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Performance of long Canadian natural fibers as reinforcements in polymers

Qingping Guo[1], Billy Cheng[1], Mark Kortschot[1], Mohini Sain[1,2], Robert Knudson[3], James Deng[3] and Ayse Alemdar[3]

Abstract

Fiber morphology has a significant effect on the mechanical properties of fiber/polymer composites. The performance of nine types of long wood fibers (initial aspect ratio of >40), two long agricultural fibers (initial aspect ratio of >40), and one short fiber wood flour (initial aspect ratio of = 5–10) are compared. The fibers were compounded in polypropylene in a Brabender mixer and subsequently injection molded. The longer natural fibers (both wood fibers and agricultural fibers) did not provide significant additional reinforcement when compared to the wood flour. The fibers were extracted from the final specimens and measured using a Fiber Quality Analyzer. They were found to be severely degraded by processing, while the wood flour morphology was only slightly modified. The degree of length degradation was found to be dependent on fiber strength.

Keywords

Degradation, mechanical properties, natural fibers, polymer–matrix composites

Introduction

Natural fibers from forest and agricultural sources may be compounded with low melting point commodity thermoplastics such as polypropylene (PP), polyethylene (PE), and polyvinylchloride (PVC) to form a composite material known as a natural fiber-reinforced polymer or NFRP. Over the past two decades, these composites have been increasingly used in automotive, construction, furniture, and other industries because of their ecological friendliness, renewability, and relatively low cost.[1–8]

Research shows that the mechanical properties of natural fiber composites depend on the properties of the fibers and matrix, the quality of the interface between them, and the dispersion and orientation of the fibers.[9–12] Much of the previous experimental work has focused on short ground fibers compounded in polyethylene and polypropylene, with a focus on the effects of various coupling agents and dispersion aids on the final mechanical properties. There has been little work on the effect of fiber morphology, and although the benefits of longer fibers are anticipated by theory, a shortage of economically viable longer fibers and the probability of degradation of fibers during conventional thermoplastic processing have limited the research into longer fiber composites.

Fiber aspect ratio (length/diameter) affects the fiber dispersion and transfer of stress between fibers and matrix, and thus plays a significant role in the determination of ultimate composite properties. The reinforcement will be inefficient if the aspect ratio of the fibers is lower than some critical aspect ratio, which depends on the combination of fibers and matrix used.

Quite often the fibers undergo size reduction during processing. The degradation of fibers is determined by their initial aspect ratio and their strength, modulus, and toughness. For example, Senapati et al.[13] have reported that breakage of Nylon 6 and PET fibers during mixing with rubber is almost negligible.

[1]Department of Chemical Engineering and Applied Chemistry, University of Toronto, Canada.
[2]Faculty of Forestry, University of Toronto, Canada.
[3]FPInnovations-Forintek Division, Canada.

Corresponding author:
Ayse Alemdar, FPInnovations-Forintek Division, 319 rue Franquet, Quebec, Quebec, Canada G1P 4R4
Email: ayse.alemdar@fpinnovations.ca

But Kutty and Nando[14] reported significant breakage of short Kevlar fibers during mixing in Brabender Plasticorder in a TPU matrix at 180°C and at 60 rpm rotor speed for 9 min.[14] The length of fibers decreased from 6 mm to approximately 1.5 mm, the diameter remaining the same.

Commercially available wood fiber composites (WFC) are usually manufactured using inexpensive wood flour derived from sawmill and furniture industry waste. These fibers have a low aspect ratio (length/diameter) and hence they are free flowing and can be fed into an extruder with conventional feed screws. The material is referred to as 'wood flour' when it is finely ground and has a low aspect ratio. Commercially available pine flour, for example, has an aspect ratio of about 4.[15]

Although such smaller, low aspect ratio particles are easier to process, long slender fibers are expected to be much more effective reinforcement agents.[16] In order to acquire a better understanding of the effect of fiber length on fiber plastics composites processing and properties, Migneault et al.[17] conducted a series of experiments using three chemi-thermo-mechanical pulps with different lengths, distributions, and length-to-diameter ratios (L/D) obtained by mechanical refining. They used a two-stage extrusion process, and by adjusting the processing parameters according to the fiber size distribution, they claimed they obtained good quality WPC with long fibers. They found that increasing the fiber length had beneficial effects on the tensile and flexural modulus of elasticity and the toughness of the WPC. However, they did not measure the fiber morphologies after compounding, and it is possible that the benefit of the two-stage extrusion process used was an improvement in the fiber dispersion, which would also improve the mechanical properties.[18]

Woodhams et al.[19] suggested that the larger fiber aspect ratios of wood pulp fibers can impart significantly greater strength and modulus values to WFRP. However, under a mixing condition of 225°C and 90 rpm in a Brabender mixer, and further granulating and injection processes, the wood pulp fibers with larger aspect ratios were significantly degraded.

In contrast, Yam et al.[20] suggested that during processing, mechanical properties and fiber length are sensitive to extrusion parameters such as screw configuration and compounding temperature. Their experimental results show that shorter fibers resulted in better mechanical properties when compared to longer fibers because they were more readily dispersed in the HDPE matrix. However, the fiber lengths were all quite close (0.61, 0.96, and 0.92 mm), and they did not quantify the dispersion of the three fibers.

Julson et al.[21] studied various filler/matrix combinations, but did not find any obvious relationship between fiber morphology and mechanical properties. This may be because the range of filler sizes they used (40, 60, 80 mesh) was not wide enough to cause significant differences. Le Baillif and Oksman[22] concluded that double extrusion improved fiber dispersion in the matrix but considerably reduced fiber length; however, tensile properties were affected by neither dispersion nor fiber length in their study.

Canada has an abundant wood supply and the manufacturing capacity to produce long wood fibers at relatively low cost using processes originally designed for MDF production. In this study, PP composites containing nine different wood fibers, two agricultural fibers, and one commercial wood flour sample were prepared using a Brabender mixer. Test samples for mechanical property measurement were prepared using injection molding. A solvent-based fiber extraction procedure was used to extract intact fibers from the processed composites for characterization.[23] The objective of the study was to determine if superior mechanical properties could be obtained using relatively high aspect ratio fibers produced by a MDF fiber refining process, the extent of fiber degradation by polymer compounding and molding, and the relationship between species, degradation, and final properties of the composite.

Experimental

Materials

In this study, nine wood fibers, two agricultural fibers, and one grade of wood flour were used as reinforcing fillers. The fibers were provided by FPInnovations and the ground wood flour (F06) was provided by P.J Murphy Forestry Products Corp. (Tables 1 and 3). Polypropylene (PP: Pro-fax 6323) was provided by Lyondell Basell. A maleic anhydride modified polypropylene (Polybond 3200), provided by Chemtura Corperation, was used as coupling agent (CA) for some samples. When used, the coupling agent concentration was set at 2% of dry fiber weight.

Processing

The PP pellets were melt-compounded with dry fillers in a Brabender mixer. Here, the word 'filler' is used as a generic term to describe all wood and agricultural reinforcement used in this study. The compounding conditions were the same for all fillers: the compounding temperature was 180°C and the compounding time was 10 min. For each kind of filler, two composites with 30% and 50% filler loading were prepared. In the composite with 30% filler loading, 2% coupling agent (MAPP) of dry filler weight was added

Table 1. The list of fibers used in this study

	Filler name	Raw material type	Target fiber characteristic
Wood fibers	MPB	MPB lodgepole pine	Long, low final content
	BS	Black spruce	
	WC	West cedar	
	AB	Abies balsamea	
	HL	Hemlock	
	DF	Douglas fir	
	BP	Birch (Betula papyrifera)	
	AS	Aspen	
	SM	Sugar maple	
Agricultural fibers	HP1	Hemp 1/16″	
	HPB	Hemp bast	
	Flax	Flax	
Wood flour	F06	Hardwood flour	

(denoted as B302I in this context). The composite with 50% filler loading was prepared both with and without adding 2% coupling agent (denoted as B502I and B500I, respectively, in this context). Then, the composite compounds were granulated using a Brabender granulator. After the granules were oven dried, they were introduced in the hopper of an injection molding machine for preparing the test samples. The tensile strength and modulus (ASTM D 638), the flexural strength and modulus (ASTM D 790), and the impact strength (ASTM D 256) were tested for each sample. By comparing the mechanical properties of composites with 30% and 50% filler (i.e., B302I and B502I), the filler loading reinforcement was obtained. By comparing the properties of composites (50% fillers) made both with and without coupling agent (i.e., B500I and B502I), the effect of coupling agent on reinforcement was obtained. Finally, the fiber dimensions were measured. The original fibers (longer natural fibers) were characterized using a Fiber Quality Analyzer (FQA) (LDA02, Op Test Equipment Inc.). Post-processed fibers were extracted from the composites using boiling xylane. After PP was completely washed off from fibers, the fibers was dried and measured by using FQA.

Results

Figures 1–5 show the effect of wood fiber, agricultural fiber, and wood flour loading on the tensile strength and modulus, flexural strength and modulus, and impact strength of composites. In all figures, the black line (PP) represents the 100% PP control. The strength data (Figures 1 and 3) show that both the tensile and flexural strengths of composites were increased above that of the pure polypropylene for all fiber types except the composites with 30% agricultural fibers and wood flour (F06, HP1, and Flax) with coupling agent. In all cases, the 50% fiber composites outperformed those with 30% reinforcement. There was a little difference between the various fiber species, with the exception of the HL fibers, which produced slightly stronger composites.

The tensile and flexural moduli (Figures 2 and 4) increased with filler loading. The flexural data showed only marginally greater improvement, indicating that the fibers are of relatively low aspect ratio in the composites. High aspect ratio fibers are aligned in the skin regions of an injection molded specimen, and in this case the flexural modulus should be significantly higher than the tensile modulus, because the flexural test weights the skin regions more heavily. Nevertheless, in all cases, an increase in fiber loading from 30% to 50% produced an increase in modulus; however, this increase was not sensitive to the species of fiber.

The impact strength was only weakly affected by the addition of wood fibers; however, the impact strength was reinforced by adding agricultural fibers and F06 ground wood flour, especially F06 (Figure 5).

The effects of a 2% addition of coupling agent on the mechanical properties of composites are shown in Figures 6–10. As expected, the coupling agent had little effect on modulus except for wood flour F06. The modulus is measured at stresses too low to cause interface failure, and consequently, the strength of the interface is not critical provided it is sufficient to carry load for low strains. In contrast, the tensile strength depends strongly on the quality of the interface, as evidenced by the improvement associated with coupling agent addition. The coupling agent had little effect on the impact strength for our specimens except in the case of F06 (Figure 10).

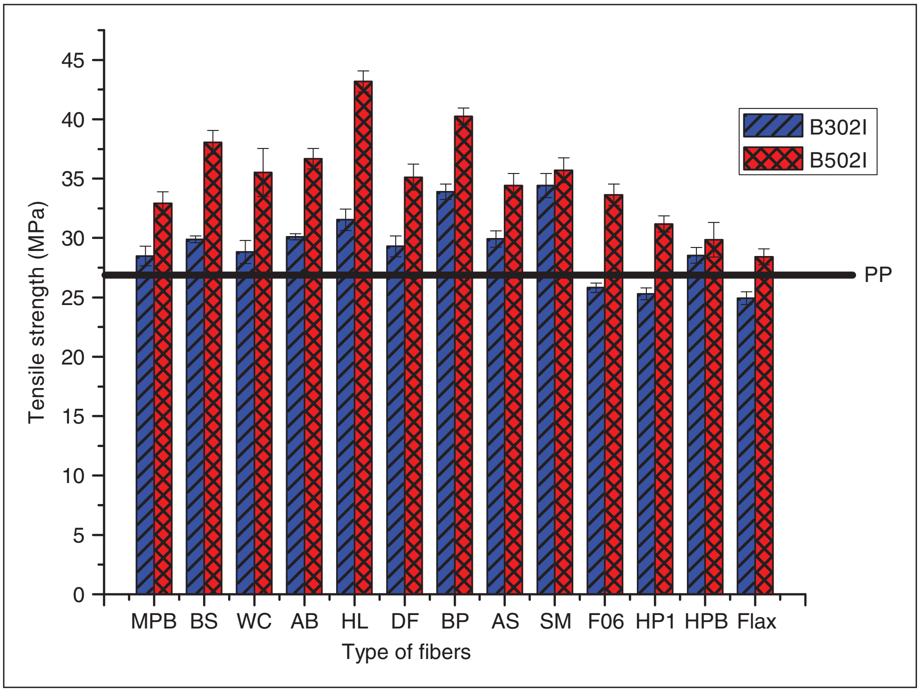


Figure 1. The effect of filler loading on tensile strength.

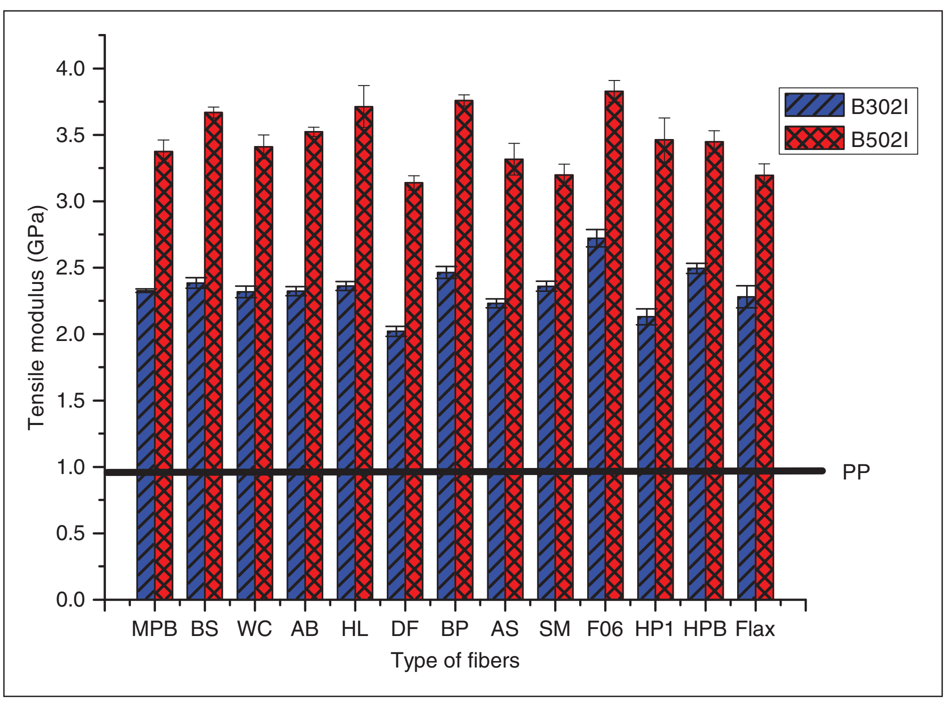


Figure 2. The effect of filler loading on tensile modulus.

Discussion

Table 2 presents a summary of the effects of fiber loading, coupling agent, and fiber type on the mechanical properties of the composites tested in this study. In general, the effect of wood flour on both the strength and modulus is similar to the effect of wood fibers. Although the wood fibers initially had a much higher aspect ratio, which was expected to facilitate stress transfer, the results did support this hypothesis. Coupling agent addition improved the tensile strength by improving the interface adhesion between the fibers

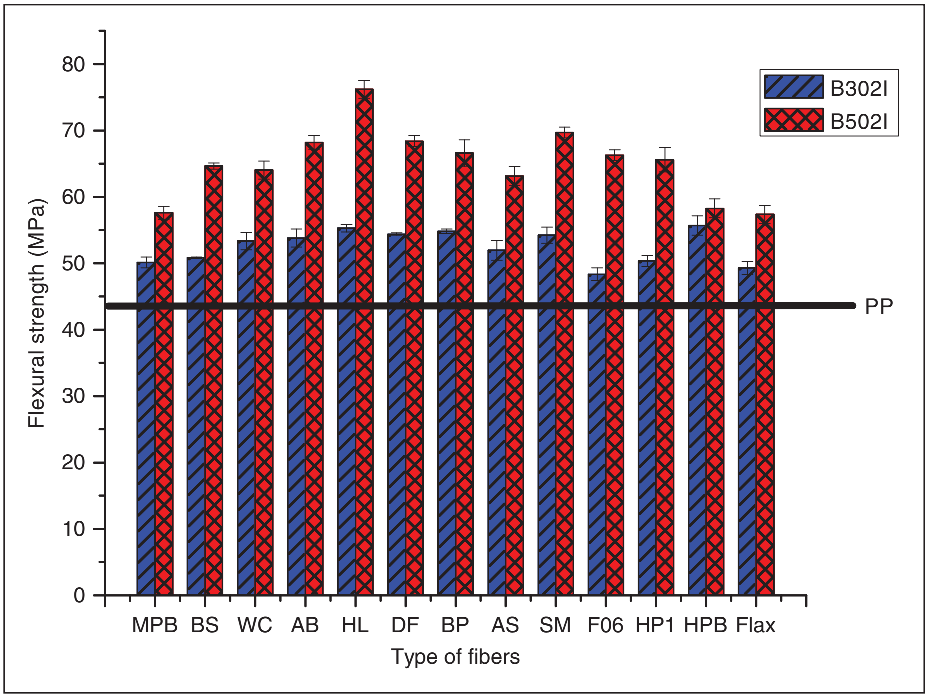


Figure 3. The effect of filler loading on flexural strength.

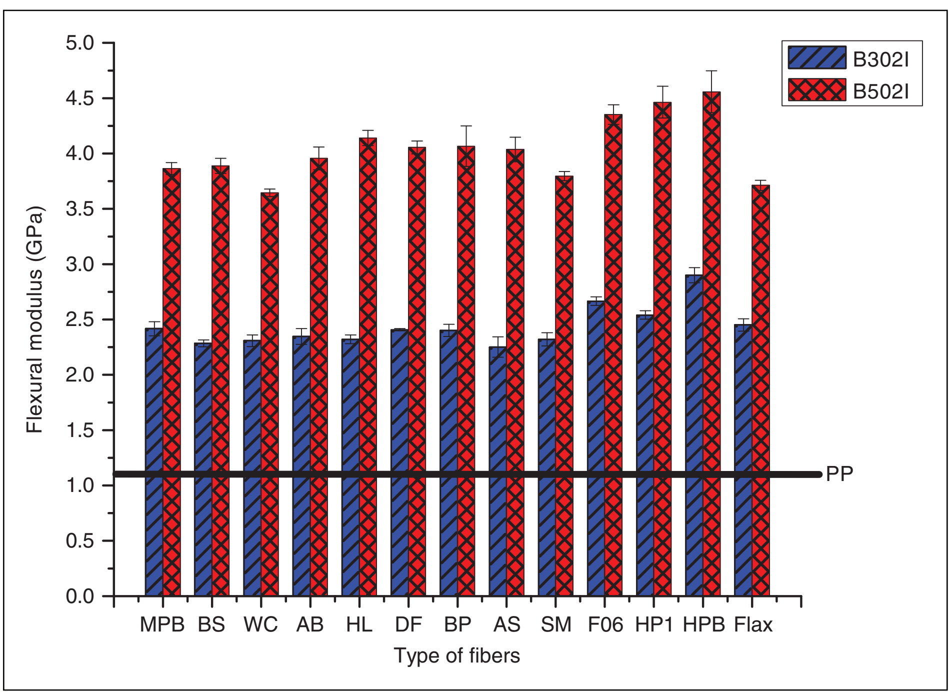


Figure 4. The effect of filler loading on flexural modulus.

and PP matrix, but this had little effect on the modulus or impact strength.

Of course, it has long been known that conventional thermoplastic processing can result in fiber degradation, so fibers were solvent extracted from the as-processed composites in order to measure the extent of degradation in our study. Table 3 shows the fiber morphologies before and after processing (only two wood fibers (MPB and BP) are shown here; others were similar). For all fibers, the degree of degradation was significant after processing, but wood flour morphology changed only slightly.

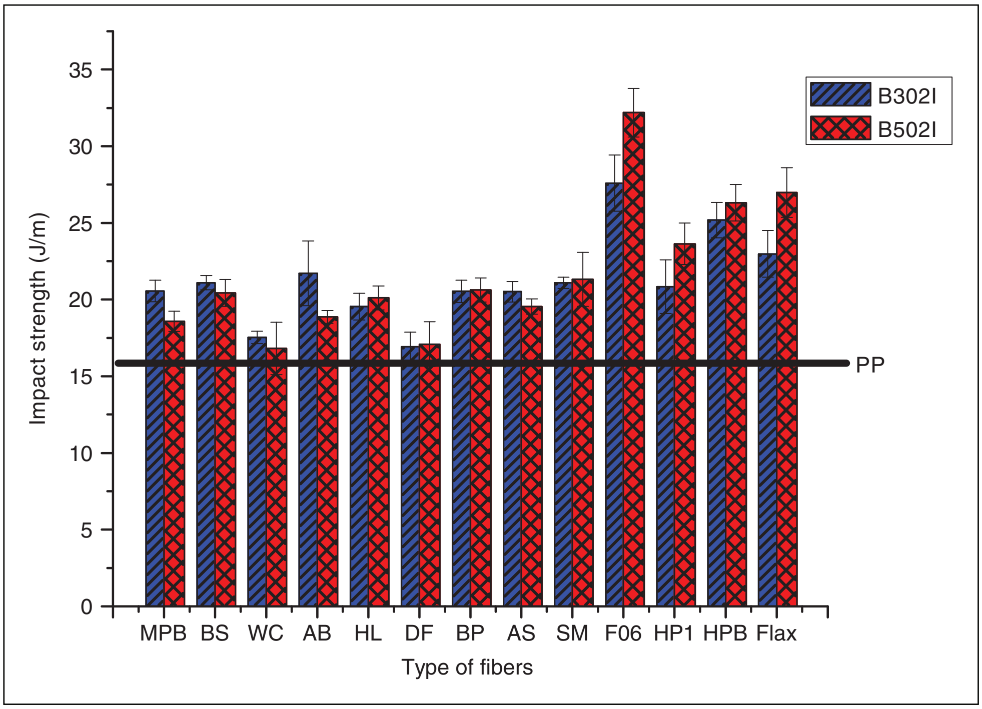


Figure 5. The effect of filler loading on the impact strength.

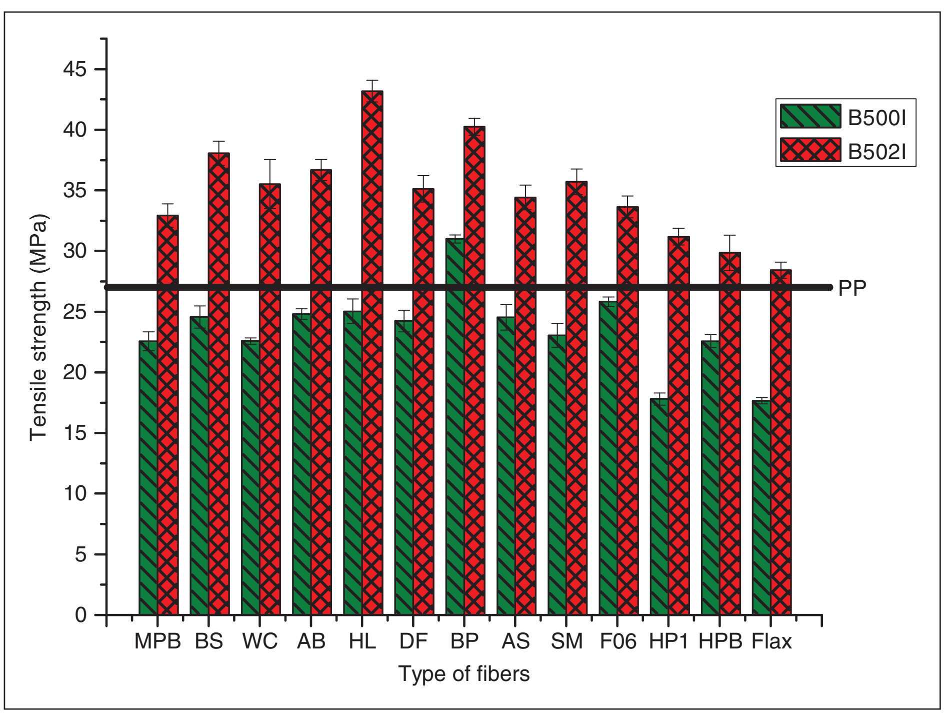


Figure 6. The effect of coupling agent on tensile strength.

After processing, the particles of the wood flour and the agriculture fibers (Hemp 1/16, Flax) in composites are much bulkier than wood fibers while the particles of hemp bast are similar to wood fibers. The bulky, low aspect ratio particles play a different reinforcing role in composites: (1) they may derive more benefit from coupling agents that improve the interfacial bond between the fillers and matrix;

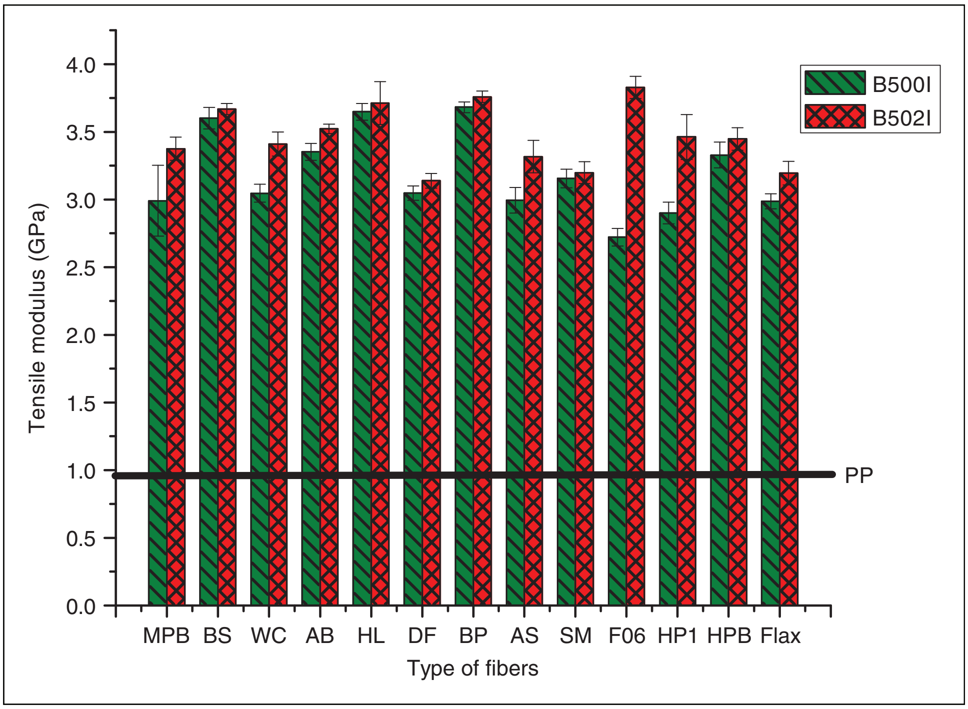


Figure 7. The effect of coupling agent on tensile modulus.

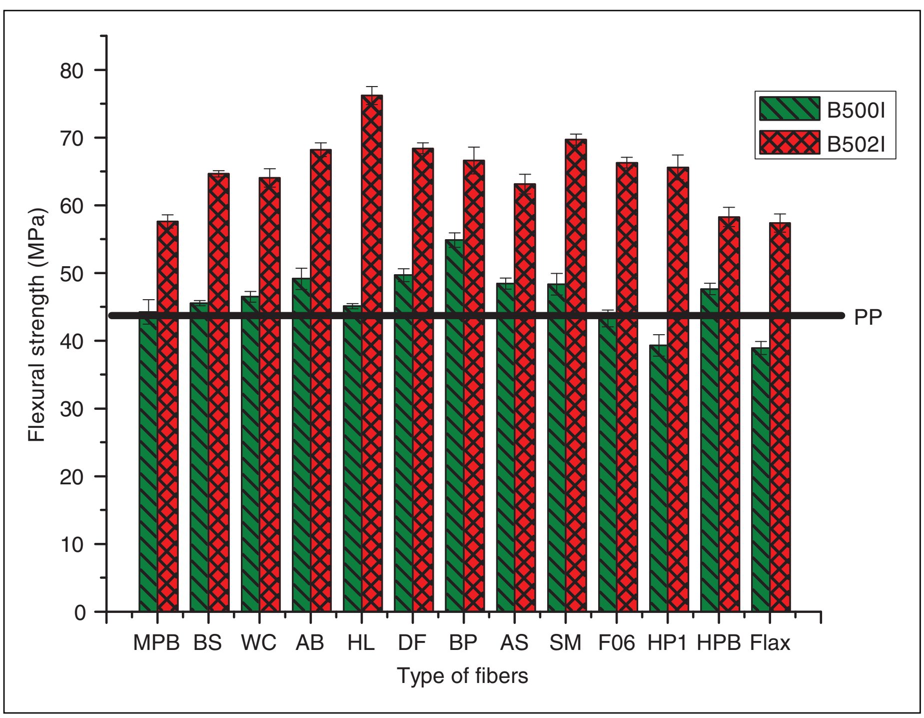


Figure 8. The effect of coupling agent on the flexural strength.

and (2) their size and shape may play an important role in improving the impact strength of composites. For example, in Figure 1, the interfacial bonding between the filler surface and matrix for the three bulkier particles was apparently poor, even in the presence of 2% coupling agent, and the resulting tensile strength of these three composites (at 30% filler content) was decreased compared with PP control. However, the agricultural fibers and wood flour outperformed wood fibers in reinforcing the impact strength of

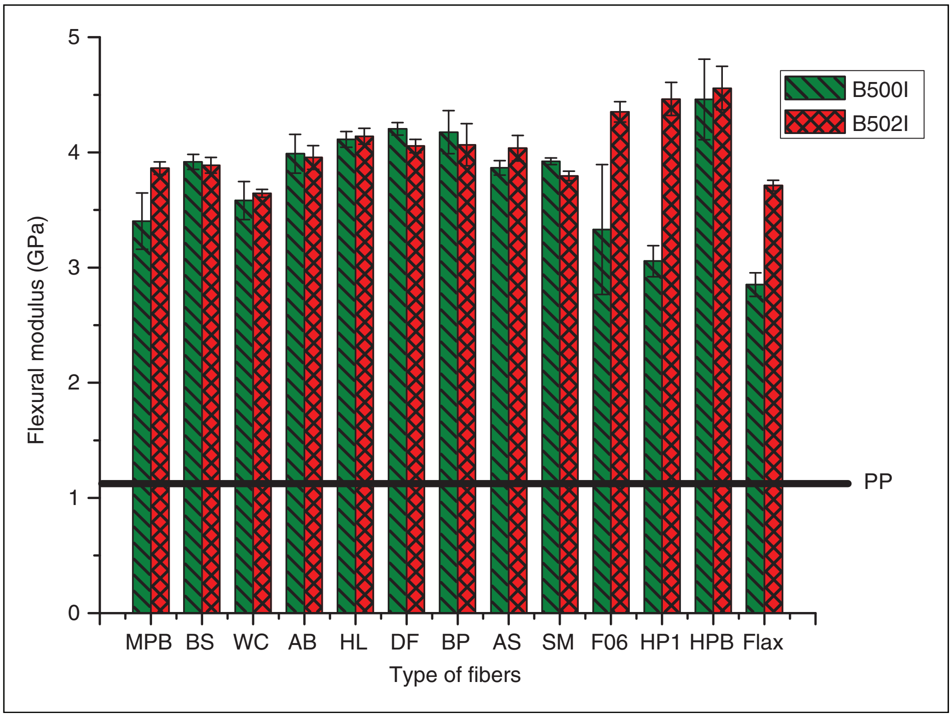


Figure 9. The effect of coupling agent on the flexural modulus.

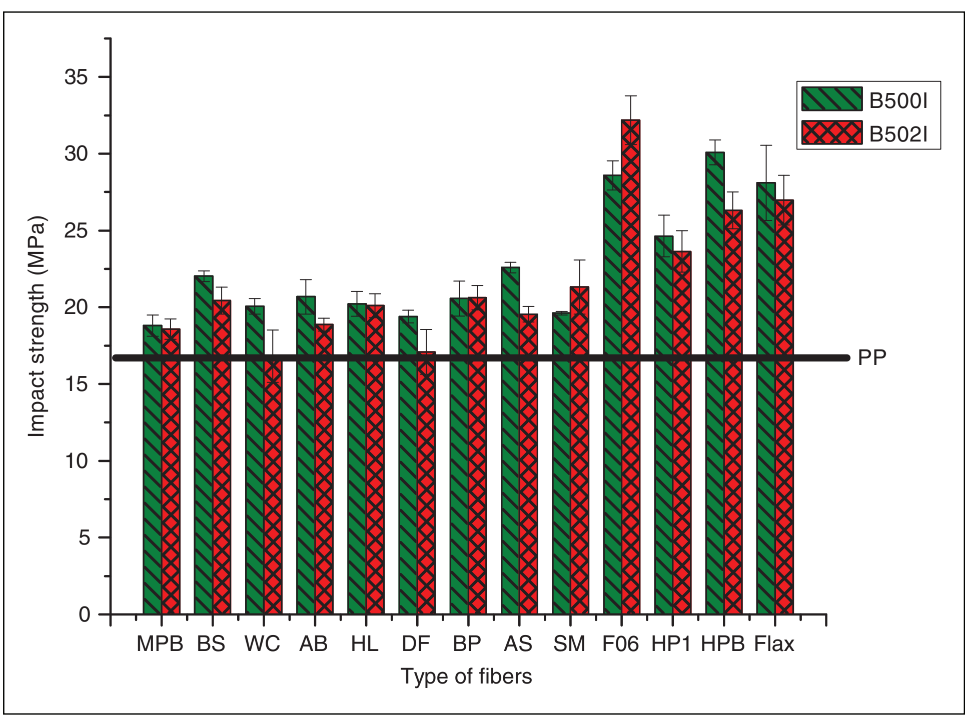


Figure 10. The effect of coupling agent on the impact strength.

composites both with (Figure 10) and without (Figure 5) coupling agent.

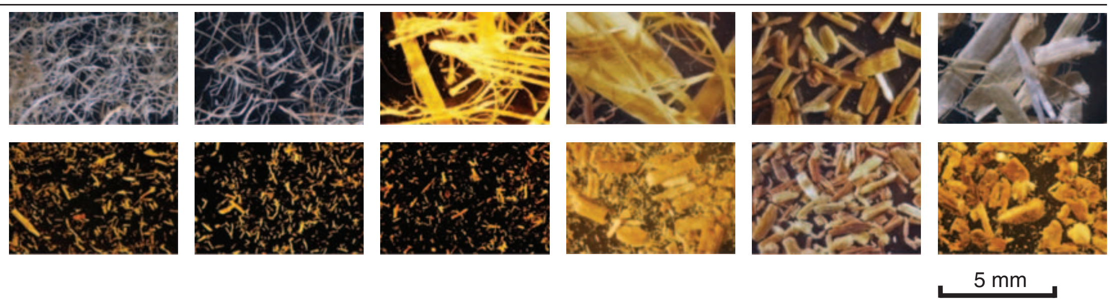
Based on the images in Table 3, both wood fibers and agricultural fibers were severely degraded during processing. The fiber lengths were characterized in an Optest FQA, and Figure 11 shows the average length-weighted length of wood fibers both before and after processing. The initial fiber lengths varied significantly, and the longer fibers were generally slightly longer after processing, but the processing equipment effectively

Table 2. Summary of the effects of fiber loading, coupling agent, and fiber type on mechanical property

		Composites of wood fibers	Composites of agricultural fibers	Composites of wood flour
Filler loading	Tensile and flexural strength	++	0 to +	++
	Tensile and flexural modulus	+++	+++	+++
	Impact strength	0	++	++
Coupling agent	Tensile and flexural strength	+++	+++	+++
	Tensile and flexural modulus	0	+	++
	Impact strength	0	-	++

Table 3. The fiber morphology before and after processing

	MPB	BP	Hemp Bast	Flax	F06	Hemp 1/16
Before processing						
After processing						



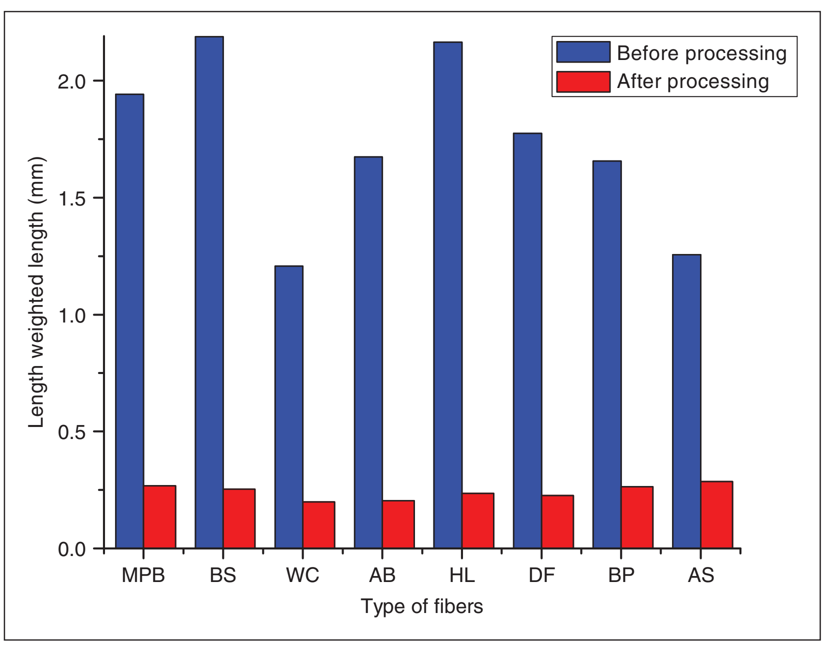


Figure 11. The degradation of wood fibers after processing.

reduced all fibers to a relatively constant length of about 0.25 mm.

Figure 12 shows the relationship between the zero-span tensile strength (a measure of fiber strength) and the weighted length of wood fibers after processing. The fiber strength had a clear effect on the fiber length after processing: the stronger the wood fibers, the longer the final average length of wood fibers are after processing. However, in Figure 13, it is clear that longer post-processed fibers do not lead to greater composite strength. This may be due to problems in dispersing longer fibers, or it may simply be that the

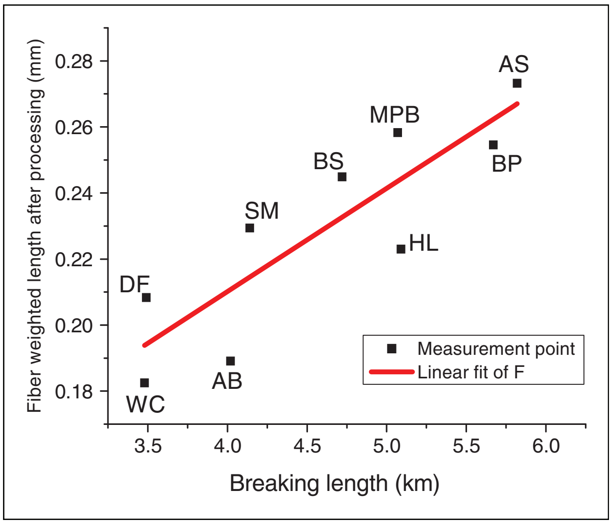


Figure 12. The relationship between fiber length and zero-span breaking length (normalized strength).

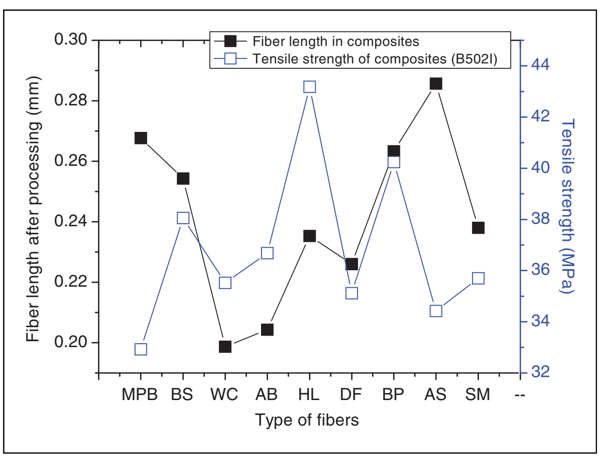


Figure 13. The comparison of trends of tensile strength and wood fiber length.

post-processed fiber lengths did not span a wide enough range to reveal an underlying trend.

Conclusions

The tensile and flexural strengths and moduli were significantly increased with increases in fiber loading. However, only the bulkier agricultural fibers and wood flour improved the impact strength of the composites tested in this study. The tensile strength and flexural strength for all fiber composites were significantly increased by adding coupling agent, while tensile modulus, flexural modulus, and impact strength were not significantly affected. The coupling agent has a larger effect on the properties of composites with bulky, low aspect ratio particles, as expected.

Both wood fibers and agricultural fibers were severely degraded during processing. Longer initial fibers did result in marginally longer final fibers, but the strength of fibers had a much more significant effect on the fiber length after processing, with stronger wood fibers holding up better during processing. Even so, the stronger, longer fibers did not result in better mechanical properties.

Acknowledgment

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