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EFFECTS OF PHYSICAL TREATMENT OF WOOD FIBRES ON FIBRE MORPHOLOGY AND BIOCOMPOSITE PROPERTIES

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Abstract

In the study, the effect of refining and fractionation of wood fibres on fibre morphology and biocomposite properties were determined. Kraft pine pulp and thermomechanical pulp were selected for the fibre treatments. The effects of physical treatment on fibre morphology were analysed with a fibre analyser and microscopy techniques. For the composites, polylactic acid was used as a polymer matrix. Composites were produced by melt processing to a fibre content of 30 w-%, and the mechanical properties of the injection-moulded biocomposites were investigated. In general, TMP fibres improved the mechanical properties of PLA more than pine pulp fibres did. Two different fibre fractions with distinct fibre properties were obtained by fractionation, and the use of a long fibre fraction provided improved mechanical properties for composites. The refining of pine pulp led to clear fibrillation of the fibre surface, but it had a negative effect on the mechanical properties of biocomposites.

Keywords: WPC; biocomposites; wood fibres; refining; fractionation.

Introduction

In conventional wood plastic composites (WPCs), sawdust or wood powder is usually used as a filler for polyolefins. For reinforced plastic applications, increasing interest in the use of wood fibres instead of wood powders can be detected.[1,2,3] To enable the full reinforcement capacity of wood-based fibres, problems such as insufficient fibre dispersion and adhesion to the matrix, inconsistent fibre uniformity and durability during processing have to be solved. To overcome these obstacles, options such as physical, chemical or enzymatic fibre treatments can be studied. In papermaking, two common physical treatments for wood fibres are fractionation and refining.

Fibre fractionation is a process in which pulp fibres are sorted based on their physical properties, and the resulting fibre fractions can have significantly different fibre properties. Today, there are two main industrial-scale devices used to fractionate pulp fibres: pressure screens and hydrocyclones. In pressure screens, the sorting of fibres is based on the dimensional properties (length and width) of the fibres, whereas in hydrocyclones it is mainly based on the density and

specific surface area (fibre surface area per gram) of the fibres.[4] Many other parameters than those related to fibre morphology also affect the efficiency of fractionation and the quality of fractions. General overviews of these methods can be found in the references.[5,6]

Refining of pulp is known to increase the strength properties of paper.[7] The bonding capacity of the fibres is improved by increasing the formability and the bonding surface area of the fibres. Refining is also known to affect fibres in many other ways. The most important effects are external fibrillation, fines production, length reduction and internal changes in the wall structure. Refined fibres are therefore more collapsed (flattened), fibrillated and flexible than unrefined fibres.[7] In addition, McIntosh and Uhrig[8] have found that refining does not greatly decrease the tensile strength of a single fibre.

This study looks at the effects of refining and dimensional-based fractionation on fibre morphology and the mechanical properties of WPCs. Refining increases the surface area of the fibres, which can lead to better fibre-matrix adhesion in composite applications. Fractioning, on the other hand, offers a great opportunity to affect fibre material uniformity and the possibility of using longer fibres with more reinforcing potential.

Experimental

Fibre treatments

Chemically bleached kraft pine pulp and thermomechanically ground (peroxide-bleached) spruce pulp (TMP) provided by UPM Kymmene were used in the experiments. Fractionation of the TMP pulp was performed using the Metso FS-03 laboratory screen-type sorter. After fractionation, thick A4 sheets were prepared and cut into chad by a paper-shredder device before compounding. Pine pulp was refined by the Escher Wyss laboratory refiner LR1. The fibres were pelletised with a planar matrix pelletising machine (Amandus-Kahl) before compounding.

Biocomposite processing

NatureWorks® polylactic acid (PLA) injection moulding grades 3001D and 3051D were used as matrix polymers of biocomposites. Physically treated fibres were compounded with PLA to a fibre content of 30 w-% using a co-rotating twin-screw extruder (Berstorff ZE 25x33D). The diameters and lengths of the screws were 25 mm and 870 mm respectively. To feed the materials, gravimetric side-feeders were used. A temperature gradient from 60°C in the feeding section to 190°C in the melting zones and the die, and a screw speed of 200 rpm were used. After processing, the compounds were injection moulded with an injection-moulding machine (Engel ES 200/50 HL) into ISO 3167 tensile test specimens with a length of 150 mm and a 10 mm wide by 4 mm thick by 80 mm long centre section. After injection moulding, the tensile test specimens were kept in a room with standard conditions (23°C, 50% relative humidity) for at least five days before testing.

Measurements

The fibre dimension measurements were performed using the L&W STFI FiberMaster device. The total amount of lignin in the TMP fractions was determined by the Klason lignin method. Light microscope imaging of fibres was performed with a Nikon Eclipse ME600 device. The CCD camera used was the PCO SensiCam LongExposure. Photos were taken at 50 or 100x magnification. Wet fibres were stained with methylene blue colour.

Laboratory paper sheets were prepared using the SCAN-C 26:76 standard, and the tensile index of paper was determined using the SCAN-P 38:80 standard. The refining level of pulp was analysed using the Canadian Standard Freeness (CSF) measurement (SCAN-C 21:6). The ISO-527 and ISO-179 standards were used in the mechanical testing of the composite specimens. To determine the fibre lengths of the injection-moulded specimens, the PLA matrix was dissolved with chloroform and the remaining fibres analysed with the L&W STFI FiberMaster device.

Results and discussion

Fibre morphology

Reference pulps

Untreated TMP and chemically bleached kraft pine pulp were selected as reference pulps, and their initial fibre properties were measured from pulp before any treatments.

Table 1 Fibre dimension measurements of TMP and kraft pine pulp

Measured properties	Unit	Initial TMP	Initial pine pulp
Fibre length*	mm	1.39	2.37
Fibre width*	µm	34	30
Aspect ratio	-	41	78
Fines	%	27	6.89
Lignin content	%	26	<0.20

*Length weighted value (measured using a FiberMaster device). The fibre properties were measured from the pulp.

Thermomechanical pulp and chemically bleached pine pulp differ in many ways (Table 1). The amount of lignin in TMP is high, whereas bleached pine pulp is almost lignin-free. TMP contains clearly more fines than kraft pine pulp, and TMP fibres are shorter and have a lower aspect ratio than pine pulp fibres. In literature, mechanical pulp fibres are described as stiffer, coarser and straighter than chemical pulp fibres.[9]

Fractionation

TMP pulp was fractionated by fibre dimensions into short and long fractions in nine sorting steps. The fibre properties were measured from rewetted chads and the results are shown in Table 2. Preparation of the sheets and shredding them into chad affected the fibre properties of the reference fibres slightly, and the results of the fibre measurements therefore differ from the initial fibre values (Table 1).

Table 2 Fibre dimension measurements and pulp properties of different fractions of TMP

Measured properties	Unit	Unfractioned TMP	Short fraction of TMP	Long fraction of TMP
Canadian standard freeness (CSF)	ml	80	<10	240
Fibre length*	mm	1.11	0.74	1.23
Fibre width*	µm	34	29	34
Aspect ratio	-	33	26	37
Fines	%	27	47	20
Lignin content	%	26	30	24
Tensile index of paper	Nm g-1	44	47	41

*Length weighted value (measured using a FiberMaster device). The fibre properties were measured from the rewetted shredded sheets before compounding.

Two fractions with different fibre dimensions were obtained by fractionation. The fibre length and width of the long fraction were clearly higher than of the short fraction, thus the aspect ratio of the long fraction was also higher. The short fraction also had a higher fines content. As is known from literature, fines have a large surface area, which improves bonding between fibres.[9] Consequently, the tensile strength of the paper from the short fraction was a bit higher than from the long fraction.

In addition to the effect on fibre morphology, the lignin content of the TMP fractions was also influenced by fractionation. The short fraction of TMP had a higher lignin content than the reference and the long fraction fibres because fine particles include more lignin than long fibres.[10]

Long fibres and a high aspect ratio of fibres are expected to lead to better mechanical properties of composites than short fibres because the composite strength and stiffness are directly proportional to the length and diameter of the reinforcing fibre.[11] The long fibre fraction produced with decreased fines content should therefore provide increased composite properties.

Refining

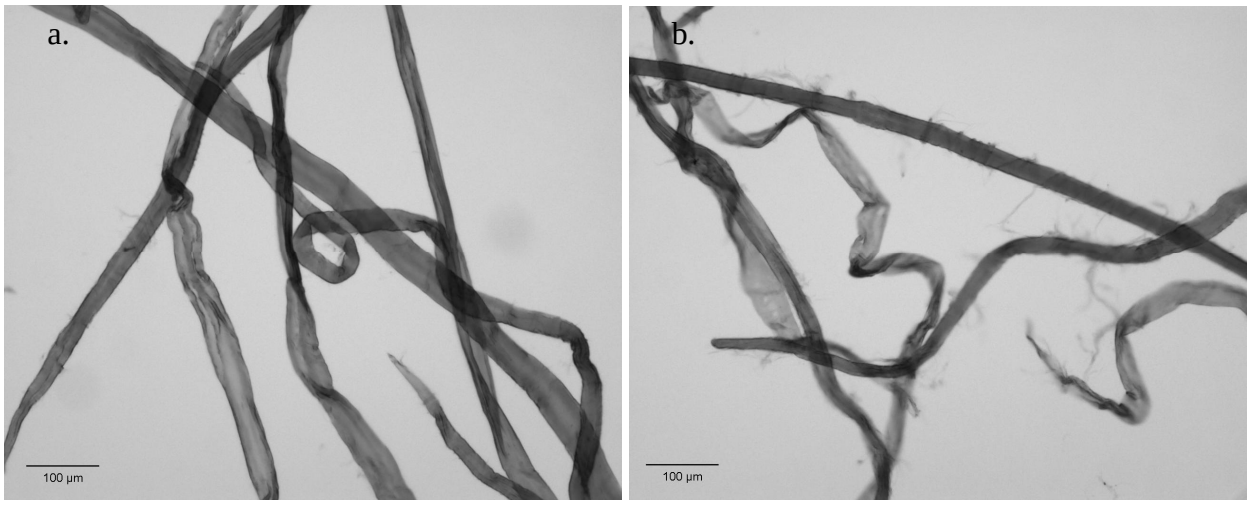
Chemically bleached pine pulp was refined at two levels with refining energies of 120 and 200 kWh t-1. The pelletising process influenced fibre properties. The measured values in Table 3 therefore differ from the initial values of the chemical pine fibres (Table 1).

Table 3 Fibre dimension measurements and pulp properties of unrefined and refined pine pulps

Measured properties	Unit	Unrefined pine pulp	Refining Level 1	Refining Level 2
Specific energy consumption (SEC)	kWh t-1	0	120	200
Canadian standard freeness (CSF)	ml	680	570	390
Fibre length*	mm	2.17	2.10	2.02
Fibre width*	µm	31	31	32
Aspect ratio	-	70	67	63
Fines	%	6.50	9.50	13
Tensile index of paper	Nm g-1	36	59	66

*Length weighted value (measured using a FiberMaster device). The fibre properties were measured from rewetted pellets.

Table 3 shows that reasonable refining of pine pulp fibres did not affect fibre dimensions significantly. The fines content of the pulp increased during refining and even doubled at a refining level of 200 kWh t-1. As the bonding capacity and fines content of the fibres were increased by refining the pulp, the tensile index of the laboratory sheets was increased radically.
The effects of refining on the fibre surface can be seen clearly from the light microscope images (Fig. 1 and Fig. 2). Fig. 1 was taken in the wet state from fibres of rewetted pellets and Fig. 2 in the dry state from the fibres pulled out from dry pellets.



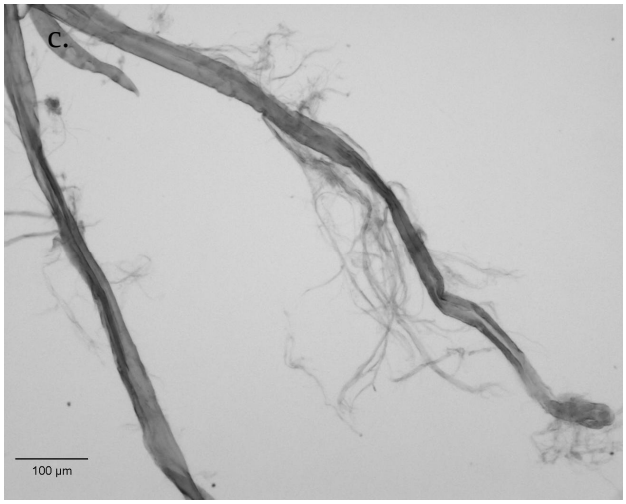


Fig. 1 Light microscope images from wet and stained pine pulp fibres at 100x magnification: a. unrefined fibres, b. 120 kWht-1 refined fibres, c. 200 kWht-1 refined fibres

It can be seen that the surface of the unrefined pine pulp fibres was intact and that there is no external fibrillation on the fibres (Fig. 1 a). Fig. 1 b. and c. show that strong external fibrillation of refined fibres exists. It is said that external fibrillation exists only in water suspension and that the removal of water brings external fibrils back to the fibre surface.[9] Fig. 2, however, shows that there is a clear difference between the dried unrefined and refined fibres. The surface of the refined fibres is much rougher than the surface of the unrefined fibres.

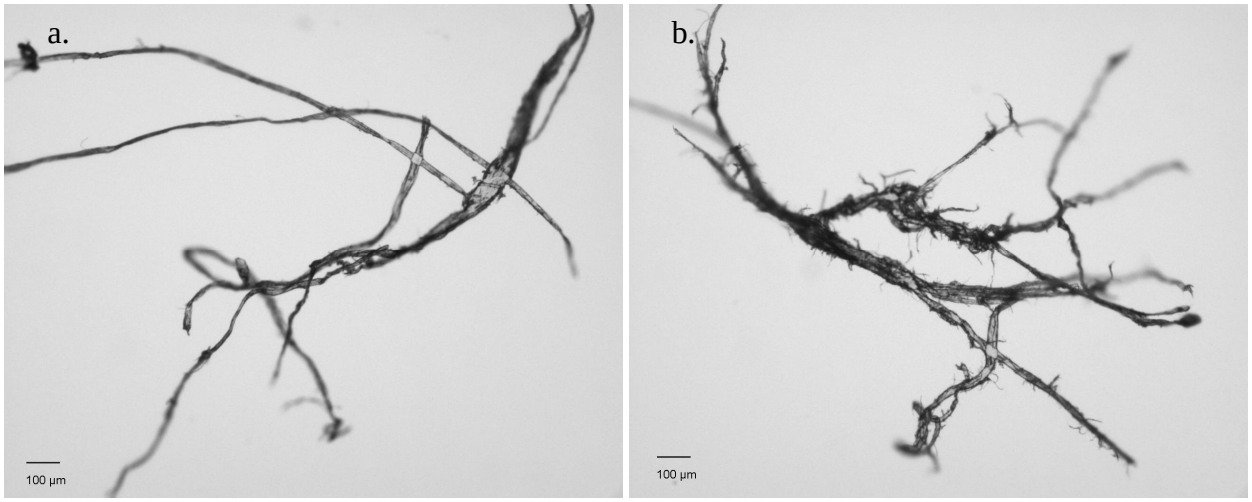


Fig. 2 Light microscope images from dry pine pulp fibres at 50x magnification: a. unrefined fibres, b. 200 kWht-1 refined fibres

By increasing the surface area of the fibres, the adhesion between the fibre and polymer might be expected to improve. Improved mechanical properties of composites may be obtained through better fibre-matrix adhesion.

Biocomposite properties

Reference composites

Reference composites with untreated TMP and chemically bleached kraft pine pulp fibres and two grades of PLA were produced according to the method described earlier. Table 4 summarises some of the primary properties of the two PLA grades used in the study. The clearest difference between the two PLA injection-moulding grades is the melt flow index: PLA 3051D is more viscous compared with PLA 3001D at the same melting temperatures.

Table 4 Some properties of NatureWorks® PLA grades[12]

Grade	Specific gravity	Melt flow index, g/10min	Melt temperature, ˚C
PLA 3001D	1.24	10-30 (190˚C, 2.16kg)	200
PLA 3051D	1.25	10-25 (210˚C, 2.16kg)	200

Fig. 3 represents colour differences between the composites with 30 w-% fibres using the same processing conditions when two different PLA grades are used. As can be seen, samples with PLA 3051D are darker than PLA 3001D specimens. This is due to the higher shear forces that exist during processing, which in turn depend on the higher viscosity of PLA 3051D. This again leads to increased pressure and temperature at the processing step, inducing discolouration of the temperature-sensitive wood fibres. The use of TMP fibres leads to a darker colour than chemically bleached (lignin-removed) pine pulp, as the lignin-containing TMP fibres are yellowish by nature.

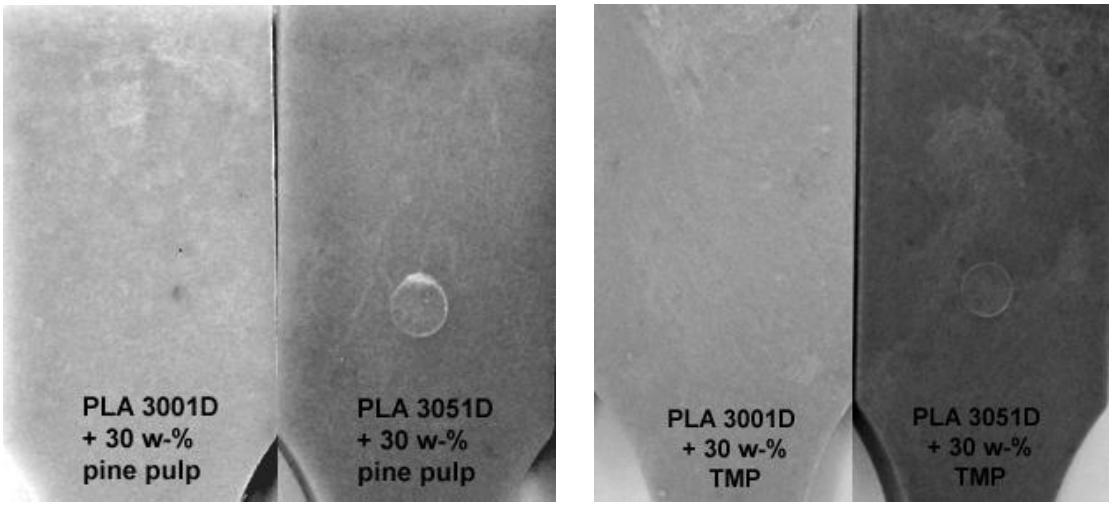


Fig. 3 Colour differences of pine pulp and TMP composites depending on the PLA grade matrix

The mechanical properties of the neat PLAs and reference composites can be seen from Table 5. The addition of 30 w-% fibres increased the elastic modulus, or stiffness, of both PLAs dramatically. The tensile strength also increased, whereas the elongation decreased. The ductility

(or impact strength) of PLA 3001D was slightly reduced when fibres were added, though the fibre addition increased the ductility of PLA 3051D.

Table 5 Mechanical properties of pure PLAs and composites with 30 w-% fibres

Polymer	Fibre	Fibre form	Polymer / Fibre, w-%	Tensile strength, MPa	Elastic modulus, GPa	Impact strength, kJ m-2	Elongation, %
PLA 3001D	-	-	100 / 0	65.1 ± 0.5	3.601 ± 0.045	18.2 ± 0.4	3.0 ± 0.2
PLA 3001D	Pine pulp	Chad	70 / 30	67.8 ± 1.3	6.190 ± 0.053	15.5 ± 2.0	1.6 ± 0.1
PLA 3001D	TMP	Chad	70 / 30	75.2 ± 1.2	6.192 ± 0.376	14.9 ± 1.9	1.7 ± 0.2
PLA 3051D	-	-	100 / 0	61.5 ± 0.3	3.640 ± 0.032	15.8 ± 0.6	4.9 ± 0.6
PLA 3051D	Pine pulp	Pellets	70 / 30	71.6 ± 0.5	6.375 ± 0.086	16.1 ± 3.7	2.1 ± 0.2
PLA 3051D	TMP	Pellets	70 / 30	77.0 ± 3.7	6.476 ± 0.140	18.9 ± 2.1	1.8 ± 0.3

When comparing the fibre types in PLA composites, TMP fibres showed better mechanical properties than pine pulp fibres in general, with only a few exceptions. This was the case although the higher aspect ratio and lower fines content of the initial pine pulp fibres could have offered better reinforcement for composite applications. There are some explanations for TMP fibres providing better mechanical properties for PLA composites than pine pulp. The advantage of the higher aspect ratio of pine pulp fibres was actually lost in the processing step (Fig. 4). The shear forces during compounding and injection moulding led to extensive fibre cutting, as longer fibres are more susceptible to length reduction than shorter fibres or fines. Full processing of the composites resulted in greater length reduction of the pine pulp fibres, leading to almost the same aspect ratio as fully processed TMP fibres. With the aspect ratios at the same level, the reason for the higher composite mechanical properties might be that TMP fibres are stiffer than chemically bleached pine pulp fibres. On the other hand, the lignin in TMP fibres might act as an adhesion promoter between the PLA polymer and the fibres, thus improving fibre-matrix adhesion.[13] With its hydrophobic nature, lignin can act as a compatibiliser between the hydrophilic fibres and the hydrophobic matrix polymer, thus strengthening the fibre-matrix interface and leading to improved composite properties.

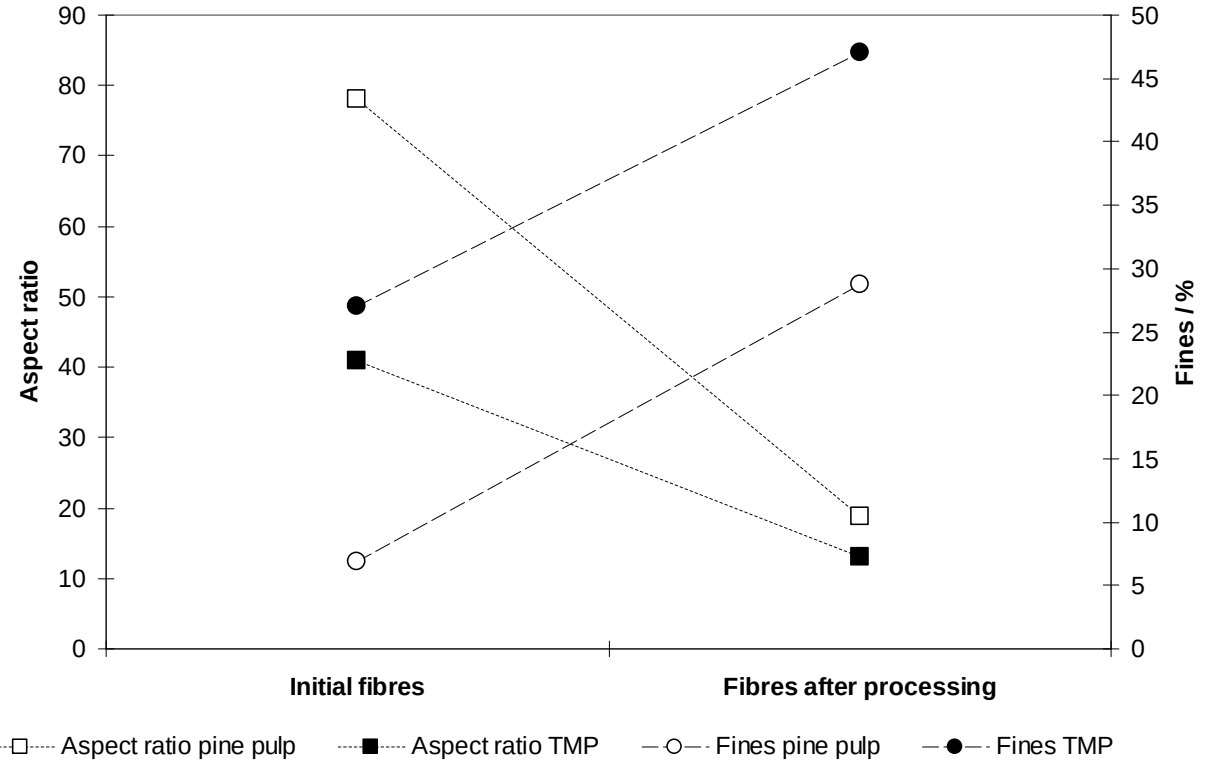


Fig. 4 Effect of processing (compounding and injection moulding) on the aspect ratios and fines contents of pine pulp and TMP fibres. To measure the fibres after processing, the PLA 3051D matrix was dissolved from the injection-moulded specimen and the remaining fibres were analysed.

Fractionation

Two fractions of TMP fibres (short and long fractions) were compounded with PLA 3001D, and the mechanical properties of the composites were studied. The results can be seen in Table 6.

Table 6 Effect of fractioning on the mechanical properties of PLA 3001D / TMP composites

PLA/pine, w-%	Fibre fraction	Tensile strength, MPa	Elastic modulus, GPa	Impact strength, kJ m-2	Elongation, %
70 / 30	Unfractioned	75.2 ± 1.2	6.192 ± 0.376	14.9 ± 1.9	1.7 ± 0.2
70 / 30	Short	67.7 ± 3.3	5.982 ± 0.316	10.4 ± 2.1	1.5 ± 0.2
70 / 30	Long	80.3 ± 1.2	6.652 ± 0.369	18.7 ± 1.9	2.2 ± 0.3

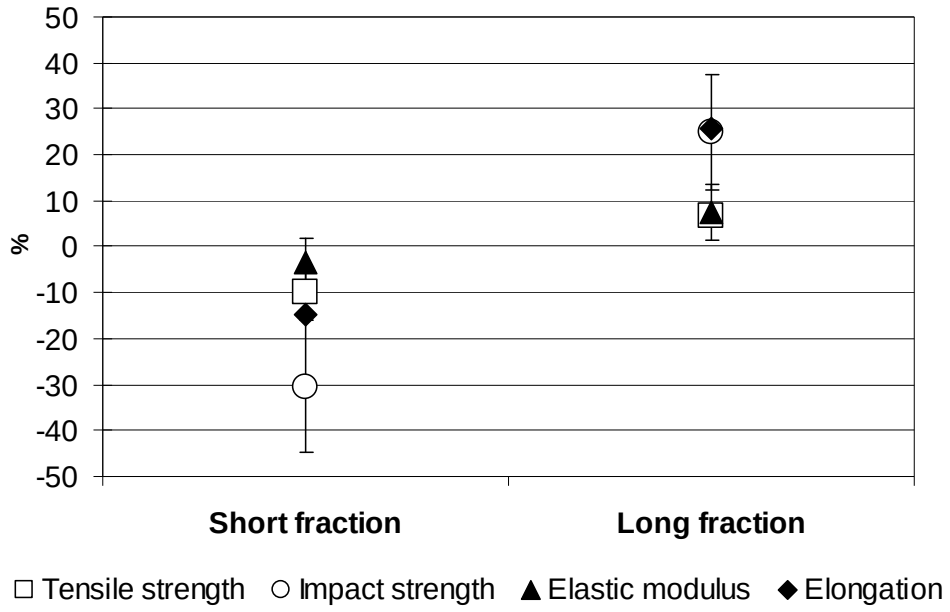


Fig. 5 Effect of fractionation of TMP fibres on the mechanical properties of PLA composites containing 30 w-% fibres. The results are normalised with respect to composites containing TMP reference fibres (no fractioning).

The influence of the use of short and long fibre fractions with respect to the TMP reference (untreated) fibres is represented in Fig. 5. The fractionation of TMP had a clear effect on the mechanical properties of the composites. The use of the short fibre fraction in PLA composites led to reduced mechanical properties: tensile strength -10%, impact strength -30%, elastic modulus -3% and elongation -15%. The long fibre fraction, however, provided improved composite mechanical properties with improvements in the tensile strength of 7%, impact strength 25%, elastic modulus 7% and elongation 26%. By removing the fines from the TMP, longer, more uniform fibres were obtained that provided a better reinforcement capability. The bonding capability of the longer fibres was also weaker, which probably led to better fibre dispersion and thus greater reinforcement of the polymer. The improved fibre dispersion of the composites with a long fibre fraction and reduced fibre dispersion with a short fibre fraction can be seen in Fig. 6, where clear agglomerates can be observed in the short fraction case.

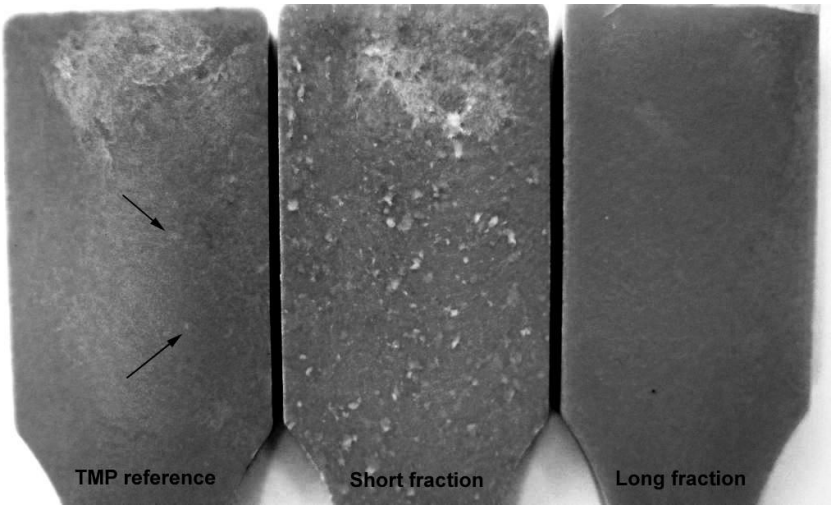


Fig. 6 Macro-scale fibre dispersion of unfractioned and fractioned (short and long fractions) TMP fibres in a PLA matrix with a fibre content of 30 w-%

Refining

Pine pulp fibres refined into levels (120 kWh t-1 and 200 kWh t-1) were compounded with PLA 3051D, and the mechanical properties of the specimens were measured (Table 7).

Table 7 Effect of refining on the mechanical properties of PLA 3051D / pine pulp composites

PLA/pine, w-%	Refining	Tensile strength, MPa	Elastic modulus, MPa	Impact strength, kJ m-2	Elongation, %
70 / 30	Unrefined	71.6 ± 0.5	6.375 ± 0.086	16.1 ± 3.7	2.1 ± 0.2
70 / 30	120 kWh t-1	60.4 ± 1.7	6.200 ± 0.039	9.1 ± 1.17	1.2 ± 0.1
70 / 30	200 kWh t-1	61.4 ± 0.3	6.131 ± 0.088	9.4 ± 1.63	1.3 ± 0.0

The effect of fibre refining on the mechanical properties of composites with unrefined pine pulp fibres can be seen in Fig. 7. The refining of the fibres led to a decrease in composite properties, although the fibrils in the fibre surface could have improved the adhesion between the fibres and matrix polymer. The reduction is due to the increased fines content, which leads to a decreased fibre reinforcement capability of very short fibres as well as fibre dispersion, the latter of which is due to the stronger bonding capability of short fibres. The effect of refining on fibre dispersion can be seen in Fig. 8, in which the poor fibre dispersion of the samples containing refined fibres can be detected from the visible fibre agglomerates.

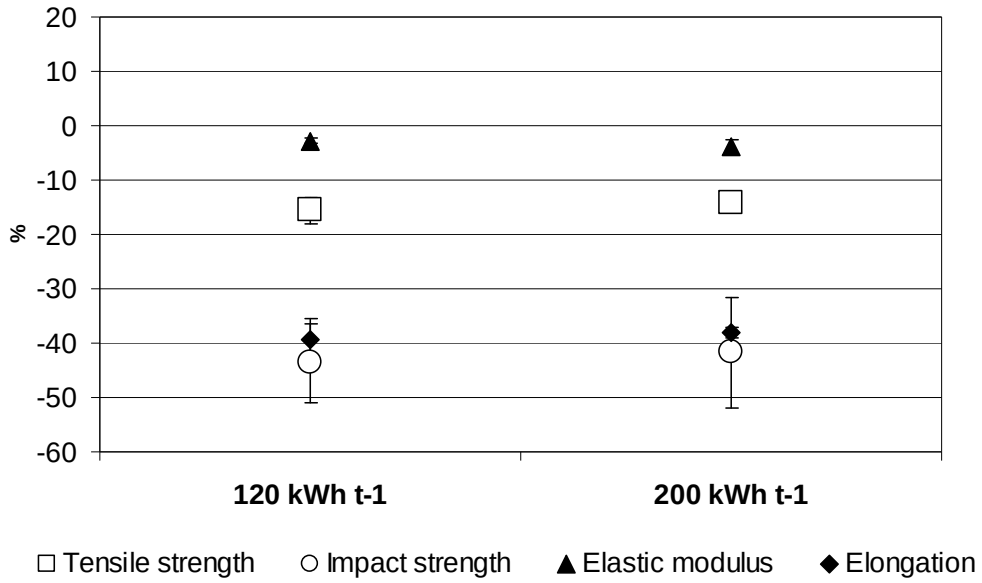


Fig. 7 Effect of refining pine pulp fibres on the mechanical properties of PLA composites containing 30 w-% fibres. The results are normalised with respect to composite containing pine pulp reference fibres (no refining).

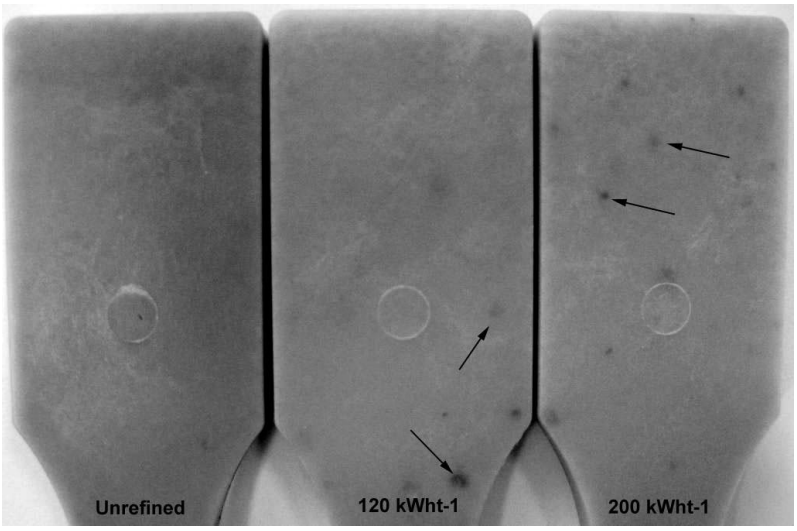


Fig. 8 Macro-scale fibre dispersion of unrefined and refined pine pulp fibres in a PLA matrix with a fibre content of 30 w-%

Conclusions

Two different kinds of pulp were chosen for the study: thermomechanical pulp (TMP) and chemically bleached kraft pine pulp. TMP contains lignin, which is removed from bleached pine pulp during the pulping process. In addition, TMP fibres are shorter, stiffer and have a lower aspect ratio than chemical pulp fibres. In PLA composites, TMP fibres showed better mechanical properties in general than chemically bleached kraft pine pulp fibres. The higher aspect ratio of pine pulp fibres was lost in the composite processing step, leading to aspect ratios of the same level for both fibres. Consequently, the better mechanical properties of TMP may be due to the higher

stiffness of the fibres. In addition, lignin in TMP fibres may act as a compatibiliser between the fibres and PLA polymer, thus improving fibre-matrix adhesion.

The fractionation of TMP fibres led to two distinct fibre fractions with different fibre properties. The longer fibre length, higher aspect ratio and lower fines content of the long fibre fraction offered a higher reinforcement capability for WPCs, thus clearly improving the mechanical properties of the produced biocomposites. The lower fines content and more uniform fibres in the long fibre fraction improved the dispersion of the fibres in the polymer matrix, whereas the poor fibre dispersion in the short fibre fraction composites containing more fines could clearly be detected.

The refining of kraft pulp fibres showed clear fibrillation of the fibre surfaces. This led to an increase in the surface area of the fibres, thus improving the fibre bonding and tensile properties of the paper. However, the refining did not give any benefits in composite manufacturing. The use of unrefined pine pulp fibres in composite applications gave better WPC mechanical properties than refined fibres did, and the distribution of fibres was also better. One reason for this is that the refined fibres formed very strong bonds between the fibres, and the pellets made from these were too tough to be able to open up and disperse well in the polymer melt during the compounding step. The refined fibres therefore remained as agglomerates rather than single fibres, and fibre dispersion decreased. The use of chemicals that weaken the fibre-fibre bonds before pelletising can improve this situation. The fines content increased when the pulp was refined, providing another explanation as to why the mechanical properties of the composites and the fibre dispersion suffered.

Acknowledgements

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