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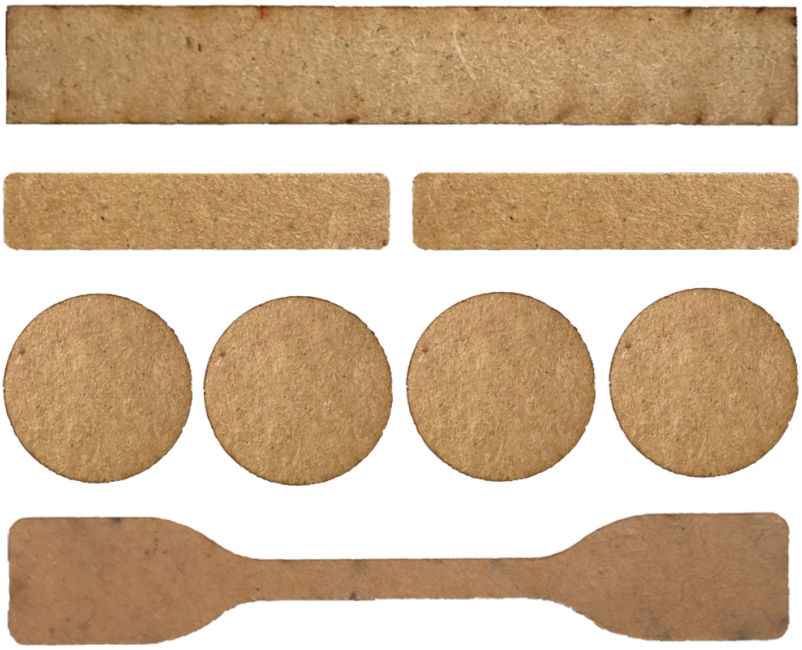

Properties of sheet formed wood fiber and polymer fiber composite materials

Master's thesis in Materials Engineering

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CHALMERS UNIVERSITY OF TECHNOLOGY
Gothenburg, Sweden 2018

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Cover: Test specimens for mechanical testing and water absorption testing

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Abstract

This project was carried out at Chalmers University of Technology in Gothenburg in collaboration with one of the largest companies in the forest industry. The company that is currently interested in possibilities to produce sheet formed composite materials with high wood fiber content, which could be formed into 3D objects by conventional thermoforming. The goal of this project was to develop a moulding process and evaluation methods to produce and analyze a novel composite material.

A search for standardized characterization methods from the leading organizations was performed and standards for different classifications of materials were compared. Considering that the end products are structural applications, a final testing protocol was designed based on the standard ASTM 1037:12 for wood based fiberboard.

A compression moulding process with four stages was developed using tensile testing to ensure the favourable parameters which enhanced strength and stiffness of the material. This process was then used to characterize 12 samples with different composition and pre-processing. Characterization was done to assess tensile, bending, internal bond strength and water absorption properties.

The optimal parameters for the process developed in this project differ between the four stages. A higher processing temperature was favourable to increase the tested mechanical properties. It was shown that the polymer content did not affect mechanical properties significantly. However, higher amounts decreases water absorption drastically. If water absorption is set aside, a polymer content of 10 % is enough to ensure easier handling of the raw material. Altogether, it can be concluded that the material is not sensitive for small changes in process parameters or composition.

Keywords: wood, composite, polymer, bicomponent fiber, air-laid, mechanical testing, water absorption, compression moulding, standardized characterization methods.

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Victoria Johansson & Ingrid Öberg, Gothenburg, June 2018

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1
Introduction

Wood is the material in history that has been used most frequently among the renewable materials on earth. Consumption of wood continues to increase in industries, in wide fields of applications [1].

This project is carried out in collaboration with a company that is currently interested in possibilities to produce sheet formed composite materials with high wood fiber content, which could be formed into 3D objects by conventional thermoforming. Thermoforming methods that could be used for this type of composite material are derived from plastic and metal sheet processing. The possibility to use standard processing is important due to interests in mass production without expensive and complex changes in the production lines.

The raw material used in this project is processed with air-laid technology, meaning that a mixture of wood and synthetic polymer bicomponent (bico) fibers are spread by air to form a mat. The synthetic bico fibers are referred to as polymeric binder throughout this report. The process is different from the regular paper making process used by the company, where water is used as carrying medium. The use of air is important to create a porous structure with a minimum of fiber-fiber bonds, which makes the material more flexible. The reason for using this type of raw material is that the flexibility is assumed necessary to form the mat into 3D geometries by compression moulding [2].

The use of paper products such as newspapers is decreasing due to digitization and therefore, new ways of using wood raw material are of interest. The applications for the end product of this new composite processing technique span a wide area including furniture [3, 4], construction and heavy packaging. Furthermore, developing this new kind of wood based composite material could introduce the company to the automotive interior market, where environmental regulations constantly gets tougher [5, 6]. A wood based composite with suitable mechanical properties would be a strong competitor to the polymer based interior currently used [7]. Low density, easy handling, recycling capabilities and a lower price are some of the benefits [8, 9].

In Sweden, certain markets have producer responsibility. This means that the producers are responsible for collecting and handling the products after functional use. The purpose of this is to motivate companies and producers to make products that are more resource efficient, easier to recycle and without hazardous substances. This applies to industries producing cars, paper and packaging, among others [5, 10]. In

order to reduce energy consumption and carbon dioxide emissions, natural fibers are seen as an alternative to synthetic fibers used in aforementioned industries [8,9]. Natural fibers promote an almost CO_2-neutral life cycle, as they can only emit the same amount of carbon dioxide the plant assimilated during its lifetime [9].

1.1 Objective

The main purpose of this project is to investigate how the mechanical properties and water absorption of an air-laid compression moulded material is affected by different fractions of polymer fibers as well as the compression moulding processing parameters. The fiber binding capacity is mainly depending on the moulding temperature, pressure and time as well as the binder content. Different combinations of moulding parameters are expected to give different results in mechanical and water absorption properties, which will be analyzed. Characterization will be performed to assess:

- ◇ Tensile properties
- ◇ Bending properties
- ◇ Internal bond properties
- ◇ Water absorption

Since there are no existing standardized methods specifically for characterization of air-laid wood based materials, another purpose of this project is to evaluate existing and applicable characterization methods for other material groups and, if necessary, modify these to suit the material. Based on the most appropriate standards, a testing protocol will be suggested.

The objective of the project can be further defined in the following research questions.

- ◇ What would be a suitable testing protocol considering that the end products are structural applications?
- ◇ How does the wood fiber and polymer fiber ratio affect the mechanical properties of the material?
- ◇ How does the moulding parameters affect the mechanical properties of the material?

1.2 Limitations

The materials investigated in this project are air-laid mats composed of two different wood pulp fibers and different contents of a polymeric bico fiber binder. The characterization methods used are mainly focused on parameters that are relevant for structural applications such as furniture and automotive interior. Due to limited access to laboratory with controlled atmosphere, the preferred testing environment might not be possible to use. Furthermore, the project is limited to equipment accessible at Chalmers University of Technology.

2
Background

2.1 Material

This section covers relevant background for the materials that have been used throughout the project.

2.1.1 Wood

Wood is composed of cells which are elongated in the longitudinal direction. Depending on their shape, the function varies and contributes to different properties such as mechanical properties and water transport properties [11]. Wood mainly consists of cellulose, hemicellulose and lignin like all natural lignocellulosic material [11–14]. Cellulose molecules are arranged in structures which together form fibrils and fibril aggregates that act as the supporting structure of the cell wall. The cellulose fibers are surrounded by hemicellulose and lignin which all together constitutes the natural composite of wood [11, 13, 14].

Pulp from Norway spruce is predominantly used for papermaking and is also the starting material used in this project. In the manufacturing of pulp, the purpose is to separate the fibers. This can be done mechanically or chemically [13]. The approximate distribution of the components in mechanical pulp is similar to that of wood and can be seen in Table 2.1 [15].

Table 2.1: Approximate ratio of components in Norway spruce [15]

Norway spruce (Picea abies)	
Cellulose	42%
Hemicellulose	27%
Lignin	27%
Extratives	4%

The different components are bond together differently. Cellulose molecules are mainly coupled to hemicellulose and lignin with hydrogen bonding while hemicellulose and lignin are coupled with chemical bonding [11]. The cellulose has crystalline and amorphous zones. The amorphous zone is more reactive than the crystalline zone, which causes a complex thermal decomposition of cellulose [14]. It has been shown that after reaching an internal temperature of 150 °C, additional heat treatment of wood at higher temperatures for 200-300 sec could increase mechanical

properties such as strength and stiffness [16].

2.1.2 Thermal properties of wood

Wood is known to develop adhesive properties when heated above a certain temperature. This transition is called thermal softening. Since wood mainly consists of the three components cellulose, hemicellulose and lignin, the softening temperatures of the material can be divided into three regions. Lignin is the component that softens at the lowest temperature, in the range of 127-193 °C, while hemicellulose softens at 167-181 °C. Cellulose is the last component to soften at temperatures above 240 °C. When combined in the complex hierarchical structure of wood, the softening temperature for the whole structure lies in the range of 180-200 °C. Thermal softening is believed to be determined by the morphology of wood as well as by chemical bonds between the components. Some softening occurs already at temperatures below 180 °C due to plasticization by water. The softening above 180 °C can be explained by two phenomena, the degradation reactions that cleaves the chemical components and polymerization of lignin. The polymerization products from lignin are highly adhesive which indicates that wood products with higher lignin content will show greater strength and stability after heat treatment above 180 °C [17].

Furthermore, it has been shown that the transition temperatures including thermal softening is depending on the moisture content in the material. There is a critical limit at 10-15 % moisture where the transition temperatures decrease drastically [17, 18]. This implicates that the temperature at which the whole wood structure starts to soften decreases from around 180 °C to 160 °C when the moisture content is greater than the critical limit. This indicates that higher processing temperatures are required for wood with lower moisture contents to ensure that adhesive properties are obtained.

2.1.3 Density effect

It is well understood that material density is an important factor for the mechanical properties [19]. Conventional fiberboard can be divided into three categories depending on their density: low, medium and high density fiberboard (LDF, MDF and HDF). The mechanical properties of wood fiber and polymer fiber composites and different types of fiberboard materials, with varying densities, are shown in Table 2.2. The polymeric binder in the composite material is PP and the ratio of wood fibers (W) to PP fibers is 90/10. The comparison with MDF is motivated by the similarity to the material being investigated in this project. Furthermore, it is often the current choice of material for some of the possible applications listed in the introduction.

In general, a lower density gives the material lower mechanical properties. This includes stiffness, tensile and bending strength as well as impact resistance. The internal bond strength decreases with decreasing density. This could be expected due to a more open structure where fibers are not as closed packed and therefore,

fewer fiber-fiber bonds are created. The water absorption however increases as density decreases. Similar changes in mechanical properties for varying densities can be seen for both W/PP composites and MDF [20, 21].

Table 2.2: Mechanical properties of wood fiber and polymer fiber composites and MDF with different densities [20, 21]

	W/PP	W/PP	LDF	MDF	HDF
Density [g/cm^3]	0.40	1.0	<0.64	0.64-0.80	>0.80
Tensile modulus [GPa]	0.53	4.20			
Tensile strength [MPa]	2.2	12.5			
Bending modulus [GPa]	0.37	2.99	1.4	2.4	3.45
Bending strength [MPa]	4.0	25.5	14	24	34.5
Internal bond strength [MPa]	0.07	0.28	0.30	0.6	0.75
Water absorption [%]	229.6	54.7		21.2	

2.1.4 Binder

The process of making loose wood fibers attach to each other can be done by adding a binder. The binder can be polymer fibers that are mixed with the wood fibers, where melting of the polymer fibers makes it possible to adhere to the wood fibers. Polymer fibers used as binder can be of bico type. A bico fiber is a fiber consisting of two components. The components can be two different polymers that are extruded separately and combined to form a single fiber, while keeping their own properties within their phase in the fiber. These kind of fibers are produced to combine properties of different materials to obtain unique and improved characteristics compared to homogeneous fibers [22]. Another benefit of using bico fibers is to combine polymers with different thermoplastic behavior. By doing this, it is possible to achieve a fiber where the core has a higher melting temperature than the sheath material. Typically, PE is used as sheath material which should melt at fairly low temperatures to bond fibers. This can be further enhanced by adding a coupling agent to the sheath polymer for a stronger attachment to the wood fibers. The core is made of a polymer with a higher melting temperature. The idea is to provide structural integrity and strength to the material by avoiding to melt the core in the binding process [23].

2.1.5 Binder content dependency

The mechanical properties are expected to change depending on polymeric binder content [7]. As seen in Table 2.3, there are quite small changes in strength even when doubling the binder content from 15 % to 30 %. However, the water absorption decreases drastically when increasing the binder content. This could be expected because there are less hydrophilic wood fibers and more hydrophobic PP fibers in the material. Based on this information, the binder content parameter is not expected to influence the strength properties but have great effect on water absorption.

Table 2.3: Mechanical properties of wood fiber and polymer fiber composites with different binder content [7]

	85W/15PP	70W/30PP
Density [g/cm^3]	1.0	1.0
Tensile modulus [GPa]	3.95	3.24
Tensile strength [MPa]	15.7	14.6
Bending modulus [GPa]	2.17	1.88
Bending strength [MPa]	34.2	34.7
Impact resistance [J]	10.3	10.8
Water absorption [%]	45.8	23.4

2.2 Air-laid technology

Structural applications containing natural fibers are mainly manufactured in large scale by compression moulding of nonwoven fibrous mats [9]. The nonwoven mats can be produced in various ways by wet-laid, dry-laid or polymer-laid technologies as well as electrospinning. These processes includes separation of fibers, arrangement into a fibrous mat, bonding of the fibers and lastly finishing to achieve the desired properties of the material [22].

In dry-laid technologies, fiber separation and air-laying are the main steps in the process. Separation processes similar to carding are done to separate and mix polymeric and natural fibers into a homogeneous fiber blend. This can be done by mechanical processing where the material moves through rotating rollers covered by metallic spikes. Mechanical fiber separation generates a web that is oriented depending on roller direction and the length of the fibers. To avoid this orientation of the fibers, aerodynamic separation can be used to get a mat with homogeneous properties [9]. The process of air-laying involves using air as a transport medium to disperse and distribute fibers onto a perforated substrate, typically a wire mesh or a fabric, to create a web of fibers [22]. After separation and air-laying, bonding of the fibrous web is required. Bonding can be achieved by needle punching, hydroentanglement, chemical bonding or thermal bonding. The process of needle punching involves mechanical treatment of the web using barbed needles which will entangle and interlock the fibers. Hydroentanglement is a similar technique which also creates bonding by entanglement, but uses water jets instead of barbed needles. Chemical bonding is achieved by using a chemical binder or resin to bond the fibers. If the binder is a thermoplastic material with fairly low melting temperature, the web can be thermally bonded. Thermoplastic binders are typically polymer fibers, bico fibers or polymer powders which are melted by calendering or hot air processes. Furthermore, molten polymer can be directly mixed with natural fibers where it is allowed to solidify and adhere [24].

2.3 Compression moulding

Compression moulding is a forming method which in its most simple form consist of a force and a two-piece mould. Usually, the process is divided into three separate stages called low pressure, high pressure and cooling. In some cases, pre-heating is included as a separate stage.

Heat and pressure are used to shape the raw material in a cavity of a mould. The raw material is usually pre-heated before it is put in the mould while the mould is heated. The heat from the mould is used to soften the binding material and pressure is applied to force the material to replicate the inner surfaces of the mould, to redistribute the material components and to form the desirable shape. The cavities of the mould typically have the shape of the part to be moulded. The temperature and pressure vary depending on the raw material, the binding material and the shrinkage intended [25].

2.4 Standardized characterization methods

Standardized characterization methods are used for safety, reliability and good quality in the development of new products and services. They enable comparison between products that have been characterized according to the same standard, which makes it possible to weigh products and results from different countries and markets against each other. The standards also acts as an insurance for the customers of the product. The two leading organizations for providing international standardized methods are The International Standard Organization (ISO) and ASTM International [26, 27].

3
Experimental

This chapter covers the experimental details of the project as well as specific characterization methods and parameters.

3.1 Standardized characterization methods

A search for standardized characterization methods from the leading organizations was performed and standards for different classifications of materials were compared. This was done to design a suitable testing protocol for characterization of the material in this project. Methods for characterizing materials such as wood based panels, polymers, composites and paper were evaluated. Major interest was devoted to wood based panels, motivated by the similarity to the material being investigated in this project. Furthermore, a wood based panel is often the choice of material for some of the applications listed in the introduction.

3.2 Material

Spruce pulp fibers of two different types, Wood Fiber 1 and Wood Fiber 2 (WF1, WF2), obtained from the company were used as the wood component in the material. Both fiber types were mechanically refined in only one stage, without any additives.

As binder, a polymeric bico fiber was used. The core of the fiber consisted of a polymer with a melting temperature around 160 °C and the sheath material was another polymer with a melting temperature around 130 °C.

A total of 12 samples with varying composition and pre-processing was used in the project. Details regarding composition are presented in Table 3.1. Samples with the same fiber type and binder content are further distinguished by other parameters from the air-laid process such as density and grammage.

Table 3.1: The composition of sample types studied

Sample	Fiber type	Binder content [%]
1	WF1	20
2	WF1	20
3	WF1	20
4	WF1	20
5	WF1	30
6	WF1	30
7	WF1	10
8	WF1	10
9	WF1	40
10	WF1	40
11	WF2	30
12	WF2	30

3.3 Sample preparation

To prepare the raw material before compression moulding, different kinds of cutting equipment were tested. Hand-held saws, scalpels, scissors and knifes did not give a satisfying result. Instead, an automatic circular saw was used which gave sharp edges and corners. After compression moulding, the test specimens were manufactured. Different methods are available on the market to create test specimens and one of the most common techniques is to use a cutting die with desired shape. Another option is to use a laser cutter.

In the present study, the test specimens used for tensile tests, bending tests and internal bond tests were cut using a laser cutter. This was motivated by the fact that the material used in this project was relatively hard and brittle and die cutting did not yield a satisfying result. The laser cutter used was a GCC LaserPro Spirit LS, with user settings, speed: 4 %, power: 60 %. For the water absorption test, a cutting die was used. The reason for changing method was to prevent the laser used in a laser cutter from affecting the cut edges which could hinder the absorption.

There were 5-10 cut specimens for each compression moulded sample leading to a total of 60-120 specimens per test method. One exception was for the internal bond tests where an entire test series could not be performed and only a few test specimens were used. A list of the geometries and number of specimens used for the different tests are given in Table 3.2.

Table 3.2: Specimen specification for different test methods

Test	Specimen size [mm]	Specimen shape	Total number of specimens
Tensile properties	85x4	Dogbone	120
Bending properties	85x13	Rectangular	120
Internal bonding	⌀ 25	Circular	-
Moisture absorption	50x10	Rectangular	60

3.3.1 Conditioning

Conditioning of the samples and specimens was done in a salt chamber where a supersaturated solution of $Mg(NO_3)_2$ gave a relative humidity of 50±5 % in ambient temperature of 24±3 °C. This was done to achieve equal starting condition for all samples and test specimens prior to processing and characterization. All samples and test specimens were kept in the chamber for at least 24 h. By oven drying the samples at 105 °C for 24 h, the moisture content of the samples after conditioning was found to be 4-4.5 % depending on wood fiber type.

3.4 Compression moulding

Compression moulding was performed to produce flat samples to be used for further characterization of the material. Prior to moulding, the samples were pre-heated in the drying chamber Binder VD 53 to evenly distribute the temperature throughout the material. The samples were moulded using a Fontijne Holland TP 200 with a capacity of 200 kN and mould dimensions of 225x300 mm. The mould was an open type, consisting of flat plates. Water cooling of the mould plates was used to ensure dimensional stability.

Development of the compression moulding process was done to answer one of the main questions of this project, regarding how the compression moulding parameters affect the mechanical properties of the material. Compression moulding can be divided into four different stages where the material is exposed to different conditions. These four stages are pre-heating, low pressure stage, high pressure stage and cooling. The optimal parameters of the stages were defined in an initial process development. Further tests were made to evaluate the influence of temperature on the mechanical properties. A temperature span ranging from 150 °C to 230 °C was investigated. Specimens of both wood fiber types with a binder content of 20 % were tested for their tensile properties to analyze the results.

To further analyze the temperature dependency for the samples containing WF1, a lower moulding temperature was used to compare with the optimal temperature defined in the initial process development. This was done to see how big changes in mechanical properties a lower temperature would generate. All WF1 samples with varying composition and pre-processing were moulded at a lower temperature and at the optimal temperature, followed by tensile testing to evaluate the properties.

3.5 Characterization

To evaluate and compare the samples with varying composition, the following characterization methods were used.

3.5.1 Tensile properties

Tensile testing was performed using a Zwick Z2.5 testing machine with flat, pneumatic grips. Testing was done according to ASTM D1037:12-10, with some modifications. The grip separation was 35 mm, the pre-load was set to 0.1 N and the loading speed was 4 mm/min. The software Zwick testXpert was used to record force and displacement data. Tensile traces were obtained by normalizing the data according to Equations 3.1 and 3.2.

$$\sigma = \frac{F}{A} \tag{3.1}$$

Where

$$\sigma = stress\ [Pa]$$

$$F = force\ [N]$$

$$A = cross\ sectional\ area\ [m^2]$$

$$\epsilon = \frac{\Delta L}{L_0} \cdot 100 \tag{3.2}$$

Where

$$\epsilon = strain\ [\%]$$

$$\Delta L = change\ in\ length\ [m]$$

$$L_0 = original\ length\ [m]$$

The tensile modulus of elasticity was calculated as the slope of the initial linear part of the tensile traces according to Equation 3.3.

$$E_t = \frac{\Delta \sigma}{\Delta \epsilon} \tag{3.3}$$

Where

$$E_t = tensile\ modulus\ of\ elasticity\ [Pa]$$

$$\Delta \sigma = change\ in\ stress\ [Pa]$$

$$\Delta \epsilon = change\ in\ strain$$

3.5.2 Bending properties

Static bending was performed using a Zwick Z2.5 testing machine with three point bending setup. Testing was done according to ASTM D1037:12-9, with some modifications. The radii of the support and crosshead tips were 5 mm. The pre-load was set to 0.1 N and the loading speed was calculated according to Equation 3.4. A support span of 60 mm and an average specimen thickness of 1.17 mm, together with the outer fiber strain rate of 0.005 min^{-1} required by the standard, gave a crosshead speed of 2.5 mm/min.

$$N = \frac{zL^2}{6d} \tag{3.4}$$

Where

$$N = crosshead\ speed\ [mm/min]$$
$$z = outer\ fiber\ strain\ rate\ [min^{-1}]$$
$$L = support\ span\ [mm]$$
$$d = thickness\ of\ specimen\ [mm]$$

The software Zwick testXpert was used to record force and displacement data. The data was plotted and the slope of the initial linear part of the trace was used to calculate the bending modulus of elasiticity according to Equation 3.5.

$$E_b = \frac{L^3}{4bd^3} \cdot \frac{\Delta P}{\Delta y} \tag{3.5}$$

Where

$$E_b = bending\ modulus\ of\ elasicity\ [MPa]$$
$$L = length\ of\ span\ [mm]$$
$$b = width\ of\ specimen\ [mm]$$
$$d = thickness\ of\ specimen\ [mm]$$
$$\frac{\Delta P}{\Delta y} = slope\ of\ the\ initial\ curve\ [N/mm]$$

3.5.3 Internal bond properties

Internal bond strength testing was performed using a Zwick Z2.5 testing machine with flat, pneumatic grips. Testing was done according to ASTM D1037:12-11, with some modifications. The grip separation was 20 mm, the pre-load was set to 0.1 N and the loading speed was 4 mm/min. The software Zwick testXpert was used to record force and displacement data. Maximum stress was calculated according to Equation 3.1.

In order to carry out the tests, an appropriate adhesive was needed. Several adhesives were tested for joining samples to disposable aluminum holders. Glue or tape was applied to the holders before they were mounted onto the specimens. The setup was then compressed for varying times and with varying pressures before testing using the tensile testing machine. The setup with aluminum holders and test specimen can be seen in Figure 3.1.

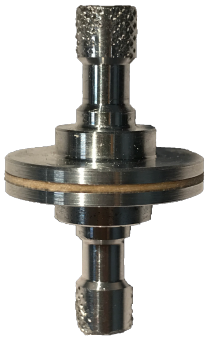

Figure 3.1: Setup with disposable aluminum holders for testing internal bond strength

3.5.4 Water absorption

Water absorption testing was performed according to ASTM D1037:12-23, using a customized setup. The setup can be seen in Figure 3.2. Method A was used and absorption after 2 h and 2+22 h water submersion was calculated according to Equation 3.6.

$$Absorption\ in\ wt.\ \% = \frac{W_{after} - W_{before}}{W_{before}} \cdot 100 \tag{3.6}$$

Where

$$W_{before} = weight\ before\ submersion\ [g]$$

$$W_{after} = weight\ after\ submersion\ [g]$$

Figure 3.2: Setup for testing water absorption

3.6 Analysis

To analyze the results obtained through mechanical characterization and water absorption, the software MATLAB was used. Graphs, curves and standard deviations were visually presented and calculations were made. Simple calculations were also made by using Microsoft Excel.

4
Process development

In the following chapter, the initial development of the compression moulding process is presented including motives, methods and results. The stages of the moulding process and the trials to determine the favourable parameters are listed below.

4.1 Pre-heating

Pre-heating is usually achieved by heating the samples in an oven. A pre-heating protocol was compiled from the company which is shown in Table 4.1 [28].

Table 4.1: The protocol for pre-heating provided by the company [28]

Pre-heating protocol	
Dwell temperature	150 °C
Heating time	8-10 min
Dwell time	5 min
Convection (fan speed)	High as possible

In order to verify the pre-heating protocol for the materials used in this study and to ensure if it was compatible with the available drying chamber, a pre-heating trial was done. A previously known difference between the equipment was that there was no fan in the drying chamber being used for this project. This could lead to some differences in temperature distribution in and between samples.

Five different samples (1, 6, 7, 9 and 12) were selected to cover the range of binder contents and fiber types. The raw material was cut in 100x100 mm squares and conditioned. The five different samples were placed, one at the time, in the drying chamber for 14 min (heating time + dwell time) at 150 °C. Afterwards, a thermo-coupling element was inserted into the middle of the sample to measure the internal temperature that had been reached. The results can be seen in Table 4.2.

None of the samples reached the internal target temperature. To improve the pre-heating, Equation 4.1 was used to calculate the pre-heating time. According to the equation, it would take infinite time to reach an internal temperature of 150 °C when the oven is set to 150 °C. Therefore, a higher temperature of 165 °C was chosen.

$$t_1 = t_2 \cdot \frac{ln\left(\frac{4}{\pi}\left(\frac{T_{material}-T_{oven}}{T_{target}-T_{oven}}\right)_{target}\right)}{ln\left(\frac{4}{\pi}\left(\frac{T_{material}-T_{oven}}{T_{measured}-T_{oven}}\right)_{real}\right)} \tag{4.1}$$

Where

$$t_1 = time\ to\ heat\ the\ sample\ to\ an\ internal\ temperature\ of\ 150\ ^\circ C$$

$$t_2 = time\ used\ during\ trials$$

$$T_{material} = temperature\ of\ sample\ before\ entering\ the\ oven$$

$$T_{oven} = temperature\ of\ oven,\ T_{target} = desired\ temperature$$

$$T_{measured} = temperature\ measured\ after\ t_2\ in\ sample$$

The results are shown in table 4.2, indicating the need for increased time to reach the target temperature. To validate if this had an effect on the results, tensile testing was performed with samples pre-heated according to the company protocol as well as according to the present study. The results showed that there were no significant difference in tensile modulus, ultimate tensile strength (UTS) nor strain at UTS. This indicated that the moulding process that followed the pre-heating was enough to reach equal performance even at lower pre-heating temperature and time, as in the protocol from the company. Thus, the provided pre-heating time and temperature can likely be used to minimize lead time without any significant difference in mechanical performance.

Table 4.2: Result of the pre-heating trial

Sample	Temperature [°C] after 14 minutes at 150 °C	Time [min] to reach 150 °C in the material at 165 °C
1	120.4	24.95
6	124.7	23.26
7	126.3	22.65
9	129.8	21.33
12	126.0	22.76

4.2 Low pressure stage

During the low pressure stage, a low pressure was applied to compress the material. This was done to ensure contact between the mould plates and the sample to increase the thermal conduction. The low pressure stage was studied by varying the pressure and time. The temperature was held constant at 210 °C to be well above

the expected softening temperature of wood. The pressures tested were 0.1, 0.5 and 1 MPa during 1, 3 and 5 min. By combining all parameters, a total of nine samples were moulded and analyzed. The material used contained WF2 and 20 % polymeric binder fibers. An overview of the parameter combinations for the different samples is shown in Table 4.3. During testing of the low pressure stage, pre-heating according to the protocol provided by the company and a high pressure stage based on literature were applied, with the parameters 210 °C, 3 MPa and 1 min.

Table 4.3: The low pressure parameters studied for compression moulding of samples

Sample	Temperature [°C]	Pressure [MPa]	Time [min]
1	210	0.1	1
2	210	0.1	3
3	210	0.1	5
4	210	0.5	1
5	210	0.5	3
6	210	0.5	5
7	210	1	1
8	210	1	3
9	210	1	5

To analyze the low pressure stage, five test specimens from each sample were cut using a laser cutter and the tensile properties were investigated. Based on tensile traces that can be seen in Figure 4.1, the preferred low pressure parameters were determined for use in further trials.

The low pressure parameters that gave the highest strength according to the tensile traces were defined as the optimal parameters. Two of the parameter combinations gave similar results, whereby they were compared in the same plot as can be seen in Figure 4.2. From this plot, sample 7 was chosen since there were less differences between the curves than for sample 1. Based on these results, the optimal parameters to use for the low pressure stage were determined to be 210 °C, 1 MPa and 1 min.

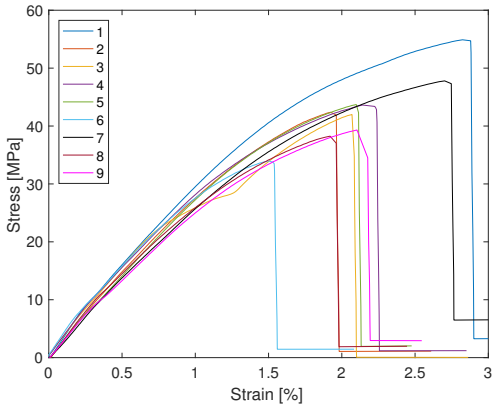

Figure 4.1: Comparison of average value of each sample from the low pressure compression moulding series

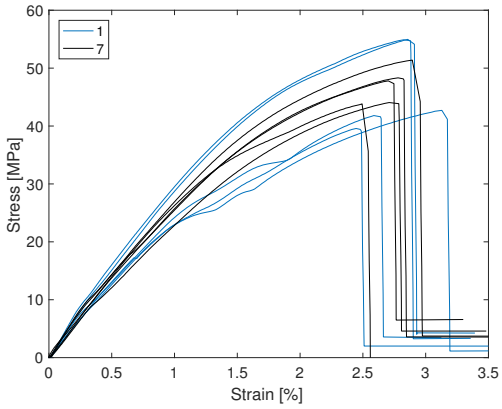

Figure 4.2: Comparison of all specimens of sample 1 and sample 7 from the low pressure compression moulding series

4.3 High pressure stage

During the high pressure stage, a high pressure was applied to shape the material and make the molten components flow. Based on the literature, three levels for the moulding parameters pressure, time and temperature were investigated. Temperatures of 170, 190 and 210 °C, pressures of 1, 3 and 7 MPa and times of 15, 30 and 60 sec were investigated. When varying one parameter, the others were held constant, leading to a total of seven samples.

To analyze the high pressure stage, five test specimens from each sample were cut using a laser cutter and the tensile properties were investigated. Errorplots were used to see significant differences or trends in the mean values and standard deviations.

The results showed that a higher pressure gave higher values for tensile modulus, UTS and strain at UTS. Significant difference was shown between 1 and 7 MPa but not between 3 and 7 MPa, therefore 3 MPa was defined as the optimal pressure. There were no significant differences regarding UTS or tensile modulus for the varying times. Based on this, the shortest time was chosen as optimal to reduce the moulding process time. Regarding the temperature, WF1 and WF2 gave different results. These results are shown as the three middle values in Figure 5.1. No significant difference was shown for WF2 but there was a maximum in mean UTS at 190 °C, whereby 190 °C was chosen as the optimal temperature to use for WF2. As for the samples containing WF1, a higher temperature gave a higher tensile modulus as well as a lower strain at UTS. Based on the significant increase in tensile modulus, the highest temperature of 210 °C was chosen as the optimal temperature for WF1.

4.4 Cooling

The aim of the cooling stage was to decrease the temperature to below 100 °C to solidify the polymeric binder. This step was done under pressure to maintain the shape of the sample. Water cooling of the mould plates was used and the time it took for the plates to decrease below 100 °C was measured to approximately 5 min. A trial was performed to see if the cooling stage had any effect on the mechanical properties but no significant difference was shown. However, one of the main purposes of this stage was to ensure dimensional stability of the moulded part. Based on this, a cooling time of 5 minutes was used in all further trials.

5
Results

5.1 Standardized characterization methods used

From the comparison of different standardized characterization methods, it was decided to use ASTM D1037:12 Standard Test Methods for Evaluating Properties of Wood-Base Fiber and Particle Panel Materials with modifications where needed. The test specimens for all methods were modified with respect to size and geometry. The used sizes and geometries can be seen in Table 3.2. The conditioning environment was also modified to a slightly lower relative humidity. Furthermore, the calculations for tensile testing parallel to the surface was performed according to Equation 3.1 - 3.3. The dry weight used for the water absorption calculations was the weight after conditioning and not the oven dry weight.

Considering that the end products are structural applications, the final testing protocol was based on the following standardized methods.

D1037:12 - 9	Static Bending
D1037:12 - 10	Tension Parallel to Surface
D1037:12 - 11	Tension Perpendicular to Surface (Internal Bond)
D1037:12 - 23	Water Absorption and Thickness Swelling

5.2 Mechanical properties of produced samples

This section covers results obtained during the project. Process development of compression moulding with focus on the temperature parameter as well as the final moulding protocol are presented. The results from mechanical characterization of the material are included.

5.2.1 The influence of processing temperature on mechanical properties

The mean values and standard deviations for compression moulded samples at varying temperatures ranging from 150 °C to 230 °C can be seen in Figure 5.1. The samples contained 20 % polymeric binder. In general, the tensile modulus and strength increased significantly with increased temperature while the strain at UTS

decreased. Both WF1 and WF2 samples showed lower mechanical properties at the lowest temperature. The modulus leveled out above 190 °C for WF2 and 210 °C for WF1, but the stress and strain behaved differently depending on fiber type. Between 210 °C and 230 °C, stress and strain decreased for WF2 and increased for WF1. However, due to the fact that the samples got a dark burned color at the highest temperature, it was not considered favourable for any fiber type.

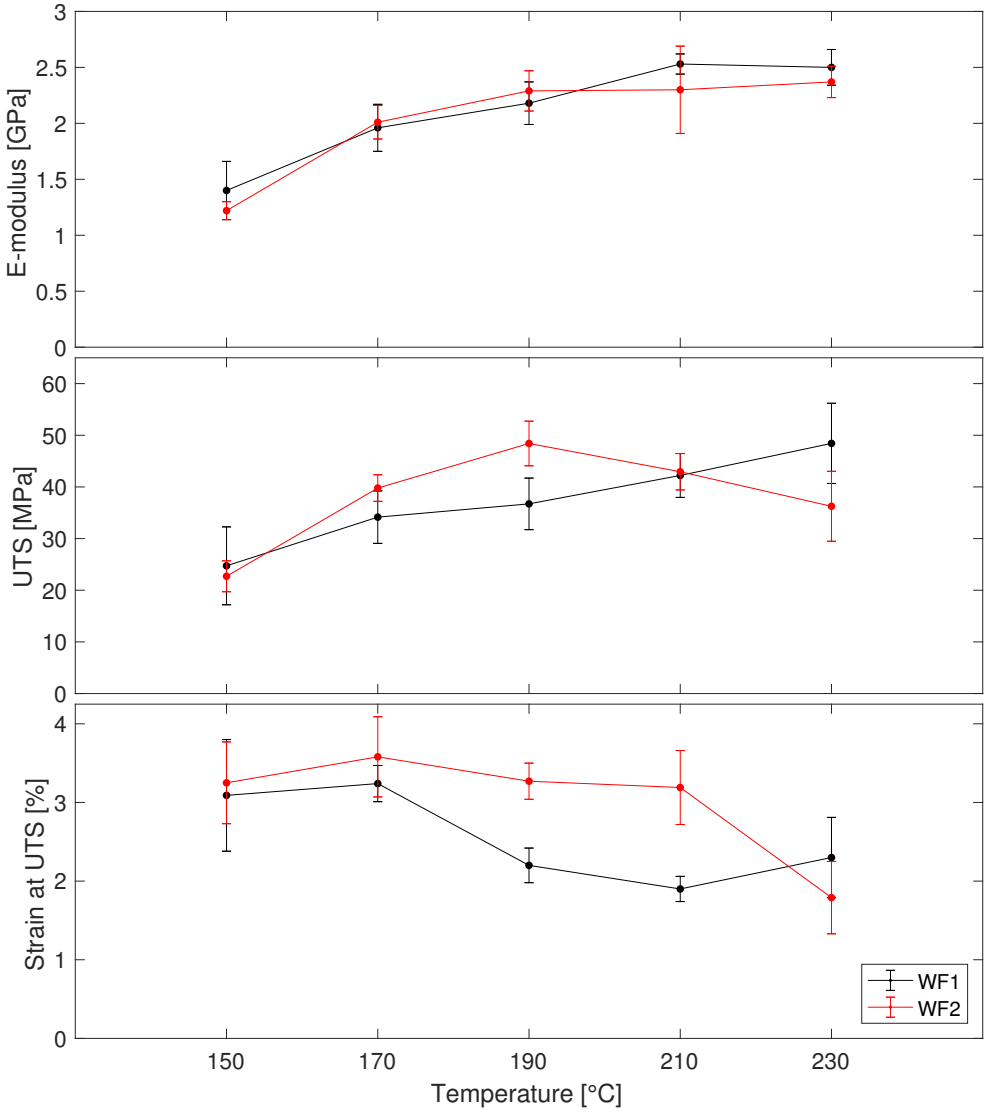

Figure 5.1: Comparison of mean values and standard deviations for different processing temperatures

To compare the mechanical properties for all WF1 samples after compression moulding at a lower temperature of 190 °C and at the optimal temperature 210 °C respectively, the mean values and standard deviations were plotted, as can be seen in Figure 5.2. The specification of the samples can be seen in Table 3.1. The results showed that a higher temperature increased tensile modulus and strength. The difference was significant for some of the samples that had lower amounts of polymeric binder. Regarding the strain, no clear trends could be seen. All together, these

results indicated that a higher processing temperature generally gave higher tensile properties but not always with significant difference. A lower temperature would be favourable if the color of the material was to be a very important aspect, since the higher temperature gave a slightly darker result. The highest binder content was not favourable when it came to geometrical stability of the samples. The polymeric binder made the samples flow and deform.

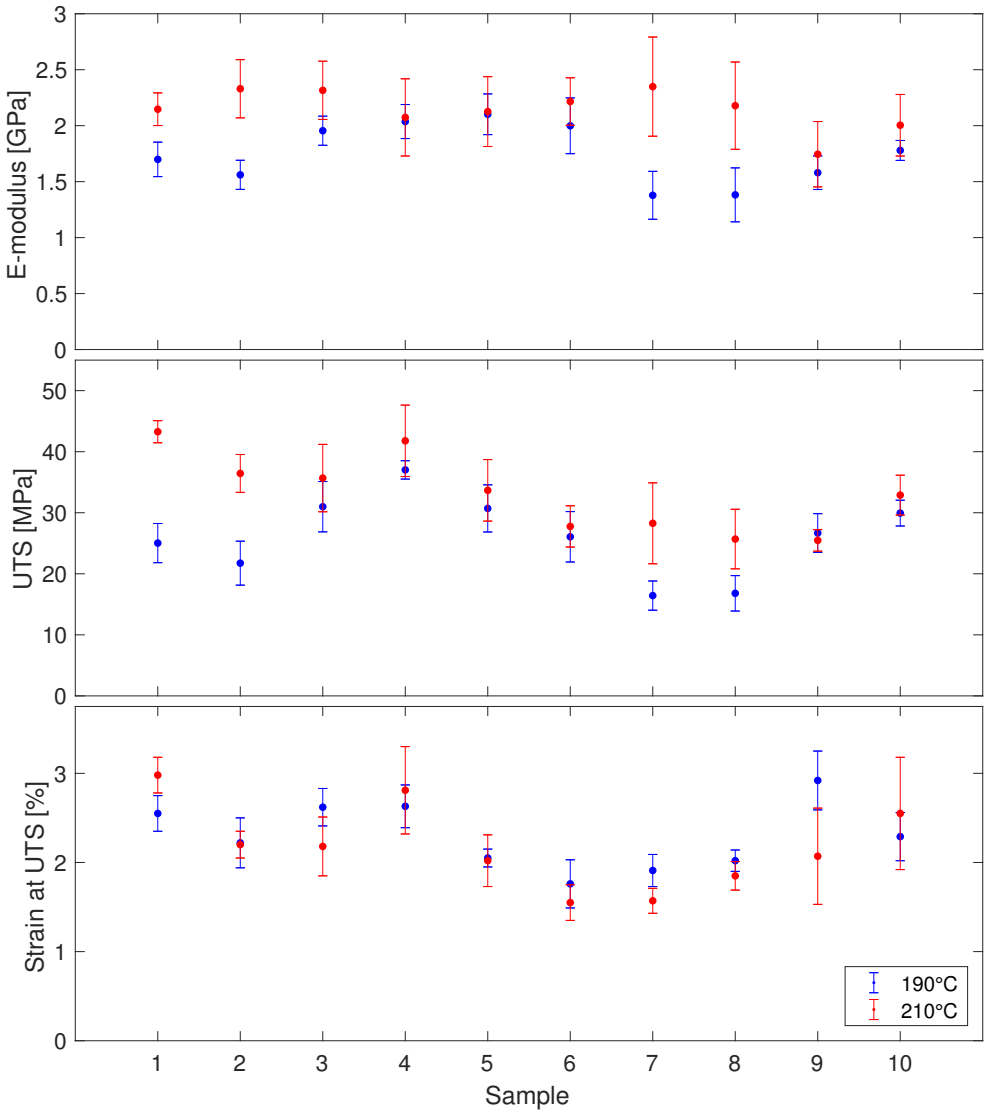

Figure 5.2: Comparison of mean values and standard deviations at two different temperatures

All previous results regarding compression moulding were summarized into a final moulding protocol shown in Table 5.1. From the pre-heating study, it was shown that an internal temperature of 150 °C in the material was not crucial for the studied properties when using the moulding process developed in this project. Therefore, a lower temperature and shorter time can be used to decrease the lead time of the process and the energy consumption. Two different temperatures were found to be optimal for WF1 and WF2 respectively. The time of the high pressure stage did not affect the results, whereas a shorter time was preferable to shorten the lead time. There were no significant differences in mechanical properties when increasing the pressure of the high pressure stage and thus, a lower pressure was preferable to save energy and time. Cooling did not affect the results significantly but is recommended to ensure dimensional stability of the moulded part.

Table 5.1: Compression moulding protocol

Stage	Temperature [°C]	Time [min]	Pressure [MPa]
Pre-heating	150	14	-
Low pressure	190/210	1	1
High pressure	190/210	0.25	3
Cooling	<100	5	3

5.2.2 Tensile properties

The three properties analyzed were the tensile modulus, UTS and strain at UTS. Mean values and standard deviations of the three properties can be seen in Figure 5.3. None of the samples showed significant better results compared to all of the other samples. The effect of varying binder contents, densities before pressing and densities after pressing was evaluated. It was not possible to draw any conclusions regarding the results when it comes to the various compositions and densities of the 12 different samples.

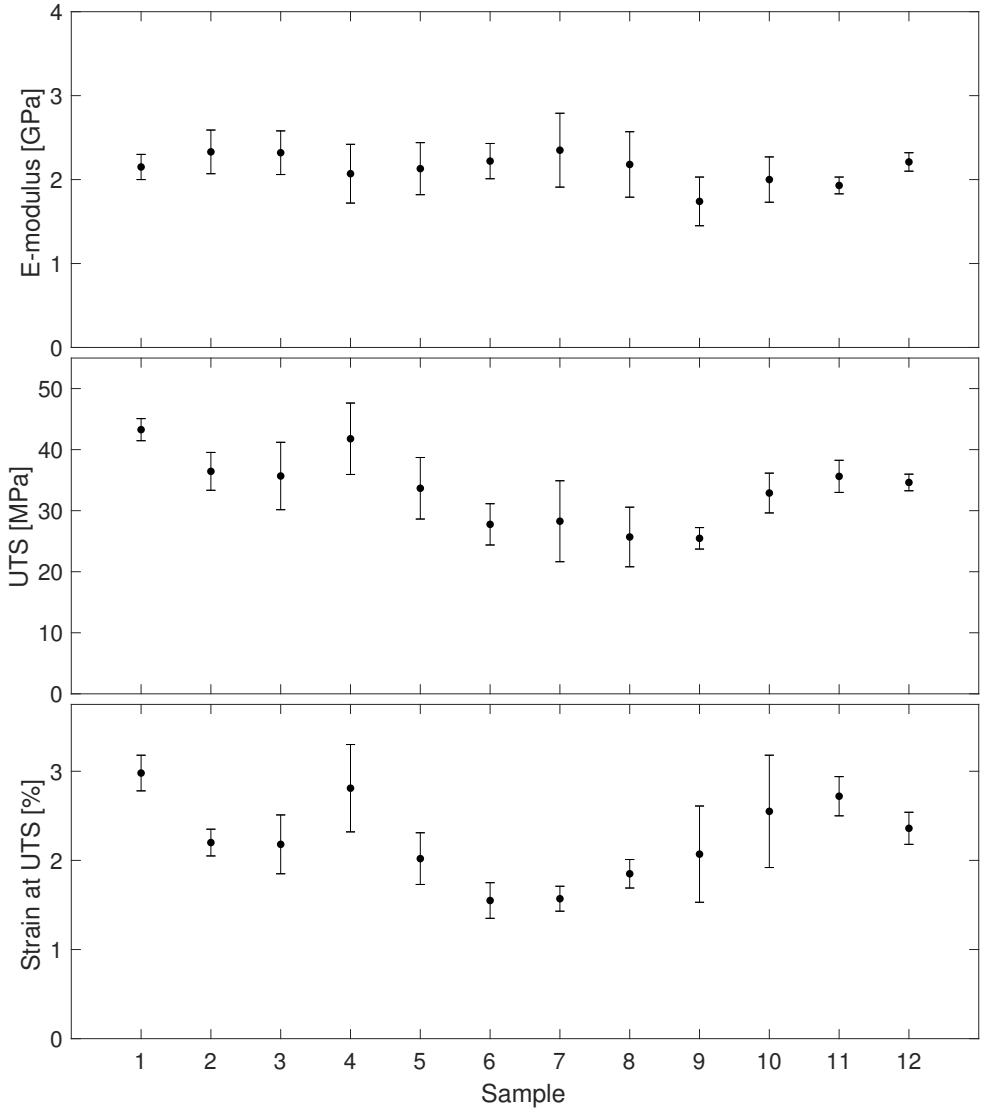

Figure 5.3: Comparison of mean values and standard deviations for tensile properties

5.2.3 Bending properties

The bending modulus was significantly higher than the tensile modulus for all 12 samples, which can be seen in Figure 5.4. This result indicated that a stiffer outer layer was formed during the compression moulding. Although the temperature may be rather homogeneous throughout the material, the outer layers will always have the target temperature for a longer period of time leading to the formation of a stiffer layer.

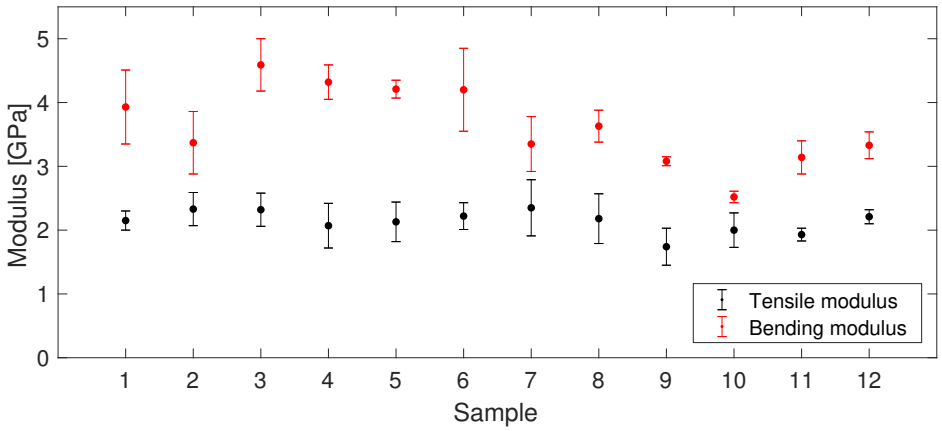

Figure 5.4: Comparison of mean values and standard deviations for tensile and bending modulus

5.2.4 Internal bond properties

The hardest part of the internal bond testing was to find a suitable adhesive and proper testing equipment.

Several types of glue and tape were tested but only one of them was promising. It was the glue Cascol Polyurethane from Casco. The test could not be performed with the machine and load cell available without error due to excessive loads. In order to induce a fracture, the test specimen was reduced by turning to a smaller diameter of 11.55 mm. In the fourth attempt, fracture occurred, partially in the middle layer of the sample but also in the adhesive bond. The UTS obtained was 4.17 MPa. All the other specimens failed due to failure of the adhesive bond, an example of this can be seen in Figure 5.5. All the results from the tests can be seen in Table 5.2. Since no test specimens got a valid fracture this might not be a suitable method for testing internal bond strength. It also indicated that there were no weaker layers in the material up to about 4 MPa and that the material was relatively homogeneous in the cross-section.

Table 5.2: Results from testing internal bond strength

Adhesive	Sample	UTS [MPa]
3M Scotch Pressure Sensitive Tape	11	1.23
3M Scotch Skirting Board Tape	11	1.02
High Tech AB Acrylic	12	0.83
SCANTECH Cyanofirm	12	1.34
Casco Cascol Polyurethane	12	4.17

During testing it was proved to be beneficial to roughen the surface of the specimen to get better adhesion.

Figure 5.5: Failure of the adhesive bond

5.3 Water absorption

The water absorption tests had two measurements. One after 2 h water submersion that simulates a product used indoors and one after 2 + 22 h water submersion that simulates a product used outdoors. The result clearly showed that higher binder content is advantageous if low water absorption is desirable. The test also showed that the material continued to absorb after 2 hours and did not get saturated. It is shown in Figure 5.6 how the absorption is affected by the binder content for both time intervals and fiber types.

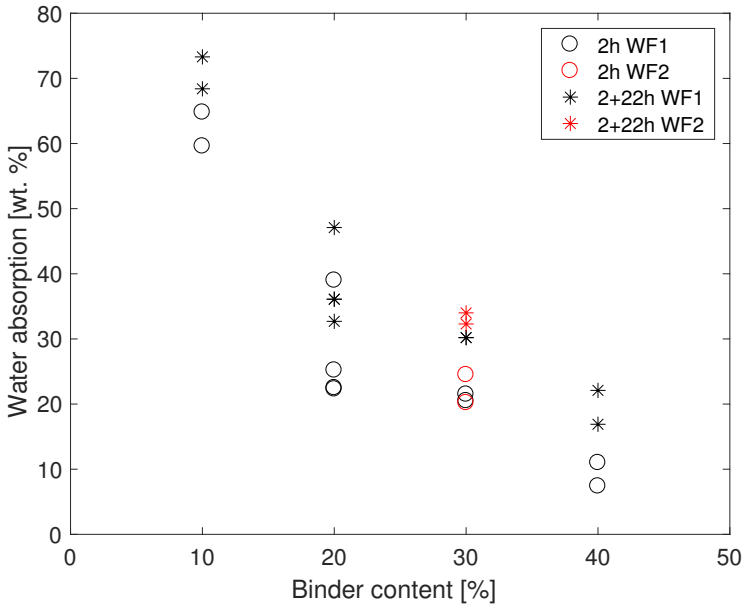

Figure 5.6: Binder content dependency of water absorption

6
Discussion

Classification of the novel composite material of this project was done by comparing standardized characterization methods for different established classes of materials such as paper, polymers, composites and fiberboards. Composites and fiberboards were the two classes that were considered closest related to our material. However, the definition of composites in the standards was often based on a polymer matrix as main constituent with varying types of reinforcement. Since the main constituent of our material is wood fibers and the synthetic material is only used as binder, the standards for composite materials were found inappropriate. Another considered aspect was the types of materials that are currently used for the possible applications of the project. With possible applications such as furniture, car panels and heavy packaging, standards for wood based fiberboard were found most suitable. However, there were some issues regarding the specimen sizes given by the standards. Since wood based fiberboards are typically produced in large quantities and used as large panels, the standardized specimen sizes were too large and not possible to apply. Because of this, modified specimen sizes were used for all mechanical characterization.

When developing the moulding process, the focus was set to obtain the highest possible tensile strength and stiffness of the material due to the possible end products. The decrease in strain that follows an increase in strength and stiffness have not been taken into account. Furthermore, the final color of the material have not been a crucial aspect, although it has been noted. Depending on the requirements of the end product, the final moulding protocol could be altered. If a bright color is required, the moulding temperature could be decreased at the expense of the studied strength and stiffness.

The moulding parameters were found to affect the mechanical properties differently for the different stages of the moulding process. The internal temperature of the material from the pre-heating stage was found to be less important than thought. Lowering the temperature to around 125 °C instead of 150 °C did not affect the mechanical properties significantly. However, there might be a critical limit where the pre-heating temperature starts to affect the final result negatively due to uneven distribution of heat. It should also be stated that the optimal moulding parameters obtained is only valid for the equipment, geometries and material used in this project. When applied to other materials or when using different equipment, verification of the methods and results should be made.

Given that the aim for our material is to be used for structural applications, a testing protocol has been developed in accordance to this. Tensile testing was done to obtain the stiffness, strength and strain at UTS of the material. Furthermore, bending tests were in our case done to indicate whether or not there were any differences in stiffness between the outer layers of the material compared to the bulk. Loading in bending is also interesting to evaluate because it is important for product performance. The reason for testing internal bond strength was to evaluate if a homogeneous result was obtained from compression moulding. If the material would not have reached target temperature throughout the material during moulding, a weaker layer could have occurred in the middle of the material. Testing of internal bond strength would reveal this weaker layer as the fracture would take place there. The fact that we did not succeed to test internal bond strength with the available adhesives indicated that no layer weaker than about 4 MPa was formed and that the compression moulding process resulted in a rather homogeneous material. Evaluation if the material was penetrated by the adhesives has not been performed.

Lastly, water absorption was included in the testing protocol. As wood is a highly hydrophilic material, water absorption should always be considered when developing products made of it. The results clearly show that a higher amount of polymeric binder decreases the water absorption which was expected due to the hydrophobic nature of the polymer fibers and the results found in the literature. This should be kept in mind when designing with this type of material, so that a composition with higher amount of polymer is chosen for products that should absorb less water. The wood fiber and polymer fiber ratio was not expected to have great impact on mechanical properties according to literature, which was also shown in our results where no trends or significant differences were found between the 12 samples of different composition.

When comparing the results from mechanical characterization in the present study with the results found in literature, this is preferably done by looking at similar densities. The 90 % wood and 10 % PP composite with a density of 1.0 g/cm^3 and HDF with a density higher than 0.8 g/cm^3 were the materials from the literature that were closest to the material produced in this project. The tensile properties differed from the W/PP composite in the literature, with a slightly lower elastic modulus but significantly higher UTS. The bending modulus was higher than for the W/PP composite and similar to the HDF. This indicates that the W/PP composite did not develop the stiffer outer layer during the manufacturing process that were observed in this study. When comparing the results for water absorption, the present results show a slightly higher absorption after 24 h than for the W/PP composite in literature. However, the densities obtained were somewhat lower than in the literature, ranging around 0.9 g/cm^3 for the samples with 10 % polymeric binder. According to previous results, this may affect the absorption drastically.

6.1 Future work

For further development of this project, it would be beneficial to use equipment with better control of parameters. If time and temperature are correct and stable during a whole test sequence, this would lead to a result with less deviations, which could give more distinct results.

Pre-heating has proved not to be as sensitive as earlier believed. Therefore, it would be good to do a new study that clarifies what time and temperature is really needed. The result of pre-heating is depending on the following compression moulding process and this has to be taken into account.

In order to manufacture products that are of interest to the industry, further development should be focused towards producing 3D geometries instead of flat sheets as in this project. It is also important to investigate how strain in the material affects the formability and evalute how the density of the raw material affects this.

When it comes to internal bonding, it would be good to focus further investigations into finding a good adhesive. This is required to be able to carry out this type of mechanical test and according to the standard, a suitable adhesive should be used. Epoxy has not been tested in this project due to safety hazards, but could be an alternative for further investigations. During curing of the adhesive it is important to compress the specimen, preferably in the same machine as the tensile test. Furthermore, it is also necessary to use bi-axial grips to ensure that no torque is induced during the tensile test and that the specimen is completely aligned.

Since all of the bico fibers are melted during compression moulding, the polymer fibers may be of homogeneous polymer instead. For large scale production, the binding process would be interesting to further evaluate. This could be done based on the available techniques described in the background.

6.2 Sources of error

During compression moulding, the largest sources of error were linked to the equipment. The drying chamber was unstable which gave some differences during preheating. Heat differences in upper and lower mould plates during compression moulding, both while heating and cooling, also likely affected the result in a negative way. Some of the sample preparation was done by using a laser cutter. This is not a standardized method and is usually not used. This means that it is unclear how it affects specimens. The size of the specimens was also changed and this could affect the results, especially since this material contains fibers which sometimes was longer than the shortest dimension in the test specimen. The fibers were non-uniformly distributed in the material which means that each fiber could result in a defect that could lead to a break. This has larger influence when using smaller specimens.

7
Conclusions

This thesis have concluded that the most suitable standardized characterization methods for the novel composite material investigated are the ones developed for wood based fiberboards. The compression moulding process consist of four stages: pre-heating, low pressure, high pressure and cooling. The optimal parameters for the process developed in this project differ between the four stages. A higher processing temperature is favourable to increase the studied mechanical properties but increases the risk for discoloration of the material. Lower temperatures can be chosen to ensure bright color at the expense of mechanical strength and stiffness. Furthermore, it has been shown that the polymer content does not affect mechanical properties significantly. However, it decreases water absorption drastically. If water absorption is set aside, a polymer content of 10 % is enough to ensure easier handling of the raw material. Higher amounts of binder content also makes the material deform during compression moulding. Altogether, it can be concluded that the material is not sensitive for small changes in process parameters or composition.

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