FIBRE-BASED GRADIENT STRUCTURES

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ABSTRACT

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Plastic remains a widely used packaging material despite its environmental drawbacks. Although its sustainability has improved over time, the industry continues to shift towards plastic-free alternatives. Moulded fibre products (MFPs) offer a promising solution due to their versatility and eco-friendliness. However, their current use is mostly limited to low-grade packaging, and enhancements are needed to broaden their appeal. The aim of this thesis was to explore the use of fibre gradient structures combining softwood (SW) and hardwood (HW) fibres, and whether they can help maintain strength while reducing raw material costs, thereby enhancing the viability of MFPs as sustainable alternative to conventional plastic-based packaging.

Two types of gradient structures were studied: thickness gradients and surface gradients. Hand sheets were prepared with five different SW/HW ratios and two fibre orientations (isotropic and anisotropic). Hot pressing trials were also conducted on thickness gradient sheets to simulate moulding conditions. Material performance was evaluated through porosity and various strength measurements.

Results showed that thickness gradients improved strength with increasing HW content, both with and without hot pressing. Surface gradients did not show the same improvement, but their properties remained stable. Minor strength reductions were observed in anisotropic orientations. Overall, fibre gradients demonstrated potential for cost-effective material optimisation without compromising performance, highlighting their practical potential in industrial applications, particularly in advancing the performance and cost-effectiveness of MFPs. This approach also supports the transition toward environmentally responsible packaging that would meet both functional and economic demands.

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Muovi on laajalti käytetty materiaali ympäristöhaasteistaan huolimatta. Sen kestävyyttä on parannettu vuosien varrella, mutta materiaalikehitys suuntautuu edelleen kohti muovittomia vaihtoehtoja. Muovatut kuitutuotteet (MFP:t) ovat monipuolinen ja kestävä lähestymistapa kertakäyttömuovien korvaamiseen. Tällä hetkellä niiden käyttö kuitenkin rajoittuu toisiopakkauksiin, ja parannuksia tarvitaan MFP:iden houkuttelevuuden lisäämiseksi.

Tämän diplomityön tavoitteena oli tutkia, voisiko kuitujen yhteiskäyttö gradienttirakenteissa parantaa MFP-tuotteiden kannattavuutta perinteisten muovipakkausten korvaajana. Kuitugradienttien soveltuvuutta tarkasteltiin yhdistämällä havupuu- (SW) ja lehtipuuselluja (HW) käsiarkeissa, luoden paksuus- ja pintasuunnan gradienttirakenteita. Kuitugradienttiarkkeja tutkittiin viidellä eri SW/HW-suhteella sekä isotrooppisella ja anisotrooppisella kuitusuuntauksella. Lisäksi paksuusgradienttiarkeille tehtiin kuumapuristuskokeita muovausprosessin simuloimiseksi. Materiaalien ominaisuuksia arvioitiin huokoisuuden ja lujuusmittausten avulla.

Tuloksista havaittiin, että HW-pitoisuuden lisäys paransi sekä puristettujen että puristamattomien paksuusgradienttiarkkien lujuutta. Vaikka pintagradienteissa ei havaittu vastaavaa kehitystä, niiden ominaisuudet pysyivät johdonmukaisina. Anisotrooppinen kuitusuuntaus aiheutti joitakin heikentymisiä. Tulosten perusteella voidaan todeta, että gradienttirakenteet ovat lupaava lähestymistapa kustannustehokkaassa materiaalikehityksessä, parantaen sekä MFP-tuotteiden suorituskykyä että kestävyyttä ja samalla tukien siirtymistä kohti ympäristöystävällisempiä pakkausratkaisuja.

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SYMBOLS AND ABBREVIATIONS

Roman characters

A	strain at break	[%]
b	bulk	[cm^3/g]
d	sheet density	[kg/m^3]
F	force	[N]
g	basis weight	[g/m^2]
K	tensile stiffness	[kN/m]
KI	tensile stiffness index	[MNm/kg]
L	breaking length	[mm]
l	sample length	[mm]
R	tearing resistance	[mN]
RI	tear index	[mNm^2/g]
T	tensile strength	[kN/m]
TI	tensile index	[Nm/g]
t	thickness	[µm]
w	sample width	[m]

Abbreviations

BOD	Biological Oxygen Demand
CD	Cross Direction
CMP	Chemi-mechanical Pulping
CTMP	Chemi-thermo-mechanical Pulping
D	Dry sheet

DSF	Dynamic Sheet Former
HW	Hardwood
MD	Machine Direction
MFP	Moulded Fibre Product
NSSC	Neutral Sulphite Semi-chemical
PGW	Pressure Groundwood
RBA	Relative Bonded Area
RH	Relative Humidity
RMP	Refiner Mechanical Pulp
RO	Reverse Osmosis
S	Slurry
SBS	Specific Bond Strength
SGW	Stone Groundwood
SW	Softwood
TGW	Thermo Groundwood
TMP	Thermo-mechanical Pulp
WRV	Water Retention Value

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1 Introduction

Plastic remains widely used material in various industries despite its environmental drawbacks. While recycling efforts have improved its sustainability, the material development is increasingly shifting towards plastic-free alternatives. Paper-based solutions have been used as sustainable substitutes in some application areas, but applications like rigid packaging and tableware demand high strength and rigidity which limit the utilisation of conventional paper.

Moulded fibre products (MFPs) offer a promising alternative for single-use plastics and paper-based packaging. MFPs are already used in low-grade applications such as egg cartons, secondary packaging and cushioning. Recently, interest has grown to expand MFP utilisation even further, to more demanding applications due to their many advantages. (Debnath et al., 2022; Didone et al., 2017; Dislaire et al. 2021; Freville et al., 2024.)

The key appeal of MFPs is their environmental benefits. Made from renewable feedstock MFPs can be biodegradable, recyclable, and support circular economy principles. (Debnath et al., 2022; Didone et al., 2017; Freville et al., 2024.) MFPs are also customisable and can be formed into various shapes to meet diverse requirements. Despite the many advantages, increased implementation of MFPs is not straightforward. Fibre-based materials are quite different compared to conventional plastics and thus the limited knowledge of the material and processing behaviour limits them from the high-performance applications (Debnath et al., 2022; Freville et al., 2024). Therefore, it is important to gain better understanding on how fibre-based material behaves and enable the flexibility on their usage for different application.

One way to enhance the appeal of MFPs is by improving cost-competitiveness. Although the MFP production process is relatively cost-effective, raw material costs – especially for virgin softwood (SW) pulp – remain high (Debnath et al., 2022). SW pulp is preferred for its strength, but its cost limits the economic viability of MFPs. (Eskelinen, 1999; Rebola, Azevedo, and Evtuguin, 2021; Johansson, 2011; Laukala et al., 2019). Therefore, this thesis investigates the possibility of combining relatively cheaper short-fibred hardwood (HW) materials with long-fibred SW pulp to reduce costs while maintaining desired properties.

To date, the majority of moulding studies have focused on process conditions and additives rather than raw material combinations (Debnath et al., 2022; Dislaire et al., 2021; Kaplan, Johansson, and Östlund, 2025). However, fibre blending is common in papermaking, where different pulps are usually mixed to balance strength and surface properties (Karlsson, 2007; Kiviranta, 2000). For pulps, long and short fibres have been combined as mixtures in so-called reinforcement pulps (Johansson, 2011; Karlsson, 2007). In both cases, utilisation of multiple fibres has been found beneficial. Additionally, some studies exist where long and short fibres have been mixed to see how these mixtures and blending ratios affect sheet properties. While some studies have examined fibre mixtures, results vary depending on fibre characteristics and other variables, making outcomes difficult to predict. (Chauhan, Kumari, and Ghosh, 2013; Mansfield, Kibblewhite, and Riddell, 2004; Retulainen, 1992.)

Existing research often combine fibre mixing with treatments like refining, making it hard to isolate the effects of fibre types. (Chauhan, Kumari, and Ghosh, 2013; Hiltunen, 2003; Mansfield, Kibblewhite, and Riddell, 2004). Thus, this study aims to address that gap by investigating fibre combinations with minimal processing, enabling to understand the role of fibre types and their combinations more systematically. In addition, since the properties of fibre mixtures seem to be quite difficult to control, perhaps a different approach to utilising fibre combinations should be applied. Therefore, this study introduces the concept of fibre gradient structures – layered configurations of different fibres – and evaluates their feasibility in paper sheets. The goal is to determine whether this approach could enhance MFP performance and cost-effectiveness. The objective of this thesis can be concluded with the following research questions:

- Can fibre gradients be created effectively?
- How different gradient structures affect final properties?
- Can fibre gradients be tailored to achieve specific final properties?
- Are fibre gradients a viable solution for MFP applications?

This thesis consists of two parts: a literature review and an experimental section. The literature review covers fibre-based processes including pulping, papermaking and moulding. The most common wood-based fibres and fibre characteristics and their effect on sheet properties are described. The experimental section describes the implementation of two types of fibre gradients in laboratory hand sheets and Dynamic sheet former (DSF) sheets. During these experiments, the composition of fibre gradients is altered to find optimal

fibre combinations. Finally, the behaviour of different structures is evaluated based on the physical and mechanical properties of sheets.

2 Fibre dimensions and properties

Fibre dimensions and properties are key factors that affect product properties. Fibre properties depend on fibre source, such as type of wood and treatment method used. (Hakkila and Verkasalo, 2009; Mboowa, 2021.) This chapter outlines various, chemical and mechanical treatment methods used for fibres. In addition, a comparison of fibre properties based on wood type, and treatment method are presented.

2.1 Pulping methods

The goal of pulping is to separate lignin and other extractives from fibres with minimal fibre breakage, allowing the fibres to form web-like structures freely. Depending on the interpretation, pulping methods can be divided into three to five groups. Two main groups are chemical and mechanical pulping. Methods that combine both treatments may be classified under mechanical pulping, or as chemi-mechanical pulping (CMP). Semi-chemical pulping, also known as high-yield chemical pulping, contains modified chemical pulping methods. A more recent addition is biological pulping, in which lignin is softened and removed with fungi or enzymes, achieving energy and chemical savings compared to mechanical and chemical pulping methods. (Bajpai, 2018f; Iglesias et al., 2020; Li et al., 2023; Mboowa, 2021.)

Bleaching is a chemical treatment that can be applied to all pulp types. Its objective is to increase pulp brightness, though the bleaching mechanism is determined by the pulp type. For mechanical pulps which contain a high amount of lignin, the bleaching process replaces coloured lignin structures with colourless. In chemical pulping, high brightness is achieved through lignin removal. Additionally, bleaching can remove extractives and hemicelluloses which can improve certain pulp properties. (Chirat et al., 2011; Jäkärä, Persson, and Mårtens, 2009; Mboowa, 2021.)

2.1.1 Chemical pulping

Chemical pulping methods use chemical treatments to remove lignin. Chemical pulps are low-yield pulps, but their advantage is that the resulting pulp has low lignin content and intact fibres with low breakage and good bonding capabilities. The most common chemical pulping methods are kraft and sulphite pulping. Although other chemical pulping methods exist including organosolv and dissolving pulp, this study will focus only on kraft and sulphite processes. (Bajpai, 2018f; Iglesias et al., 2020; Li et al., 2023; Manfredi, 2024; Mboowa, 2021.)

Kraft pulping is the most common pulping method used worldwide. Its roots go all the way back to the 19th-century. The universality of kraft pulping is due to the numerous advantages of the process. For example, the kraft process provides superior mechanical strength properties for pulp, which is favourable in papermaking and many other pulp applications. Moreover, all wood types and even non-wood materials can be pulped with the kraft process and mills can function as significant electricity providers when hemicellulose and lignin are burned to produce steam and electricity. (Gustafsson et al., 2011; Iglesias et al., 2020; Mboowa, 2021.)

Kraft pulping is a combination of elevated temperature, high pressure and strongly alkaline conditions created by sodium sulphate and sodium hydroxide. During the process lignin is broken down into ionic fragments as fibre swells. These ionic fragments then dissolve into cooking liquor. The use of chemicals in the process has been optimised through time and chemicals are recycled and recovered efficiently in the process. (Bajpai, 2018f; Gustafsson et al., 2011; Iglesias et al., 2020; Mboowa, 2021.) Still, extracted kraft lignin is mainly burned to generate energy. In recent years there has been a lot of research done about possible new applications for extracted lignin to enhance sustainability and improve cost-effectiveness. (Argyropolulos et al., 2023; Dessbesell et al., 2020; Kumar et al., 2020.)

The second most common chemical pulping method is sulphite pulping. Its history goes back to the 20th century, around 60 years after kraft pulping was patented. Sulphite pulping has three modifications: acid, neutral and alkali sulphite pulping. In addition to the chosen pH, the process uses elevated temperature and pressure with sulphur dioxide and bisulphite combined with cations, such as calcium, sodium or potassium. During the process, lignin is separated from fibres through sulphonate formation. Of the three variations of sulphite

pulping, the acid process is rarely used because its low pH causes corrosion in the process and decreases the quality of fibres. (Bajpai, 2018f; Gustafsson et al., 2011; Iglesias et al, 2020; Mboowa, 2021.) The other two methods are used more, but mainly as a part of chemi-thermo-mechanical pulping (CTMP) or in the production of speciality pulps like neutral sulphite semi-chemical (NSSC) and dissolving pulp. (Iglesias et al., 2020.)

Sulphite pulps can compete with kraft pulp in brightness, yield, costs and flexibility that results from a wide range of possible pH conditions. However, the process has also significant defects that have determined its status as a secondary chemical pulping method. Sulphite pulps and papers produced from it struggle with strength, whereas the process is slow and has poor sustainability because of difficult chemical recovery. The applicability of the sulphite process is also poor because it is only suitable for wood sources with low extractive content. (Bajpai, 2018f; Gustafsson et al., 2011; Iglesias et al, 2020; Mboowa, 2021.)

2.1.2 Mechanical pulping

Mechanical pulps are commonly used even though chemical pulping methods are more widely utilised. In mechanical pulping, wood is pulped under mechanical stress through grinding, refining and shearing producing coarse pulp. Given that no chemicals are used during mechanical pulping, lignin is only softened under mechanical stress, which results in a very high yield, typically over 90 %. Mechanical pulps are mostly produced from SW, commonly spruce, to improve the strength properties of the pulp. (Bajpai, 2018f; Manfredi, 2024; Mboowa, 2021; Lönnberg, 2009.)

Mechanical pulps can further be divided into groundwood (GW), thermo-mechanical pulp (TMP), refiner mechanical pulp (RMP) and CTMP (Bajpai, 2018f; Mboowa, 2021). GW pulps can be further divided into stone groundwood (SGW), pressure groundwood (PGW) and thermo groundwood (TGW) (Karojärvi et al., 2009). Among all mechanical pulping processes, RMP is rarely used, and its descendant TMP is preferred. The same goes for SGW and TGW: the two are barely used and the majority of GW pulps are produced with PGW. (Bajpai, 2018f.) Thus, RMP, SGW and TGW will not be discussed further.

PGW uses steam at moderately high temperatures (70-120°C) to soften wood before it is ground into pulp. By softening wood chips before grinding, fewer fibres are broken ensuring better pulp strength and lower energy consumption. (Bajpai, 2018f; Karojärvi et al., 2009.) Although PGW is better than SGW and TGW processes the pulp is significantly weaker and coarser than TMP or CTMP. Therefore, the use of the PGW process is uncommon. (Bajpai, 2018f.)

TMP and CTMP are the most used mechanical pulping methods. In TMP the pulping is done in two stages. In the first stage chips are refined in refiners that are pressurised with steam and operated in high temperature. This process step is done to liberate fibres. In the second stage, the aim is to prepare liberated fibres for papermaking, and it is done with refiners in lower temperatures and atmospheric pressure. This two-step pulping method causes significantly less fibre cutting than RMP or GW methods, ensuring also higher pulp strength. (Bajpai, 2018f; Mboowa, 2021; Tienvieri et al., 2009.)

CTMP is the same as the TMP process but with the addition of mild chemical treatment before refining. When the TMP process uses steam to soften fibres before refining, CTMP combines steam with chemicals. Commonly utilised chemicals include peroxides and carbonates, but the most used ones are sodium hydroxide and sodium sulphite. (Bajpai, 2018f; Mboowa, 2021.) To reach a similar separation of fibres that the TMP process can achieve, less mechanical energy is needed because of chemical treatment (Mboowa, 2021). Simultaneously, chemical treatment is considerably milder compared to chemical treatments in chemical pulping. (Bajpai, 2018f; Mboowa, 2021.)

Compared to TMP, the strength properties of CTMP are better, while also maintaining high yields. In addition, TMP struggles with the utilisation of hardwoods (HW), whereas CTMP works well with both SW and HW. On the downside, the CTMP process produces high biological oxygen demand (BOD) and high colour effluents that are hard to treat, which makes the process environmentally challenging. (Bajpai, 2018f; Nickull and Pitkänen, 2009; Mboowa, 2021.) Although CTMP is classified under mechanical pulping, it is often used as a replacement for chemical pulps (Bajpai, 2018f).

2.2 Important fibre properties

Fibre properties and dimensions are important factors affecting pulp properties which further influences the behaviour of paper. Important fibre properties include length, width, coarseness, strength, cell wall thickness, bonding properties, flexibility, collapse and swelling. All these properties are highly dependent on the raw material. Whether the material source is SW, HW, chemical pulp, or mechanical pulp, these properties vary greatly. (Bajpai, 2018g; Brännvall and Annergren, 2009; Manfredi, 2024; Sirviö, 2008.) It should also be noted, that although fibre properties affect for example, strength and optical characteristics of paper, no fibre property alone should be used to explain differences in paper or pulp behaviour (Manfredi, 2024).

Fibre length is related to pulp and paper strength. Up to some point, longer fibre means that there are more possible contact points for fibre-fibre bonds, which then increases the bonding capacity and strength of the product. However, long fibres do not automatically indicate good paper strength because fibre length is not the only property affecting it. (Brännvall and Annergren, 2009; Manfredi, 2024). As for other paper properties, long fibres can affect negatively on the formation of paper, since long fibres get entangled with one another, causing uneven fibre distribution. (Brännvall and Annergren, 2009; Manfredi, 2024; Mangasha, 2019.)

Fibre strength describes the strength of a single fibre. It is greatly affected by all treatments, such as chemical treatments that often decrease fibre strength. (KnowPulp, 2024d; Manfredi, 2024). The strength of fibre does not necessarily mean strong paper, since paper strength is affected by fibre-fibre bonding and not only individual fibre strength (Bajpai, 2018g). Accurate quantification of fibre strength is also problematic, which is why it is rarely used in the characterisation of fibres. (Brännvall and Annergren, 2009; KnowPulp, 2024d.)

Cell wall thickness and fibre width affect the strength properties of paper but in a different way than fibre length. Whereas the effects of fibre length on paper strength are more straightforward, width and cell wall thickness improve strength through fibre flexibility. When the fibre cell wall is thick the fibre is usually dense. The same goes for fibre width. When the fibre is narrow or the fibre wall is thin fibres are more flexible, which improves fibre bonding thus increasing the strength of the paper. (Heikkurinen, 1999; KnowPulp, 2024c; Manfredi, 2024.)

Coarseness is a property that can explain multiple paper properties. Coarseness affects fibre strength, specific surface area and flexibility of a fibre (KnowPulp, 2024g). Coarseness is a relation between fibre length, cell wall thickness and fibre width. Therefore, this one property can in a way take three properties into account. (KnowPulp, 2024d; Manfredi, 2024.) The relation between coarseness and strength is as follows: often high coarseness means low strength. Reduction in strength is the result of poor fibre flexibility. (Manfredi, 2024.)

Bonding properties are one of the most important fibre characteristics. Bonding affects the optical and strength properties of paper. (Brännvall and Annergren, 2009; Manfredi, 2024.) The most significant bonding properties include bonding strength which can be illustrated as specific bond strength (SBS) and relative bonded area (RBA). RBA describes the bonded area of a fibre compared to all available fibre surface area (Manfredi, 2024; Sirviö, 2008). SBS is the minimum force that is needed to break the fibre bonding area, and it has a strong effect on strength properties like tear, tensile and burst strength. RBA, on the other hand, has a greater influence on optical properties. (Brännvall and Annergren, 2009; Manfredi, 2024; Sirviö, 2008.)

Collapse and flexibility are quite similar properties with the same effect on paper behaviour. Fibre width, cell wall thickness and fibre length affect the two properties, which then further influence fibre bonding through the available bonded area. (Brännvall and Annergren, 2009; Manfredi, 2024; Mangasha, 2019.) Moreover, collapsed and flexible fibres create improved fibre networks because fibres can adapt better to their environment while simultaneously increasing paper strength (Brännvall and Annergren, 2009; Manfredi, 2024).

Collapsing ability and flexibility are strongly dependent on fibre swelling. Swelling is said to be the most important fibre characteristic because it has a great influence on product properties, but also on pulp processing. Fibre swelling is caused by water that penetrates the cell wall. This property is described by water retention value (WRV) and is often manipulated with refining. (Bajpai, 2018g; Brännvall and Annergren, 2009; Manfredi, 2024; Mangasha, 2019; Sirviö, 2008.)

2.3 Fibre properties of different wood types

Fibre properties are dependent on many varied factors. For wood-based fibres, the effect of wood type on fibre properties is essential to acknowledge. Wood types can be divided into two groups: SW (evergreen) and HW (deciduous). SW species include pines and spruces. HW species are a broader group and include for example birch, acacia, eucalyptus, aspen and poplar. The most significant HW species in the pulp and papermaking field are birch and eucalyptus. (Iglesias et al., 2020; Li et al., 2023; Mboowa, 2021; Sirviö, 2008.)

In addition to fibre characteristics that were described in Chapter 2.2, it is also important to consider the chemical properties of fibres. The chemical composition of wood-based fibres is broadly divided between cellulose, hemicellulose and lignin. Moreover, there are small amounts of extractives present. (Hakkila and Verkasalo, 2009; Iglesias et al., 2020; Mboowa, 2021.) Rough estimates for chemical properties of SW and HW species are presented in Table 1.

Table 1. Estimates of different wood-based fibres' chemical composition (Hakkila and Verkasalo, 2009; Knowpulp, 2024d; Mboowa, 2021).

Wood species	Cellulose, [%]	Hemicellulose, [%]	Lignin, [%]	Extractives, [%]
Pine	40-45	25-30	25-30	< 5
Spruce	40-45	25-30	25-30	≈ 5
Birch	40-45	35-40	20-25	< 5
Eucalyptus	45-50	20-25	25-30	< 5

Table 1 shows that SW has generally lower cellulose content than HW. Hemicellulose and lignin contents are nearly equal for both wood types, except for birch which has noticeably higher hemicellulose content and lower lignin content. Extractives are similar for all presented wood species, with spruce having slightly higher content than other three.

The chemical composition of fibres defines their behaviour. It can also affect process performance. For example, the yield of the chemical pulping process is partly dependent on the cellulose content of fibres. If cellulose content is high, yield is high and vice versa. High cellulose content correlates with increased fibre strength as well. Hemicellulose content has an effect on fibre swelling in a way that lower hemicellulose content leads to less swelling. Although hemicellulose type is different for SW and HW the same correlation stands.

Finally, lignin content influences process yield, pulp and paper strength, as well as stiffness of fibres. Higher lignin content causes lower yields and strength, and higher fibre stiffness. Therefore, in chemical pulping lignin is removed from the biomass. (Hakkila and Verkasalo, 2009; KnowPulp, 2024d; Mangasha, 2019.)

The main difference between physical properties of SW and HW is fibre size. SW fibres are longer and thicker than HW fibres. At the same time, HW fibres' cell walls are often thicker, providing HW fibres with stiffness, whereas SW fibres are more flexible and prone to collapse. Finally, SW fibres are coarser than HW fibres. In addition to differences between SW and HW, there are differences within species of both wood types. Averages of basic dimensions of SW and HW are presented in Table 2. (Bajpai, 2018h; Hakkila and Verkasalo, 2009; Iglesias et al., 2020; Sirviö, 2009.)

Table 2. Average fibre dimensions of SW and HW (Bajpai, 2018e and h; Hakkila and Verkasalo, 2009; Iglesias et al., 2020; Knowpulp, 2024c; Sirviö, 2009).

Dimension	SW	HW
Length, [mm]	2-6	0.8-1.6
Width, [µm]	25-50	10-30
Coarseness, [mg/m]	0.10-0.30	0.07-0.08
Cell wall thickness, [µm]	2-5	2-3

As seen from Table 2, there is a lot of variation in dimensions within wood types. While the presented values provide a general range for these dimensions, the actual variation is even greater. The variations depend on the part of the tree from which dimensions have been measured, but also the age of the wood, environment and even time of year can have considerable influence. (Bajpai, 2018f; Sirviö, 2009.) Therefore, the presented values should only be taken as indicative.

2.4 Fibre properties of different pulps

Treatment of wood fibres has a significant influence on fibre properties. Mechanical and chemical pulping methods are used to treat fibres to make them suitable for applications like papermaking. Because mechanical and chemical pulp fibres have very different properties,

the pulps are used to achieve different paper properties. (Bajpai, 2018f; Iglesias et al., 2020; Manfredi, 2024.)

The difference between chemical and mechanical pulp fibre properties resembles those between SW and HW fibres, although the cause of these similarities is distinct. Mechanical pulping methods, excluding CTMP, do not chemically soften wood, which leads to significant fibre breakage and the production of short, stiff fibres with high coarseness and low bonding ability due to undissolved lignin and extractives on the surface. This results in weak fibre network with limited strength. (Bajpai, 2018f; Manfredi, 2024; Sirviö, 2009; Lönnberg, 2009; Sirviö and Tuovinen, 2009.) In contrast, chemical pulping softens fibres and removes the majority of surface lignin, preserving fibre length while producing fewer fines. The resulting fibres are more flexible, collapsed, and swollen, which enhances their bonding potential and conformability, creating stronger fibre networks. (Bajpai, 2018f; Lehto, 2004; Sirviö, 2009.) Some of the pulp and fibre properties of mechanical and chemical pulps are gathered in Table 3.

Table 3. Comparison of chemical and mechanical pulp fibre properties (Retulainen, 1992; Sirviö, 2009).

Subject	Property	Chemical pulping	Mechanical pulping
Pulp properties	Hemicellulose content	Low	High
	Lignin content	Low	High
	Water affinity	Hydrophilic	Hydrophobic
Fibre size distribution	Long fibre content	High	Low
	Short fibre/fines content	Low	High
Fibre properties	Length	Long	Short
	Width	Narrow	Wide
	Stiffness	Flexible	Stiff
	Coarseness	Smooth	Coarse
	Straightness	Curly	Straight
	Level of collapse	Collapsed	Less collapsed

As shown in Table 3 both chemical and mechanical pulps have characteristics that make them more suitable for specific applications. Chemical pulps are typically preferred when high strength is required, whereas mechanical pulps perform better in applications where superior optical properties are essential. (Bajpai, 2018f; Manfredi, 2024: Lönnberg, 2009.)

2.5 Effect of mechanical treatment on pulp and fibre properties

Mechanical treatments apply mechanical force to influence fibre properties, and their effects vary depending on whether they are applied before or after pulping. As discussed in Chapter 2.1.2, mechanical treatments before pulping aim to liberate fibres and soften surface lignin. Post-pulping treatments, mainly refining, go beyond fibre separation. When optimised, refining can influence fibre length, stiffness, RBA, as well as fibre swelling and collapse. Collectively, these effects can significantly improve both the optical and mechanical properties of the pulp. (Bajpai, 2018g; Gharehkhani et al., 2015; Johansson, 2011; Manfredi, 2024.)

In addition to direct effects on fibre properties, mechanical treatments generate fines, which can have both favourable and unfavourable effects on pulp performance. The two main types of fines are lamellar and fibrillar, with the latter being more favourable due to their higher swelling capacity and surface area. While fines can enhance certain pulp and paper properties, they are usually considered problematic. (Bajpai, 2018g; Gharehkhani et al., 2015; Johansson, 2011; Manfredi, 2024.)

3 Effect of fibres on sheet properties

Fibres and their properties are considered when manufacturing paper. The previously discussed wood types and treatment methods are used in the papermaking process based on desired paper characteristics. To be able to characterise pulp in a way that the required properties are achieved, one must understand the mechanisms that affect the final behaviour of paper. (Johansson, 2011; Manfredi, 2024.) In this chapter, sheet properties are going to be discussed, including a comparison of paper properties between wood and pulp types.

3.1 Important sheet properties

Sheet properties can be divided roughly into three groups: optical, chemical, and physical properties. Optical properties include colour, brightness, opacity, and gloss. These properties are especially important in applications such as printing paper. (Bajpai, 2018b; Manfredi, 2024; Pauler, 2008; Vaarasalo, 1999.) Optical properties result from absorption and reflectance of paper surfaces, and they are both influenced by free surface area. For example, opacity improves with increasing free surface area. Free surface area means that there are no fibre bonds on the light reflecting surface which is why good optical properties usually equal poor strength. (Bajpai, 2018b; Manfredi, 2024; Pauler, 2008.)

The chemical properties of paper are rarely measured because the same properties can be measured directly from the pulp. The most significant chemical property measured is ash content. It describes the inorganic content of paper. A small amount of inorganic matter originates from wood, but most of it is from pulping and different fillers used in papermaking. In addition to ash cellulose, hemicellulose and lignin contents of pulp impact paper properties. (Bajpai, 2018b; Hausalo and Söderhjelm, 1999; Manfredi, 2024.)

Physical properties can be further divided into smaller groups like mechanical, structural and surface properties. Surface and structural properties do not have a reliance on fibre bonds. One of the most measured surface properties is roughness while structural properties consist of thickness, formation, porosity, bulk, and air resistance. (Bajpai, 2018b; Levlin, 1999; Manfredi, 2024.) Roughness is affected by fibre length. Longer fibres are coarse, and they have higher mass compared to short fibres. Therefore, short fibres form smoother surfaces

for sheets, reducing the roughness value. (Alava, 2008; Bajpai, 2018b; Manfredi, 2024.) The formation is also influenced by fibre length, but coarseness and flexibility affect it, too (Manfredi, 2024). Formation describes how well fibres are oriented and how uniform the sheet is. It is one of the most impactful properties because it plays a role in many paper characteristics including mechanical and optical properties. (Bajpai, 2018b; Manfredi, 2024.)

Many structural properties are dependent on the same fibre characteristics. For example, bulk, thickness and density are all affected by fibre flexibility. This is because stiffer fibres do not pack as densely making low-density sheets with high thickness and high bulk. (Bajpai, 2018b; Kajanto, 2008; Manfredi, 2024.) Fibre deformations like curls and kinks can also influence these three structural properties because they inhibit the formation of tight structures when fibres cannot get so close to each other. However, fibre deformations cause losses in mechanical properties, which is why they are not utilised in bulk or thickness manipulation. (Manfredi, 2024.)

Porosity is easier to describe through its dependence on bulk and density rather than specific fibre characteristics. For high-density sheets porosity is usually low because there are fewer voids between fibres compared to bulkier sheets. However, fines can cause the opposite behaviour. High bulk material often has high fines content, but contrary to normal assumption of high porosity, those fines can fill the voids between fibres causing porosity reductions. Porosity is measured with air permeance, which describes the amount of air that flows through the paper when a pressure difference is subjected between its sides. Air resistance is the inverse of porosity. If sheet porosity is high air resistance is low. Thus, high-density sheets have small air permeance and high air resistance. (Karlsson, 2007; Manfredi, 2024.)

Mechanical properties are the most important sheet characteristics in many applications. Mechanical properties include tensile strength, tearing resistance, burst strength, bond and stiffness properties. (Bajpai, 2018b; Levlin, 1999; Manfredi, 2024.) These properties are dependent on fibre bonding and fibre type as well as pulping. Most mechanical properties are related to a fibre's collapsing abilities, specific surface area and flexibility since these properties define the bonding abilities. (Alava and Niskanen, 2008; Bajpai, 2018b; Levlin, 1999; Manfredi, 2024.) Tensile and burst strength are mainly affected by the bonding abilities of fibres and the strength of these bonds. Moreover, fibre length and strength impact

tensile strength whereas burst strength is influenced by the stretchability of fibres. (Bajpai, 2018b; Karlsson, 2007; Levlin, 1999; Manfredi, 2024.)

Tearing resistance is influenced by fibre bonds, as are other mechanical properties but in addition, it is strongly affected by fibre length. Due to the dependence on fibre length and as opposed to other mechanical properties tear strength is decreased with mechanical treatment because of reduced fibre length. A secondary property that affects tearing resistance is fibre strength. (Karlsson, 2007; Levlin, 1999; Manfredi, 2024.) The bond strength or internal strength of paper is highly reliant on fibre length like tearing resistance. Additionally, fibrillar fines content influences its value. High fibrillar fines content means that there are a lot of available contact points for bonding which results in high binding capabilities. (Kajanto, 2008; Manfredi, 2024.) Finally, the stiffness properties of paper behave oppositely to the mentioned mechanical properties. This is because stiffness improves when fibre length is reduced. Stiff, short fibres create thick sheets with high bulk, adding resistance to the bending of paper. (Kajanto, 2008; KnowPulp, 2024f.)

3.2 Comparison of softwood and hardwood sheet properties

Differences between SW and HW sheet properties result from differences in the fibres. SW fibres provide strong, well-bonded sheets whereas HW fibres create sheets with good optical properties. In more detail, the strength of SW sheets originates from long fibres that have numerous available contact points between fibres, as flexible fibres conform better, creating strong fibre networks, and collapsibility which increases fibre surface area that is utilised for fibre bonds. As a result, tensile and burst strengths are high. Tear and internal bonding strengths are also high for SW sheets because of fibre length. Good strength properties further provide good runnability. (Johansson, 2011; Karlsson, 2007; Levlin, 1999; Manfredi, 2024.)

The optical properties of SW sheets are poor because there is very little free surface area that reflects light. After all, most surface area is used by fibre bonds. Formation is also poor as a result of the coarseness and length combined with the high mass of fibres, which causes sheets with high roughness. Finally, due to fibre flexibility SW sheets are dense, with low bulk, thickness, porosity, and high air resistance. As a result of low bulk SW sheets also have low stiffness abilities. (Bajpai, 2018h; Johansson, 2011; Manfredi, 2024.)

In contrast to SW sheets, HW sheets have good formation and high opacity because of short and stiff fibres with free surface area. HW sheets are also significantly smoother than SW sheets due to good formation. (Bajpai, 2018g; Johansson, 2011; Manfredi, 2024.) Due to the fibre stiffness, HW creates sheets with high bulk and low density. Bulky sheets are also relatively thicker than dense sheets which makes HW sheets stiffer with high porosity and low air resistance. Strength properties are noticeably weaker compared to SW sheets because there is significantly less bonded area and contact points on fibres. Therefore, the resulting fibre networks lack tensile, burst, bonding and tear strengths. (Karlsson, 2007; Levlin, 1999; Manfredi, 2024.) A comparison of SW and HW sheet properties is compiled in Table 4 below.

Table 4. Comparison of SW and HW sheet properties (Bajpai, 2018h; Iglesias et al., 2020; Johansson, 2011; Karlsson, 2007; Levlin, 1999; Manfredi, 2024).

Group	Property	SW sheet	HW sheet
Optical	Opacity	Low	High
Surface	Roughness	High	Low
Structural	Formation	Poor	Good
	Bulk	Low	High
	Thickness	Low	High
	Porosity	Low	High
	Air resistance	High	Low
Mechanical	Tensile strength	High	Low
	Burst strength	High	Low
	Tear strength	High	Low
	Bonding strength	High	Low
	Stiffness	Low	High

It should be noted that Table 4 describes properties in comparison between SW and HW. For example, the low tensile strength of the HW sheet does not mean that appropriate strength properties could not be achieved with HW. HW sheet strength is just typically lower than of SW sheet. The second thing to consider is that sheet properties are not only dependent on wood type, but specific species and many other factors also have an effect.

Due to differences between sheet properties, SW and HW are used in different applications. SW is preferably used when high strength is needed, like paper board, printing and writing papers. HW is more useful in cases where optical properties are of high importance, like printing and art papers. In some applications such as paperboard production, SW and HW

are both used depending on the layer. The top layer is often made from HW because it provides a good surface finish. SW is used more in middle layers to define the strength of the board. Also, paper and board-making processes utilise different chemicals and fillers in achieving the desired properties, which makes raw material less significant. (Haarla, 2000; Kimari, 2000; Kiviranta, 2000; KnowPulp, 2024e.)

3.3 Comparison of chemical and mechanical pulp sheet properties

The comparison between chemical and mechanical pulp sheet properties resembles the comparison between SW and HW in the previous chapter. However, despite the similarities, the mechanisms behind these characteristics do not occur naturally but are achieved through chemical or mechanical manipulation. The differences between mechanical and chemical pulp sheets are primarily from variations in lignin content and fibre length. Lignin's hydrophobic nature restricts fibre swelling and bonding, making mechanical pulp sheets with high lignin content significantly weaker than chemical pulp sheets, which are hydrophilic due to the removal of lignin during chemical processing. (Bajpai, 2018f; Lehto, 2004; Manfredi, 2024; Sirviö, 2008.) Fibre length affects both mechanical and optical properties of sheets. As a result, mechanical pulp sheets exhibit superior optical performance. (Heinemann and Vehniäinen, 2009; Lehto, 2004; Manfredi, 2024; Sirviö, 2008.) Comparison of chemical and mechanical pulp sheet properties is comprised in Table 5.

Table 5. Comparison of chemical and mechanical pulp sheet properties (Bajpai, 2018f; Heinemann and Vehniäinen, 2009; Manfredi, 2024; Sirviö, 2008).

Group	Property	Mechanical pulp sheet	Chemical pulp sheet
Optical	Opacity	High	Low
Surface	Roughness	Low	High
Structural	Formation	Good	Poor
	Bulk	High	Low
	Thickness	High	Low
	Porosity	High	Low
	Air resistance	Low	High
Mechanical	Tensile strength	Low	High
	Burst strength	Low	High
	Tear strength	Low	High
	Bonding strength	Low	High
	Stiffness	High	Low

The illustrated properties of each pulp type determine their suitability for different applications. Chemical pulp is used where high strength is essential, such as surface layers of board and fine papers. In contrast, mechanical pulp is favoured for applications requiring good optics and bulk, such as printing papers. Additionally, mechanical pulp is the more cost-effective option, which is why it is blended with chemical pulp in various applications. In general, the proportion of mechanical pulp is maximised to achieve cost savings while meeting performance requirements. (Haarla, 2000; Kimari, 2000; Kiviranta, 2000; KnowPap, 2024a; KnowPulp, 2024a and b.)

4 Pulp-based forming processes

Pulps can be utilised in many applications including boards, textiles and tissue products. The most conventional product is, however, paper. (Bajpai, 2018f; Manfredi, 2024.) Its production can be done on different scales from laboratory sheets that are used in quality control to industrial paper (Brännvall and Annergren, 2009; Hiltunen, 1999). In this chapter, three paper-forming procedures are discussed: hand sheet forming, DSF and the conventional papermaking process.

4.1 Preparation of hand sheets

Laboratory hand sheets are used to evaluate the quality of pulp and its applicability to the desired final purpose. Hand sheets do not guarantee that the final product, for example, paper is high quality and properties measured from hand sheets often differ noticeably from the properties of the final product. The primary reason for that is the fibre orientation of sheets. Laboratory hand sheets are isotropic which means that fibres in the sheets are randomly oriented in different directions. On the other hand, in machine-made paper fibres are mostly oriented in the direction of the machine. (Brännvall and Annergren, 2009; Hiltunen, 1999; Johansson, 2011.) However, hand sheets are suitable for evaluating the behaviour and quality of pulp (Brännvall and Annergren; Hiltunen, 1999).

There are standards for laboratory hand sheet forming from for example TAPPI and ISO (ISO 5269-1:2005, 2005; TAPPI T 205 sp-24, 2024). Other standards exist as well but these two are perhaps the most commonly considered. Fundamentally, sheet forming consists of pulp preparation, sheet forming and drying before testing. In more detail, the process starts from pulp disintegration in water. After disintegration, some pulp tests can be performed if desired. If mechanical pulp is used, the next step is the actual sheet forming. However, in the case of chemical pulp, the pulp-water suspension is often refined first with a laboratory refiner. After sheet forming, the next steps are wet pressing, drying, and conditioning. When the hand sheets are fully conditioned, they are ready to be tested. (Brännvall and Annergren, 2009; Hiltunen, 1999.) An example of a laboratory sheet former is presented in Figure 1 below.

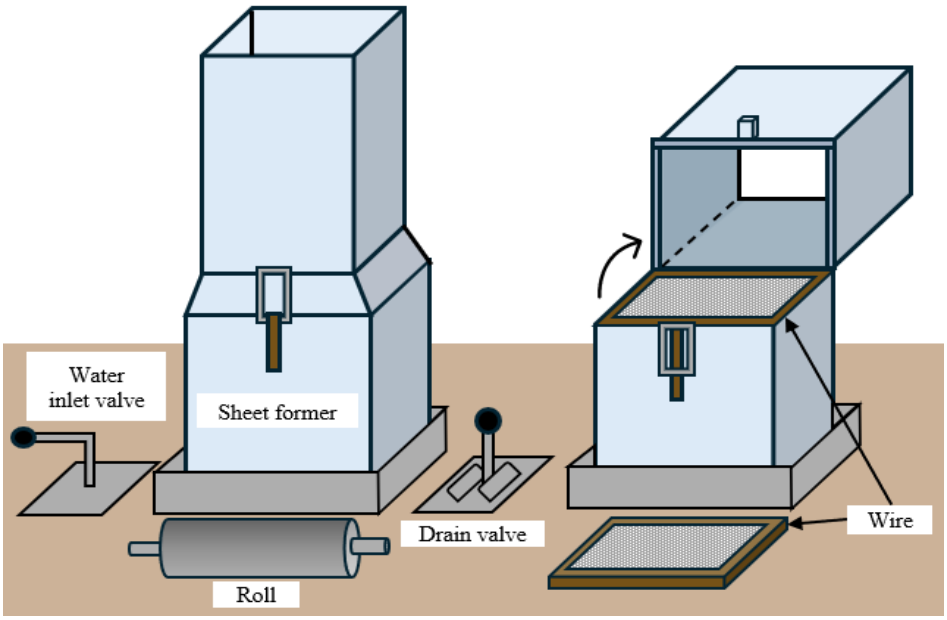

Figure 1. Laboratory hand sheet former (Adapted: Hiltunen, 1999).

In Figure 1, the illustrated hand sheet former is a so-called squared model. Other models like rectangular and circular formers also exist. The water inlet valve and drain valve shown in Figure 1 are used to control water levels and drainage in the former. The wire is inserted inside the former and hand sheets are formed on top of it during water drainage. After sheet formation, sheets are rolled before gathering them for pressing and drying. Formed sheets are often made to 60 g/m² basis weight to simulate paper production but other weights can also be used to evaluate pulp behaviour in other applications. (Brännvall and Annergren, 2009; Hiltunen, 1999.)

There are two basic methods to form laboratory sheets: fresh water and whitewater circulation methods. They can also be called open and closed white water systems. The fresh water method is used more often whereas white water circulation is used mainly with mechanical pulps because of their high fines content. In the freshwater method, water used during the forming is changed after each formed sheet. On the other hand, the closed white water system circulates the same water for all sheets. Circulation of white water is necessary for mechanical pulps to retain similar fines content in the sheets as in original pulp. (Brännvall and Annergren, 2009; Hiltunen, 1999.)

4.2 Dynamic sheet former

DSF is the next step from conventional laboratory sheet forming towards the papermaking process. It is used similarly to laboratory hand sheets to evaluate pulp quality but the process and formed sheets are significantly different. In the DSF process, the water-pulp slurry is sprayed through a nozzle on a rotating drum. As the drum rotates at high speed, some fibres align themselves in the rotating direction. Created sheets are thus more anisotropic than laboratory hand sheets and closer to commercially produced paper. (Ali, 2020; Neagu, Gamstedt, and Lindström, 2005.) A rough illustration of DSF is presented in Figure 2.

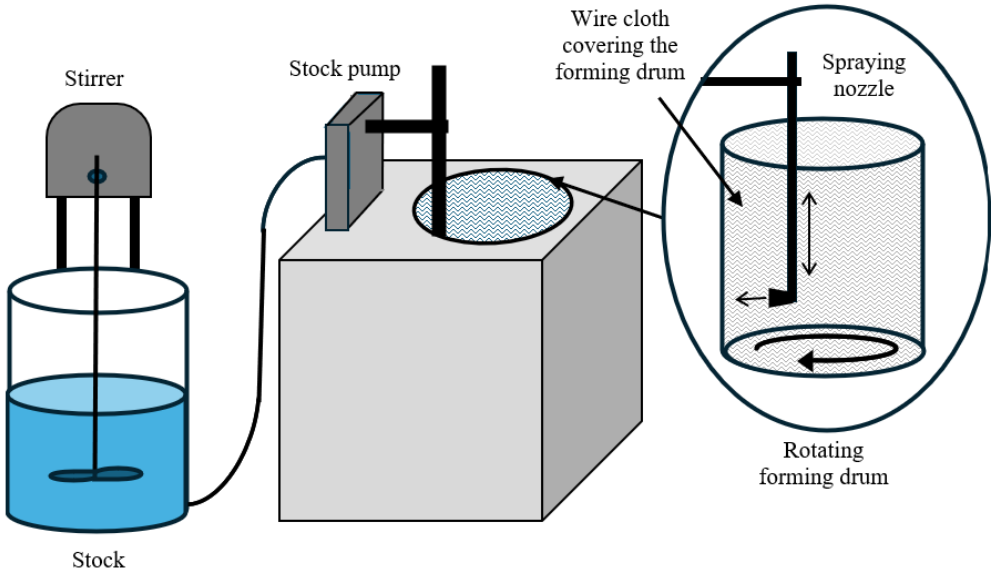

Figure 2. Simplified illustration of DSF. Rotating forming drum is perforated, enabling water drainage. Drum is covered with a wire cloth for easy sheet removal. (Adapted: Neagu, Gamstedt, and Lindström, 2005.)

In the DSF process, before starting the sheet forming, a so-called water wall is created behind the wire that goes around the drum to assist the sheet formation. Continuously stirred water-pulp slurry is then pumped to DSF, where the slurry is sprayed through the nozzle on the cylindrical rotating drum. The spraying nozzle moves up and down during the process, spreading slurry evenly across the height of the drum. As the drum rotates at high-speed, fibres align themselves in the rotating direction. Once the sheet reaches the targeted basis weight, water is drained from the drum. Lastly, the sheet is carefully removed from the drum, followed by pressing and drying as in laboratory hand sheet making. (Ali, 2020; Neagu, Gamstedt, and Lindström, 2005; Pickering and Le, 2016; Sunny, Pickering, and Lim, 2017.)

4.3 Papermaking process

Ideally, material development progresses from laboratory to piloting and further to commercial production. In case of paper development, similar steps could mean starting from preparation of hand sheets (Chapter 4.1), proceeding to DSF (Chapter 4.2) and finally to papermaking process, which will be presented in this chapter. In this type of development, it is important to acknowledge the differences between the process scales, how products differ and what is the significance of the changes. Therefore, this chapter will also highlight reasons behind the behavioural differences of industrial and laboratory scale sheets.

Papermaking starts at the wet end of the paper machine, which consists of the web-forming and press sections. As the name describes, web-forming section is where wet fibre web is formed on the wire and drained to inhibit web breakage in later steps. Paper properties such as basis weight, formation and uniformity are mostly defined during this first section of the machine. (Bajpai, 2018d; Norman, 2008.) An example of a web-forming configuration is presented in Figure 3.

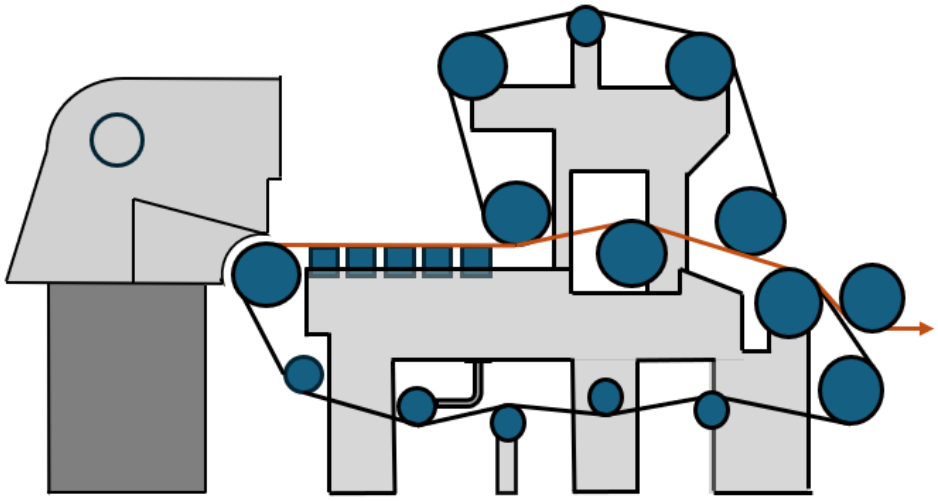

Figure 3. Web forming section of a paper machine (Adapted: KnowPap, 2024c).

In the press section of the wet end, water is removed by compression when paper runs between rolls and felts. During this section paper usually gains its wet strength through improvements in fibre bonds. Other paper properties that are also affected are smoothness, porosity and bulk. (Bajpai, 2018d; Paulapuro, 2008.) An example of a press section is presented in Figure 4 below.

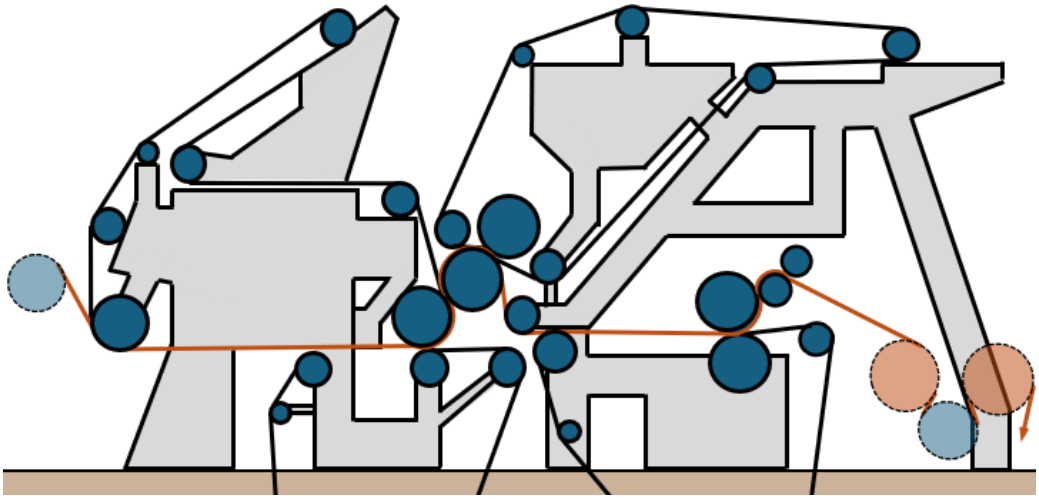

Figure 4. Press section of a paper machine (Adapted: KnowPap, 2024b).

The wet end is followed by the dry end which is generally the longest section with the highest energy consumption of the paper machine. In the dry end water is removed from the paper through evaporation when paper runs between hot cylinders, reaching dry content as high as over 90 %. (Bajpai, 2018c; Juppi, 2010.) The dry end influences many paper properties including strength and surface properties as well as brightness (Bajpai, 2018c; Pakarinen et al., 2010). A part of the drying section is illustrated in Figure 5.

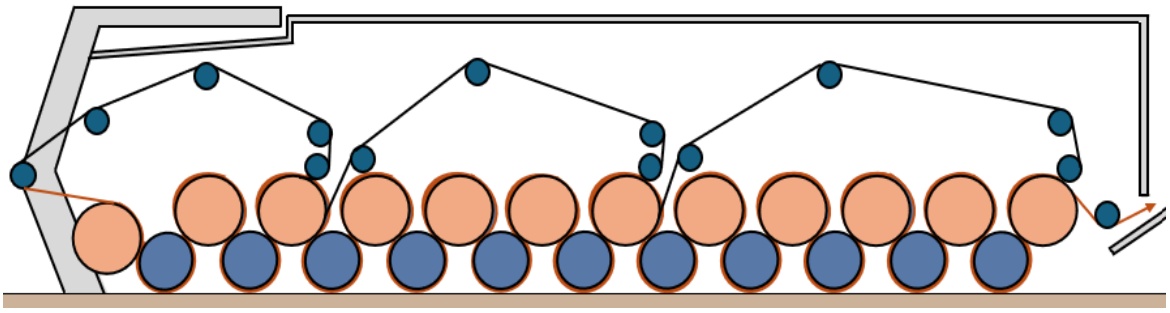

Figure 5. Drying section of a paper machine (Adapted: Juppi, 2010).

The final part of the paper machine is the finishing section. The finishing section can contain sizing, coating, calendering and reeling, and the applications of the steps depends on the final product (Ahlstedt et al., 2010; Enwald et al., 2010; Paltakari, Lehtinen, and Imppola, 2009). Some of the properties defined during finishing steps include internal strength, thickness, gloss, smoothness, and opacity (Ahlstedt et al., 2010; Bajpai, 2018a and c; Paltakari, Lehtinen, and Imppola, 2009).

The papermaking process adds constant stress on paper due to its high speed, temperature and pressure, which need to be considered when transitioning from laboratory to industrial scale. One of the most significant factors is fibre orientation. In commercial papermaking, fibre orientation is anisotropic, as fibres align in the machine direction (MD), causing differing properties in MD and cross direction (CD). This orientation is better estimated with DSF sheets than in laboratory hand sheets. (Brännvall and Annergren, 2009; Hiltunen, 1999; Johansson, 2011.) Another key difference is sheet formation. Laboratory hand sheets typically have more uniform fibre distribution, while commercial sheets can contain flocs, primarily formed from long fibres, that reduce paper strength (Brännvall and Annergren, 2009; Hiltunen, 1999; Johansson, 2011; Norman, 2008).

Pressing also differs as laboratory sheets undergo a longer but gentler pressing, while a paper machine applies considerably higher pressure on paper for a shorter time. Pressure load of the paper machine does not yield as good benefits as the gentler pressing of laboratory sheets. (Brännvall and Annergren, 2009; Johansson, 2011.) Finally, drying in laboratory conditions better controls shrinkage compared to the drying of the paper machine, where fibre alignment and shrinkage can reduce stretch properties. (Brännvall and Annergren, 2009; Johansson, 2011; Pakarinen et al., 2010.) All of the above highlight the problems in estimating commercial paper performance with laboratory results.

5 Moulded fibres

Moulded fibres are cellulose-based, three-dimensional products that have long been produced in various forms, such as egg cartons and box cushions. Recently, there has been a surge in research on moulded fibres as an environmentally friendly alternative to petroleum-based plastic packaging. There are two moulded fibre processes: the wet moulding, and the dry moulding process. (Debnath et al. 2022; Dislaire et al., 2021; Freville et al., 2024.) In this chapter, these two processes are described in detail. In addition, some of the biggest challenges of MFPs are discussed.

5.1 Wet moulding process

Wet moulding, originally inspired by the conventional papermaking process, is used to produce MFPs (Freville et al., 2024). These products can be classified into one-cast, transfer moulded, thermoformed, and processed MFPs based on raw material, end use, and production method. Processed MFPs do not follow a specific moulding process. Instead, the term refers to products that have undergone additional coating or processing steps. The key criterion is that they must remain biodegradable and recyclable, just as MFPs produced by the other three methods. (Debnath et al., 2022; Freville et al., 2024; Zhang et al., 2022.)

The simplest version of wet moulding process is one-cast, where fibres are drawn from a pulp suspension onto the surface of a porous mould, dewatered using vacuum, and then oven dried. Transfer moulding follows the same initial steps as one-cast moulding until dewatering step. After that, the product is transferred to a transfer mould, which compliments the formed shape and creates a more even surface on both sides. The product is then oven dried. The process can be extended further into thermoforming, where instead of oven drying, the product is dried using heated moulds pressed on both sides. (Debnath et al., 2022; Dislaire et al., 2021; Freville et al., 2024.) As the process progresses from one-cast to thermoforming, the quality of the MFPs improves. While one-cast and transfer moulded products are typically used for support and cushioning, thermoformed MFPs are suitable for high-quality applications such as cosmetics packaging and tableware. (Debnath et al., 2022; Dislaire et al., 2021; Freville et al., 2024; Jäntti, 2024; Zhang, 2022.)

5.2 Dry moulding process

The dry moulding process is still a fairly novel approach and barely used at industrial scale. This moulding process itself is short and quick, but it utilises fluff pulp as feedstock which needs to be produced separately. (Freville et al., 2024; Kaplan, Johansson, and Östlund, 2025; Pasquier et al., 2024.) Therefore, both fluff pulp process and dry moulding process are described in following chapters.

5.2.1 Fluff pulp production process

Fluff pulp is a bulky and strong materials with high absorption capacity and low absorption time (Ismaeilimoghadam et al., 2022; Rebola, Azevedo, and Evtuguin, 2021). It is manufactured for tissue applications like diapers, absorbent towels, and feminine hygiene (Ismaeilimoghadam et al., 2022; Freville et al., 2024; Rebola, Azevedo, and Evtuguin, 2021). In recent years, fluff pulp has also been utilised in dry moulding applications (Freville et al., 2024; Pasquier et al., 2024).

Fluff pulp production comprises of two steps: forming and fluffing. The forming step is similar to the papermaking process, although all steps are performed to produce a low-density sheet for easy fluffing, instead of strong and dense paper sheet (Kakonke, 2019; Rebola, Azevedo, and Evtuguin, 2021). In the fluffing step, the dry fluff pulp sheets are defibrated (fluffed) using equipment such as garnet mill or hammer mill, aiming to separate the sheet into individual fibres. The resulting fluff contains fibres of various lengths, along with fibre bundles and knots. (Eskelinen, 1999; Ismaeilimoghadam et al., 2022; Kakonke, 2019; Rebola, Azevedo, and Evtuguin, 2021.)

Fluff pulp is primarily produced from SW kraft pulp, which forms stronger fibre networks than HW (Eskelinen, 1999; Rebola, Azevedo, and Evtuguin, 2021). However, the utilisation of SW has its challenges due to difficult fluffing and high amount of produced dust (Rebola, Azevedo, and Evtuguin, 2021). As a result, HW fluff pulp has gained increasing interest in the market. Another common challenge in fluff pulp production is the formation of knots, which can cause deteriorations in absorption performance. Knot formation can be prevented by enhancing fibre separation with debonding agents. (Ismaeilimoghadam et al., 2022; Rebola, Azevedo, and Evtuguin, 2021.)

5.2.2 Dry moulding of fluff pulp

Dry moulding takes place after the fluffing of fluff pulp rolls. While the wet moulding process uses fibre slurry dry moulding utilises dry fibre fluff. First, fluffed pulp is pressed or air-laid on a vacuum belt forming a sheet of randomly oriented fibres. The dry fibre sheet is then moulded into shape with high temperature and pressure which promotes strong fibre bonds creating strong product. (Freville et al., 2024; Kaplan, Johansson, and Östlund, 2025; Pasquier et al., 2024.) The dry moulding process is illustrated in Figure 6 below.

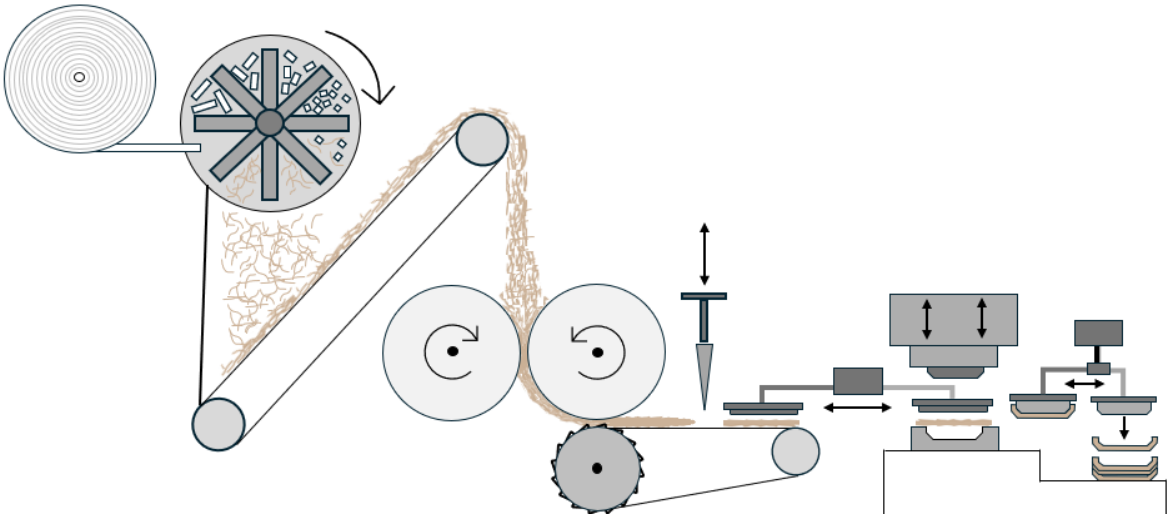

Figure 6. Dry moulding of fluffed pulp sheet (Adapted: Freville et al., 2024; Yangi, 2024).

As of now, MFPs are not advanced enough to replace plastic packaging without any additives or other processing before end use. Just as processed wet moulded products exist, dry moulded products can also be coated and treated to achieve better end properties. Coating is done on the dry fibre mat before pressing. Different additives can be either sprayed on the surface or mixed with fibres. (Freville et al., 2024; Kaplan, Johansson, and Östlund, 2025; Pasquier et al., 2024.)

5.3 Comparison and challenges of moulded fibres

Moulding processes offer promising solutions for the packaging industry. However, there are still significant challenges that either limit their applicability or overall are undesired in an industrial scale. Economically, dry moulding is the more appealing solution of the two as

it uses dry fibre mat instead of fibre slurry, leading to reduced energy consumption and faster drying. (Freville et al., 2024; Kaplan, Johansson, and Östlund, 2025; Pasquier et al., 2024.)

Regardless of the economic advantages, dry moulding remains mainly unused at an industrial scale, whereas wet moulding is used, despite its high energy and water consumption. One affecting factor is limited product depth of dry moulded products, typically around 3 to 6 cm (Freville et al., 2024). Additionally, wet moulded products are considerably stronger because the water used during moulding improves fibre bonding due to two mechanisms: capillary forces, which pull fibres closer together during water removal, and fibre swelling, which softens fibre surface and enhances interfibre contact and bonding. Low strength is perhaps the most substantial disadvantage of dry moulding, as strength is of great importance for MFPs. (Kaplan, Johansson, and Östlund, 2025; Pasquier et al., 2024.)

Another challenge relative to strength of dry moulded product is fibre length. When a fluff pulp sheet is subjected to mechanical stress during defibration, the average fibre length is reduced (Ismaeilimoghadam et al., 2022; Sorvari et al., 2001). As in papermaking, shorter fibres and fines weaken the product. Therefore, it is important to find a good length-to-strength ratio that maintains good product strength despite decreased fibre length (Bajpai, 2018g; Johansson, 2011; Rebola, Azevedo, and Evtuguin, 2021). A similar balance should be also found in terms of cost-effectiveness by balancing between SW and HW fibres. While SW is preferably used for its long and strong fibres, it is expensive, hence utilisation of cheaper HW pulp would be an attractive alternative to investigate (Bajpai, 2018f; Laukala et al., 2019).

Besides strength, another important property that needs to be developed in MFPs is hydrophobicity, yet it is a complicated matter. Hydrophobicity is an essential characteristic because MFP should retain its shape and strength even when subjected to moisture. (Debnath, 2021; Freville, 2024; Zhang et al., 2022.) However, fluff pulp used as raw material is strongly hydrophilic due to its usual objectives of use (Kakonke, 2019; Rebola, Azevedo, and Evtuguin, 2021). Moreover, hydrophobicity reduces the strength of inter-fibre bonds which decreases already crucial product strength (Manfredi, 2024). In conclusion, it is challenging to create strongly hydrophobic MFP from hydrophilic fluff pulp while balancing between strength deterioration caused by both moisture and moisture resistance.

6 Fibre mixtures

Fibre mixtures aim to modify the fibre structure in materials. In papermaking, combined use of different fibres like mechanical and chemical pulps simultaneously is rare but not unheard of. For example, chemical pulps are often used in surface layers of paper where strength is of utmost importance. In the middle layer, however, strength is not as important and mechanical pulp can be used to achieve other desired properties like high bulk. (Haarla, 2000; Karlsson, 2007; Kimari, 2000; Kiviranta, 2000.) Utilisation of actual fibre mixtures is rarer, but it could offer cost savings and improve the product customisability (Bajpai, 2018f; Laukala et al., 2019; Manfredi, 2006). In this chapter, SW/HW and chemical pulp/mechanical pulp mixtures are discussed based on several literature sources to get a general idea of how mixtures affect sheet properties.

6.1 Softwood – hardwood mixtures

As discussed in previous chapters, SW is a good raw material when creating strong products. Because of the strengthening abilities of SW, it can be added to HW pulp as reinforcement agent. HW can in turn be used to improve the stiffness, bulk and surface properties of SW sheets, but the addition of shorter and stiffer fibres can cause deterioration in the strength of the paper. (Johansson, 2011; Karlsson, 2007, Manfredi, 2024.) Thus, it is important to consider the mixing ratio of short and long fibres to find the best configuration for maximum enhancement and minimal deterioration. Some literature sources describe the effects of mixing SW and HW fibres for hand sheets. Main findings of these studies are summarised in Table 6 below.

Table 6. Available research on fibre mixtures with SW and HW.

Fibre blend	Property	Effect of HW	Notes	Reference
Euca/SW	Tensile strength Tear strength Burst index	+ - +	▪ Corefined samples ▪ Increased refining for high SW% causes decreased strength of HW fibres	Chauhan, Kumari, and Ghosh, 2013
Euca/SW	Opacity Smoothness Tear index	+ + -	▪ Mixtures are refined to specific tensile index ▪ High HW% needs more refining to reach wanted tensile index	Manfredi, 2006
Euca/SW	Light scattering Tensile index Tear index Fracture toughness	+ - - -	▪ Type of SW affects results ▪ Refining affects results	Mansfield, Kibblewhite, and Riddell, 2004
Euca/SW	Folding endurance Tear index Tensile index Burst index	+ - - -	▪ Wet papermaking process ▪ Tensile index first decreases, then remains quite stable ▪ Optimal ratio 94:6 (HW/SW)	Miao et al., 2023
Euca/SW	Folding endurance Tear index Tensile index Burst index	- - - -	▪ Solution casting ▪ All properties reach best results around 85% HW ▪ Optimal ratio 88:12 (HW/SW)	
Euca/SW	Air permeance Bulk Tensile index Roughness Burst index Tear index	+ + - - - -	▪ Best tensile index with ratio 25:75 (HW/SW)	Pego and Bianchi, 2021; Pego, Bianchi, and Yasumura, 2022

As seen from the table, the addition of HW to SW causes a decrease in strength properties including tensile, tear, burst index, and fracture toughness. Concurrently, increasing the fraction of HW improves optical properties like opacity and light scattering, as well as physical properties including air permeance and bulk. However, there are contradictions in the results. Chauhan, Kumari and Ghosh (2013) found that with increasing HW fraction tensile strength and burst index increased while tearing resistance decreased. All other listed papers concluded that strength properties decrease with the addition of short fibres (Manfredi, 2006; Mansfield, Kibblewhite, and Riddell, 2004; Miao et al., 2023; Pego, Bianchi, and Yasumura, 2022). The reason behind this difference could be found in refining. Chauhan, Kumari and Ghosh (2013) co-refined eucalyptus with SW. When the fraction of SW was increased refining was intensified as well. However, the refining conditions were

too intense for HW fibres, causing a decline in their strength. (Chauhan, Kumari, and Ghosh, 2013.)

Literature sources approach fibre mixtures from varying points of view. Miao et al. (2023) combined eucalyptus and SW in hand sheet forming and solution casting. In both scenarios, HW/SW ratios were tested on a scale of 100:0 to 79:21 to find the point where the addition of SW would be optimal in enhancing strength properties. The optimal ratio was 94:6 for the wet papermaking process and 88:12 for solution casting. (Miao et al., 2023.) On the other hand, Pego and Bianchi (2021) concentrated on a broader scale of HW:SW ratios from 100:0 to 0:100 and found that tensile strength was the best at a ratio of 25:75. Because of differences in all research it is difficult to compare them. It could be said that at some level, HW fibres can improve product properties while maintaining the strength of SW-based products. However, results are dependent on many factors including refining, additives and so on. (Chauhan, Kumari, and Ghosh, 2013; Manfredi, 2006; Mansfield, Kibblewhite, and Riddell, 2004; Miao et al., 2023; Pego, Bianchi, and Yasumura, 2022.) To get a better understanding of the effect of fibre types more research needs to be done with minimal treatment of fibres before sheet preparation.

6.2 Chemical pulp – mechanical pulp mixtures

As for SW/HW combinations, the idea behind chemical and mechanical pulp blends is either to reduce costs by adding cheaper mechanical pulp to expensive chemical pulp or to reinforce mechanical pulp with strong chemical pulp (Johansson, 2011; Karlsson, 2007; Retulainen, 1992). Chemical and mechanical pulps can be compared to SW and HW pulps because of their similar characteristics and behaviours. In addition to properties that mechanical pulp and HW share, the mechanical pulp has high lignin content which decreases fibre-to-fibre bonds and the strength of the material. (Heinemann and Vehniäinen, 2009; Lehto, 2004; Manfredi, 2024; Sirviö, 2008.) Similarly to SW/HW mixtures the mixing ratio is of utmost importance for blends containing chemical and mechanical pulp. However, existing literature is about understanding trends rather than investigating ideal ratios for certain properties. Table 7 below comprises said literature sources and the trends that have been observed.

Table 7. Available research on fibre mixtures with chemical and mechanical pulp.

Fibre blend	Property	Effect of mechanical pulp	Notes	Reference
TMP/Kraft (TMP: summer- or springwood)	Light scattering Density Tear index	+ - -	▪ Small changes for tensile index and stiffness ▪ TMP is either summer- or springwood	Retulainen, 1992
TMP/Kraft CTMP/Kraft CMP/Kraft	Apparent density	-	▪ Smallest changes for CTMP/Kraft combination	Görres et al., 1996
TMP/Kraft TMP/Unbleached Kraft	Tensile index Strain Tear index Fracture toughness	- +/- - -	▪ Effect of refining on pulp combinations ▪ No different ratios used; comparison is done against pure Kraft sheet	Hiltunen, 2003
TMP/Kraft GW/Kraft	Density Tensile index Scott Bond Tear index Fracture energy Light scattering	- - +/- - - +	▪ Tensile index is somewhat equal with short fibre content of 30-100 % ▪ Scott Bond decreases with short fibre content of 0-30% but increases after 30%	Honkasalo, 2004

Table 7 shows similar trends as in Table 6 for SW/HW mixtures. The addition of mechanical pulp decreases tensile index, tear index, fracture toughness, and density, while enhancing properties such as light scattering and bulk. Strain and Scott Bond were marked with +/- since the strain was not significantly affected by the addition of mechanical pulp fibres, while both increases and decreases were observed for Scott Bond at different mechanical pulp contents. Because of the differences between experiments and raw materials results shown in Table 7 should only be used as indicative.

6.3 Advantages and challenges of fibre mixtures

Fibre mixtures are advantageous in many applications compared to single fibres. They can offer improvements from different viewpoints like characteristics and applicability of product as well as economic viability. Economic viability can be improved through cost savings not only for raw materials by increasing the content of cheaper materials, but also in transportation expenses. That is because products made from only SW or chemical pulp have high density, but if bulkier material is added the product achieves desired stiffness and

thickness with lower weight. (Görres, 1996; Johansson, 2011; Karlsson, 2007; Manfredi, 2024.) Regarding characteristics and applicability, fibre blends can offer improvements in many properties and overall functionality of the product. The possibility to modify fractions of different pulps and to tailor specific properties in desired ways makes fibre blends adjustable and customisable, further enhancing their advantages. (Chauhan, Kumari, and Ghosh, 2013; Görres, 1996.)

On the other hand, despite the advantages there are multiple challenges with fibre blends. For example, while fibre blends are used to improve chosen properties, other properties are decreased. These property deteriorations are inevitable and cannot be prevented. Perhaps the most significant challenge is however the unpredictability of fibre mixtures. The effect of fibre blends and blend ratios rarely follow linear trends and all properties behave differently. This can make optimisation of the mixture rather difficult. (Honkasalo, 2004; Mansfield, Kibblewhite, and Riddell, 2004; Retulainen, 1992.)

Another factor of unpredictability is the number of different variables affecting the final properties. Chemical composition and fibre morphology, expectedly, affect results. (Bajpai, 2018g; Hakkila and Verkasalo, 2009; Manfredi, 2024; Mboowa, 2021.) However, factors like refining, age or whether the wood is summerwood, or springwood can cause variations as well. (Chauhan, Kumari, and Ghosh, 2013; Hiltunen, 2003; Retulainen, 1992). Finally, within chemical and mechanical pulps there are many different pulp types like GW, TMP and CTMP, or even bleached and unbleached which all behave differently (Görres, 1996; Hiltunen, 2003; Honkasalo, 2004). Therefore, the utilisation of fibre mixtures in different applications is a complicated process that requires a lot of knowledge and balancing between all variables.

6.4 Fibre mixtures in MFPs

There are no reports of research or utilisation of fibre mixtures in moulding applications. Despite the lack of related research, fibre mixtures could be promising to use in MFPs to increase their adjustability and further improve their suitability for different objectives. Similarly, as discussed in Chapter 6.3 fibre mixtures could also provide cost reductions to otherwise expensive moulding processes (Debnath et al., 2022; Görres, 1996; Johansson, 2011; Karlsson, 2007). However, the unpredictability of fibre mixtures could be even greater

challenge, especially for dry moulding, which already falls short in strength properties compared to wet moulded MFPs (Kaplan, Johansson, and Östlund, 2025; Pasquier et al., 2024). Because of such shortcomings, it is not desirable to implement something as unstable which in the worst-case scenario can cause more property deterioration than improvements.

Due to the complex nature of fibre mixtures, other possible approaches to combined use of different fibre types should be considered. Perhaps an easier, more predictable and more beneficial way would be the papermaking type of approach. As described before, in papermaking, different layers can be made from different fibres, for example, HW in the middle layer and SW in the top and bottom layers (Haarla, 2000; Karlsson, 2007; Kimari, 2000; Kiviranta, 2000). Thus, the best characteristics of both fibre types can be efficiently utilised without compromising predictability. In this type of method, fibres are not properly mixed, but rather blended between individual fibre sections, forming a so-called fibre gradient. In moulding applications, such fibre gradients could be formed in two directions: thickness and surface. Illustrations of thickness and surface gradients are presented in Figure 7.

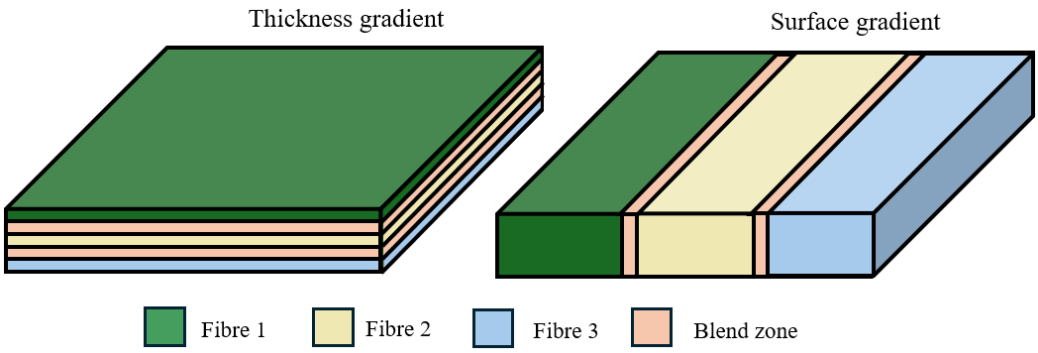

Figure 7. Thickness and surface gradients on a hand sheet. The three-fibre gradient structure can be formed from three different fibres or, for example, using one fibre on outer areas (Fibre 1 and Fibre 3) and second type of fibres in the middle (Fibre 2).

As seen from Figure 7, in thickness gradient different fibres are layered on top of each other. For the surface gradient, different fibres form completely separate sections next to one another. In MFPs, not all sections of the product require similar properties, for example strength. Areas like corners and edges are more relevant in maintaining the shape and enduring stress compared to areas such as lids or sides. Weaker structures could therefore be utilised in areas where strength is not as important. For thickness gradient, such an approach could mean a layered structure with high short fibre content while areas that require good

strength could mainly consist of longer fibres. In the case of surface gradients, strength-requiring areas could be made entirely from long fibres and other areas could utilise short fibres. However, factors including the behaviour of different layered structures and properties of blend zones need to be investigated to determine whether fibre gradients could be beneficial approach in MFP production.

7 Materials and Methods

The objective of the experimental part of this thesis was to investigate different fibre gradient structures and their effect on sheet properties, as well as to simulate how these structures could behave in moulding applications. Two types of gradient structures – thickness gradient and surface gradient - were employed with varying setups, combining short and long-fibred materials. This approach aimed to provide a better understanding of both structural and compositional factors influencing final properties. Sheets were prepared using a laboratory hand sheet former and DSF. The pulps used in the experiments are listed in Table 8.

Table 8. Pulps used in the experiments.

Application	Pulp grade	Origin
HW	Bleached eucalyptus kraft pulp (BEK)	UPM Paso de los Toros, Uruguay
SW	UPM Conifer (bleached pine kraft)	UPM Kaukas, Lappeenranta, Finland

All trials were performed using single pulp batches to ensure minimal quality variations caused by fluctuations between batches. Pulps were obtained from mills as dry sheets.

7.1 Sheet forming methods

Before sheet forming, dry pulp sheets were soaked for four hours in reverse osmosis (RO) water (conductivity ≤ 0.25 mS/m at 25°C). Soaked pulp sheets were then disintegrated into pulp slurry with a Voith-Sulzer Laboratory Refiner. The prepared slurry acted as a base solution that was diluted according to the targeted number and desired basis weight of sheets in different experimental setups. Sheets were produced using two sheet forming methods described in the following chapters.

7.1.1 Laboratory hand sheet former

The laboratory hand sheet former used is presented in Figure 1 in Chapter 4.1.1. An open white-water system with a 120-mesh wire was applied to all laboratory hand sheets prepared during the experiments.

In the first step, the furnish tank of the sheet former was filled with water until the water surface reached 255 mm above the wire. The amount of stock required for one sheet was then poured into the former, followed by 10 seconds of compressed air mixing. After mixing, the stock was left to settle for 10 seconds before the drain valve was opened. Water was drained until the water level was below the wire and then for five more seconds to remove water from the formed sheet. The drain valve was closed and the furnish tank opened for sheet removal.

Excess water was removed from the sheet using blotting papers, by placing them on top of the sheet, followed by rolling with a couch roll. Rolling was done for around 20 seconds, consisting of five rolls across the sheet. Once the sheet was removed from the wire, it was placed – together with the blotting paper attached to its surface – on top of a dry blotting paper in the sheet template. A drying plate was placed on the free surface of the sheet. These steps were repeated until the prepared diluted stock was finished, or the target number of sheets was reached.

Once the laboratory sheets were prepared and stacked in the sheet template, they were transferred to pressing. The formed pile of sheets, blotting papers, and drying plates were placed in the press. First, 410 kPa pressure was applied for five minutes to remove water from the sheets. After pressing, all blotting papers were replaced with new ones, followed by the second pressing. The second pressing applied the same 410 kPa pressure but for a shorter period of two minutes.

Final drying and conditioning were done in a temperature of 23°C and relative humidity (RH) of 50%, according to the standard ISO 187:2022. Sheets were placed inside drying arches against the fabric, while the other side of the sheets remained attached to drying plate to prevent shrinkage. Sheets were left to dry and condition overnight, after which they were detached from the drying plates, cut to the correct size (141 mm x 141 mm), and weighed to ensure the correct basis weight.

7.1.2 Dynamic sheet former

A simplified illustration of the used DSF (Andritz) is presented in Figure 2 (Chapter 4.2). First, the feed stock was prepared inside the feed tank. Two tanks were used – one for each fibre type. The feed stock in the tanks was continuously mixed with pressurised air to ensure even stock consistency and to prevent clogging.

Before starting the sheet forming process, the forming drum was rinsed and covered with a felt and wet wire cloth. The feed pressure was also checked to be under 4 bar, indicating that the spraying nozzle was not clogged. The sheet forming process started with the waterwall forming sequence, where the drum began to rotate at high speed while water was sprayed onto its lower part. As the water wall became visible as a grey interface, the sequence was stopped, and excess water was drained from the drum.

After draining, parameters including the number of strokes and drum rotating speed were defined, and the sheet forming sequence was initiated. During sheet forming, the rotating speed of the drum was 2500 rpm, while the spraying nozzle unit (jet), moved back and forth along the height of the drum, simultaneously spraying pulp slurry onto the wire cloth.

As the set number of strokes was reached, the jet stopped at the lowest position and the water removal sequence began. During the water removal sequence, water drainage was achieved through centrifugal forces by using high rotating speed for five minutes, after which the rotating drum was stopped, and the jet was moved to the up position. The sheet was removed from the stagnant drum and pressed twice to remove excess water, first under 410 kPa and then 590 kPa pressure. Final drying was done by leaving the sheets inside moulds overnight, keeping them straight and inhibiting the formation of any bumps. Before further measurements, dry sheets were conditioned in 23℃, and 50% RH.

7.2 FS5 Fibre analyser

Valmet FS5 (Valmet Fiber Image Analyzer) was used in the experiments to examine fibre properties and the size distribution of various fibre mixtures. The measured properties are listed in Table 9.

Table 9. Measured fibre properties of FS5 fibre analyser

Measured property	Unit
Length weighted fibre length	mm
Fibre width	µm
Fibre curl	%
Optical coarseness	mg/m
Kink / m	1/m
Kink angle	°
Fibre fractions	%
Fibrillation	%
Flake fines	%
Fibrillated fines	%
Fines	%
Dry weight	mg
Number of particles	pcs
Number of fibres	pcs
Number of fibres / gram	million fibre / g

FS5 analysis was conducted for all SW/HW fibre blend ratios utilised throughout the experiments, as well as for pure SW and pure HW. Samples were prepared both from disintegrated pulp slurries and hand sheets to ensure consistent fines content throughout the sheet forming process.

For wet samples, approximately 50 mL of pulp slurry was measured and diluted to 300 mL. In the case of fibre mixtures, the 50 mL volume was split between the two fibre types. For dry sheet samples, about 0.5 g of dry sheet was torn and soaked in 300 mL of water (20°C) for at least 30 minutes. The soaked sheet pieces were then disintegrated using Bamix disintegrator at the lowest speed setting for two minutes. During this time, the disintegrator was turned on and off in short cycles at different angles and heights.

Once a 300 mL dilute slurry was prepared, either from slurry or dry sheet, two to three spoonfuls were transferred into a sample container and further diluted to 100 mL. The final diluted samples were then placed in the FS5 sample container unit. All samples were analysed using the Conifer operating model, except for pure eucalyptus sample, which was analysed with the Eucalyptus operating model.

7.3 Formation of fibre gradients

Fibre gradients were applied in both thickness and surface directions. In each case, long fibres were used in the outer layers of the sheet, while short fibres were placed in the centre. A total of five different sheet compositions were used during the experiments. These compositions were based on alternating the basis weight of individual sections while maintaining a total sheet weight of 400 gsm. The compositions are presented in Table 10.

Table 10. Sheet compositions used in the experiments. In the first column, *a* and *b* are equal in weight, where *a* represents a SW layer and *b* a HW layer.

Composition number	Sheet composition	HW%	Basis weights of layers		
			Layer 1 (SW)	Layer 2 (HW)	Layer 3 (SW)
1	[2a : b : 2a]	20	160 gsm	80 gsm	160 gsm
2	[a : b : 2a]	25	100 gsm	100 gsm	200 gsm
3	[a : b : a]	33	133 gsm	133 gsm	133 gsm
4	[a : 2b : 2a]	40	80 gsm	160 gsm	160 gsm
5	[a : 2b : a]	50	100 gsm	200 gsm	100 gsm

Reference samples consisting entirely of SW or HW were also prepared to assess the impact of gradient structures on sheet properties in comparison to conventional hand sheets. Additionally, sheets with the same SW/HW weight ratios as the gradient sheets were produced, but with fibres mixed together before sheet forming rather than using them in separate sections.

7.3.1 Thickness gradient

All five sheet compositions listed in Table 10 were used to create thickness gradients. These gradient structures were formed in laboratory hand sheets by preparing separate wet sheets from SW and HW, then pressing them together before drying. First, a SW sheet was formed using the method described in Chapter 7.1.1. After formation, the sheet was removed from the wire using blotters and a couch roll. The number of blotters was adjusted to facilitate easy removal while retaining sufficient moisture for good inter-layer bonding. The SW sheet was then set aside while the HW layer was formed.

The HW layer was prepared in the same way. However, once the HW sheet was formed on the wire, the wet SW sheet was placed on top, followed by blotters and a couch roll to attach the layers. This two-layer sheet was then set aside, and the third layer (SW) was formed and attached in the same manner. Once all three layers were assembled, a drying plate was placed on top, and the layered sheet was ready for pressing and drying. An illustration of the thickness gradient formation process is shown in Figure 8 below.

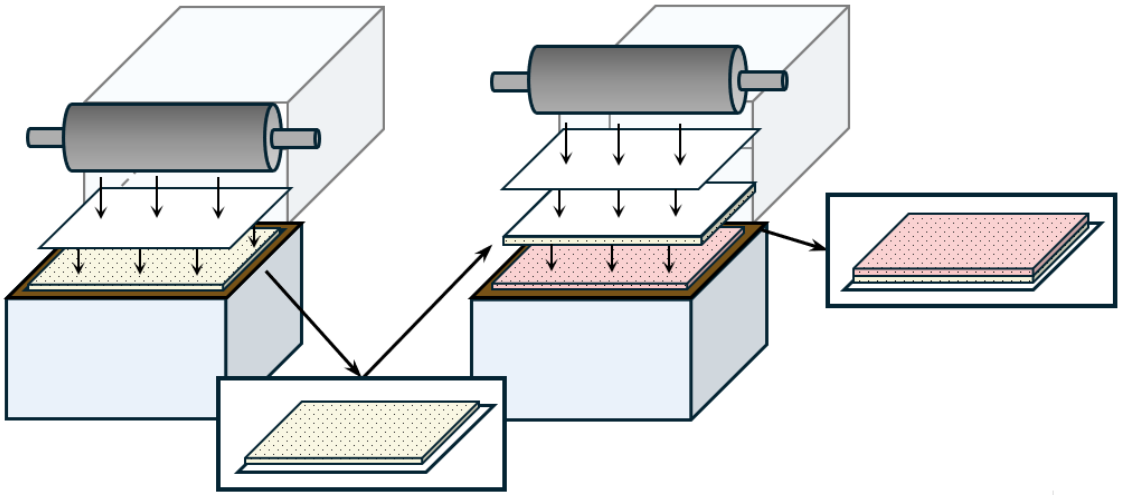

Figure 8. Illustration of the process for creating a thickness gradient in a laboratory hand sheet.

Pressing and drying were carried out as described in Chapter 7.1.1. Thickness gradient sheets were also produced using DSF. The process began by spraying SW slurry until the desired basis weight was reached. The feed was then switched to a tank containing HW slurry for the second layer, and finally back to the SW tank for the third layer. The three-layered sheet was removed from the forming drum, pressed, and dried according to the method described in Chapter 7.1.2.

7.3.2 Surface gradient

Surface gradients were created using sheet compositions 1, 3 and 5 (from Table 10). Compositions 2 and 4 were excluded because their SW sections had different basis weights, which could result in an uneven HW section during blending. The goal of the surface gradient was to produce sheets with three distinct areas: two outer sections from SW and a

central HW section. To achieve this, two partition plates were installed in the laboratory hand sheet former to divide the furnace tank into three equal sections. The height of the partition plates was defined to keep all three sections separate during mixing but allow blending during drainage between the plates and the wire. This setup enabled surface gradients to form at the interfaces between SW and HW. The described setup is illustrated in Figure 9.

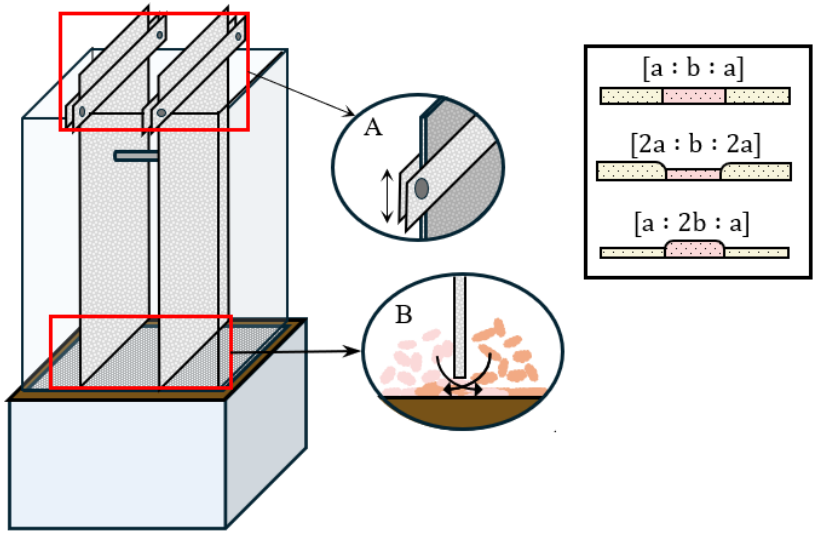

Figure 9. Illustration of creating a surface gradient to laboratory hand sheet. The three sheet compositions used are shown in the top right corner. (A) The height of the partition plates can be adjusted using plastic plates attached with a screw. (B) The partition plates do not touch the wire, allowing fibre blending between sections.

In the actual surface gradient sheets, the thickness differences between the sections were smaller than shown in Figure 9 due to fibre blending. This effect was especially noticeable in composition [a : 2b : a], where all sections appeared relatively level. The HW section (2b) blended more readily into the SW sections (a) than vice versa, due to the shorter fibre length of HW, which facilitated better blending.

In addition to laboratory hand sheet former, surface gradient sheets were formed with DSF. Unlike at laboratory scale, DSF surface gradient sheets were created with only two sections for easier operation. SW and HW were sprayed separately, alternating the area they were sprayed on – the top or the bottom half. Spraying areas were defined in a way that enabled some fibre layering and blending in the middle of the drum, creating the so-called gradient structure. The formed surface gradient compositions and their comparable laboratory hand sheet structures are shown in Table 11.

Table 11. Experimental setups for surface gradient sheets using the laboratory hand sheet former and DSF.

Fibre ratio (a = SW, b = HW)	Laboratory (isotropic)	DSF (anisotropic)
a > b	[2a : b : 2a]	[2a : b]
		[b : 2a]
a = b	[a : b : a]	[a : b]
		[b : a]
a < b	[a : 2b : a]	[a : 2b]
		[2b : a]

As seen from the table above, for each laboratory hand sheet former setup there were two DSF configurations. That is because in DSF fibres were sprayed to either the bottom or top half of the drum. Weight distribution of the fibres is often a bit uneven on the drum. To take this type of variations into account, the fibre on the different sections of the drum was alternated for each trial point. After the sheet forming, pressing and drying steps were performed normally, as in thickness gradient trials.

7.4 Hot pressing of hand sheets

A hot press (Bürkle LA 160) was used on selected gradient sheets to produce samples simulating MFPs. The objective was to identify and evaluate any significant changes in material behaviour. Pressing experiments were conducted on pure SW and HW sheets, as well as on all five mixed-fibre compositions and thickness gradient sheets. Surface gradient sheets were excluded because of their uneven thickness across the sheet, which would have led to inconsistent pressure distribution and densification during pressing.

The hot pressing was performed at a temperature of 150°C and a pressure of 160 bar. The selected temperature is within the typical range of moulding temperatures (140°C-200°C) (Macrodyne, 2025). Moulding pressure was selected to ensure the densification of the sheet. Generally, moulding can be performed at any pressure above 100 bar. In this thesis, the applied pressure of 160 bar was determined by the operational limits of used equipment. Pressing time of 10 seconds was chosen for several reasons. First, pressing durations in moulding processes vary from a few seconds to several minutes, depending on whether a dry or wet moulding process is used (Yangi, 2025). Since the pressing was performed on dry samples, the conditions simulated a dry moulding process, justifying a shorter pressing time.

Second, the equipment used was not designed for such small sample sizes (141 mm x 141 mm), and a shorter pressing time helped minimise the risk of uneven pressure distribution. Additionally, longer pressing at high temperatures could lead to fibre degradation.

Prior to pressing, all samples were conditioned in 23°C, and 50% RH. Because the Bürkle press was not located under standard conditions, the conditioned samples were sealed before pressing to retain moisture and enhance fibre bonding. Preliminary tests were conducted on conditioned sheets to ensure blistering would not occur. These tests showed no signs of blistering, and the pressing trials were performed as planned. Each sample was pressed individually using the parameters described above.

7.5 Sheet property measurements

This chapter provides details about the measurements used to evaluate sheet properties. The measured properties include thickness, bulk, sheet density, air permeance, air resistance, as physical properties. In addition, mechanical properties were evaluated using tensile strength, tensile stiffness, tearing resistance, and their index values, as well as strain at break, breaking length, and internal bonding strength.

7.5.1 Physical properties

All physical property measurements were performed in 23°C, and 50% RH. The thickness of the sheet was measured from ten parallel samples using a thickness gauge (L&W Micrometer, code 221). Bulk and density of the sheet were then calculated based on the thickness results. Sheet density was calculated using Equation (1) below.

$$d = \frac{g}{t} \tag{1}$$

Where d is sheet density [kg/m³], g is basis weight [g/m²] and t is sheet thickness [µm].

Bulk was calculated from sheet density as described in Equation (2).

$$b = \frac{1}{d} \tag{2}$$

Where b is bulk [cm³/g].

Air permeance and air resistance values were measured from five sheets for each sample set, using a porosity meter (L&W Air Permeance Tester). During the measurements, the meter created a pressure difference between the sides of the sample. The meter then measured the air flow through the sample, providing air permeance in unit [ml/min] and air resistance in [s/100ml].

7.5.2 Mechanical properties

As mechanical properties, tearing resistance, Scott bond, tensile strength, and tensile stiffness were measured. All measured samples were conditioned in 23°C and 50% RH prior to testing. Tearing resistance and tear index were measured with L&W SE 009 Tearing Tester. Sample strips of 62 mm x 50 mm were cut from prepared hand sheets, and six sample pieces were measured, one at a time, for each trial point. During the measurements, the sample was set between clamps, and a small cut was made at the bottom of the sample. A pendulum was then released to tear the sample. Tearing resistance values were acquired from the device and tear index values were calculated according to Equation (3).

$$RI = \frac{R}{g} \tag{3}$$

Where R is tearing resistance [mN] and RI is tear index [mNm²/g].

Internal bonding strength of samples was measured with Huygen 1314 Internal Bond Tester. Before analysis, samples were cut into 25.4 mm x 178 mm pieces. One sample piece was used for each trial point. To prepare the samples, double-sided tape was first placed between the setting pins on top of the anvils. The sample strip was attached to the tape, and a second layer of double-sided tape was applied to fully cover the sample strip. The upper part of the press, equipped with aluminium angles, was then pressed against the sample and tape assembly at 690 kPa pressure for three seconds. This process ensured better adhesion between the tapes and sample, as well as tape, aluminium angles and anvils. After pressing, the upper press component was lifted off, and each sample strip was cut into five 2.54 mm x 2.54 mm pieces. These sample pieces were individually placed in the testing fasteners. A pendulum was released to strike the aluminium angle and to split the sample piece along the bonded area. Results were obtained in the unit [J/m²].

Tensile properties were measured with TIRA test 2820 device. For each trial point measured with TIRA, 10 sample strips of 15 mm x 141 mm were prepared. Universal clamps to secure the samples during testing. During the measurements sample pieces were positioned individually between the clamps and samples were pulled until break. Strain at break was the only property acquired directly from the results, in the unit [%]. Tensile strength was calculated according to Equation (4).

$$T = \frac{F}{w} \tag{4}$$

Where F is force [N], T is tensile strength [kN/m], and w is sample width [m].

Tensile index was calculated from tensile strength and basis weight as illustrated in Equation (5).

$$TI = \frac{T}{g} \tag{5}$$

Where TI is tensile index [Nm/g].

Tensile stiffness values were also obtained from TIRA measurements with Equation (6).

$$K = \frac{F}{\frac{A}{100} * l} \tag{6}$$

Where A is strain at break [%], K is tensile stiffness [kN/m], and l is sample length [mm].

Tensile stiffness index was calculated as follows.

$$KI = \frac{K}{g} \tag{7}$$

Where KI is tensile stiffness index [MNm/kg].

The last measured strength property was breaking length of sample strips, and it was calculated as described in Equation (8).

$$L = l * \left(1 + \frac{A}{100}\right) \tag{8}$$

Where L is breaking length [mm].

Although all above mentioned strength properties were measured and calculated, only the most relevant ones were utilised in the comparative analysis of samples. Index values were prioritised as they consider all differences in basis weights across the trial points.

8 Results and Discussion

The main objective of this thesis was to investigate fibre-based gradient structures and their influence on hand sheet properties. Two types of gradient structures – thickness and surface gradients – were created using a laboratory hand sheet former and DSF. These forming methods were employed to investigate the effects of isotropic and anisotropic fibre orientations. In efforts to simulate moulding applications, high temperature pressing was also applied to selected hand sheets to assess whether their properties differed from those of unpressed samples. The hand sheets were evaluated based on their physical and mechanical properties. Additionally, FS5 analysis was employed to evaluate fibre properties of different sheet compositions, as well as to ensure consistent fines content throughout the experiments. The pulps used in the experiments were conifer (pine) and eucalyptus pulps.

8.1 FS5 analysis of used fibres and fibre mixtures

FS5 analysis was first employed to understand the fibre length distributions of selected pulps and then to validate that similar distribution is intact in the produced sheets. Regarding the fibre length distributions of selected pulps, fines contents were also analysed to determine whether the fibre fines content would decrease during hand sheet formation compared to the original pulp slurries. In the analysis, fines were defined as fibres with length of 0-0.2 mm. Fines content reduction of few percent can be expected due to the standard error between sample batches in visual analysis like FS5 and said reduction should not have a substantial effect on measured properties.

Two types of fines contents were studied: total fines and the fines fraction of length weighted distribution. Total fines calculate the number of fine particles compared to total number of fibres, whereas in length weighted size distribution long fibres have bigger influence, taking fibre size and its effect on product performance better into account. Analysis was done on three SW/HW combinations with the highest HW content: 33%, 40% and 50%. Total fines of sheets and pulp slurries are shown in Figure 10.

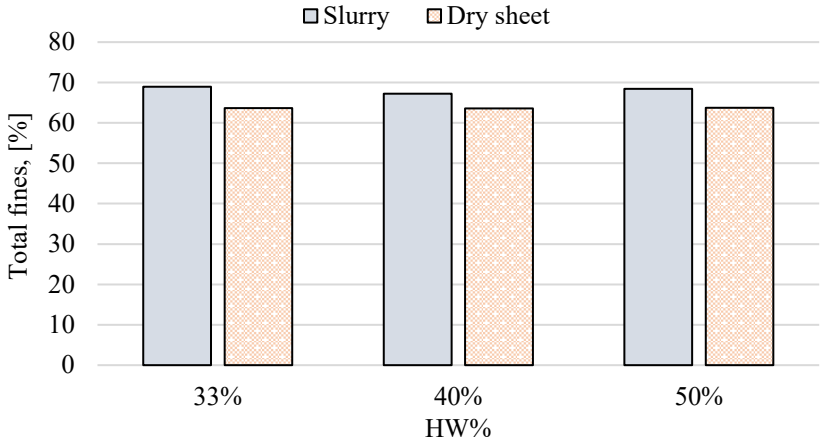

Figure 10. Effect of hand sheet forming on total fines content.

As shown in Figure 10, all three slurry consistencies behave correspondingly, regardless of increasing HW%. Fines content decreases around four percent, which is typical, and within batch-to-batch variations of pulp and open white-water hand sheet forming process and is known to have no significant effect on sheet properties. Length weighted fibre fractions of the dry sheets and slurries are illustrated in Figure 11.

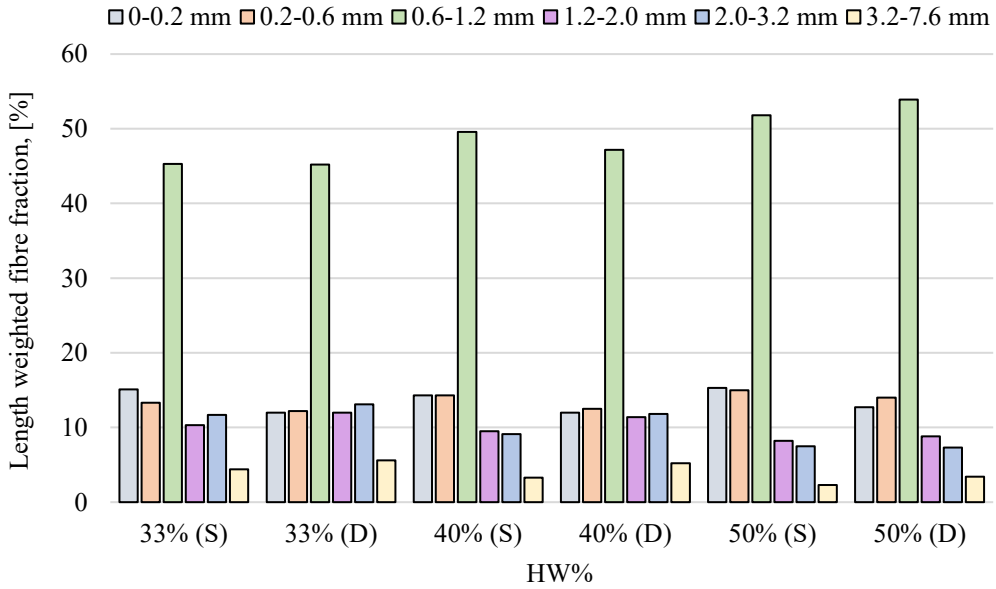

Figure 11. Length weighted fibre fractions of slurries (S) and dry sheets (D) with HW% of 33, 40 and 50.

The trends in Figure 11 mirror those observed in total fines contents. The fines fraction decreases by 2 – 3 % for all three consistencies, while the second fraction (0.2-0.6 mm) shows a reduction of one percent. Based on the results sheet consistencies remain consistent

with the original slurries. Therefore, the sheet property measurements can be considered reliable and representative.

To further investigate how different slurry consistencies affect fibre properties, FS5 analysis was performed on all five SW/HW ratios, as well as pure SW and HW. The first properties compared are length weighted fibre length and fibre width as shown in Figure 12.

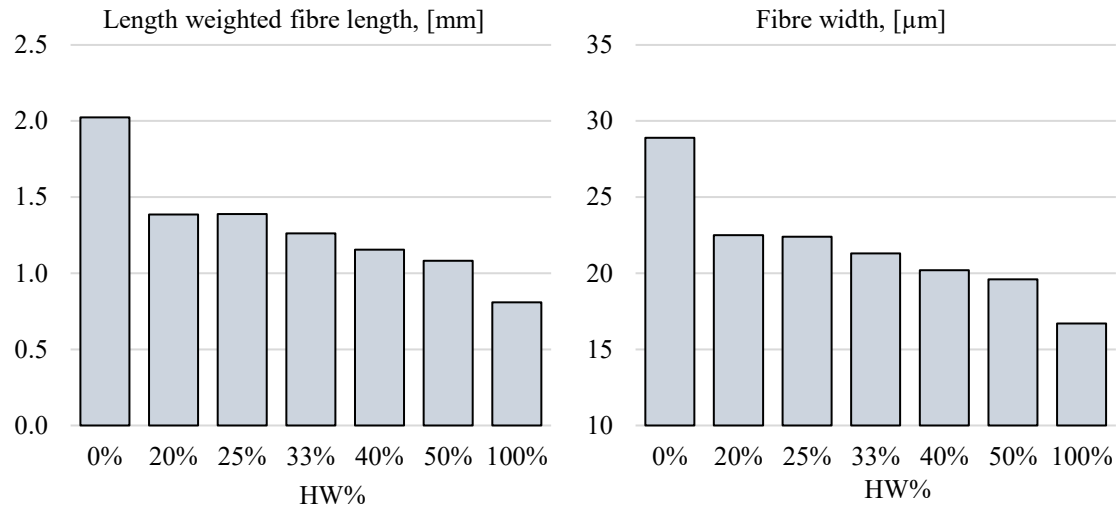

Figure 12. FS5 analysed length weighted fibre length and fibre width for different SW/HW ratios.

Figure 12 shows clear and consistent reduction in both measured properties with increasing HW content. This result is expected, as HW fibres are generally shorter and thinner than SW fibres. In fact, HW fibres were found to be 1.2 mm shorter and over 10 µm narrower.

Fines content is a critical factor affecting material behaviour. FS5 analysis not only quantifies fines content but also identifies whether the fines are flake-like or fibrillated. As discussed in Chapter 2.2, fibrillated fines enhance fibre bonding and mechanical strength due to increased surface area, whereas flake-like fines contribute less to material strength. Flake-like and fibrillated fines contents are shown in Figure 13.

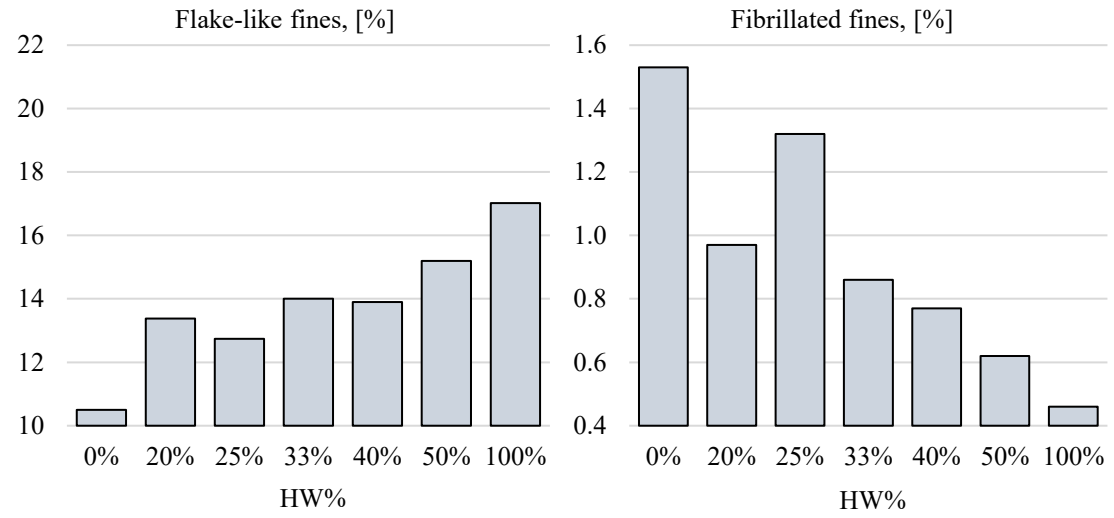

Figure 13. Proportion of flake-like and fibrillated fines in pulp slurries of different HW contents.

The results indicate that SW pulp contains more fibrillated and fewer flake-like fines than HW pulp. This aligns with the fact that typically SW materials have better strength properties compared to HW. These trends correspond with known differences between the fines of chemical and mechanical pulps: longer fibred materials seem to have more fibrillated fines, while flake-like fines are more common with short fibres. Finally, length weighted size distributions of the varying slurry compositions are compared, as shown in Figure 14. distribution.

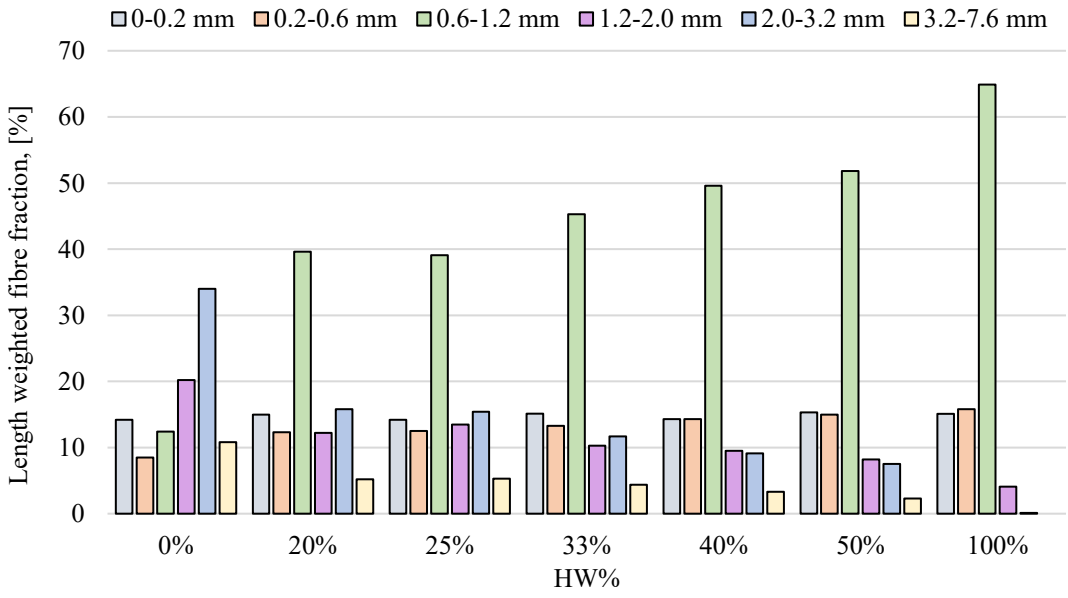

Figure 14. Length weighted size distributions with increasing HW%.

The fines fraction (0-0.2 mm) remains fairly constant for all samples. All other fractions show distinct size distribution in the pure SW sample. Overall, the changes in fibre size distributions follow logical trends with increasing fraction of shorter fibres along with rising HW content.

8.2 Comparison of layered and mixed sheet structure on sheet properties

Thickness gradient sheets were created using five different consistencies, as described in Table 10. The same consistencies were also used to create sheets where SW and HW were fully mixed together. These experiments were done to examine how sheet structure – layered versus mixed – affects properties. First, bulk and air permeance of layered and mixed sheets are compared. As discussed in Chapter 3.1, air permeance describes porosity of the material: high air permeance indicates high porosity. Preferred porosity of the material depends on its objective, but in packaging where good barrier characteristics are of high importance, low porosity is more desirable. Bulk and air permeance values are presented in Figure 15.

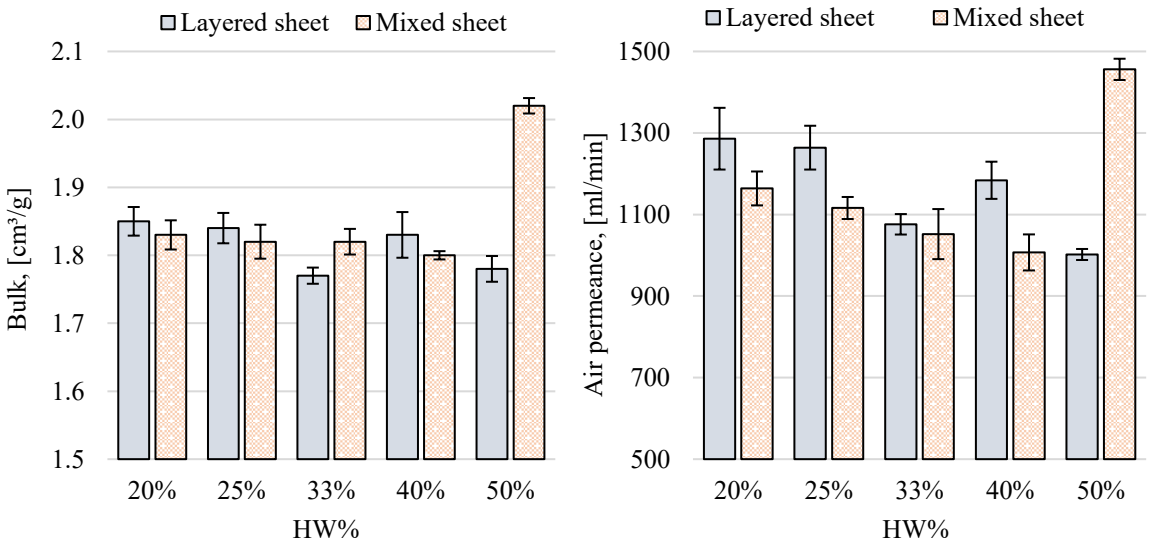

Figure 15. Bulk and air permeance of mixed and layered laboratory hand sheets with varying HW content. Standard deviations are presented as error bars.

For all HW contents except 50%, bulk values are relatively similar whether the sheet structure is mixed or layered. The mixed sheet containing 50% of HW is the only sample out of order, showing a noticeable increase in bulk. Based on these results it seems that once

HW content reaches certain level, its short and dense fibres begin to inhibit dense fibre packing, resulting in a bulkier sheet.

Porosity of both sheet structures is reduced when HW content is increased. However, layered sheets tend to be slightly more porous than mixed sheets – except for the mixed sheet containing 50% of HW, which shows unquestionably higher porosity than any other samples. Higher porosity of layered sheets may be due to the SW layers. Long SW fibres do not pack as tightly as HW fibres. In mixed sheets, shorter fibres can fill the voids between longer fibres, but in layered sheets, the separation of layers inhibit HW from mixing with SW, resulting in top and bottom layers with higher porosity. Existing HW layer with its relatively low basis weight, cannot compensate the effect. As for bulk, porosity of mixed sheet increases significantly when HW content of 50% is reached. Therefore, it seems that at a certain level (most likely HW content of 50%≤) the amount of HW fibres exceeds the void capacity, created by SW. Because of the stiffness of the fibres, the surplus of HW starts to create a more porous structure.

Tensile index and tensile stiffness index are very important properties for packaging applications, as they ensure that the material remains intact and endures stress. Tensile and tensile stiffness index values of measured samples are presented below.

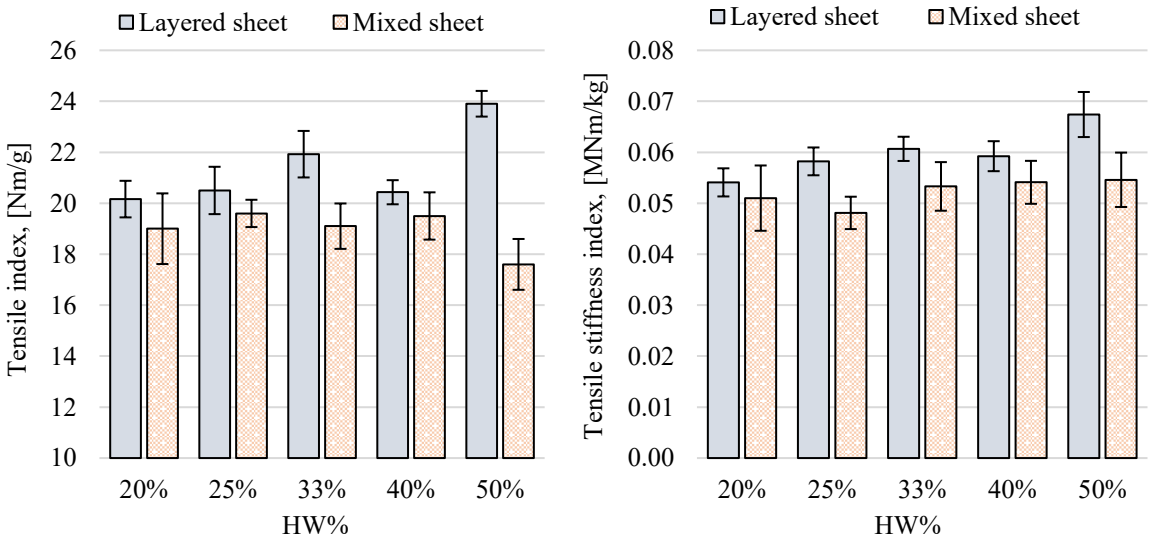

Figure 16. Tensile index and tensile stiffness index of mixed and layered laboratory hand sheets with varying HW content. Standard deviations are presented as error bars.

Results show that layered sheets have consistently higher strength than mixed sheets. While the strength of mixed sheets remains relatively constant for all five samples, the layered sheets show a steady improvement in both properties with increasing HW content.

These results are promising and indicate the potential of layered structures to enhance mechanical properties of sheets. In addition to strength properties, tear strength and internal bonding strength were also measured, as shown in Figure 17.

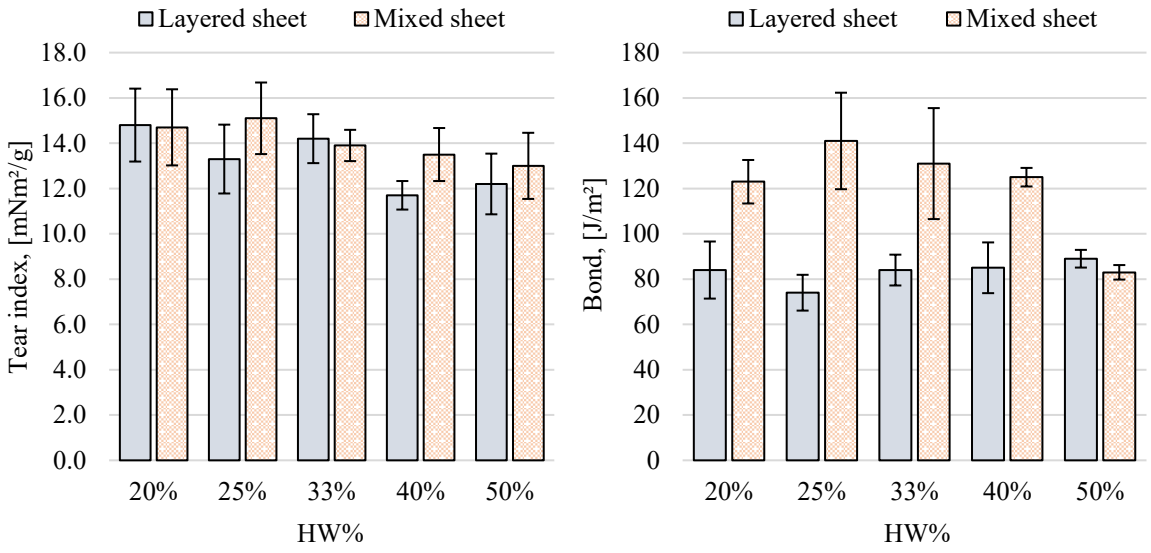

Figure 17. Tear index and internal bonding strength of mixed and layered sheets with varying HW content. Standard deviations are presented as error bars.

Tear index results do not show significant differences between the two sheet structures. Although mixed sheets may have slightly better tear strength, the standard deviations in the data make it difficult to draw conclusions. On the other hand, internal bonding strength shows a lot clearer trends. Mixed sheets seem to have a lot better bonding, first with a noticeable peak when HW content is increased from 20 to 25%. After 25 % bonding strength declines. Layered sheets show the opposite behaviour with less significant variations.

Small variations in layered sheets could result from their formation process. Since the wet layers are formed separately and then pressed together, inter-layer bonding might not develop as well as in mixed hand sheets. In addition, the first layer is couch rolled twice during its attachment to the middle layer, while the third layer is rolled only once, possibly resulting in weaker bonding. However, it does seem that when HW% is increased, the bonding strength of layered sheets will eventually develop bonding strength as good or even better than mixed sheet's.

The performance of mixed sheet structures exhibited unpredictability through sudden peaks, inconsistent results, and the lack of behavioural trends related to varying HW content. Similar behaviour has been reported in previous research on blended sheets, regardless of whether chemical/mechanical pulps, or SW/HW mixtures were used (Honkasalo, 2004; Mansfield, Kibblewhite, and Riddell, 2004; Retulainen, 1992). Some studies have attempted to improve fibre mixture predictability by developing calculation models, using refined fibres instead of pure fibres. However, the accuracy of formed correlations has varied depending on the specific properties and mixing ratios. (Chauhan, Kumari, and Ghosh, 2013; Mansfield, Kibblewhite, and Riddell, 2004; Pego and Bianchi, 2021; Retulainen, 1992.) Although the studies primarily utilised refined pulps rather than pure fibres, they – along with this thesis – effectively demonstrate the unpredictable nature of fibre mixtures.

In contrast, layered sheet structures demonstrated a more consistent relationship between HW content and sheet performance. These findings suggest that combining HW and SW fibres in a layered structure offers a promising alternative to mixed sheet configurations. The layered configuration also indicated strength improvements, with increased tensile index and tensile stiffness index values. Consistent results were reported by Radomska, Olejnik, and Pospiech (2024), who investigated mixed and layered sheet structures with a basis weight of 150 g/m². Their study compared one-ply and three-ply mixed sheets to three-ply layered sheet, which is comparable in composition to the 33% HW layered sample used in this thesis and found the layered structure to have better strength than both mixed sheet configurations (Radomska, Olejnik, and Pospiech, 2024). Given that basis weight is not expected to significantly influence sheet behaviour, and the study utilised similar sheet structures and fibre types, their findings further support the results presented in this thesis.

Although internal bonding strength was lower, it could be explained with the sheet formation method rather than the structure itself. The effects of HW and SW content, as well as the influence of thickness gradient composition on sheet properties, will be discussed in more detail in the following chapters.

8.3 Effect of thickness gradient structure on sheet properties

Five different sheet compositions with thickness gradients were created. In addition, pure SW and HW sheets were formed for comparison. This section investigates the effects of SW and HW content, as well as sheet composition on various sheet properties.

8.3.1 Effect of softwood and hardwood on thickness gradient sheets

Experiments were done to investigate, how increasing SW or HW content influences sheet properties. Experiments were done with layered sheets and total basis weight of 400 g/m². For the experiments, comparable sheet compositions were selected. Compared sheet compositions are shown in Table 12 below.

Table 12. Samples used to investigate the effects of SW and HW on thickness gradient sheet properties.

Effect of SW, samples	**SW content**
[a : 2b : a]→[a : 2b : 2a]	50% → 60%
[a : b : a]→[a : b : 2a]	67% → 75%
Effect of HW, samples	**HW content**
[2a : b : 2a]→[a : b : a]	20% →33%
[a : b : a]→[a : 2b : a]	33%→50%

The effects of increasing SW and HW content are presented side by side in the following figures. Changes in tensile index values are shown in Figure 18.

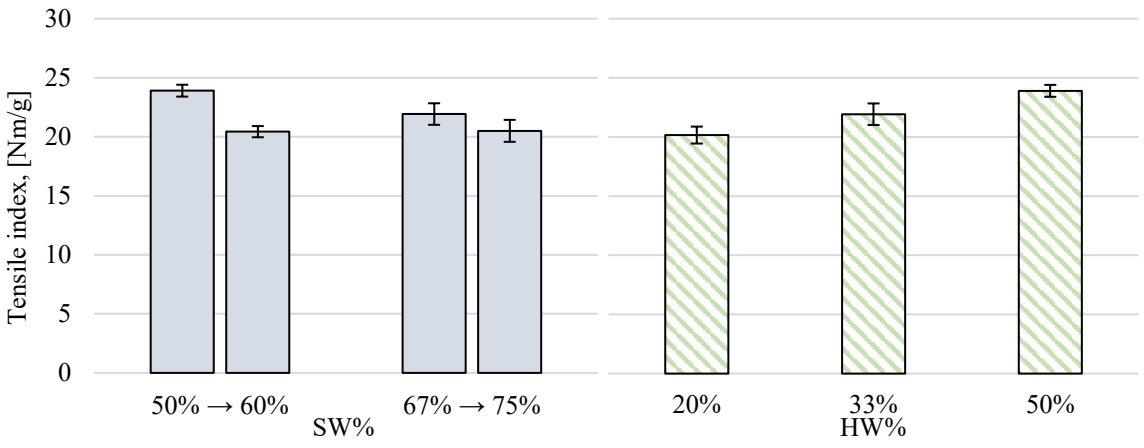

Figure 18. Effect of increasing SW content (left) and HW content (right) on tensile index. Standard deviations are presented as error bars.

The results show consistent trends: increasing SW content in layered structures leads to a decrease in tensile index, while increasing HW content results in slight improvements. Next figure represents changes in tensile stiffness index values.

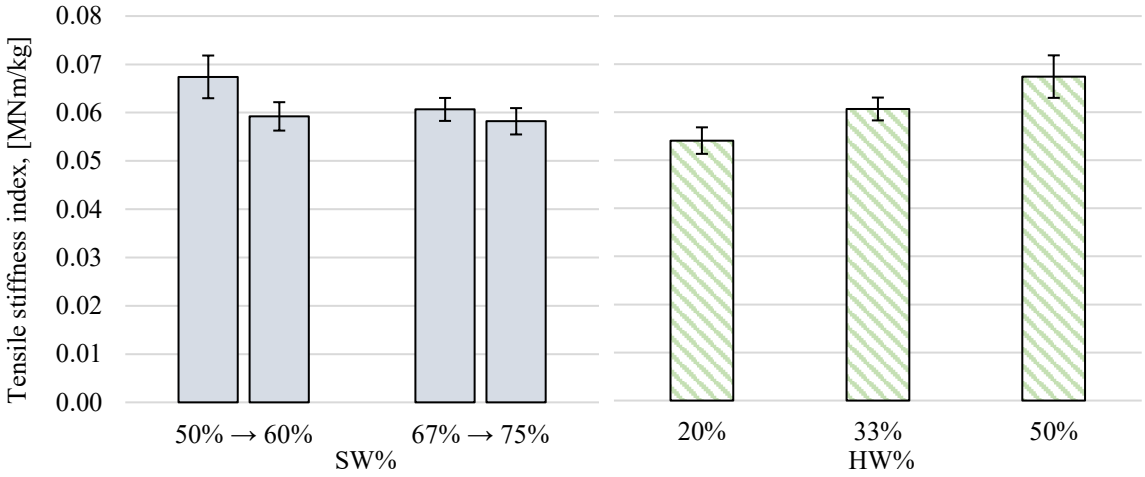

Figure 19. The effect of increasing SW content (left) and HW content (right) on tensile stiffness index. Standard deviations are presented as error bars.

Tensile stiffness index follows a similar pattern. Higher HW% has a positive effect on both tensile and tensile stiffness index values. The final compared property, air permeance, is presented in Figure 20.

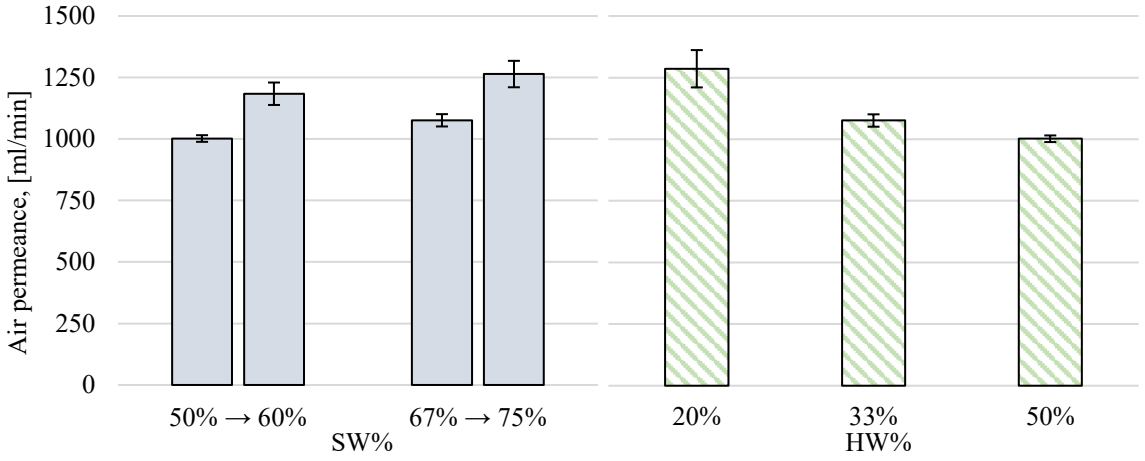

Figure 20. The effect of increasing SW content (left) and HW content (right) on air permeance. Standard deviations are presented as error bars.

As the results show, air permeance (porosity) increases with higher SW%. This phenomenon is due to the long fibres of SW that create pores to sheets, as already explained in Chapter

8.2. On the contrary, addition of HW fibres, being shorter, tend to fill these gaps, creating a denser, less porous sheet.

As discussed in Chapter 3.2, addition of HW is often associated with reduced strength. However, the results of this thesis showed a contrary behaviour. Increased SW content did not provide significant advantages, while sheets with higher HW content exhibited improved performance. Chauhan, Kumari, and Ghosh (2013) similarly found an increase in tensile strength with higher HW contents. In contrast, other studies have observed the opposite behaviour, where increasing HW content led to reduced strength and increased porosity (Mansfield, Kibblewhite, and Riddell, 2004; Miao et al., 2023, Pego and Bianchi, 2021). It is important to note, however, that the mentioned studies focused on mixed sheets produced from refined SW and HW pulps, making their findings not directly comparable to those of this thesis.

Since mixed sheets did not illustrate corresponding advantages, as discussed in Chapter 8.2, the described improvements must be caused by the structural change of the sheet. This makes the utilisation of HW potentially favourable approach in layered sheet structures.

8.3.2 Effect of thickness gradient composition on sheet properties

Previous chapters have highlighted the advantages of layered structures over mixed sheets. Moreover, increased HW content showed beneficial behavioural changes, strengthening the appeal of gradient structures as a way to enhance the economic viability of the moulding process. In this chapter five thickness gradient compositions are compared to determine the most favourable structures. In all figures, samples are colour coded based on the comparability of the external SW layers in sheet composition: 20%, 33% and 50% HW are labelled *blue*, while 25% and 40% are labelled as *orange*. Measured tensile index, tensile stiffness index, tear index, and bond strength values are presented in Figure 21 below.

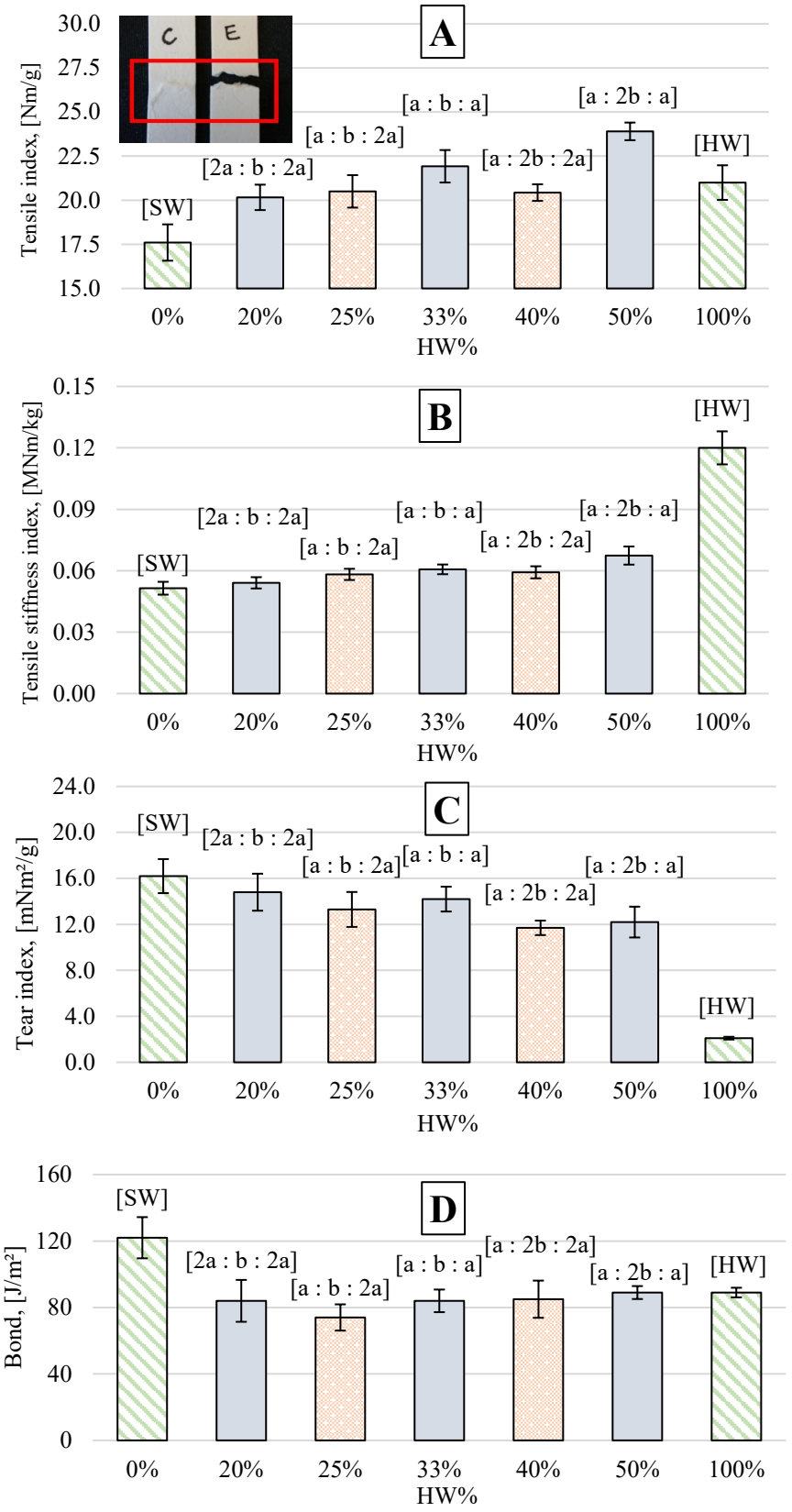

Figure 21. Tensile index (A), tensile stiffness index (B), tear index (C), and bond strength (D) values of different thickness gradient structures. Sheet structures are shown for all HW contents. Comparable structures with similar external SW layers are colour coded and referred to as *blue,* and *orange* samples. Error bars represent standard deviations.

Figure A shows that pure SW exhibits the lowest tensile index. This, however, may not be accurate due to improper breakage of SW samples during testing. While HW samples fractured completely, SW samples remained intact, showing only minor breakage, as illustrated in the inset of Figure A. Although the area of the fracture is visible, the SW samples could sustain significantly more stress before complete break. As tensile index values describe the stress at full breakage for HW and only the initial fracture for SW, the results are not entirely comparable. In moulding applications, partial fractures may not compromise product functionality if the structure remains intact. Therefore, despite the lower measured tensile index, SW may offer more favourable performance in the application.

A similar breaking behaviour was observed in the layered samples, as the SW layers remained intact while the middle HW layer broke. This suggests that the layered structure contributes positively to overall mechanical strength, with the SW layers enhancing durability under stress. Among the gradient samples, the blue series shows clear improvements in tensile strength with increasing HW content, whereas orange samples remain relatively unchanged. These differing trends imply that the improvements are primarily driven by structural changes rather than fibre content alone.

As expected, pure HW sheets demonstrate the highest tensile stiffness (B) due to the short and stiff HW fibres. Other samples show only minor variations, with a slight positive trend in the blue series. Although the stiffness of all other samples is significantly lower than that of pure HW sheet, the objective is to partially replace the more expensive SW with cheaper HW. From this perspective, comparing the layered samples with pure SW sheets is more relevant and the results show a positive development in stiffness performance with increasing HW content in layered structures.

Tear index (C) behaves quite the opposite to presented tensile index and tensile stiffness index. Pure HW show the lowest tear strength, which is consistent with the known influence of fibre length: short fibres provide less resistance to tearing than longer fibres. As HW content increases, tear strength decreases. However, blue samples maintain tear strength more effectively than orange samples, indicating a more optimal structure.

Pure SW has evidently the best bonding strength (D), due to the longer fibres and enhanced fibre-fibre bonding. In layered structures, bonding strength improves with increasing HW

content, although the trends differ slightly between the blue and orange series. Neither of the structures demonstrate a clear advantage, suggesting that both structures offer comparable bonding performance. Lastly, air permeance is presented in Figure 22.

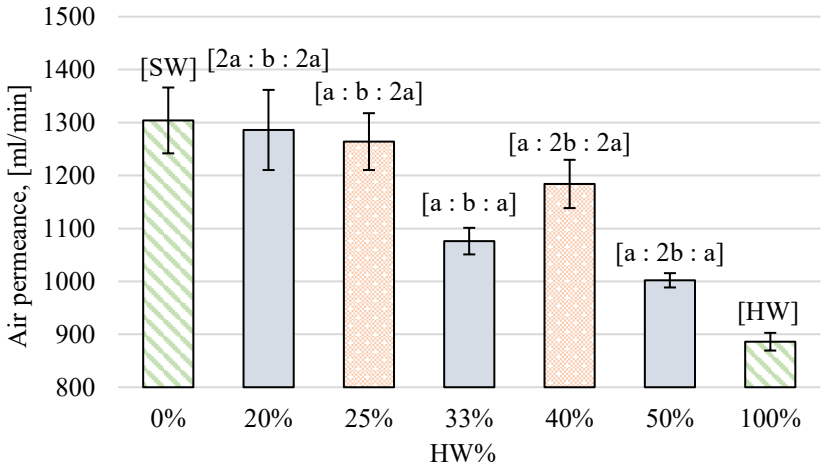

Figure 22. Air permeance values of different thickness gradient structures. Comparable structures with similar external SW layers are colour coded and referred to as *blue,* and *orange* samples. Error bars represent standard deviations.

Air permeance is the highest for pure SW. Once HW content is increased porosity starts to decrease reaching its lowest point with pure HW. Again, blue and orange samples behave similarly but porosity of orange samples reduces less than of blue samples. Based on these results, the uneven distribution of SW in orange samples inhibits pore blockage, caused by HW, more efficiently.

As described in Chapter 8.3.1, existing research on the combined use of fibres primarily focuses on fibre mixtures rather than layered structures (Chauhan, Kumari, and Ghosh, 2013; Mansfield, Kibblewhite, and Riddell, 2004; Miao et al., 2023, Pego and Bianchi, 2021). Although these studies are not directly comparable to this thesis, they generally report a reduction in strength with the addition of HW, with the exception of Chauhan, Kumari, and Ghosh (2013), who observed the opposite behaviour. Radomska, Olejnik, and Pospiech (2024) observed that layered structures are generally more favourable compared to single-layer or mixed fibre structures. However, research specifically addressing the effects of varying layered structures remains limited. Therefore, the observed positive influence of increased HW content in this study is likely due to the structures rather than fibre composition.

In conclusion, among the five gradient structures, the blue samples clearly stand out. In these samples, the increased HW% leads to a more substantial improvements in most mechanical properties. Tear index is the only property that suffers from deteriorations, while other essential strength properties improve. In addition, porosity values exhibit favourable, decreasing trends. Although the orange samples follow similar trends, their overall performance is weaker and shows less variation in response to structural changes. Based on these findings, the blue samples demonstrate the most promising results and therefore, they were selected for further evaluation in DSF trials.

8.4 Effect of surface gradient structure on sheet properties

As mentioned previously in Chapter 7.3.2 and illustrated in Figure 9, only three different sheet structures were used to form surface gradients on laboratory hand sheets. These structures had HW contents of 20%, 33% and 50%, achieved by varying the basis weights of different fibre sections while maintaining a total basis weight of 400 gsm. Properties of the sheets were measured across three to five zones, depending on the analysing method. The sheets were divided into SW, HW and a blended area. Properties of the blended area were measured either across the entire section or separately from the SW and HW sides of the partition walls. For tensile and tensile stiffness tests, the cross direction (CD) of the sheets was also analysed. The layout of these sections is illustrated in Figure 23.

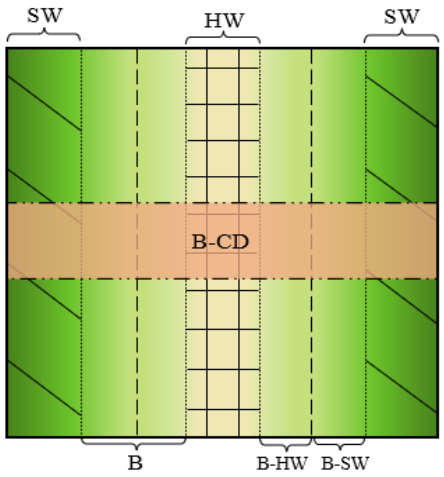

Figure 23. Different fibre areas of surface gradient sheet. Letter B represents blended area where both SW and HW fibres are likely to mix together. B-HW and B-SW illustrate HW and SW sides of blended area (B). B-CD describes a sample piece cut in cross direction.

It should be noted, that in surface gradients different HW% samples cannot be properly compared due to their differing fibre contents. For example, the HW area has more fibres in 20%, and 50% HW content compositions. Therefore, the middle of 20% HW sheet is expected to behave entirely different than the one of 50% HW sheet. The main thing to follow is that there are no deteriorations between pure and blended areas. Tensile index and tensile stiffness index values were measured from five different areas: SW, HW, B-SW, B-HW, and B-CD. Results are shown in Figure 24.

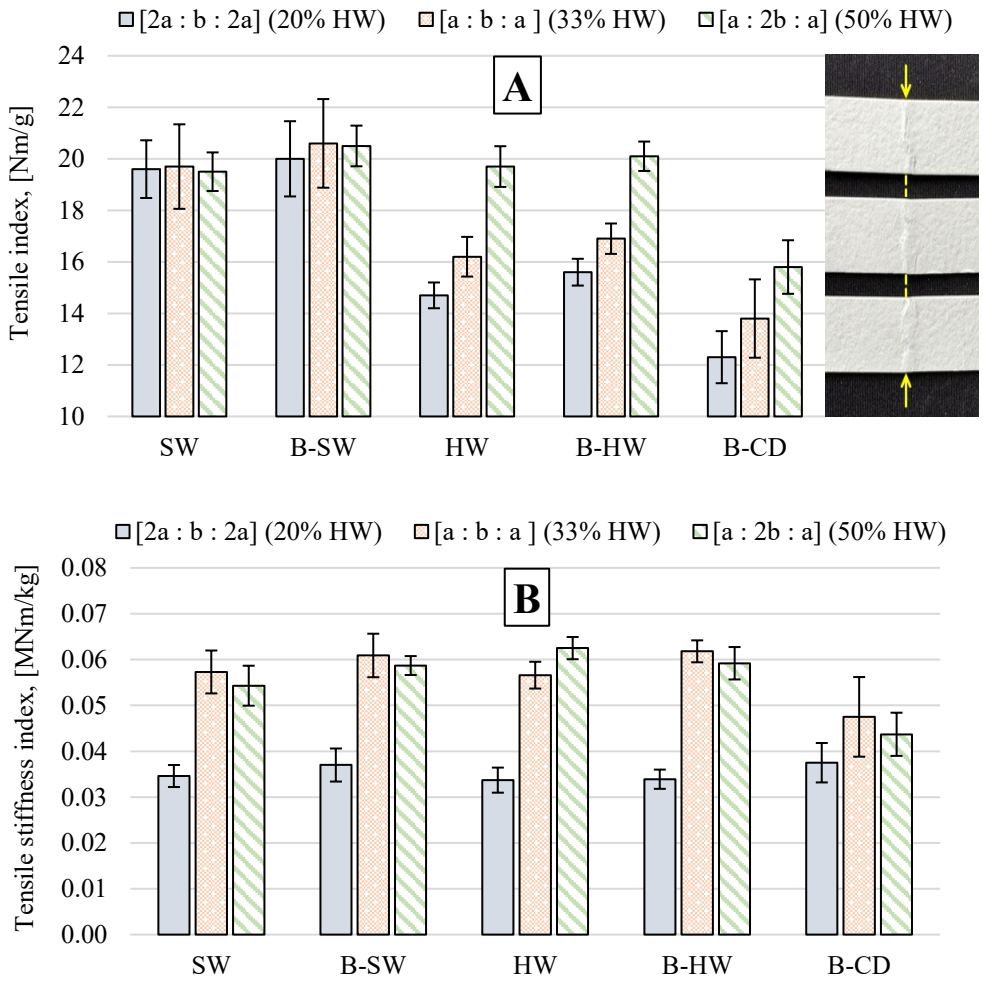

Figure 24. Tensile index (A) and tensile stiffness index (B) values for different areas of the surface gradients. Standard deviations are marked with error bars. Strips on the right show breaking points of CD samples.

Tensile index values (A) in SW and B-SW, as well as HW and B-HW regions show congruent results. Across all three surface gradient structures, blended sections outperform pure sections (B-SW/SW and B-HW/HW), although the differences are mainly within the

standard deviations. The overall lowest tensile index values are found for B-CD samples. This is, however, expected, as the samples broke consistently near the partition walls, indicating a structural weak spot, perhaps caused by restricted blending or thickness. This breaking can be seen from the right side of Figure A, as all three CD sample stripes are broken from the exact same spot. Overall, tensile index results showcase positive effects of blended area compared to pure SW and HW sections and no significant strength reductions were detected.

Stiffness results (B) show that all three sheet consistencies maintain stiffness regardless of the measured area. For sheets with 33% and 50% HW, there are small reductions in cross directional stiffness. Best stiffness values are found for 33% and 50%, which supports the idea, that high HW% can be beneficial in surface gradient structures. Next property, tear index, was measured from four areas and the results are shown in Figure 25.

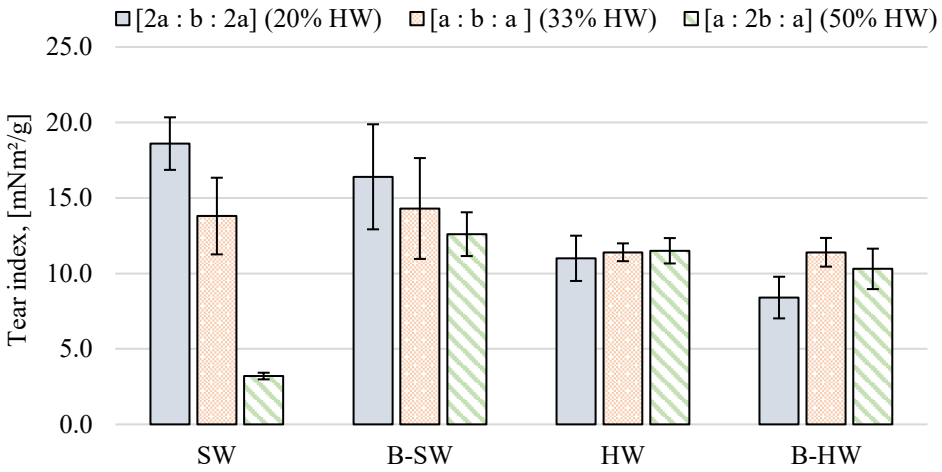

Figure 25. Tear index values for surface gradient sheets. Standard deviations are marked with error bars.

As for tensile properties, tear strength remains mainly stable across all compositions. However, small reductions can be seen in 20% HW content from both SW and HW to their corresponding blended areas. The biggest difference between pure and blended area is found in 50% HW content sheets in the transition from SW to B-SW: there is a significant increase in strength towards blending zone. These changes illustrate the structure of surface gradient sheets well: in the sheets of 20% HW content the number of fibres should decrease towards the centre (HW area), reducing the tear strength. In the case of 50% HW, the sheet behaviour is the opposite. In MFP applications, due to the need for high stress resistance, tensile

strength and stiffness are more critical than tear resistance. Therefore, seen reductions in tear resistance should not inhibit the utilisation of gradient structures in practise.

Internal bonding strength was measured from SW, HW and blended sections. However, bonding strength remained mainly unchanged and therefore, the results will not be discussed further. As the final analysed property, air permeance values were measured only for blended area and measured values were compared to pure SW and HW porosities, as seen in Figure 26.

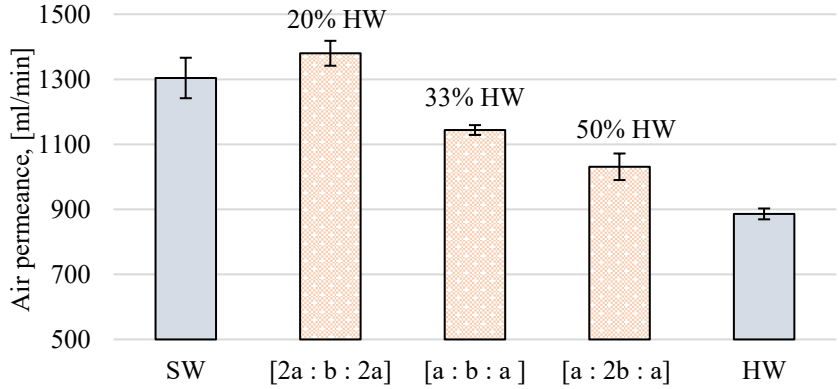

Figure 26. Air permeance values for blended area, SW and HW. Standard deviations are marked as error bars.

In surface gradient sheets porosity decreases with increasing HW, just like in thickness gradients and mixed sheets. Compared to layered structure, porosity of surface gradient sheets reduces slightly less. However, this difference is far from significant and does not need to be investigated in more detail.

Due to the novelty of the approach, previous studies employing similar sheet structures are extremely limited or non-existent. Therefore, the findings presented in this thesis cannot be directly compared to existing literature. This highlights the innovativeness of surface gradients and the need for further research on the subject. The results demonstrate promising performance in all key properties. Blended areas behave relatively good and for tensile and tensile stiffness index values, blended sections perform better than pure SW or HW areas. Altogether, no critical property deteriorations were observed. Based on the above, surface gradient structures appear applicable for objectives requiring strength and stress resistance.

8.5 Effect of hot pressing on sheet properties

To simulate the effects of dry moulding on sheet properties, thickness gradient sheets, mixed sheets, and pure HW and SW sheets – prepared using a laboratory hand sheet former – were subjected to hot pressing. The pressing trials were carried out with the temperature of 150°C, under a pressure of 160 bar, and for a duration of 10 seconds per sheet. A comparison of tensile index, tensile stiffness index, and tear index values between pressed and unpressed samples is shown in Figure 27.

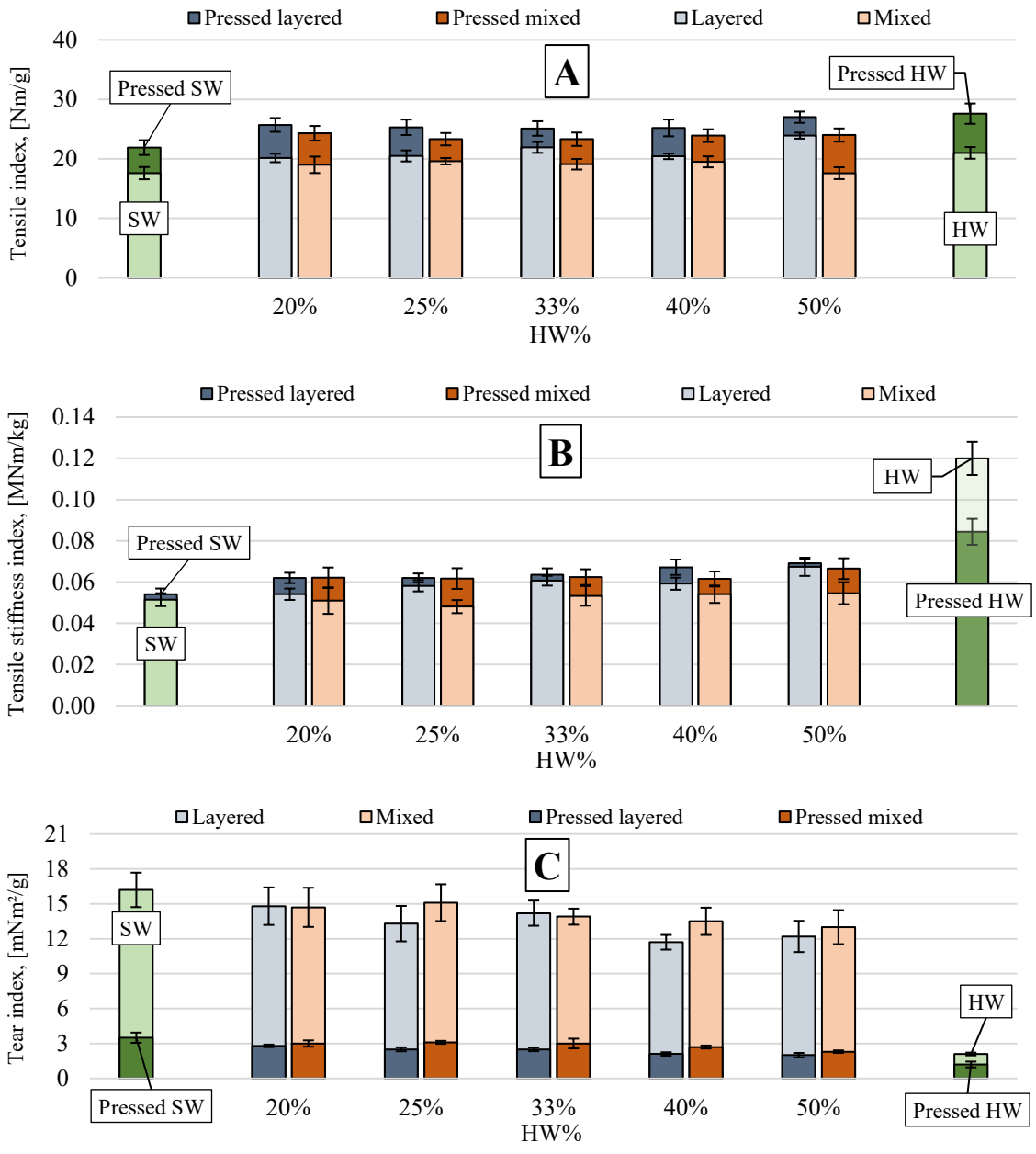

Figure 27. Tensile index (A), tensile stiffness index (B), and tear index (C) values of hot-pressed samples. Error bars describe standard deviations.

Overall, tensile index values (A) follow similar trends in the pressed and unpressed hand sheets. Layered structures show improvements compared to mixed sheets, and SW samples remain slightly weaker than HW. As observed previously in Chapter 8.3.2, HW samples break completely during tensile strength tests, whereas SW samples show only partial breakage. This suggests that SW samples are stronger than HW samples, despite them appearing weaker in tensile test results.

Hot-pressed samples have increased tensile strength compared to unpressed sheets. The improvements are not significant but still well visible. Since the pressed sheets do not have any substantial behavioural changes, and they seem to be stronger than unpressed sheets, it is reasonable to expect similar behaviour from MFPs.

Tensile stiffness (B) follows the same trends as tensile strength. The only notable exception is pure HW sample, where tensile stiffness is reduced after pressing. This behaviour is likely due to the higher thickness of unpressed HW sheets, where loosely packed stiff fibres can offer more resistance. What is also worth noticing, is that hot pressing improves the stiffness of mixed sheets more than that of layered sheets. These improvements result in nearly equal stiffness values between the two structures. However, the improvements are very small and some of them are within the range of standard deviations. Thus, tensile stiffness can be considered somewhat unchanged.

Tear resistance (C) is significantly reduced after hot pressing, except for pure HW samples, where tear index remains nearly unchanged. The reductions in tear index values can be a result of decreased thickness of the pressed sheets. Aside from this, there are no such variations that should be considered, since layered and mixed sheets behave consistently, whether the samples are pressed or not. Finally, air permeance values are shown in Figure 28 below.

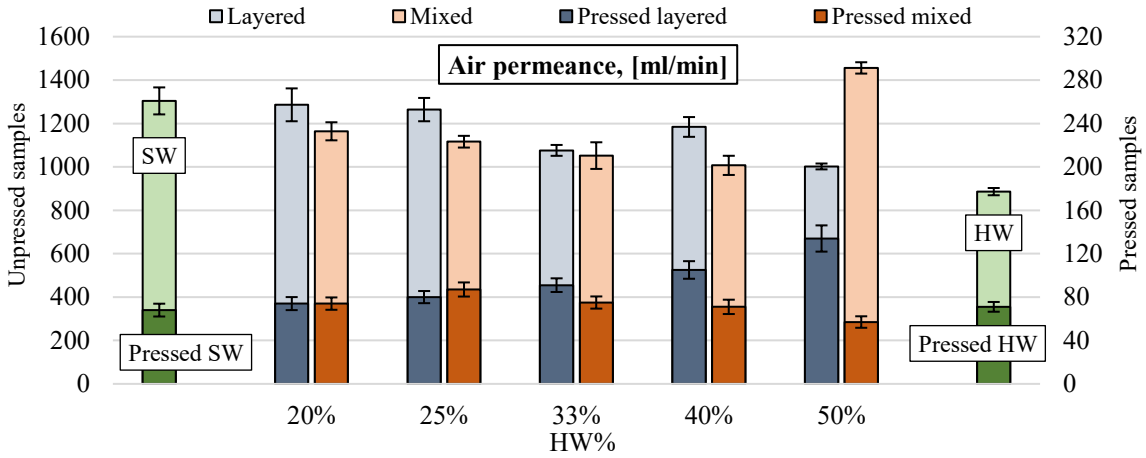

Figure 28. Air permeance values of hot-pressed samples. Error bars describe standard deviations.

Air permeance values show decreasing porosity when sheets are pressed. These results are not only desired but also expected, because of the densification caused by hot-pressing: fibres are forced to pack densely, which minimises gaps between the fibres. Unlike other properties, air permeance values do not behave alike within unpressed and pressed samples. In unpressed sheets, porosity decreases with increasing HW content. However, in pressed samples, mixed sheets maintain their porosity quite well, while layered sheets show increasing porosity with higher HW%. Based on these results, the change is caused by structural differences. It could be, that stiff HW fibres resist pressing a bit better. When the amount of HW is increased, the share of stiff fibres grows as well, creating more pores compared to high SW% sheets where pressing moulds flexible fibres towards tighter formation.

All in all, the results indicate that hot pressing does not cause significant deteriorations in sheet properties. On the contrary, it offers small improvements in strength. The only notable reduction was observed in tear index. However, as discussed in previous chapters, stiffness and tensile strength are more critical in MFP applications. Similar trends have been reported by Joelsson et al. (2020a and b), who studied hot pressing across range of temperatures and pulps with varying lignin contents, including TMP, CTMP, and unbleached kraft pulps. Their findings showed that hot pressing at temperatures comparable to this thesis, improved both tensile strength and tensile stiffness across all pulp types. Although their studies focused on single pulp sheets instead of layered or mixed structures, their results provide a good view for understanding the general impacts of the treatment. (Joelsson et al., 2020a and b.)

The findings of this thesis, together with those of Joelsson et al. (2020a and b) support the conclusion that hot pressing does not compromise, and may even enhance, product strength. Structure wise, while mixed sheets show only limited improvements, layered structures demonstrated clear advantages, even outperforming pure SW sheets. The results discussed in this chapter reinforce the idea that layered structures are potential approach to utilise in moulding applications, offering both economic savings and performance benefits through increased use of HW fibres.

8.6 Effect of fibre orientation on thickness and surface gradients

To investigate the effect of fibre orientation on key final properties, DSF trials were conducted to create both thickness and surface gradient sheets. Thickness gradient sheets were produced with HW contents of 20%, 33%, and 50%, and the results were compared to those of laboratory hand sheets. Tensile index and air permeance values of thickness gradients in both isotropic and anisotropic fibre orientations are presented in Figure 29.

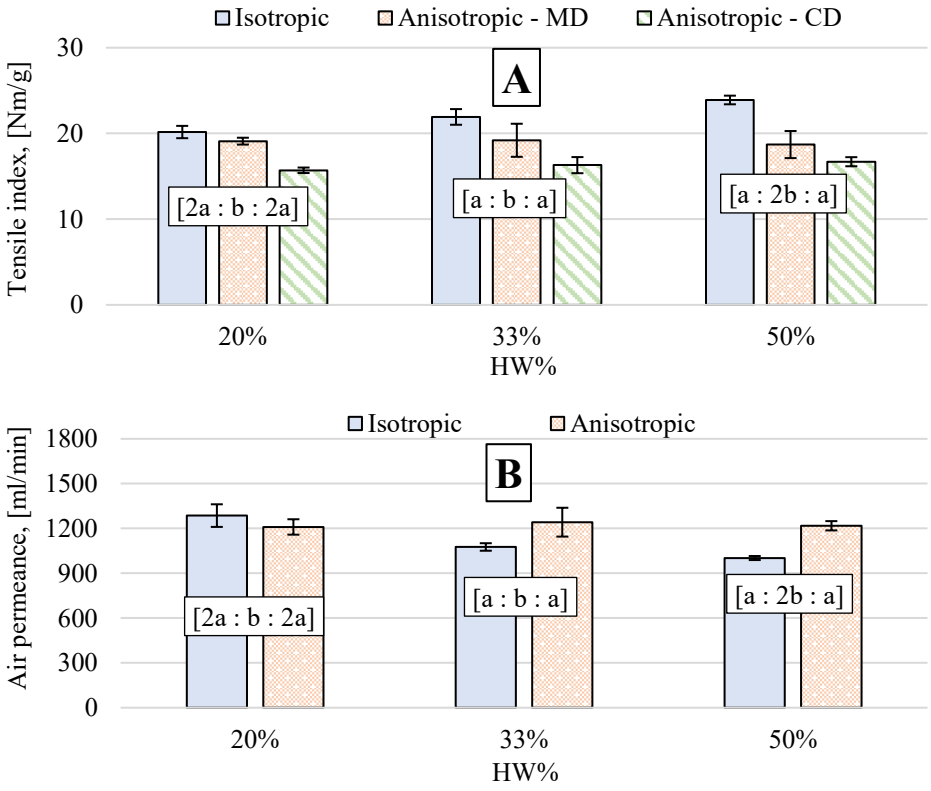

Figure 29. Tensile index (A) and air permeance (B) values of thickness gradient sheets formed with laboratory hand sheet former and DSF. Standard deviations are marked with error bars.

The results show some variation in tensile strength (A) depending on fibre orientation. The highest strength values are found for isotropic laboratory hand sheets, while the lowest is measured in CD samples. As discussed in earlier chapters, increasing HW content in layered laboratory hand sheets appears to improve sample strength. However, in anisotropic DSF sheets, no similar trends are visible – tensile index values remain consistent regardless of the HW%. Overall, although there is a small reduction in strength between isotropic and anisotropic sheets, as well as between MD and CD, the differences are relatively small, and good strengths are still achieved.

As with tensile index results, porosities (B) of isotropic sheets are slightly more favourable compared to anisotropic fibre orientation. At 20% HW content, isotropic sheets have slightly higher porosity, though the difference is negligible. With increasing HW content, anisotropic sheets maintain their porosity, while the porosity of isotropic sheets is decreased.

Based on the results shown above, isotropic fibre orientation is generally more favourable in layered structures. However, the observed reductions in strength and increases in porosity of anisotropic sheets are not severe, indicating that thickness gradient structures could be viable in moulding applications regardless of the fibre orientation.

Surface gradient structures were also created with anisotropic fibre orientation. As in Chapter 8.4, sheet properties were measured for pure SW and HW, blended areas, and CD. Tensile index values in MD and CD of formed anisotropic sheets are compared in Figure 30.

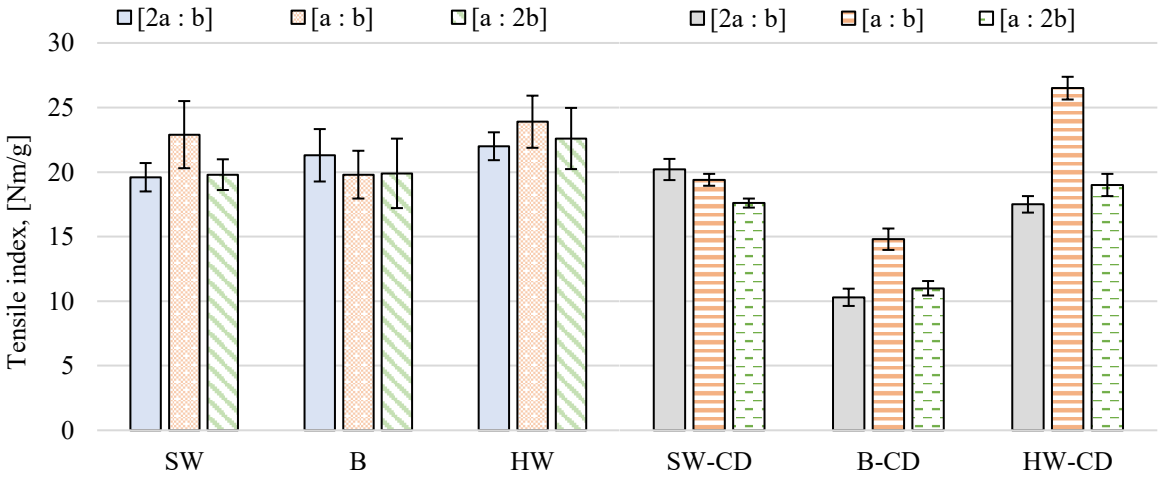

Figure 30. Tensile index values of anisotropic surface gradient sheets measured in MD and CD. Standard deviations are marked with error bars.

CD samples give slightly lower strength than MD samples, though the differences are small and often within standard deviations. The most significant change occurs in the blended area in CD. Similar to the results shown in Figure 24 for laboratory-made surface gradient sheets, this deterioration indicates a structural weak spot. In MD, the blended area was measured from a uniform strip, whereas in CD, the measurement included transition sections from SW and HW to blended area, which are where the breakage most likely occurs. Overall, the results confirm that surface gradient structures can be formed, and the properties remain consistent, regardless of measuring direction. Comparison of tensile index and porosity values for isotropic and anisotropic surface gradient sheets are illustrated in figure below.

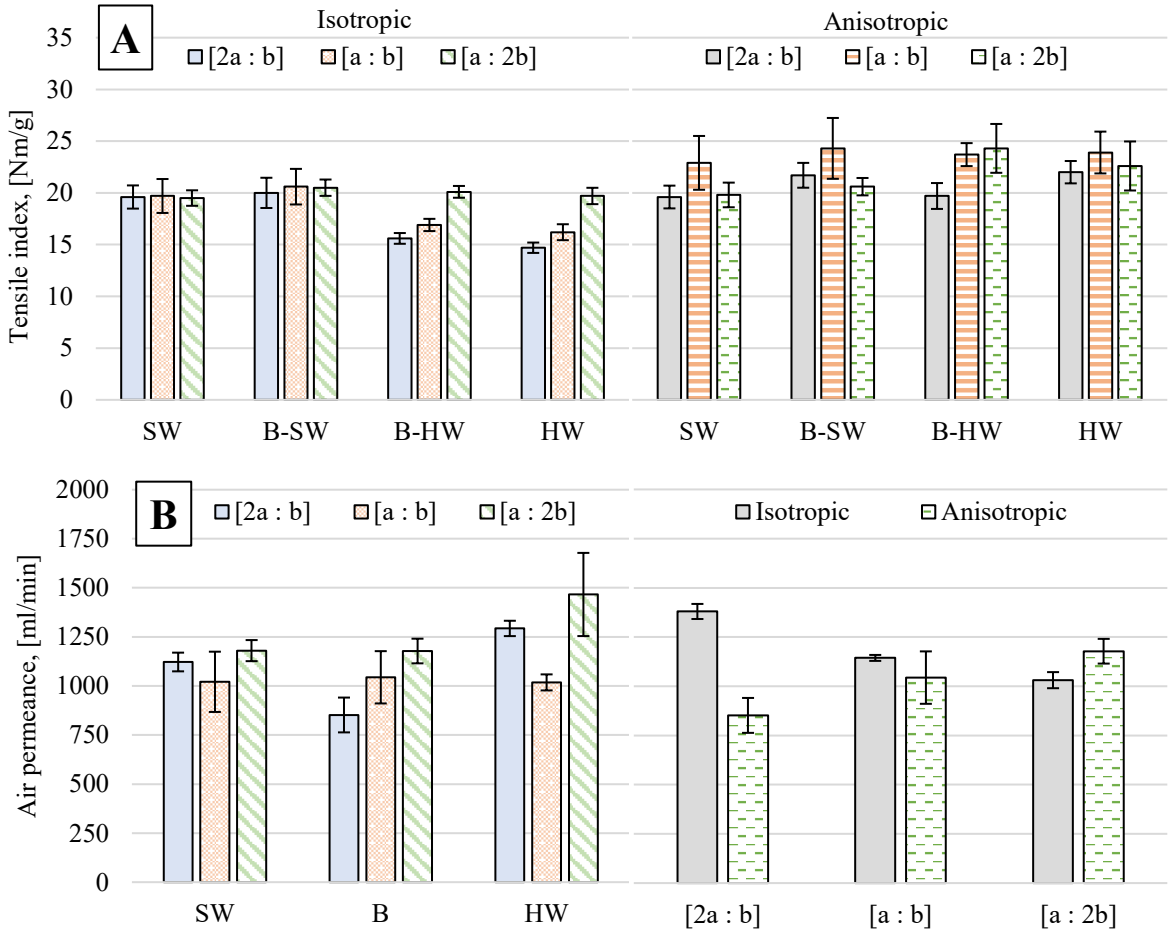

Figure 31. Tensile index (A) and air permeance (B) values of isotropic and anisotropic surface gradient sheets in different sheet compositions. The left side of Figure B shows porosities of anisotropic sheet compositions, while the right side compares isotropic and anisotropic blended areas. Error bars indicate standard deviations.

For tensile index (A), both fibre orientations have stable strength between pure and blended areas, with slight improvements in some cases. Anisotropic sheets display congruent tensile

index values across sheet compositions, but in isotropic sheets there is some variation. The variations are due to the differences in result calculations, since in isotropic sheets it was not possible to measure the basis weights of different sections. In anisotropic sheets, tensile index values were calculated with consideration of the varying section weights. Therefore, comparisons should focus on differences between pure and blended areas within each sheet composition, rather than the differences between all structures.

The air permeance (B) results on the left side of the figure show high deviation in porosity among anisotropic sheets. In the [a : 2b] composition (twice as much HW as SW), porosity increases towards the HW section. In the even SW/HW structure, porosity remains consistent, while in the SW-dominant composition, porosity initially decreases and then increases towards the HW section. Based on these mixed results, it is difficult to draw definite conclusions about porosity behaviour.

On the right side of the figure, the trends are more visible. In isotropic sheets, increasing the share of HW reduces porosity. In contrast, anisotropic sheets show increasing porosity with higher HW content. The measured area in isotropic sheets included two blended sections and the middle HW section, while in anisotropic sheets, only the blended area was measured. The difference between the measured areas is that DSF-formed anisotropic sheets had higher fibre content and basis weight in the centre. For example, in the [2a : b] composition, the middle part of the isotropic sheet is thin due to low HW content, whereas in DSF sheets, both SW and HW have been sprayed in the centre, resulting in a much higher basis weight. Thus, porosity in isotropic sheets is influenced by fibre content, while in anisotropic sheets it is defined by structural consistency.

Overall, anisotropic fibre orientation was more pronounced in thickness gradient sheets, with CD samples consistently measuring the lowest tensile strength across all sheet compositions. CD samples were also the weakest in surface gradient sheets. However, defining the exact difference between MD and CD is challenging due to the presence of clear structural weak spots in the sheets. According to Hiltunen (1999), tensile strength is a reliable indicator of fibre orientation because of its dependence on fibre alignment. It was found in this thesis, that tensile strength varied with the measured direction, indicating that some level of fibre orientation was successfully created with DSF.

Literature suggests that in anisotropic fibre orientation, long-fibred materials such as SW tend to form flocs, which create weak spots between them. In contrast, short fibres, such as those of HW, flocculate less, which can lead to more or less equal strength of HW and SW. (Brännvall and Annergren, 2009; Hiltunen, 1999; Johansson, 2011.) In anisotropic thickness gradient sheets, the results remained consistent despite the variations in HW content. This behaviour could be due to the SW fibre flocculation, which reduces the strength difference between SW and HW, thereby minimising the effect of HW%. A similar trend was observed in anisotropic surface gradient sheets, where both SW and HW had consistent strengths.

In conclusion, isotropic fibre orientation proved to be more favourable in both gradient structures with higher strength and reduced porosity. In addition, surface gradient sheets showed no deteriorations between pure and blended areas. Overall, the results align well with existing literature, suggesting that sheet behaviour follows predictable trends. Based on these findings, fibre orientation does not prevent the utilisation of fibre gradients but rather supports their potential for moulding applications.

9 Conclusions

The primary objective of this thesis was to develop and examine the effects of various fibre gradient structures as a potential structural strategy for MFPs, which could serve as sustainable alternatives to single-use plastics. Two types of gradient structures were explored in laboratory hand sheets: thickness gradients and surface gradients. Surface gradient sheets were primarily used to assess whether mechanical strength could be maintained across blended fibre regions. In contrast, the thickness gradient study involved multiple experimental setups to investigate the influence of SW/HW content, sheet composition, and fibre orientation. Additionally, hot pressing trials were conducted to simulate the behaviour of these gradients under moulding conditions.

Across all experiments, key mechanical properties were measured: tensile strength, tensile stiffness, tear resistance, internal bonding strength, and porosity. Tear and bonding strength often declined with increasing HW content. In some instances, these reductions could be attributed to high variability within the test data; however, in other cases, the deterioration was clear. In the context of moulding applications, it is essential for the final three-dimensional product to retain its shape under mechanical stress. Therefore, tensile strength and stiffness were identified as the most critical properties, while tear resistance was considered particularly relevant at the edges of the product. Bonding strength, although important, did not dominate in any specific structural region. As such, tensile properties were prioritised in evaluating the viability of gradient structures.

Thickness gradient structures were successfully fabricated using a laboratory hand sheet former. These structures outperformed mixed fibre sheets, particularly as HW content increased. Among the tested compositions, three – 20%, 33%, and 50% HW – demonstrated superior strength and reduced porosity. These configurations featured symmetrical SW layers flanking the HW core. In contrast, the 25% and 40% HW compositions, which had asymmetrical SW layering, did not perform as well. Due to their favourable performance, the three symmetrical structures were selected for subsequent trials focusing on fibre orientation.

Hot pressing trials yielded consistent results, with layered structures outperforming mixed sheets and showing improved properties with increasing HW content. A notable exception

was observed in porosity behaviour: while unpressed sheets exhibited decreasing porosity with higher HW content, hot-pressed thickness gradient sheets showed an increasing trend. This suggests that the stiffness of HW fibres can contribute to a more porous structure, depending on the layout and resist compression during pressing. Nevertheless, as expected, hot-pressed sheets were significantly less porous than their unpressed counterparts.

Surface gradient structures were also successfully produced in three different compositions. In all cases, CD samples exhibited the lowest strength, indicating a structural weakness introduced during the sheet forming process. Aside from this, the sheets maintained consistent strength across both pure and blended regions, with no significant deteriorations observed.

Both gradient types were also produced using a DSF to introduce anisotropic fibre orientation. For thickness gradients, isotropic sheets had slightly better tensile strength than anisotropic ones, though the difference was minor. Tensile strength increased with HW content in isotropic sheets but remained stable in anisotropic ones.

For anisotropic surface gradient sheets, the forming method was adapted to accommodate the operational constraints of the DSF. While the resulting structures resembled those produced in the laboratory, key differences emerged in the blended regions. In isotropic sheets, blending occurred naturally, whereas in DSF-formed sheets, the process was controlled. This distinction was reflected in opposing porosity trends: in isotropic sheets, porosity was governed by fibre content, while in anisotropic sheets, it was influenced by structural consistency. Despite the controlled forming process, a similar weak zone was observed at the interface of the blending region in CD samples, leading to reduced strength. However, tensile strength remained consistent across both fibre orientations, and the blended regions retained their strength without further deterioration.

In summary, both gradient structures demonstrated strong potential for use in MFPs. HW and SW fibres were successfully combined in various ratios and configurations without significant change in performance, suggesting that fibre gradients could offer a viable means of reducing raw material costs. Surface gradient sheets maintained consistent strength throughout, while thickness gradient sheets showed potential for performance enhancement. The observed differences between sheet compositions highlight the versatility and customisability of fibre gradient structures. This adaptability allows for tailoring sheet

properties to meet specific performance requirements – such as prioritising tear resistance or tensile strength – based on the intended application.

The positive outcomes from hot pressing and fibre orientation trials further support the feasibility of fibre gradients in MFP applications. This approach could enhance the economic and functional appeal of dry moulding processes by increasing the versatility, flexibility, and customisability of the final products. In the long term, such improvements could facilitate the broader adoption of MFPs as sustainable alternatives to single-use plastics, thereby contributing to more environmentally responsible packaging solutions.

Despite these promising results, further research is needed. Firstly, the existing research on fibre gradient structures is extremely limited. Most prior studies focus on mixed or layered fibre configurations, with little attention given to pure, unrefined fibres or their behaviour in gradient structures. Moreover, research on moulding processes tends to emphasise process parameters rather than the influence of raw material composition. As such, the findings of this thesis should be validated through additional studies.

Secondly, while this thesis presents fibre gradients as a promising approach for MFPs, their actual performance in three-dimensional products remains unproven. Although hot pressing and fibre orientation trials provide encouraging indications, the complexity of combining fibres in three-dimensional structures makes it difficult to predict final properties. Future research should therefore include practical testing of surface and thickness gradient structures in MFPs, with variations in HW content and composition across different regions (e.g., edges, sides, corners, and bases). Another rather promising avenue would be to explore the incorporation of mechanical or refined pulps into the gradient structures due to their low costs.

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