Extrusion Processing of Wood Raw Materials for Use in Wood-Polymer Composites

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April 2011
Abstract

The interest in wood-polymer composites and their use in different applications has been growing over the last 10-15 years. Environmental issues and demands on lower material costs are the driving forces behind the increasing use of renewable materials such as wood and other natural fibres as reinforcement in polymer composites.

Wood flour consisting of small wood particles is currently used as the main wood raw material in commercial wood-polymer composites. However, the reinforcing potential of wood flour is limited. A better reinforcement could be achieved by using wood fibres with a higher aspect ratio as raw material, but individual fibres are seldom used in composite manufacturing due to processing problems and high cost. Therefore, the objective of the work was to study the possibility to use wood chips as raw material and separate individual fibres with higher aspect ratios from the wood chips during the composite manufacturing process.

First, the effect of the extrusion process only on wood raw material was studied without a matrix polymer, and then composites using polypropylene as matrix were made. The main goal was to produce wood particles/fibres with high aspect ratio during the manufacturing of wood polymer composites. The effects of extrusion parameters, different screw configurations, raw materials and raw material pre-treatments were evaluated. The size of the separated wood particles and fibres was measured using optical fibre analysis methods. Microstructure of wood particles as well as the fractured surfaces of prepared composites were examined using electron microscopy. The mechanical properties of the composites were measured using flexural and impact testing.

The results showed that wood chips can be used as raw material in a one-step manufacturing process of wood-polymer composites. Also, individual fibres with a higher aspect ratio than wood flour were separated from the wood chips with suitable processing conditions.
List of papers

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Paper II

Paper III

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Hietala, M., Samuelsson, E., Niinimäki, J. & Oksman, K. The effect of pre-softened wood chips on wood fibre aspect ratio and mechanical properties of wood-polymer composites. To be submitted.
1 Introduction

1.1 Wood-polymer composites

Wood-polymer composites (WPCs) are materials which combine the properties of wood and matrix polymer. In general, wood-polymer composite materials can contain varying contents of wood, plastics and additives and they are processed by the same techniques as thermoplastic polymers.

The use of wood-polymer composites in different applications has been steadily growing since the 1990s. So far, the major markets for wood-polymer composites have been North America and Asia, although the use of natural fibre and wood reinforced polymer composites is also on the increase in Europe. Currently, the main use of WPC is in building and construction to replace impregnated wood in outdoor applications such as decking, railings, and window and door frames. Other uses of WPC are, e.g. automotive interior panels and interior materials such as flooring and furniture. Even electric guitars are manufactured from wood-polymer composite [1]. Examples of some WPC products are shown in Fig. 1. [2]

![Fig. 1. Examples of wood-polymer composite products.](image)

The use of natural cellulose materials such as wood fibres and wood flour as reinforcement/filler in thermoplastics has many advantages over the inorganic and synthetic fillers traditionally used in polymer composites. Natural fibres are light and inexpensive, they are abundant and they have high
specific stiffness and strength. The environmental aspects are also important; natural fibres are renewable, recyclable and biodegradable materials. [2]

However, there are also problems related to the use of natural fibres in thermoplastic composites. Major drawbacks are the poor interfacial adhesion between the hydrophobic matrix and the hydrophilic fibres and the difficult dispersion of fibres in the matrix. Due to the hydrophilic nature of fibres, they also need drying before compounding. The processing temperatures are limited due to the thermal degradation of the wood fibres in temperatures above 200°C, hence limiting the range of suitable matrix polymers. [2]

1.1.1 Raw materials

The polymer matrix forms a continuous phase which surrounds the wood component in WPC. Due to the low thermal stability of wood, only polymers with processing temperatures lower than 200°C are typically used in WPC. The polymers used are mostly low-cost commodity thermoplastics which flow easily when melted, and the most common polymers used are polyethylene (PP), polypropylene (PP) and polyvinylchloride (PVC). [3]

The wood raw material used in wood-polymer composites is either wood flour or wood fibres. The term fibre refers to a single spindle-shaped wood cell, even though it is sometimes used inaccurately to describe wood particles. Individual fibres can be separated from wood using various chemical or mechanical methods familiar from papermaking processes. Fibres have typically high strength and quite high aspect ratios (length-to-diameter ratio, l/d); therefore, their reinforcing potential is also good. However, due to the higher cost and difficulties in processing, especially in feeding and metering, wood fibres are not used as much as wood flour in commercial manufacture of wood-polymer composites. [3]

Wood flour is the most commonly used material in manufacturing of wood-polymer composites; it is less expensive and feeding and metering wood flour is easier compared to wood fibres. Wood flour consists of small wood particles composed of fibre bundles and the aspect ratio of the particles is low, typically between 1 and 5. Therefore, the reinforcing capability of wood flour is not as good as fibres. Wood flour is provided in various particle size ranges; however, typically the particle size of wood flour is within 180-840 μm. Wood flour can be obtained directly from the forest product
companies or from companies specialized in producing wood flour [4]. Wood flour is mainly produced from the residues of wood processors, and the main processing steps are usually: 1) size reduction with mills and 2) size classification by screening. Wood flour typically contains at least 4% moisture when delivered, and it is normally dried before compounding. [3]

In addition to wood and matrix polymer, wood-polymer composites contain small amounts of additives. These additives affect the processing and performance of the material. Most common additives are coupling agents, light stabilizers, pigments, lubricants, fungicides and foaming agents. Coupling agents (or compatibilizers) are especially important in wood-polymer composites. They are used to improve the interfacial adhesion between the hydrophobic matrix and the hydrophilic wood. Maleic anhydride grafted polypropylene, MAPP, is one of the most commonly used coupling agents in WPC. MAPP is believed to enhance the interfacial adhesion via two mechanisms: 1) anhydride forms an ester bond with a wood cell wall hydroxyl group, and 2) PP segment attached to the anhydride entangles with the PP or PE network in the melt. [4]

1.1.2 Processing

Processing of wood-polymer composites typically consists of three steps: 1) wood raw material processing, 2) compounding and 3) moulding of the products by injection moulding, profile extrusion or compression moulding. [2]

The purpose of the wood processing step is to increase the value and quality of the wood raw material by separating it into different sizes and species and drying it before the compounding step. There are also wood fibre companies which provide pre-processed wood fibres and fillers for many applications. In the compounding step the actual composite is manufactured by blending the reinforcement or filler with the matrix polymer. The compounding is typically done in a twin-screw extruder. The compounded material can be immediately pressed or shaped into an end product or formed into pellets for further processing. The products can be manufactured, for example, using sheet or profile extrusion, injection moulding, calendering, thermoforming or compression moulding. [2, 4]

Direct extrusion is the most common technique in the manufacture of WPC. With this technique the raw materials are melt-compounded and extruded into a continuous profile by forcing the molten
material through the die in the same process step [4]. In direct extrusion either profiles or sheet materials for compression moulding can be manufactured. A schematic picture of a direct extrusion line for profile manufacture is shown in Fig. 2. [2]

With the pelletizing technique raw materials are melt-compounded prior to moulding into a product. One of the simplest methods of producing pellets is the strand pelletizing method, whereby the composite melt is pressed through a die with one or several holes, and the resulting strands are cooled in a water bath. After cooling the strands are fed into an air blowing unit to remove excessive water, and then the strands are fed into a pelletizer unit in which the strands are cut into small granules. The pellets can be used in injection moulding or profile extrusion. [5]

1.1.3 Properties

Because of the wide variety of wood-polymer composites, the properties and performance of the composites vary considerably. The individual properties of wood filler and matrix polymer determine the properties of a composite. The stiffness is especially influenced by the form, size, dispersion and content of the wood filler in the composite. The interfacial adhesion between the filler and matrix determine the strength, moisture uptake and long-term properties of wood-polymer composites. [2]
In general, it can be said that addition of a wood component enhances the stiffness of the polymer matrix. However, at the same time, the materials become more brittle. When compared to solid wood, WPCs are typically less stiff. When fibres are used instead of wood flour as reinforcing filler, the mechanical strength, elongation and the unnotched impact strength properties of the composites usually increase. [6]

The fungal resistance and dimensional stability of wood-polymer composites is better than solid wood because they absorb moisture more slowly due to the thermoplastic matrix. However, the mechanical properties such as creep resistance, stiffness and strength are lower than those of solid wood. Therefore, wood-polymer composites are currently used only as non-structural members in building applications. However, a lot of research is being done to improve the mechanical performance of wood-polymer composites. In addition, much of the research is concentrated on studying the durability and service life of wood-polymer composites. [6]

1.2 Wood properties

Wood consists of cellulose, hemicelluloses, lignin and extractives. The structure and anatomy of wood makes it a stiff, strong, tough and lightweight material which can effectively perform tasks critical for tree survival, such as moisture transport. In comparison with commodity synthetic polymers, wood is a cheaper, stiffer and stronger material, which makes it a good candidate for polymer filler or reinforcement. [3]

1.2.1 Macrostructure

The general structure of a tree stem is shown in Fig. 3. As Fig. 3 shows, the stem can be divided into three planes: 1) transverse (or cross-section) surface, 2) radial surface and 3) tangential surface. Wood appearance and properties vary depending on which plane is studied. [7]
Fig. 3. The macroscopic structure of a tree stem showing the main planes: tangential, radial and transverse surface [7].

From Fig.3 it can be seen that in the centre of the stem is the pith, which is a small core of soft tissue formed during the first growth year. The pith is surrounded by heartwood. Heartwood consists of dead wood cells and it functions as a mechanical support for the tree. Heartwood is usually darker than sapwood due to the fact that resinous organic compounds deposit in its cell walls and cavities. Sapwood is the physiologically active part of a tree and, besides being a structural support, it acts as a food reservoir and it enables water conduction up from the roots. Sapwood is physiologically active and it communicates with the cambium and phloem through sap flow. The cambium is a thin layer of tissue between the bark and the sapwood. Cell divisions in the vascular cambium result in wood thickness growth. The inner bark, or the phloem, is a narrow layer of tissue through which the sap moves up and down. Inner bark is usually light-coloured. The outer bark (rhytidome) consists of dead wood cells and it is dark-coloured. [7, 8]

In the boreal and temperate climates the cambium has a yearly growth cycle, which is the reason for the annual rings of a tree. The wood volume growth is usually most rapid in the beginning of the growth period and it slows down and finally stops at the end of the growing season. This causes a band of light and darker wood every year. The wood formed early in the growing season is called earlywood and the wood formed late in the season is called latewood. [7]

1.2.2 Wood cells

Depending on their function in a tree, wood cells can be divided into three categories: 1) conducting cells, 2) supporting cells and 3) storage cells. The conducting and supporting cells are called
prosenchymatous (prosenchyma) and storage cells are called parenchymatous (parenchyma). Procenchyma cells are thin, long cells which are narrower at the ends, and parenchyma cells are short cells which are either rectangular or round [9]. In all wood cells there are also small recesses, pits, which interrupt the secondary wall. Through these pits fluids and gases are able to pass from cell to cell. [7]

Hardwoods and softwoods differ somewhat in terms of cell structure. In general, hardwoods have a more complex structure and a larger number of different kinds of cells than softwoods [7]. The vertical structure of softwoods consists mainly of long and tapering cells called tracheids. Tracheids are typically considered as fibres in softwood species. The average length of Scandinavian softwood tracheids is 2-4 mm and the average width in the tangential direction is 0.02-0.04 mm [9]. Some species also have vertical resin canals. Horizontally, wood consists of narrow rays. These rays are usually one cell in width but several cells in height. Ray cells can be either ray parenchyma or ray tracheid cells. All tree species have ray parenchyma cells but only some have ray tracheid cells. Fig. 4 shows different types of softwood tracheid and parenchyma cells. [8]

The main vertical structure of hardwoods consists of long, narrow cells called libriform fibres and shorter and wider cells called vessel elements. Water is distributed through the vessels. Hardwoods also have a vertical parenchyma system and a horizontal or ray parenchyma system. In addition, hardwoods contain hybrids of the already mentioned cell types, which are called fibre tracheids. Different types of hardwood cells are presented in Fig. 5. In hardwoods the term fibre is mostly used to describe libriform cells and fibre tracheids. The average length of birch libriform fibres is 1.1-1.2 mm and the average width is 14-40 μm. [8, 9]
Fig. 4. Different types of softwood cells [10].

Fig. 5. Different types of hardwood cells: earlywood vessel elements of birch (a), aspen (b) and oak (c), latewood vessel elements of birch (a₁) and oak (c₁), longitudinal parenchyma cell of oak (d), ray parenchyma cell of aspen (e) and of birch (f), tracheids of oak (g) and birch (h) and a libriform fiber of birch (i). [10].
1.2.3 Cell wall structure

The structure of a wood fibre is very complex, and it can be considered as a composite structure itself, because it is composed of several layers. The typical cell wall structure of a softwood tracheid is shown in Fig. 6. The cell wall consists of middle lamella (ML), primary wall (P), and secondary wall (S) with three layers and lumen (L) (Fig. 6). These layers have different structures and chemical compositions, but the main difference is in their microfibril alignments. Microfibrils are cellulose molecules bundled together \[8, 10\].

The middle lamella (ML) does not actually belong to the cell wall because it separates two adjacent tracheids. Middle lamella has very high lignin content and it is amorphous. \[8, 10\]

Fig. 6. Cell wall structure of softwood tracheid [11].

Primary wall (P) is the outermost layer of the cell wall of wood fibre. It is a thin layer consisting mainly of lignin and pectins. Secondary wall (S) has three layers: S\(_1\), S\(_2\) and S\(_3\). The layers differ from each other on different microfibril alignments and compositions. S\(_1\) is a thin and lignin-rich outer layer, S\(_2\) is a thick middle layer and S\(_3\) is a thin inner layer. The S\(_2\) layer of softwood tracheid is technically the most valuable layer because of its high cellulose content. The inner layer of secondary wall, S\(_3\), is also called as tertiary wall (T), and it consists mainly of hemicelluloses. In the middle of softwood tracheid there is a canal called lumen (L). In all softwoods and in some hardwoods there is also a thin amorphous membrane in the inner surface of the cell wall called the warty layer \[9\]. \[7, 10\]
1.2.4 Chemical composition

The main components of wood are cellulose, hemicelluloses, lignin and extractives. The amounts of each of these components in softwoods and hardwoods are presented in Table 1.

<table>
<thead>
<tr>
<th>Component</th>
<th>Softwood</th>
<th>Hardwood</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cellulose</td>
<td>42 ± 2%</td>
<td>45 ± 2 %</td>
</tr>
<tr>
<td>Hemicelluloses</td>
<td>27 ± 2 %</td>
<td>30 ± 5 %</td>
</tr>
<tr>
<td>Lignin</td>
<td>28 ± 2 %</td>
<td>20 ± 4 %</td>
</tr>
<tr>
<td>Extractives</td>
<td>3 ± 2 %</td>
<td>5 ± 3 %</td>
</tr>
</tbody>
</table>

Cellulose is the most important substance in wood because it determines the character of the fibre. The chemical formula of cellulose is \((\text{C}_6\text{H}_{10}\text{O}_5)_n\), and \(n\) is the number of repeating sugar units or the degree of polymerization, DP. The repeating unit in cellulose consists of two glucose molecules. This unit is also called *cellobiose*. The DP varies depending on the cellulose source and the treatments it has received. The DP of most papermaking fibres is between 600 and 1500. The properties of cellulose-containing materials depend strongly on the DP of cellulose molecules. Cellulose can have organized, crystalline regions or unorganized, amorphous regions, and the crystalline cellulose is stronger. [8]

Hemicelluloses are branched polymers consisting of five different sugars: glucose, mannose, galactose, xylose and arabinose. Hemicelluloses degrade and dissolve more easily than cellulose, and their amount in pulp is always less than in the original wood. Besides cellulose and hemicelluloses, wood contains lignin and extractives. Lignin is an amorphous and highly polymerized substance whose primary purpose is to form the middle lamella and glue the fibres together. The structure of lignin is very complex. In addition, different kinds of substances, e.g. resin acids, fatty acids, turpenoid compounds and alcohols, are also present in wood fibres. These substances are typically called extractives. [8]

1.3 Mechanical separation of fibres

To produce uncut fibres mechanically from wood it is very important that the polymeric wood material is softened before mechanical treatment. Because wood is a polymer composite with
viscoelastic behaviour its response to mechanical treatment depends on the temperature, moisture and time under load. Thus, the glass transition temperatures \((T_g)\) of wood polymers (cellulose, hemicelluloses and lignin) are very important from a fibre separation point of view. [12]

Lignin polymer has a major influence on the wood behaviour in mechanical fibre separation processes. As Fig. 7 shows, moisture content has an effect on the softening temperatures of wood polymers. In dry conditions the softening temperature \((T_g)\) of lignin and hemicelluloses is between 180 and 220°C. However, in water-saturated conditions the softening temperature \((T_g)\) of hemicelluloses and amorphous cellulose is close to 20°C. For water-saturated isolated lignin the softening temperature is between 80°C and 90°C. The softening temperature of lignin as a part of wood matrix is typically from 100°C to 130°C. Therefore, lignin softening is the critical factor in mechanical separation of wood fibres. If lignin remains stiff, the fracture between the fibres occurs in an uncontrollable way, and more broken fibres are formed. [12]

![Fig. 7. Influence of moisture content on the softening temperature of wood][13].

For practicality, the wood for mechanical fibre separation processes should have a moisture-wood ratio of 0.3-0.4 or even higher to ensure sufficient moisture content, and the temperature should be above the lignin glass transition temperature. Due to the viscoelastic nature of wood, increase of the frequency of the mechanical action also increases the softening temperature of wood polymers. [12]
1.4 Objective of this study

The objective of this work was to study the possibility to use wood chips as raw material and separate individual fibres with higher aspect ratios from the wood chips during the composite manufacturing process. In addition, the effect of wood chips as raw material on the mechanical properties of wood-polypropylene composites was studied. The aspect ratios of the extruded wood particles and fibres were measured using optical fibre analysis methods. Microstructure of wood particles as well as the fractured surfaces of prepared composites were examined using electron microscopy. The mechanical properties of the composites were measured using flexural and impact testing.
2 Experimental procedure

2.1 Materials

Spruce pin chips and pre-crushed spruce were used as wood raw materials in the experiments. Polypropylene in powder form (MFI = 32 g/10 min, 230°C/2.16 kg and 16 g/10 min, 200°C/2.16 kg) supplied by Borealis Polyolefins, Austria, was used as matrix polymer. Maleic anhydride grafted polypropylene (MAPP, Epoline E-43, Eastman Chemical Company, USA), lubricant (TPW 113, Struktol, USA) and antioxidant agent were used as additives. In the chemical pre-treatment of wood chips sodium sulphite (Na₂SO₃, Merck KGaA, Germany) was used.

2.2 Extrusion

A co-rotating twin-screw extruder with twin-screw side feeder, Coperion W&P ZSK-18 MEGALab (Stuttgart, Germany) equipped with K-Tron gravimetric feeders (Niederlenz, Switzerland), two atmospheric vents and a vacuum vent was used in the experiments (Fig. 8). The screw shafts (Fig. 9) in Coperion W&P ZSK-18 MEGALab twin screw extruder have an elemental structure and the screw configurations can therefore be varied.

Fig. 8. Co-rotating twin-screw extruder, Coperion W&P ZSK-18 MEGALab [14].
2.3 Characterization

In the characterization of wood particles the emphasis was to determine the size and shape of the particles after extrusion. For this purpose, the polymer matrix was dissolved from the composites using boiling xylene. In the characterization of the composites the focus was on mechanical testing and microscopy.

2.3.1 Fractionation

The fractionation method was used in this work to divide the wood particle samples into different size categories and to obtain images of the samples for the image analysis. The fractionation was done using a tube flow fractionator (Metso Automation, Kajaani, Finland). Tube flow fractionation is a method in which the sample (in water suspension) is injected into a continuous plug flow of water, which then enters a long plastic tube. As the sample flows in the tube it is fractionated by size. The fractionation device is equipped with a CCD camera, which records approximately 600 images of the sample during the analysis. The basic principle of the device is presented in Fig. 10. Tube flow fractionation is typically used in the analysis of wood pulp samples for papermaking purposes.

Fig. 9. The screw shafts of Coperion W&P ZSK-18 MEGALab co-rotating twin-screw extruder.
Fig. 10. Schematic description of the tube flow fractionator [15].

In addition to the tube flow fractionator, a Bauer McNett classifier was used in Paper I to divide samples for microscopic analysis.

2.3.2 Image analysis

The length and width of the wood particles were measured using an optical fibre analyzer FiberLab (Metso Automation, Kajaani, Finland, Paper I) and image analysis software kajaaniIMG (Metso Automation, Kajaani, Finland, Papers II-IV). The average length (length weighted) and average width measurements were then used to calculate the aspect ratio of the wood particles and fibres. The images captured by the tube flow fractionator were used in the kajaaniIMG image analysis. In brief, the kajaaniIMG software uses several image analysis algorithms to measure the properties of the objects (particles, fines and fibres) in the captured images. A projection area is determined for each object, and based on the shape of the object, parameters such as length and width are decided.

2.3.3 Microscopy

Microscopic methods were used to study the morphology of the wood particles as well as the fractured surfaces of the prepared composites. Optical and scanning electron microscopy (SEM and FESEM) was used to capture images of wood particle samples. Scanning electron microscopy was used to study the microstructure of the fractured surfaces of the composites. The fractured surfaces were created by first freezing the specimen using liquid nitrogen and then breaking it. The wood
particle samples were freeze-dried before microscopy. The samples for scanning electron microscopy were sputter-coated with platinum or gold before observation.

2.3.4 Mechanical testing

Mechanical properties of the composites were measured using flexural (Papers II-IV) and impact testing (Paper IV). Flexural testing of the wood chip-PP composites was performed according to ASTM D790 standard and with a Shimadzu Autograph AG-X universal testing machine (Shimadzu Corp, Kyoto, Japan). A falling weight impact test was used to measure the impact properties of the prepared composites. Testing was done with a Dynatup Minitower (Instron, UK) falling weight impact testing machine and according to ASTM D3763 standard.

2.3.5 Density

The density of the composites was measured using either a pycnometer (AccuPyc 1330, Micromeritics Instrument Corp, USA, Paper II and III) or the water displacement method according to ASTM D792 standard (test method A, Paper IV). The pycnometer measurement is based on gas displacement technique.
3 Summary of appended papers

Paper I: The effect of processing parameters on wood raw material in an extruder used for manufacturing of wood-polymer composites (WPC)

Paper I was a preliminary study to examine the effect of extrusion parameters on different types of wood raw material in the manufacture of wood-polymer composites. The aim was to produce wood particles with as high aspect ratio as possible. The study consisted of two parts: 1) the breaking and separation of individual fibres from wood chips during the extrusion process, and 2) the effect of chemical pre-treatment and reversed screw elements (RSE) on pre-crushed wood raw material. This study showed that a twin-screw extruder can be used to separate individual fibres from wood chips, and the separated fibres have higher aspect ratios than the wood flour particles typically used in wood-polymer composites. When more fibrous and chemically softened wood raw material was used, fibres with even higher aspect ratios were obtained.

Paper II: Wood Chips as Raw Material in Wood Plastic Composites

In Paper II wood chip-polypropylene composites were manufactured using extrusion. The aim was to study the effect of the extrusion process on wood chip aspect ratio and to measure the mechanical properties of the prepared composite materials. Two different compounding methods were used in manufacturing the composites. Dried and undried wood chips were used as wood raw materials. The results showed that both the processing method and the wood chips’ moisture content had an effect on the wood particle size and aspect ratio. Individual fibres and wood particles with high aspect ratio could be separated from the wood particles when wet wood chips were used as raw material. The composites manufactured with dry wood chips had slightly better flexural properties compared to composites manufactured with wet wood chips, despite the fact that the aspect ratio of wood particles in wet wood chip composites was higher.
Paper III: Processing of Wood Chip-Plastic Composites: Effect on Wood Particle Size, Microstructure and Mechanical Properties

Paper III is a revised version of Paper II, which is a conference paper. In addition to the results presented in Paper II, the densities of the composites were measured and the flexural properties of the composites were normalized to respond 50 wt% wood content. Also, new images were taken from the fractured surfaces of the composites to achieve better understanding of the interfacial adhesion between wood particles and polymer matrix in the composites.

Paper IV: The effect of pre-softened wood chips on wood fibre aspect ratio and mechanical properties of wood-polymer composites

In Paper IV chemically pre-treated wood chips were used as raw material for wood-polypropylene composites. The aim was to study the effect of sulphonation pre-treatment and moisture content of wood chips on the wood particle aspect ratio and the mechanical properties of the composites. The results showed that the wood chip pre-treatment enhanced both the aspect ratio of wood particles and the mechanical properties of the composites. Combination of pre-treated, undried wood chips resulted in the highest wood particle aspect ratio and the highest flexural strength and impact properties of the composites. As in Paper III, the flexural modulus of the composites decreased when undried wood chips were used as raw material.
4 Conclusions

This study shows that wood chips can be successfully used as raw material for wood-polymer composites. A twin-screw extruder can be used to separate individual fibres from wood chips, and the separated fibres have higher aspect ratios than the wood flour particles typically used in wood-polymer composites. When undried wood chips are used as raw material, fibres and wood particles with high aspect ratio can be separated. It is possible to use undried wood chips as raw material and reach stiffness values similar to the composites in which wood flour has been used as raw material, thereby decreasing the raw material cost for the final product. If the bound moisture from the undried wood chips can be removed during the process, it might be possible to improve the composites’ mechanical properties even further.

5 Future work

Further development and optimization of the extrusion process for wood chip-polymer composites is necessary. The removal of all the moisture from the undried raw material during extrusion would be a particularly significant improvement. As proper softening of the wood raw material is essential for separation of wood particles and fibres with a higher aspect ratio, pre-softening of wood raw material and even more prior processing are of interest.
6 Acknowledgements

This work was carried out as part of a joint project between Luleå University of Technology, Sweden and University of Oulu, Finland.

I would like to thank both of my supervisors, Professor Kristiina Oksman and Professor Jouko Niinimäki for their valuable help in making this work possible. My sincere gratitude goes also to all of my colleagues in Luleå and Oulu, who have helped me in the lab and outside the lab.

Last, but not the least, I wish to thank the most important persons in my life, Jani and my parents and sisters, for their support.
7 References


Paper I
The effect of processing parameters on wood raw material in an extruder used for manufacturing of wood-polymer composites (WPC)

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Abstract

This study was made as a preliminary study to examine the effect of extrusion parameters on different types of wood raw material in manufacturing of wood-polymer composites. The aim was to produce wood particles with as high aspect ratio as possible. The study consisted of two parts, the first part was to break and separate individual fibres from wood chips during extrusion process and in the second part the effect of chemical pre-treatment and reversed screw elements (RSE) on wood raw material was evaluated. Statistical analysis was made in purpose to evaluate the most important factors affecting wood particle size in extrusion. The statistical analysis showed that the screw speed is the main factor affecting wood fibre length in twin-screw extrusion of wood chips. Higher speed resulted in higher aspect ratio. This study showed that a twin-screw extruder can be used to separate individual fibres from wood chips, and the separated fibres have higher aspect ratios than the wood flour particles typically used in wood-polymer composites. When more fibrous and chemically softened wood raw material was used, fibres with even higher aspect ratios were achieved.

Keywords: Wood, extrusion, statistical analysis, microscopy, particle analysis
1 Introduction

Wood-polymer composites (WPC) are typically described as materials which combine the properties of wood and thermoplastic materials. The biggest market for WPCs at the moment is outdoor building materials, but also automotive parts and furniture are manufactured from WPCs [1]. When compared with inorganic and synthetic fillers and reinforcements traditionally used in polymer composites, wood is considered as low-cost and more environmentally friendly raw material.

Most of the commercially manufactured WPCs today are made by compounding wood flour consisting of small wood particles with polyolefin polymer, e.g. polyethylene or polypropylene as matrix [2]. Wood flour is produced from the wood residue of various wood processors by size reduction with different kinds of mills and size classification of the pulverised wood by screening [3]. The particles in wood flour consist actually of fibre bundles, not individual fibres, and therefore the aspect ratio (length-to-diameter ratio) of wood flour is quite low, typically between 1 and 5 [4]. Because of the low aspect ratio the reinforcing potential of wood flour is limited, and wood flour acts mainly as filler material in composites. The aspect ratio of a softwood fibre, for example, can be 100 [5]. Earlier studies have also shown that wood fibres with a higher aspect ratio have better reinforcing capability in wood-polymer composites [6, 7]. However, fibres are seldom used in manufacturing of commercial WPCs because feeding and metering of low-bulk-density fibres is difficult and they are also more expensive than wood flour [2, 3]. Problems with poor dispersion of longer wood fibres in composites have also been reported [8, 9].

WPC research has strongly concentrated on enhancing the interfacial adhesion between the wood and polymer [10-15] and the effect of wood characteristics is less frequently reported. The effect of wood particle size and aspect ratio on composite properties has been studied to some extent [7, 16-
but only a few studies can be found where the effect of extrusion compounding on the dimensional properties of wood fibres and particles is examined [21-24], even though twin-screw extrusion is one of the main processing methods used in the manufacture of WPC.

In twin-screw extrusion shear forces are subjected into the material, and the amount of shear forces depends greatly on the used screw configuration. The shear forces could also be used to reduce the particle size and to separate fibres with a higher aspect ratio from the wood particles if suitable processing conditions can be found. Thus, the wood raw material would not need as much pre-processing as the commonly used wood flour requires before it can be compounded with plastic in an extruder. If larger, undried wood particles can be used as raw material in WPCs, use of cheaper wood residues in composite manufacturing without pre-processing could be possible.

This study is a preliminary study to examine the effect of extrusion parameters on different types of wood raw material in manufacture of wood-polymer composites. The main goal was to produce wood particles/fibres with as high aspect ratio as possible. Basically the study consisted of two parts. In the first part, the possibility to break down and separate individual fibres from wood chips in a co-rotating twin-screw extruder was examined. Statistical analysis was made in purpose to evaluate the most important factors affecting wood particle size in extrusion. The effect of raw material moisture content, screw configuration and screw speed on the aspect ratio of wood particles was evaluated using multiple regression analysis. In the second part of the study the effect of chemical raw material pre-treatment and reversed screw elements (RSE) on more fibrous wood raw material was evaluated. The raw material and pre-treatment were chosen because the pre-treatment performed in the first part of the study was considered insufficient.
The dimensional properties of the extruded wood fibres and particles were measured using optical fibre analysis. Scanning electron microscopy was also used to study the morphology of the samples. To avoid the laborious matrix removal, all the experiments were carried out without addition of matrix polymer in the extruder.

2 Materials and methods

2.1 Materials

Spruce wood chips (Fig. 1) were used as raw material in the first part of the study. The average wood particle size was of 16.2 mm × 3.3 mm. Wood chip particle size was determined by measuring the length and the width of 100 particles manually. Wood chips were stored frozen until use. The wood chips were used as untreated (defrosted) and as pre-treated in boiling water for 4 minutes immediately before extrusion. The purpose of the pre-treatment was to soften the wood chips and thus avoid crushing them into small particles and thereby facilitate the separation of individual fibres. The dry matter content of the non-treated wood chips was 57 wt% and the dry matter content of the pre-treated wood chips was 30-35 wt%.

Fig. 1. Spruce wood chips used in this study.  
Fig. 2. Pre-crushed spruce used in this study.
In the second part of the study pre-crushed spruce (Fig. 2) was used as a raw material. This raw material was more fibrous in comparison with wood chips. The pre-crushed spruce was used as such (untreated) and as chemically pre-treated. The pre-treatment was a mild sulphonation, and it was done to achieve more softened raw material. The sulphonation was done at 90 °C and at pH 9 for 210 minutes. The raw material was diluted into 10 % consistency and 5 % of Na$_2$SO$_3$ was used in the treatment. The dry matter content of the untreated, pre-crushed spruce (referred as PC-U) was 46.3 wt%, and the dry matter content of sulphonated pre-crushed spruce (referred as PC-S) was 36.3 wt%.

2.2 Extrusion equipment

Co-rotating twin-screw extruder Coperion W&P ZSK-18 MEGALab (Stuttgart, Germany) without the extrusion die was used in the experiments. Feeding of the wood chips into the extruder was done by hand due to the fact that the feeding equipment was not suited for materials used in the study. The extruder screws were fed as full as possible to keep the filling degree of the screws similar between experiments.

2.3 Characterization

The average fibre length (length weighted) and average fibre width used to calculate the aspect ratio (l/d) were measured using an optical fibre analyzer FiberLab (Metso Automation, Kajaani, Finland). Morphology of selected samples was studied using field emission scanning electron microscope, FESEM (Zeiss ULTRA Plus, Carl Zeiss SMT AG, Jena, Germany) and scanning electron microscope, SEM (Jeol JSM-6400, Jeol Ltd., Tokyo, Japan). The samples for scanning electron microscopy were freeze dried and coated with platinum (FESEM) or gold (SEM) before examination.
In the experiments in which wood chips were used as raw material, it was necessary to fractionate the samples into four different size categories before measuring the fibre lengths. The fractionation was done using a tube flow fractionator (Metso Automation, Kajaani, Finland). The purpose of the fractionation was to remove the first fraction containing the largest wood particles (shives) from the sample, and thus to avoid the blocking of the FiberLab analyzer. The average fibre lengths were then measured by combining the fractions 2, 3 and 4 from the tube flow fractionator into a one sample. The tube flow fractionation method is described in more detail in following publications: Laitinen et al. 2006 [25] and Krogerus et al. 2003 [26].

In addition, Bauer McNett classifier was used to divide samples into fractions R14, R28, R48 and R200 before examination with SEM in the second part of the study. The R14 fraction is typically considered as the shives fraction, R28 as the long fibre fraction, R48 as the short fibre fraction and R200 as the fines fraction.

2.4 Experimental

2.4.1 Screening of the important factors

The aim of the first part of this study was to find out the most important factors affecting wood aspect ratio of in a co-rotating twin-screw extruder. MODDE 8 (Umetrics) statistical software and multiple linear regression analysis (MLR) were used to analyse the experimental data. Confidence level of 95 % was used in the regression analysis. The aspect ratio of wood particles after extrusion was used as a response.

The experiments were performed according to a non-replicated $2^3$ full-factorial design with three centre points. The total amount of experimental runs was 11. Experimental design is presented in Table 1. The experiments were performed in randomized order. The factors studied were 1)
rotational speed of the screws, 2) screw configuration and 3) raw material treatment. The rotational speeds used were 100, 300 and 500 rpm. 300 rpm was functioned as a centre point in the design. Two different screw configurations were used: A) screw consisting of conveying elements and one wide kneading element (WKE), and B) screw consisting of conveying elements with one wide kneading element (WKE) followed by reversed screw element (RSE). The raw material used was either non-treated or pre-treated wood chips.

Table 1. Experimental design.

<table>
<thead>
<tr>
<th>Exp. no.</th>
<th>Run order</th>
<th>Raw material</th>
<th>Speed</th>
<th>Configuration</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>11</td>
<td>non-treated</td>
<td>100 rpm</td>
<td>A</td>
</tr>
<tr>
<td>2</td>
<td>2</td>
<td>non-treated</td>
<td>500 rpm</td>
<td>A</td>
</tr>
<tr>
<td>3</td>
<td>10</td>
<td>non-treated</td>
<td>100 rpm</td>
<td>B</td>
</tr>
<tr>
<td>4</td>
<td>3</td>
<td>non-treated</td>
<td>500 rpm</td>
<td>B</td>
</tr>
<tr>
<td>5</td>
<td>1</td>
<td>pre-treated</td>
<td>100 rpm</td>
<td>A</td>
</tr>
<tr>
<td>6</td>
<td>6</td>
<td>pre-treated</td>
<td>500 rpm</td>
<td>A</td>
</tr>
<tr>
<td>7</td>
<td>8</td>
<td>pre-treated</td>
<td>100 rpm</td>
<td>B</td>
</tr>
<tr>
<td>8</td>
<td>4</td>
<td>pre-treated</td>
<td>500 rpm</td>
<td>B</td>
</tr>
<tr>
<td>9</td>
<td>5</td>
<td>non-treated</td>
<td>300 rpm</td>
<td>A</td>
</tr>
<tr>
<td>10</td>
<td>7</td>
<td>non-treated</td>
<td>300 rpm</td>
<td>A</td>
</tr>
<tr>
<td>11</td>
<td>9</td>
<td>non-treated</td>
<td>300 rpm</td>
<td>A</td>
</tr>
</tbody>
</table>

* A: screw configuration with WKE, B: screw configuration with WKE and RSE.

The factor levels used in the experiments are presented in Table 2. The factor levels were chosen in a way that they would be as extreme as possible. There were some limitations when selecting suitable levels, e.g. reasonable processing time and maximum motor load had to be taken into consideration. To avoid excessive drying or burning of the wood in the extruder, the barrel temperature was set to 80°C in all experimental runs.
Table 2. Factor levels used in the statistical analysis.

<table>
<thead>
<tr>
<th>Factor</th>
<th>Factor levels</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>-1</td>
</tr>
<tr>
<td>Screw speed, rpm</td>
<td>100</td>
</tr>
<tr>
<td>Configuration</td>
<td>A</td>
</tr>
<tr>
<td>Raw material treatment</td>
<td>non-treated</td>
</tr>
</tbody>
</table>

The screw configurations used in the experiments are presented in Fig. 3. The screw elements were chosen so that configuration A would be gentler than B. The reversed screw element, RSE, in configuration B has threads in the opposite direction than the other elements, and therefore it resist the material flow effectively [27]. Pre-runs made with different screw configurations also showed that using of wide kneading element in front of RSE was necessary to avoid motor overload of the extruder when using wood chips as raw material.

![Fig. 3. Screw configurations A and B. WKE = wide kneading element, RSE = reversed screw element.](image)

2.4.2 Effect of raw material and screw configuration

Because the pre-treatment made for the wood chips in the first part of the study did not increase the wood aspect ratio, it was decided to use a more fibrous raw material with and without chemical pre-treatment in the second part of the study. Due to the change in raw material, reversed screw element,
RSE, could now be used as the first screw element. Three different screw configurations were used in the experiments (Fig. 4). Configuration C had one reversed screw element (RSE), configuration D had two RSEs and configuration E had two RSEs with a thin kneading element between them (TKE) in addition to conveying elements (Fig. 4). The temperature of the extruder barrel was set to 70°C to avoid unwanted drying of the raw material. Screw speed in all experimental runs was 500 rpm. The experimental plan with extrusion parameters is presented in Table 3.

![Screw configurations C, D and E. RSE = reversed screw element, TKE = thin kneading element.](image)

Table 3. The experimental settings.

<table>
<thead>
<tr>
<th>Exp.</th>
<th>Temperature, °C</th>
<th>Screw speed, rpm</th>
<th>Configuration</th>
</tr>
</thead>
<tbody>
<tr>
<td>PC-U1</td>
<td>70</td>
<td>500</td>
<td>C</td>
</tr>
<tr>
<td>PC-U2</td>
<td>70</td>
<td>500</td>
<td>D</td>
</tr>
<tr>
<td>PC-U3</td>
<td>70</td>
<td>500</td>
<td>E</td>
</tr>
<tr>
<td>PC-S1</td>
<td>70</td>
<td>500</td>
<td>C</td>
</tr>
<tr>
<td>PC-S2</td>
<td>70</td>
<td>500</td>
<td>D</td>
</tr>
<tr>
<td>PC-S3</td>
<td>70</td>
<td>500</td>
<td>E</td>
</tr>
</tbody>
</table>

PC-U=untreated pre-crushed spruce, PC-S=sulphonated pre-crushed spruce.
3 Results and discussion

3.1 Screening of the important factors

The average fibre length (length weighted) and fibre width as well as aspect ratios (l/d) calculated from the length and width measurements are presented in Table 4. As Table 4 shows, the experiment no. 8, which was made using pre-treated chips, 500 rpm screw speed and screw configuration B (configuration with WKE and RSE), had the highest wood particle aspect ratio among the experimental runs.

Table 4. Average fibre length, average width and the aspect ratio of the samples.

<table>
<thead>
<tr>
<th>Exp.</th>
<th>Raw material</th>
<th>Speed, rpm</th>
<th>Screw configuration</th>
<th>Length, L(l), mm</th>
<th>Width, g80</th>
<th>Aspect ratio, l/d</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>non-treated 100 A</td>
<td>0.65</td>
<td>31.1</td>
<td>20.9</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>non-treated 500 A</td>
<td>0.68</td>
<td>31.2</td>
<td>21.8</td>
<td></td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>non-treated 100 B</td>
<td>0.55</td>
<td>30.6</td>
<td>18.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>non-treated 500 B</td>
<td>0.68</td>
<td>30.9</td>
<td>22.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>pre-treated 100 A</td>
<td>0.59</td>
<td>29.9</td>
<td>19.7</td>
<td></td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>pre-treated 500 A</td>
<td>0.67</td>
<td>29.4</td>
<td>22.8</td>
<td></td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>pre-treated 100 B</td>
<td>0.55</td>
<td>28.9</td>
<td>19.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>pre-treated 500 B</td>
<td>0.67</td>
<td>27.8</td>
<td>24.1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>9</td>
<td>non-treated 300 A</td>
<td>0.64</td>
<td>30.7</td>
<td>20.8</td>
<td></td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>non-treated 300 A</td>
<td>0.67</td>
<td>30.3</td>
<td>22.1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>11</td>
<td>non-treated 300 A</td>
<td>0.63</td>
<td>30.5</td>
<td>20.7</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

However, the statistically significant factors affecting the aspect ratio of wood raw material in twin-screw extrusion were estimated using MODDE 8 statistical software and multiple linear regression analysis (MLR). Fig. 5 shows the coefficient plot for the aspect ratio before reducing the insignificant factors from the model and the results from the regression analysis for the reduced model are presented in Tables 5 and 6. Table 5 shows the regression coefficients for the significant
(α = 0.05) factors affecting the aspect ratio of wood raw material in a co-rotating twin-screw extruder together with the $R^2$, $R^2_{(adj.)}$ and $Q^2$ parameters measuring the goodness of fit of the model. The analysis of variance, ANOVA, for the fitted model is presented in Table 6. From Table 5 it can be seen that according to the MLR analysis, screw speed (Scr) had the largest effect on aspect ratio, and it was also the only significant factor with a 95% confidence level affecting the aspect ratio. The $R^2$ value for this model was 0.704. The $R^2_{(adj.)}$ for the model was 0.671, and the $Q^2$ value for the model was 0.536. These parameters indicate that the model fits the data moderately. The analysis of variance (ANOVA) for the fitted model is presented in Table 6. The P-value for lack of fit for this model was 0.399 indicating that there is no lack of fit.

![Fig. 5. The scaled and centred regression coefficients for aspect ratio before model reduction.](image)

Table 5. Results from the regression analysis for the fitted model for aspect ratio.

<table>
<thead>
<tr>
<th>Response</th>
<th>Constant</th>
<th>Scr</th>
<th>Con</th>
<th>Raw</th>
<th>Scr×Con</th>
<th>Scr×Raw</th>
<th>Con×Raw</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aspect ratio</td>
<td>21.015</td>
<td>1.638</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

$R^2 = 0.704$, $R^2_{(adj.)} = 0.671$, $Q^2 = 0.536$

Scr = screw speed, Con = configuration, Raw = raw material treatment.
Table 6. The analysis of variance (ANOVA) for the fitted model for aspect ratio.

<table>
<thead>
<tr>
<th>Response</th>
<th>Source</th>
<th>DF</th>
<th>SS</th>
<th>MS</th>
<th>F-value</th>
<th>P-value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aspect ratio</td>
<td>Regression</td>
<td>1</td>
<td>21.45</td>
<td>21.45</td>
<td>21.44</td>
<td>0.001</td>
</tr>
<tr>
<td></td>
<td>Lack of Fit</td>
<td>7</td>
<td>7.79</td>
<td>1.11</td>
<td>1.82</td>
<td>0.399</td>
</tr>
<tr>
<td></td>
<td>Residual</td>
<td>9</td>
<td>9.01</td>
<td>1.00</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Pure Error</td>
<td>2</td>
<td>1.22</td>
<td>0.61</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Total</td>
<td>11</td>
<td>4919.33</td>
<td>447.21</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

DF = degrees of freedom, SS = sum of squares, MS = mean squares.

Even though the regression analysis showed that the screw configuration did not have a significant effect on aspect ratio and it had only a very small effect on fibre length, it was thought that the real effect of screw configuration was not clearly seen in this experiment. The wide kneading element, WKE, was used in both screw configurations as the first element to avoid motor overload, and it was suspected that actually the results show that the reversed screw element (RSE) had a small fibre shortening effect after the wood had first gone through the wide kneading element. Thus it is possible that the kneading element was responsible of the extensive fibre shortening.

FESEM pictures from experiments no. 2, 4, 6 and 8 are presented in Fig. 6. These samples were chosen for the analysis because they had the longest fibre lengths and highest aspect ratios according to the analysis. The FESEM images in Fig. 6 support the view that screw configuration had a larger effect on wood fibre properties than the statistical analysis showed. The screw speed in these four experiments was 500 rpm. From Fig. 6 it can be seen that the samples in which screw configuration A (configuration with wide kneading element. WKE) was used (exp. 2 and 6) contain more coarse material (more shives) in comparison with the samples in which the screw configuration B was used (exp. 4 and 8).
Fig. 6. Scanning electron microscopy images of experiments no. 2, 4, 6 and 8.

3.2 Effect of raw material type and reversed screw elements

The results from the average fibre length and fibre width measurements together with the calculated aspect ratios are presented in Table 7. In these experiments the pre-crushed, more fibrous wood raw material was used and as Table 7 shows, the aspect ratios of the extruded samples are significantly higher when compared to the aspect ratios measured from the samples in which wood chips were used as raw material (Table 4).

Table 7. Fibre properties of the pre-crushed spruce after extrusion with three different screw configurations.

<table>
<thead>
<tr>
<th>Exp.</th>
<th>Configuration</th>
<th>Length, L(l), mm</th>
<th>Width, μm</th>
<th>Aspect ratio, l/d</th>
</tr>
</thead>
<tbody>
<tr>
<td>PC-U1</td>
<td>C</td>
<td>1.21</td>
<td>27.3</td>
<td>44.3</td>
</tr>
<tr>
<td>PC-U2</td>
<td>D</td>
<td>0.82</td>
<td>25.0</td>
<td>32.8</td>
</tr>
<tr>
<td>PC-U3</td>
<td>E</td>
<td>0.85</td>
<td>25.5</td>
<td>33.3</td>
</tr>
<tr>
<td>PC-S1</td>
<td>C</td>
<td>1.59</td>
<td>30.0</td>
<td>53.0</td>
</tr>
<tr>
<td>PC-S2</td>
<td>D</td>
<td>1.20</td>
<td>27.9</td>
<td>43.0</td>
</tr>
<tr>
<td>PC-S3</td>
<td>E</td>
<td>1.24</td>
<td>27.8</td>
<td>44.6</td>
</tr>
</tbody>
</table>

PC-U = pre-crushed, untreated spruce. PC-S = pre-crushed, sulphonated spruce.
Based on the results received from the first part of this study, three different screw configurations were used in the experiments. The highest aspect ratios were achieved using screw configuration C (samples PC-U1 and PC-S1) with one reversed screw element. The screw configurations with two reversed screw elements (configuration D and E) were most likely too harsh and therefore the aspect ratios were lower in the samples made with these. As expected, the sulphonation pre-treatment affected the aspect ratio of the extruded fibres positively. This indicates that a proper softening of the wood raw material is very important to gain high aspect ratio fibres in extrusion of wood raw material.

In Fig. 7 the SEM images of R14, R28, R48 and R200 mass fractions of two of the samples (PC-U1 and PC-S1) are shown. The samples were divided into fractions to achieve a better understanding of the morphology of the fibres. The images in Fig. 7 show that both of the samples contain individual fibres and only a small amount of shives. When comparing the images taken from the different fibre fractions, especially R14 fractions of the samples, it seems that PC-U1 contains coarser material than the PC-S1 sample (Fig. 7).

Fig. 7. Scanning electron microscopy images of R14, R28, R48 and R200 fractions of samples PC-U1 and PC-S1. PC-U = untreated pre-crushed spruce, PC-S = sulphonated pre-crushed spruce.
4 Conclusions

This study showed that a twin-screw extruder can be used to separate individual fibres from wood chips, and the separated fibres have higher aspect ratios than the wood flour particles typically used in wood-polymer composites. When more fibrous and chemically softened wood raw material was used, fibres with even higher aspect ratios were achieved.

The statistical analysis showed that the screw speed is the main factor affecting the aspect ratio of wood raw material in twin-screw extrusion of wood chips. Higher speed resulted in higher aspect ratio. However, the effect of screw configuration on aspect ratio can’t be totally discarded, due to the choice of configurations in the experimental design. More extensive experimental plan with replicates could give more reliable data regarding on the most important factors affecting aspect ratio in extrusion.

5 References


Paper II
11th International Conference on Biocomposites:
Transition to Green Materials

Wood Chips as Raw Material in Wood Plastic Composites

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Abstract

In this study, wood chips were used as raw material for wood-plastic composites (WPCs) and the effect of extrusion process on wood chips was studied. WPCs with wood content of approximately 50 wt% were manufactured by using two different compounding methods. In the first method the wood chips were added to molten PP via side feeder. In the second method the feeding order was the opposite: wood chips were fed first, ground into smaller particles and PP was added as a second component using the side feeder. The screw configurations used in these two methods differed from each other, and they were built to enable the size reduction of wood chips. Dried and undried wood chips were used to investigate whether the drying process could be carried out in the same step. Wood particle properties, such as aspect ratio and size distributions, were measured using optical fibre analysis and microscopical methods were used to examine the microstructure of wood particles. Furthermore, the prepared composites' mechanical properties were studied.

1 Introduction

Wood-plastic composites (WPCs) are materials which have a wood-like appearance but can be processed like plastic materials. When compared to solid wood, WPCs have better durability and lower maintenance requirements. Wood is also considered a low-cost and renewable material in comparison with inorganic and synthetic fillers traditionally used in polymer composites. Currently the biggest market for WPCs is outdoor building materials, but automotive parts and furniture are also manufactured from WPCs [1].

WPCs are manufactured by compounding wood particles with thermoplastic matrix. The most typical processing methods used to produce WPCs are extrusion compounding followed by profile extrusion or injection moulding. The matrix polymer is usually a low-cost polymer with a processing temperature below the wood degradation temperature. The most common matrix polymers in WPCs are polyethylene (PE), polypropylene (PP) and polyvinyl chloride (PVC) [2].

The most widely used wood-derived raw material in commercially manufactured WPCs is wood flour. Wood flour is produced from the scrap materials of various wood processors by size reduction with different kinds of mills and size classification of the pulverized wood by screening [3]. The particle size of commercially manufactured wood flour is usually smaller than 425 µm, and the aspect ratio (length-to-diameter ratio) is between 1 and 5 [4]. The low aspect ratio limits the reinforcing potential of wood flour [5], and studies have shown that wood fibres with a higher aspect ratio have better reinforcing capability [6, 7]. Especially when good adhesion between fibre and matrix exists, fibres provide better stress transfer to the fibres [5]. However, fibres are seldom used in manufacturing of commercial WPCs because feeding and metering of low-bulk-density fibres is difficult and they are more expensive than wood flour [2, 5]. Problems with poor dispersion of wood fibres in composites have also been reported [8, 9].

A great deal of WPC research has concentrated on enhancing the interfacial adhesion between hydrophilic wood and hydrophobic thermoplastic matrix due to the incompatibility problems of these components. Particularly, the use of different coupling agents and fibre surface treatments has been studied extensively [10-15]. Research activities focusing on the effect of wood characteristics and processing conditions on WPC properties have been less frequently reported. The effect of wood particle size on composite properties has been studied to some extent [7, 16-19], but only a few studies can be found where larger wood particles have been used in the
In addition, there have been few studies on the effect of extrusion compounding on wood particle size and fibre length [21, 22], even though twin-screw extrusion is one of the main processing methods used in the manufacture of WPC.

In the study by Bledzki and Faruk [17] the physical and mechanical properties of composites made of PP and different types of wood fibres and wood chips were compared. They concluded that wood chips-PP composites showed better tensile and flexural properties than the other wood-PP composites when 5% coupling agent was added. Balasuriya et al. [20] used wood flakes in wood-HDPE -composites in their study of the effect of wood flake distribution and wetting on the structure-property relationship of WPC. The size of these wood flakes was 1-4 mm × 0.1-2 mm. They also measured the flake length and width distributions after compounding and reported that the length of wood flakes was reduced.

Other studies have also shown that wood particle size and fibre lengths are reduced during the compounding [21-24]. Usually, this reduction is considered a disadvantage, but the shear forces in extrusion could also be used to reduce the particle size of larger wood particles. It is possible that individual fibres with a higher aspect ratio can be separated from wood particles if an optimal screw configuration and processing method is found. Thus, the wood raw material would not need as much pre-processing as the commonly used wood flour requires before it can be compounded with plastic in an extruder. If larger, undried wood particles can be used as raw material in WPCs, use of cheaper wood residues in composite manufacturing may be possible.

In this study the use of wood chips as a raw material in a wood plastic composite is investigated. The high shear forces in a co-rotating twin-screw extruder are utilized to grind wood chips into smaller particles, possibly even into individual fibres, which are then compounded with polymer matrix in the same processing step. Two different extrusion processing methods are used, and both dried and undried wood chips are used to study the possibility of carrying out the drying process of wood in the same process step. In addition, the effect of wood moisture content on wood particle size during extrusion is examined. Wood particle properties such as aspect ratio and size distributions after extrusion are analysed. Furthermore, the mechanical properties of the prepared composites are studied.

### 2 Experimental

#### 2.1 Materials

Spruce pin chips received from UPM Kymmene (Lappeenranta, Finland) were used as wood raw material (Fig 1.).

![Figure 1. Spruce wood chips used in this study](image)

Wood chip particle size was determined by sieving and by measuring the length and the width of 100 particles manually. The results from the sieve analysis are presented in Table 1. A vibratory sieve shaker (Fritsch Analysette 3 PRO) and sieves with aperture size of 8 mm, 4 mm, 2 mm, 1 mm, 500 µm and 250 µm were used in the sieve analysis.

<table>
<thead>
<tr>
<th>Particle size</th>
<th>Mass fraction (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>&gt; 8 mm</td>
<td>0.3</td>
</tr>
<tr>
<td>4-8 mm</td>
<td>10.8</td>
</tr>
<tr>
<td>2-4 mm</td>
<td>54.8</td>
</tr>
<tr>
<td>1-2 mm</td>
<td>25.6</td>
</tr>
<tr>
<td>0.5-1 mm</td>
<td>6.9</td>
</tr>
<tr>
<td>250-500 µm</td>
<td>1.3</td>
</tr>
<tr>
<td>&lt; 250 µm</td>
<td>0.3</td>
</tr>
</tbody>
</table>

According to manual measurements, average particle length was 16.2 mm and average particle width was 3.3 mm. Initial moisture content of the wood chips was 40 wt% based on total weight. Half of the wood chips were dried at 105°C for 24 h to reach moisture content below 1 wt% (based on dry weight) before processing. The other half of the wood chips used was undried.

Polypropylene in powder form (MFI = 32 g/10 min, 230°C/2.16 kg and 16 g/10 min, 200°C/2.16 kg) supplied by Borealis Polyolefins, Austria was used as matrix polymer. Maleic anhydride grafted polypropylene, MAPP (Epolene E-43), lubricant (Struksol TPW 113) and antioxidant agent were used as additives.
2.2 Processing of WPC

2.2.1 Extrusion compounding

A co-rotating twin-screw extruder with twin-screw side feeder, Coperion W&P ZSK-18 MEGALab (Stuttgart, Germany) equipped with K-Tron gravimetric feeders (Niederlenz, Switzerland) was used to compound the wood chip-PP composite materials. Before extrusion the polypropylene and the additives were pre-mixed. Gravimetric feeders were used to feed PP into the extruder while wood chips were fed into the extruder manually because it was not possible to use these feeders for this kind of material.

Four types of WPCs were manufactured using dry and wet wood chips (undried wood chips) and two different processing methods, method I and method II. Method I: PP and additives were fed into the first feeding zone (A) and wood chips were fed into the third zone (B). Method II: wood chips were fed into the zone A and PP and additives were fed in to the zone B. Extrusion set-ups for methods I and II are shown in Fig. 2 and 3. Different screw configurations were used in method I and II. The configuration used in method I (PP fed first) is presented in Fig. 4, and the configuration used in method II (wood chips fed first) is presented in Fig. 5. The screw elements used in the configurations were based on the feeding order of the materials. The compounded composites were profiled into rectangular profile with a cross section of approximately 5 x 20 mm.

Processability of composites with dry wood chips was quite good, though there was some heat generation at the extrusion zones where kneading screw elements were located, due to the high shear forces. This was especially evident when processing method II and dry wood chips were used. Processing of composites with wet wood chips was slightly more difficult, due to the fact that the moisture present in the wood chips was vaporized during extrusion.

The wood content of composites was calculated according to the feeding rate of PP (kg/h) and sampling time. The initial goal was to prepare composites with a wood content of 50 wt%. The exact material composition of each material is shown in Table 2.

Table 2. Material compositions of the wood chip-PP composites

<table>
<thead>
<tr>
<th>Materials/Method</th>
<th>Weight%</th>
</tr>
</thead>
<tbody>
<tr>
<td>PP-Dry chip/I</td>
<td>39.3</td>
</tr>
<tr>
<td>PP-Wet chip/I</td>
<td>42.5</td>
</tr>
<tr>
<td>Dry chip-PP/II</td>
<td>40.0</td>
</tr>
<tr>
<td>Wet chip-PP/II</td>
<td>43.8</td>
</tr>
</tbody>
</table>

1 Polypropylene includes 0.18 wt% of antioxidant agent.

The composites manufactured with processing method I had a rougher surface in comparison with composites manufactured with method II, as can be seen from Fig. 6. The composites with dry wood chips had a typical WPC colour, while the composites with wet wood chips were lighter in colour (Fig. 6).
2.2.2 Compression moulding

Test specimens for mechanical testing were moulded with a conventional compression moulding press (Fjellman Press AB, Mariestad, Sweden). The mould temperature was 50°C and the pressure applied on the samples was approximately 70 MPa. Before the moulding, extruded profiles (34g batch weight) were heated in a hot air oven at 200-210°C for 30 minutes.

In addition, the composite samples in which wet wood chips were used as raw material were dried at 75°C for 12-34 h (PP-Wet chip and Wet chip-PP) before compression moulding in order to remove possible moisture left in the composites.

2.3 Characterization of wood particles

2.3.1 Extraction of matrix polymer

To measure fibre length and wood particle size after the compounding, PP matrix was extracted from composites. Prior to extraction, extruded pieces of each composite material were pressed into thin sheets with a heat press (2 min preheating and 30 s pressing at 200°C). 5 g of each material was kept in boiling xylene until the matrix was completely dissolved (15-35 h). The solution was filtered and washed with hot xylene. After this, the sample was washed with ethanol to remove xylene and filtered. Finally, the sample was washed with water to remove ethanol and filtered again.

2.3.2 Fractionation

To achieve better understanding of particle size distributions in each sample, the extracted samples consisting of wood particles and fibres were divided into four different size categories (fractions) by using the tube flow fractionation method (Metso Automation, Kajaani, Finland). After fractionation, mass fractions of each fraction were determined by filtering.

Tube flow fractionation is a method in which the sample (in water suspension) is injected into a continuous plug flow of water, which then enters a long plastic tube. As the sample flows in the tube it is fractionated by size. The fractionation device is equipped with a CCD camera, which records approximately 600 images of the sample during the analysis. The images recorded of each sample were saved and used in the image analysis. Typically, when fractionating pulp samples the first fraction (FR1) is the shives fraction, the second (FR2) and the third (FR3) fraction are middle fractions, and the fourth fraction (FR4) is the fines fraction of the original sample. The tube flow fractionation method is described in more detail in publications by Laitinen et al. and Krogerus et al. [25, 26].

2.3.3 Image analysis

To measure wood particle and fibre dimensions, the images taken by the tube flow fractionation device were analysed using kajaaniIMG image analysis software supplied by Metso Automation (Kajaani, Finland). During image analysis, captured images are segmented to regions that may have one or several objects. The objects (particles, fines and fibres) in one region are then interpreted based on geometrical criteria. To measure properties of each object, several image analysis algorithms are performed. The basis for these algorithms is the model on interaction of light and various objects. This enables the use of so-called sub-pixel algorithms to improve the resolution. For each of the objects, the projection area is determined. Based of the shape of the object, the other parameters such as length and width are then decided. These parameters were then used to calculate the aspect ratio of the wood particles. Settings used in the image analysis are presented in Table 3.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>value</th>
<th>unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Minimum fibre width</td>
<td>1</td>
<td>µm</td>
</tr>
<tr>
<td>Maximum fibre width</td>
<td>500</td>
<td>µm</td>
</tr>
<tr>
<td>Minimum fibre length</td>
<td>0.01</td>
<td>mm</td>
</tr>
<tr>
<td>Maximum fibre length</td>
<td>7.6</td>
<td>mm</td>
</tr>
</tbody>
</table>
2.3.4 Microscopy

To study the morphology of wood particles, microscopic images were taken from the fractionated samples. A Leica MZ LIII stereomicroscope was used to take images of wood particle fractions FR1, FR2 and FR3. Due to the small size of wood particles in fraction FR4, images were taken with field emission scanning electron microscope, FESEM (Zeiss ULTRA Plus). All the microscopy samples were freeze-dried and FESEM samples were sputter-coated with platinum before observation.

2.4 Characterization of composites

2.4.1 Mechanical properties

Flexural testing of the wood chip-PP composites was performed according to ASTM D790 standard (three-point bending). A Shimadzu AG-X universal testing machine with a load cell of 1 kN was used for testing. A minimum of five specimens of each material were tested.

2.4.2 Electron microscopy

Fractured surfaces of composites were studied with a Zeiss ULTRA Plus field emission scanning electron microscope (FESEM). The sample surfaces were sputter-coated with platinum before observation.

3 Results and discussion

3.1 Characterization of wood particles

3.1.1 Fractionation

The extracted samples consisting of wood particles and fibres were divided into four different size categories (fractions) using tube flow fractionation device. After fractionation, mass percentage of each fraction was determined by filtering. The mass fractions of analyzed samples are shown in Fig. 7.

Fig. 7 shows that all the samples have dissimilar particle size distributions, indicating that both the processing method and the moisture content of raw material effected wood particle size during extrusion. It can be seen from Fig. 7 that the share of shives fraction FR1 is larger in samples where processing method I is used as compared to samples where processing method II is used. This is reasonable, because in method I wood chips were fed from the second feeding port, and thus were exposed to less shear forces caused by the screw elements. The use of wet wood as raw material also resulted in a larger share of bigger particles (FR1) compared to samples in which dry wood was used (Fig. 7). This was especially evident when processing method I was used (PP-Wet chip). The amount of fines (fraction FR4) was largest when method II and dry wood chips were used, indicating that this was the harshest processing combination (Fig. 7).

The images recorded of each sample were also saved and used in the image analysis. Examples of the images taken by the fractionation device are shown in Fig. 8.
3.1.2 Image analysis

Results from image analysis of fractionated samples are shown in Table 4. It can be seen from the results that when wet wood chips have been used as raw material the samples contain longer particles in comparison with dry wood chip samples. This is most evident when method I and wet wood chips have been used as raw material. Comparison of aspect ratios between fractionated samples is illustrated in Fig. 9. As Fig. 9 shows, wood particles extracted from composite specimens manufactured with processing method I using wet wood chips (PP-Wet chip) had clearly the greatest aspect ratios.

Table 4. Results from the image analysis

<table>
<thead>
<tr>
<th>Material</th>
<th>Fraction</th>
<th>Mass-%</th>
<th>Average length, L(1), mm</th>
<th>Aspect ratio, l/d</th>
</tr>
</thead>
<tbody>
<tr>
<td>PP-Dry chip</td>
<td>FR1</td>
<td>15.6</td>
<td>0.59</td>
<td>3.5</td>
</tr>
<tr>
<td></td>
<td>FR2</td>
<td>18.1</td>
<td>0.64</td>
<td>8.5</td>
</tr>
<tr>
<td></td>
<td>FR3</td>
<td>32.7</td>
<td>0.22</td>
<td>5.3</td>
</tr>
<tr>
<td></td>
<td>FR4</td>
<td>33.7</td>
<td>0.08</td>
<td>4.4</td>
</tr>
<tr>
<td>PP-Wet chip</td>
<td>FR1</td>
<td>55.1</td>
<td>1.15</td>
<td>19.9</td>
</tr>
<tr>
<td></td>
<td>FR2</td>
<td>11.6</td>
<td>1.12</td>
<td>34.3</td>
</tr>
<tr>
<td></td>
<td>FR3</td>
<td>22.0</td>
<td>0.59</td>
<td>27.9</td>
</tr>
<tr>
<td></td>
<td>FR4</td>
<td>11.4</td>
<td>0.08</td>
<td>5.6</td>
</tr>
<tr>
<td>Dry chip-PP</td>
<td>FR1</td>
<td>49.1</td>
<td>0.08</td>
<td>2.4</td>
</tr>
<tr>
<td></td>
<td>FR2</td>
<td>11.0</td>
<td>0.50</td>
<td>8.8</td>
</tr>
<tr>
<td></td>
<td>FR3</td>
<td>42.1</td>
<td>0.35</td>
<td>12.2</td>
</tr>
<tr>
<td></td>
<td>FR4</td>
<td>34.9</td>
<td>0.10</td>
<td>4.8</td>
</tr>
<tr>
<td>Wet chip-PP</td>
<td>FR1</td>
<td>11.9</td>
<td>0.44</td>
<td>6.2</td>
</tr>
<tr>
<td></td>
<td>FR2</td>
<td>11.0</td>
<td>0.50</td>
<td>8.8</td>
</tr>
<tr>
<td></td>
<td>FR3</td>
<td>42.1</td>
<td>0.35</td>
<td>12.2</td>
</tr>
<tr>
<td></td>
<td>FR4</td>
<td>34.9</td>
<td>0.10</td>
<td>4.8</td>
</tr>
</tbody>
</table>

Figure 9. Aspect ratios of the fractionated wood particle samples

3.1.3 Microscopy

Microscopic images of the whole wood particle samples as well as particle fractions FR1, FR2, FR3 and FR4 of each sample are presented in Fig. 10. The microscopic images show that in each of the four cases wood chips have been significantly broken down during extrusion compounding. Additionally, it can be seen from Fig. 10 that there are obvious differences in the particle shape and size between samples. From Fig. 10 it can be seen that when dry chips were used as raw material, the particle size was greatly reduced, but wood has remained in particulate form. The shape of wood particles extracted from composites in which wet wood chips were used as raw material differ greatly from samples in which dry chips were used (Fig. 10). Unlike samples “PP-Dry chip” and “Dry chip-PP”, individual fibres can be also seen in the microscopic images of samples in which wet wood chips have been used as raw material (Fig. 10).

Microscopic observations confirm the results obtained from fractionation and image analysis. Processing method I, where PP was fed before wood chips, appeared to be gentler on wood chips because, after extrusion, the wood particle lengths and aspect ratios were higher in comparison with samples for which method II was used. The use of wet wood chips as raw material resulted in wood particles and fibres with high aspect ratio.

Figure 10. Microscopic images of the whole samples (images on top row) and fractions FR1, FR2, FR3 and FR4 of each sample.
3.2 Characterization of composites

3.2.1 Mechanical properties

Flexural properties of the manufactured WC-PP composites were compared with each other. Additionally, the flexural properties of composite materials with wet wood chips and without coupling agent were measured (PP-Wet chip* and Wet chip-PP*) to find out whether the use of coupling agent had any effect on the properties of wet wood chip composites. Flexural properties of the manufactured composite materials are presented in Table 6.

From Table 6 it can be seen that the moisture content of wood chips had the greatest influence on the flexural properties of the manufactured wood chip-PP composites and the properties were not affected significantly by the processing method used in compounding. The results show that the flexural modulus was highest with the composites in which dry wood chips were used as raw materials. Composites with wet wood chips had a somewhat lower wood content compared to composites with dry wood chips (Table 2), which might partly explain the results. From Table 6 it can be seen that the strength properties of all the composites with MAPP were quite similar, though the properties of “Wet chip-PP” were slightly lower. Addition of MAPP enhanced flexural modulus and strength properties of the wet wood chip composites.

In general, the mechanical properties of composites manufactured with wet wood chips were lower or similar when compared to composites with dry wood chips, despite the higher aspect ratio of wood particles. It is possible that moisture was not removed from the wood chips during the extrusion, which then decreased the mechanical properties of these materials. It is also possible that the coupling agent used was affected by the moisture in the wood chips.

**Table 6. Flexural properties of composites**

<table>
<thead>
<tr>
<th>Material</th>
<th>Modulus (GPa)</th>
<th>Strength (MPa)</th>
<th>Strain (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PP-Dry chip</td>
<td>4.6 ± 0.2</td>
<td>40.5 ± 3.1</td>
<td>1.7 ± 0.4</td>
</tr>
<tr>
<td>PP-Wet chip</td>
<td>3.8 ± 0.3</td>
<td>43.6 ± 4.5</td>
<td>2.2 ± 0.5</td>
</tr>
<tr>
<td>Dry chip-PP</td>
<td>4.4 ± 0.4</td>
<td>39.1 ± 7.0</td>
<td>1.6 ± 0.6</td>
</tr>
<tr>
<td>Wet chip-PP</td>
<td>3.6 ± 0.4</td>
<td>36.2 ± 4.1</td>
<td>2.0 ± 0.5</td>
</tr>
<tr>
<td>PP-Wet chip*</td>
<td>3.1 ± 0.2</td>
<td>28.7 ± 6.0</td>
<td>2.5 ± 0.7</td>
</tr>
<tr>
<td>Wet chip-PP*</td>
<td>3.2 ± 0.4</td>
<td>30.6 ± 4.4</td>
<td>2.4 ± 0.4</td>
</tr>
</tbody>
</table>

* Composites without coupling agent, MAPP.

3.2.2 Electron microscopy

FESEM images of the fractured surfaces of the composites are shown in Figures 11 and 12. It can be seen from Fig. 11 that when dry wood chips are used as raw material the wood particles are larger in comparison with wet wood chip composites in which wood is in a more fibrous form.

![Figure 11. Overviews of the fractured surfaces of wood chip-PP composites: (a) PP-Dry chip, (b) PP-Wet chip, (c) Dry chip-PP and (d) Wet chip-PP.](image)

Fig. 11a and Fig. 11b show the fractured surfaces of composites manufactured with method I. Details of the fractured surfaces of these samples can be seen in Fig. 12a and 12b. When dry wood chips are used as raw material, a lot of fibre pull-outs can be seen. The
composite manufactured with wet wood chips has less fibre pull-outs in comparison with the dry wood chip composite. Also, fractured fibres can be seen, indicating good adhesion in “PP-Wet chip” (Fig. 11b and Fig. 12b).

Fig. 11c shows the fractured surface of composite manufactured with method II and dry wood chips. Large wood particles can be seen in Fig. 11c, similar to method I and dry wood chips, but there seems to be fewer fibre pull-outs in comparison with “PP-Dry chip”. Furthermore, fractured wood particles can be seen in Fig. 12c. The fractured surface of composite manufactured with method II and with wet wood chips is shown in Fig. 11d. When compared with wet wood chip composite manufactured with method I (PP-Wet chip) more fibre pull-outs and cleaner fibre surfaces can be seen, implying that the adhesion between wood and matrix is not as good as with method I. The lower strength properties (Table 6) also implied that the adhesion in the composite manufactured with method II and with wet wood chips (Wet chip-PP) was not as good as with the composite manufactured with method I and wet wood chips. It was thought that the reason for poorer adhesion was the remaining moisture in the composites with wet wood chips. However, presence of the porosity, caused by the moisture, was not seen (Fig. 11 and Fig.12). It is possible that the moisture affected the coupling agent used and, therefore, the mechanical properties of the composites were lower.

4 Conclusions

When wood chips were used as raw material in wood plastic composites, both the processing method and the wood chips moisture content had effect on the wood particle size and aspect ratio. Individual fibres and wood particles with high aspect ratio could be separated from the wood particles when wet wood chips were used as raw material. When dry wood chips were used as raw material, the particle size was significantly reduced, but wood remained in particulate form.

The processing methods used in this study did not have a great effect on the mechanical properties of the composites. Composites manufactured with dry wood chips had slightly better flexural properties compared to composites manufactured with wet wood chips, despite the fact that the aspect ratio of wood particles in wet wood chip composites was higher.

Acknowledgments

The financial support of Franz och Carl Kempes Minnestiftelse, Sweden, is gratefully acknowledged. The authors wish to thank Borealis Polylefins, Austria, for supplying polypropylene and UPM Kymmene, Finland for supplying wood chips used in this study.

References


Paper III
Processing of wood chip–plastic composites: effect on wood particle size, microstructure and mechanical properties

M. Hietala1,2, J. Niinimäki2 and K. Oksman*1

Wood chips were used as raw material in extrusion of wood–plastic composites. Wood–plastic composites with ~50 wt-% wood content were manufactured by using two different compounding methods. Dried and undried wood chips were used to investigate the effect of wood moisture content on the wood particle size and whether the drying process could be carried out in the same step. Wood particle properties were measured using optical fibre analysis. Microscopical methods were used to examine the microstructure of wood particles. Furthermore, the prepared composites’ mechanical properties were studied. The particle size of wood chips was significantly reduced during extrusion in both processing methods. The undried wood chips had higher aspect ratios in comparison with the dried wood chips after extrusion. Despite the higher aspect ratio, the mechanical properties of composites manufactured with undried wood chips were not better than the properties of composites with dried wood chips.

Keywords: Wood–plastic composites, Extrusion, Wood particle size, Mechanical properties

This paper is part of a special issue on Manufacturing and Design of Composites

Introduction

Wood–plastic composites (WPCs) are materials which have a wood-like appearance but can be processed like plastic materials. When compared to solid wood, WPCs have better durability and lower maintenance requirements. Wood is also considered a low cost and renewable material in comparison with inorganic and synthetic fillers traditionally used in polymer composites. Currently the biggest market for WPCs is outdoor building materials, but automotive parts and furniture are also made from WPCs.1

Wood–plastic composites are manufactured by compounding wood particles with thermoplastic matrix. The most typical processing methods used to produce WPCs are extrusion compounding followed by profile extrusion or injection moulding. The matrix polymer is usually a low cost polymer with a processing temperature below the wood degradation temperature. The most common matrix polymers in WPCs are polyethylene, polypropylene (PP) and polyvinyl chloride.2

The most widely used wood derived raw material in commercially manufactured WPCs is wood flour. Wood flour is produced from the scrap materials of various wood processors by size reduction with different kinds of mills and size classification of the pulsed wood by screening.3 The particle size of commercially manufactured wood flour is usually smaller than 425 μm, and the aspect ratio (length/diameter ratio) is between 1 and 5.4 The low aspect ratio limits the reinforcing potential of wood flour,5 and studies have shown that wood fibres with a higher aspect ratio have better reinforcing capability.6,7 Especially when good adhesion between fibre and matrix exists, fibres provide better stress transfer to the fibres.5 However, fibres are seldom used in manufacturing of commercial WPCs because feeding and metering of low bulk density fibres is difficult and they are more expensive than wood flour.2,5 Problems with poor dispersion of wood fibres in composites have also been reported.8,9

A great deal of WPC research has concentrated on enhancing the interfacial adhesion between hydrophilic wood and hydrophobic thermoplastic matrix due to the incompatibility problems of these components. Particularly, the use of different coupling agents and fibre surface treatments has been studied extensively.10–15 Research activities focusing on the effect of wood characteristics and processing conditions on WPC properties have been less frequently reported. The effect of wood particle size on composite properties has been studied to some extent,7,16–19 but only a few studies can be found where larger wood particles have been used in the manufacture of WPCs.17,20 In addition, there have been few studies on the effect of extrusion compounding on wood particle size and fibre length,21,22 even though twin screw extrusion is one of the main processing methods used in the manufacture of WPCs.

In the study by Bledzki and Faruk17 the physical and mechanical properties of composites made of PP and
different types of wood fibres and wood chips were compared. They concluded that wood chips–PP composites showed better tensile and flexural properties than the other wood–PP composites when 5% coupling agent was added. Balasuriya et al. used wood flakes with a size of (1–4) mm in wood–high density polyethylene composites in their study of the effect of wood flake distribution and wetting on the structure–property relationship of WPCs. They noticed that the length of wood flakes was reduced after compounding. Other studies have also shown that wood particle size and fibre lengths are reduced during the compounding. Usually, this reduction is considered a disadvantage, but the shear forces in extrusion could also be used to reduce the particle size of larger wood particles. It is possible that individual fibres with a higher aspect ratio can be separated from wood particles if an optimal screw configuration and processing method is found. Thus, the wood raw material would not need as much preprocessing as the commonly used wood flour requires before it can be compounded with plastic in an extruder. If larger, undried wood particles can be used as raw material in a WPC, the use of cheaper wood residues in composite manufacturing is possible.

In this study the use of wood chips as a raw material in a WPC is investigated. The high shear forces in a co-rotating twin screw extruder are utilised to grind wood chips into smaller particles, possibly even into individual fibres, which are then compounded with polymer matrix in the same processing step. Two different extrusion processing methods are used, and both dried and undried wood chips are used to study the possibility of carrying out the drying process of wood in the same process step. In addition, the effect of wood moisture content on wood particle size during extrusion is examined. Wood particle properties such as aspect ratio and size distributions after extrusion are analysed. Furthermore, the mechanical properties of the prepared composites are studied.

Experimental

Materials

Spruce pin chips were used as wood raw material (Fig. 1). Wood chip particle size was determined by measuring the length and the width of 100 particles manually. According to manual measurements, average particle length was 16.2 mm and average particle width was 3.3 mm. Initial moisture content of the wood chips was 40 wt-% based on total weight. Wood chips were used as dried and as undried. The dried wood chips were kept at 105 °C for 24 h to reach moisture content below 1 wt-% (based on dry weight) before processing. The undried wood chips were used as such.

Polypropylene in powder form (melt flow index = 32 g/10 min, 230 °C/2.16 kg and 16 g/10 min, 200 °C/2.16 kg) supplied by Borealis Polyolefins (Austria) was used as matrix polymer. Maleic anhydride grafted PP (MAPP, Epoleine E-43; Eastman Chemical Company, USA), lubricant (TPW 113; Struktrol, USA) and antioxidant agent were used as additives.

Processing of WPCs

Extrusion compounding

A co-rotating twin screw extruder with twin screw side feeder, Coperion W&P ZSK-18 MEGALab (Stuttgart, Germany) equipped with a K-Tron gravimetric feeder (Niederlenz, Switzerland), two atmospheric vents and a vacuum vent was used to compound the composite materials. Before extrusion the PP and the additives were premixed. The gravimetric feeder was used to feed PP into the extruder while wood chips were fed into the extruder manually because it was not possible to use the gravimetric feeder system for this kind of material. Two different processing methods (methods I and II) were used in the manufacturing of the composites. The methods differed from each other in feeding orders of the raw materials, in screw configurations and in temperature profiles. In method I, PP and additives were fed first and wood chips were fed using the side feeder. In method II, wood chips were fed first and PP and additives were fed using the side feeder. Extrusion set-ups for methods I and II are shown in Figs. 2 and 3. The screw configuration used in method I (PP fed first) is presented in Fig. 4, and the screw configuration used in method II (wood chips fed first) is presented in Fig. 5.
The screw configurations were based on the feeding order of the materials.

The compounded composites were profiled into rectangular profile with a cross-section of 5 x 20 mm. Altogether four types of WPCs were manufactured using two different processing methods and two kinds of wood chips:

(i) PP–DWC: composite made with method I and dried wood chips
(ii) PP–UWC: composite made with method I and undried wood chips
(iii) DWC–PP: composite made with method II and dried wood chips
(iv) UWC–PP: composite made with method II and undried wood chips.

Owing to the manual feeding of the wood chips, the wood content of composites was calculated according to the feeding rate of PP (kg h\(^{-1}\)) and sampling time used to collect the composite material. The initial goal was to prepare composites with a wood content of 50 wt-%; however, the actual wood contents varied to some extent.

Characterisation of wood particles

Extraction of matrix polymer

To measure wood particle size and shape after the compounding, PP matrix was extracted from composites. Before extraction, extruded pieces of each composite material were pressed into thin sheets with heated press (LPC-300; Fontjine Grotnes, Netherlands) by first preheating the material for 2 min and then pressing for 30 s at 200 °C. Five grams of each pressed material was kept in boiling xylene until the matrix was completely dissolved (15–35 h). The solution was filtered and washed with hot xylene. After this, the sample was washed with ethanol to remove xylene and filtered. At last, the sample was washed with water to remove ethanol and filtered again.

Fractionation

To achieve better understanding of particle size distributions in each sample, the extracted samples consisting of wood particles and fibres were divided into four different size categories (fractions) by using the tube flow fractionation method (Metso Automation, Kajaani, Finland). After fractionation, mass fractions of each fraction were determined by filtering.

Tube flow fractionation is a method in which the sample (in water suspension) is injected into a continuous plug flow of water, which then enters a long plastic tube. As the sample flows in the tube it is fractionated by size. The fractionation device is equipped with a CCD camera, which records approximately 600 images of the sample during the analysis. The images recorded of each sample were saved and used in the image analysis. The tube flow fractionation method is described in more detail in publications by Laitinen et al. and Krogerus et al.\(^{25,26}\)

Image analysis

To measure wood particle and fibre dimensions, the images taken by the tube flow fractionation device were analysed using image analysis software developed by Metso Automation for research purposes. During image analysis, captured images are segmented to regions that may have one or several objects. The objects (particles,
Based on gas displacement technique. The average specimen size was $30 \times 10 \times 5 \, \text{mm}$. Three replicates for each material were tested.

**Mechanical properties**

Flexural testing of the wood chip–PP composites was performed according to ASTM D790 standard (three-point bending). A Shimadzu Autograph AG-X universal testing machine (Shimadzu Corp., Kyoto, Japan) with a load cell of 1 kN was used for testing. A minimum of five specimens of each material were tested.

**Microstructure**

Fractured surfaces of composites were studied with a Jeol JSM-6460 SEM (Jeol Ltd, Tokyo, Japan). The specimen from flexural testing were first frozen using liquid nitrogen and then broken to create the fractured surface. The sample surfaces were sputter coated with gold before observation. Acceleration voltage of 15 kV was used in the observation.

**Results and discussion**

**Processing of WPCs**

Table 2 shows the material compositions of the manufactured composites. Owing to the manual feeding of the wood chips, there was some variation in the wood contents between the composite materials. The composites with dried wood chips had a higher wood content compared to those with undried wood.

The processability of the composites with dried wood chips was better than composites with undried due to the fact that the moisture present in the wood chips was vaporised during extrusion. During processing the actual processing temperatures increased slightly from the set temperatures in the mixing zones where the kneading elements were located, due to the high shear forces. This was observed especially when method II (wood fed first) and dried wood chips were used.

The extruded profiles of the manufactured composites are shown in Fig. 6. The composites manufactured with processing method I had a more rough surface in comparison with those manufactured with method II, as can be seen in Fig. 6. The composites with dried wood chips (PP-DWC and DWC-PP) had a typical WPC colour, while those with undried wood chips (PP-UWC and UWC-PP) were lighter (Fig. 6).

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with the samples with dried wood chips. The wood particles extracted from composite specimens manufactured with processing method I and undried wood chips (PP-UWC) had the greatest aspect ratios, as shown in Table 3.

**Microscopy**

Microscopic images of the whole wood particle samples as well as fractions FR1, FR2, FR3 and FR4 of each sample are presented in Fig. 8. The images show that in each of the four cases wood chips have been significantly broken down during extrusion compounding. There are also clear differences in the particle shape and size between samples. Especially the shape of wood particles extracted from composites made with undried wood chips differs greatly from that of samples extracted from composites in which dried wood chips were used. With dried wood chips, the particle size is greatly reduced, but wood has remained in particulate form. The particles are more fibrous and also individual fibres can be seen in the samples extracted from composites in which undried wood chips were used as raw material.

**Image analysis**

Results from image analysis of fractionated samples are shown in Table 3. It can be seen from the results that when undried wood chips have been used as raw material the samples contain longer particles in comparison to samples with dried wood chips. Processing method I (PP fed first) appeared to be gentler on wood chips because, after extrusion, the wood particle lengths and aspect ratios were higher in comparison with samples in which method II was used. The use of undried wood chips as raw material resulted in wood particles and fibres with highest aspect ratio.

**Characterisation of composites**

**Density**

The densities of the wood–chip composites are presented in Fig. 9. There were small differences between all the densities of the composites, but especially the density of the composite manufactured with method I and with undried wood chips (PP-UWC) was lower than others. The difference is possibly due to the lower wood content in the composites with undried wood chips. However, porosity may also cause the lower density of the composite.

**Mechanical properties**

The flexural properties of the manufactured composites are presented in Figs. 10 and 11. The results from flexural testing were normalised to respond 50 wt-% of

<table>
<thead>
<tr>
<th>Material</th>
<th>Fraction</th>
<th>Mass-%</th>
<th>Average length, mm</th>
<th>Average width, μm</th>
<th>Aspect ratio (length/diameter)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PP-DWC</td>
<td>FR1</td>
<td>15.6</td>
<td>0.59</td>
<td>167.4</td>
<td>3.5</td>
</tr>
<tr>
<td></td>
<td>FR2</td>
<td>18.1</td>
<td>0.64</td>
<td>74.7</td>
<td>8.5</td>
</tr>
<tr>
<td></td>
<td>FR3</td>
<td>32.7</td>
<td>0.22</td>
<td>42.3</td>
<td>5.3</td>
</tr>
<tr>
<td></td>
<td>FR4</td>
<td>33.7</td>
<td>0.08</td>
<td>17.9</td>
<td>4.4</td>
</tr>
<tr>
<td>PP-UWC</td>
<td>FR1</td>
<td>55.1</td>
<td>0.15</td>
<td>57.6</td>
<td>19.9</td>
</tr>
<tr>
<td></td>
<td>FR2</td>
<td>11.6</td>
<td>1.12</td>
<td>32.8</td>
<td>34.3</td>
</tr>
<tr>
<td></td>
<td>FR3</td>
<td>11.4</td>
<td>0.08</td>
<td>21.1</td>
<td>27.9</td>
</tr>
<tr>
<td></td>
<td>FR4</td>
<td>11.4</td>
<td>0.08</td>
<td>13.7</td>
<td>5.6</td>
</tr>
<tr>
<td>DWC-PP</td>
<td>FR1</td>
<td>11.8</td>
<td>0.48</td>
<td>100.6</td>
<td>4.8</td>
</tr>
<tr>
<td></td>
<td>FR2</td>
<td>11.9</td>
<td>0.44</td>
<td>71.0</td>
<td>6.2</td>
</tr>
<tr>
<td></td>
<td>FR3</td>
<td>11.0</td>
<td>0.50</td>
<td>56.3</td>
<td>8.8</td>
</tr>
<tr>
<td></td>
<td>FR4</td>
<td>34.9</td>
<td>0.10</td>
<td>20.3</td>
<td>4.8</td>
</tr>
<tr>
<td>UWC-PP</td>
<td>FR1</td>
<td>11.9</td>
<td>0.44</td>
<td>71.0</td>
<td>6.2</td>
</tr>
<tr>
<td></td>
<td>FR2</td>
<td>11.0</td>
<td>0.50</td>
<td>56.3</td>
<td>8.8</td>
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<tr>
<td></td>
<td>FR3</td>
<td>34.9</td>
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<td>20.3</td>
<td>4.8</td>
</tr>
</tbody>
</table>

*PP: polypropylene; DWC: dried wood chips; UWC: undried wood chips.
The results from the flexural testing show that the addition of the coupling agent and the moisture content of wood chips had the greatest influence on the composites flexural properties and that the processing method did not affect the properties significantly.

Figure 10 shows a trend that the composites with dried wood chips (PP–DWC and DWC–PP) had the highest flexural modulus being around 4 GPa while the composites with undried wood chips (PP–UWC and UWC–PP) had a bending stiffness around 3–5 GPa. The manufactured composites with dried wood chips had similar bending stiffness, to that reported earlier by...
Oksman and Sanadi for wood polymer composites with 50 wt-% wood flour. The composites maximum strength values when the coupling agent was added were between 35 and 41 MPa, with no significant differences as can be seen in Fig. 11. However, the composites with undried wood chips manufactured with method I (PP–UWC) showed a trend with higher strength if compared to the others. The composites with no coupling agent showed lower properties and also very high scattering in the data. The composites with wood chips as raw material showed lower flexural strength values when compared to earlier reported values with wood flour when the strength was reported to be 59 MPa. The reason to test the properties without the coupling agent was because it was suspected that the moisture in the undried wood chips could react with MAPP, and therefore affect the composites properties negatively (Figs. 10 and 11). However, the results showed that the addition of MAPP had a positive effect on both flexural modulus and strength (PP–UWC and UWC–PP) in comparison with the composites without coupling agent (PP–UWC* and UWC–PP*).

In general, it can be concluded that the mechanical properties of composites manufactured with undried wood chips were lower or similar when compared to composites manufactured with dried wood chips, despite the higher aspect ratio of wood particles. It is possible that bound moisture was not removed from the wood chips during the extrusion process, which then act a plasticiser for the wood and decreased the stiffness of the composites.

**Microstructure**

The SEM images from the fractured surfaces of the composites are shown in Fig. 12. The wood particles are significantly larger in the composites with dried wood chips (Fig. 12a and c) in comparison with undried wood chips (Fig. 12b and d). Wood particles are also clearly fibrous in the composites made with undried wood chips. Even long individual fibres can be seen in the fractured surfaces of the composite made with method II and undried wood chips (Fig. 12d).

However, cavities from fibre pull-outs and clean fibre surfaces can be seen in all of the composites indicating that the adhesion between the matrix and the wood particles is not very good in any of these composites. The composites with dried wood chips (PP–DWC and DWC–PP) have large cavities from fibre pull-outs and the composites with undried wood chips have plenty of smaller fibre pull-outs, as the fractured surfaces of the composites show (Fig. 12).

The results from flexural tests showed a trend that the composites stiffness was lower when undried wood chips...
were used which was unexpected when seeing the micrographs of these materials. The undried wood chips with larger aspect ratios should result in higher bending stiffness than composites with particular wood. One possible reason for this behaviour could be the remaining moisture in the wood. The moisture can cause porosity in the matrix polymer and it can also act as a plasticiser for the wood. However, when the fractured surfaces of the composites were studied, no obvious signs of porosity in the matrix could be seen (Fig. 12b and d). Therefore, the reason the composites did not reach better stiffness might be that the moisture was not efficiently removed during processing and that softened to wood fibres.

Conclusions

When wood chips were used as raw material in WPCs, both the processing method and the wood chips moisture content affected the wood particle size and the aspect ratio. In the case of dried wood chips, the particle size was significantly reduced, but wood remained in particulate form. Individual fibres and wood particles with high aspect ratios were separated during the compounding process when undried wood chips were used as raw material.

The used processing methods did not have a great effect on the mechanical properties of the composites. Composites manufactured with dried wood chips had slightly better flexural properties compared to those manufactured with undried wood chips, despite the fact that the aspect ratio of wood particles in undried wood chip composites was higher.

This work shows that it is possible to use undried wood chips as raw material for WPCs and reach stiffness values similar to the composites where wood flour is used as raw material and by that decrease the raw material cost for the final product. Furthermore, if the bound moisture from wood chips can be removed during the process it might be possible to improve the mechanical properties because the wood is in the form of fibre. Fibres have larger aspect ratios than wood particles and should therefore be more efficient as reinforcement for polymers. It might be possible to remove the moisture by increasing the residence time in the extruder by using lower processing speeds, a larger extruder or an extruder with a higher length/diameter ratio.

Acknowledgement

The authors wish to thank Borealis Polyolefins, Austria, for supplying polypropylene.

References

Paper IV
The effect of pre-softened wood chips on wood fibre aspect ratio and mechanical properties of wood-polymer composites

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b) Laboratory of Fibre and Particle Engineering, University of Oulu, Finland
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Abstract

The objective of this work was to study the effect of chemical pre-treatment and moisture content of wood chips on the wood particle aspect ratio after compounding in a twin-screw extruder and on the mechanical properties of wood-polymer composites (WPC). Composites with 50 wt% wood content were manufactured using pre-treated and untreated wood chips. The effect of wood moisture content on composite properties was studied by using dried and undried wood chips. The mechanical properties and fractured surfaces of the composites as well as the microstructure and aspect ratio of wood particles after compounding were studied. The highest wood particle aspect ratio after extrusion was achieved by using pre-treated, undried wood chips as raw material. The chemical pre-treatment was found to enhance the fibrillation of wood chips as well as mechanical properties of the composites.

Keywords: Wood, Polymer-matrix composites, Mechanical properties, Extrusion.
1. Introduction

Wood-polymer composites (WPC) are materials which combine the properties of wood and thermoplastic polymers. They are considered as more environmentally friendly options for wood and plastic products in several applications, such as outdoor building materials, furniture and interior panels in automotives [1]. WPCs are manufactured by compounding wood raw material with a molten matrix polymer together with additives such as lubricants, antioxidants and coupling agents in various compositions. In commercial manufacturing of wood-polymer composites the main wood raw material used is wood flour. Wood flour is typically produced from the residues of wood processors by milling and screening the material into specific particle size range [2]. Usually the aspect ratio (length-to-diameter ratio) of wood flour particles is low, between 1 and 5 [2]. Earlier studies have shown that wood and other natural fibres with higher aspect ratio give better reinforcement than wood particles with low aspect ratio [3, 4]. Especially when there is a good adhesion between fibres and the matrix, a better stress transfer to the fibres is provided [2]. However, since the processing of longer, bulky fibres is more difficult and fibres are more expensive than wood flour, wood fibres are not often used in the manufacture of wood-polymer composites [5].

The focus in wood-polymer composite research has strongly been on the improvement of the interfacial adhesion between the hydrophilic wood and hydrophobic polymer matrix. Studies have concentrated especially on various coupling agents and chemical treatments to improve adhesion between these two components [6-10]. The amount of research focused on studying the effects of wood particle properties on composite properties is much less, and these studies are mainly about how the wood particle size and wood species affect composite properties [11-14]. Only a few studies can be found where the effect of the aspect ratio of wood fillers has been studied [15-17], even though it has been suggested that the aspect ratio and not the particle size of wood filler has
the main influence on the mechanical properties of the wood-polymer composites [4]. The possibility to use larger wood particles as raw material for WPC has been studied only by few [18, 19], though it would be an economical benefit if wood residues could be used directly in the compounding process of wood-polymer composites without pre-processing steps such as milling, sieving and drying. In our earlier study it was shown that wood chips can be used directly as raw material for WPC, and it is even possible to separate individual fibres from the wood chips during the extrusion compounding if undried wood chips are used as raw material [20].

The objective of this work was to investigate the effect of chemical pre-treatment and moisture content of wood chips on the wood particle aspect ratio after compounding in a twin-screw extruder and on the mechanical properties of prepared composites. The purpose of the chemical pre-treatment was to soften the wood raw material prior extrusion. In the earlier study by Hietala et al. 2011 a decrease in the flexural properties was noticed when undried wood chips were used as raw material, even though the aspect ratio of wood particles was increased [20]. The high moisture content (40 wt%) of the wood raw material feed was believed to be the reason for this, and therefore in this work the effect of lower moisture content of the undried wood raw material was also studied.

2. Experimental

2.1. Materials

Spruce chips were used as wood raw material (Fig 1.). Wood chips were used as: 1) untreated and undried (UU), 2) untreated and dried (UD), 3) pre-treated and undried (PU), and 4) pre-treated and dried (PD). The moisture content of the undried wood chips was 33 wt% based on total weight, and the moisture content of the dried wood chips was below 1 wt% based on the dry weight. The drying of the wood chips was done in an oven at 105°C for 24 h.
Fig. 1. Spruce wood chips.

The matrix polymer used was polypropylene, PP, in powder form (MFI = 32 g/10 min, 230°C/2.16 kg and 16 g/10 min, 200°C/2.16 kg) supplied by Borealis Polyolefins, Schwechat, Austria. Maleic anhydride grafted polypropylene, MAPP (Epolene E-43, Eastman Chemical Company, Longview, Texas, USA), lubricant (TPW 113, Struktol, Stow, Ohio, USA) and antioxidant agent were used as additives.

2.2. Wood chip pre-treatment

Sodium sulphite, Na₂SO₃ (Merck KGaA, Darmstadt, Germany), was used in the chemical pre-treatment. The wood raw material was diluted into 10% consistency and 5% of Na₂SO₃ (based on the total mass of the mixture) was used in the treatment. After the pre-treatment, wood chips were washed with water and they were left to dry in room temperature until moisture content of 33 wt% was reached.

2.3. Compounding process

A co-rotating twin-screw extruder with twin-screw side feeder, ZSK-18 MEGALab (Coperion W&P, Stuttgart, Germany) equipped with a K-Tron gravimetric feeder (Niederlenz, Switzerland), two atmospheric vents and a vacuum vent was used to compound the composite materials. Polypropylene and the additives were pre-mixed prior extrusion. The gravimetric feeder was used to
feed PP into the extruder while wood chips were fed into the extruder manually because it was not possible to use the gravimetric feeder system for this kind of material. The wood content of composites was calculated according to the feeding rate of PP (kg/h) and sampling time used to collect the composite material. The aim was to prepare composites with a wood content of 50 wt%, but the actual wood contents varied between 45 wt% and 54 wt%.

Fig. 2. The extrusion set-up and the screw configuration used in the study.

The extrusion set-up and the screw configuration used in the compounding of the composites are shown in Fig. 2. The extrusion conditions were the same for all prepared composites. The screw configuration used was chosen based on the results reported on an earlier study [20]. The compounded composites were profiled into rectangular profile with a cross section of 5 mm x 20 mm. Four types of wood chip-PP composites were manufactured:
a) **UD-PP**: composite made with untreated and dried wood chips

b) **UU-PP**: composite made with untreated and undried wood chips

c) **PD-PP**: composite made with pre-treated and dried wood chips

d) **PU-PP**: composite made with pre-treated and undried wood chips

### 2.4. Characterisation

#### 2.4.1. Specimen preparation

Test specimens for flexural testing were moulded with a conventional compression moulding press (Fjellman Press AB, Mariestad, Sweden). Before moulding, the extruded profiles were heated in a hot air oven at 200°C for 20-45 minutes. The mould temperature was 50°C and the pressure applied on the samples was approximately 70 MPa. Plate-shaped test specimen for impact testing were made with a LPC-300 Fontijne Grotnes (Vlaardingen, Netherlands) heat press by first preheating the extruded material for 10 min without pressure in 200°C and then pressing for 2 min at 150 kN in 200°C. The average dimensions of the test plates were 1.6 mm × 10.0 mm × 10.0 mm.

#### 2.4.2. Mechanical properties

Flexural testing of the wood chip-PP composites was performed according to ASTM D790 standard (three-point bending). A Shimadzu Autograph AG-X universal testing machine (Shimadzu Corp, Kyoto, Japan) with a load cell of 1 kN was used for testing. A minimum of 13 specimens of each material were tested. Impact properties of the prepared composites were measured with a falling weight impact testing machine, Dynatup Minitower (Instron, Norwood, MA, USA) according to ASTM D3763 standard. The total mass of the impactor was 2.31 kg and speed of testing was 2.8 m/s. At least 3 specimens of each material were tested.
2.4.3. Density

Density of the compression moulded composite specimen was measured using water displacement method according to ASTM D792 standard (test method A). The specimen size was 65 mm × 10 mm × 5 mm. Four replicates for each material were tested.

2.4.4. Extraction of matrix polymer

To study the wood particle size and shape after compounding, PP matrix was extracted from composites. Prior to extraction, pieces of each composite material were pressed into thin sheets with a heat press (LPC-300, Fontijne Grotnes, Vlaardingen, Netherlands) by first preheating the material for 5 min without pressure in 200°C and then pressing for 2 min at 150 kN in 200°C. Few grams of each pressed material were kept in boiling xylene until the matrix was completely dissolved (24h). After extraction, the wood particles were washed with ethanol and water to remove xylene.

2.4.5. Aspect ratio of wood particles

For the determination of the aspect ratio of wood particles extracted from the composites, the samples were first analysed with a tube flow fractionator (Metso Automation, Kajaani, Finland). The tube flow fractionator is typically used to fractionate wood fibre samples into different size categories. However, in this case the device was used to capture images of the wood particle samples for the image analysis, because during analysis approximately 600 images of the sample are recorded. The tube flow fractionation method is described in more detail in publications by Krogerus et al. and Laitinen et al. [21, 22].

After fractionation the images were analysed using kajaaniIMG image analysis software supplied by Metso Automation (Kajaani, Finland). During image analysis, captured images are segmented to
regions that may have one or several objects. The objects (particles, fines and fibres) in one region are then interpreted based on geometrical criteria. Several image analysis algorithms are performed to measure the properties of each object. The basis for these algorithms is the model on interaction of light and various objects. This enables the use of so-called sub-pixel algorithms to improve the resolution. For each of the objects, the projection area is determined. Based of the shape of the object, the other parameters such as length and width are then decided. The aspect ratio of the wood particles was calculated using the length and width parameters. The settings used in the image analysis are presented in Table 1. The settings were chosen so, that the smallest wood particles would be left out from the analysis.

Table 1. Settings used in the image analysis.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>minimum fiber width</td>
<td>10 μm</td>
</tr>
<tr>
<td>maximum fiber width</td>
<td>3000 μm</td>
</tr>
<tr>
<td>minimum fiber length</td>
<td>0.1 mm</td>
</tr>
<tr>
<td>maximum fiber length</td>
<td>7.6 mm</td>
</tr>
</tbody>
</table>

2.4.6. Morphology and microstructure

Scanning electron microscope (Jeol JSM-6460, Jeol Ltd, Tokyo, Japan) was used to observe the size and shape of wood particles after extraction of the polymer matrix as well as the fractured surfaces of the manufactured composites. The wood particle samples were freeze-dried before microscopy. The fractured surfaces of the composites were created by freezing flexural testing specimen in liquid nitrogen and then breaking the frozen specimen. All the specimens were sputter-coated with gold before observation. Acceleration voltage of 10 kV was used in the observation of
the fractured surfaces and acceleration voltage of 5 kV was used in the observation of the wood particles.

3. Results and discussion

3.1. Composites

In Table 2 the compositions of the wood chip-PP composites are shown. The wood contents of the composites varied to some extent due to the manual feeding of the wood chips in the extruder. In general, the composites with dried wood chips had higher wood content compared to the composites with undried wood. The extruded profiles of the composites are presented in Fig. 3. From Fig. 3 it can be seen that there are colour differences between the composites and the wood particles are more visible in the composites made with dried wood chips (UD-PP, PD-PP) in comparison with the composites made with undried wood chips (UU-PP, PU-PP).

Table 2. Material compositions of the wood chip-PP composites.

<table>
<thead>
<tr>
<th>Material</th>
<th>Weight%</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>PP&lt;sup&gt;1&lt;/sup&gt;</td>
</tr>
<tr>
<td>UD-PP</td>
<td>41.4</td>
</tr>
<tr>
<td>UU-PP</td>
<td>44.1</td>
</tr>
<tr>
<td>PD-PP</td>
<td>45.0</td>
</tr>
<tr>
<td>PU-PP</td>
<td>49.5</td>
</tr>
</tbody>
</table>

<sup>1</sup>Polypropylene includes 0.18 wt% of antioxidant agent.
3.1.1. Mechanical properties

The results from the mechanical testing are shown in Table 3. Because there were differences in the wood contents of the composite materials, the results from flexural and impact testing were normalized to respond 50 wt% wood content.

<table>
<thead>
<tr>
<th>Material</th>
<th>Flexural properties</th>
<th>Impact properties</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Strength (MPa)</td>
<td>MOE (GPa)</td>
</tr>
<tr>
<td>UD-PP</td>
<td>43.6 ± 3.9</td>
<td>4.5 ± 0.3</td>
</tr>
<tr>
<td>UU-PP</td>
<td>48.6 ± 3.4</td>
<td>3.8 ± 0.3</td>
</tr>
<tr>
<td>PD-PP</td>
<td>50.5 ± 2.6</td>
<td>4.9 ± 0.5</td>
</tr>
<tr>
<td>PU-PP</td>
<td>57.7 ± 5.4</td>
<td>4.3 ± 0.5</td>
</tr>
</tbody>
</table>

PP = polypropylene, UD = untreated and dried wood chips, UU = untreated and undried wood chips, PD = pre-treated and dried wood chips, PU = pre-treated and undried wood chips.

From Table 3 it can be seen the chemical pre-treatment affected the flexural properties positively: the composites with pre-treated wood chips had better strength properties and higher flexural modulus (MOE) when compared with composites with untreated wood chips and the same wood chip moisture levels. The flexural properties on the composites were also affected by the moisture content of the wood chips: the use of undried wood chips with a moisture content of 33 wt%
enhanced the flexural strength but simultaneously the modulus of elasticity was decreased. The composite manufactured with pre-treated, undried wood chips (PU-PP) had the highest flexural strength and the composite with pre-treated, dried wood chips (PD-PP) had the highest flexural modulus among the prepared composites. The lower modulus of the composites with undried wood chips could be explained by the fact that moisture makes wood more flexible. From Table 3 it can be seen that the strains of the composites with undried wood chips were larger than with the composites with dried wood chips.

The impact properties of the composites were studied using a falling weight impact test, and the results are presented in Table 3. Characteristic load-deflection and energy-deflection curves from the impact testing are shown in Fig. 4. The shape of the curves was similar for all the materials. It can be seen from Table 3 that the peak load and total impact energy values were somewhat higher for the composites in which the undried wood chips were used (UU-PP, PU-PP). The use of pre-treated wood chips resulted in higher peak load and total impact energy values comparison with the composites with untreated wood chips.

Fig. 4. Characteristic load and energy vs. deflection - curves from impact testing.

The results from the mechanical testing showed that the chemical pre-treatment enhanced the mechanical properties of the composites. The flexural strength and modulus of the composites with
pre-treated wood chips were higher in comparison with the composites with untreated chips. Also the impact properties, peak load and total impact energy, were slightly higher for the composites with pre-treated wood chips. The enhancement of properties of the composites with pre-treated wood chips is believed to be caused mainly by the higher aspect ratio of wood fibres and particles due to lignin softening. Especially this is assumed when undried wood chips have been used. It is also possible that pre-treatment with sodium sulphite changed the surface properties of wood particles [23], and thus improved the mechanical properties of the composites by improving the adhesion between wood and polypropylene.

3.1.2. Density

The results from the density measurements are shown in Fig. 5. The results were normalized to respond 50 wt% wood contents. The composite with pre-treated, undried chips (PU-PP) had the highest density, 1.23 g/cm³, and the composite with untreated, dried chips had the lowest density, 1.03 g/cm³. According to Clemons 2008 [2], increased density of the wood-polymer composites is often related to the collapsing or filling of the hollow wood fibres. Therefore, the reason for the higher density of the PU-PP composite might be that there are more individual wood fibres which collapse or fill more easily in comparison with the larger wood particles in the other composites.

![Fig. 5. Densities of the wood chip-PP composites.](image)
3.1.3. Fractured surfaces

Figs. 6 and 7 show SEM images taken from the fractured surfaces of the wood chip-PP composites. The images in Fig. 6 are taken with a smallest magnification to show a general view of the fractured surface of the composite. From Fig. 6 it can be seen that the fractured surface of composites with dried wood chips (Fig. 6a and 6c) is rougher in comparison with composites with undried wood chips (Fig. 6b and 6d). Also large cavities from pull-outs and parts of large wood particles can be seen in the fractured surfaces of the composites with dried wood chips. The fractured surfaces of the composites with undried wood chips are smoother, most likely due to smaller wood particles and fibres in them.

Fig. 6. Overviews of the fractured surfaces of the wood chip-PP composites. (a) UD-PP, (b) UU-PP, (c) PD-PP and (d) PU-PP.
Fig. 7. Details from the fractured surfaces of the wood chip-PP composites. (a) UD-PP, (b) UU-PP, (c) PD-PP and (d) PU-PP.

Fig. 7 shows details from the fractured surfaces of the wood chip-PP composites. It can be seen that wood particles have not been separated into individual fibres when dried wood chips have been used as raw material (Fig. 7a and 7c), even though the particle size appears to be greatly reduced. The presence of individual fibres in the composites with undried wood chips is obvious (Fig. 7b and 7d). Also larger wood particles could be seen in the fractured surfaces of the composites with undried wood chips, but the amount was significantly lower than in the composites with dried wood chips. No obvious differences between the composites with untreated and pre-treated wood chips could be seen in the SEM images, and the adhesion appeared to be moderately good in all the composite materials. No large gaps between the matrix and the wood particles could be seen in the images; however fibre and particle pull-outs were be observed in all the composites.

3.2. Wood particles

3.2.1. Microstructure

Fig. 8 shows SEM micrographs of the wood particles after matrix removal. Individual fibres are clearly seen in the images which show the wood particles extracted from the composites manufactured with undried wood chips (Fig. 8b and 8d). In addition, more individual fibres can be
seen in Fig. 8d in comparison with Fig. 8b, indicating that the wood chip pre-treatment improved the fibre separation from the undried wood chips. When dried wood chips have been used as raw material, the wood has remained in particulate form, as can be seen from Fig. 8a and c. There are no apparent differences between the wood particles extracted from the composites made with dried, untreated wood chips (Fig. 8a) and dried, pre-treated wood chips (Fig. 8c).

![Fig. 8. Wood particles after matrix removal. (a) UD-PP, (b) UU-PP, (c) PD-PP and (d) PU-PP.](image)

3.2.2. Aspect ratio

The average aspect ratios (l/d) as well as the average lengths and widths of the wood particles extracted from the composite samples are shown in Table 4. The aspect ratios were calculated from the length and width measurements determined using image analysis software.

<table>
<thead>
<tr>
<th>Material</th>
<th>Length, L(l) mm</th>
<th>Width, μm</th>
<th>Aspect ratio, l/d</th>
</tr>
</thead>
<tbody>
<tr>
<td>UD-PP</td>
<td>0.3</td>
<td>46.3</td>
<td>7.9</td>
</tr>
<tr>
<td>UU-PP</td>
<td>0.6</td>
<td>34.1</td>
<td>23.1</td>
</tr>
<tr>
<td>PD-PP</td>
<td>0.3</td>
<td>38.3</td>
<td>8.9</td>
</tr>
<tr>
<td>PU-PP</td>
<td>0.7</td>
<td>35.7</td>
<td>25.3</td>
</tr>
</tbody>
</table>

*length weighted average length.
As it can be seen from Table 4, the wood particle samples from the composites in which dried wood chips were used (UD-PP, PD-PP) had similar aspect ratios as wood flour typically used in wood-polymer composites. The use of undried wood chips as raw material resulted in significantly greater wood particle aspect ratios after compounding. The wood chip pre-treatment slightly increased the wood particle aspect ratio, which can explain the improved mechanical properties of the composites with pre-treated wood chips. The wood particles with the highest aspect ratio were extracted from the composite made with pre-treated and undried wood chips (PU-PP), which also had the highest flexural strength and impact properties of the composites (Table 3).

4. Conclusions

In this work the effect of chemical pre-treatment and moisture content (dried and undried wood chips) in manufacture of wood chip-PP composites was studied with a aim to create more individual wood fibres during the processing as well as improve the composites mechanical properties. This study showed that chemical pre-treatment with sodium sulphite improved the composites mechanical properties and that the wood particle aspect ratio was increased in the extrusion process.

The use of pre-treated wood chips enhanced the flexural and impact properties of the wood chip-PP composites. The use of undried wood chips resulted in improved flexural strength and impact properties in comparison with the composites with dried wood chips, but the flexural modulus was decreased. Study of the aspect ratios of wood particles extracted from composites showed that the use of pre-treated wood chips resulted in higher aspect ratio after compounding.
The overall results are promising, the study showed that it is possible to use wood chips as raw material for the production wood-polymer composites and that the wood chips can be fibrillated during the composite manufacturing process. When a simple chemical pre-treatment was used to soften the wood a more fibrillated wood structure could be achieved.

5. Acknowledgements

Authors wish to thank Borealis Polymers for supplying the polypropylene used in the study.

6. References


