abfall. In deutschen Sägewerken fallen jährlich etwa 4 Millionen m^3 Rinde an. Nur ein kleiner Teil dieser Rinde wird weiterverarbeitet. Der größte Teil wird in den Sägewerken nass verbrannt. Das wirtschaftlich untergeordnete Interesse an Rinde sowie die hohe Variabilität der Rindenstrukturen untereinander und sogar innerhalb eines einzelnen Baumes sind Gründe warum dieses Material im Vergleich zu Holz noch recht unerforscht ist. Das Ziel dieser Arbeit war es mit Methoden aus der materialwissenschaftlichen Grundlagenforschung und dem Design, ausgewählte Rindenarten zu untersuchen und die gewonnen Erkenntnisse in hochwertige Anwendungsmöglichkeiten im Bereich Design und Architektur einfließen zu lassen.

Rinde von brandenburgischer Waldkiefer (Pinus sylvestris, L.), Stieleiche (Quercus Robur, L.), Europäischer Lärche (Larix decidua, Mill.) und Europäischer Weißbirke (Betula pendula, Roth) wurde manuell geschält, um große Rindenstücke zu erhalten. Nach der Ernte wurde die Rinde an der Luft getrocknet und mit verschiedenen Verfahren bearbeitet.

Im ersten Verfahren wurden zwei oder mehrere Rindenstücke mit der Borkenseite zueinander gewandt und in Faserrichtung um 90° verdreht und in einer Heißpresse zu Platten verpresst. Mechanische Tests zeigten, dass eine Biegesteifigkeit und -festigkeit sowie die Querzugfestigkeit ähnlich wie bei Holzwerkstoffen wie Spanplatten erreicht werden konnten. Es wurde ein Einfluss der Faserorientierung insbesondere auf die Biegeeigenschaften festgestellt. Versuche mit unterschiedlichen Wassergehalten in der unverpressten Rinde zeigten, dass die Feuchtigkeit während des Druckprozesses das spätere Quellverhalten und die mechanischen Eigenschaften beeinflusst. Eine Erhöhung der relativen Luftfeuchtigkeit führte zu einem stärkeren Quellen der Platten in Querrichtung. Teilweise blieb diese Quellung auch nach dem Trocknen dauerhaft bestehen. Ein positiver Effekt, der Verpressung, ist die hohe Oberflächenqualität der Rindenplatten. Jene ist vergleichbar mit geschliffenen Oberflächen von Massivholz und könnten für den Möbelbau interessant sein. Zusätzlich wurde das Verpressen in 3D-Formen mit Eichenrinde realisiert.

Im zweiten Verfahren wurde Baumrinde mit einer Glycerinwasserlösung infiltriert. Dadurch behielt die Spiegelrinde der Kiefer auch nach dem Trocknen ein gewisses Maß an Flexibilität.

Eiche und Birke ließen sich nicht und Lärche nur teilweise flexibilisieren. Es ist anzunehmen, dass für die Infiltration ein hoher Anteil an leitendem Phloem nötig ist sowie eine glatte mit wenigen Borkenschuppe übersäte Rinde, die eine freie Bewegung in alle Richtungen zulässt. Ein bemerkenswerter Unterschied zwischen Rinde und Holz bestand darin, dass die Glycerinbehandlung von Rinde zu größeren Dehnungen führte als die Wasserlagerung. Dies deutet darauf hin, dass dieser Effekt nicht mit Veränderungen in den verholzten Zellwänden zusammenhängt, wie dies bei Holzzellen der Fall ist, sondern dass Spiegelrinde der Kiefer einen potenziell hohen Zellulose-Mikrofibrillenwinkel (wie Druckholz) aufweist. Sowohl das haptische Gefühl als auch die Verarbeitungsmöglichkeiten von flexibilisierter Rinde waren mit Leder vergleichbar. Weitere Zugversuche mit zwei verschiedenen Lederproben zeigten, dass die flexible Kiefernrinde bei Beanspruchung in Faserrichtung mehr als fünfmal steifer ist. Die höheren mechanischen Eigenschaften von Leder hängen vor allem mit dessen Struktur und chemischer Zusammensetzung zusammen, die sich grundlegend von Baumrinde unterscheidet.

Das dritte Verfahren - die Verwendung von Rindenasche, war durch die Idee motiviert, alle Reste der natürlichen Ressource Rinde zu verwenden. Baumrinde ist für ihre im Vergleich zu Holz unterschiedlichen chemischen Zusammensetzungen bekannt. Verschiedene Arten von Baumrindeaschen wurden mittels Massenspektrometrie mit induktiv gekoppeltem Plasma (ICP) untersucht und anschließend mit transparenten Porzellanglasuren vermischt und gebrannt. Die Rindenglasuren zeigten farbig glasierte Schalen, in für die Elemente Kalium und Mangan charakteristischen Farben beige, gelb, rosa bis hin zu dunkelbraun. Calcium fungierte als Flussmittel und erzeugte netzartige Oberflächenmuster in der Fichtenascheglasur. Insgesamt ließen sich die Ergebnisse nur vereinzelt reproduzieren.

Die entwickelten Verfahren wurden erfolgreich in Designprototypen überführt. Flexibilisierte Baumrinde konnte – angelehnt an die biologische Schutzfunktion – für Modeanwendungen in Form einer Jacke gezeigt und durch den Einsatz von Webtechniken optimiert werden. Außerdem wurden Schuhe mit Robinienrindensohle gefertigt, die weitere Anwendungsmöglichkei- ten für Baumrinde eröffneten. Um das Potenzial von Rinde in architektonischen Anwendun- gen zu beleuchten, wurde eine Installation/ Kugel gebaut, in der Personen stehen und die Erfahrung machen können, vollständig von Rinde umgeben zu sein. In Form einer begehbaren Hohlkugel wurde eine dreidimensionale Fertigung - weitgehend frei von geometrischen Begrenzungen (des Ausgangsmaterials Rinde) - demonstriert. Die abstrakte Form einer Kugel lässt genügend Raum für eigene Assoziationen und kann die Kommunikation und den Austausch über Rinde als mögliches Material für zukünftige Anwendungen fördern.

Contents

1 Introduction

1.1 Motivation: Waste Material from the Wood Processing Industry

"Cellulose is the most abundant polymer on Earth and trees are a major part of this biomass. Modern forestry technologies concentrate on harvesting stems as their most valuable part, leaving reaction wood, branches and bark as forest residues and waste wood. The main reason is that the ultrastructure of the material outside the main part of the wood is more variable and, therefore, more costly to directly put into use. Some typical usages of trees are shown in figure 1. Plants grow in an adaptive way depending on the local environmental conditions, such as the land slope, side winds or other mechanical challenges. Trees develop reaction wood in some parts of the stem as well as in branches to compensate for loads that change over time (Du and Yamamoto 2007). The resulting properties of the wood material are, therefore, more variable in these regions and more difficult to put into commercial use."[1]

"While bark and softwood branches are typically only used for low-value applications and as an energy source, they appear to be understudied compared to stem wood, which is often used for advanced applications (Fig. 1.1). Wood is one of the oldest useful materials of mankind: knowledge of the properties of various woods is documented already for the Neolithic period, i.e. 5000 years ago (Kraft 2009). This ancient tradition is still contained in the etymology of the term, insofar as the ancient Greek term Ὕλη (hyle, Engl.: matter/material) or in Latin translation materia and materies primarily meant wood, both the forest and the material (timber) that was used for the construction of houses, ships and as ship masts (Detel 1980). In this respect, wood stands in as an 'archetype' for both the general concept of matter and for that of functional material (Bensaude-Vincent 2011). Practical access to the wood as material ranges from the mythical 'primitive hut' referred to in architectural theory since Vitruvius (first century BC) (Vitruvius 1867) to modern timber building systems of prefabricated modules such as the 'Packaged House System' by Konrad Wachsmann and Walter Gropius (1940s), which could 'grow' additively in all directions, i.e. be extended. What is changing,

[1] *Wenig, C., Dunlop, J. W., Hehemeyer-Cürten, J., Reppe, F. J., Horbelt, N., Krauthausen, K., … & Eder, M. (2021). Advanced materials design based on waste wood and bark. Philosophical Transactions of the Royal Society A, 379(2206), 20200345*

however, is the way the material is handled: while it was initially an object of selection due to its complex structure and consequent properties (hygroscopicity, elasticity, inhomogeneity), in the twentieth century it was often used in a decontextualized way, i.e. homogenized into a passive building material (e.g. medium density fiber board) by crushing and in combination with resistant adhesives (Kraft 2009). Only recently have attempts been made to transfer the structural potential of wood (its ability to react to environmental conditions) into constructions, for example in the experimental architectural projects of Menges & Reichert (Menges and Reichert 2015)."[2]

"Natural, unmodified wood is again a material with highly desirable properties. Not only is it aesthetically pleasing, with applications in furniture making, and the production of veneers (a semi-finished product), it has a low thermal conductivity and can act as an insulator. Wood's high mechanical performance means that it is used as a structural material for constructions and buildings, especially when loaded along the fiber direction. For applications requiring a minimal mass for a given performance, such as beams in construction, wood outperforms most synthetic materials and at much lower cost (Ashby 2005), (Wegst and Ashby 2004). Besides approaches to improve mechanical properties, numerous modification methods for a higher dimensional stability, durability, etc., are researched and applied (e.g. (Hill 2007)). In summary, both natural wood and modified wood with excellent mechanical properties and comparably low biological variability are retrieved from mature stems, ideally having a low curvature of the tree rings and a small micro fibril angle (MFA). However, not all parts of trees can be used for the described 'high value applications' due to their geometries or material properties. The side cuttings of the sawed tree stems and the upper parts of trees with low diameters, low densities and larger MFAs are typically classified as industrial wood (Fig. 1.1). Industrial wood is reduced to smaller pieces ranging from strands to small chips and re-assembled to different types of panels (e.g. OSB, particle board). A considerable amount of industrial wood is raw material for making pulp, an energy and chemistry-intensive process for further disintegration involving the removal of lignin. The quality of pulp depends on the applied process (e.g. sodium sulfate, sodium sulfite process) but also on the raw material. To give an example, softwood pulp with much longer fibers than hardwood pulp is more suited for high-strength paper as used for packaging. Besides being the raw material for

[2] *Wenig, C., Dunlop, J. W., Hehemeyer-Cürten, J., Reppe, F. J., Horbelt, N., Krauthausen, K., … & Eder, M. (2021). Advanced materials design based on waste wood and bark. Philosophical Transactions of the Royal Society A, 379(2206), 20200345*

fiber-boards or paper, pulp is the starting material for wood-based textile fiber production (e.g. viscose and lyocell) but also for the production of cellulose nanocrystals (CNCs), which are used and researched for biomedical devices, nanocomposites, as emulsions, gels and foams (Vanderfleet and Cranston 2021) and for optical materials due to their ability to self-assemble (Tran, Boott, and MacLachlan 2020)."[3]

The given examples illustrate the wide variety of how wood is currently used. The structure and composition of the raw material as well as the final product is essential for the performance, and the geometric element 'fiber', found at various length scales (cellulose fiber at the molecular level and fiber at the microscopic level), is the key parameter for the material properties."[4]

"The examples show that tree material with predictable properties such as stem wood is typically used in an almost unmodified way. With increasing biological variability as observed in, for example, branches, the degrees of disintegration and modification of the raw material for the desired applications increase, involving environmental footprints which are possibly not neglectable and require further research effort to fully quantify them. For example, it was shown recently that for CNCs, the environmental impact by water and energy consumption, carbon footprint and missing biorefinery concepts were unclear with hardly any comprehensive lifecycle assessments available products made of these renewable raw materials (Vanderfleet and Cranston 2021)."[5]

[3] *Wenig, C., Dunlop, J. W., Hehemeyer-Cürten, J., Reppe, F. J., Horbelt, N., Krauthausen, K., … & Eder, M. (2021). Advanced materials design based on waste wood and bark. Philosophical Transactions of the Royal Society A, 379(2206), 20200345*

[4] *Wenig, C., Dunlop, J. W., Hehemeyer-Cürten, J., Reppe, F. J., Horbelt, N., Krauthausen, K., … & Eder, M. (2021). Advanced materials design based on waste wood and bark. Philosophical Transactions of the Royal Society A, 379(2206), 20200345*

[5] *Wenig, C., Dunlop, J. W., Hehemeyer-Cürten, J., Reppe, F. J., Horbelt, N., Krauthausen, K., … & Eder, M. (2021). Advanced materials design based on waste wood and bark. Philosophical Transactions of the Royal Society A, 379(2206), 20200345.*

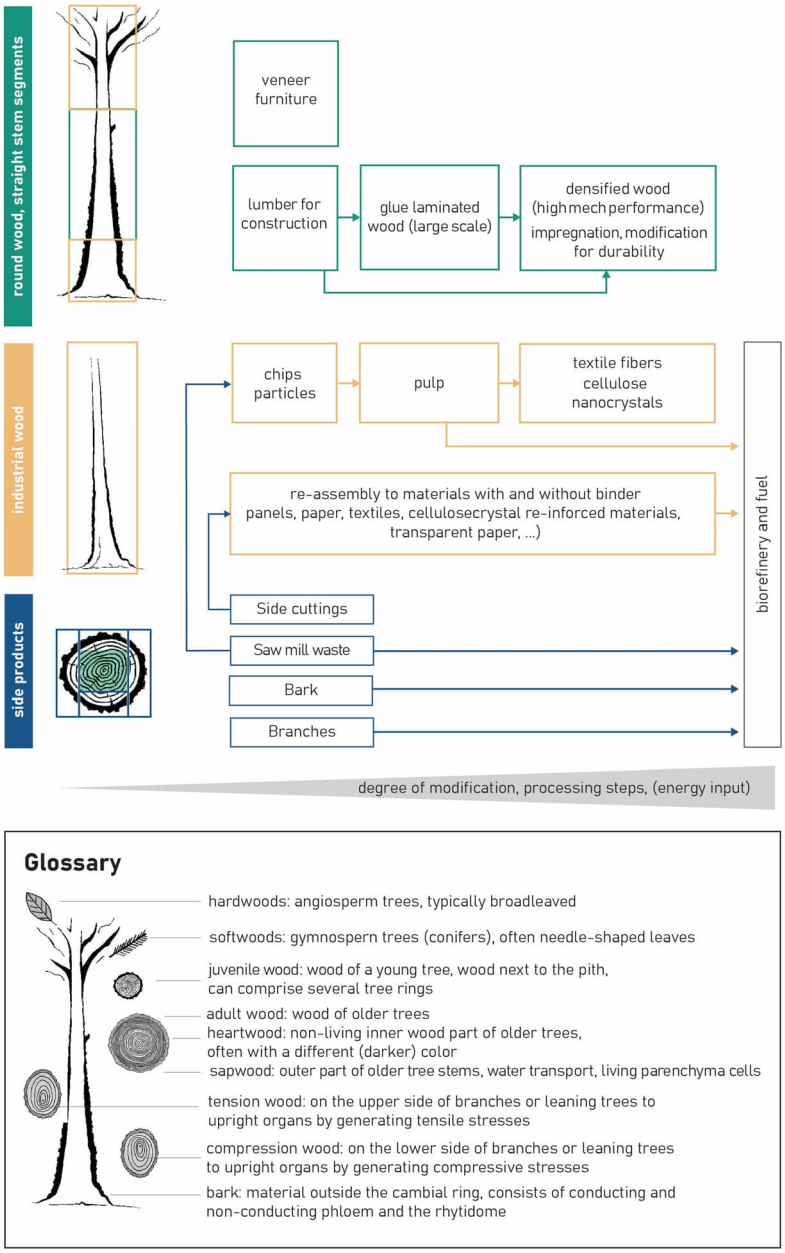

Figure 1.1: *Tree material use. The trees on the left side illustrate an old tree (top) and a young tree (below). The disc of the stem illustrates proportions of wood and bark and side cuttings. The chart of tree material use shows possible applications as well as treewaste (parts of trees) which go directly into the biorefinery or are used as fuel. The glossary at the bottom describes different parts of the tree with hardwood reaction wood on the left and softwood reaction wood on the right (excentric cross-sections).*[6]

[6] *This graphic is based on "Advanced materials design based on waste wood and bark", by Wenig et.al, 2021*

The research fields which are presented above, show the effort undertaken to generate more value for all parts of wood. Side products like bark are economically less important and less studied than wood. Reason for this can be the more complex structure and the higher variability of bark structure (inter- and intra-species wise) compared to wood. High variability evolves from a subsequent change through cell divisions and cell growth of/between the primary and secondary tissue (phloem). The proportion of bark in tree trunks is in the range of ~ 10 - 20 % (1). Currently industrial use of bark is in the range a low cascade usage. That means that bark is either directly burnt or used for horticulture, chemical extractions or as an additive in the wood panel production.

Tress are renewable/biological resources associated with land use and also become subject to scarcity caused by climate change. Consequently, it is necessary to use all parts of a tree as long and as efficiently as possible.

The motivation of this thesis is to focus on bark as a relevant raw material for the production of a long lifecycle of bark by the exploration of possible high cascade usages of different locally sourced barks.

1.2 Comprehensive Understanding: Science and Design for a Better Material Understanding and Use

As described in the previous section, bark of trees is mainly seen as low value or even waste material. There are many reasons for this. For example, there are minor production facilities and a lack of knowledge about the material bark compared to wood. Dealing with such complex problems is usually approached by one profession (e.g wood engineering, product design, biology, etc.). As a result, questions can become one-sided and solutions are usually found within the scope of this one profession. By combining science and design, the motivations to understand the structure of waste materials (bark) and find new fields of application (with bark) converge. For example, tree bark properties can be characterized by applying scientific methods. This knowledge about characteristic can be used in further fields of architectural and design applications.

Combining basic material characterization with application scenarios require upscaling approaches from micro- to macrostructures. For this scale transfer, the knowledge of both sides

must be combined. One example of this is the effect of water on the properties of bark. Different types of tree bark swell and shrink differently in different anatomical directions. Researching swelling properties can help for the appropriate construction of objects made of tree bark and potential areas of application (indoor or outdoor). The scientific and design connections result in a comprehensive understanding in which both sides learn from each other.

1.3 Objectives and Scope of Work

From this comprehensive understanding, the following hypothesis is generated: By analyzing the structure and properties of local bark species, it is possible to find applications for tree bark, adding value to the raw material. For this purpose, it is necessary to understand: i) the material structure and composition of investigated barks, ii) which processing methods from craft and industry could be adapted and applied, iii) how bark reacts to these methods and how the modification changes the material characteristics of bark, iiii) in which application fields tree bark could be used as a material for the design of applications.

2 State of the Bark

In the following chapter, the structural composition of tree bark and wood is presented (chapter 2.1). Following, the main types of tree bark structure are presented and explained (chapter 2.1.1). The four relevant tree bark species for this thesis, are presented and their structures are described in the chapters 2.1.3 – chapter 2.1.6. In the following, the techniques of debarking (chapter 2.2.1), the chemical components and their application (chapter 2.2.2) as well as established industrial processing methods of tree bark are discussed (chapter 2.2.3, chapter 2.2.4). In the subchapter objects (2.3), the use of tree bark in history and in the current design are presented.

2.1 Structure and Properties

2.1.1 Structure of Wood and Bark

"Plants make up by far the largest amount (approx. 80%) of biomass on Earth (Bar-On, Phillips, and Milo 2018). Their sizes range from small unicellular organisms such as green algae to the heaviest known organism, the giant aspen clone 'Pando' (Grant, Mitton, and Linhart 1992; Mitton and Grant 1996). Parts of the living organism, the plant, can be seen as and are used as materials, with the proportions of useful material increasing with growth. But what is a 'plant material'? Typically, plant materials are taken to be materials derived from or extracted from plants. These plant materials can include seeds/husks (e.g. cotton, kapok, coir), stems (wood, flax, hemp, bamboo, sugar cane), bark (cedar bark, cork of oak, birch bark), leaf fibers (sisal, agave) or roots (spruce root, willow root, cedar root), waxes and resins."[7]

"To expand this viewpoint, we first look at the cell, the smallest living building block. One characteristic of plant cells is the presence of a cell wall, located outside the plasma membrane. The plasma membrane contains protein complexes, called cellulose synthase units (CESAs), that move within the membrane to produce and secrete stiff, strong and long cellulose micro fibrils on the outside of the cell. Hemicelluloses and pectins are synthesized by enzymes localized in the Golgi and transported by the endomembrane system towards the plasma membrane (Sinclair, Rosquete, and Drakakaki 2018). The spatial arrangement of the cell wall

[7] *Wenig, C., Dunlop, J. W., Hehemeyer-Cürten, J., Reppe, F. J., Horbelt, N., Krauthausen, K., ... & Eder, M. (2021). Advanced materials design based on waste wood and bark. Philosophical Transactions of the Royal Society A, 379(2206), 20200345*

polymers, together with growth directions of the tissues and external restrictions of the neighboring cells, determines cell shapes and geometries (Sapala et al. 2018). After cell growth has ceased, many cells form secondary cell walls—composites of cellulose, hemicelluloses and lignin, a complex polymer consisting of phenolic precursors. Compared to the primary cell walls, secondary cell walls are much thicker and the cellulose fibrils are arranged mainly parallel to each other (Salmén 2004b)—at least in fibrous cells. Many cells with secondary cell walls die shortly after formation allowing them to provide a function to the plant in water transport and mechanical stability. Agglomerations of dead cells, such as large proportions of wood or bark, can thus be seen as functional material even though they are still essential parts of a living organism.

Trees, due to their abundance and large size, are of particular interest in the context of plant materials. Despite their size, large proportions of trees—wood fibers, tracheids, vessels and the rhytidome (Figure 2.1)—consist of dead cells with secondary cell walls. The cellulose microfibrils, indicated as black lines in figure 2.1 (bottom circles and diagram), are the essential building blocks of the cell walls. With a very high stiffness (greater than 120 GPa) and strength (7GPa) (Salmén 2004b), (Moon and Martini), they are reinforcing structural elements, akin to the stiff glass fibers in glass-fiber-reinforced polymers. Their spatial arrangement with respect to loading directions (diagram in Figure 2.1) determines mechanical (Eder et al. 2013) but also swelling properties (Scheuring, Boudier, and Sturgis 2007),(Perré and Huber 2007). Cellulose orientation is defined by the microfibril angle (MFA), given by the angle of the majority of cellulose microfibrils with respect to the longitudinal axis (Figure 2.1). The MFA plays an important role, especially in fibrous cells, which are abundant in wood and to a lesser extent in bark. These cells can also be found in fruits and other seed containers. Cells with a small MFA are stiff and strong in tension, while cells with a large MFA are less stiff and strong (Eder et al. 2013). For other cells, such as isodiametric cells, pavement cells or three-dimensional puzzle cells, relatively little is known about the cellulose orientation and its role on the mechanics of cells.

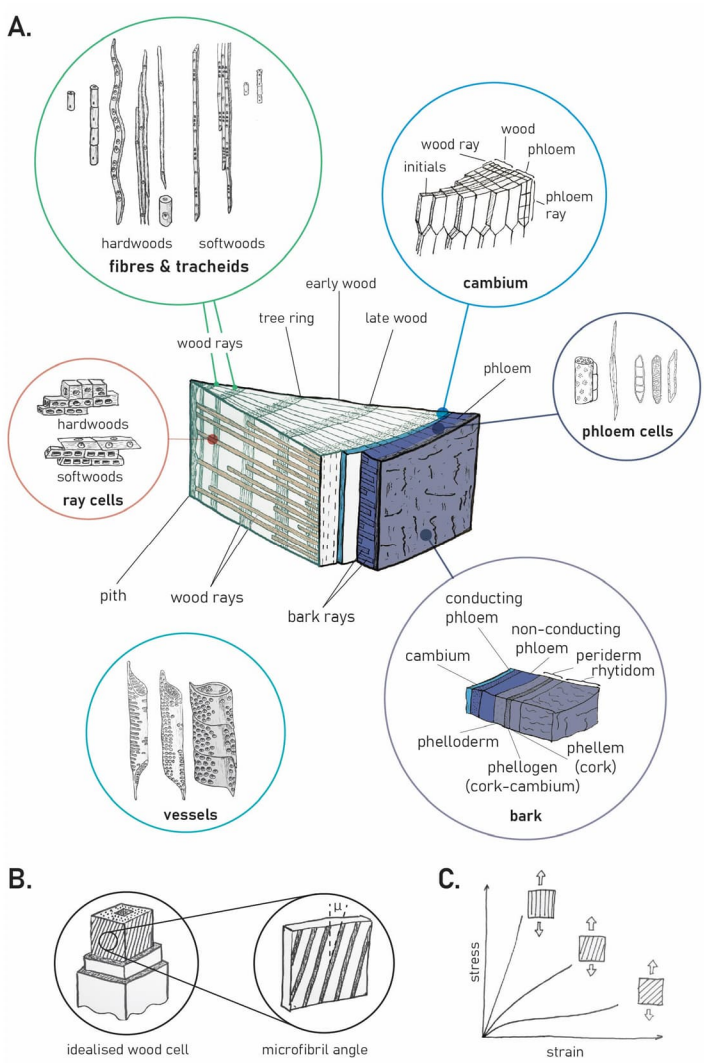

Figure 2.1: *Schematic of the stem of a tree showing different tissue and cell types. The 'piece of cake' shows a macroscopic segment (several centimeters) of a tree stem from the pith to the outer bark with its different tissue types. From the outside: bark (blue and grey), cambium (turquoise), wood (green) with ray cells (orange). Coloured circles show the cell types and geometries of these tissues at higher magnifications (cell diameters between 5 and 40 µm, lengths up to 3 mm). Grey circles on the bottom show secondary cell wall structure with parallel cellulose orientation (mi- crofibril diameters approx. 30 nm) and the diagram illustrates the effect of the cellulose orienta- tion on the mechanical properties. Drawings partly inspired by the figures in [(Evert 2006),(Nultsch 1991),(Britannica 2021).* [8]

[8] *This graphic is based on "Advanced materials design based on waste wood and bark", by Wenig et.al, 2021*

Wood cells are formed by the cambium at the periphery of the stem (Figure 2.1). The wood cells (Figure 2.1) are arranged in specific patterns (Figure 2.2) that provide functionality, such as water and nutrient transport, storage and mechanical stability. This requires not only a detailed physicochemical characterization but also needs to be understood in the context of the sessile plant's environmental conditions and the needs of the plant. This can be illustrated by the flexibility of a young tree, which bends rather than breaks upon wind or animal loading. Young trees consist to a large extent of wood with a low density and a large MFA (Lichtenegger et al. 1999). As the tree becomes larger, stiffness is needed to sufficiently support the crown. The absence of tissue remodelling processes in trees means that they can only adapt to changing conditions by growth—by adding new cells with various geometries and cell wall structures in regions of the tree where needed. To support the increasingly heavier crown, trees grow the newer layers of wood with denser cells, and with lower MFAs. Once a tissue is formed, the microstructural features such as density and MFAs are imprinted and stored within the material for the life of the tree and beyond. Hence, a simple, straight stem consists of wood with a wide variety of properties. The inner material remains less dense with the larger MFA and the outer material being denser with smaller MFAs. Woven within and between the fibers, vessels and tracheids are the ray cells (Figure 2.2), which can provide increased radial strength (Burgert and Eckstein 2001).

The dead material in the centre of the tree can still be modified by the tree long after the cells have formed, with numerous tree species forming heartwood. After the sapwood tracheids stop water transport and the parenchyma cells die, extractives are incorporated into the cell walls to increase natural durability (Taylor, Gartner, and Morrell 2002). This heartwood formation is a one-way process, leading to permanent changes in local properties. Towards the top of the tree are stem segments of juvenile wood without heartwood formation. Since trees are bound to a location, they also need to be able to react to changing loading conditions. Tilted stems, but also branches, form reaction wood. In the case of softwoods, compression wood is formed on the lower side to push the stem or branch by the generation of compressive stresses. Hardwoods form tension wood on the upper side and pull the organs in the desired direction (Barnett, Gril, and Saranpää 2014). The process of stress generation is based on structural differences at the cell wall, cell and tissue level (Burgert et al. 2007), (Goswami et al. 2008), resulting in altered chemical and mechanical properties compared to 'normal wood'. The ability of plants to move is not restricted to stems or branches. Seed structures especially are known to perform complex motions; however, due to their limited availability

and small size, these structures are of little interest as a plant-based raw material. These materials rather serve as bioinspiration, for example of self-actuating (façade) elements (Menges and Reichert 2015), (Vailati et al. 2018), (Guiducci et al. 2016) and provide a fertile research area for the development of bioinspired self-moving materials (Pandolfi and Izzo 2013).

Besides wood, bark is also available on a large scale and in huge quantities; however, muchless is known about the properties of bark than about wood. Recent publications partly address this missing information giving comprehensive information about bark anatomy of various species (Angyalossy et al. 2016), (Schweingruber, Steiger, and Börner 2019), macroscopic bark structure (Schweingruber, Steiger, and Börner 2019) and chemical composition (Bianchi 2016), (Vangell 2020). Bark is the boundary of a tree stem to its environment and the term bark contains all the tissues from the cambium towards the outside (e.g. (Evert 2006), Figure 2.1). As the cambium synthesizes xylem cells (wood) towards the inside, it synthesizes to a lesser extent secondary phloem cells (sieve cells and tubes and bark ray cells, Figure 2.1) towards the outside. The phloem consists of the inner conducting and the nonconducting phloem, which is located a bit further outside (Figure 2.1). Even though the nonconducting phloem loses its conductive capacity after being formed, storage functions and meristematic potential remain. The outermost layer of a young tree seedling is the epidermis, which is not able to grow sufficiently to serve as a permanently protecting layer. At some stage, the epidermis ruptures and is replaced by the periderm, a secondary protective covering. The periderm consists of phellogen or cork cambium, a meristematic tissue, which produces the watertight phellem or cork towards the outside and the periderm towards the inside. The periderm separates non-functional phloem from the living inner part of the stem towards the outside, which forms the rhytidome. The location of the phellogen(s) in the plant plays an important role for the bark structure.

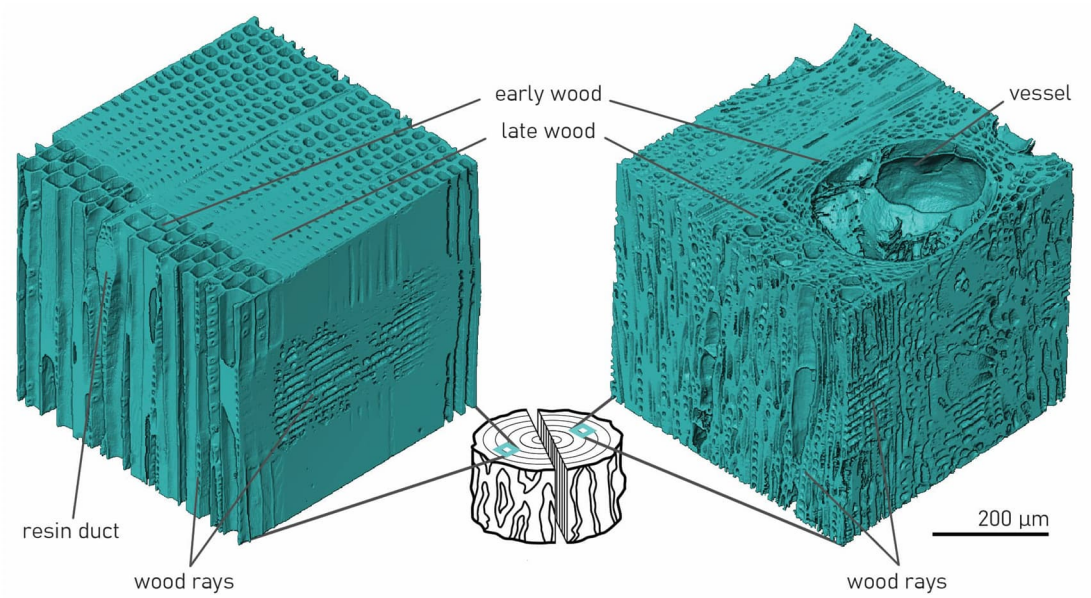

Figure 2.2: *µCT scans of spruce and oak wood. The CT scan of spruce wood (left) shows the distinct growth ring boarder with thin-walled early wood cells for efficient water transport and thick- walled latewood cells contributing more to the mechanical stability. The wood rays are narrow and ray cells are visible on the radial plane. Radial resin duct on the tangential plane. The µCT scan of oak shows the higher complexity of angiosperm wood with vessels, tracheids, libriform fibers, narrow and very broad rays.* [9]

The main functions of bark are water and nutrient transport and storage. The outer bark protects the tree from external challenges such as frost, heat, fire or microbes. Owing to its peripheral location on the stem and hence its large second moment of area, bark also contributes to the mechanical integrity of tree stems and twigs, despite its lower modulus compared to wood (Niklas 1999). Similar to other parts of the tree such as roots or leaves, bark is in permanent interaction with the environment and shaped by the environment and 'bark-inhabitants'. It provides 'housing' for numerous other organisms: fungi, lichens, mosses, plants but also animals such as insects and birds. It is, therefore, not surprising that its properties are highly variable—not only between different tree species but also within species and even within a single tree.

[9] *This graphic is based on "Advanced materials design based on waste wood and bark", by Wenig et.al, 2021*

2.1.2 Structural Differences between different Barks

Bark, in terms of its composition and structure, is at least as diverse as wood and differences between species may display different environmental challenges in different ecosystems. Trees in cold regions need to protect the living cells against intracellular ice formation. In very cold regions, this protection is mainly biochemical (Parker 1963), (Strimbeck et al. 2015) while a physically protective role of the bark is possible in regions with more random and shorter frost events, as seen in savannahs (De Antonio, Scalon, and Rossatto 2020). Trees growing in fire-prone regions often possess fire-resistant barks which protect the underlying meristematic tissues against high temperatures (Bauer et al. 2010), at the same time protection against mechanical damage may play an important role, as recently shown for giant sequoias growing in regions with regular rockfalls (Bold et al. 2020).

Changes in bark properties between tree species, within the same species and even within a tree can be highly diverse as, for example, in pine: *Pinus sylvestris*, possesses a thin so-called mirror bark when young and further up the stem and a thick, flaky bark at the bottom of older trees (Figure 2.3). ”[10]

Bark differs in many factors such as: its appearance, structure and material thickness. The proportion of bark depends on the type of bark, the age of the tree, the trunk diameter, the trunk height, as well as the site conditions and the sociological position of the tree. In general, pioneer tree species (Salix, Populus, Betula) and species growing in locations with more light exposure (such as Larix, Pinus, Quercus and Robinia) are more likely to form thick bark than species growing predominantely in the shade (Abies, Picea and Fagus) (Schweingruber, Steiger et al. 2019).

[10] *Wenig, C., Dunlop, J. W., Hehemeyer-Cürten, J., Reppe, F. J., Horbelt, N., Krauthausen, K., ... & Eder, M. (2021). Advanced materials design based on waste wood and bark. Philosophical Transactions of the Royal Society A, 379(2206), 20200345.*

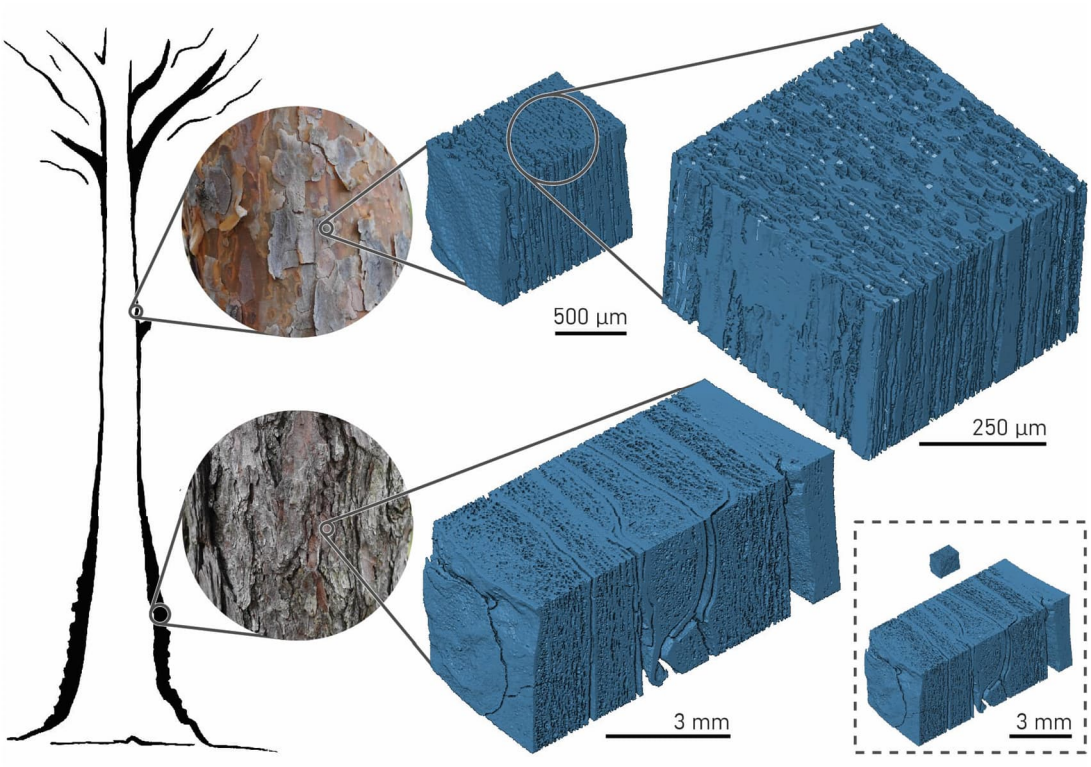

Figure 2.3: *Macro- and microstructure of pine bark. Photographs show mirror bark and flaky bark from the bottom of the trunk. µCT scans of mirror bark, at high magnification low-density regions with large pores show dilated parenchyma, high density regions are collapsed sieve tubes and periderm remnants, elongated crystal tubes appear bright, fibers are missing—a characteristic for pine bark (Schweingruber, Steiger, and Börner 2019). µCT scan of flaky bark show periderm layers (dense) as well as large volume fractions of rhytidome. Grey bark cubes display size relation between flaky old bark and mirror bark.*[11]

[11] *This graphic is based on "Advanced materials design based on waste wood and bark", by Wenig et.al, 2021*

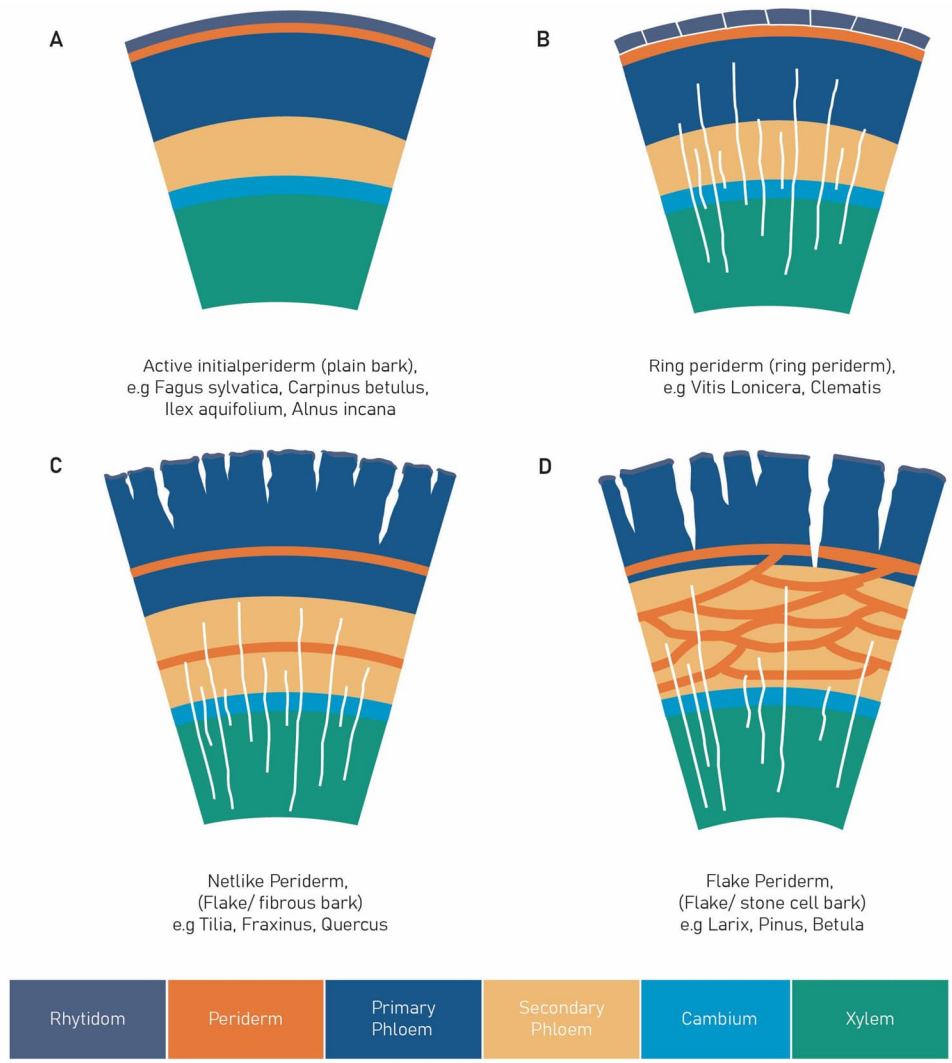

Figure 2.4: *Schematic drawing of a radial cut. Shows the structural composition of the different types of bark, (a) tree bark with active intial periderm in which the rhytidom layer remains mostly smooth and undamaged, (b) ring periderm in which the thickness growth causes the rhytidom to detach circumferentially or in longitudinal strips from the trunk, (c) netlike periderm in which the thickness growth causes deep cracks into the primary phloem. The phloem layer, which is deeply fissured along the stem, is solid, (d) flake periderm forms irregular follicular periderm rings. The individual periderms are arranged in a flake-like manner, which detach from the trunk in flakes over time. Graphic is based on structural drawings of Vaucher H. Baumrinden. F. Enke; 1990 (Vaucher 1990).*

In some species, the initial periderm remains for the entire life of the tree (orange line in Figure 2.4.a). This permanent initial periderm/surface periderm usually does not result in fissured bark structure (like Figure 2.4 b,c,d). Trees with this bark type (e.g. *Fagus sylvatica, Carpinus betulus, Ilex aquifolium, Alnus incana*) are only surrounded by a thin layer of bark (Kupferschmid 2001).

In species with deep/successive periderms (Figure 2.4 b,c,d), there are two characteristic patterns that determine the structure of the bark (due to their different successive periderms). Periderm, which runs cylindrically and parallel to the bark surface, is called ring periderm (e.g. *Thuja, Juniperus, Clematis, Vitis* and *Lonicera).* First, ring periderm is made out of closed layers. By cell growth and resulting thickness growth periderm layers open up circumferential (Figure 2.4 b).

The majority of local tree species[12] form (concave) flake periderms resulting in a so-called flake bark (Figure 2.4 d). In this case, periderms overlap each other (orange line Fig. 2.4 d). Secondary phloem is the conducting tissue of bark and produced by the cambium towards the outside. It causes the thickness growth of the whole bark structure. The composition of the secondary phloem also determines the appearance of the bark surface. This is largely determined by the fiber content and the amount of sclereids in the particular bark. If the fiber content in the bark is high, the surface will crack in a net-like pattern as the bark continues to grow (Figure 2.4 c), forming flake fiber bark (e.g. *Tilia, Fraxinus, Quercus*). Flake barks with a high sclereid content form sheet-like flake barks/flake-stone cell barks from e.g *Larix, Pinus*).

All barks studied in this thesis (Table 1) belong to species with deep/successive periderms where thickness growth can cause deep cracks into the rhytidom and the primary phloem (Figure 2.4 b,c,d). Barks with a net or flake periderm can reach high thicknesses (Vaucher 1990).

[12] *Barks used in this work were harvested around Potsdam, Germany.*

Species	Structure	Thickness (mm)	Volume %	Appearance
Pinus sylvestris	Flake Bark	Up to 50	5–18	Young stem: thin, flaky, orange brown Older stem: thick, scaly, dark gray brown
Betula Pendula	Flake/Stone-Cell Bark	5–50	7 -13	Young stem: white, smooth, peeling off in papery flakes Older stem: less white, more black bark spots
Quercus Robur	Flake /Fi-brous Bark	6–40	11–20	Young: smooth, dark brown Old stem: (deeply) fissured
Larix decidua	Flake Bark	Up to 50	16–24	Young: smooth, gray, brown Old stem: dark blue green, scaly with long furrows

Table 2.1: Characteristics of selected bark species (Hohlheide 1951), (Kupferschmid 2001), (Schweingruber, Steiger, and Börner 2019)

2.1.3 Pine *(Pinus sylvestris)*

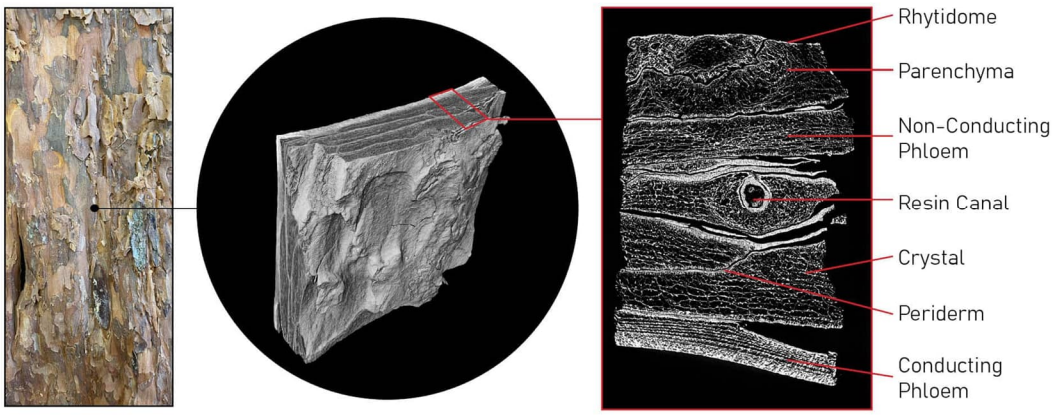

Figure 2.5: *Structure of "Pinus sylvestris". The left image represents the appearance of pine bark on a tree stem. The center image shows a 3D µCT scan of a pine bark sample. The right image is a 2D µCT scan in tangential-radial direction. The different structural parts of pine bark are marked in the right image.*

The habitat of *Pinus sylvestris* extends from Eurosiberia, northeast Spain to Scandinavia and the Siberian to the Pacific coast. This makes it one of the trees with the largest distribution area. It grows on rocky, sandy or swampy soils. This prevents it from competing with other species on deeper soils. It can be found at altitudes up to 2600 m.

The young phloem consists of regular tangential layers. The absence of fibers and isolated sclereids is characteristic for *Pinus sylvestris*. Rectangular sieve cells are in 3 to 5 seriate tangential rows and collapse within a few years. While in juvenile phloem parenchyma cells are small and in oval shape, in adult phloem they are enlarged. Rays crossing the parenchyma are radially corrugated. The phellem is irregularly layered and consists of rectangular thick-walled sclereids. In the early stages (near the cambium), annual rings can also be seen in the bark of *Pinus sylvestris*. As the phloem progresses, these are only indistinctly visible, due to the collapse of the sieve cells and the expansion of the parenchyma cells (Schweingruber, Steiger, and Börner 2019).

2.1.4 Larch *(Larix decidua)*

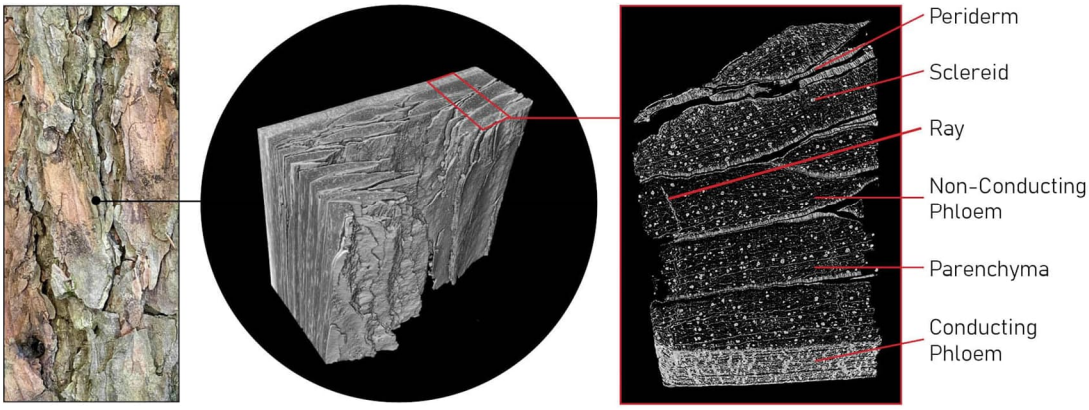

Figure 2.6: *Structure of "Larix decidua". The left image represents the appearance of larch bark on a tree stem. The center image shows a 3D µCT scan of a larch bark sample. The right image is a 2D µCT scan in tangential-radial direction. The different structural parts of larch bark are marked in the right image.*

Larix decidua is native in the Alpine regions and in some areas of the Carpathians (east Maritime Alps east, lower Austria and Croatia, in the hills of southern Poland). They grow in areas between 150 m up to 2900 m altitude (Pâques 2013). For economic reasons, the European

larch has been cultivated for centuries outside its natural habitat, even in the Central European low mountain ranges.

The bark structure of the young phloem is arranged in tangential layers and located as annual rings for several years near the cambium. The periderm remains active for several years. With the formation of the adult phloem annual rings become more and more indistinct, due to the collapse of the sieve cells. These are previously arranged in 3-5 fold tangential rows before they collapse within a few years.

Isolated round and thick-walled sclereids are characteristic for larch bark. Outside the sclereids small prismatic crystals can be found. Parenchyma cells, which are small and oval in the juvenile phloem, enlarge in the adult phloem. There are no fibers in larch bark. The phloem is permeated by radially wavy rays. The phellem layer is formed by rectangular and thin-walled cells (Schweingruber, Steiger, and Börner 2019).

2.1.5 Oak *(Quercus robur)*

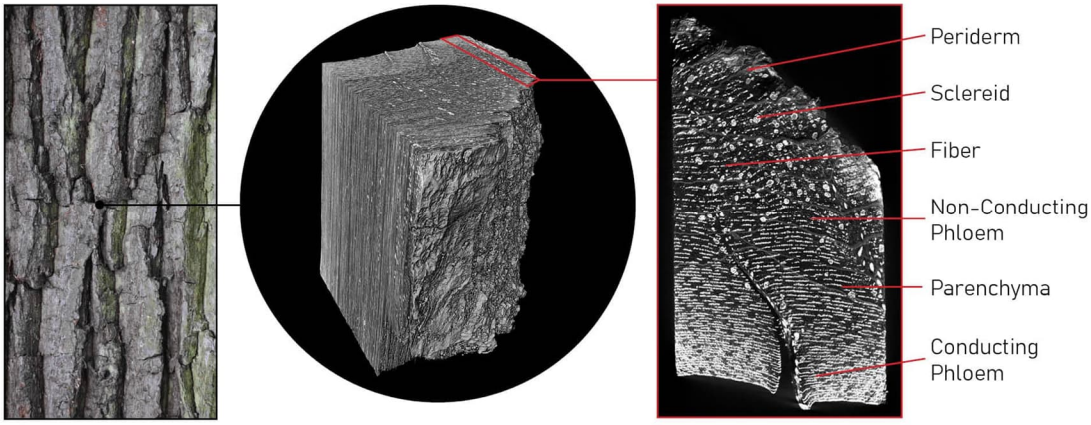

Figure 2.7: *Structure of "Quercus robur". The left image represents the appearance of oak bark on a tree stem. The center image shows a 3D µCT scan of an oak bark sample. The right image is a 2D µCT scan in tangential-radial direction. The different structural parts of oak bark are marked in the right image.*

Quercus robur is widespread throughout temperate Europe (northeast of Portugal, southern Sweden, around Lake Ladoga and the Volga, the Caucasus, south to northern Turkey, the Balkans, and Italy). Oak prefers moist to wet, deep and loamy soils and can be found up to 1200 m altitude.

The bark structure of *Quercus robur* is tangentially layered. The parenchyma cells are round and arranged radially. The sieve tubes located in the parenchyma are squared and collapse shortly after their formation. Fiber bands alternate with zones of sieve tubes and parenchyma cells. In the phloem there are round sclereids groups. Rays are uniseriate straight or wavy and have the possibility to expand/dilate. The phellem consists of rectangular, thin-walled cork cells built up in layers (Schweingruber, Steiger, and Börner 2019).

2.1.6 Birch *(Betula pendula)*

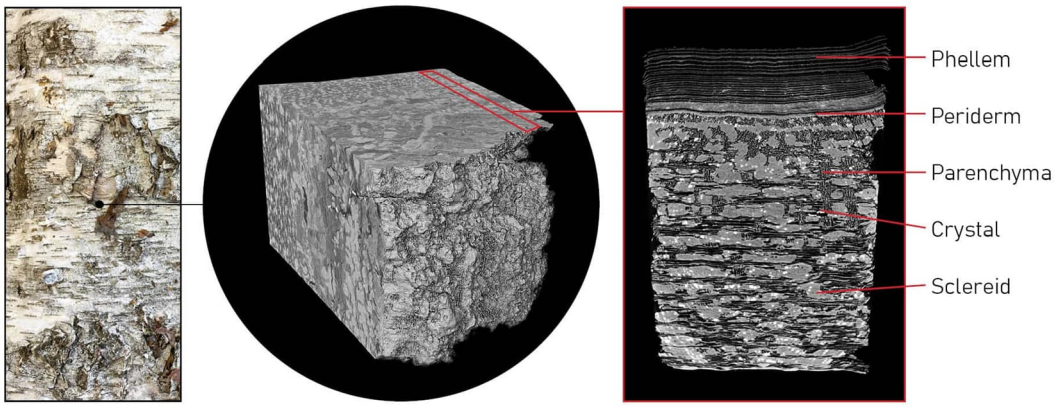

Figure 2.8: *Structure of "Betula pendula". The left image represents the appearance of birch bark on a tree stem. The center image shows a 3D µCT scan of a birch bark sample. The right image is a 2D µCT scan in tangential-radial direction. The different structural parts of birch bark are marked in the right image.*

Betula pendula is found in temperate forests (Eurosiberia, northeast of Catalonia, whole of Scandinavia, the northern Yenisei River, Altai Mountains, south to northern Iran, Macedonia and Sicily). As a pioneer tree, it has minimal demands on the growing location. Preferably it grows on a moist to dry, moderately nutritious and stony, turfy to sandy soil at an altitude of up to 1800m.

Parenchyma cells are round shaped in birch. Crystals and fibers are completely absent. The groups of sclereids, which are very present, are often slightly tangentially layered. Sieve tubes are irregularly shaped. Shortly after their formation they collapse. Ray cell dilatation happens without cell divisions. The periderm of birch consists of rectangular cells with thin to thick walls (Schweingruber, Steiger, and Börner 2019).

2.2 Processing and applications of bark

2.2.1 Industrial Debarking

With industrialization and globalization, wood processing was centralized in many parts of the world. Since the 20th century, large quantities of wood were felled in in many areas worldwide (e.g. Scandinavia, Russia and North America) and transported to distant sawmills for further processing (Harkin 1971). During transport, bark serves as a protective layer for wood against contaminants (sand, dirt), insects, fungi, desiccation and damage during handling. The debarking process is mostly centralized in the sawmill shortly before the wood is further processed. The large throughput in saw mills and other wood processing industries led to the accumulation of huge amounts of bark within short time periods (Harkin 1971).

Some of the most established debarking-techniques are the rosser-head debarker, hammer-type debarker, ring debarker, rotor debarker, drum debarker, water jet and high frequency debarker ((Baroth 2005),(Ressel 2006), (Hargitai 2003)). The bark is chipped to small pieces with all mentioned techniques. The choice of debarking technique is influenced by several factors: tree species, dimensions of logs, internal bark structure, seasonality-related moisture contents ((Einspahr, Van Eperen, and Fiscus 1984), (Chow and Obermajer 2004), (Baroth 2005)) and bark shape.

Figure 2.9: *Debarked logs. Rotor debarking is the most common method of debarking in sawmill and veneer industry. Logs are driven through the machine separately and rotating blades scrape the bark along the cambium layer. The image shows debarked logs with characteristic debarking lines.*

After removal from tree logs, bark is stored outside in piles (Figure 2.9). In some cases, bark is further processed in hammermills, choppers , shredders , grinders for pulverizing or shredding (Harkin 1971). Several applications with bark as bulk material or supplement for the panel industry or chemical extraction is possible and will be described in the next subchapters.

2.2.2 Chemical Extraction of Bark

Characterizing bark as a material in general is challenging. Between tree species it differs with regard to its topography, appearance and chemical composition (Srivastava 1964). General mechanical properties of bark are influenced by its structure but also by its chemical composition.

Compared to wood the mechanical properties of tree bark are lower (Fengel and Wegener 2011). Reasons based on the chemical composition of bark could be the different distribution of the main chemical components between bark and wood (Figure 2.10). While bark has a higher content of polyphenols/extractives and suberin (blue section, Figure 2.10) (Fengel and Wegener 2011), its proportion of polysaccharides is considerably lower (orange section, Figure 2.10).

Furthermore, differences within the bark are common, eg between phloem and rhytidome. As Fengel and Wegener (1983) stated, the amount of extractives and polysaccharides generally declines while the content of lignin and polyphenolic substances increases from the inner towards the outer bark (Fengel and Wegener 2011).

Additionally, defining the exact amount of all components in bark is extremely difficult. Results can differ drastically depending on both the extraction procedures applied, and the extraction used (chemicals/solvents). This is the case for components like lignin and polysaccharides (Fengel and Wegener 2011). The influence of the extraction technique was proven for the bark of loblolly pine (*Pinus taeda*): McGinnis and Parikh (1975) determined an extractive content of 19.9 %, while the technique used by Labosky (1979) results in 27.5 % for the same bark species (Fengel and Wegener 2011).

As stated above, the determined chemical composition depends on many factors. However, the following chapter gives an overview of the main components of the bark species studied in this thesis and compares them to the reference material wood.

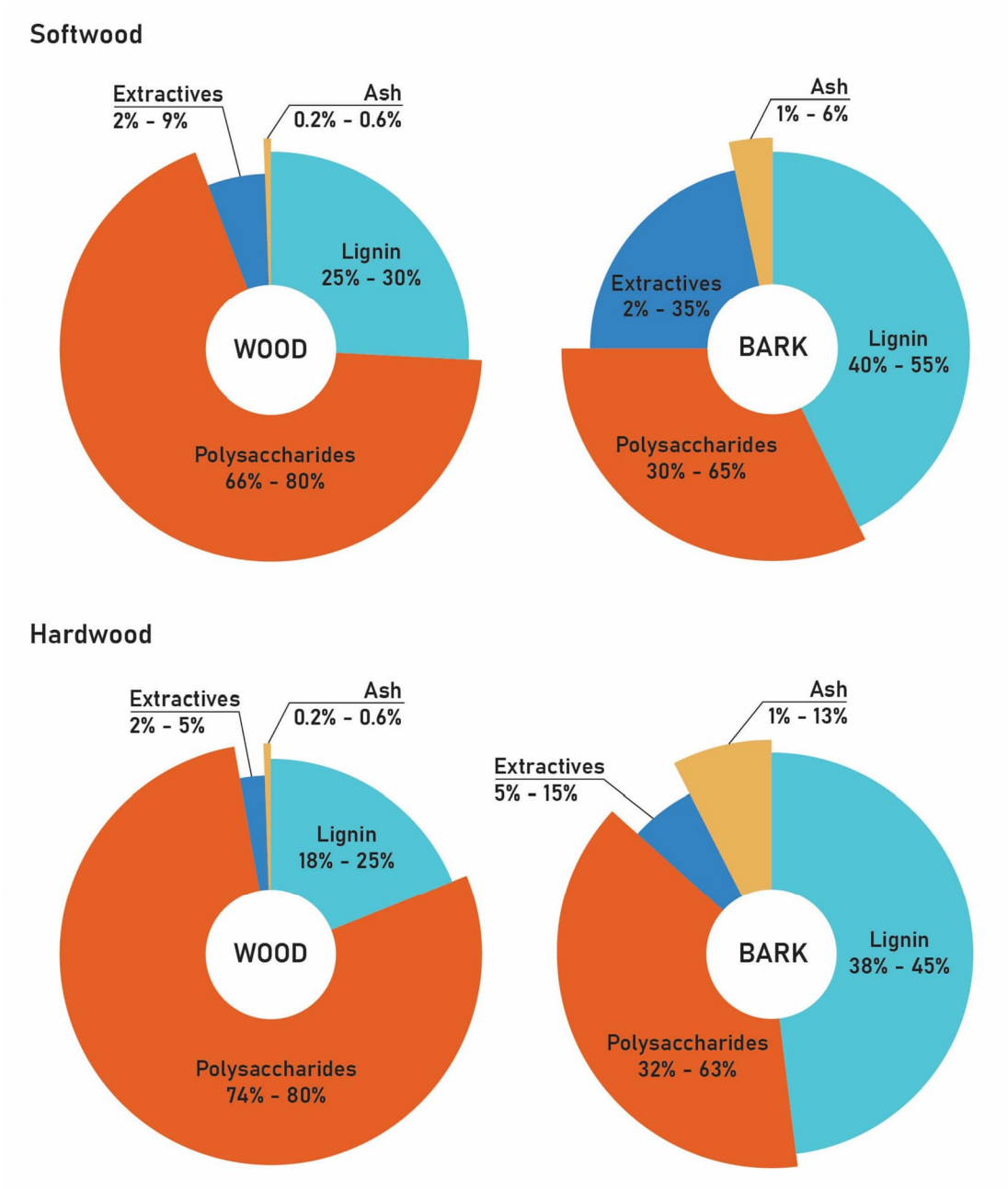

Figure 2.10: *Chemical composition of wood and bark. Pie charts show the distribution of main chemical compositions of hardwood and softwood and tree bark of hardwood and softwood. All percentages are approximated (Harkin 1971), (Fengel and Wegener 2011), (Kupferschmid 2001), (Feng et al. 2013a)*

Polysaccharides

Polysaccharides are the major component in all barks and play an important role for the material properties. This large fraction consists of cellulose, hemicellulose and pectins (Rowell et al. 2005). Cellulose is a linear polymer that is made from single glucose monomers. Together with lignin and suberin, polysaccharides create the structural cell wall constituents in bark.

With ~ 66 – 80 wt %, polysaccharides are also the main component of wood (Fengel and Wegener 2011; Van de Vyver et al. 2011). However, their proportion in bark is estimated at around ~30-65 wt% (Fengel and Wegener 2011), (Harkin 1971) and is therefore substantially lower. The amount of cellulose also differs between bark species. For example, pine shows a cellulose percentage of 20.2 % while in oak 32.6 % are detected (Dietrichs et al. 1978). The typical value for wood ranges from 40 to 60 wt % (Schutyser et al. 2018). Bark and wood cellulose have the same crystal lattice, but the degree of crystallinity is lower in bark (Fengel and Wegener 2011). Also the degree of polymerization is lower in bark cellulose (Fengel and Wegener 2011).

As Fengel and Wegner (1983) concluded after listing a selection of polyose, bark and wood have the same chemical structure, but show differences in their composition. Different kinds and quantities of polyose also exist in different bark species, e.g. those of softwood and hardwood. While beech (*Fagus sylvatica)* contains up to 23.1 % polyoses, only 9.3 % were measured for oak (*Quercus robur*).

Lignin

In contrast to wood there is little known about lignin in bark. Overall, standard measurements for lignin contents (i.e. Klason lignin) have shown a higher lignin content in bark than in wood (Rowell et al. 2005). Again, the quantity of lignin differs between bark species (Rowell et al. 2005). Analysis (Srivastava 1964) has shown that lignin can be localized in cell walls of bark fibers, in rhytidome and periderms cells, as well as in sclereids (Fengel and Wegener 2011). Fengel and Wegner summarized (1983) several studies on lignin structure and concluded that there is no difference between wood and bark lignin structures. The difference is the amount found in the respective system (Fengel and Wegener 2011).

Polyphenols

Natural polyphenols occur in plants as secondary plant substances and are a structurally broad group of chemical components. In particular, softwood bark is rich in phenolic compounds, and specifically in (condensed) tannins (Fengel and Wegener 2011).

Tannins as an example of polyphenols

Tannins are an extensively researched group of polyphenols. They are almost ubiquitous in all plant tissues (Haslam 1989). Furthermore, tannins are the most abundant renewable source of phenolic molecules after lignin (Porter and Hemingway 1989). Tannins can be separated into two main classes of chemical compounds: hydrolysable tannins and condensed tannins. Both are able to form complexes with proteins and polysaccharides but have different functions in the plant metabolisms (Khanbabaee and Van Ree 2001).

The predominant biological role of tannins is the protection against herbivores, microorganisms (Barbehenn and Constabel 2011) and parasites. Like other polyphenols, they also regulate the slower decay of bark compared to wood in litter (Ganjegunte et al. 2004). Both functions are based on the complex and almost irreversible bonds that tannins form with cellulose, pectin and proteins (Haslam 1989; Porter and Hemingway 1989).

In addition, their ability to form complex bonds has a positive impact not only on the bark, but also on the entire ecosystem that surrounds it. Tannin-protein complexes degrade slowly. With that they regulate the nitrogen release in soil (Bianchi 2016).

The production of tannins and thus their concentration and molecular structure are based on seasonal variations (the annual growing cycle) and tissue aging. Porter (1989) measured a general minimum in early spring and a maximum in summer/early autumn of the whole phenolic extraction yield (Porter and Hemingway 1989).

The exemplary use of the polyphenol group - tannins

Tannins have been utilized in many different forms by humans for centuries. The most well-known example is their traditional use in the leather industry as tanning agents (Pizzi 2008), but the interest in plant extracts rich in tannins goes beyond. They are also used in traditional

silk dyeing and contribute to tannin-containing foods and beverages, having a typical astringent taste (Haslam 1989). Because of their significantly higher tanning effect and a far more efficient production, today's large-scale leather industry predominantly uses synthetic tanning agents. However, there is an ambition to use natural tanning agents again, and, for ecological reasons, preferably those from industrial waste. This effort is currently a subject of research.

The bark of coniferous trees, for example, is an important by-product of the wood and paper industry. Sauro Bianchi (Bianchi 2016) investigated it in his thesis in order to define possible tannin source potentials.

In addition to the production of leather, tannins also show interesting properties for industrial wood-based products. Most commonly wood boards are bonded by standard urea-formaldehyde adhesives (Bekhta et al. 2021) with the disadvantage of emitting formaldehyde, a substance classified as carcinogenic (Gupta, Ulsamer, and Preuss 1982), (Zhou et al. 2014). It has been shown that formaldehyde emissions can be lowered by using tannin-based formulations (Pizzi, Valenezuela, and Westermeyer 1994), (Gönültaş 2018).

The suitability of tannin-rich bark extracts as natural substitutes in thermosetting resins is not limited to wood adhesives. Foamed material based on mixtures of condensed tannins and furfuryl alcohol have been widely investigated and characterized (Tondi et al. 2009). These products showed noteworthy insulation and fire-resistance properties comparable to synthetic phenolic foams. Tannin resins were used as a matrix in composite materials containing sisal, flax or jute fibers (Sauget, Nicollin, and Pizzi 2013; Barbosa Jr et al. 2010).

Due to their heavy metal chelation ability, softwood bark extracts can also be used as flocculants in wastewater treatments (Yamaguchi et al. 1992; Sánchez-Martín, Beltrán-Heredia, and Gibello-Pérez 2011).

Pharmacological effects of condensed tannins such as antibacterial, antiviral, enzyme inhibitor, antimutagenic and antitumoral, next to some specific interactions with vascular and cardiac systems, were also reported (De Bruyne et al. 1999). These characteristics of phenolic monomers and oligomers are mostly documented for soft wood bark.

Suberin

Suberin is an aliphatic polyester present, which is located in the cell walls of phellem or cork cells in the outer bark (Graça 2015). This insoluble constituent contributes to insulation, protection and reducing water loss (Graça 2015).

To analyze suberin separately from cork cells, a saponification with an alkali treatment must be undertaken first. The individual components can then be extracted and analyzed by chromatographic methods (Fengel and Wegener 2011).

The suberin content varies greatly between barks of different tree species. Suberin content in bark species range from almost no suberin to 40-45 wt% for cork of cork oak (*Quercus suber*) bark (Pereira 2013). But also cork cells of other bark species produce a significant amount of cork, and, thus, suberin. Even the Swedish birch bark (*Betula verrucosa*) contains 43.3% of suberin in its cork cells (Fengel and Wegener 2011). While both barks contain approximately the same amount of suberin, the composition is different. Other notable examples are birch (*Betula pendula*), douglas fir (*Pseudotsuga menziesii*) and Chinese cork oak (*Quercus variabilis*), all having a suberin content of more than 30 wt% (Leite and Pereira 2017).

Being cultivated in the Mediterranean climate, cork oak is famous for its mechanical and chemical properties, its high cork content and its ability to regrow. It is mostly known for the production of wine stoppers but also other products like furniture and textiles can be made out of cork (Pereira 2011).

Minerals

Mineral constituents in bark are analyzed as ash content in most methods after the complete incineration of bark. In general, the ash content and the number of elements of barks is much higher compared to that of wood (Kurth 1947; Fengel and Wegener 2011).

Ash contents of more than 10 wt% have been reported for some hardwood barks, whereas for wood, these values are typically below 1 wt% (Rowell et al. 2005; Fengel and Wegener 2011). The amount of mineral components again varies between different bark species. This is e.g. shown by a study on 14 different barks of Canadian species with bark of softwood and hardwoods. The analyzed ash contents in soft wood was found in the range of 2.0-4.2 wt %, while hardwood bark remained 1.8-8.1 wt % of ash (Bryers 1996).

The main element found in the mineral fraction is calcium with 82-95%, followed by potassium and magnesium (Bryers 1996). Also other elements, mostly metal oxides, can be found (K_2O, MgO, Na_2O, CaO, and Fe_2O_3, etc.) but in most cases with less than 1% (Fengel and Wegener 2011).

All minerals are located in the cell walls with a certain concentration in the rays and resin canals. A reason for the high calcium content is the presence of calcium oxalate crystals which are located in sieve cells and longitudinal parenchyma in the phloem and phelloderm (Evert 2006; Hudgins, Krekling, and Franceschi 2003).

However, these concentrations are not evenly distributed and showed large differences especially between the individual cell structures of bark and wood. One bark, which was further investigated in this respect, is the bark of pine (*pinus sp.*) by Fossum et al (1972) (Fossum, Hartler, and Libert 1972). They have shown that the contribution of Magnesium (Mg) and Manganese (Mn) is changing within the bark structure. While the Ca-content from wood to bark is increasing, a decrease of Mg and Mn-content is reported.

The applications for bark ash are very limited. In general, bark is mostly burned moist and contains less volatile matter and a higher percentage of fixed carbon than wood (Feng et al. 2013b). Both tend to lower the heating values. In order to benefit from this energy consuming process, research is being conducted on e.g. bark ash as an additive for concrete production (Rukzon and Chindaprasirt 2014). Furthermore, mineral constituents are being investigated for their use as supplementary materials in horticulture (Pérez-Cruzado et al. 2011). Overall, the fields of application for bark ash are still small.

2.2.3 Bark as Bulk Material

The efforts undertaken to study bark for use as a bulk material are limited (Harkin 1971). Currently, bark mostly remains as an industrial by- or waste-product.

The devaluation of tree bark is already evident in the middle of the 20th century. Increasing progress in wood harvesting and processing created an enormous and problematic amount of bark. At that time, bark was mostly delivered to landfills (Feng et al. 2013b).

According to Feng et al. (Feng et al. 2013b) in Canada more than 17 million m^3 of bark is produced yearly and more than half of it is burned or still delivered to dumps. The strategy to burn bark is nowadays widely established (Barbu 2011). Bark waste, which is mostly burnt

in wet conditions directly on site of e.g. sawmills for heat and/or power production, significantly reduces the efficiency of energetic utilization, as most of the energy is needed to evaporate the moisture content. Also, as combustion properties of bark, such as ash content or heating value are less favorable compared to wood (Nosek, Holubcik, and Jandacka 2016), the use of bark for energy production is of only medium interest.

Alternatively, Melin (Melin 2008) produced bark pellets, motivated by the lower price and assuming advantageous physical and mechanical qualities : bark (extractives) melts easily under high pressure and therefore could form a strong bond among particles (Melin 2008). Nevertheless the disadvantage of high ash content in the bark (chlorides, sulfates) needs to be considered (Lehtikangas 2001).

Next to energy production, bark is mainly used as mulching materials for agriculture and gardening. Most common barks are shredded spruce, scotch pine and oak. The high moisture content of bark is a great benefit for mulching. It reduces evaporation on the soil surface and helps the reproduction of microorganisms. Also the decay of bark is slower than wood (Pasztory et al. 2016).

2.2.4 Bark for Panel Industry

Wood-based materials with bark content have been developed and improved since the middle of the 20th century (Pasztory et al. 2016). In the 1970s the mechanical influence of bark particles in particle boards was investigated.

In the first attempts the bending strength of the particle boards decreased due to a higher bark ratio. Therefore these particle boards could not meet the standard bending strength (requirements) (Kain et al. 2012). Further research showed that to some extent, bark particles can replace wood particles to produce particle boards with acceptable (mechanical) properties (Pedieu, Riedl, and Pichette 2008). In 1984, Muszynski and McNatt made wood panels with a maximum bark content of 30% (*Picea abies* and *Pinus sylvestris*), which were suitable for the production of furniture (Muszynski and McNatt 1984). In addition bark particles were used in medium-density fiberboard (MDF), hard-density fiberboard (HDF) and insulating panels. The development and production of adhesive-free bark particleboards as well as pure bark panels were topics in recent studies (Pasztory et al. 2016).

As described in chapter 2.2.2, chemical composition and structure of bark is different compared to wood. Mechanical properties, such as strength are lower when bark is used in particle boards (up to 100% bark are possible). This can be explained by lesser fiber compared to wood (Harkin 1971). Contaminants on the bark surface (sand and soil) also challenge the production of particle boards.

Investigation for the production of medium-density fiberboard (MDF) with bark showed promising results with refined bark fibers (of four tree species hybrid poplar, jack pine, larch and spruce). These MDF boards had bigger volume density compared to the control wood group (Xing et al. 2006). In addition, hygroscopic properties of the composite boards were studied (Yemele et al. 2008). Bark particleboards showed increased swelling and shrinkage (Gößwald et al. 2021), which affects mechanical strength (Yemele et al. 2008).

Another approach is not only to understand the influence of bark on wood particle boards but also to use species-specific properties, such as water resistance. Pedieu et al (Pedieu, Riedl, and Pichette 2008) investigated how water-resistant outer birch bark particles (*Betula papyrifera*) can improve the dimensional stability of mixed particleboards when used in the surfaces layers.

Efforts were also made to produce pure bark particle boards (Yemele et al. 2008) and fiberboards. To increase the directional properties, Kain et al. (Kain et al. 2018) pressed bark particles with sizes between 10 – 30 mm using urea formaldehyde resins. The resulting hot-pressed insulation panels showed densities ranging from 200 kg/m^3 to 450 kg/m^3 depending on the particle orientation. However, the use of such panels should be reduced as formaldehyde is classified as carcinogenic (Union 2012). The extract of some barks are rich in phenol compounds and could replace phenol-formaldehyde (PF) glues (Gößwald et al. 2021; Kain et al. 2012). While most bark panels were produced by adding resin, Burrows (Burrows 1960) showed already in 1960 that it is possible to produce resin-free bark particle boards by activating the natural gluing capability of the material through hot-pressing. Burrows assumed that the plasticization mechanism may occur due to the lignocellulosic character of bark and due to the presence of water as a plasticizer (Gößwald et al. 2021).

Considering barks' low heat conduction compared to wood (Niemz and Sonderegger 2016) the use in insulation boards might be an interesting application. The thermal conductivity of bark-based panels was evaluated in various studies. For example, in insulation boards bonded with natural tannins (Kain, Tudor, and Barbu 2020; Kain et al. 2018) or with larch, pine, spruce, fir, and oak tree bark is impregnated with urea-formaldehyde, melamine urea-, and

tannin-based adhesives (Gößwald et al. 2021). Special focus is given to the possibility of using formaldehyde-free tannin resin (Barbu et al. 2020) and the ambition to achieve density values in relation to the targeted mechanical and physical properties (Kain et al. 2014).

Recently, the formaldehyde-binding functions of bark boards (Barbu et al. 2020) and adhesive-free low-density insulation panels produced with spruce bark fibers were investigated (Gößwald et al. 2021).

Insulation boards from particles of larch bark (*Larix decidua Mill.*) bonded with a formaldehyde-free tannin resin interestingly shows that the resin amount did not influence the mechanical board properties as strongly as expected. Increasing proportions of bark particles are another influence on insulation performance (Kain, Tudor, and Barbu 2020). Due to its low density (Miles 2009) and relatively high proportion of cork cells (Holdheide and Huber 1952; Pereira 2015) larch bark insulation panels are mechanically stable compared with common insulation materials (Kain et al. 2012). Additionally larch extractives could be seen as a self-defense mechanism against microorganisms (Doi and Kurimoto 1998).

Also investigation on the production of insulation panels, made out of pine (*Pinus sylvestris L.*) bark have shown the possibility to press comparatively light panels (\500 kg/m3) bonded with UF resin (competitive with existing boards in terms of thermal conductivity (0.06–0.09 W/(m*K)) (Kain et al. 2012).

But not only the bark species and the particle size correlate with the board density, thus influencing the thermal conductivity (Kain et al. 2016). Further research shows that particle orientation, bark board porosity and pore size are important factors for the optimization production of insulation panels (Kain et al. 2018). In their research on larch bark insulation boards, Kain et. al (Kain et al. 2016) showed that the thermal conductivity of bark boards with particles aligned horizontally to the panel plane is on average 13% lower than with orthogonally oriented particles.

Apart from thermal insulation, bark-based composites can be used as sound-absorbing panels. (Tudor et al. 2020; Li et al. 2020) or for fire protection

However, the contemporary use of bark as described above relies mostly on bark pieces and thus on crushed material. Moreover, in some cases the manufacturing of panels involves heavy processing. As a consequence, the inherent structural properties of natural bark are destroyed and no advantage can be taken out of the potential of being a natural material, available in large scales and quantities.

2.3 Traditional Uses of Bark

While wood is valuable enough to be stored until dry, bark—with the exceptions of the cork of oak trees and some barks with valuable substances for extraction—is often seen as less beneficial. Therefore it is frequently burnt in the wet condition to free storage capacities at the saw mills. Because resources are limited, the values of bark-which is a by-product of large volumes in the wood industry-should be addressed. For instance bark was used for a big variety of purposes in different cultures throughout history. The finding of an approximately 8000 year old bark cloth beater in southern China is one of the earliest pieces of evidence for bark cloth making (Li et al. 2014). Early bark cloth is non-woven and made by beating the plant fibers after retrieving the inner bark. With human migration, the knowledge as well as the plants (e.g. paper mulberry) expanded to various regions (Chang et al. 2015) (Cameron 2008). Bark cloth is still made in islands of the Oceania as 'tapa' as well as in Central America and Uganda (Rwawiire, Luggya, and Tomkova 2013). However, the inner bark material was also spun into cord or yarn and used for tying and knotting. This bast-like material could be knitted, woven and braided into two- and three-dimensional objects—bags knitted from bark yarn have been found in Australia, for example, and mats woven and braided from birch bark (from useful underlays to coverlets diligently worked with quills with feathers), boxes (from salt vessels to bags) and useful tools such as the pot scourer (a ball made from rolled-up strips of bark) but also bark scrolls with drawings on it have been found in the Finnish/Russian region of Karelia.[13] The objects from Karelia show that the resistance and durability of birch bark material in combination with its pliability made it possible to weave and braid the bark after it was cut into narrow strips. The advantage of the inner bark material consists in the fact that after processing (watering and beating), it behaves as a kind of *solides souples* (Engl.: *soft solids*) and in some aspects as an analogue to paper (Leroi-Gourhan 1971). This is confirmed by the use of birch bark for writings such as in the Gandh¯aran Buddhist manuscripts that date from approximately the first century CE and are believed to have been created in

[13] *All examples are taken from the collection of the British Museum. See museum numbers Oc.4061 (netted bag made of bark fiber cord, made by aboriginal Australian, nineteenth century), 2016,8030.1 (bag made of plaited strips of birch bark from Karelia in Russia/former Finland, 1900–1940), Am1928,-.92 (mat made of birch bark, with quill works and feathers, from North America, bought 1928), 2011,8036.35 (pot scourer in the form of a ball, made of curled birch bark strips, from Finland, 2011), Am1949,22.170 (scroll made of birch bark, with zoomorphic drawings, made by the Ojibwa, Minnesota, bought 1949), Oc1894,0814.4 (Oblong sheet of barkcloth with geometrical designs from Taveuni in Fiji/Oceania, nineteenth century). www.britishmuseum.org/collection (last accessed 6 March 2021).*

Afghanistan.[14] However, the outer bark was known as a useful material and even as a construction material: for hundreds of years, North American Indians used birch bark for their canoes (Adney and Chapelle 1964), and in Austria an old tradition is remembered where the loggers used bark as construction material for temporary huts in the forest (e.g. bark huts in Hof close to Salzburg or in the Salzburger Freilichtmuseum) (Ast 2011). […][15]

Until the 1920s, huts were used as dwellings, for example, in the saline area around Vienna. How these huts were designed and how they were built is only poorly documented. The discovered scriptural sources as well as pictures were used to reconstruct the basic manufacturing process of the huts (Figure 2.11). Depending on the area, these are mostly spruce or also fir bark. While in Austria the huts were essential for the forest workers, bark huts can also be found throughout history as garden decor. The bark kitchen as well as the Eremitage in the park Sanssouci in Potsdam are both oak bark huts that served only for a temporary rest (Lehmann 2004).

The most frequently used bark species throughout all historical periods and regions is birch. Its wide distribution area and the possibility to easily remove the outermost periderm layer without having to cut down the tree for this purpose, makes it suitable for many applications all over the world. In Finland, for example, birch bark is strongly interwoven with Scandinavian culture and objects. Boxes, shoes or even whole suits were made out of birch and many more objects can be found in the Finnish National Collection[16].

Birch bark is still used today. Applications range from boxes and containers for household uses (Mergelsberg 2019), furniture (Koshcheeva-Rasehorn 2021), to flooring (Mergelsberg) and house facades (Blocksdorf 2021). Besides birch, also spruce is used for the production of furniture and decor (Polakova 2018). In order to better understand the properties of barks and thus find more applications, there is a trend in current research to include other professions in the investigations. The "Chemarts Project" at Aalto University creates an environment

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[14] The manuscripts were found at the beginning of the 1990s and came to the British Museum in 1994. See Or. 14195.17, 19–21, 27–27, 35. www.britishmuseum.org/collection (last accessed 6 March 2021).

[15] Wenig, C., Dunlop, J. W., Hehemeyer-Cürten, J., Reppe, F. J., Horbelt, N., Krauthausen, K., ... & Eder, M. (2021). Advanced materials design based on waste wood and bark. Philosophical Transactions of the Royal Society A, 379(2206), 20200345

[16] Many objects can be found in museums all over Europe. For example, in Northern Europe in Finland various household items were made of birch bark. All examples are taken from the collection of the National Museum of Finland. See museum numbers KB2664b (woven bag made of birch bark, made before 1893), KA7424 (shoes made of birch bark, made before 1906),Photos credit: Photo: Finnish Heritage Agency, Johnny Korkman). www.finna.fi (last accessed 23.8.2021).

for design and chemistry students. Both fields are jointly investigating processes and applications for spruce- (bark), willow- (bark) and other cellulose-based materials (Aalto University's Master's Degree Programme 2020).

Overall, the application fields of bark as a bulk material are still very limited (with the exception of birch bark). Considering the quantities of tree bark accumulating in industry, an expansion of the used types of bark and their fields of application is desirable.

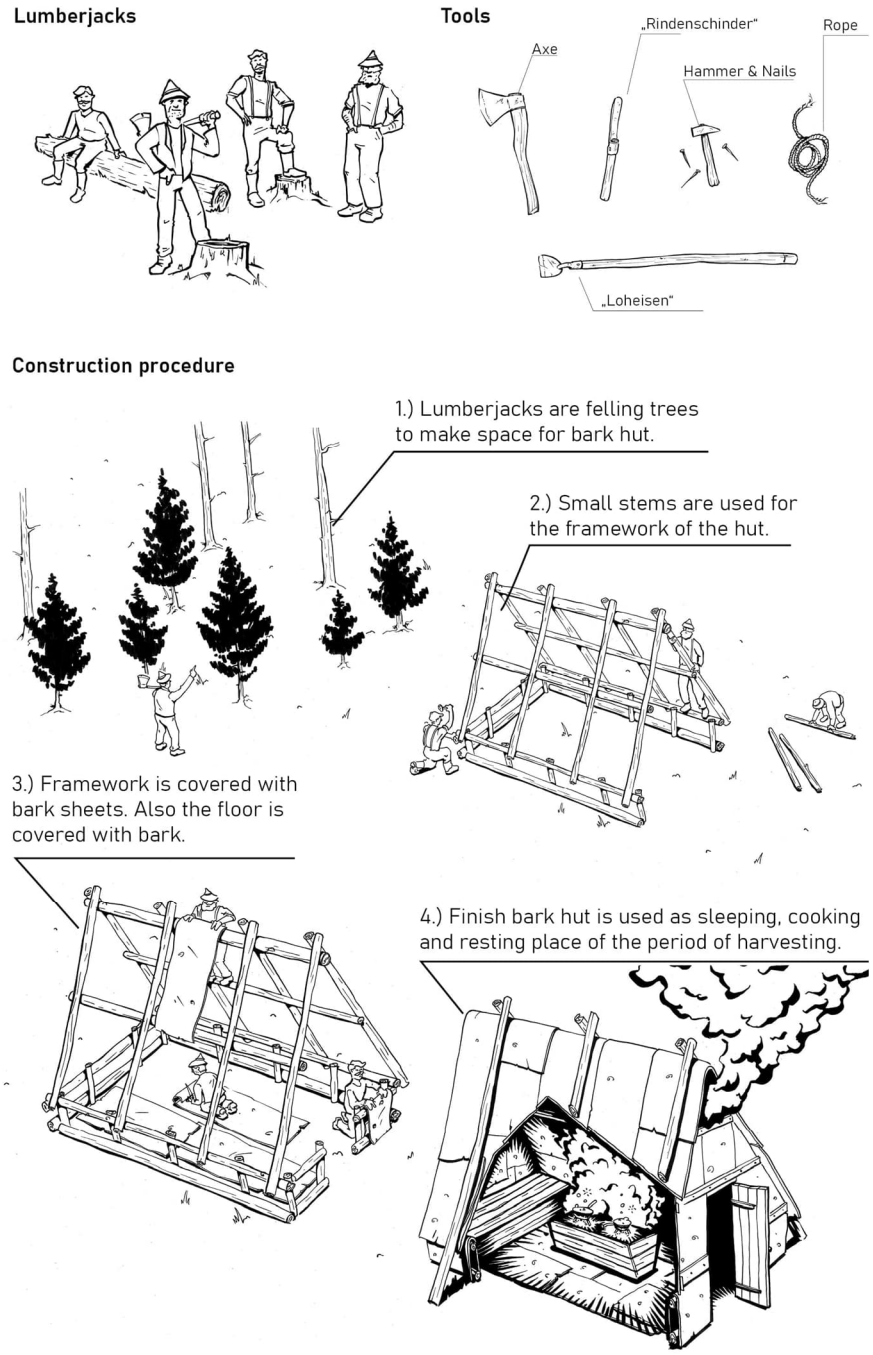


Figure 2.11: Construction of historical bark huts. *The illustration shows the construction of bark huts, which were used in the Austrian Saline region until the beginning of the 20th century. First, seasonal lumberjacks created open space near the harvesting area. For this purpose, smaller trees of no interest to the lumberjacks were felled. Then, these logs were used to construct the hut structure. Afterwards, the bark of the felled trees (mostly fir or spruce) was used for the roof, the walls, the floor and the interior covering. The size of the bark hut varied depending on the number of lumberjacks and the length of time the hut was used. In most cases, the huts were no longer used after the end of the season and therefore became dilapidated. Comic made by Florian Weisz.*

3 Material and Methods

This chapter is divided into two subchapters. The first part (chapter 3.1), material processing, describes the harvesting and production of bark (materials), which are then used for material characterization and design experiments. In the second part (chapter 3.2) the individual material characterization methods are presented.

3.1 Material Processing

3.1.1 Peeling/Harvesting

Freshly felled birch (*Betula pendula*, Roth), larch (*Larix decidua*, Mill.), oak (*Quercus robur*, L.), pine (*Pinus sylvestris*, L.) and Spruce (*Picea abies*), robinia (*Robinia pseudoacacia*), lime *(Tilia sp*.) were debarked manually in May-June 2018 -2020 in the forests of Potsdam and Birstein, Germany. The tree trunks were cut to ~ 60 cm - 100 cm long pieces (Figure 3.1.1, a). Afterwards a longitudinal cut was placed in the bark including the bast and cambium (Figure 3.1.1,b).Pieces of bark with lengths between 600 – 1000 mm and widths of 300 – 600 mm were removed by hand or with a handle (Fig. 3.1.1,c).

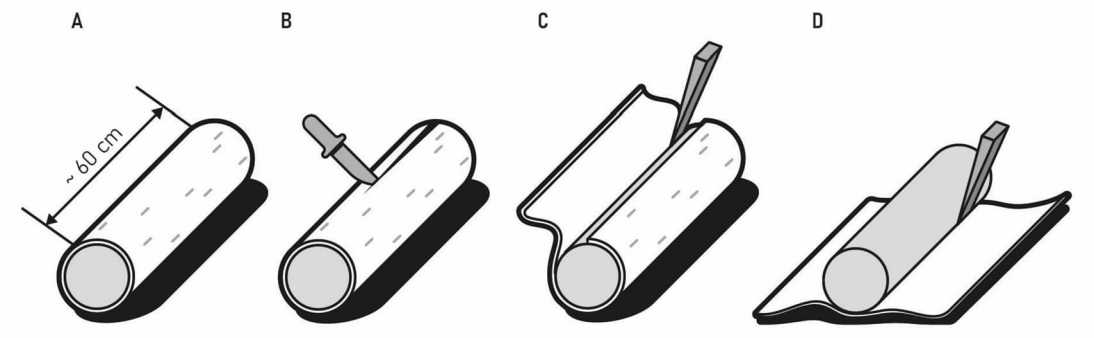


Figure 3.1.1*: Process of bark peeling. Cut of the stem into needed length for bark peeling (A). By creating a longitudinal cut (B), the bark including bast and cambium becomes a starting point for the peeling (C). By hand or with a handle (D) the bark is removed in one piece.*

3.1.1.1 Drying

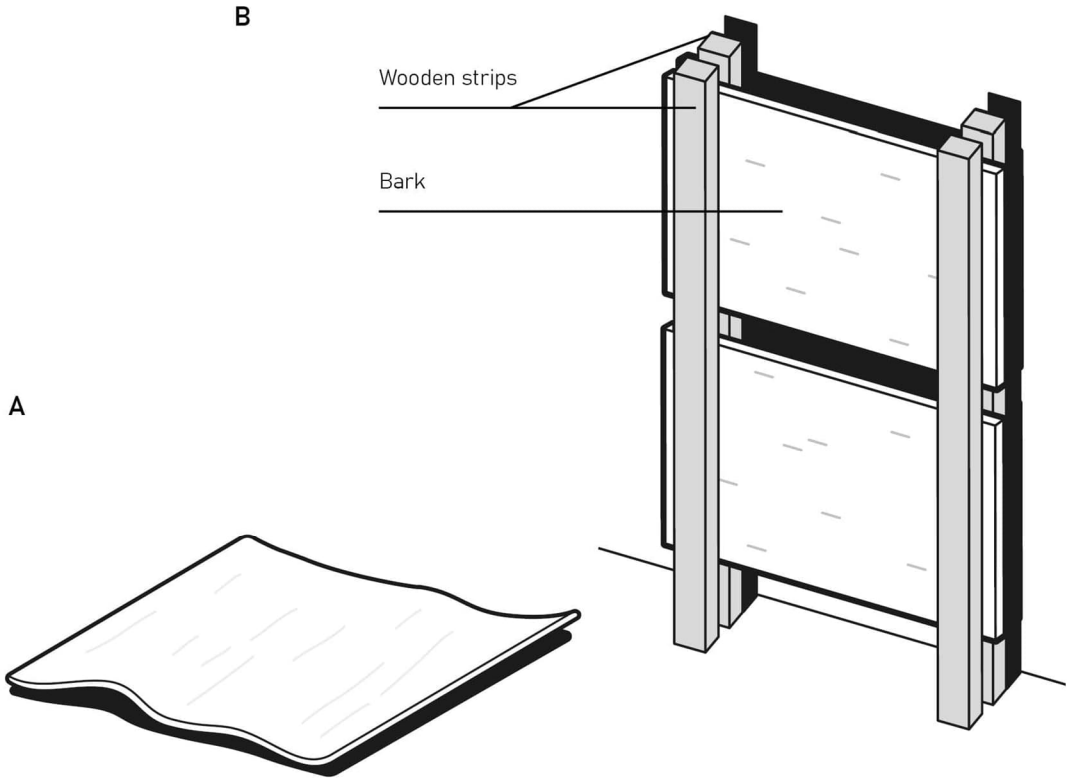


***Figure 3.1.2**: Different options of bark drying depending on bark species. Bark piece is laying on the ground for drying without fixed fixation. This option is used for pine bark (A). To prevent curling, freshly cut bark (e.g. oak, larch, spruce, birch) is fixed between wood logs (B).*

After peeling the bark is very flexible. To prevent oak, larch, robinia, lime, birch and spruce from curling upon drying, all samples were fixed with screws between wood strips (Figure 3.1.2, b). Without fixation all bark rolled up after a few hours. Because Pine bark is not curling when drying it was laid out on the ground without any fixation (Figure 3.1.2, a). The drying process for pine takes around 3 days. Afterwards pine bark was piled up and stored under a shelter until further use. Other barks were also stored under an outdoor shelter for 6 weeks. After this initial natural drying the wood strips were removed and the flat bark pieces were stored outside under a shelter for at least another 8 weeks. Samples with dimensions of 10 cm x 10 cm x bark thickness, 22 cm x 22 cm x bark thickness and 30 cm x 30 cm x bark thickness were cut and stored under laboratory conditions until use.

3.1.2 Densification

3.1.2.1 Panels

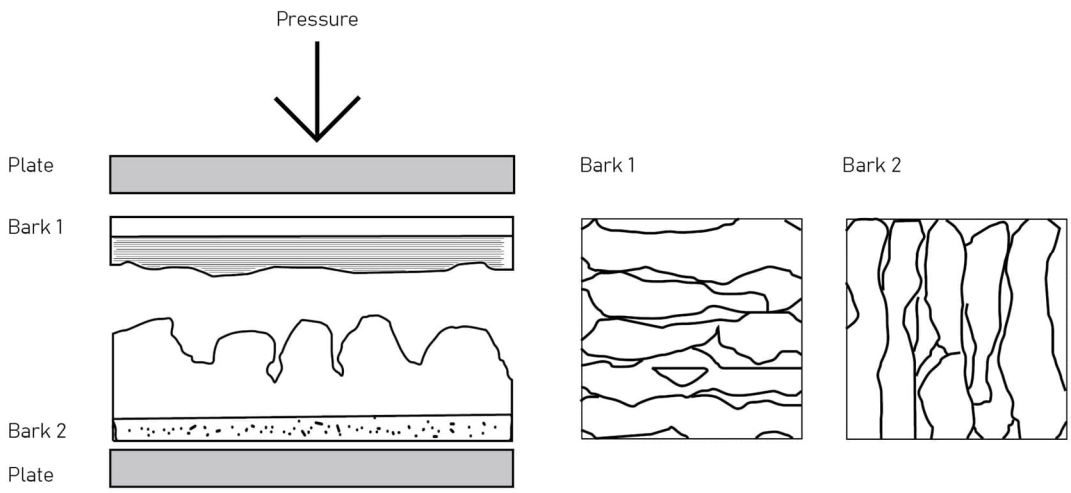


***Figure 3.1.3**: Production of flat panels. Arrangement of the bark pieces in the hot press (left). Two bark pieces were combined crosswise (right).*

Production of flat panels (FP1)

Laboratoy-stored pieces of bark with in-plane dimensions of 10 cm x 10 cm, 22 cm x 22 cm and 30 cm x 30 cm were pre-heated at 90 °C in a closed oven without circulating air for 20 – 30 minutes. Directly after pre-heating two bark pieces were combined crosswise, with the rhytidome sides facing each other (Fig. 3.1.3), in a heated hydraulic press (ZSCHOKKE WARTMANN, IMEX). To keep specific pressure constant, the oil pressure was set to 20 bar and 97 bar for 10 x 10 cm^2 and 22 x 22 cm^2 samples, respectively. The panels were pressed for 20 minutes at 90 °C before cooling them down outside the press at room temperature (22° C).

Production of flat panels made of bark with different moisture contents (FP2)

In a second test series, to examine the influence of the moisture content of the raw bark on the panel properties, barks of oak and larch (available in large quantities) with four different defined moisture contents were used for panel production. For both tree species, 16 square

bark pieces each (25 x 25 cm) were stored at 20 °C and relative humidities (RH) of 15 %, 40 %, 65 % and 95 % until the mass constancy of the samples was reached. The bark panels were pressed crosswise on a Siempelkamp hot press (G.Siempelkamp GmbH & Co., Maschinen- und Anlagenbau, Krefeld, max press area 50 x 100 cm) with a specific pressure of 97 bar at 90 °C and for 20 minutes. Immediately after the pressing process, the panels were removed and allowed to cool in an air-circulated standing position at room temperature for 30 min. This was followed by storage at 20 °C and a RH of 65 % until equilibrium mass was reached.

3.1.2.2 Curved elements

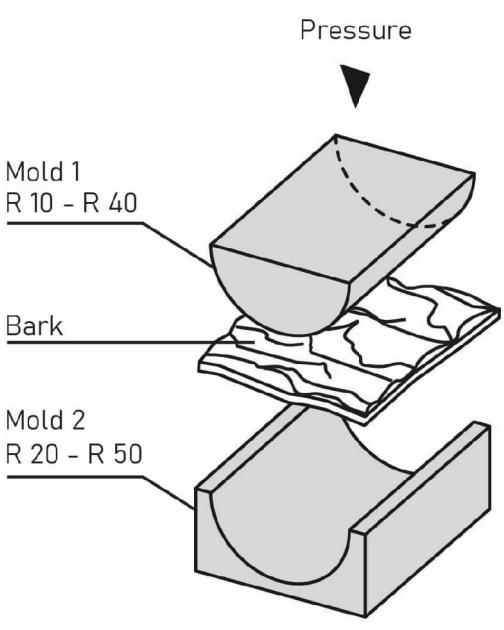


***Figure 3.1.4**: Production of curved and densified bark samples. Bark sample is placed between both mold parts and pressed. Radius of press curves range from 10mm up to 50mm*

In another approach the feasibility to create 3D shaped geometries was tested. Larch, pine, oak and birch bark without glue or resin (Fig 5.2.2) were molded with the goal to explore limits in achievable curvatures. For this, bark with in-plane dimensions of 10 cm x 10 cm were pre-heated at 90 °C. Directly after pre-heating the bark piece was placed in curved molds with the axial bark direction following the curve. Different diameters (10mm/20mm;

20mm/30mm; 30mm/40mm; 40mm/50mm) as shown in Figure 3.1.4 were used. The mold was placed in a heated hydraulic press (ZSCHOKKE WARTMANN, IMEX). For 10 x 10 cm samples the oil pressure was set between 3 and 20 bar -depending on the dimensions of the mold and the piece of bark. The samples were pressed for 20 minutes at 90 °C. Afterwards the curved panel were removed from the mold for cooling down at room temperature (22°C).

3.1.2.3 Form

To examine the possibility to create 3D geometries with corners and edges in bark, densification tests were made. Larch, pine, oak and birch bark without any adhesives were pressed into a self-designed mold (Figure 3.1.5). The mold itself consists of 2 parts of milled aluminum. The bark with in-plane dimensions of 10cm x 10cm was pre-heated to 90° before transferring it to the mold (Figure3.15, A).

The mold was then placed in a heated hydraulic press (ZSCHOKKE WARTMANN, IMEX). For 10 x 10 cm samples, the oil pressure was set by 15 bar (Figure 3.1.5, B). The samples were pressed for 20 minutes at 90 °C (Figure, C). Afterwards the densified panel was removed from the mold for cooling down at room temperature (22°C) (Figure 3.1.5, D).

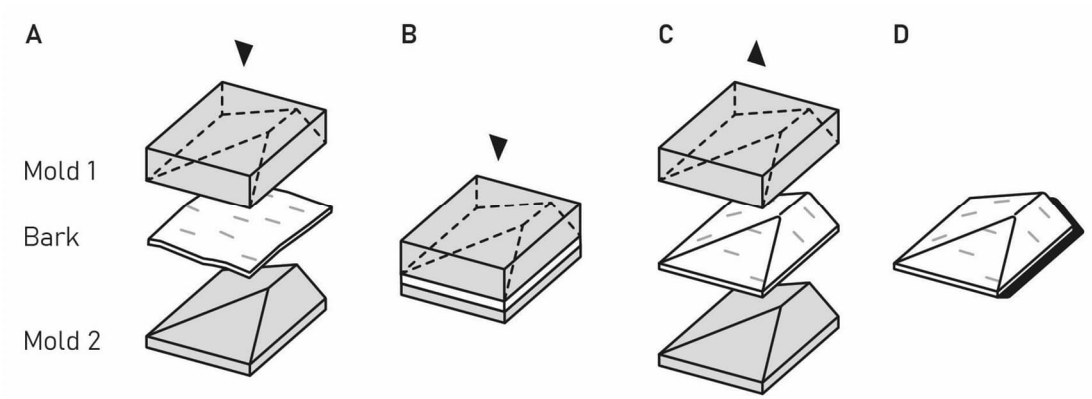


Figure 3.1.5: *Production of three-dimensional shapes. Schematic illustration of the pressing process of bark pieces into three-dimensional shapes. (A) Placing bark between open forms, (B) densification, (C) opening step, (D) pressed bark piece*

3.1.3 Flexibilization

For wood and also for bark, mechanical properties like flexibility are strongly dependent on their moisture content (Eder et al. 2021). With drying bark becomes harder and brittle. To keep bark in a flexible state, selected bark species were infiltrated with water-glycerol solutions (Figure 3.1.6). Sample flexibility was tested before and after the treatment with water-glycerol solutions.

Flexibilization process

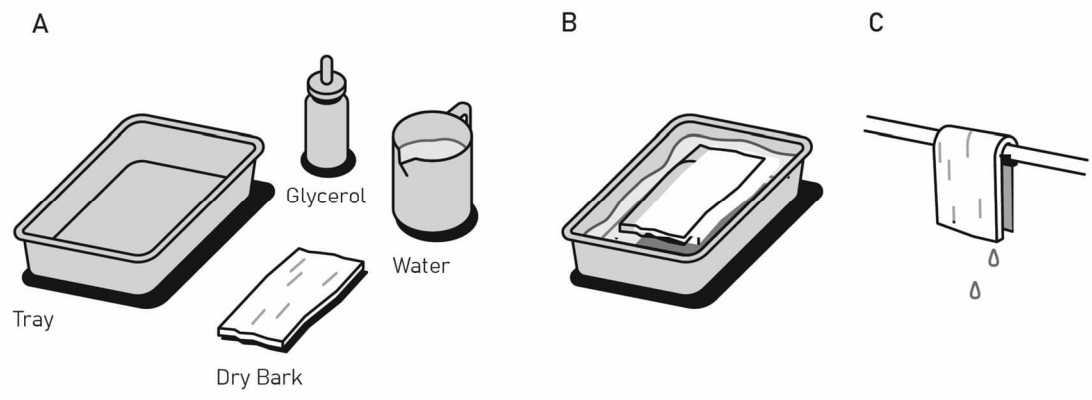


Figure 3.1.6: *Flexibilization of bark with a glycerol-water solution. schematic illustration of flexibilization process (A) items need for flexibilization, (B) bark is placed in an open try with a water glycerol solution, (C) after infiltration time, bark is removed from the solution and hung in the lab for air drying for several days*

Oak harvested in 2019 and stored outside under a roof for 4 monthes was moved into the lab (lab conditions) for further sample preparation. With a circular saw, samples were cut to pieces with dimensions of 10 cm (along the fiber) x 10 cm (across the fiber) x bark thickness. A few oak bark pieces were planed on the rhytidome side (via electric planer, Oberlin Werk Potsdam) and as a result had a lower material thickness than the other oak samples. Pine bark (harvested in Katharinen Holz, Potsdam 2018/2020) was taken from sprawling thick bark (Borke) and younger thinner parts (called mirror bark). Samples from both parts were cut to 10 cm x 10 cm x bark thickness. Birch bark was ordered from Sagaan (Mergelsberg 2019).

The delivered bark was not flat, possibly caused by changing moisture conditions during transport. Hence that preparation of flat samples with precise dimensions was not possible. Still, two samples of the outer rhytidome of birch were cut with a circular saw. Samples of larch (Katharinen Holz, Potsdam 2018) were cut with a circular saw to 10 cm x 10 cm x bark thickness. While one larch sample includes all parts of bark layers, one has the wooden flakes (outer rhytidiom) partly removed. The third sample was planed in Oberlin Berufsbildungs-werk, Potsdam.

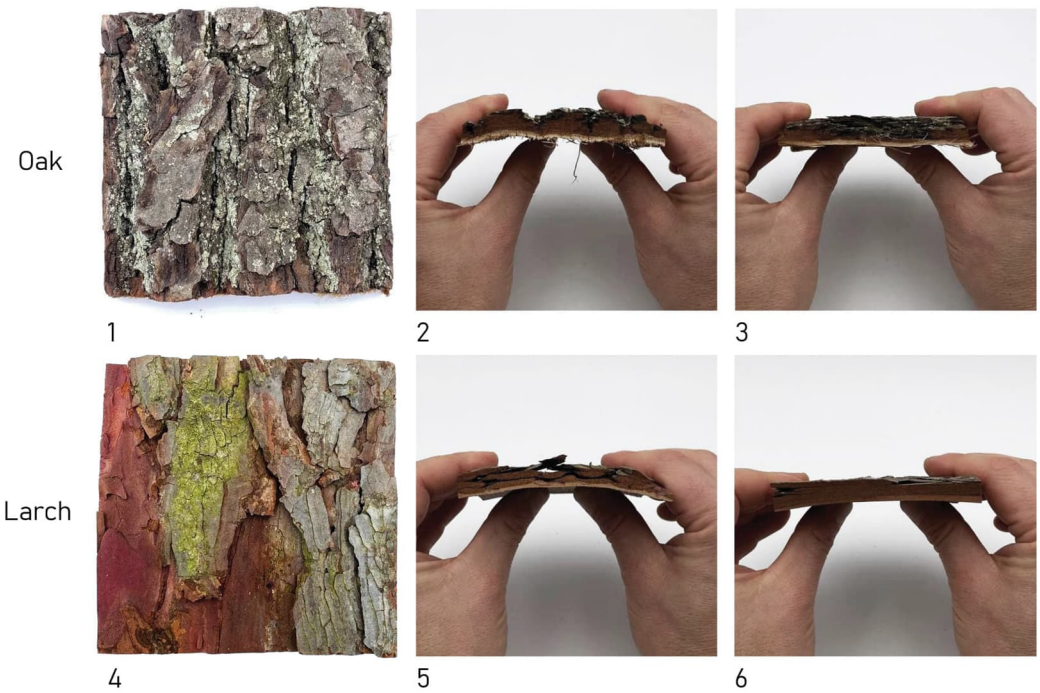


Figure 3.1.7: *Qualitative testing of flexibility of bark sample before treatment. Flexibility of oak (1) and larch (4) bark: image 2 shows flexibility of oak bark and image 5 of larch bark before treatment radial, image 3 shows flexibility of oak bark and larch bark (6) before treatment longi- tudinal*

Before flexibilization, all samples were qualitatively tested for their flexibility (Figure 3.1.7) by bending them gently between the hands in longitudinal (Figure 3.1.7, 3,6) and perpendicular fiber direction (Figure 3.1.7 2,5). Afterwards all samples were species wise immersed in glycerol-water solutions at 20 °C consisting of 1 volume part glycerol (SV Liquid Production GmbH, Glycerol 99.7% USP/EP) and 4 volume parts water. Bark samples were kept immersed

in an open tray under lab condition for 48h. Afterwards all samples were taken out of the trays and allowed to dry at room temperature (22°C) for several days (5 days). Then the qualitative flexibility test was repeated in both directions.

Treatment of pine mirror bark with glycerol-water-solutions with different concentrations

To examine the influence of the concentration of the water-glycerol mixture on the properties of the mirror pine bark, dry bark samples were infiltrated with varying glycerol concentrations. Pine (Michendorf 2020) was dried outside for 2 month and stored under lab conditions for 3-5 days until infiltration. Dry bark was broken by hand into ~ 15cm x 30cm pieces (along the fiber direction). All pieces broke brittle before treatment. Pieces of pine mirror bark were immersed in glycerol -water solutions with ratios of 1:1,1:2,1:4,1:6,1:8 for 48h. The steps used are similar to those studied by E. Paparozzi and D. McCallister (Paparozzi and McCALLISTER 1988) for the preservation of different cultivars of *Limonium sinuatum*. All pieces were removed after infiltration and allowed to dry under lab conditions. Next, the pieces were taken one by one in the hands and carefully bent in all directions.

3.1.4 Burning/Ash

For the first incinerations, several kilograms of tree bark were used. To achieve a similar composition of industrial (bark) waste, bark from the sawmills "Furnierwerk Merkscha GmbH" in Gratwein and the "Furnierwerk Prignitz GmbH und CoKg" in Pritzwalk was used. Oak *(Quercus robur)*, pine *(Pinus sylvestris)*, robinia *(Robinia pseudoacacia)*, spruce *(Picea abies)*, larch *(Larix decidua)* and birch *(Betula pendula)* were used in the first burning procedure. Before incineration, the bark was stored under lab conditions (~ 22 °C and ~50% RH) until further processing. Tree bark was burned in a gas furnace at the Hochschule für Künste in Bremen, Germany and at "Müritzkeramik" in Lärz, Germany. Barks were burned at 600°C for 3 hours.

The gas furnace (self-built for university purposes), which was kindly provided by the University of the Arts, did not obtain a fire temperature of 700°C. In order to follow the targeted temperatures better, a wood furnace was used. Tree bark from the same sources (the two woodworking companies) as well as residues of specifically peeled bark (lime (*Tilia*)), were burned. Another batch of bark was burned in a wood furnace at "Müritzkeramik" in Lärz, Germany. All bark samples were placed in clay pots (~ 10l) (Figure 3.1.8) and weighed before and after burning. During the first firing, the barks were burned with a maximum temperature of ~ 665°C and a holding time of 1h. Since there was still too much charcoal after the first

firing, the barks were burned another time with a maximum temperature of ~ 1000°C and a holding time of 5.5h. All fires were conducted under ambient pressure.

Figure 3.1.8: *Production of bark ash. Furnace with bark before burning, Müritz Keramik, Close up of bark samples inside separate open burning vessels (left images), illustration shows that bark samples are placed in open vessel (A) and then fired in a gas furnace (B). Photograph taken by Müritz Keramik, Markus Böhm*

3.2 Material Characterization

3.2.1 Surface Characterization: Panels

The surfaces of bark panels (FP1) were investigated with a digital microscope (Keyence VHX-5000 Microscope) at a magnification 200x. Profile lines were drawn across the surface and the profile values were exported to a tabular format. Origin 2019 was used to filter the profiles: data of bark panels by means of a 2nd derivative spline baseline substraction over 8 points by a 3rd order polynomial fit. For the determination of the average surface roughness (R_z), the sampling length of 5 equal and consecutive segments was chosen to 0.25 cm, resulting in a total length of 1.25 cm, according to DIN EN ISO 4288. For each segment, the difference between minimum and maximum profile height was calculated. The average of the values for all 5 segments give the maximum height of profile for one sample.

3.2.2 3D Microstructure: Raw Bark and Panels

The natural bark material structure and the changes in structure caused by the pressing process (pressure and heat) were characterized by µCT measurements. Native bark samples and bark panels (FP1) were cut with a band saw. The native bark samples (n=4) had a cross section of ca 1 x 1 cm and varying lengths in radial direction, which corresponded to the thickness of the bark. The panels (n=4) were cut to strips with a width of ca. 1 cm, the cut was 0° and 90 ° to the fiber orientation of the pressed bark pieces. The samples were placed in a µCT scanner (RX Solutions, EasyTom150/160) and scanned with a microfocus X-ray tube unit (Hamamatsu Photonics K.K. made in Japan) at a tube voltage of 60kV and 150 µA tube current. The frame rate of the flat panel detector was 2 and the frame averaging was 8. The acquired radiographs of the native bark samples and the panels were reconstructed with the software XAct2 (RX Solutions) and visualized with the software Amira (Thermo Fisher Scientific).

3.2.3 Density: Raw Bark and Panels

To assess the degree of densification during the pressing process, the density of both natural bark pieces and the produced panels (FP1) was determined, largely following DIN EN 323. Oak, larch, birch and pine (n=6 / tree species) bark was cut with a circular saw into 2.5 x 2.5 cm large pieces. Additional samples of strongly matured, thick bark of oak and birch were cut to the size of 5 x 5 cm^2. The pieces were stored in a climate chamber (VÖTSCH, VCL 4010) at 20 °C and 65 % relative humidity and weighed every 24 hours until the constancy in weight was reached. To account for the rough bark surface of raw bark samples, volume determination was performed with a 3D imaging approach. The conditioned samples were scanned with a micro computed tomograph (EasyTom 150/160 RX Solutions) with a microfocus tube operated at 40 kV and 300 µA in the large focal spot mode. The framerate of the flat panel imager was 12.5 without averaging, resulting in scan times of approximately 5 minutes. The recorded radiographs (896) were reconstructed with the software XAct and further processed with Amira. The bark pieces were segmented and volume was calculated. Immediately after the scan the samples were stored again in the climate chamber until their weight changed less than 0.1% within 24 hours. The weight was recorded and the density of each sample was calculated with the previously determined volume. The density of the bark panels was determined on 5 cuboids per species with in-plane dimensions of 2.5 x 2.5 cm. The panel thickness

was variable and ranged from 5.7 mm – 11.9 mm. The cuboids were stored in a climate chamber at 20 °C and 65 % relative humidity until their weight changed less than 0.1 % within 24 hours. The sample dimensions were determined with a caliper directly after weighing.

3.2.4 3-Point Bending Test: Panels

To get an initial idea about the mechanical properties of the bark panels (FP1), samples with a width of 2.5 cm and a length of 21 cm were cut for a 3-point bending test. Care was taken to cut at 0 ° and 90 ° angles to the grain of the panels. The samples were then stored until testing at 20 °C and 65 % relative humidity until their weight did not change more than 0.1 % within 24 hours. The mean thickness and mean width of each sample was calculated from 3 measurements, taken with a caliper at the center and on both support points. Bending tests were conducted on a Zwick universal testing machine equipped with a load cell with a maximum capacity of 10 kN. Two test series were performed for each wood species with the grain orientation either along or across the tension side of the bending sample. The test span was 15 cm and the test speed was set at 0.1 mm s^{-1}. The samples ($oak_{long.}$ n=7, $oak_{perp.}$ n=6, $larch_{log.}$ n=8, $larch_{perp.}$ n=11) were tested until fracture, strain was measured via cross-head displacement. The determination of the bending modulus E was calculated according to eq. 1:

$$E = \frac{\Delta F * l_1^3}{4 * b * t^3 * \Delta\varepsilon}$$

where ΔF is the force increase in the linear region, l_1 the distance between the supports, b the sample width, t the sample height and Δε the deformation increase in the middle of the beam.

3.2.5 Transverse Tensile Test: Panels

Determination of transverse tensile strength (FP2)

To characterize the effect of moisture content during pressing on the connection between the crosswise pressed bark pieces, transverse tensile strength perpendicular to the plane of the panel was determined according to the standard DIN EN 219. The climatized bark panels were cut to samples with a size of 50 mm × 50 mm x panel thickness. Ten samples cut out of 2 panels pressed with the same moisture content and conditions were mounted with polyurethane glue (Kleiberit PUR 501) between beech plywood yokes. The glued samples were clamped

into prefabricated templates using screw jaws. Additional wooden spacers prevented the specimens from slipping sideways between the yokes. The transverse tensile tests were carried out on an universal tensile machine "Zwick Roell GmbH, 1484" equipped with a load cell with a maximum capacity of 200 kN.

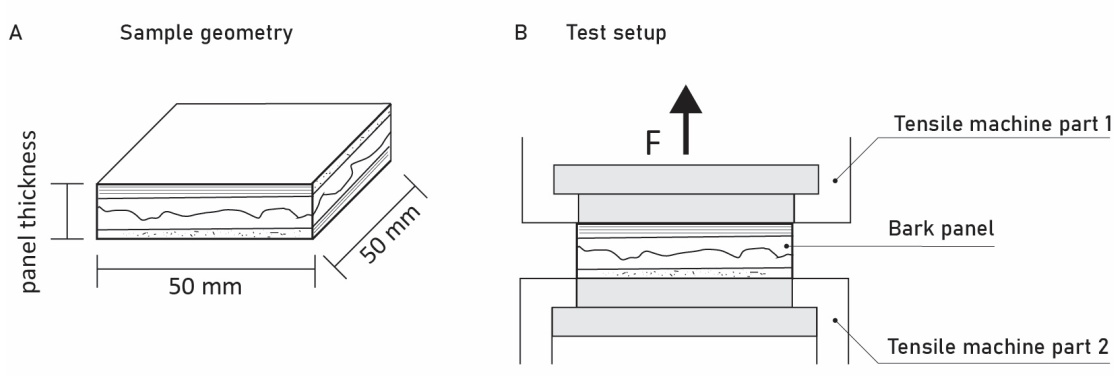


Figure 3.2.1*: Transverse tensile test. Samples with a length of 50 mm and a width of 50 mm were cut out of panels. Sample thickness corresponded with panel thickness (A). For tensile test, samples were glued on beech plywood (gray) and placed in the test set up (B).*

Prior to material testing, the normal densities and equilibrium moisture contents were determined. The determination of the transverse tensile strength was calculated according to eq. 2:

$$\sigma_Z = \frac{F_{max}}{a * b}$$

Where σ_Z is the transverse tensile strength, Fmax the maximum force and a*b the cross sectional area.

The results of the panels manufactured of bark with different moisture contents were compared with each other and statistically analyzed with the Mann-Whitney test (OriginPro 2021b).

3.2.6 Swelling and Shrinkage: Raw and Panels

Swelling and shrinkage of native bark cubes

Materials in applications are continuously exposed to changing environmental conditions (relative humidity and temperature) and water contact is one of the big challenges encountered by using lignocellulosic materials as it strongly influences the material properties. While directional swelling movements of wood and derived timber products are comparably well described (Engelund et al. 2013), (Eder et al. 2021), only little is known about swelling and shrinkage of bark.

To compare the swelling and shrinkage behavior of the native bark and the compressed panels, bark cubes without cracks of oak and larch were produced. Small sized strips were cut from the dried bark from oak and larch bark. For each species two subseries of specimens (oak n=12, larch n =10) were produced originating from two different trees. While the first series included both the phloem and the phellem tissue the second series exclusively comprised phellem tissue where the adjacent phloem has been cut off. The strips were planed square on two adjacent sides (radial/tangential plane) with a small hand plane. Afterwards both opposite sides were trimmed with a microtome (Leica RM 2255) to a thickness of 10 mm each. The resulting square strips were cut to a length of 10 mm with an electric mitre saw. Areas which included cracks or visible voids were discarded. The resulting bark cubes were conditioned in a climate chamber (Vötsch VCL 4010) at 65 %, 40 %, 20 %, 85 %, and 0 % relative humidity at 20 °C. Throughout the experiment the climatic conditions were measured with a calibrated sensor (SHT85, Sensirion). The lengths of the samples along their three anatomical directions were measured with an outside micrometer at constant weight (difference < 0.1 % / 24 h, balance: PCE AB100C) which was usually reached after 3-4 days of conditioning. The dry state of 0 % relative humidity was achieved by a direct input of compressed air into the chamber (dew point below -30 °C). For the evaluation, the results of the state at 65 % relative humidity served as reference.

Swelling and shrinkage properties of larch and oak panels pressed at different moisture contents (FP2)

To investigate the sorption, swelling and shrinkage properties of the panels, larch and oak samples (55 mm x 55 mm x panel thickness, (n=10 per species)) were exposed to five climatic

cycles. A climatic test chamber (Feutron type "035/09") was used for climatization. Each cycle was completed when all samples had reached an equilibrium state. The dimensions of the panels were measured with a digital sliding caliper (accuracy of ±0,01 mm), changes in thickness and length were analyzed. The equilibrium sample dimensions of the starting cycle 1 (20 °C, 65 % RH), were defined as reference point for the calculations of the percentage swelling and shrinkage dimensions, according to standard DIN EN 317. To investigate the shrinkage of the panels, the relative humidity was reduced from the starting cycle 1 to cycle 2 (20 °C, 40 % RH) and to cycle 3 (20 °C, 20 % RH). This was followed by an increase in humidity in cycle 4 (20 °C, 85 % RH) for the investigation of swelling. Cycle 5 was drying at 80°C and laboratory RH.

3.2.7 Tensile Test: Wet, Dry, Flexible Pine Bark

The mechanical properties of dry and water soaked (48 h) pine mirror bark and mirror bark treated with differently concentrated water-glycerol mixtures were analyzed with tensile tests. Tensile specimens with dimensions of 160 mm - 200 mm in the longitudinal and 10 mm in the tangential direction were prepared by cutting them with scissors or a scalpel. The prepared samples (excluding the wet sample) were then stored under lab conditions (23,5 °C and ~50% RH) until testing.

To be able to compare the mechanical properties of the flexibilized barks with similar materials, leather (pit tanned and drum tanned), adult normal spruce wood and branch compression spruce wood were cut to (if possible) similar dimensions and tested as described above.

Two types of leather were selected based on a haptic comparison of flexible bark with different leather types. Test bodies of the same length and width as bark samples were prepared (200 mm in the longitudinal and 10 mm in the tangential direction).

Using a handsaw, small blocks of ~ 60 mm x 10 mm x 20 mm (LxRxT) were cut from adult normal wood of the outermost growth rings of an adult spruce trunk and from branch compression wood of the lower side of a spruce branch close to the base of the branch. The small wood blocks were soaked in water over night before they were cut into consecutive150 µm thick longitudinal-radial slices with a rotary microtome (Zeiss RM 2255). The resulting slices were cut to a final width of ~4 mm with a razor blade. Two groups of randomized wood samples were allowed to dry over night under ambient conditions. To prevent twisting the slices were put between metal grids while drying. After drying one group was placed into a glycerol-

water solution with a mixing ratio of 1:4 for 48 h and then dried under lab conditions. The second group was stored under lab conditions and the third group was kept in water until testing.

Before the test the width of each sample was measured 3 times along the test span with a caliper and the thickness was measured with a micrometer. The samples were tested with a Zwick universal testing machine (Zwicki Z2.5) equipped with a load cell with a maximum capacity of 1kN. The test span was 100 mm for the bark and leather samples and 30 mm for the wood samples. The test speed was set at 0.04 mm s^{-1} for the bark samples, 0.5 mm s^{-1} for the leather samples. To compensate for the shorter test span, the wood samples were tested at a speed of 0.5 mm s^{-1}. The samples were tested until fracture, strain was measured via an in-built video extensometry.

3.2.8 Creating the Bowls

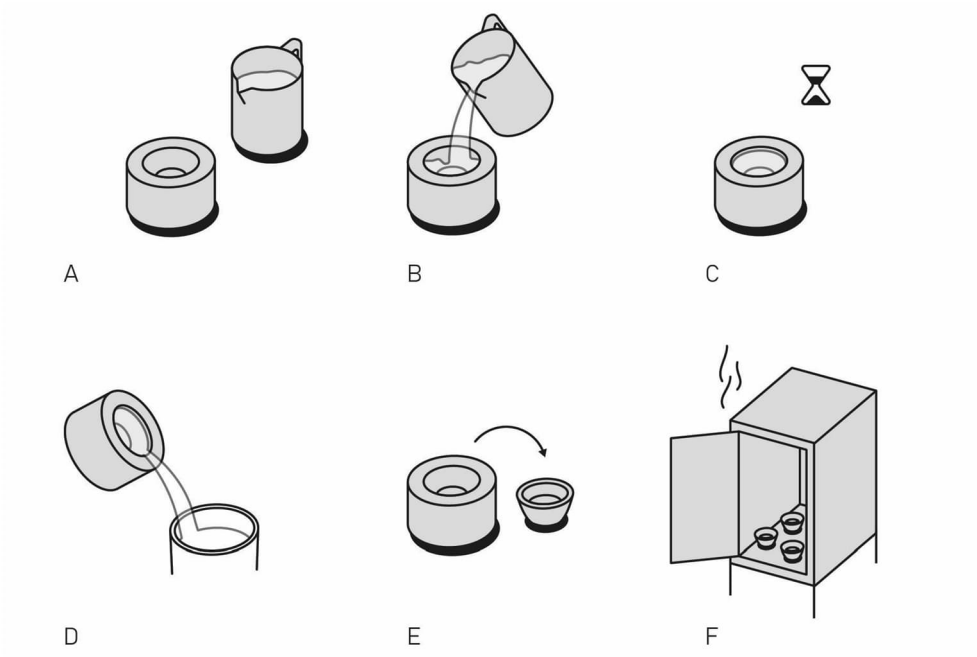


Figure 3. 2. 2: *Production of bowls by using positive casting molds. A plastic sheet is formed into a cylinder according to the required diameter and placed on the table (A). Using fresh clay, the cylinder is fixed on the table and filled with liquid plaster (B). After the plaster has dried from liquid to moldable consistency, the bowl is pressed into the plaster (C). After drying, the cylinder and the bowl can be removed. The molds are filled with porcelain (D) and after approximately 5-15 minutes the remaining porcelain is poured out. After another 20 minutes, the raw porcelain mold can be removed (E).). The raw porcelain bowls are fired in a gas furnace (F).*

For testing the glazes, bowls were produced (Figure 3.2.1). All the bowl sizes were cast into the positive mold made out of plaster (Stucco plaster, Formula Saint-Gobain, Germany). The material used was limos (Limoges PC010B, Imerys Ceramics, France). The raw porcelain bowls dried in the plaster molds and were then fired in a gas furnace (the small bowls at the Hochschule für Künste in Bremen and the watch glasses at the Kunsthochschule Weißensee) in a biscuit firing (~850°C to 1000°C for at a holding time of 30 minutes) (Figure 3.2.1 (E)). The porous and fragile bowls were thus transformed into a solid and more robust state.

3.2.9 Inductively Coupled Plasma (ICP)

Ash from 7 different bark species (Oak *(Quercus robur)*, pine *(Pinus sylvestris)*, robinia *(Robinia pseudoacacia)*, spruce *(Picea abies)*, larch *(Larix decidua)*, birch *(Betula pendula)* and lime *(Tilia)*) were analyzed with ICP. All bark ash was filtered with a fine-mesh sieve and then weighed. Afterwards, samples were dissolved in 500 µl aqua regia (167 µl HNO3 + 333 µl HCl) at 90°C for 2h and filtered with a filter paper. The produced solutions were diluted at a 1:100 ratio with Millipore H2O. Emission spectra were captured with an ICP-OES analyzer (Optima 8000, Perkin-Elmer). To atomize the sample solutions a concentric glass nebulizer (CemCon) equipped with a glass cyclonic spray chamber and an Argon plasma torch were used. Spectra were acquired with a dual échelle polychromator in combination with a CCD detector. The software WinLab32 was used for sample analysis.

3.2.10 Bark Glaze Recipe

Filtered bark ash was added to the glazes before the firing process. The recipe for the transparent porcelain glaze (borate glaze) was provided by the ceramic workshop supervisor Ute Fischer (Hochschule für Künste Bremen). The recipe for the glaze is given in table 3.2.1.

Water	120 ml

Silica	33.39 g
Whiting	15.3 g
Georgia kaolin	10.91 g
Nepheline Syenite	15.55 g
Gerstley Borate	8.45 g
Magnesium	1.86 g
Bark ash	30 g

Table 3.2.1: Recipe for transparent porcelain glaze with borate (borate glaze)

All chemicals were mixed with 120 ml of water. Afterwards, glazes were applied by pouring glaze into the form. After a few seconds the excess glaze solution was poured out. After drying for 48h, pre-glazed bowls were fired in a gas furnace (University of Arts, Bremen) for 4 hours at 650°C and finally at 1250°C for 30 minutes.

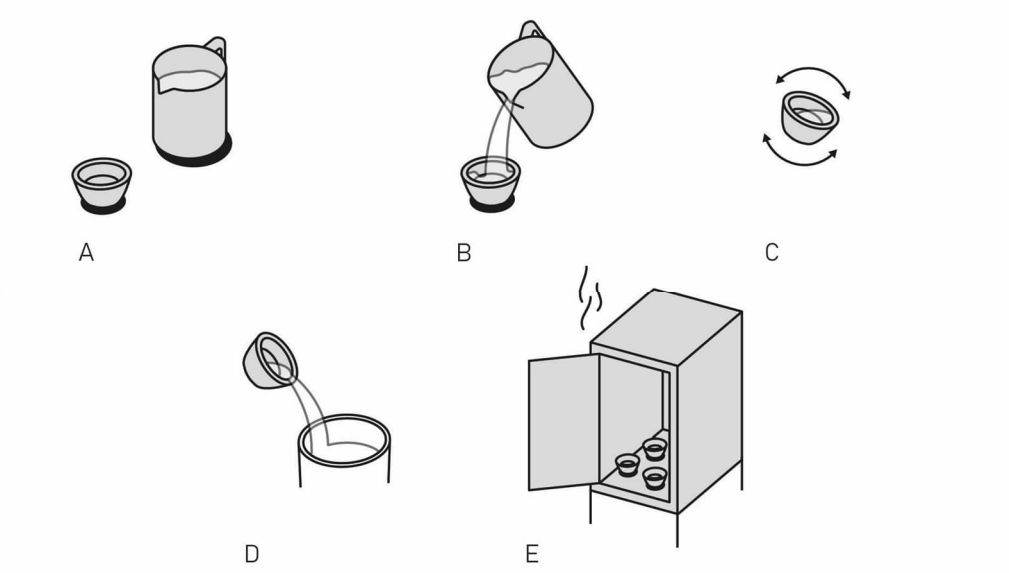


Figure 3.2.3: *Steps of glazing the bowls. Bowls are burnt in a biscuit firing and ready for glazing* (A), *glaze is poured into the bowl (B), bowl is gently moved to civer all inside areas with glaze (C), excess glaze is poured out (D), glazed bowls are dried over 24h and afterwards burnt in the furnace (E)*

Further glazing experiments were kindly realized in cooperation with the ceramist Dirk Friese (Hismalin, Kotzen, Germany). In these experiments bark ashes of Oak *(Quercus robur)*, pine *(Pinus sylvestris)*, robinia *(Robinia pseudoacacia)*, spruce *(Picea abies)*, larch *(Larix decidua)*, birch *(Betula pendula)* and lime *(Tilia)* were used. As a further test series, a recipe for the production of a mat transparent glaze (Tab. 3.2.3), a glossy transparent glaze (Tab. 3.2.2)

and a crystal glaze (Tab. 3.2.4) were used. For applying glazes on the test bowls, all glazes are poured into the bowls. Excess glaze was poured out short after.

Water	100 ml
Silica	25 g
Whitin	10 g
Kaolin	20 g
Zinkfritte FM5021	5 g
Na-Feldspat LF90	24 g
Dolomit	10 g
calciumboratfritte M9152	5 g
Titandioxid	2,5 g
Bentonit	2,5 g
Bark ash	20 g

Table 3.2.2 : Recipe for the transparent porcelain glaze

Water	100 ml
Kali-Feldspat D 75	23 g
Silica	23 g
Kaolin	23 g
Dolomit	23 g
Zinkfritte – FM 5021	8 g
Bark ash	30 g

Table 3.2.3 : Recipe for matt porcelain glaze

Water	100 ml
Zinkfritte FM 77355	60 g
Titandioxid	9 g
Strontiumkarbonat	8 g

Lithiumkarbonat	3 g
Bentonit 57	5 g
Whiting	5 g
Na-Feldpsat LF 90	10 g
Bark ash	30 g

Table 3.2.4 : Recipe for crystal porcelain glaze

In addition, ash from oak *(Quercus robur)* and lime *(Tilia)* was used without any other additives, i.e. glaze. To provide adhesion of the ashes and a possible melting process, ashes were mixed with 12%, 24% and 36% flux (melter) respectively. Both, lime *(Tilia)* and oak *(Quercus robur)* glaze were applied to the porcelain using a brush.

4 Results and Discussion

The following chapter presents results and subsequent discussions on the main topics of the thesis. In chapter 4.1, the peeling and drying process of bark is discussed, which was essential for collecting all bark needed for following experiments. Chapter 4.2 shows the results of the densification of bark and its structural and mechanical changes compared to the non-densified raw bark. Chapter 4.3 discusses the influence of glycerol on mechanical properties of bark. These results are discussed and compared with mechanical characteristics of wood and leather. Objects presented in chapter 5 were mainly designed with flexibilized bark. The connection of the mechanical properties of glycerol treated pine bark and its effect on the design is referred to in flexibilization section 4.3. In section 4.4, bark ashes and their effect in porcelain glazes are discussed. Finally, chapter 4.5 discusses the possibilities of sustainable long-term use of bark.

4.1 Harvesting and drying of bark

4.1.1 Peeling /Harvesting

Figure 4.1. 1: *Peeling bark in the forest. Larch bark was peeled with the aid of simple wedge-shaped tool (left image). Spruce bark is carefully peeled with a metal handle (right). Photographs taken by Florian Weisz*

Like wood, bark is a large-scale material. To retrieve it from a tree, the traditional method of hand-peeling, typically applied in early spring when trees are full of water, was used (Figure 4.1.1).

The manual peeling process of bark pieces provided initial, qualitative information about the peeling properties: birch, oak, pine, larch, spruce, robinia and pine can be peeled with human strength in spring, indicating that it is easily feasible by human power for small numbers of trees. However, manual debarking of Scots pine was classified with a heavy workload intensity (Çağlar 2021). Since the peeling direction was around the stem axis, it is conceivable that industrial processes, similar to the ones in veneer production, could be adapted for large-scale bark harvests.

4.1.2 Drying of Harvested Bark

Figure 4.1.2: *Drying of freshly peeled bark. Pieces of pine drying in the sun with the cambium side facing the sun (left image). Oak bark fixed between wood logs (right).*

All harvested barks were dried with sufficient air supply to avoid fungal attack. With insufficient air supply the cambium of oak experienced fungal contamination even within two days, possibly caused by a high sugar content of the cambium which can be easily accessed by microbes. Another drying effect was the curling of bark pieces after peeling with the exception of pine mirror bark. Efforts to flatten curled dry bark samples were not successful.

Reasons for the curling resistance of pine mirror bark could be the lack of a thick layer of non-conducting phloem; instead, only thin pieces of dead phloem are loosely attached to the living phloem. This might allow for a more equal air drying on both sides of pine bark (Figure 4.1.2, right).

Larch, spruce and older pine bark possess pronounced layers of tessellated bark flakes. It can be assumed that this side is drying slower compared to the cambium side and hence promotes curling. The deep longitudinal furrows of robinia, (thick/matured) birch and oak bark may further promote curling.

4.2 Densification

4.2.1 Panels

For using bark in high quantities in product and furniture design or in architecture, it would be beneficial if the material could be processed with conventional manufacturing techniques. To provide standardized fabrication techniques like cutting, CNC milling, etc., an equal material thickness of bark is favorable. To achieve a flat, panel like material, bark was pressed. First densification test with one piece of bark resulted in a destroyed bark sample (Figure 4.2.1, A). The cracks caused by the pressing procedure always started from the weakest spots/smallest thickness of the material. In the second experiment, two bark samples were placed with the rhytidome side facing each other (Figure 4.2.1, B). In this test, the bark samples also broke at the weakest spots. In the third test, two bark samples were cross-wise placed and pressed. This densification was successful for all 4 types of bark (Figure 4.2.1, C).

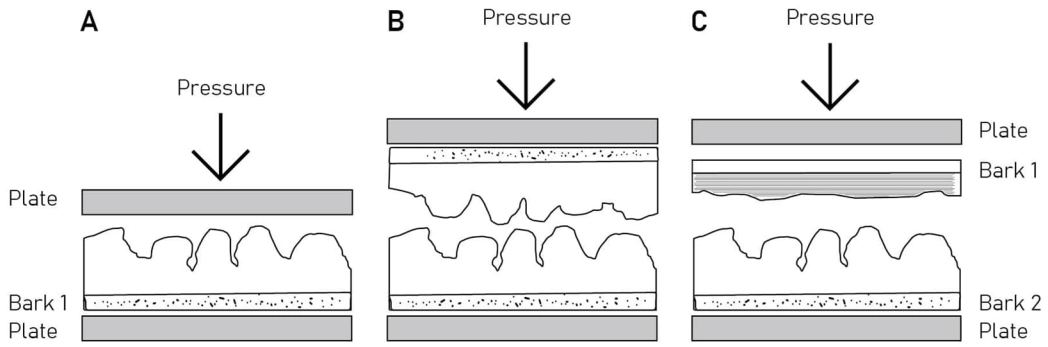


Figure 4.2.1: Different densification test. *One bark sample was pressed (A), two bark sample were placed with the rhytidome side facing each other and both were placed parallel in fiber di- rection to each other (B), two bark samples were placed cross-wise and with the rhytidome side facing each other.*

Production process

The first experiments on laboratory-stored and pre-heated barks (FP1) were successful for all barks of the four tree species birch, oak, pine and spruce. The cross-wise placed and hot-pressed bark pieces (Fig 3.1.3) resulted in flat panels with species-characteristic textures and colors (Fig. 4.2.2). The experiments with bark stored at defined relative humidities previously to the pressing process revealed, that bark stored at a RH > 95% could not be pressed to pan- els since it showed a liquid-like behavior in the hot-press. Furthermore, storage at 95 % RH

promoted growth of microorganisms on the cambial side of the bark. Similar to the observations during drying after harvest, this might be attributed to an availability of easily accessible nutrients for microbes (e.g. sugars).

Figure 4.2.2: *Bark panels made of selected bark samples. Produced panels with in-plane dimensions of 10 x 10 cm^2 showing a random thickness caused by the natural structure of the pressed bark pieces*

The panels can be easily manufactured by milling or cutting. Panels, as shown in Figure 4.2.2 were cut with a circular saw, revealing the interior of the panels. The boundary lines between the phloem (living during harvest) and the rhytidome (dead bark) are clearly visible.

4.2.2 Curved Panels

To evaluate the potential of densification into more complex forms, tests for different 3D curvatures were made. The test to create 3D shaped geometries showed that radii of 40 – 50 mm along the fiber direction can be realized for oak bark pieces without visible damage of the raw material.

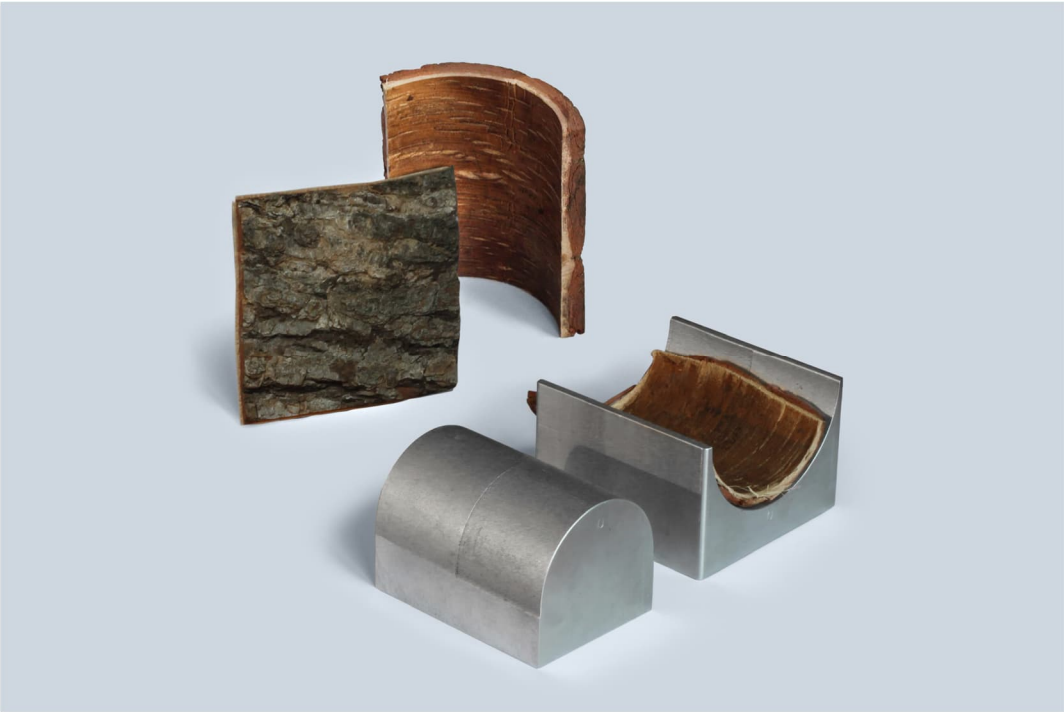


Figure 4. 2.3: *Aluminum mold and produced densified bark curves. Image shows a densified oak bark sample with a radius 50mm and a random thickness caused by the natural structure of the pressed (oak) bark piece.*

Radii between 40 and 20mm can be realized as well, although with a certain fiber relocation. Structures with radii below 20 mm could not be realized. The same experiments were also performed with birch, larch and pine bark. They developed cracks in all directions upon molding It is conceivable that the stability is based on inherent gluing potentials, e.g. resins, tannins, becoming activated by the applied pressure and heat (Kain et al. 2014). The findings show, that only bark with long fibers (e.g. oak) can be deformed three-dimensionally without fiber damage (Figure 4.2.3). This gives the possibility to create 3D shapes which otherwise would need CNC fabrication techniques, creating waste by milling.

4.2.3 Form

The test to create sharp edges in 3D geometries showed that the approach was successful for oak bark, without visible damage of the raw material (Figure 4.2.4, middle and right image). Birch, larch and pine bark developed cracks in all directions upon molding. Birch and pine broke immediately into pieces. Molded larch bark retained an unstable 3D geometry. It can be assumed that the weak form stability is based on inherent gluing potentials, e.g. resins, tannins, becoming activated by the applied pressure and heat (Kain, Güttler et al. 2014). By comparing oak and larch it can be hypothesized that only bark with long fibers (e.g. oak) can be molded.

Figure 4.2.4: *Larch bark and oak bark pressed in 3D form. The structure of larch bark is broken but creates a stable shape (left), oak bark can is molded into 3 dimensional shape without any major visible damages (middle and right image)*

4.2.4 Material Characterization

4.2.4.1 Surface characterization of Panels

Since in-plane surfaces appeared smooth after the pressing process, their roughness was determined 90 ° to the grain direction on the raw panels and gave Rz values of 33.9 µm for larch, 15.1 µm for pine, 13.2 µm for oak and 21.6 µm for birch. These roughnesses are comparable with sanded surfaces of solid wood (Sinn et al. 2004). Considering their natural appearances (Fig. 4.2.2) and the machinability, applications for furniture and paneling without extensive surface treatment can be suggested.

4.2.4.2 3D Microstructure: Raw and Panels

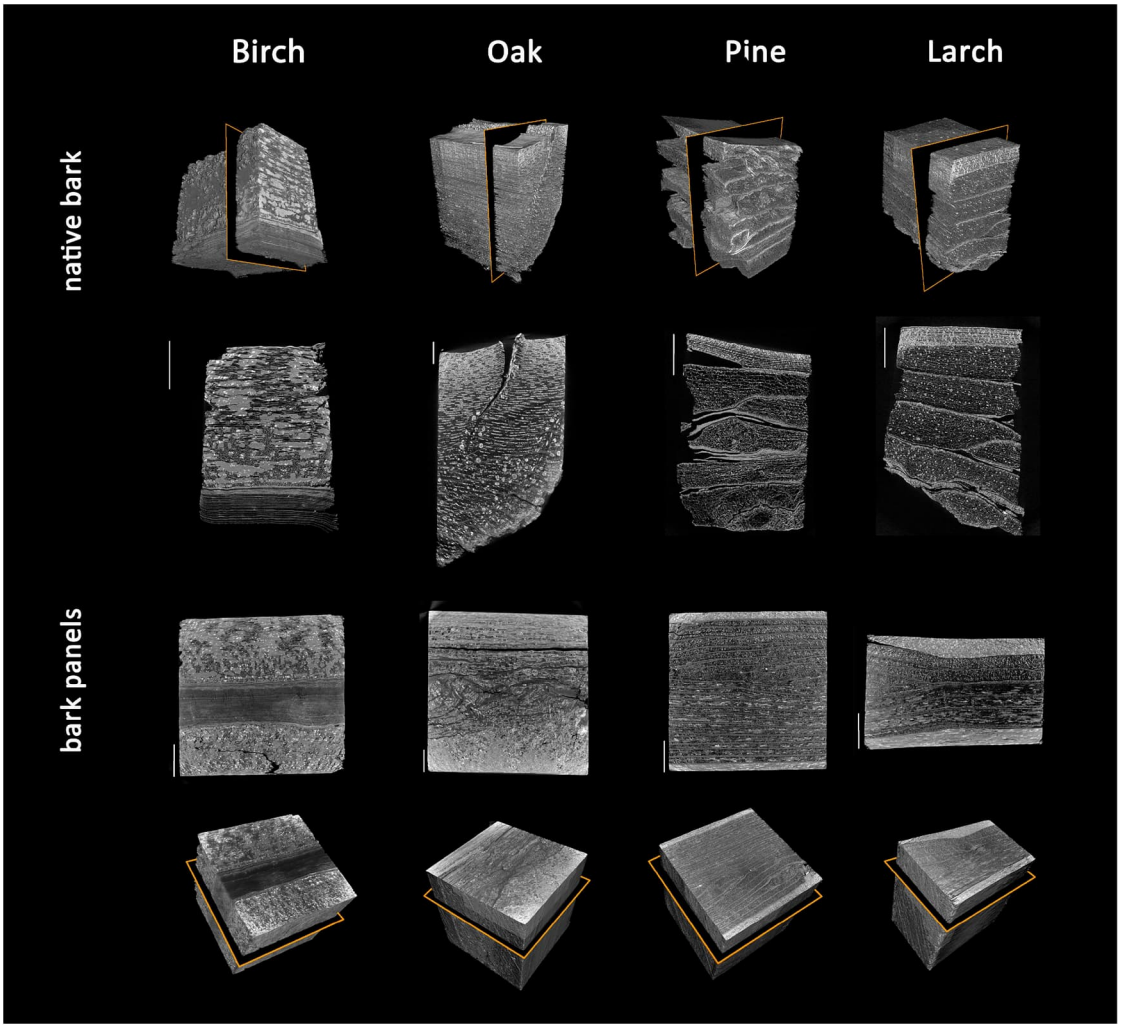


Figure 4.2.5: *µCT scans of native bark pieces and compressed bark. Black slices in rectangular volumes indicate the virtual cutting directions of the slices next to the volumes. Bars 2mm.*

The effects of the pressing process on the bulk material are visualized with µCT data of native bark and panels of all four tree species (Fig. 4.2.5). The flake-like native barks of pine and larch possess numerous cracks, mainly in the periderm or at the interface periderm – the dead part of the phloem. These cracks disappear upon pressing. In addition, the pressing process leads to a strong densification of the sieve cells and dilated parenchyma of the phloem (both young and old), whereas the sclereids of larch phloem and many phellogen cells of larch and pine retain their shapes. In comparison, the microstructures of the native hardwood barks of oak and birch are fundamentally different to the two described softwood species. The fiber-less birch bark is characterized by groups of sclereids in the phloem, the phellem consists of the prominent papery layers. In oak bark numerous fiber bands are present and sclereids are

found in groups. The compression process for panel formation results in a collapse of sieve tubes and parenchyma cells, and the distance between fiber bands decreases considerably. In birch, on the other hand, the irregularly distributed groups of sclereids seem to prevent the thin-walled elements (sieve tubes and parenchyma) from further compression. The sclereid clusters may also explain the random crack formation in the 3D curved structures which seem to appear in the tissue between the sclereids.

4.2.4.3 Density: Raw Bark and Panels

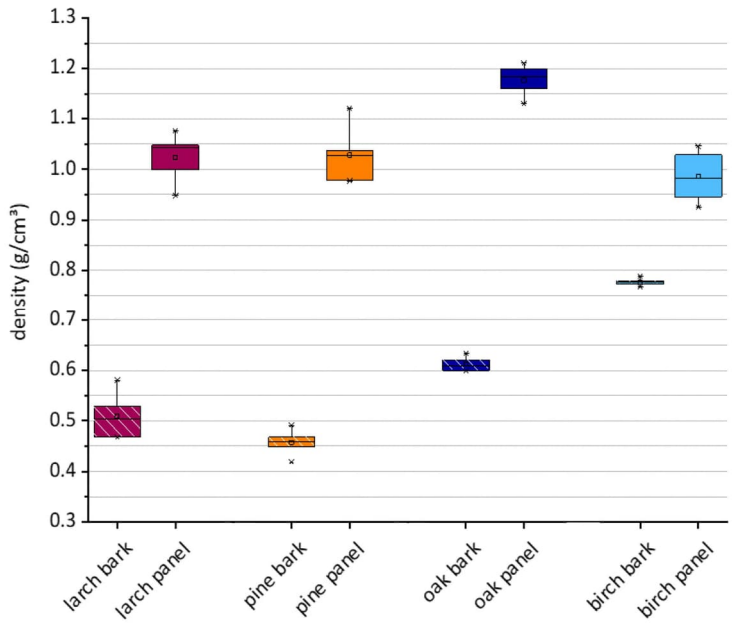


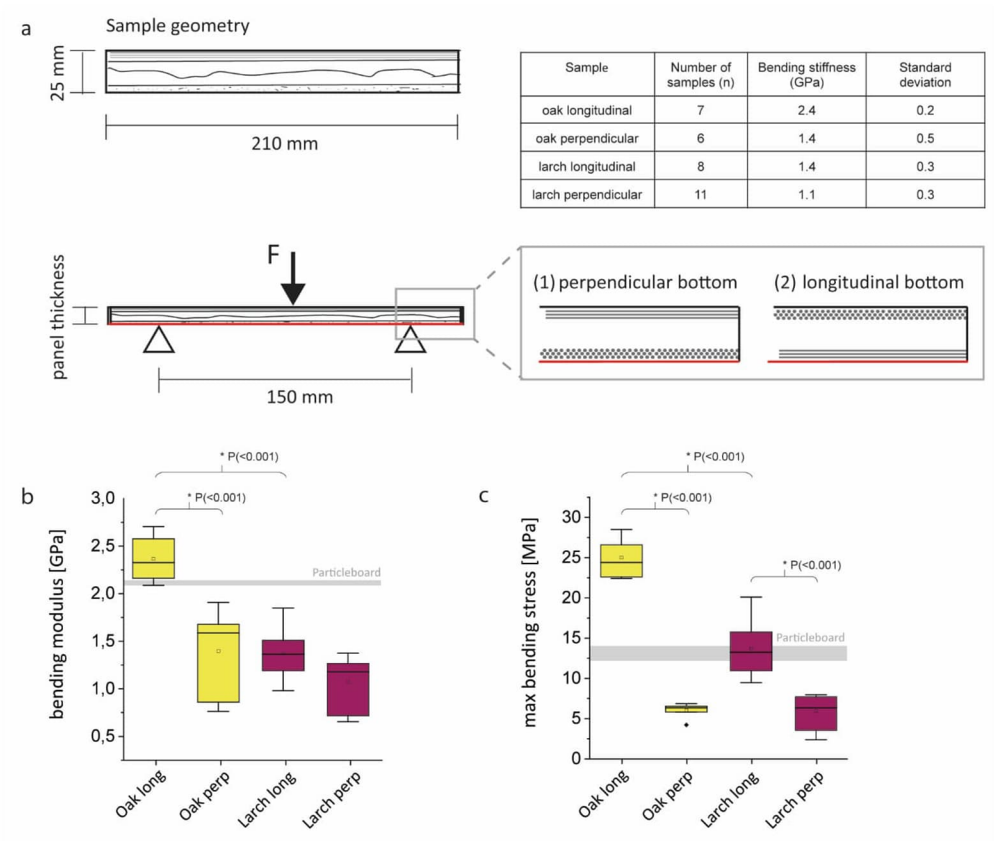
Figure 4.2.6: *µCT scans of native bark pieces and compressed bark for density measurement. (left) Box plot shows densities of native bark pieces and compressed bark (FP1).*

The previously described anatomical differences are also reflected in the densities of the native bark and the panels (Fig. 4.2.6). Due to their structural composition densities of bark samples increase differently with pressing procedure. Native birch bark has a high density between ~ 0.78g/cm^3 and ~ 0.83 g/cm^3, which can be explained by the high amount of dense sclereid groups. The pressing procedure resulted in a slight increase in density (0.99 g/cm^3). A possible explanation is that the maximal compression is reached when sclereid groups get in contact with each other. Oak bark was less dense than birch with a mean density of 0.71 g/cm^3 (thin pieces) and 0.61 g/cm^3 for thick pieces. The compression process led to panels with a density as high as 1.18 +/- 0.03 g/cm^3. The bark of the softwood species had lower

densities with 0.51 g/cm^3 for larch and 0.46 g/cm^3 for pine. However, the same pressing parameters led to panels with densities of 1.03 +/- 0.06 g/cm^3 (pine) and 1.02 +/- 0.05 g/cm^3 (larch) which are similar to the values for birch.

4.2.4.4 3-Point Bending Test: Panels

For an initial evaluation of the mechanical properties of bark panels (FP1), 3-point-bending tests were performed only on larch and oak bark panels. Limited availability of raw material did not allow tests on bark panels of the other tree species. For comparative reasons and deviation of sample geometries from standard tests (Fig 4.2.7), particle board samples with the same geometries were tested and included in the analysis.



Sample	Number of samples (n)	Bending stiffness (GPa)	Standard deviation
oak longitudinal	7	2.4	0.2
oak perpendicular	6	1.4	0.5
larch longitudinal	8	1.4	0.3
larch perpendicular	11	1.1	0.3

Figure 4.2.7: *Mechanical data of three-point-bending experiments. (a) samples with a length of 210 mm and a width of 25 mm were cut out of the panels and tested with a test span of 150 mm. Sample thickness corresponded with panel thickness, the orientation of the fibers at the bottom was either longitudinal (2) or perpendicular (1). (b) bending modulus of the tested samples (c) max bending stress of the specimens. Boxes show 25 % – 75 % percentile, small rectangle in box is the mean and the line the median of the samples, stars correspond to outliers.*

The three-point-bending tests showed a strong effect of the fiber orientation on the modulus (Fig 4.2.7, b) and on the maximum bending stress (Fig 4.2.7, c). This effect is more pronounced for oak and can possibly be attributed to the presence of numerous and massive fiber bundles in the phloem. Strong correlation of fiber orientation on the tension side of bending beams are not surprising and is also known from other fiber-based composites, e.g. plywood and veneer-based wood products (Xue and Hu 2012). The calculated bending moduli (Fig. 4.2.7) are in the same order of magnitude than reported values from wood based panels such as fiber or particle boards (Niemz and Sonderegger 2016) and are considerably lower than those of larch or oak bulk wood which typically exceed 1O GPa and more (Niemz and Sonderegger 2016). The consideration of the different panel densities (oak ~1.2 g/cm^3, larch ~ 1 g/cm^3) leads to less pronounced differences in specific properties between the different types of panels (Gößwald et al. 2021). However, the bending properties of oak panels with longitudinal fiber orientation at the bottom layer are superior compared to all other panels. This first attempt to create pure bark panels and the initial bending experiments suggest promising properties for binderless bark panels. However, optimization steps of the processing conditions (temperature, pressure and water content of raw material) are still needed.

4.2.4.5 Transverse Tensile Test: Panels

To optimize process conditions, effects of moisture content of the raw material was considered for making the of FP2 panels. The evaluation of the connection between the two bark pieces of a panel (FP2) was based on transverse tensile tests (Fig 3.2.1).

Storage conditions raw bark [°C/% RH]	Bark sample	Transverse tensile strength [MPa]	Bark sample [g/cm^3]	Equilibrium moisture content u_{65} [%]
P1 (20 / 15)	Oak	0,24 ± 0,02	1,11 ± 0,03	8,12 ± 0,19
	Larch	0,14 ± 0,03	1,00 ± 0,03	8,71 ± 0,27
P2 (20 / 40)	Oak	0,46 ± 0,10	1,13 ± 0,05	8,95 ± 0,25
	Larch	0,17 ± 0,04	1,04 ± 0,07	10,08 ± 0,40
P 3 (20 / 65)	Oak	0,50 ± 0,10	1,22 ± 0,22	11,02 ± 0,53
	Larch	0,13 ± 0,05	0,95 ± 0,09	12,90 ± 0,30

Table 4.2.1: transverse tensile strength of panels made of bark with different moisture contents

The transverse tensile strengths of panels were obtained. The panels made with oak bark, stored at 20°C and 65% RH before panel production, achieved twice as high ((0.5 ± 0.10) MPa) transverse tensile strengths compared to the panels made of bark stored at 20°C and 15 % RH (Table 4.2.1, Figure 4.2.8).

Statistical analysis showed no significant differences between panels made with oak bark stored at 20°C and 40% RH and 20°C and 65% RH. For the larch panels, there was no clear effect of the moisture content of the raw material on the transverse tensile strength which was generally lower compared to the oak bark panels. A possible explanation for the low transverse tensile strength of larch could be its bark structure: natural larch bark appears flake-like and contains numerous "weak spots" and cracks. In the panels they might serve as starting points for crack initiation, crack growth and fracture. In the future, a particular focus should be on the bonding of the flakes within larch bark pieces as well as on the connection between two pieces. In contrast, the water content of oak bark during pressing affected the bonding between and possibly, but less likely, within the bark pieces: a water-induced reduction in hardness of the raw material, similar to wood (Wang and Wang 1999), might lead to a deeper indentation of the serrated bark pieces into each other and hence to a stronger mechanical connection. The formation of hydrogen bonds might also play a role as well as a faster heat transport due to a higher moisture content (Kollmann 1987).

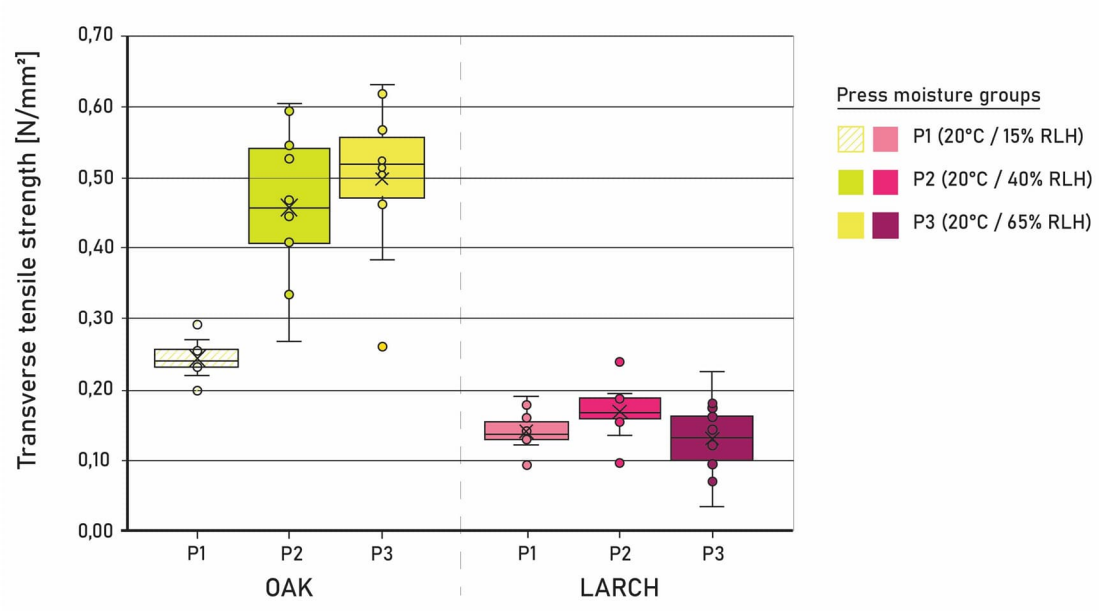


Figure 4.2.8: *Transverse tensile strengths of the bark panels divided into the press moisture groups P1, P2 and P3 of both bark types. Boxplots with median as middle line and arithmetic mean as cross. The whisker length is a maximum of 1.5 times the interquartile range.*

The achieved transverse tensile strengths of oak (P3, Fig. 4.2.8) is comparable to the transverse tensile strengths of an ordinary MDF board (0.55 N/mm^2), a UF resin-bonded particleboard (0.65 N/mm^2) and a UF resin-bonded particleboard with 40% spruce bark (0.58 N/mm^2) (Wang and Wang 1999).

4.2.4.6 Swelling and Shrinkage: Raw and Panels

Swelling of native bark

To evaluate the hygroscopic behavior of bark and bark panels, swelling properties of the native barks were determined in the different anatomical directions on undamaged isolated pieces of material.

The dimensions of the samples stored at 20 °C and 65 % RH were defined as the original length [0]. The most pronounced swelling and shrinkage movement of the bark cubes was measured in transverse direction for larch and oak. Upon drying the samples in radial direction, oak and larch shrank ~ 2 % and ~ 3 % respectively. Increasing the relative humidity to 85% led to swelling of oak and larch ~ 2,3 % and ~ 3,3 %. In tangential direction samples shrank ~ 2% (oak) and ~ 2,2 % (larch) and increased up to ~ 1,4 % (oak) and ~ 0,7 % (larch). In the longitudinal direction, the swelling is much less pronounced (Figure 4.2.9)

The present and previously reported work (Raczkowski, 1979 (Raczkowski 1979)) found the most pronounced swelling in the transverse direction. To summarize, the highest amount of swelling takes place in the radial direction, which is compressed during panel fabrication. Slightly different swelling values of our work compared to Raczkowki (1979) can be explained by different humidity levels (85 % RH here and water immersed in (Raczkowski 1979)). The tangential swelling of Raczkowskis work was more pronounced than the radial swelling. A reason for this could be differences in bark structure since bark in general is known for its high variability (Fengel and Wegener 2011).

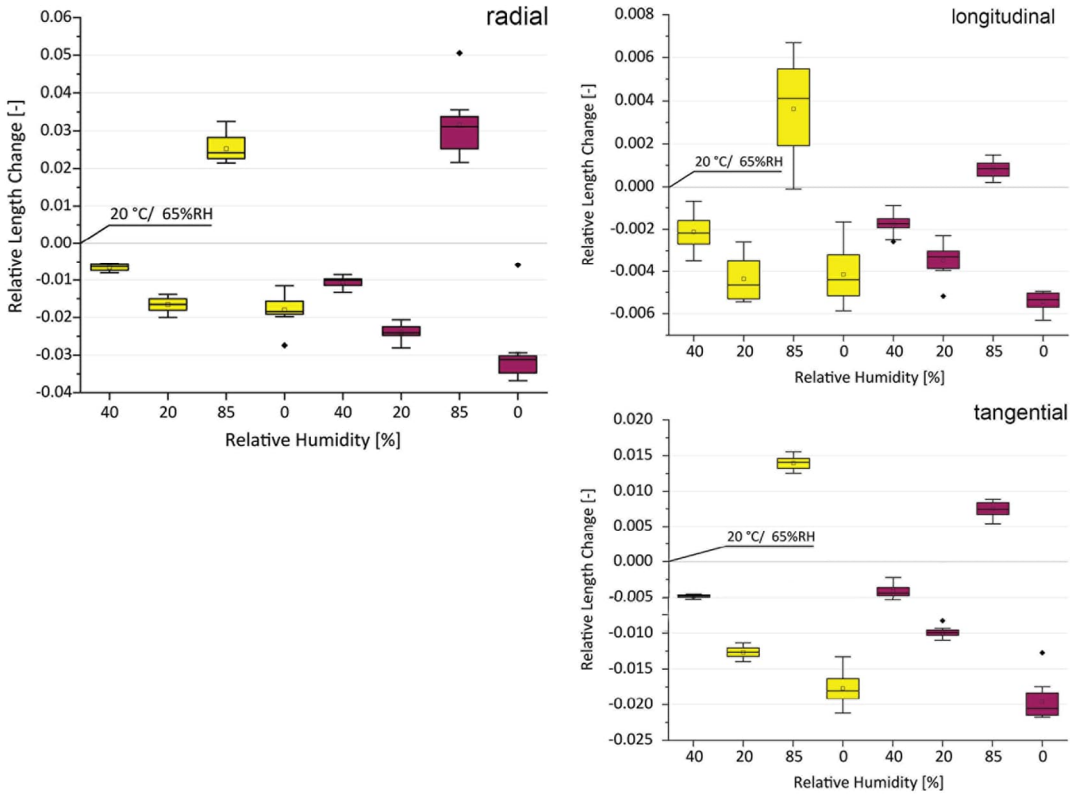


Figure 4. 2.9: *Swelling and shrinkage behavior of native oak (yellow) and native larch (purple) exposed to a sequence of different relative humidities Thickness swelling i s compared to original sample dimension length after storage at 20°C and 65% rh [0].*

Swelling of panels

To examine the impact of the moisture content of the raw bark on the panel characteristics, larch and oak bark pieces were stored at 20 °C and relative humidities (RH) of 15 %, 40 % and 65 % until constant mass was reached. Afterwards all bark samples of the different moisture groups were pressed. The swelling experiments on oak and larch panels reveal clear differences between the samples of press moisture groups 20°C / 15% RH, 20 °C / 40 % RH and 20 °C / 65 % RH. All samples showed pronounced thickness swelling/shrinkage compared to the samples initially stored at20 °C / 65 % RH. Figure 4.2.10 shows their percentage thickness swelling upon changing humidities. The dimensions of the samples stored at 20 °C and 65 % RH were defined as the original length [0]. Upon reducing the RH to 40 % and then 20 % panel thicknesses decreased for all barks pressed with different moisture contents (15% RH, 40% RH, 65% RH). Increasing moisture to 85 % RH during the 4th humidity step led to pronounced thickness swelling. Large differences were observed between panels fabricated with different moisture contents of the raw material. Raw material equilibrated at 65 % RH previously to

hot-pressing (20°, 65% RH) showed a higher dimensional stability compared the containing smaller amounts of water (20°C, 15% RH and 20°C, 40% RH). Interestingly, the swelling of the panels did not exceed swelling of the native bark cubes (Figure 4.2.9). This is a strong indication that there is no spring-back effect due to the formation of internal bonds induced by the heat- and pressure-exposure. The amount of water in the raw material seems to play an important role. The exact mechanism, however, remains unknown and further studies are required.

Surprisingly dry storage at 80 °C did not lead to shrinkage in all panels. It is possible that drying from 85 % RH to ~0 % RH causes drying-induced cracks within the boards which lead to an increase of gaps between panel pieces.

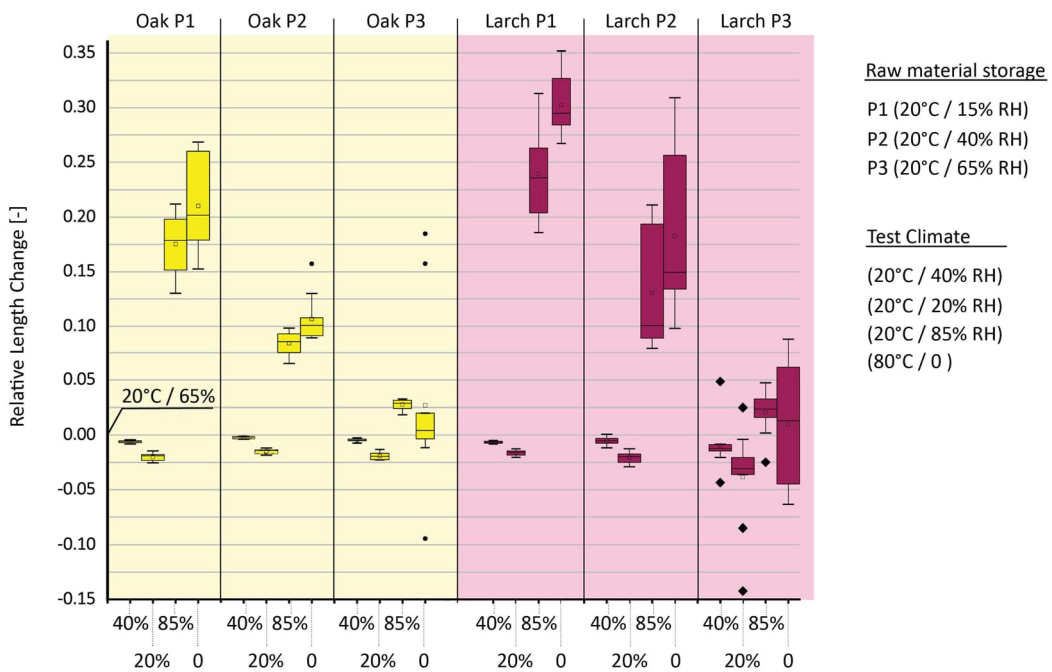


Figure 4.2.10: *Swelling and shrinkage behavior of oak (yellow) and larch (pink) panels in the climate test.*

4.3 Flexibilization of Bark

The effect of glycerol treatment on barks of four different tree species and how their mechanical properties change due to the water- glycerol treatment are presented in this chapter. Wet, dry and glycerol treated bark was qualitatively and quantitatively characterized, the latter by

performing tensile tests to determine the mechanical properties. Since the applications described in Chapter 5.1. suggest that flexibilized bark shows characteristics similar to the closely related material wood and to leather, samples of both materials were tested to underpin these haptic impressions with quantitative mechanical values.

4.3.1 Material Processing

4.3.1.1 Flexibilization: Process with Different Barks

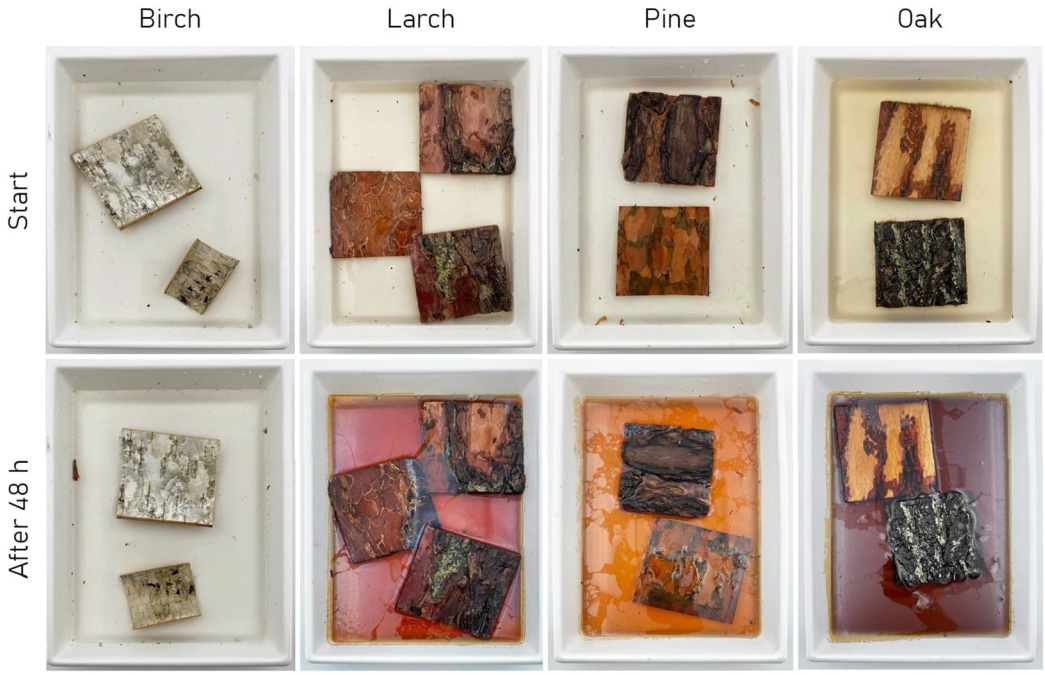


Figure 4.3.1: *Bark samples in glycerol -water solution. After 48 h, a clear coloration of the solution is visible for most barks. from left to right: birch shows no optical changes, larch shows a reddish-brown dark discoloration as well as a light film on the surface of the solution, pine has a strongly orange solution with film, oak has a dark brown solution with a film.*

Glycerol -water treatments of tree barks were performed for the first time within the framework of this thesis (to the best of our knowledge). Since both glycerol and water are solvents the solutions were visually investigated after infiltrating barks of larch, pine, birch and oak bark for 48 hours (Figure 4.3.1). The solution of oak showed a dark red brown color and can

be considered the most intense in terms of color and saturation. The larch solution was reddish brown and the coloration less intense and more red-tinted compared to the oak. Solution of pine led to a clear orange and birch to no visible coloration of the solution.

The glycerol -water treatment resulted for pine bark in a flexible material, (Fig. 4.3.2, 2) which can be bent by hand in both fiber directions (Fig. 4.3.2). Both oak and birch samples ((Fig. 4.3.2, 1) and (Fig. 4.3.2, 3)) broke in longitudinal direction. Larch (Fig. 4.3.2, 4) showed an interesting behavior. While all dead bark (rhytidome) could be broken easily, the inner bark showed flexibility, similar to pine samples.

Since pine bark showed the most promising results regarding its flexibilization potential it was selected for a more detailed investigation on the influence of glycerol concentration on treated bark. Tests with glycerol water solutions with mixing ratios of 1 : 0, 1 : 1, 1 : 2, 1 : 4, 1 : 6, 1 : 8 for 48 h showed that a ratio of 4 parts water and 1 part glycerol led to a flexible bark which still was dry to the touch. Bark immersed in 100% glycerol stayed stiff and brittle and the surface became sticky. Bark treated with 1:1 and 1:2 showed increased flexibility but a sticky surface, which makes it difficult to use for applications. Bark immersed in solutions with a ratio 1:6 and 1:8 showed a smooth and dry surface after drying but had reduced were less possible to bend without damages compared to 1:4.

The tests showed that the degree of flexibilization can be influenced by the mixing ratio of the water- glycerol mixture. Based on the qualitative evaluation of haptics and flexibility the best result was achieved with the 1:4 solution.

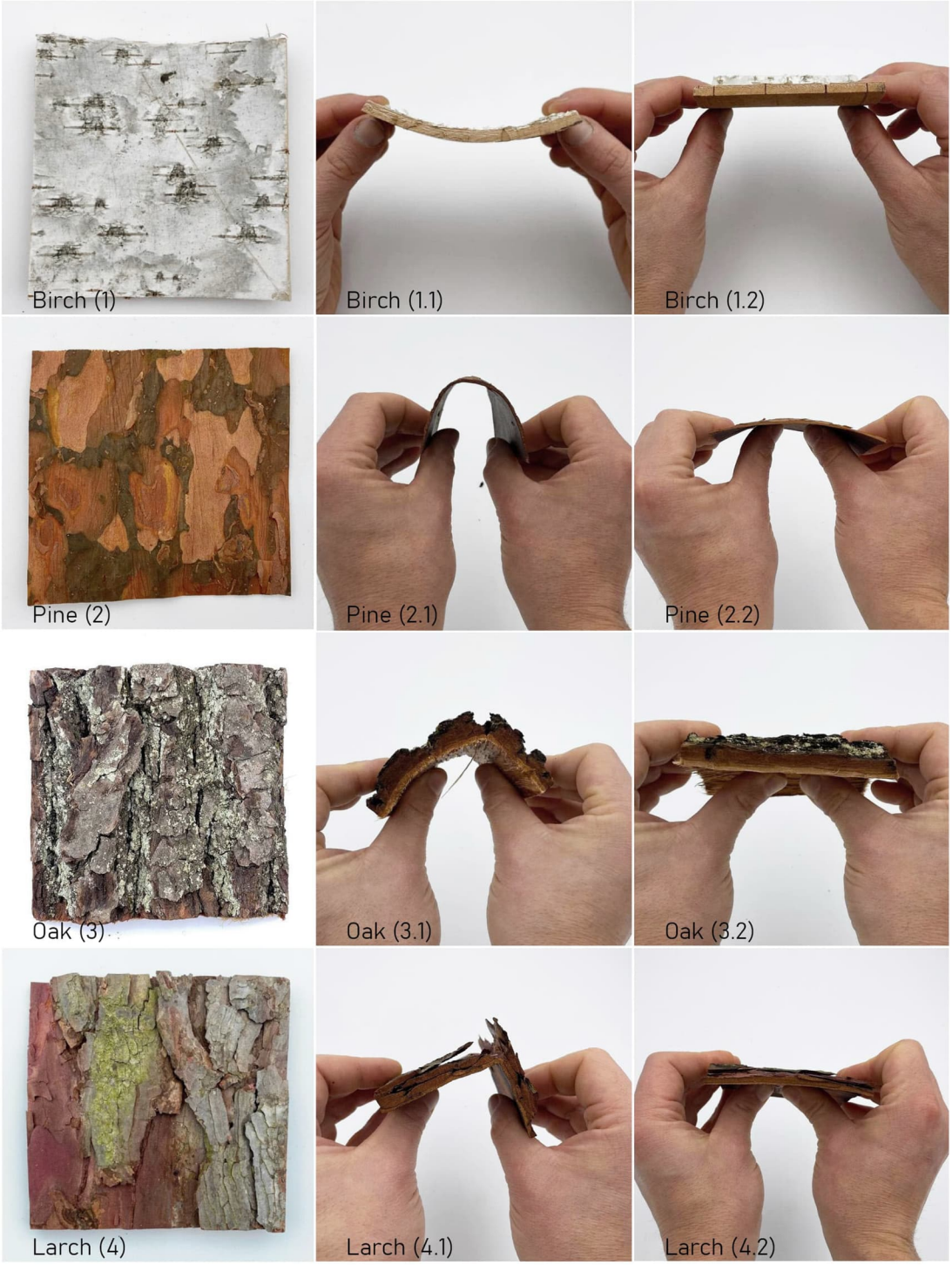


Figure 4.3.2: *Flexibility of bark after drying. (1) birch bark is not flexibilized, (2) pine bark is flexible in both directions, (3) oak bark is stiff longitudinal and brittle in tangential direction, (4) larch is flexible in tangential direction and not longitudinal*

The potential and limits of preserved flexibility for different barks

Tree bark is flexible directly after peeling but loses the flexibility upon drying. The aim of the present work was to preserve flexibility and to keep the texture and color of the fresh bark by immersing bark in a glycerol-water solution, a method known from leaf preservation (Babu et al. 2018). The glycerol replaces a certain amount of water within the plant material and hence keeps it flexible (Babu et al. 2018). Reported experiences show that most leaves – also tree leaves (e.g. oak, magnolia and eucalyptus) can be preserved using this method (Babu et al. 2018), (Smith and Laschkewitsch 1992).

The presented experiments show, that it is possible to flexibilize pine bark and the phloem of larch bark, while oak and birch could not be flexibilized. The differences in flexibility between the barks can be related to structural heterogeneity and in particular to the different proportions of conductive phloem. The successful flexibilization of pine is presumably based on the connection between the periderm and the outer rhytidom and the high content/share of conductive phloem and cambium remnants, while only thin dead bark pieces (flakes) are present on mirror pine bark. Due to their loose connection with the phloem, bark flakes have the tendency of falling off upon bending the glycerol treated material (Figure 4.3.3, B).

Compared to pine mirror bark, larch bark possesses tessellated pieces of outer rhytidom, stacked on top of each other and interlocked with each other (Figure 4.3.3, A). On more mature pine tree stems, the flakes grow stronger and their connection within the bark becomes more similar to larch (Figure 4.3.3, A). This transition from mirror pine bark to more pronounced matured pine bark remain unclear and require further research.

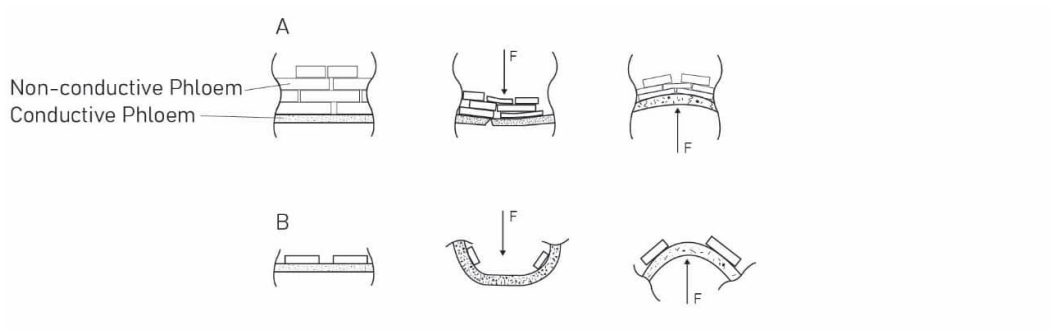


Figure 4.3.3: I*nfluence of non-conductive phloem on the flexibility of bark samples. (A)Pro- nounced "tessellated" bark part causing cracks inside flexibilized phloem (B) Phloem without hard and unflexible bark pieces is able to move in different directions*

The resistance of birch and oak against flexibilization is possibly caused by thick rhytidomes with numerous sclereids (Figure 4.2.5, (Fengel and Wegener 2011)) and fiber bundles (Figure 4.2.5) composed of lignified, thick walled cells – similar to cells in wood. The presence of fibers and/or sclereids depends on species and phloem age. Very pronounced fiber and sclereid bundles/clusters are mostly described for angiosperms (Einspahr, Harder, and Parham 1978) (Evert 2006). Examined gymnosperms also contain fibers and sclereids, but compared to the oak and birch bark to a lower extent (Evert 2006). The dissolved substances in the glycerol-water-solution after bark treatments were not analysed. Literature data suggest, that the high content of water-soluble tannins in oak, larch and pine caused the coloring of the solution (Ashok and Upadhyaya 2012) .

4.3.1.2 Tensile Test: wet, dry and flexible Pine Bark

Dry, wet and glycerol *treated pine bark strips*

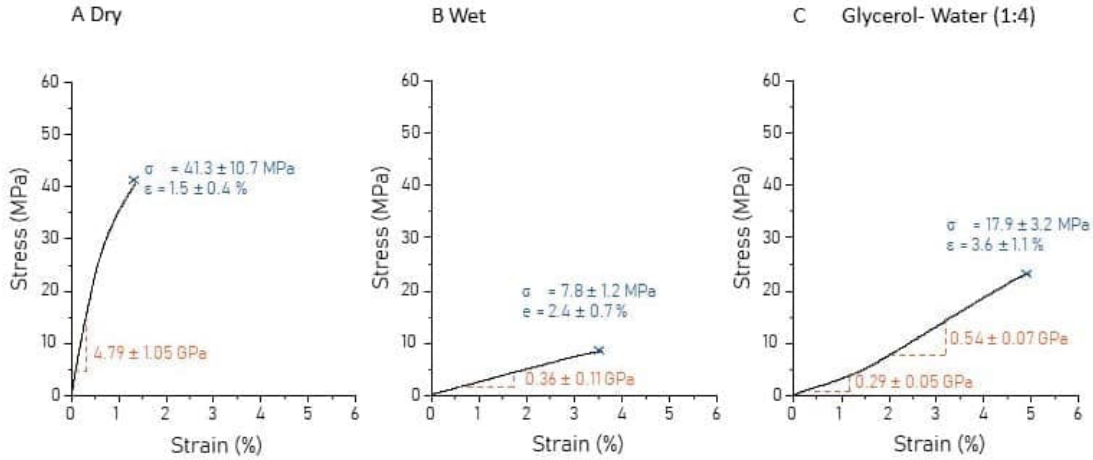


Figure 4.3.4: *Representative stress–strain curves of dry (laboratory conditions), wet and glycerol-treated mirror bark of pine. The values show arithmetic mean values and standard deviations of 20 (a), 18 (b) and 19 (c) successfully tested samples. Figure reproduced with permission from Philosophical Transactions of the Royal Society*[17]

[17] Wenig, C., Dunlop, J. W., Hehemeyer-Cürten, J., Reppe, F. J., Horbelt, N., Krauthausen, K., ... & Eder, M. (2021). Advanced materials design based on waste wood and bark. *Philosophical Transactions of the Royal Society A*, *379*(2206), 20200345.

The effect of glycerol water solutions on mechanical properties of bark were quantified by performing tensile tests on dry, wet, and glycerol treated bark strips (4 parts of water and 1 part of glycerol). The experiments confirmed the observation, that dry bark is stiffer (4.7 GPa) and more brittle than wet bark (360 MPa) (Figure 4.3.4). Glycerol treated bark samples showed improved flexibility and stress at failure compared to wet bark. Interestingly, the shape of the stress–strain curve changed to a bi-phasic behavior with an initial stiffness of 290 MPa increasing to 540 MPa (Figure 4.3.4).

Effects of varying concentrations of Glycerol in solutions

The effect of the concentration of the glycerol solution on the mechanical properties of flexibilized pine bark was quantified by tensile tests on different glycerol -water treated pine samples. The tensile tests showed, that bark immersed in water- glycerol solutions with low glycerol concentrations (1:6 and 1:8) possessed a higher stiffness compared to those immersed in solutions containing higher proportions of glycerol (Figure 4.3.5,C). Hence, bark samples with a high amount of glycerol showed improved flexibility. Remarkable is the increase in strain to failure from 1.61 % +/- 0.45 for bark treated with a 1:6 glycerol -water-mixture to 3.59% +/- 1.11 for bark immersed in a 1:4 solution (Figure 4.3.5). The highest strains of 1:2 glycerol treated bark (4.65% +/- 0.71) are almost twice as high as the maximum strain of wet bark (Figure 4.3.5, B). Remarkable is the change of the stress and strain behavior for 1:1 treated bark. From 1:8 up to 1:2 an increase of strain with a decrease of stress is measured. With the 1:1 concentration of the glycerol solution the stiffness is again increasing and the strain is lower (2.8 % +/- 0.76) compared to the tensile behavior of 1:2 glycerol treated bark. While 1:8 and 1:6 treated bark samples show a stress-strain curve similar to the wet samples, the shapes of the stress–strain curves of 1:4 (Figure 4.3.4), 1:2 and 1:1 treated bark changed to a bi-phasic behavior.

Tensile tests on dry, wet, and glycerol treated spruce wood samples

Due to the high variability and limited knowledge of bark, and to better understand the effect of glycerol water solutions on lignified tissues, tensile tests were also performed on dry, wet and glycerol treated spruce adult and compression wood (Fig. 4.3.5), known for their low and

high cellulose microfibril angles (adult wood ~ 10 °, compression wood ~ 35-50°) (Niemz and Sonderegger 2016). The experiments confirmed textbook knowledge, that dry wood is stiffer (spruce adult wood 9.66 GPa +/- 1 GPa , spruce branch compression wood 3.53 +/- 0.29) compared to wet wood (spruce adult wood 5.37 GPa +/- 0.39 GPa , spruce branch compression wood 1.64 +/- 0.13 GPa). The decrease in stiffness of adult spruce with increasing water content is much higher than reported for bulk wood samples (Niemz and Sonderegger 2016). A reason could be the small sample thickness (150 µm), possibly allowing the wood fibers to slip past each other because of the low shear strength of the middle lamellae. It is also described for cellular materials that sample thickness and cell size ratio can affect mechanical properties (Tekoglu and Onck 2005). Compared to bark, both types of wood are stiffer and have a higher tensile strength (Fig. 4.3.5). Glycerol treated adult spruce wood samples (7.44 GPa +/- 0.46 GPa) showed 3 orders of magnitude higher stiffness compared to the initial stiffness of 1:4 glycerol treated bark (0.29 GPa +/- 0.54 GPa). A change in strain could not be seen for measured adult spruce wood samples treated with glycerol. In contrast, branch compression wood treated with glycerol showed a considerable increase in strain compared to the dry state, but less than the wet samples. Interestingly, strain measurements of 1:4 mixing ratio of glycerol treated pine bark (3.59% +/- 1.11 %) and with 1:2 mixing ratio of glycerol treated pine bark (4.65% +/- 0.71%) showed almost similar strain values as spruce compression wood (1:4, 4.69%, +/- 1%).

Tensile tests on leather samples compared with 1:4 glycerol treated pine bark

To compare mechanical properties of bark and leather (pit tanned and drum tanned), tensile tests with leather were made (Fig. 4.3.6). The experiments showed, that 1:4 glycerol treated bark samples had a lower tensile strength (0.39 MPa +/- 0.05 MPa) compared to all leather samples (drum tanned leather 16.8 MPa +/- 1.95MPa, pit tanned leather 18.93 MPa +/- 2.41 MPa) (Figure 4.3.6, A). The strain of leather is ~ 6 – 7 times higher compared to glycerol treated bark (drum tanned leather 29.4%, +/- 2.6%, pit tanned leather 27.6%, +/- 4.3%, 1:4 glycerol treated bark 3.59% +/- 1.11%) (Figure 4.3.6, B). The shape of the stress–strain curve of all leather samples had also a bi-phasic behavior (Fratzl 2008) comparable with stress–strain curves of 1:4, 1:2 and 1:1 glycerol treated bark samples.

Bark in dry, different glycerine concentrations (glyc), wet

Adult wood (ad), Branch compression wood (bcw), 1:4 Glycerine treated pine bark (glyc 1:4)

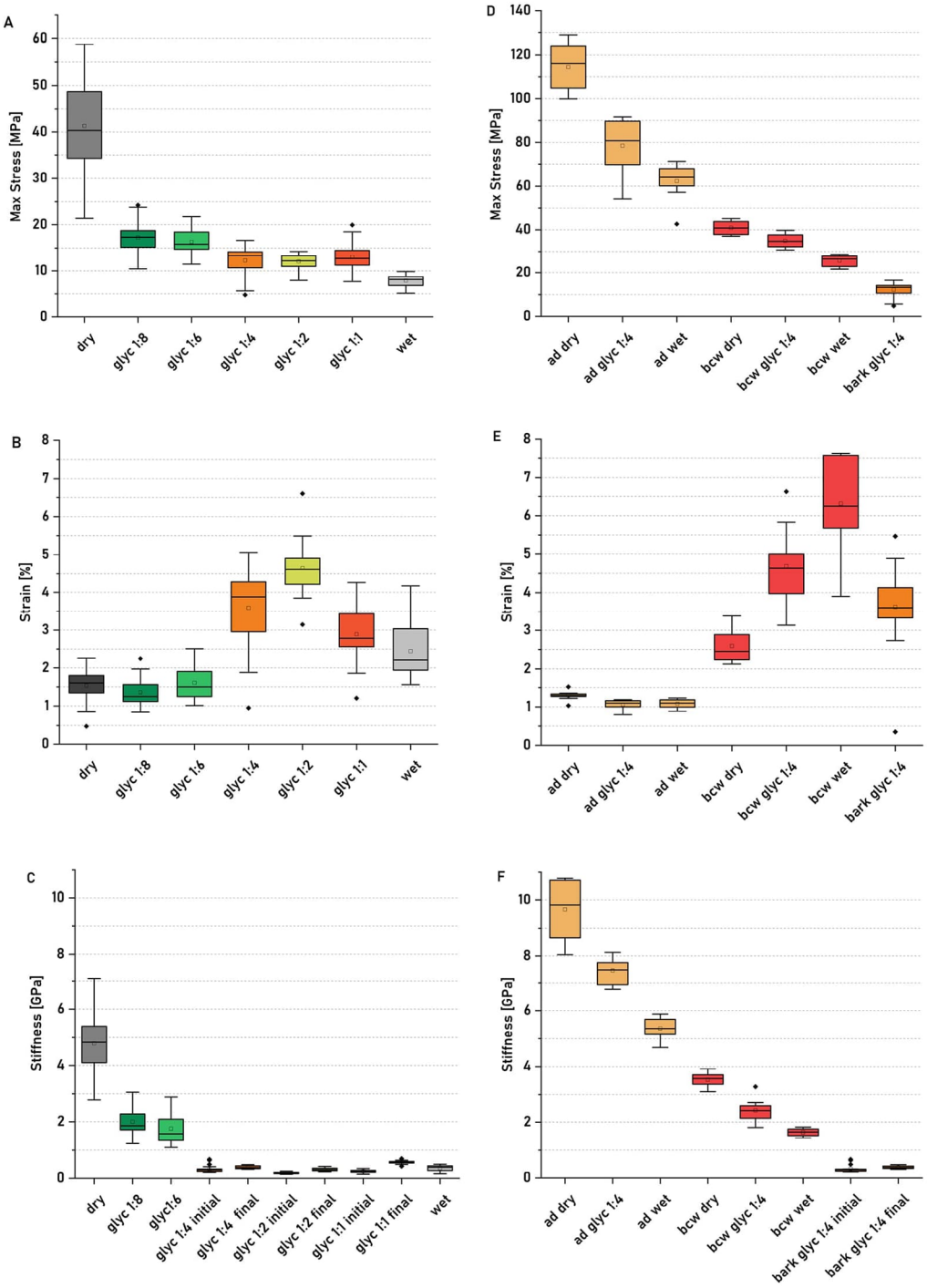


Figure 4.3. 5: *Effects of different glycerol treatments on pine bark and wood. Left column, different glycerol solution treatments of pine bark (orange): maximum tensile stress (A), strain (B) and stiffness (C) were measured; right column shows adult spruce wood (bright brown), compression wood spruce (red) and 1:4 glycerol treated pine bark (orange). Wood samples were measured dry, 1:4 glycerol treated and wet. Box plots show maximum tensile stress (D), strain (E) and stiffness (F). Boxes show 25 % – 75 % percentile, small rectangle in box is the mean and the line the median of the samples, stars correspond to outliers.*

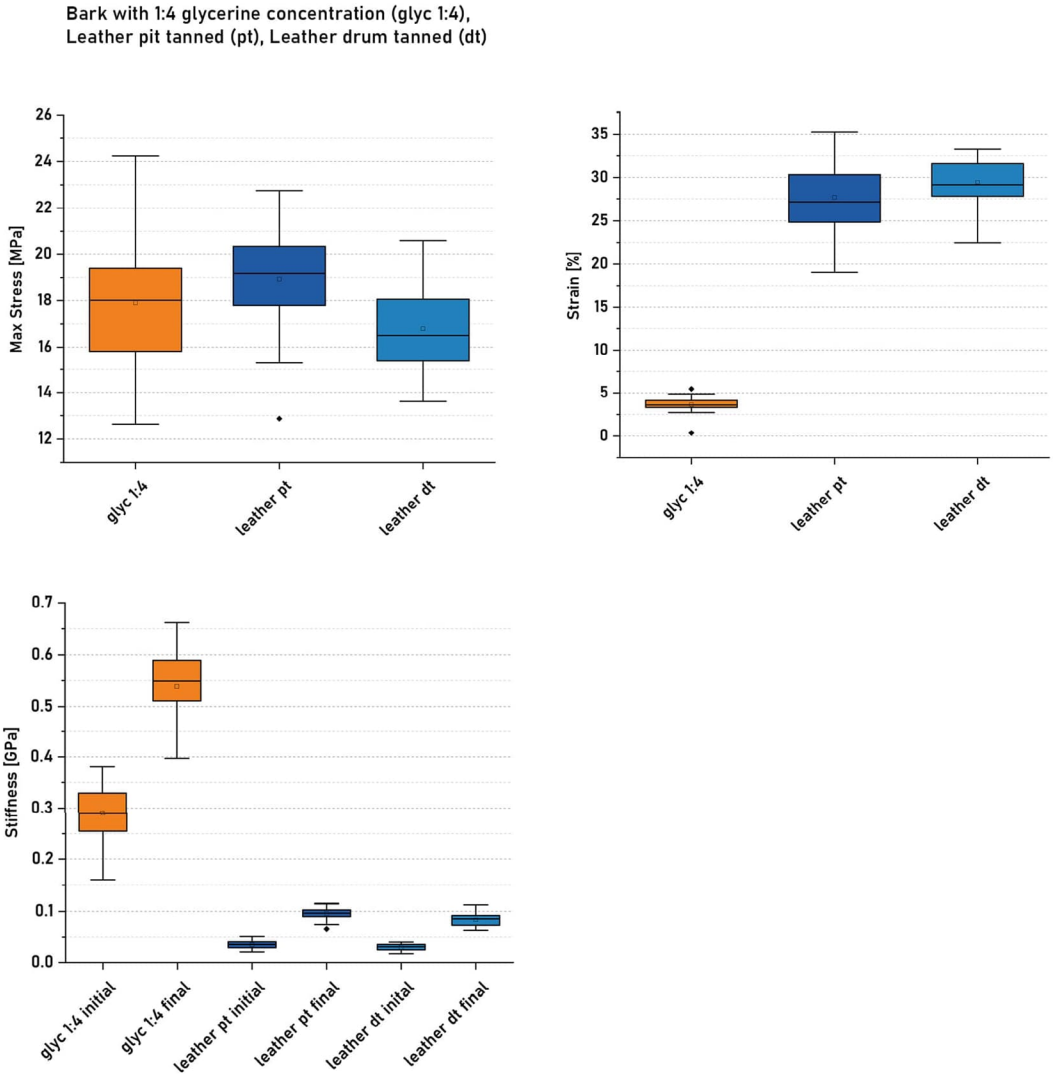


Figure 4. 3. 6 *Comparison of tensile properties of flexibilized pine bark and leather. Maximum tensile stress (A), strain (B) and stiffness (C) pit tanned leather (dark blue), drum tanned leather (blue) and 1:4 glycerol treated pine bark (orange). Boxes show 25 % – 75 % percentile, small rectangle in box is the mean and the line the median of the samples, stars correspond to outliers.*

Comparison of flexibility and tensile properties of bark, wood and leather

Flexibility is an important material property for applications for motion, such as textiles for clothing or accessories, like tents and bags. The achievement to create a pine bark which remains flexible under ambient conditions (Chapter 4.3.1.1, Figure 4.3.2) was applied to create the objects described in Chapter 5. First experiences were gained by designing and making the plain bark jacket (Chapter 5.1). A challenge for many textile applications are local stresses, e.g. when elbows are bent. Here, a favorable ratio between stress and strain is crucial to provide both longevity and convenience. A high strain, which does not fail under local stress (i.e. a maximum stress), is needed. Furthermore, the strain levels should not exceed the elastic range to prevent plastic material deformation like bulging.

The results show that tensile properties of pine bark depend on the concentration of glycerol in the solution (Figure 4.3.5). Higher glycerol contents lead to more flexibility. A similar relation was observed in the past by Dubois and Joyce for different plants used for decorative purposes (jarrah (*Eucalyptus marginata* Smith), bookleaf (*Daviesia cordata* Smith), bamboo (*Leucopogon verticilliatus* R.Br.), red kangaroo paw (*Anigozanthus rufus Labill.*). A higher glycerol concentration in water- glycerol solutions resulted in a higher infiltration of glycerol into the sample (Dubois and Joyce 1992).

Overall, with a higher amount of glycerol the strain increases while the stiffness decreases. In case of 1:4, 1:2 and 1:1 glycerol treated pine bark the strain was measured in a higher range compared to wet pine bark (Figure 4.3.5). It can be assumed that the reason for the changing mechanical properties by infiltration with different glycerol solutions is related to the moisture kept in the conductive phloem and promoted by a higher micro fibril angle. The reason for the increase in stiffness of the 1:1 treated bark remains unclear.

Tensile tests were also performed with wood to obtain more information about the influence of glycerol water treatment on mechanical properties of lignified tissues (of pine bark).

Adult and compression wood were selected, since they show pronounced differences in structure and chemical compositions (Fengel and Wegener 2011). Adult normal wood from the tree stem has a small microfibril angle (0-20°), while compression wood is characterized by a significantly larger microfibril angle (30° – 60°) (Niemz and Sonderegger 2016) which results in lower stiffness and larger strains and hence flexibility.

A remarkable difference between bark and wood is that the glycerol treatment of bark leads to larger strains compared to water immersion. Hence, the finding suggests that this effect

does not relate to changes in lignified cell walls similar to wood cells. However, the observed differences in strains upon (30° – 60°) treatment and hydration between adult and compression wood suggest a potentially high cellulose microfibril angle in mirror pine bark.

While pine bark can be flexiblized, wood showed no increase in flexibility after an infiltration with solution. This leads to the assumption, that not only the fibrous structures are the reason for an increase of flexibility. The reason for the flexibility of pine bark could be the absence of fibers and isolated sclereids which is characteristic for *Pinus sylvestris (Schweingruber, Steiger, and Börner 2019)*. Both sclereids and fibers are composed of lignified, thick walled cells, and affect the mechanical characteristic by stiffening the material. Large proportions of trees also consist of dead cells with thick lignified secondary cell walls (Salmén 2004a), (Moon et al. 2011). With the cellulose microfibril angles stiffness and strength can be modulated (Reiterer et al. 1999). While cellulose orientation in wood is well studied, only little is known about the cellulose orientation of bark cells and its effect on the mechanics of the material itself. To clarify the role of the cellulose orientation on the flexible bark, more studies are needed.

However, the mechanical values are not the only relevant quality for material use. For instance, bark treated with a 1:2 glycerol solution gets a shiny and greasy surface, with negative effects on decorative appearance and usability for further processing. This effect was frequently described for glycerol preservation (Paparozzi and McCALLISTER 1988). Consequently, the 1:2 ratio is disadvantageous, since haptics is essential for aesthetics as well as for the simplicity of processing (i.e. the material can be easily cut and touched without being sticky). This makes the 1:4 mixing ratio the best compromise. In the following only bark treated with this mixture will be discussed.

Flexibilized pine bark feels softer than dry wood or dry bark. As described in chapter 5.1.3 it is possible to cut and to sew flexibilized pine bark. Both the haptic feeling and the processing possibilities of flexibilized bark were comparable with the feeling of leather and its processability.

Tensile tests with two different sets of leather samples showed that flexible pine bark is more than five times stiffer when stressed along the grain – even though the qualitative manual comparison of leather and bark samples showed similar haptic feel and flexibility. As a consequence, this high stiffness in the longitudinal direction restricted the flexibility of pine bark and became obvious in the first design object, the bark jacket (Chapter 5.1), where it limited the free movement of the person wearing it.

Even though bark feels as soft as leather and also the color and structure is similar, this first jacket was too stiff that the model could not lift his arms without causing damage to the material (Chapter 5.1.4). Additionally, the flexibilized bark was fragile perpendicular to the longitudinal direction.

The higher mechanical properties of leather are related to its structure. Collagen is the main structural protein in skin (Thomson 2006). Collagen molecules are extremely long in relation to their cross-section. During their formation they arrange into fibrils and bundles of fibrils. These fibril bundles interweave in a three-dimensional manner through the skin (Haines 2006a). This natural fibrous weave is preserved in the final leather (Haines 2006b) and it is this fibrous structure that gives its good physical properties including flexibility, a relatively high tensile strength and resistance to tearing (Kite and Thomson 2006).

The comparison between the mechanical properties of both wood and leather with bark reveal the unique character of bark. On the one hand, flexible pine bark is as flexible as leather, but leather has a higher maximum strain. On the other hand, flexibilized pine bark is in its strain comparable to wet or glycerol treated branch compression wood but shows a much lower stiffness. Consequently, mechanical data like maximum stress, stiffness and strain are needed to meet challenges in the designing process. Failures in the first prototype of the jacket – for example the arm lifting problem – could have been avoided by a previous analysis of flexibilized pine bark. It emphasizes the relevance of hands on experiments in the closest possible cooperation between design and science for the development of innovative materials and manufacturing processes.

4.4 Ceramics

The majority of tree bark is burned in the wood processing industry. This creates large amounts of bark ash which is rarely used. Ash in general is an important additive in ceramics and glass crafts (Wolf 2012). To explore possibilities of using bark ashes for ceramics, barks of different tree species were burned. Afterwards, glazes were prepared with different glazing recipes to investigate the influence of bark on the glazing process. The glazes were applied on self-made porcelain forms.

4.4.1 Material Processing

The use of ashes always played an important role in glass and glaze production. Records date the use of wood or grass ash to the early beginnings of glass and ceramic craft (Wolf 2012). Depending on its chemical composition, ashes of different plants provide a variety of colors and surface qualities to the glazing process (Bezborodov 1975). Tree bark is known for its variability in chemical compositions within and between species (Fengel and Wegener 2011). Its ash consist of different mineral parts. Different types of tree bark ashes were (each) mixed with a recipe for a transparent porcelain glaze. Hand-made porcelain bowls were glazed and then fired. The results are analyzed visually.

4.4.1.1 Ash Production

The bark samples incinerated at 600°C in the first firing at the University of Arts in Bremen, were not completely burned, even after several hours. Except for the Robinia ash, all other samples contained pieces of charcoal (Figure 4.4.1). Smoke development during combustion made further burning not possible.

Figure 4.4.1: *Results of combustion test with bark at the University of Arts in Bremen. Left robinia bark ash, right oak bark ash with charcoal*

In order to better control the firing process and to enable a full incineration, the next combustion was performed with a ceramic specialist ("Müritzkeramik" in Lärz, Germany). In this firing, strong smoke development at 600 – 700°C was also observed. In the second consecutive firing there was no further smoke development.

The optical evaluation of the bark ashes first revealed the difference in quantity and the varying degree of complete combustion. Robinia remained (visibly) as white ash, hinting towards complete incineration (Fig. 4.4.2). The incinerated other barks all contained charcoal pieces. Particularly in the case of pine, a high proportion of charcoal remained even after the second combustion step.

Figure 4.4.2: *All bark samples after the second combustion test at "Müritzkeramik"*

Bark structure and manufacturing related challenges for a complete combustion of bark

The incomplete combustion of all barks except for robinia could be caused by an uncontrolled and rapid temperature rise to 600°C (Lehnhäuser 1985) . The expected better control of the combustion process at "Müritzkeramik", was also accompanied by smoke development and incomplete initial combustion. The strong smoke formation, may have been caused by a too high moisture content in the bark even though the bark was stored under lab conditions before burning, suggesting a low moisture content. For future burning experiments, an additional drying step before burning should be considered and moisture contents should be determined.

For entire combustion, temperature and time were strongly increased in a second firing step. After this additional firing, ash, pieces of coal and sometimes slag were found in the individual capsules/vessels. The reason for the incomplete combustion at high temperatures is still unclear.

One factor in all 3 kiln-firings could be a lack of oxygen supply. While in the first firing no active control of the ventilation was possible, in the second and third firings the design of the vessels could have complicated oxygen supply. The optimum burning temperature and burning length was possibly not reached. Furthermore, unfavorable combustion curves and progressions (too fast high temperatures or too fast cooling) may prevented complete burning.

The combustion problems of this experiment, on the other hand, prove a fundamental function of tree bark. Bark as heat protection for the cambium is increasingly being considered in research (Bär and Mayr 2020), (Kupferschmid 2001). How well bark provides heat protection and, conversely, selective burning, varies widely among bark species. Decisive factors are thermal conductivity, heat transfer, moisture content, bark thickness, density and possibly chemical composition (Pausas 2015; Spalt and Reifsnyder 1962). Therefore, targeted incineration temperatures are difficult to determine.

4.4.2 Material Characterization

4.4.2.1 Creating the Bowls

4.4.3 : *Production of test bowls for glazing experiments. Left picture shows bowls just removed from the mold and bowls still drying in the mold. Right picture shows an already dried but still unfired bowl*

The individual shapes varied between 3mm – 5mm in their material thickness. All bowls were stable after bisque firing and smooth on the inside (which was later used for glazing).

Discussion about form

The shapes of the bowls were designed to evaluate different properties of the glazes. Based on the shape of the bowls, the adhesion of glaze to walls can be assessed. The bowls demonstrate the distinct outcomes of different kinds of bark and the varying density of the glaze layers depending on the thickness of the glaze. If adhesion is good, there will still be a thicker glaze layer in the center of the shell than on the walls. Thick layers of glaze can significantly change the coloration (Matthes 1985).

4.4.2.2 Inductively Coupled Plasma (ICP)

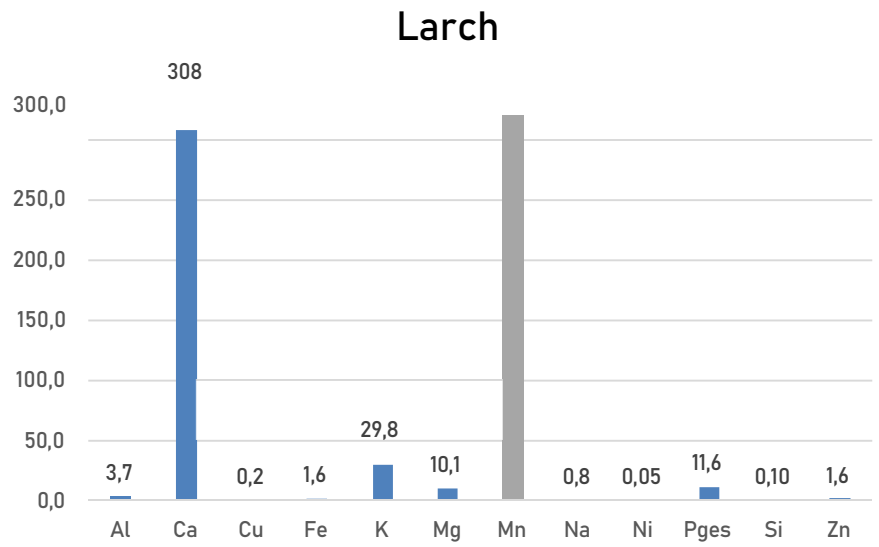
ICP was performed to track the amount of elemental constituents in bark ash. Figure 4.3.4 shows the results of all screenings. Measurements were taken in a calibration range of 0 mg/l up to 1000 mg/l. Some samples show higher amounts of some elements. Since it can be assumed that the calibration curves develop linearly, higher values can also be measured. In all

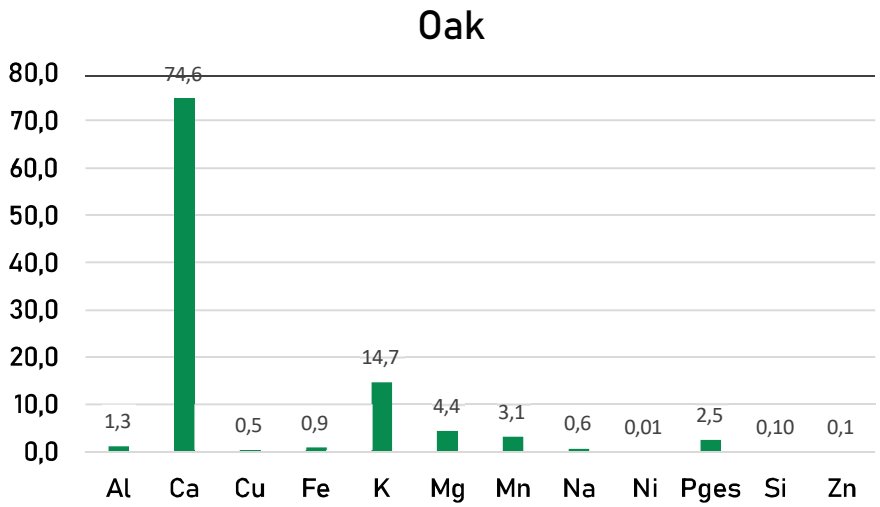
samples calcium was the predominant element, with the absolute amounts varying between bark species. While lime ash had a calcium content of 388 mg/g ± 0,78 mg/g, in oak ash only 74,6 mg/g ± 0,78 mg/g was measured. The second most abundant element was potassium. The concentration of potassium was the highest in robinia ash (54,7 mg/g ± 1,06 mg/g) and the lowest in oak ash (54,7 mg/g ± 0,49 mg/g). Magnesium and manganese were present in a substantially lower concentration. Exceptions are the high manganese content in birch ash and larch ash. Here the values of manganese were too high to make an accurate statement about the amount for this element. For a determination of the manganese content in birch ash and larch ash, the measurement needs to be repeated with a smaller amount of ash or a higher dilution.

Phosphor and phosphor compounds were measured in the lowest concentration for pine ash (1 mg/g) and the highest for larch ash (11.6 mg/g). All other elements (copper, iron, nickel, phosphor and phosphor compounds, silicon and zinc) were measured under 2 mg/g.

Bark ash	Label	Measured elements			
Oak		Aluminium	Al	Natrium	Na
Lime		Copper	Cu	Nickel	Ni
Birch		Iron	Fe	Phosphorus	Pges
Robinia		Potassium	K	Silicium	Si
Spruce		Magnesium	Mg	Zinc	Zn
Pine		Magnese	Mn		
Larch					

all results in mg/g

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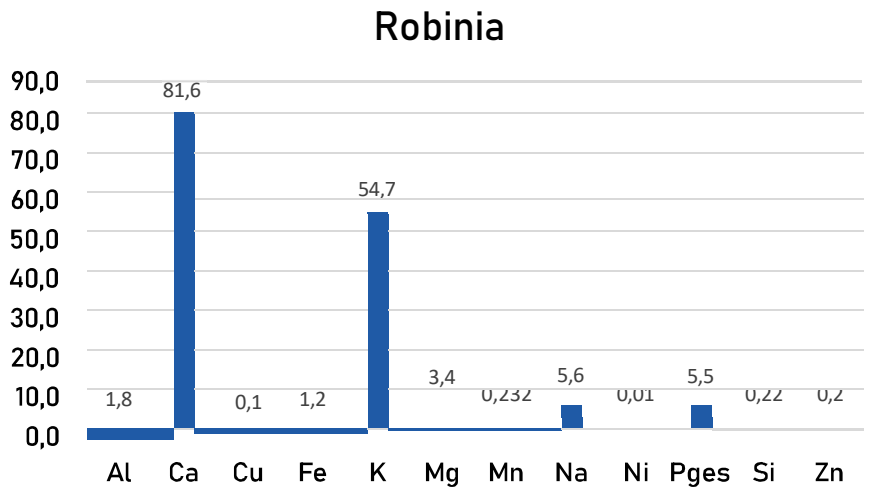
20,0
10,0
0,0
74,6
14,7
1,3
0,5
0,9
4,4
3,1
0,6
0,01
2,5
0,10
0,1
Al
Ca
Cu
Fe
K
Mg
Mn
Na
Ni
Pges
Si
Zn


Robinia
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80,0
70,0
60,0
50,0
40,0
30,0
20,0
10,0
0,0
81,6
54,7
1,8
0,1
1,2
3,4
0,232
5,6
0,01
5,5
0,22
0,2
Al
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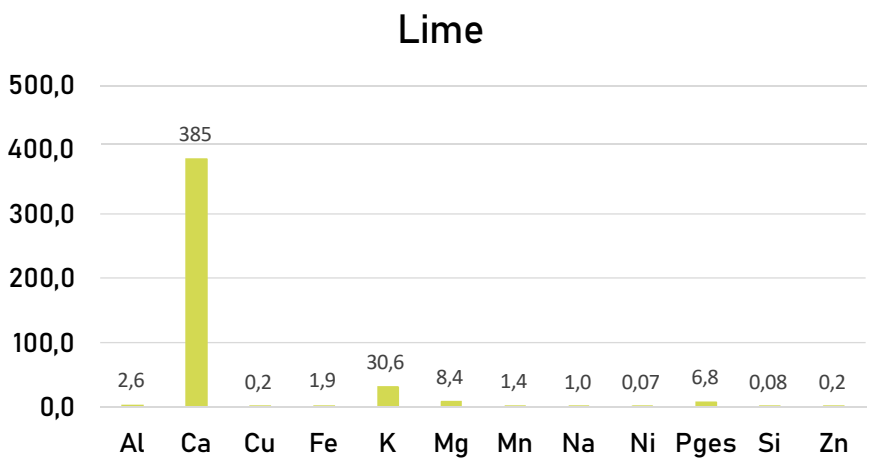
Fe
K
Mg
Mn
Na
Ni
Pges
Si
Zn


Lime
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400,0
300,0
200,0
100,0
0,0
385
2,6
0,2
1,9
30,6
8,4
1,4
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Fe
K
Mg
Mn
Na
Ni
Pges
Si
Zn

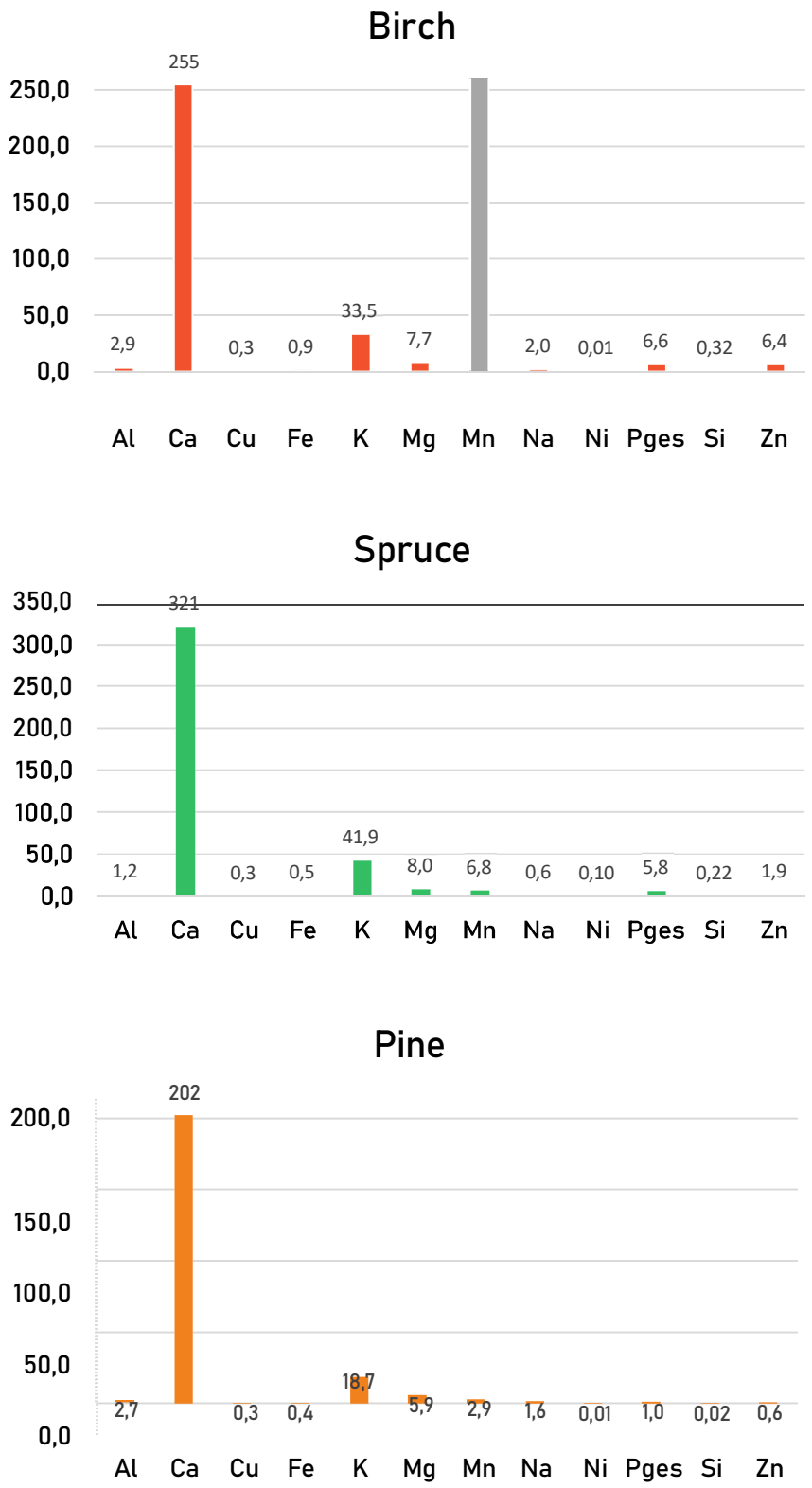


Figure 4.4.4 *ICP results of tested bark ash. Bar charts show the mineral constituents of the individual bark ashes measured with ICP. All values are measured in mg/g. The maximum measurement*

level for all samples had a calibration range of 0 mg/l up to 1000 mg/l.

Distribution of mineral constituents in different barks

Bark ash is mostly produced in the wood processing industry. To better understand the composition of "industrial bark ash", ICP measurements were made. The relative amounts of elements measured with ICP are consistent with the literature data. The main element found in all bark ash is calcium followed by potassium (Bryers 1996). A reason for the high calcium content is the presence of calcium oxalate crystals which are frequently located in sieve cells and longitudinal parenchyma in the phloem and phelloderm (Hudgins, Krekling, and Franceschi 2003), (Evert 2006). The other elements, mostly metals, (copper, iron, cornett) but also silica, magnesium, natrium and phosphorus were measured in less percentage compared to calcium and potassium. This also agrees with previous research about ash composition in bark (Fengel and Wegener 2011), (Miranda et al. 2012), (Saarela et al. 2005). Surprisingly, manganese, generally only a minor constituent of bark, is highly concentrated in birch ash and larch ash. Both bark species were peeled manually with crowbars. Manganese is used in the steel industry as an alloying constituent. Therefore, it can be assumed that the high manganese values come from the tools used and not from the barks. It is therefore difficult to evaluate the measured manganese values. Since the measured ICP values largely agree with those found in literature, it is still possible to conclude on the general composition of individual bark types. Processing methods that could use a high amount of calcium (e.g ceramics) would therefore be conceivable.

4.4.2.3 Bark Glaze Recipe

First glazing results showed that all transparent glazes in combination with bark ash were colored (Figure 4.4.5). The colors ranged from yellow, brown and blue-green tones. Most of the bark ash moved to and accumulated at the center of the bowl, where a thick layer of glaze was formed. All glazes bonded with the ceramic body. A few bowls had a transparent sheen on the interior walls, while others were matte. In the following text, only selected results will be discussed. All glazed and fired bowls are shown in a complete overview in Figure 4.4.7

Figure 4.4.5: *First glazing results with transparent porcelain glaze with borate (Tab 3.2.1)*

One of the most colorful result was the use of spruce ash (Figure 4.4.6, a). The inside walls of the bowl are only partially covered by the spruce glazes. The glaze formed a netlike pattern and flowed onto the bottom where it formed a thick layer of dark brown glaze. The larch glaze (Figure 4.4.6, b) developed a fully covered, shiny beige surface. The surface is perfectly closed and the color varies between beige, blue and purple, according to the thickness of the layer. After firing, the birch glaze (Figure 4.4.6, c) formed a closed and strongly rose-colored surface, which could be clearly seen especially in thicker layers.

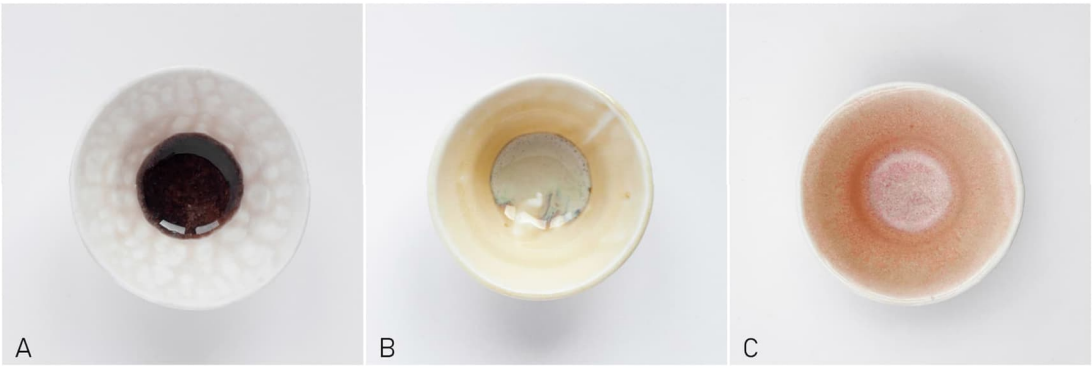


Figure 4.4.6: *Selected test bowls after the first firing test (borate glaze), (A) spruce ash glaze, (B) larch ash glaze, (C) birch as glaze*

For further investigation, the glazing experiments were expanded with different glazing recipes as well as a test series with pure bark ashes (used as glaze by itself). The previously used borate glaze recipe was also tested again.

Results are shown in Figure 4.4.7. The bark samples evaluated in these 4 test series are robinia, larch, spruce, birch. In addition, each test series includes a bowl fired without bark ash (pure glazing) as a reference sample.

Figure 4. 4.7: *Overview of all bark glazed bowls after the second firing. Four different glazing recipes were mixed with bark ash and fired (Tab 3.2.1 – Tab 3.2.4)*

Pure ash was melted with a gradually increasing addition of fluxes (Figure 4.4.8) Oak and lime show partial bonding with the bowls at all three flux concentrations. In this case, not all ash parts bond with the porcelain and remain as residual ash in the center of the bowl. There is a tendency that the higher the flux concentration, the stronger the fusion of the bark ash. While in the test series at 12% flux, the ash still covers the entire inner bowl and appears gray-black, the unmelted components of the ash decrease at 24% flux and a green-yellow coloring of the ash occurs. In the last step 36% flux oak shows a green-yellowish discoloration and lime shows single melted areas in a yellow-brown tone.

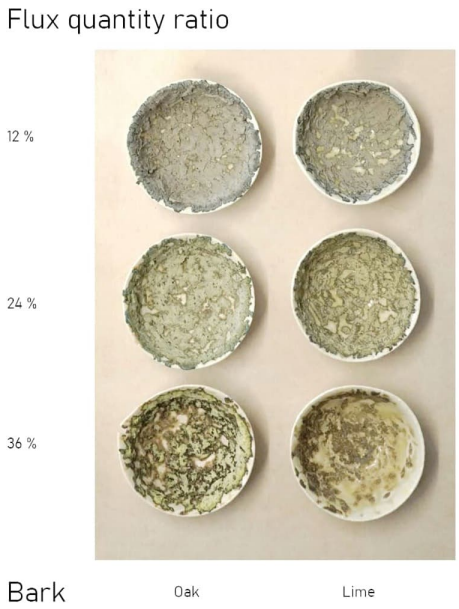


Figure 4.4.8 *Results of glazes made out of pure bark ash. Oak and lime ash were mixed with 12%, 24% and 36% of conventional glazing flux. The results demonstrate how much bark melts to form a glaze layer.*

The previously used recipe (borate glaze, Tab 3.2.1) was used a second time. Due to limited material, the repeated test was only tried on the smallest bowls (Figure 4.4.9). Robinia showed a transparent slightly yellowish glaze. The adhesion between body and glaze is good, but the surface shows slight imperfections. The result for the larch ash showed a glossy glaze in a slightly darker color this time but in the same color spectrum of reddish brown on the outer walls and bluish purple in the center of the bowl. The birch glaze also adhered well and resulted in an even rose tone with a glossy rather than matt surface. The strongest difference is seen in spruce. In contrast to the dark brown transparent glaze, the coloring was a light shiny beige with a whitish blue veil (boron veil) in the middle. The surface of the spruce glaze has developed a craquelure.

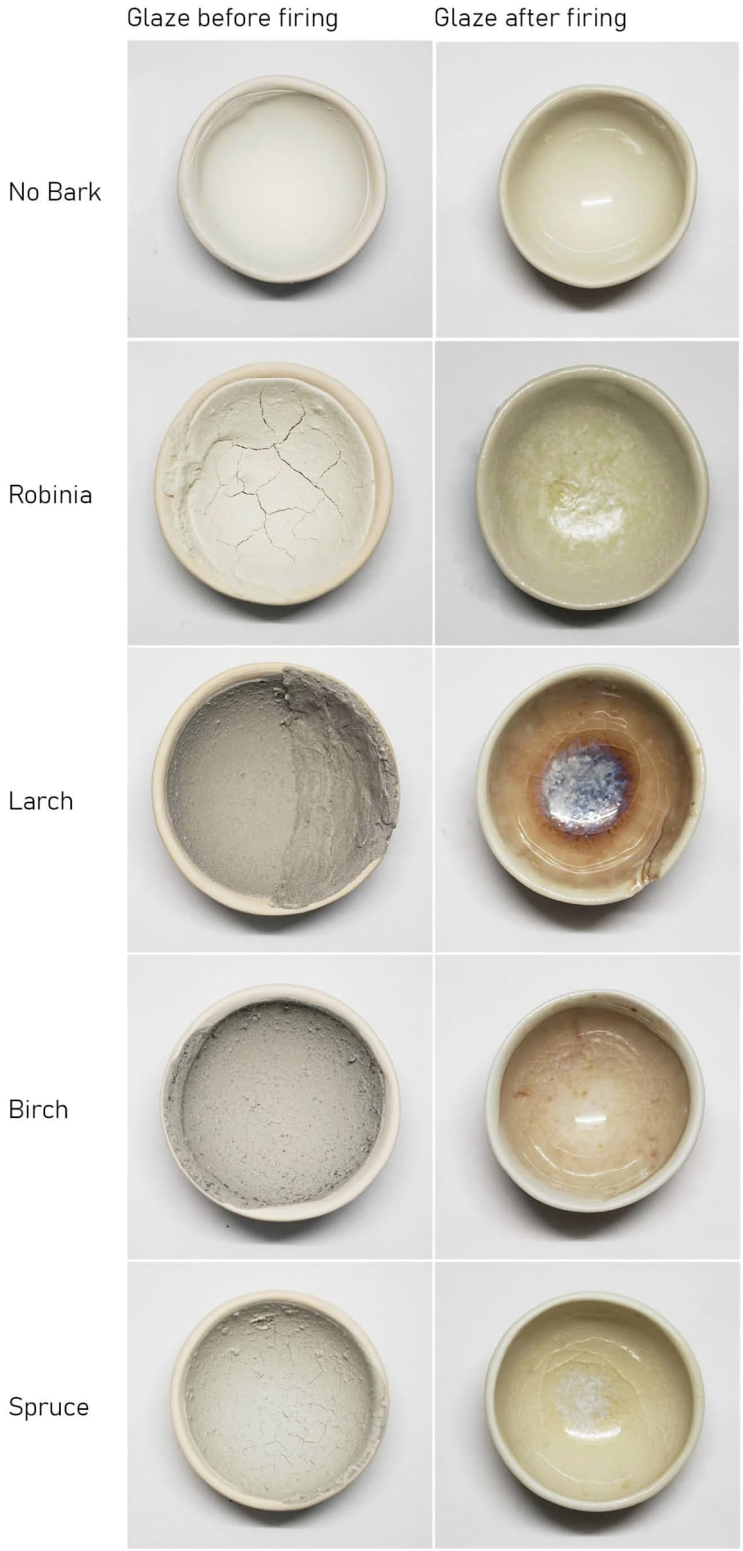


Figure 4.4.9: *Glazing results of the second test with borate glaze (Tab.3.2.1). The left column shows glazed bowls before firing, the right column shows glazed bowls after firing*

The second transparent glaze was provided by Dirk Friese (Tab. 3.2.2). The combination of this glaze with robinia produced a white matt surface with a surface structure. Birch showed a glossy coloring in deep red-brown. The glaze flowed strongly so that a lot of glaze collected in the middle of the bowl. Spruce glaze is light brownish and beige. Larch was not included in this test series because of material limitations.

The matt glaze (Tab. 3.2.3) leads to different results compared to the first glazing experiments and the transparent glaze (Tab.3.2.1, Tab. 3.2.2). The robinia glaze strongly flowed in the inner wall of the bowl and is glossy. In the center the glaze is brownish yellow. The birch glaze is glossy with a deep red-brown coloring in the bowl center. The spruce glaze is transparent yellow to orange in the center and has cracks in the glaze. The larch glaze also heavily flowed on the bowl walls. A dark purple-brownish coloration appeared in the bowl center.

Crystal glazing (Tab. 3.2.4) is frequently used for decorative purposes due to enhanced crystal growth. Already the basic glaze has a characteristic coloring. Crystals are clearly visible in the center of the reference bowl. The robinia sample shows the reference coloration at the upper edge. In the center, yellow-brown glaze is collected, which is transparent and shiny. The glaze in the center shows a craquelure with 3 grown crystal groups. Birch shows a reddish brown, strongly net-like melted glaze with bluish and uneven glaze in the center of the bowl. The spruce glaze has a net-like structure flowing into the center from light orange to deep brown. Very strong crystal growth occurred in a light orange in the center. The larch glaze shows patchy glaze with a shiny deep brown glaze accumulation in the center and no optical crystal growth.

		Glazes	Color	Surface quality	Fluidity
Reference glazes without bark ash		Transparent	- glossy transparent	- closed, smooth	- good adhesion, good dispersion
		Matt	- semi matt - white, beige	- closed, smooth	- good adhesion, good dispersion
		Borate glaze	- glossy transparent - white, transparent, bright blue spots	- closed, smooth	- good adhesion, at the center thicker
		Crystallization	- semi matt - edge: blue, grey - center: blue, white crystals	- partly uneven rough in the center	- high fluidity, at the center thicker

		Glazes	Color	Surface quality	Fluidity
Birch bark ash		Transparent	- semi matt - edge: red brown - center: dark brown, white spots	- edge: closed, smooth - center: uneven	- good adhesion, at the center thicker
		Matt	- edge: glossy beige, yellow, transparent - center, glossy, dark brown, purple, white spots	- edge: closed, smooth - center: uneven	- good adhesion, at the center thicker
		Borate glaze	- edge: glossy beige, yellow, transparent - center, glossy, dark brown, purple, white parts	closed, smooth	- good adhesion, at the center thicker
		Crystallization	- semi matt - edge: yellow, brown - center: brown, white parts, not crystals	- partly uneven, rough in the center	- high fluidity, at the center thicker
Robinia bark ash		Transparent	- semi matt - white, with brown spots	- partly uneven, rough	- good adhesion, good dispersion
		Matt	- edge: glossy beige transparent - center, glossy, yellow transparent	- closed, smooth	- good adhesion, at the center thicker
		Borate glaze	- semi matt - edge: white, beige - center: white, beige	- closed, uneven	- good adhesion, un-equally distributed thickness
		Crystallization	- edge: blue, grey - center: glossy, yellow, craquele, white crystals	- edge: closed, rough - center: closed, shiny	- high fluidity, at the center thicker

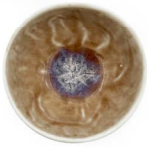
		Glazes	Color	Surface quality	Fluidity
Spruce		Transparent	- semi matt - beige, with white spots	- closed, smooth	- good adhesion, good dispersion
		Matt	- edge: glossy beige transparent - center, glossy, yellow transparent	- closed, smooth	- good adhesion, at the center thicker
		Borate glaze	- semi matt, beige, partly transparent	- partly uneven	- high fluidity, at the center thicker
		Crystallization	- edge: partly unglazed, brown - center, dark brown, orange crystals	- partly uneven - rough in the center	- high fluidity, at the center thicker
Larch		Matt	- edge: glossy beige, yellow, transparent - center, glossy, dark brown, purple, white spots and lines	- closed, smooth	- good adhesion, at the center thicker
		Borate glaze	- edge: glossy beige, red, transparent - center, glossy, brown, bright blue, white spots and lines	- closed, partly uneven	- good adhesion, at the center thicker
		Crystallization	- edge: partly unglazed, brown - center, glossy, dark brown,	- partly uneven, closed in the center	- high fluidity, at the center thicker

Figure 4.4.10: *Comparing results of all four glazing recipes. Transparent, matt, borate glaze and crystal forming glaze of birch glazes, robinia glazes, spruce glaze and larch glazes are shown and described in their color, surface quality and in their metlting behavior on the bowl (fluidity). Bowls glazed without bark are fired to provide reference of the glazing appearance without bark ash.*

Potential of bark ash as a substitute for porcelain glazes

The usage of ash for glaze has a long tradition. It goes back as far as 1500 BC, China. Various plants were burned in a specific way to create the ash (Tichane 1998). Studying the varying structured of the ash and the close observation of the changing process after burning of the glaze is an art in itself. The use of bark ash in this thesis is motivated by the idea to use any leftovers of the natural resource bark.

In the first test series, all bark ashes changed the appearance of the used glazes. The color spectrum ranges between yellow, brown and blue green tones. In historic literature these colors are also described as the colors found in the production of antique glass (Tichane 1998), caused by the oxides of copper, iron and manganese (Wolf 2012). Wood ashes usually contain larger amounts of CaO, MgO and K_2O, which can lead to a higher surface tension in the glaze and is used for decorative surface patterns (Figure 4.4.6 A). Grass ashes contain more SiO_2, Al_2O_3 and more sulphates which can lead to a highly viscous glaze and - in combination with low-iron oxide ash, can result in blue-green tones in the glaze. (Matthes 1985)

While the reference bowl is transparent and white, the spruce ash bowl shows a strong deep brown coloration, which spreads like a net along the walls into the center of the bowl (Figure 4.4.6, a). This effect is described for high calcium oxide concentrations in glazes (Wolf 2012). Spruce ash is very rich in CaO, which produces surface tension resulting in this characteristic for glazed surfaces. Birch bark shows a strong matt rose colored glaze. The change from glossy to matt can be due to many factors. If glaze is glossy (which is the base glaze), then dulling is a fault in the firing process. One reason can be traces of charcoal in the glaze, which can lead to a higher amount of gas development during the firing process. Another reason could be that the heating or cooling curve (of the glazing process) does not match the composition of the glaze, which can lead to increased devitrification (rough matting) (Lehnhäuser 1985). The rose color might come from the combination of boron (from the base glaze recipe) and the extremely high amount of manganese (Lehnhäuser 1985). According to literature (Lehnhäuser 1985), this combination can cause a violet tint in glazes.

The test series with the pure ashes shows that it is possible to use the mineral components of oak and lime (with the help of fluxes that promote the sintering process) to produce a glaze-like coating (Figure 4.4.6). Historical records state that calcium in wood ash was used as a flux (Wolf 2012). Since bark has a higher calcium content than wood (1:10), it can be assumed that tree bark ash can serve as a natural flux for glazes (Tichane 1998).The unequally distributed glaze spots in the bowls suggest an inhomogeneous distribution of the components within

both types of bark (Figure 4.4.8). This can be caused by the incomplete burning (described in 4.3.1). The bark ashes can be influenced by the amount of remained coal in the glaze. This could have a negative impact on the series of experiments, since coal burning can cause gas formation and melting behavior changes during the glazing process. Further investigations are needed for further analysis and optimization of the glazing process.

The results for the transparent glaze (tab 3.2.1) from the first test series (Figure 4.4.5) were repeated in a second series (Figure 4.4.9). Small differences appear in the color intensity (larch) and in the gloss or matt finish (birch).

For the color change if larch ash glaze, it can be assumed that the thicker layer of glaze in larch is probably the reason for the higher color intensity. In case of birch ash glazing test, the firing conditions and the glaze seem to be optimal for the birch test series, as there was no change in gloss and only minor changes in color intensity.

A completely different coloring was obtained for spruce which changed from dark brown in the first test series to light shiny beige in the second series. Reasons could be a change in the distribution of the chemical components between the first and second series. This differ in the distribution of individual glazing components was caused by incomplete mixing between the first glazing and the second glazing. Glazes usually consist of water-soluble and water-insoluble components. Therefore, it is important to mix and sieve glazes carefully before use. Since all glaze tests were performed manually, there is a risk that there was incomplete mixing between the two glazing experiments. An additional error component may have resulted from the skipping of the sieving step. This could lead to the color change of spruce glazes between the first and the second test series.

Expanding from one glaze recipe to three was intended to further the understanding of bark ash as a glaze substitute. The previously described observations from the first glaze experiments (Tab 3.2.1) were repeated in the remaining glazes (Tab 3.2.2, Tab 3.3.3, Tab 3.3.4). Most of the glaze accumulated in the center. Bark ash promoted a melting of the glaze for all samples. All results ranged from white, beige, rose, yellow to reddish brown color spectrum. These are typical colors produced by the high content of iron and manganese (Wolf, 2012 #97}. While the colors are very similar, there is a high diversity between the samples in terms of surface gloss and surface texture.

The development of a glaze during firing depends on the exact mixture of substances in the glaze, the firing curve, the exact temperature and the oxygen content in the furnace

(Lehnhäuser 1985). The variability of all results shows how precisely this process must be tuned and that a certain degree of variability of the results cannot be avoided.

In addition to a high variability in the glaze result, there were also problems with the production of the bark ash in these series of tests. Coal has a lower ash content than fully burned bark. Thus, quantified values of the required amount of ash might be incorrect. This also changes the color result and probably leads to significantly lighter colors. In addition, pieces of charcoal may be unevenly distributed in the glaze and thus lead to stains.

Presumably, these problems do not play a role in industrially produced bark ashes, since they are burned for a longer time. All the described glaze tests were made in an oxidizing fire. It is also possible to burn without oxygen supply. This could lead to other colors (Matthes 1985). Typical percentage of plant based ash is 30% w.t. for ash glazes (Tichane 1998). But also high ash glaze with 30 to 60% and 100% are possible (Tichane 1998). The possibilities of tree bark glazes are by far not exploited yet.

5 Objects

The Interlacing of Scientific and Design Research

The approach of this thesis is to combine design practice with methods of scientific research. This puts the collection of new knowledge, through design, on the same level as the natural sciences. Similar approaches have been conducted throughout human history and, depending on the zeitgeist and definition, the two disciplines have been considered either close to or distant from each other. (Myers 2012).

The relationship between science and design strongly depends on the motives of its connection. It makes a difference whether one profession simply uses methods of the other or whether the two bring their combined focus to a shared problem. The latter can lead to the creation of boundary objects, which will be discussed further in chapter 5.5.

A period in which design strongly sought connection with science was the 1960s. In the USA and Western Europe, this time marked a great openness towards experimental scientific and technological setups (Mareis 2014). The new design movement aimed at rationally and objectively recording and controlling design processes that had been carried out more or less intuitively until then (Mareis 2014). Criticism of this movement was intense. In her thesis, Claudia Mareis describes how practically motivated concerns mutated into abstract theoretical undertakings. Designers complained about the academicization of the discourse and thus about an alienation of design values (Jones 1977, Jones 1992). The reasons for this resistance probably lie in the mere adoption of non-design methods. Science was used only as a tool for restructuring design processes, but was not included in their making. The implementation of new methods can be more successful, especially if a method is understood in the context of design and shaped to its requirements.

One example of putting science into practice is the work of the architect, designer and scientist Buckminster Fuller, who coined the term "design science" as a problem solving method (Fuller 1963). He pursued a decidedly biological systematic approach, as seen in one of his most famous works, the US Pavilion at the 1967 World's Fair in Montreal. By transferring principles from nature to larger scales of biological structures, he created a strong link between the natural sciences and design.

In the world's most widely read book on the responsibility of design "Design for the real world", Viktor Papanek (Papanek 1972) stated that biology, bionic and related fields can help

design to develop more sustainable solutions (Papanek 1972). To Papanek, finding analogies was crucial to solving increasingly complex problems. He recommended setting up interdisciplinary design teams (also including end users and workers) and using biological prototypes and systems to create holistic design outcomes and products in the context of their environment.

Over the past 20 years, the role of design has increasingly transformed to engage with valued dialogue in new achievements in natural sciences and emerging technology. The exhibition "Design and the Elastic Mind'', curated by Paola Antonelli at MOMA in 2008, showcased design and its influence on science. It highlighted examples of design in innovation and in ongoing research, as well as reflections on future responsibilities of design. Developments in nanotechnology, speculative design, bionics, computer-aided fabrication and biological-based objects were represented (Antonelli, Hall et al. 2008). All these works show how an exchange between design and science is possible. Through joint work on larger questions that equally concern both professions, as well as through the use of new technology or new materials, an interlacing of both research disciplines happens. As science opens up new fields of design, the latter also becomes a creator of scientific data and materials. (Antonelli, Hall et al. 2008, Myers 2012).

In the numerous possibilities that connect science and design, this research is based in the field of biomaterials research. In the context of this thesis, I will mention three groups that deal with biomaterials in different approaches. The first group I call "design with grown materials''. This consists mostly of plants and organisms that have not been considered in industrialized design before. Two of the best known examples are seaweed and mycelium: seaweed is used as food (Lohmann 2018), for the production of yarn (AG 2020, Hoogvliet 2014 onwards), as dyes for the fashion industry (Hoogvliet 2014 onwards) and as material for interior design (Lohmann 2018), while mycelium has shown to be suitable to form solid parts of various sizes. At Dutch Design Week 2018, for example, an entire “Growing Pavilion“ by Pascal Leboucq (Leboucq 2019) was made of mycelium. But also packaging material (grown.bio 2016) and technically advanced products such as bicycle helmets (Meyer, Schmidt et al. 2020), fashion and furniture can be made from mushroom mycelium (Wang 2022). Because of its property to root in all spatial directions, mushroom mycelium is also being used to develop sustainable 3D printing material (Klarenbeek 2011).

The second design group is "design with growing materials". In this approach, materials with the desired properties are cultivated and processed. Bacterial cellulose, for example, is used

to create alternatives for the leather industry (Lee 2003, Materials 2021). Especially in the field of fashion and in the color industry, designers and biotechnologists are creating new sources for pigment and structural colors (Johansen, Catón et al. 2018, Brunato 2019).

The third design group is "design with grown leftovers''. It describes the utilization of organic industrial waste by expanding application fields for biogenic materials. This can be done with either minimal modification or complete processing of the material. Fiber plants for example are often used as fabrics in composite materials, such as facade components in architecture (Elfordy, Lucas et al. 2008) or as decor (Laposse 2017). With minimal modification, they can be manufactured into products made from straw fibers (Lavicka 2016), (Goodrum 2018). Complete processing of waste materials is widespread, especially in the production of bio-plastic alternatives. Crustaceans shells (theshellworks 2019) are used as a basis for plastic alternatives, including plastic 3D printing filaments (Mogas-Soldevila, Duro-Royo et al. 2014). It should be noted that the biomaterial applications described here cover only a small part of current developments. In this context, the boundaries between science and design are becoming increasingly fluid, and collaborations between the two disciplines are contributing to even more far-reaching development.

This thesis is located within the third group "design with grown leftovers''. *It focuses on the utilization of tree bark as a by-product of the wood industry. The design experiments and objects described in this chapter were created in close collaboration with my colleague Johanna Hehemeyer-Cürten. We worked intensively together on the experiments, conception and execution of all design objects. Development of cutting patterns as well as the production of the jackets, was done by Johanna alone.*

5.1 Simple Bark Jacket

Bark protects tree. It forms the interface between the environment and the vital cambium and xylem. But what does this protection feel like? Can bark take on protective functions for humans as well? These questions arise when peeling bark off a tree for the first time. While the flexibility of the freshly peeled, wet pine bark is very similar to that of textile materials, it is lost as soon as the bark dries and requires flexibilization to be maintained (see chapter 4.2). The aim of creating a plain bark jacket was to explore bark as a boundary layer for humans.

5.1.1 Idea

The feel and color of glycerol-treated, flexible bark is similar to leather, animal skin tanned with substances from tree bark. Leather is an attractive material in design, with designers and scientists currently looking for replacements without animal origins (Muthu 2020). Humans often use leather as a kind of second skin and as a protection from the environment, as in clothes and shoes that protect against the cold or damp. Multi-layered and based on fibrous elements, bark can be seen as the skin of plants, and hence as a structure that works as an enclosure as well as a connection to and trading zone with the environment.

However, if we want to address the design purposes of today by using waste bark material as a resource, we need to ask the following question: What structure–property–function relations can be translated from bark into the realm of human beings?

5.1.2 Concept

To evaluate different manufacturing techniques as well as the sensory aspects of the output, a jacket was selected as the research garment. Jackets cover vital, cold-sensitive regions of the body. In contrast to other garments such as skirts, they require a high amount of visual transformation, deviating noticeably from the original shape of a tree. Jackets consist of both large contiguous areas that have a warming function (back) and parts that require a high degree of mobility (arms).

5.1.3 Process

To best explore the feel of tree bark against the skin, the jacket and separate hood had to cover as much body surface as possible, as tightly as possible. For this reason, the prototype was tailored-made to the model's measures, contained no inner lining and only very few non-bark materials (sewing thread and zippers).

The six-piece pattern of the jacket was based on a classic HAKA (menswear) jacket pattern, consisting of two sleeves and a vest. The vest was made of four large pieces of bark, which were reinforced at the front with an additional layer. To be able to close the jacket, a zipper was integrated. In the neck area it was attached to a collar made of folded bark. Due to a lack of material, the sleeves had to be segmented into three parts. To make them fit even tighter,

each one of them contained a zipper. The cutting of bark was done with a scalpel. The sleeves were made with a sewing machine and then sewn by hand to the body of the jacket (Figure 5.1.3).

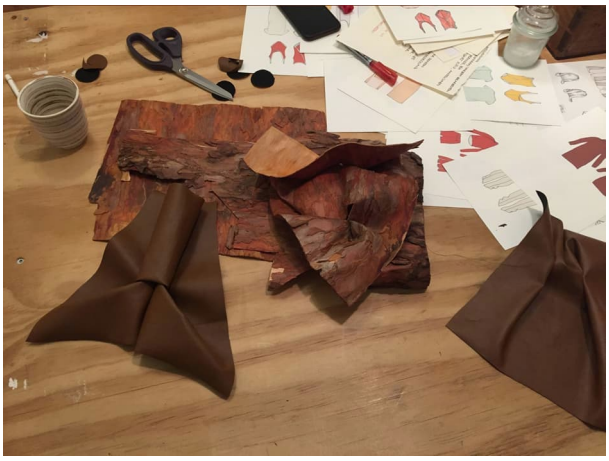


Figure 5. 1.1: *Design experiments for the production of the first jacket. Folding techniques were tried using leather first. Afterwards, the same folding technique was repeated in flexibilized pine bark.*

The hood was made separately (Figure 5.1.2). This way it could be put on first, adjusted very tightly to the head and then fixed with the collar of the jacket. No additional connecting items (zippers, buttons, etc.) were necessary. The three-piece pattern of the hood was based on a classic balaclava, with a collar that could be folded in the front. To create a three-dimensional round form fitting the shape of the head, pins were used at the side parts of the pattern. All three parts were sewn by hand. The edges of the hood were finished with a black ribbon trim in order to protect them and prevent internal tears from expanding.

Figure 5.1.2: *Hood made of pine bark. Ribbon was used to protect the edges.*

Figure 5.1.3: *The production of the first bark jacket. Sleeves are made out of three pieces of bark. The vest is sewn with a leather sewing machine and lined with a ribbon. Images show the back and front of the sleeves and vest.*

5.1.4 Result and Discussion

The aesthetics of the first bark jacket were photographically documented (Figure 5.1.4). In order to fully focus on the effect of the bark, all non-bark parts (hood and zipper) were removed.

Figure 5.1.4: *Image of the finished bark jacket, front. Image copyright by Studio Patrick Walter and Max Planck Institute of Colloids and Interfaces*

The bark jacket was examined based on two fundamental design aspects: the process of manufacturing and the feeling of wearing it. To investigate the feeling of bark on skin, the model was helped into the jacket at room temperature (Figure 5.1.5). In a short interview undertaken to obtain qualitative descriptions, he then answered questions about its comfort. Although the feedback received was subjective and the experiment was not repeated as such, it helped gaining a first impression of the haptic, sensory features of the bark jacket. At first, the model was asked to comment on the general feeling wearing the jacket. Immediately after dressing, it was described as rather pleasant, although some areas, such as the sleeves, were perceived as very tight and damp. Next, the model was asked to evaluate the fit. Although the jacket was tailored to individual measurements, the model did not feel that it fit particularly well. While the sleeves were very tight, the jacket was wide around the stomach. Overall, the

model described the jacket as aesthetically pleasing but not very comfortable. When asked what the feeling of the material on the skin was comparable to, the jacket was described as leather-like with a resinous smell

Figure 5.1.5: *Model during the interview.*

Ten minutes later the model was asked the same questions again. The comfort had decreased and the jacket was described as moderately pleasant looking. It now felt hard on the skin and very cold in the tight areas. The wider areas around the waist were a little warmer. The feeling of the material was compared to damp leather and the specific smell was no longer noticeable.

Overall, the feel and enjoyment of the jacket had deteriorated significantly.

The flexibility of the bark was highly limited, as later measured in the tensile tests (see chapter 3.2.7). The material did not stretch at all, which is a required feature for most textiles. As a consequence, the model could not lift his arms and found the jacket to be rather uncomfortable. In comparison with leather, flexible pine bark is approximately five times stiffer along the direction of the fibers. The limited mobility while wearing the jacket results from bark being highly anisotropic, since the majority of its fibrous elements run parallel to each other (Wenig, Dunlop et al. 2021). The anisotropy of bark also caused difficulties during the manufacturing process. The jacket was fragile perpendicular to the fiber grain. The bark could not be loaded

parallel to the fibers. Tension or weakening of the material by creating seams caused the bark to tear (Figure 5.1.6, right). To avoid this, seams had to be used with the largest possible opposite angle when connecting two pieces of bark (Figure 5.1.6, left). The help of sewing machines was limited by fragility and restricted flexibility. Seams turned inside and points where the material had to be greatly compressed (e.g., at the shoulder) were rather unsuitable for the use of a sewing machine.

Besides the difficult processing, the material also proves to be difficult to assemble. The need to extract large defect-free pieces of bark for sewing resulted in a substantial amount of the harvested bark remaining as waste.

Figure 5.1.6: *Assembly details. left picture: cracks in the bark, right picture: wider fiber angles were preferred for connecting two pieces of bark.*

In summary, the first jacket shows pre-treated bark to be more flexible than dry bark and is especially interesting aesthetically. However, the lack of flexibility and severe processing difficulties left room for improvement. This is due to the fact that bark properties are comparable neither to those of leather (see chapter 4.3.1.3) nor to those of conventional fashion textiles. The latter are most often made of single fibers, woven into a fabric – a promising principle for further research on the usability of tree bark. This is described generally in chapter 5.2 and is applied to bark in chapter 5.3.

5.2 Creating a Textile by Weaving

In the context of this thesis, the first prototype's lack of flexibility, the bark's anisotropy and the amount of waste material resulting from the production were addressed by weaving techniques.

Weaving is one of the most ancient crafts. Even older than ceramics, the fundamental principle of weaving remains essentially unchanged (Albers, Weber et al. 2017). Whether woven by hand or by machinery: two fibers or threads are rectangular interlacing, which can be used to either support or to impede the different threads' material characteristics (Albers, Weber et al. 2017).

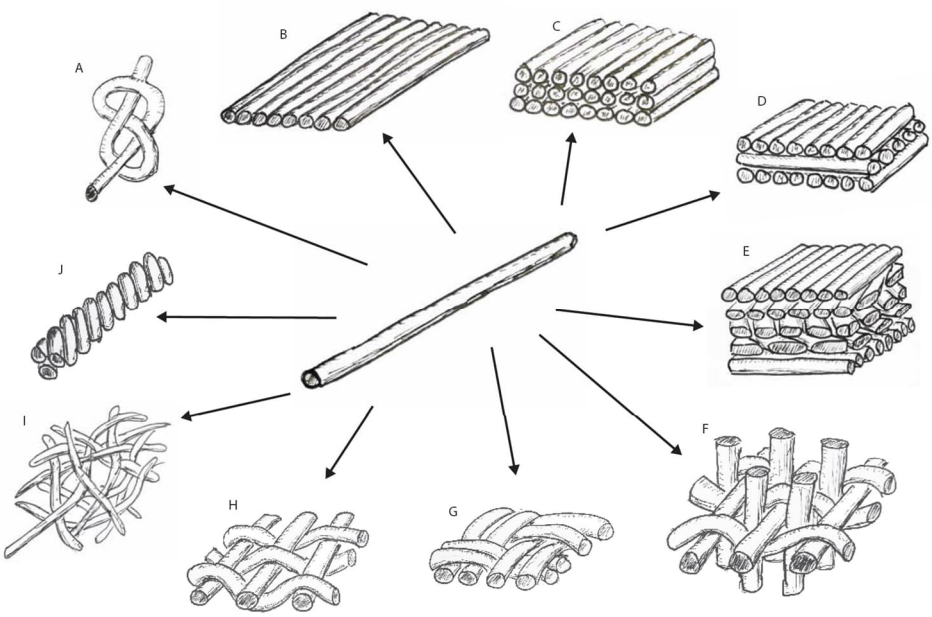


***Figure 5.2.1**: How single anisotropic fibers can be built up into macroscopic materials with new properties. Fibers can be knotted (A) arranged in layer (B) that can be stacked in different ar- rangements (C–E) giving anisotropic or almost isotropic properties. Fibers can be twisted (J), ran- domly arranged (I) or woven in two (G, H) or three dimensions (F). Figure reproduced with per- mission from the Philosophical Transactions of the Royal Society, Wenig et al. 2021*

Many structural materials in biology consist of fibrous building blocks, like cellulose in plants, with collagen silks and chitin being typical examples (Fratzl and Weinkamer 2007). The fibrous nature of these components and their inherent anisotropy mean that the 'architecture'

or the spatial arrangement of these fibers at different length scales is a fundamental parameter in controlling the properties of the resultant tissue. For fibers, there are several length-scale-independent ways to construct a material (Figure 5.2.1). Isotropic material properties can be achieved by stacking parallel-oriented layers of fibers on top of each other with alternating twists, as is well known in plywood construction (Hull and Clyne 1996). Many such features can be seen in biology (osteons in bone, insect cuticles, plant cell walls) and are thought to improve toughness (Giraud-Guille 1998, Gordon, Cohen et al. 2015, Reznikov, Bilton et al. 2018, Neville 1993). Twisted fibers (such as those used in making cables and ropes) can allow for the transmission of tensile loads between a large number of shorter fibers, retaining transverse flexibility (Costello 1997). Weaving or alternating crossing of fibers under and over each other can create two-dimensional fabrics, with the weave pattern controlling properties in particular directions (Albers, Weber et al. 2017). In principle, weaving can also be done in three dimensions, with fibers interlocking creating functionality in multiple directions (Bilisik 2012, in some way reminiscent of interwoven wood cells (rays and fibers). Random patterning (such as is found in fiber arrangements in paper manufacturing, or in the production of filters or felts) can give uniform properties throughout a plane.

These general thoughts on how fibers can be explored in order to achieve different functionalities can be seen at multiple length scales going from the molecular up to the macroscopic scale of ropes or plywood structures. Although the link between macroscopic properties and the underlying architecture of fibrous materials is understood, the biological processes that are responsible for the assembly of the fibers are still unclear. As a consequence, many research groups are currently working on understanding how 'fibrous patterning' works in living materials such as wood, bone and cuticle."[18]

5.2.1 Processing of Flexibilized Bark

For the weaving test, pine bark was cut into strips of different thicknesses along the fiber. The thickness of the strips varied from 1mm to 2cm. The length of the strips was determined by the total length of the bark used for cutting (Figure 5.2.2). All strips were cut by hand using a

[18] Wenig, C., Dunlop, J. W., Hehemeyer-Cürten, J., Reppe, F. J., Horbelt, N., Krauthausen, K., ... & Eder, M. (2021). Advanced materials design based on waste wood and bark. *Philosophical Transactions of the Royal Society A*, *379*(2206), 20200345

scalpel, a simple but very time-consuming process. Experiments with a laser cutter or mechanical cutting knife indicate that this procedure can be optimized drastically.

Figure 5.2.2: *Different material thicknesses of hand-cut bark strips. The strips are between 1mm and 2cm in length.*

5.2.2 Creating Weaving Patterns

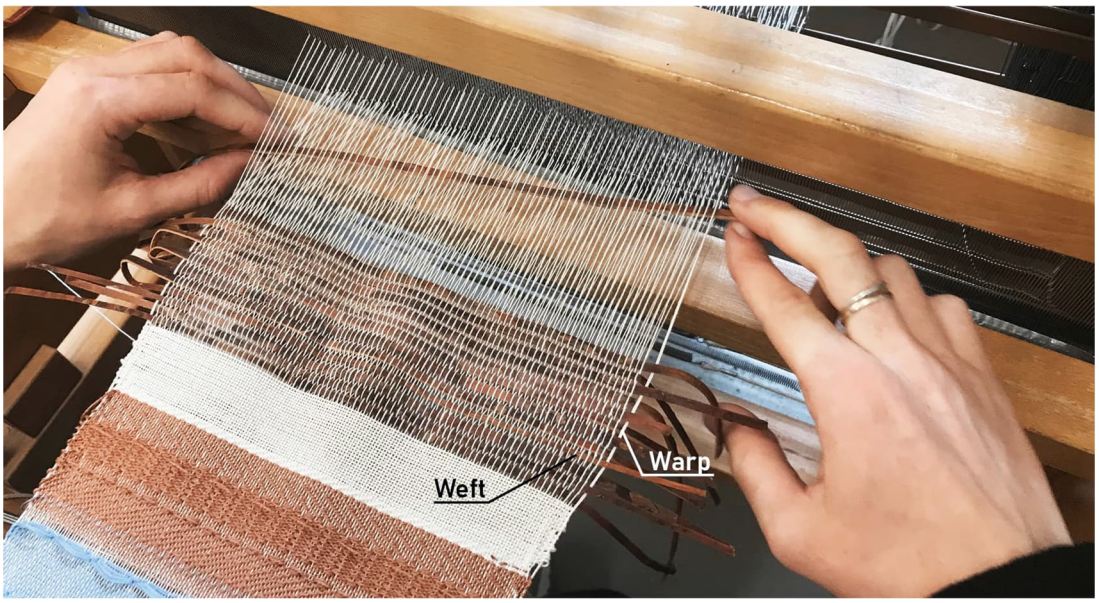


Figure 5.2.3*: Weaving test on a manual weaving chair. The warp (cotton fiber) is set into the manual weaving chair, while the wheft (flexibilized pine bark) is interwoven.*

The weaving tests were made on a manual weaving chair at the Weißensee School of Art, where a cotton yarn was set as the warp (Figure 5.2.3). The bark strips were inserted and woven by hand with the help of controllers so that it was possible to lift and lower threads in a self-selected rhythm. Whenever the bark strips were long enough, they were re-inserted at the ends of the warp, forming a continuous pattern. Short strips had open ends on both sides of the weaving sample. For the test, the three basic weaving patterns were applied: plain weave, twill weave and satin weave. Patterns involving a long float (i.e. long stretches where the bark thread is free to move) were avoided since the moving bark could easily tear from fabric twisting, thus destroying the entire textile pattern. The patterns were documented and their characteristics were described.

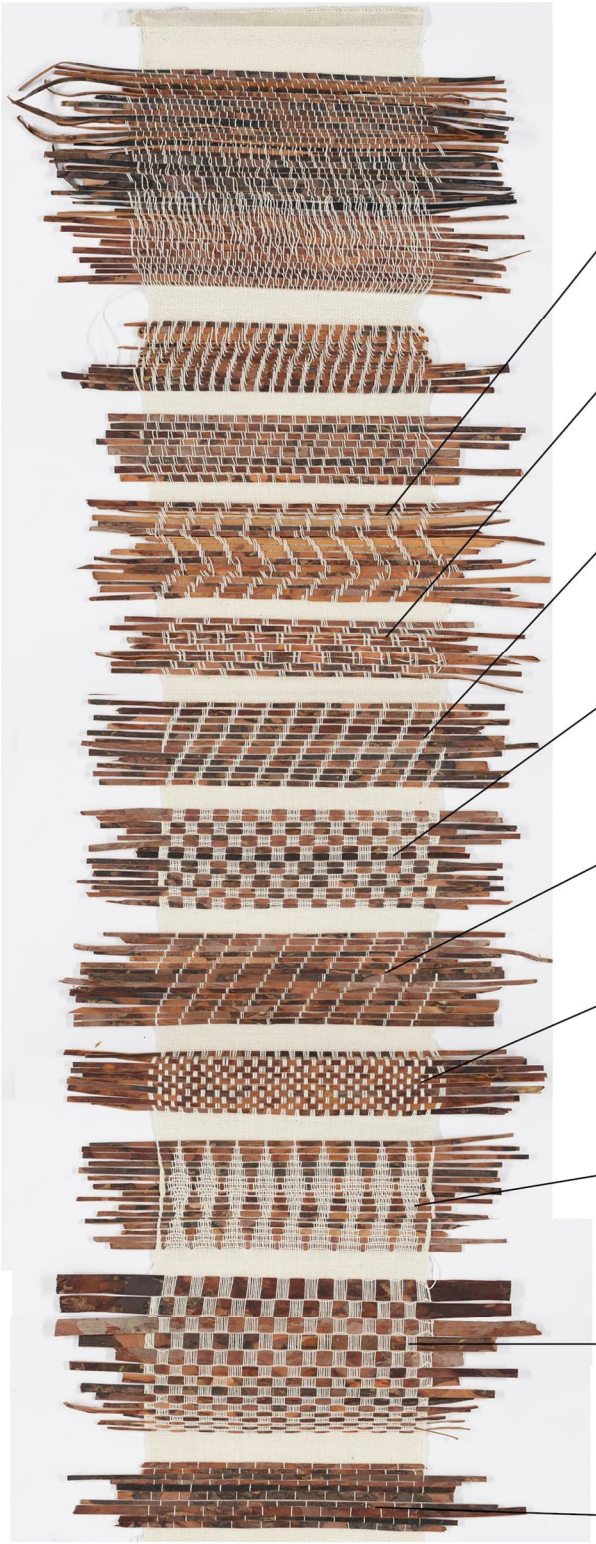
5.2.3 Evaluation of Woven Pattern

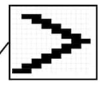
"Through weaving, it was possible to make use of the better mechanical properties along the fiber and to process narrower pieces of bark as threads (Fig. 5.2.1, c)."[19]

Different techniques resulted in different textile characteristics. For example, higher flexibility was achieved when applying a twill weave, while a plain weave created a very sturdy and tear-resistant textile (Figure 5.2.4). Due to its freely movable threads, the satin weave shows no improvement to the flexibility or resistance of the bark textile. It was not only the weaving pattern that influenced the textile; its properties were further changed by the width of the strips and the combination of strips with different widths within one sample. The narrower the bark strips, the firmer the weaving bond and the greater that flexibility that could be created by weaving. Besides the weaving pattern and the strip width, the density also had a large influence on the behavior of the entire textile. During the tests, bark strips were woven both tightly and loosely (Figure 5.2.4). This affected the textile's overall strength and weight.

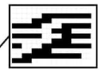
Furthermore, by combining different weaving patterns (Fig. 5.2.1D), it was possible to modulate strength and flexibility and to join bark textiles without permanent incorporation of other substances such as glue.

[19] Wenig, C., Dunlop, J. W., Hehemeyer-Cürten, J., Reppe, F. J., Horbelt, N., Krauthausen, K., ... & Eder, M. (2021). Advanced materials design based on waste wood and bark. *Philosophical Transactions of the Royal Society A*, *379*(2206), 20200345

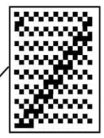




Twill weave with
changing directions
flexible in all directions

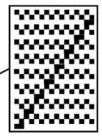


Twill weave loosly woven with two threads
rarely fixed



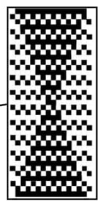
Twill weave with four threads densed woven in one direction
dense and flexible

Plain weave loose woven
dense and more flexible in one direction



Twill weave loose woven
flexible in one direction

Plain weave
dense



Plain weave combined with twill weave
partly flexible

Plain weave with different bark strip widths
thin bark strip create a more flexile and dense weave

Plain weave with one thread
loose connection

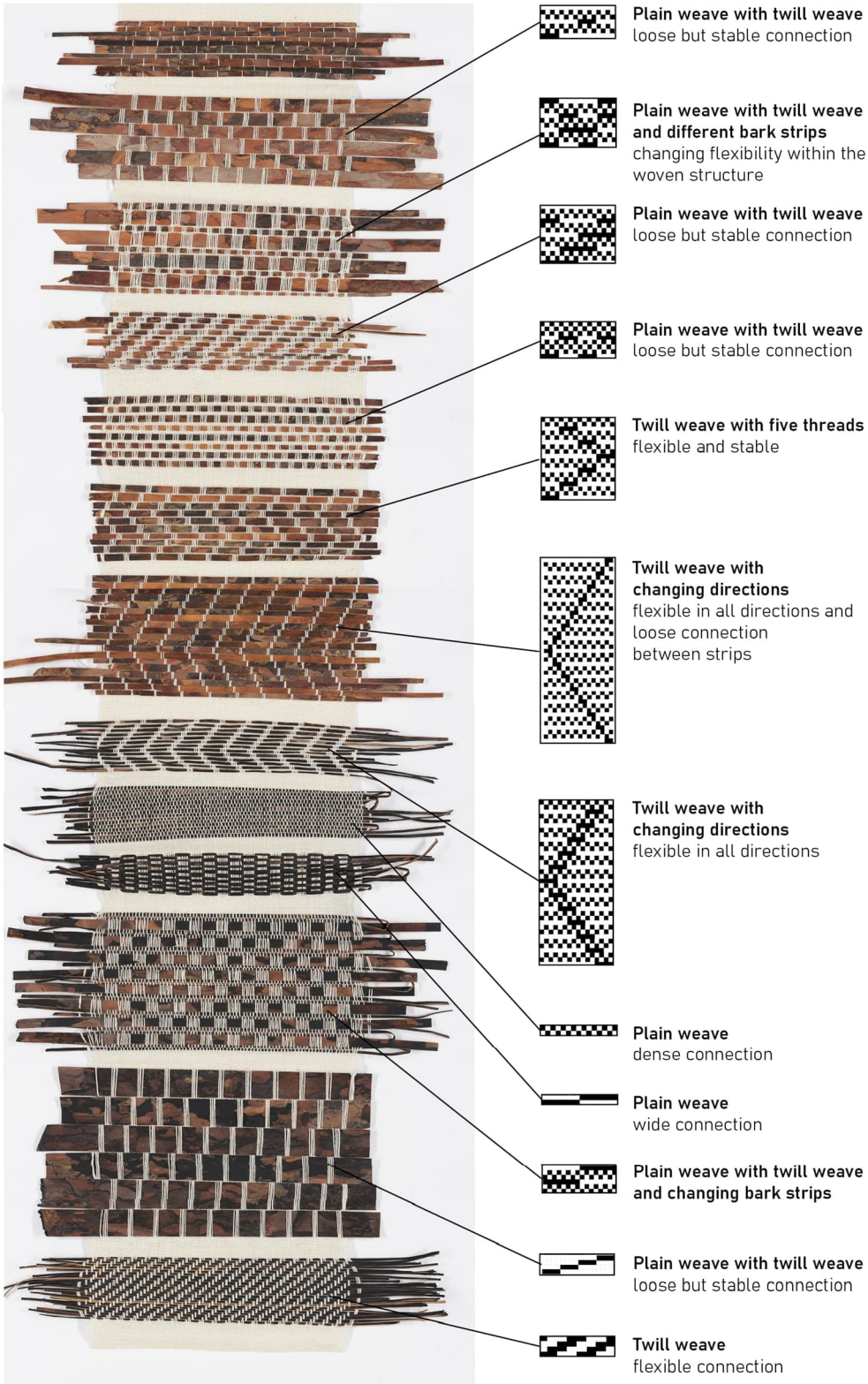


Figure 5.2.4: *Evaluation of the woven bark carpet. Different weaving pattern tested with bark.*

5.3 Woven Bark Jacket

In the first attempt to create a jacket, the aim was to cover as much of the body with tree bark as possible and to analyze the feeling of bark on skin. This was not the driving motivation for the second jacket, where the specific critical findings of the first prototype were addressed: the lack of flexibility (e.g. inability to move the arms) by processing a woven bark textile and the overall fit (e.g. feeling too tight or too loose in some areas) by applying an optimized cut. As before, special attention was paid to the processing possibilities and aesthetics.

5.3.1 Idea

Helpful findings for the production of the jacket were obtained from the weaving tests. The carpet (Figure 5.2.4) was initially an experiment to evaluate different types of weaving on their suitability for processing bark. However, compiling and documenting the test outcomes resulted in a pattern library on its own. This enabled selecting and combining weaving patterns according to their characteristics when creating the second jacket and following concepts.

For the woven bark jacket a twill weave was used (Figure 5.2.4). The weave gives the bark thread a degree of flexibility and tear resistance that is necessary for jackets.

5.3.2 Concept

Figure 5. 3.1: *Sketches of different cuts of outfits and shoes with bark cloth, image by Johanna Hehemeyer-Cürten.*

The main challenge was redesigning the sleeves, as these were to mainly consist of bark and be flexible at the same time. Additionally, the second prototype needed to be wider and more flexible in general. The front of the jacket was to consist of woven bark textile. The new jacket cut was based on that of a work wear jacket. The extra width in the back and wider arms allow for greater freedom of movement. The jacket can be adjusted at the waist with a ribbon to create an elegant silhouette.

5.3.3 Production

The cut of the jacket was determined by the width of the bark textile. The loom covered a width of 48 cm (Figure 5.3.3). The bark strips used were 2–3mm wide and tightly woven, ensuring high flexibility and tear resistance. The jacket cut was transferred to the textile using a template and then cut with scissors. The individual parts were sewn with a leather sewing machine. Unfortunately, weaving with the chosen parameters was very time-consuming, severely limiting the amount of material. Therefore, the sleeves and the front of the jacket were made of the bark textile, while the collar and the back had to be made of cotton.

***Figure 5.3.2**: Paper template for woven bark jacket. The template shows a front section of the second jacket. This was fixed to the fabric and then cut out.*

Figure 5.3.3: *Woven bark section of the second jacket. 2–3mm wide, flexibilized pine bark strips were tightly woven to create a flexible and tear-resistant textile.*

5.3.4 Result and Discussion

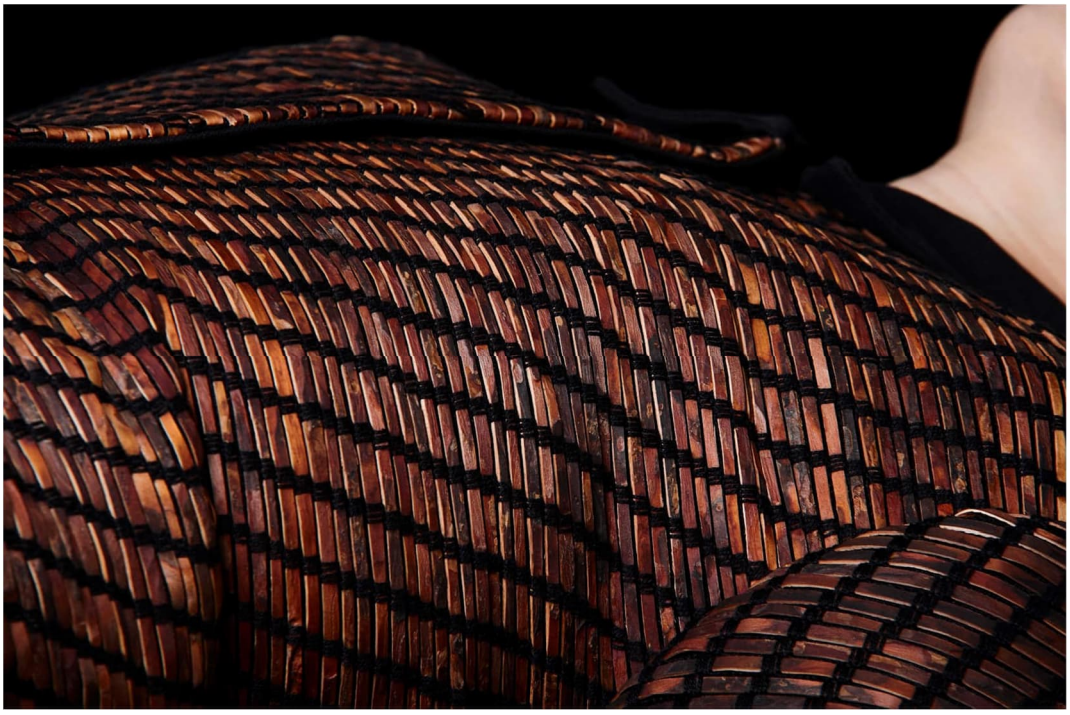


***Figure 5.3.4**: Close-up of finished woven bark jacket. Image copyright Studio Patrick Walter and Max Planck Institute of Colloids and Interfaces*

Despite the fact that the second jacket was not entirely made of bark, major improvements could be recorded in its design. Not only the flexibility increased significantly, but also the amount of processing possibilities. When producing the first bark jacket, it was almost impossible to avoid material defects, such as cracks or branch holes. By cutting the bark into strips, this could be circumvented. Weaving the bark strips ensured that tear resistance was significantly increased, and the wide range of possible weaving patterns created strong variability in textile properties (flexibility, weave tightness) and different surface aesthetics.

Comfort greatly increased since the jacket fitted more loosely. This is further supported by the chosen weaving pattern, where the bark is mostly on the outside and the cotton is on the inside neutralizing the wet and cold feeling on the skin. The sleeves were more flexible, though still not comparable to common fabric or leather.

***Figure 5.3.5**: Model with woven bark jacket and bark shoes. Image copyright Studio Patrick Walter and Max Planck Institute of Colloids and Interfaces*

Future explorations of processing woven bark textiles may even be more simplified. The possibility of using conventional sewing machines indicates that prospective garments can also be made with typical fashion material combinations, such as inner lining. The previously time-consuming process of cutting bark strips by hand can be improved by using a laser cutter. Furthermore, this was only a first attempt to produce larger amounts of bark textile. The possibilities of other processing techniques such as knitting, knotting or combinations with other materials have not been tested further in this work, but may be promising for further developments for the areas of fashion and domestic textiles.

5.3.5 Further Experiments: Bark Shoes

During the discussions on how tree bark could be used for applications on the body, the use of bark other than that of pine was also considered. The distinctive profile of oak and robinia strongly resembled the profile of hiking boots. Since shoe soles are responsible for a large amount of trash in the fashion industry (Muthu and Gardetti 2016) and many shoes are rarely worn due to fast changing consumer preferences, our idea was to try different scenarios using as much bark as possible in a shoe. For this we designed boots, high heels and sandals. The sole of all three pairs is made from robinia (dyed black in the boots). This choice was made, and not oak, because its bark is fiber free and can be easily processed by cutting and sanding. The robinia bark was cut with a hand saw and then hand sanded. Since prefabricated footbeds were used for these prototypes, it was difficult to build up in layers and use nails. Therefore, the bark was glued to the footbed with wood glue.

Overall, it turned out that the type of boots chosen was too complex to be made of bark entirely. It consisted of a prefabricated leather body which was partially combined with bark (Figure 5.3.6, right). The high heels were made of a robinia bark sole attached to a cork footbed and a felted wool upper shoe (Figure 5.3.6). The sandals consisted of a robinia bark sole, a cork footbed and a partially dyed, woven pine bark strap. They had a cooling effect on the skin due to the glycerol, which can be perceived as pleasant for a summer shoe (Figure 5.3.6, left).

Figure 5.3.6: *Shoes partly made out of bark: sandals with a robinia bark sole, a cork footbed and a partially dyed, woven pine bark strap (left), boots with bark soles and partially attached bark on the leather body (center), high heel made out of felt and a cork heel with a robinia bark sole(right). Image copyright Studio Patrick Walter and Max Planck Institute of Colloids and Inter- faces.*

5.4 Bark Sphere

Both jackets demonstrated the functional utilization of flexibilized bark on a rather limited surface. However, bark is a large-scale material and the question is if bark textile can be used more effectively by upscaling the dimensions of application. As we mentioned, bark was also used for temporary huts. In the case of the woven bark jacket as well as in the temporary hut, bark was used at larger scales, but their geometries were limited to what was available. For the early huts that are mentioned by Vitruvius in the first century BC, techniques of weaving were used and thus rudiments of geometric arrangements (Vitruvius 1867). This of course is a simple form, when branches with leaves were woven into a rough framework to create a roof or walls. But also more developed techniques are known, e.g. when mats or fabrics are woven and used as wall coverings, such as in the famous 'Caribbean hut' that Gottfried Semper documented in the nineteenth century in his theory of architecture (Semper 1863).[20]

5.4.1 Idea

The idea of the bark sphere was motivated by different sources. First, the weaving tests and the processing techniques of the bark jackets proved to be very successful and led to considering further fields of application outside of fashion. The slightly cool feeling on the skin also gave reason to not further explore objects with direct skin contact. Furthermore, the production of the mostly very narrow bark strips was very time-consuming and limited in output size by factors such as the loom width. The possibility of using pieces several meters long (the longest piece we peeled was 11m long) inspired thinking about applications on architectural scales.

[20] Wenig, C., Dunlop, J. W., Hehemeyer-Cürten, J., Reppe, F. J., Horbelt, N., Krauthausen, K., ... & Eder, M. (2021). Advanced materials design based on waste wood and bark. *Philosophical Transactions of the Royal Society A*, *379*(2206), 20200345

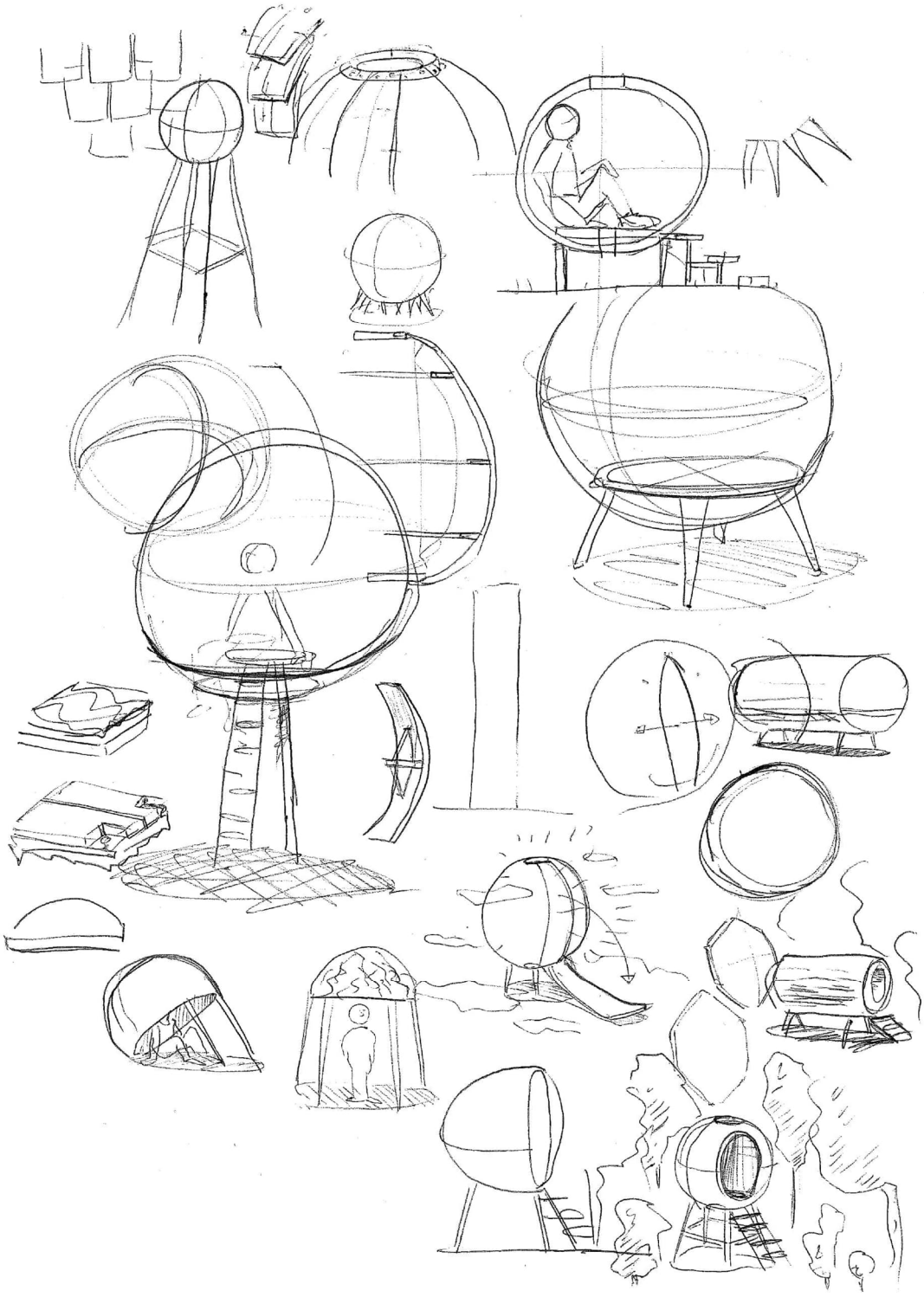


Figure 5.4.1: *Design sketches for bark sphere. First sketches for possible form of an installation on an architectural scale.*

5.4.2 Concept

Figure 5.4.2: *Inspiration for bark sphere concept. A hunting stand is a contemporary human-made hut found in forests.*

"The desire to create a clearly human-made object that integrates into the forest (Figure 5.4.2) and is not restricted to a certain geometry led to the idea of a walk-in sphere made of tree bark (Fig. 5.4.1). The use of tree bark with a woven structure allows use of the protective function of the bark in addition to controlling the object's stability by using different weaving patterns. The inherently difficult round shape of the sphere was selected to explore the limits of formability and to leave room for the viewer's own interpretation."[21] (Figure 5.4.1)

The aim for the bark sphere installation was to allow one or more people to stand inside and experience being completely surrounded by bark.

The sphere approaches the question about how, for example, the protective function of bark can be used here. Furthermore, it is not only a design for temporary architecture but also an exhibition object that encourages communication and exchange about bark as a possible future design and architectural material.

[21] Wenig, C., Dunlop, J. W., Hehemeyer-Cürten, J., Reppe, F. J., Horbelt, N., Krauthausen, K., ... & Eder, M. (2021). Advanced materials design based on waste wood and bark. *Philosophical Transactions of the Royal Society A*, *379*(2206), 20200345

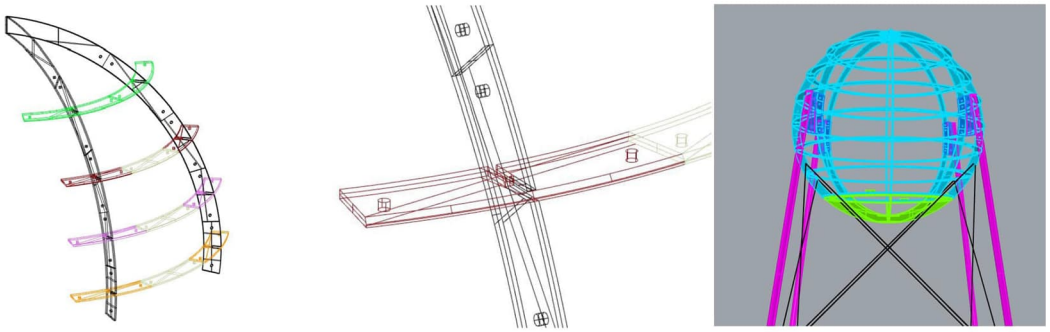


Figure 5.4.3: *3D model of the first construction ideas of the inner structure*

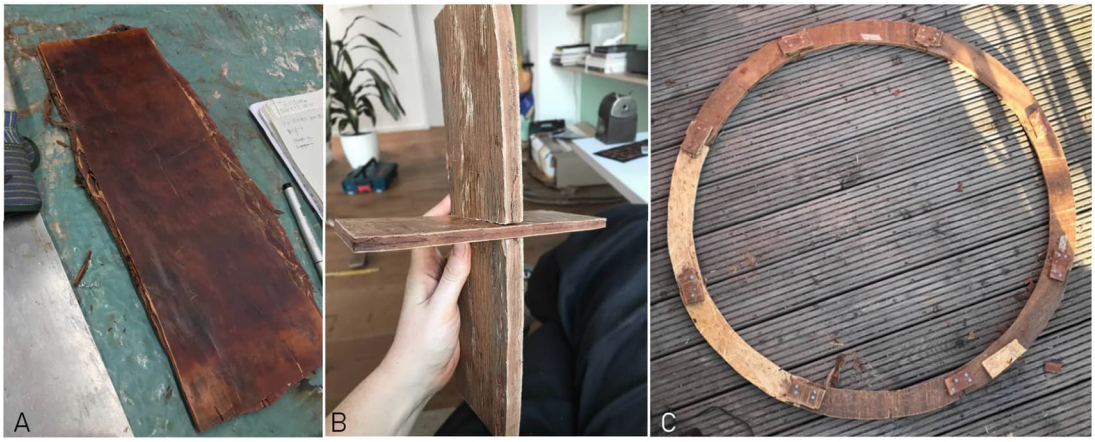


Figure 5.4.4*: Fabrication test of densified bark panels. Oak, larch and spruce panels were cut and tested for the inner structure of the sphere, (A) densified spruce panels, (B) first construction test made out of densified oak bark, (C) test ring for bark sphere partly made with densified and cut bark pieces and fiber board*

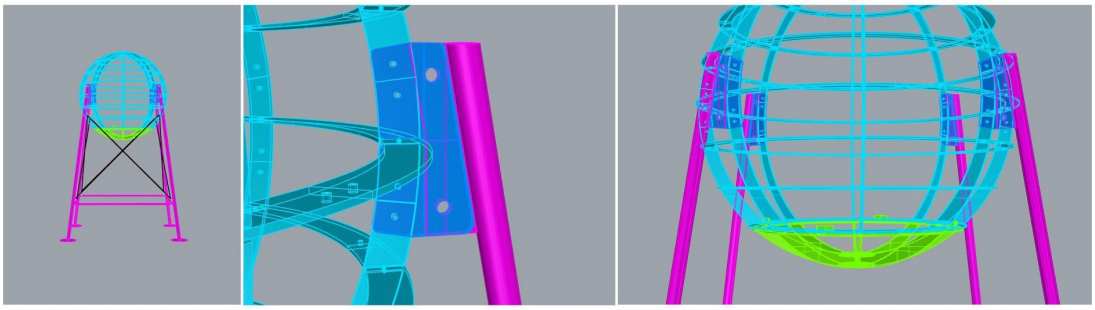


Figure 5.4.5: *Construction detail of the legs. The legs and bark sphere are connected with a self- designed hanger assembly/ suspension*

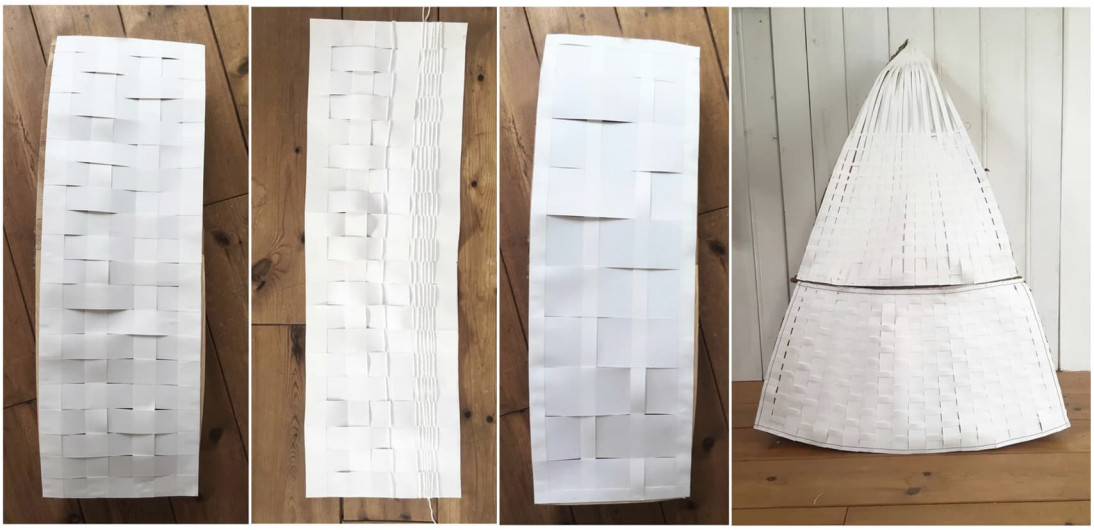


Figure 5.4.6: *Paper models used to create a three-dimensional surface. The aim was to create a shape with as little tree bark as possible. Paper models made by Johanna Hehemeyer-Cürten*

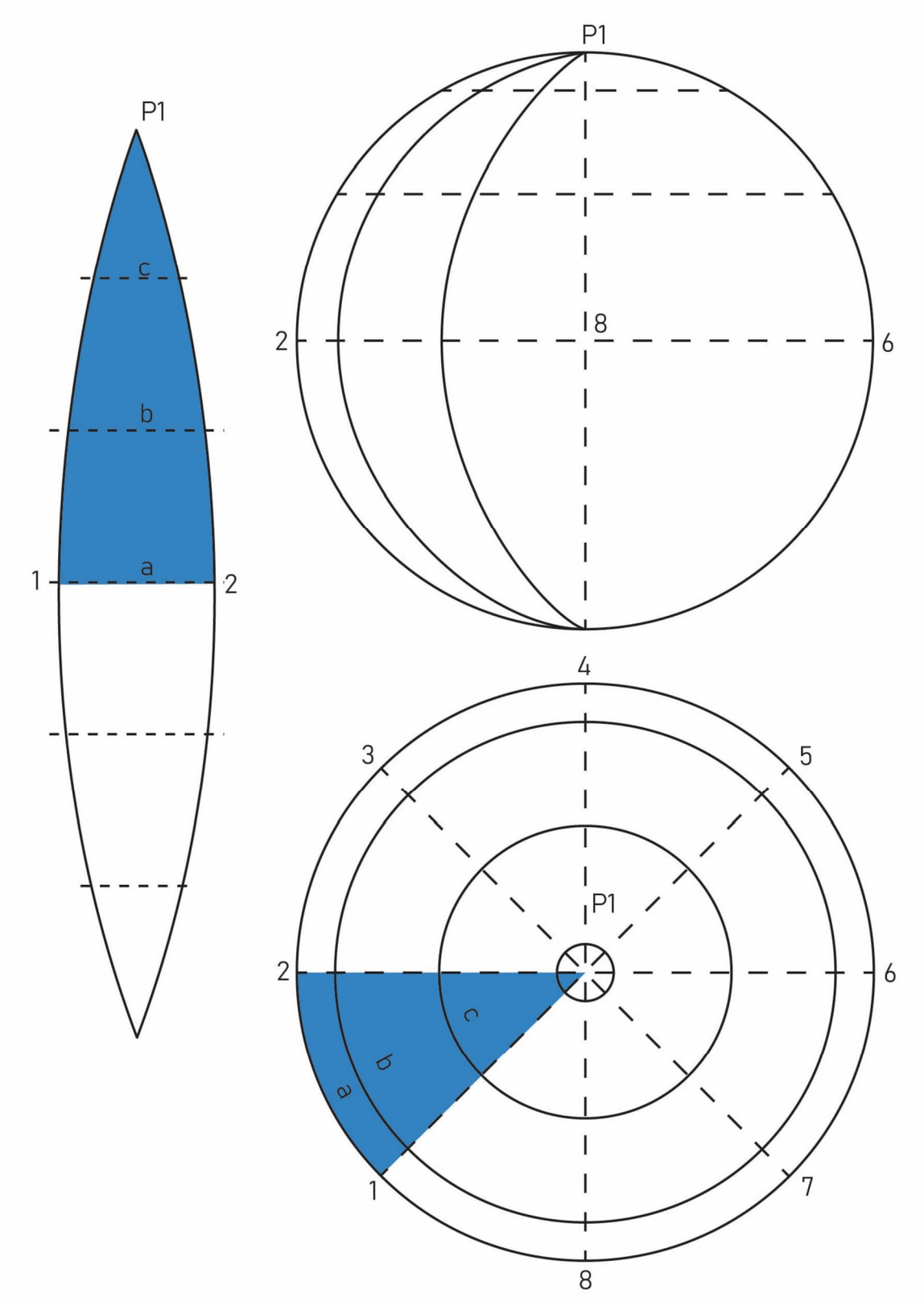


***Figure 5.4.7**: Unfolding model of the basic geometry of bark skin.*

The construction was divided into three parts: the inner structure (Figures 5.4.3, 4.4.4), the skin (Figures 5.4.6, 5.4.7) and the legs (Figure 5.4.5). With the goal of using as few non-bark materials as possible, various construction principles of spherical architectures were examined. The model finally chosen was the dome of the German Parliament, designed by the architect Norman Foster in 1995, in which the ring beams are supported in the main ribs (Figure 5.4.2). This basic principle made it possible to use panel material and to connect it with the application of a self-developed plug-in system, using a minimum number of screws (Figures 5.4.3, 5.4.4).

The densified bark panels (chapter 3.2.6) were considered for the inner structure material. The outlook was optimistic based on both the mechanical characteristics described in chapter 4.1.2.7 and the good processing capabilities with conventional jigsaws and hand routers (Figure 5.4.4) shown in initial testing. This suggested that framework components would be producible using CNC fabrication methods. However, due to the pandemic, which began in 2019, it was not possible to produce the required quantity of bark panels at ETH Zurich. Thus, individual processes, such as milling and plugging, were only tested, but not realized on a large scale.

With a diameter of 1.7m, the internal structure was divided into 8 rib arches and 10 transverse arches (Figure 5.4.3). In order to make the manufacturing of the internal structure more materially efficient, they were further divided and later connected with screws. The ribs and transverse arches of the sphere prototype were constructed as plug-in connections. Everything was designed in a way that allowed for easy replacement of the initially used material (hardboard and beech wood MDF) with bark panels later.

The legs were made of stainless steel, aiming at affecting the bark sphere as little as possible (Figure 5.4.5). For the skin of the sphere, different unfolding concepts for paper spheres were considered (Figure 5.4.6). The basic unwrapping consisted of eight strips derived from the principle in Figure 5.4.7. The eight parts were made of woven pine bark in a plain weave and fixed with rivets. The weave pattern was adjusted so that the entire surface of the sphere was made of bark and a tight weave bond was created. In order to keep the overall weight of the construction as low as possible, woven bark was only used where necessary. In the entire construction (framework, legs, skin), the use of adhesives was avoided.

5.4.3 Production

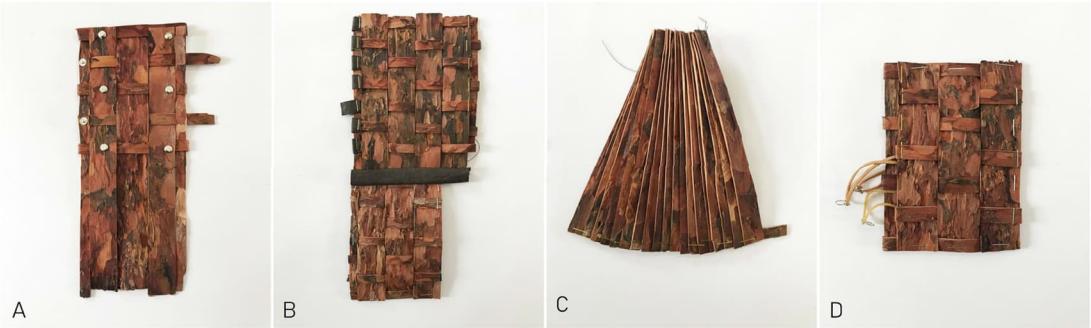


Figure 5.4.8: *Connection test with staples, rivets and sewing and rubber bands. The aim was to connect single pieces of the woven skin in a reversible way. Test was made together Johanna Hehemeyer-Cürten.*

Figure 5.4.9: *Final connection of bark skin to the inner structure. Closing the sphere. The cap is hung on the body of the sphere with a bayonet lock.*

Figure 5.4.10: *Closed bark sphere with 4m legs between pine trees. Image copyright Alexander Magerl and Max Planck Institute of Colloids and Interfaces.*

The production of the internal framework was outsourced to the Oberlin Berufsbildungswerk Potsdam. They milled the individual parts, which were assembled into the complete framework at the MPI. The legs were manufactured in MPI's metal workshop. The bark “skin” was made from self-peeled pine bark that had been flexibilized in the laboratory. With the help of templates, the bark was cut into strips which were woven by hand afterwards. The individual pieces were sewn with a sewing machine and riveted at the outer edges. The eight large strips of bark were reversibly tied to the framework with rubber bands and hooks (Figure 5.4.8, d). After the skin was attached, the bark sphere was screwed to the legs using four metal brackets specifically manufactured for this purpose.

5.4.4 Result and Discussion

Figure 5.4.11: *Johanna entering the bark sphere, located in 4m height in a forest. The legs of the sphere were adjustable in height, allowing for an approximate overall height of between 2.50m and 4m. At a height of 4m, the sphere could be interpreted as a high stand or nest and could only be entered with a ladder. Image copyright Alexander Magerl and Max Planck Institute of Colloids and Interfaces.*

Figure 5.4.12: *The closed sphere with 2.5m long legs. Image copyright Alexander Magerl and Max Planck Institute of Colloids and Interfaces.*

Figure 5.4.13: *There is a strong smell of pine in the bark sphere even months after construction.*

The first prototype of the sphere was an accessible structure that enabled the experience of being inside tree bark (Figures 5.4.11, 5.4.13). The bark was translucent in thin parts, creating different lighting effects inside the installation. The smell was very resinous and the acoustics were affected by the material and the form. Additionally, the abstract round shape allowed every visitor to make their own individual associations.

Whether the bark is suitable for temporary structures can only be determined in a long-term test. Records assume that bark huts last between one season (Ast 2011) and many years. Some examples, such as the "Eremitag" or "Borkenküche" at the Sans Souci Palace Park in Potsdam, show that bark buildings can also stay intact for over 10 years.

Overall, bark has shown diverse material potential, based on design criteria such as a variety of shapes and processing possibilities. However, the processing techniques and objects explored so far have been very time-consuming. Building on the knowledge gained in these experiments, two directions can be interesting for further research on the utilization of tree bark: considering applications outside of fashion and architecture and exploring options besides weaving to support the bark mechanically.

5.5 Further Considerations: Theory of Boundary Objects

As described in the previous chapter, the boundaries between science and design are fluid. Objects that emerge show influences from both professions. These can be seen as boundary objects. Since the present work is based on the interaction between science and design, the objects created here can also be considered boundary objects. In the following paragraphs, the main idea of this theory and its characteristics are explained with reference to this thesis.

Boundary objects are objects that have different meanings in different social worlds, while their structure is common enough to more than one world to make them a key in developing and maintaining coherence across intersecting social worlds. They can function as a tool that allows different groups to work together without consensus. This sociological concept was introduced by Susan Leigh Star and James R. Griesemer (1989), using the example of a natural history museum in the US state of California (Star and Griesemer 1989). In this study, the boundary object is the collection in the field of ornithology. Birds are studied by scientists as well as amateur observers. While the amateur observers report on different bird species and

their abundance, the scientific community uses the collected data for other aspects. The collected findings are presented in the museum, thus involving another group, the museum visitors.

In this example, ornithology is the building bridge between different groups. The science of birds is not a barrier here, it is a connection. With that the museum becomes a shared space where common tools facilitate cooperative actions (Star and Griesemer 1989).

The outcomes described in chapter 5 can also be seen as boundary objects. An important criterion for boundary objects is flexibility and relevance for all groups at the same time. The approach of this thesis can be seen as an example of this. The three prototypes have the potential to be relevant for very different recipients: scientists and designers, as well as ultimately consumers (jackets) and installation visitors (sphere). Furthermore, much of this thesis starts with design experiments followed by combination with and understanding of scientific experiments. One example of this is flexibilized bark. Motivated to obtain a flexible textile-like state of tree bark for the purpose of creating design objects, solutions with glycerol and water were used. As shown above (3.1.3), not all bark can be flexibilized, which is due to the structure of the individual species and thus raises further scientific questions.

Another vital aspect, also studied by Star and Griesemer, is the importance of communication. In an iterative process between design and science, a reliance on a common language is important. The authors describe the dynamic as a cross functional group that creates a common language despite their differences by using the same tools by defining them differently (Star and Griesemer 1989). Within this thesis, the material properties of plants, such as anisotropy, were discussed in design contexts, and inherent material properties (fiber direction) were used to address challenges by means of design (weaving).

An interpretive flexibility of the created outcome encourages development of design objects that, in their specific meaning (e.g. fashion), are no longer connected to science. Being developed on the basis of scientific data, they have the potential to build a new bridge towards society and the use of bark in everyday objects. They can encourage conversations or further experimentation without demarcation of any research field (e.g. which characteristics of local bark are not discovered, which properties of bark can be used for other applications). Through the interaction of fundamental research, design experiments and the involvement of an extended audience (through design scenarios), it would be possible to take these concepts and ideas further.

According to the principle defined by Star and Griesemer, boundary objects gain significance when they are applied to solve problems. With the bark sphere, an attempt was made to use tree bark for architectural applications. The building sector is one of the main drivers of climate change (Cellura et al. 2018). The durability of many buildings is not analogous to that of the materials used. While temporary buildings are being used from one season to only a few years, incorporated materials, such as plastics, outlast this period significantly.

With the planning of an accessible installation made of tree bark, the size of the application was scaled up and thus first possibilities for the architectural temporary use of tree bark were investigated. The finished installation (described in 5.4) again led to new scientific questions. To name only a few: Why does the bark still smell so strongly of forest even though the peeling process took place over one year ago? How does the material affect the acoustics?

The last boundary objects conditions I want to discuss are questions of scaling. By standardizing or limiting the boundary objects to a certain size or scope, overall structures are broken down. The desire to fulfill a certain standard would force the objects into an intermediate category, thereby forcing their whole meaning and infrastructure to be shaped by the conventions of a community. Star states that the solving of tensions between fields is the death of boundary objects. When terms are defined and structured, and results are fixed, the boundary object becomes a standard. With regard to the research conducted in the context of this thesis, a boundary object would cease existing as soon as an agreed outcome from both worlds (science and design) emerges. However, as every realization raises new questions and many potentially new (creative) areas open up, it is uncertain when an object is "completed" and no longer functions as a boundary object. This raises the question whether, in this case, the bark spheres would transcend the goal of application and thus fully integrated into the discipline of design, or whether they would still serve as a scientific object.

The theory of Star and Griesemer has been mainly integrated into institutions, teams and organizational structures. This work relates this theory to material-specific knowledge and applications. While Star predict the death of boundary objects as the collaboration progresses, this project assumes that the creation of interdisciplinary work could be transformed back into boundary objects with the help of other professions. The outcome of this thesis is therefore not finished design applications, but a demonstration of collected knowledge and possible future scenarios.

6 Discussion: Manufacturing and Design Strategies for Bark

Current production methods substantially contribute to climate change, consume a large amount of resources, and contribute to biodiversity reduction (Walcher and Leube 2017). The prevailing “throwaway society” is characterized by the linear "cradle-to-grave" model. Resources are used once for production and then discarded (Walcher and Leube 2017). The transformation of the current linear economy into a circular economy is part of the European Commission's environmental plan. The aim is to minimize environmental pollution and resource consumption (Walcher and Leube 2017). The basis for the circular economy principle is the theory that in a world with finite resources, only production processes with a real material cycle can be continued indefinitely (McDonough and Braungart 2010). This principle is inspired by nature, since there is no waste in nature. The goal of circular economy is to achieve cascading uses without waste (zero waste) and without emissions (zero emission). As good as this theoretical model may sound; it has been heavily criticized in recent years (Corvellec, Stowell et al. 2021). The term "circular" can be misleading, as there is no possibility of one hundred percent recycling at all product (lifetime) stages. Additionally “circular theory” ignores limitations of reprocessing technologies, the complexity of waste and social and economic issues (Corvellec, Stowell et al. 2021).

Another approach, for example that established in the German government's Climate Protection Plan 2050, is the cascade use of renewable raw materials (der Forstwirtschaft 2016). Cascade utilization means that renewable raw material is first used (several times) as a material and then – as waste – as an energy source (Arnold, Geibler et al. 2009). The principle of cascade utilization has been noted for several years as an approach to increase resource efficiency (Fehrenbach, Köppen et al. 2017). Regenerative raw materials – biological-based materials – are particularly in focus here.

Biomaterials are renewable resources associated with land use and thus also become subject to scarcity (Fehrenbach, Köppen et al. 2017). Consequently, it is also important here to use biomaterials as long, as often and as efficiently as possible, and to only to use them for energy production at the end of their product lifecycle (Fehrenbach, Köppen et al. 2017).

The material that most likely comes into most peoples' minds as a renewable resource is wood. Around 80 million cubic meters of wood were harvested in Germany in 2020 (Destatis 2021). This represents a strong increase in timber harvesting over the past years. In 2020, it was around 16.8 percent higher than the previous peak of 68.9 million cubic meters from

2019. The reason for the increased felling is ongoing forest damage due to drought and heat, which favors insect infestation (Bocksch 2021). Rising demand from Germany and other countries further complicates the growing beetle infestation (Destatis 2021). Consequently, it is reasonable to think that as much wood as possible should be used from healthy, already harvested trees.

Unfortunately, the reality is different: 44% of the wood taken from the German forests is burned without further use – more than half of it directly as energy wood, the rest as sawmill by-products (Mantau 2012). Only 19% of waste wood is recycled as additives for particleboards (Mantau 2012).

In order to extend the use of wood as well as other materials, it is essential that cascade utilization is included in the development of process chains. This has to be included in the development of products in order to enable an economic value chain (Fehrenbach, Köppen et al. 2017). This should be applied to bark as well as wood. The proportion of bark in tree trunks is in the range of ~10–20% by volume (Harkin 1971). Based on this amount of bark in one tree, approximately 4 million m^3 of "leftover" bark is produced annually in German sawmills (Wollenberg and Warnecke 2005). Given these quantities alone, the long-term utilization of bark should be a goal in order to increase the utilization efficiency of the raw material: wood/trees.

The following figure (Figure 6.1) and text outline possible optimization in the production process, expanding processing possibilities and future application scenarios. Figure 6.1 shows established implementation procedures, processing procedures and application options developed and tested in this thesis, and the speculative scenarios based on these results.

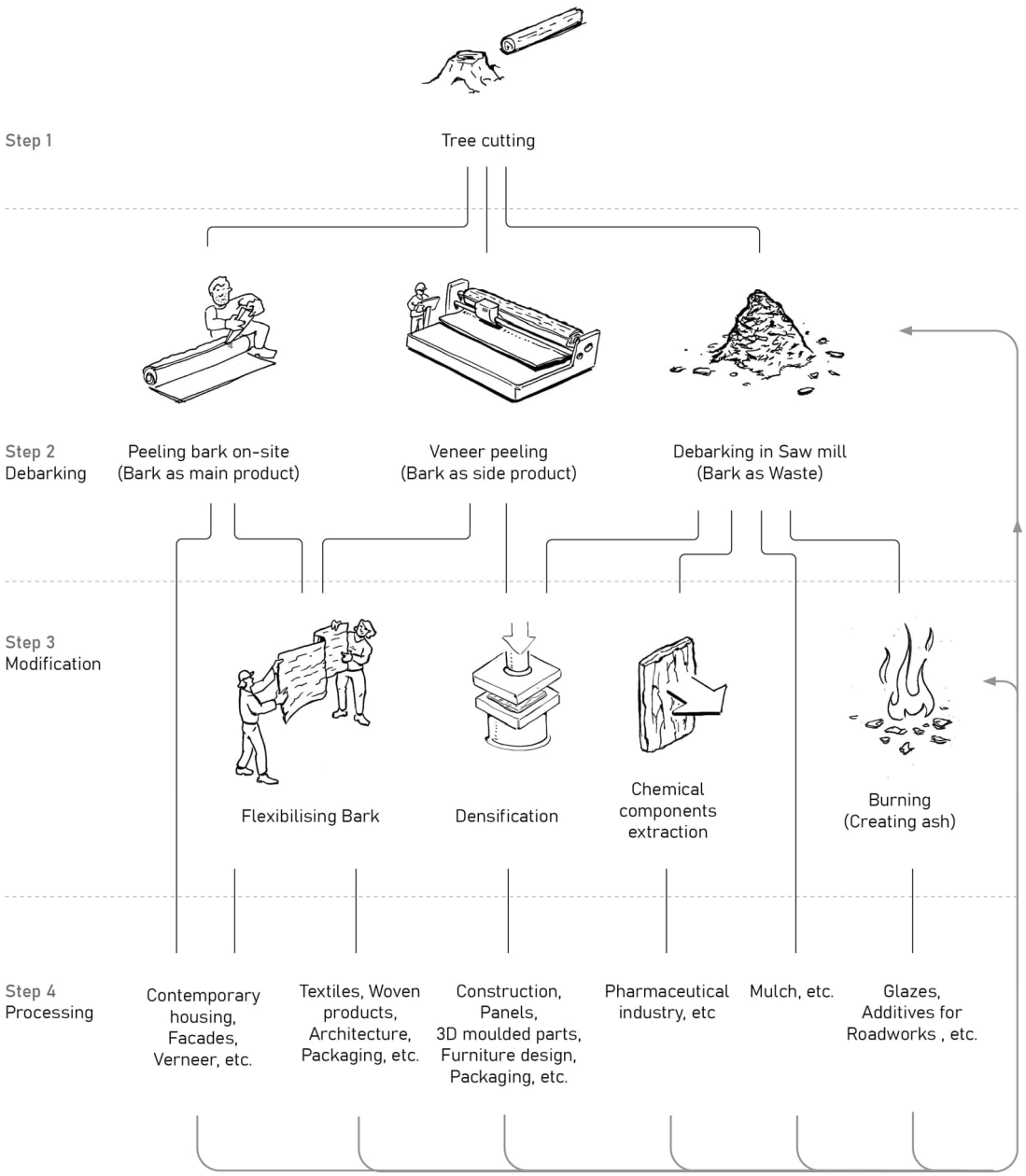


Figure 6.1 *Cascade use of bark. The figure shows established and newly developed procedures, applications, and speculative scenarios based on the results of the thesis. Illustration by Florian Weisz.*

Step 1: The process starts with the felling of the tree, by chainsaw or by harvester. Depending on the purpose, individual trees or several trees are felled, of only one species of tree or multiple ones. The felling season plays a vital role in the process: In late spring/summer, the tree is active and the bark can be peeled in large contiguous pieces with little physical effort. In fall/winter, the tree is dormant and peeling requires a greater amount of force and the bark more likely to break apart. These factors determine the state and the characteristics of the bark after felling and influences the way it can be processed and used later on.

Step 2: In the next step, the bark is removed from the log. This can be done either on site or in wood processing facilities. If bark is peeled on site, the peeling process is done manually. The bark for the processing methods and applications developed in this work was peeled in large contiguous pieces. May and June are described as the best time for peeling in historical records, due to the high amount of water inside the tree (Ast 2011). It can be assumed that a high amount of water helps to loosen the connection between the xylem and cambium. Peeling by hand is a tiring and time-consuming task (Çağlar 2021), but it enables the collection of bark in large dimensions and without major defects. This bark has different applications (see above), making a multi-stage cascade utilization possible. Due to the high effort, however, it is unlikely to be replicable at a larger scale.

If the tree is peeled in a wood processing factory, the quality of bark strongly depends on the technology of debarking (Baroth 2005). Current debarking methods shred the bark into small pieces. In this scenario, the bark is seen as a waste material. However, using established technologies such as veneer peeling is conceivable for bark peeling. Here, it would take place as a production step before the actual veneer peeling. By fixing the log in the peeling machine and peeling it in one circular move, long, contiguous pieces can be obtained. Ideally, that part of the log would be very symmetrical and knot-free; based on the characteristics of living trees described above, trees felled in spring/summer would be best suitable for peeling. In this scenario, bark would be a high-quality material.

Step 3: After harvesting and drying, the processing starts (in the following, it will be referred to as modification). If bark is obtained in large contiguous pieces, it can be used untreated. This type of application has been historically shown to be particularly suitable for building temporary huts (roofs and walls) in different regions (see chapter 2.3; (Paukner 1991), where tree bark's biological protective function is used.

Pine mirror bark can be further treated with glycerol. This leads to flexibilization of pine bark (see chapter 3.1.3).

Due to the internal bonding properties of the bark, it is possible to press these bark species into large panels (see chapter 3.1.2). The hot pressing method (see chapter 2.2.3.1) is very well established in the wood industry and therefore offers a promising option for implementation.

If the bark is not removed from the tree trunk in large pieces, but in individual small particles, it is also possible to use these for the production of panels and boards (see chapter 2.2.3.1; Pasztory, Mohácsiné et al. 2016). This form of bark utilization has been studied extensively and is already being applied in an economically feasible form. Certain types of bark are also used for gardening and horticulture (pine, spruce; Whiting, Tolan et al. 2003). One application in which the shape of the bark pieces does not play a role is in the extraction of extractives (Bianchi 2016, Harkin 1971). However, the majority of the bark is burned directly and thus has no further use (FAO 2020). This is a single-stage cascade utilization and should no longer be directly targeted in sustainable scenarios.

Applications such as the use of large untreated pieces of bark, the bark mulch and the densified bark panels have the advantage of not being modified with chemicals. These applications are compatible with the goals of the cradle-to-cradle movement.

This concept implies that all materials can be reused by classifying them into two cycles, the natural and the technical (McDonough and Braungart 2010). Tree bark would be included in the natural cycle. The materials are compostable and can thus provide new nutrients for nature (McDonough and Braungart 2010).

In this sustainability theory, the bark panels are a good example of a long-lasting application that can later be returned to a natural cycle. Due to the addition of adhesives, most current conventional panels are no longer fully recyclable and would therefore not fulfill the cradle-to-cradle principle; it is debatable whether this applies to flexibilized tree bark.

It is very important that the cycles remain clearly separated from each other. If the natural cycle is contaminated by the technical one, toxins, carcinogens or mutagens can be metabolized by and injure organisms. Conversely, natural nutrients must not be allowed to enter the technical cycle, as they not only reduce the quality of the substances but also complicate recycling (McDonough and Braungart 2010). Food grade glycerol is used to make the pine bark more flexible. Glycerol is considered to not pose a health risk and can be washed out of the bark. Its classification as a technical material is therefore questionable.

Modifications can be reiterated depending on the type of processing. Flexibilized pine bark can be used in a multi-step cascade: whether as sheet material or after being processed into a textile (through weaving), both options still allow washing out the glycerol, pressing the bark afterwards and, lastly, burning it. In the case of the densified bark panels, modification of the structure and geometry is possible, for example, by processing (by cutting, milling, sandblasting) for design applications or by shredding. Another later modification step can include chemical changes by removing extracts or burning.

Step 4: The last step exemplary shows the manufacturing/processing of (treated) bark for different purposes. In the case of bark mulch or ash, this step can take place directly after peeling.

One additional use is bark ash as a substitute in the porcelain glazing process (see chapter 4.3). A main chemical component of bark ash is calcium, known as a flux substitute in the glazing process (Lehnhäuser 1985). Other mineral constituents of bark could be used as a substitute for glaze colors. In conventional glazing, metal oxides create colorful glazes. Some of the best known metal oxides for glazes are cobalt blue, green made by chrome oxide and brown tones made by manganese and iron (Matthes 1985). In the firing process, metal oxides react with the other components of the glaze and create a non-toxic glass film (Lehnhäuser 1985). By using bark ash, a local leftover material could be used instead of metal oxides.

Depending on the firing procedures, the same glaze could appear in different colors. In general, it is difficult to achieve the same results in two different fire procedures. This problem is not a special case for bark glazes; it is also known from conventional ceramic production. There is therefore uncertainty in the large-scale industrial (re-)production of ceramics. A design concept where the color is not the main focus is therefore needed. The variability of the color could also be seen as a chance to create a unique object. By using ash, even the final bark leftovers obtain a new purpose. Bark ash is used as an additive for ceramic glazes and can become part of a long-lasting consumer product. If the ceramic object is broken, it can still be pulverized and used as an additive in road construction.

Applications that use non-toxic types of chemical modification or no chemical modification at all are preferable for all sustainable material scenarios, such as the flexibilized bark and bark panels. Both semi-finished products can be further modified in their shapes but also improved in their mechanical properties by processes like cutting, weaving and milling.

Bark panels and 3D bark panel may serve in applications without water contact (e.g. interior design). That the surface smoothness of panels is comparable with sanded wood surfaces facilitates applications in furniture as well as in packaging. Applications and products created in this way can be further processed into other applications after their period of use. In the last step, decomposition or incineration is possible.

As described, bark has a great potential for long cascade utilization. By adapting existing technologies such as weaving or veneer peeling, new applications for tree bark can be developed. This raises the question of whether bark could even replace existing materials. This includes not only plastics, the vast use of which in all fields of life is widely discussed and criticized but also biological-based materials like leather; designers and researchers currently looking for replacement materials of non-animal origin (Rathinamoorthy and Kiruba 2020). In the following paragraph, how and where bark can contribute to leather alternatives is discussed.

The feel and color of the glycerol-treated (flexible) bark is similar to leather. Leather is one of the oldest materials in human history (Kite and Thomson 2006). It creates wealth and culture and yet environmental problems at the same time. The variety of leather (in their properties and appearance) makes it impossible to replace all leather applications with the same material. Its main applications are in the lifestyle industry, in home furnishing and automobile upholstery. According to the current forecast, leather consumption will continue to grow (UNIDO 2010). Quality leather products, even with constant use, can last more than 100 years. In this context, leather may be called a highly sustainable material (Muthu and Gardetti 2016). However, if we consider how fast society and consumer products are changing – from functional and aesthetic points of view, most leather goods last a much shorter time. In the current fashion industry, footwear production is one of the largest sectors in the leather industry. By 2007, it had reached 16 billion pairs of shoes annually (UNIDO 2010). The use of bark might be a better fit for these fashion lifecycles. First attempts at replacements have been attempted for different kinds of shoes (Chapter 5.3.5). By applying minor (e.g. densification or glycerol treatment) modification or even none at all, there are numerous bark applications in fashion and home furnishing; after use, the bark could be returned into the material cycle.

In addition to its short lifespan, the production of leather is also critical. Each treatment applied to leather makes it less of a natural product. To achieve durability, animal skins are usually tanned with chromium salts. Several reports claim that tanning is one of the highest pollution-creating industries due to powerful toxic chemicals (Muthu and Gardetti 2016). Therefore, leather production is harmful for the environment, for people working in tanneries and

for consumers (Muthu 2020). The flexibilization process of bark uses water and glycerol (3.1.3). After this treatment, there is no additional chemical modification involved. To change the mechanical behavior of flexible bark, techniques like weaving could be used. The treatment and processing of flexibilized pine bark can be done without any toxins.

Furthermore, ongoing ethical discussions regarding the use of animals in the fashion and lifestyle industries has created the need for vegan leather alternatives (Muthu and Gardetti 2016). The demand for materials that look and (mechanically) behave similar is thus also challenging leather production. For example, vegan consumers refrain from buying animal-based leather but might use polyurethane- or polyvinylchloride-based "fake leather" or other plastic alternatives. Polyurethane and polyvinyl chloride belong to the group of mass plastics and are based on petroleum and therefore also have a large environmental impact (Muthu 2020).

Overall, there are leather products which could be replaced with bark, especially short-lived leather objects like fashion accessories and bags but also shoes and furniture. This would not stop the creation of short living objects but it would contribute to a better environment through its less intensive treatment process before production and easier recycling after consumer use.

The option to source bark regionally as well as the natural size of trees, enabling production of small- and large-scale (architectural) objects alike, shows tree bark to be a high-quality material of the future economy.

7 Summary and Conclusion

In the framework of this thesis, new approaches to using the bark of locally grown trees were developed. As the natural variability within and between types of bark is very high and the material is often considered as waste of the timber industry, there has been limited knowledge related to its properties.

To better understand bark as a material and to add value to its lifecycle, two biological characteristics of the material required consideration: firstly, the time of harvest (young vs old bark) and secondly, the structural and compositional differences between the investigated species. This challenge was addressed by the combined utilization of basic science tools and design experiments.

In the process of harvesting, large pieces of bark were peeled and their natural structures were preserved. To keep tree bark within a material cycle, bark was processed into products without incorporating additional substances. “Pure” one-component products do not require separation of components after their consumer lifetime. Even if the structure of the raw material is altered, the basic building blocks remain the same and hence can still be easily used for subsequent processing, such as in the extraction of chemicals or fibers, or as a fuel for energy production.

The procedure developed here for densified bark panels shows that the production of adhesive-free bark panels is possible. Bending stiffness and strength and transverse tensile strength similar to wood-based-panels such as particle boards were achieved. The densification process allows the use of bark (panels) in mass applications. Bark panels can be processed with conventional woodworking methods such as cutting, milling or even pressing into 3D shapes, suggesting potential use as a material in the furniture industry.

One local Brandenburg tree type is pine. A special characteristic of pine is the strong difference in internal structures between matured bark and young mirror bark. By using glycerol, which is plant based and widely used in the food and cosmetic industry, mirror pine bark was kept in a flexible state. The tensile strain of mirror pine bark can be increased by this procedure. Certain unwanted mechanical properties can be adjusted on a larger length scale by using various processing techniques. For example, flexibilized pine bark can be woven into different patterns, showing its potential as a flexible, textile-like material. The applications of tree bark are thus extended into the sector fashion (jackets, shoes) and temporary architecture (large-area woven fabrics for installations).

Large quantities of tree bark are burnt in sawmills, producing large amounts of ash. Using ICP, the different chemical compositions of the individual types of tree bark ash were measured. The ash was used to create colored glazes for porcelain. The outcome provides glazes for ceramic products which prolong the lifecycle of bark ash.

A different approach to prevailing design methods is the integration of material into the design process right from the start. Often a design is developed first and the material is selected at a later stage of product development. If material selection were to be implicated immediately, it would lead to material-based design. Through this lens, the natural properties of tree bark are not classified as weaknesses to be overcome, but as material boundaries to be considered in the design. The use of bark in such high-value applications – like in fashion or in other product design – allows us to use resources in a more sustainable way by bestowing them with a longer lifecycle. Therefore, cooperation between different disciplines is essential to understand the full potential of bark (and any other material).

Through a symbiosis of science, humanities and design, existing practices and knowledge can be combined, inspire each other's (practical) work and lead to new research questions. This could lead to a revolution in the use of increasingly precious raw materials, which are currently treated as industrial waste.

The historic boundaries among different research disciplines should be dissolved and an environment for co-creation and coexistence should be established. Through building a research environment, where diversity is appreciated and a variety of research fields are considered beneficial, we can create sustainable research.

List of Publications

The central findings of this thesis are presented in two publications. Both publications were implied under permission of the journal in chapters 2-5 and if necessary further developed. The relevant passages are marked in the thesis.

Paper I.

Wenig C, Dunlop JWC, Hehemeyer-Cürten JL, Reppe FJ, Horbelt N, Krauthausen K, Fratzl P, Eder M (2021) "Advanced materials design based on waste wood and bark." *Philosophical Transactions of the Royal Society"* A 379.2206 (2021): 20200345.

Author Contributions

In this publication, the role of waste wood and bark in biological context and in cultural matter as well the result of my own work are discussed, together with all co-authors. I was involved in the experimental design, material collection and have performed a series of experiments together with my colleagues Nils Horbelt, Friedrich Reppe und Michaela Eder on the performed structural and mechanical characterization of bark. I have performed a series of experiments to create the flexibilized pine bark needed for the design experiments. Together with Johanna Hehemeyer-Cürten, I have designed and created the woven bark jacket and the bark sphere. Michaela Eder and John Dunlop drafted the manuscript, Karin Krauthausen contributed with knowledge and text related to historical use of bark and wood materials. The figures were hand-drawn and/or created by John Dunlop, Michaela Eder, Friedrich Reppe and me. Peter Fratzl initiated the multidisciplinary collaboration and was involved in numerous scientific discussions related to the project. All authors discussed, commented, wrote text passages and approved the manuscript.

In Preparation Paper 2.

Wenig C, Reppe F, Horbelt N, Spener J, Berendt F, Cremer T, Frey M, Burgert I, Eder M „Adhesives free bark panels: an alternative application for a waste material"

Author Contributions

This publication is based on the production and analyzation of densified bark panels with additional substances, with the support of all co-authors. I planned the densification procedure and developed the parameters until the first successful panels. Marion Frey did first densification tests with wet bark samples. I was involved in the material collection and have performed a series of experiments for the 3D dimensional densification and further processing of the panels. My colleague Friedrich Reppe has performed surface characterization of bark panels, density measurements on raw bark and panels and swelling and shrinkage measurement of raw bark and densified bark panels. First 3-Pointbending test were performed by Nils Horbelt. In the context of his, bachelor thesis, Jaromir Spener examined influence of the moisture content of raw bark on the mechanical properties of the bark panels. For this purpose, he performed swelling and shrinkage tests of the densified bark panels as well as transverse tensile tests. Ferreol Berendt Tobias Cremer, Michaela Eder and myself, supervised this bachelor thesis. Ingo Burgert kindly provided the hot press of the ETH Zurich and was involved in numerous scientific discussions. Moreover, I have participated in writing the manuscript together with Micaela Eder and prepared the figures.

List of Abbreviations

CESA	cellulose synthase units
CNC	cellulose nanocrystals
CNCmilling	computerized numerical control milling
d.wt.	dry weight
FP1	native bark samples and bark panels
FP2	bark panels with different moisture contents
HCl	hydrochloric acid
HDF	hard-density fibreboard
ICP	inductively Coupled Plasma
µCT	micro-computed tomography
MDF	medium-density fibreboard
MFA	micro fibril angle
OSB	oriented strand board
sprRH	relative humidity
XRD	X-ray diffraction

List of Figures

List of Tables

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