

WOOD AND FLAX FIBER POLYOLEFIN COMPOSITES

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Abstract

In this work, natural fiber and wood composites based on neat and recycled polypropylene (PP) were fabricated by melt processing. Different formulations, including various reinforcement content, different types of coupling agents, different types of reactive additives, and an impact modifier were developed. The reinforcements were in the form of natural fibers like banana, flax, rice husk and palm fibers and of wood sawdust. For the long fiber composite systems, processing was done by compression molding of piles of long fiber mat and extruded polypropylene film. For the short fiber composite, the samples were prepared by extrusion followed by injection molding. The tensile, flexural and impact performance were characterized and all composites show superior mechanical properties when compared with the pristine matrix. Mechanical performance of the wood composites was also evaluated before and after conditioning in water for 1 and 7 days. Results indicate that the composites resist to humidity very well. The results also demonstrate the effect of formulations on the performance of the recycled composites.

INTRODUCTION

Although much of the pioneering work in wood-filled thermoplastic composites was done in the 1970s [1-3], only recently there has been substantial development. The market for thermoplastic wood composites in North America was 320,000 tonnes in 2000 and the volume is expected to more than double by 2005 [4].

Wood plastic composites are gaining market share because of their advantages over wood and metal in terms of longevity, appearance, life-cycle cost, and value. They are attractive, insect and rot resistant, and paintable while they can be made to have a wood look. In addition, they are stiffer than plastic products while at the same time they can be worked, cut, glued and fastened with the same screws, nails, or staples as wood.

The great majority of reinforced thermoplastic composites available commercially use as their reinforcing fillers inorganic materials like glass, clays, minerals, etc. These materials are heavy and abrasive to processing equipment. In contrast with conventional reinforcements, wood and natural fibers possess the advantages of low cost, light weight and non-abrasiveness. In addition, compared to natural fibers, wood fibers are very cheap, therefore it is not surprising that wood fibers are more attractive for composite production [5-16].

At this stage in the technology, the processing of wood thermoplastic composites has been well explored. Wood thermoplastic composites can be processed either by co-rotating or counter-rotating or parallel or conical twin-screw extruders, or single-screw extruders as well. Wood thermoplastic composites can be compounded by an inline or offline procedure, using continuous or batch processes. However it is essential that the wood reinforcement must be

dried well before compounding with the polymer matrix in order to avoid degradation and formation of bubbles/voids. Furthermore, water in the extruded materials can also reduce extrusion speed, as well as destroy the surface finish and mechanical properties. Several different approaches have been developed to meet this requirement but each of them poses different challenges. For example, in a separate step, wood can be dried in a kiln or oven (suitably explosion-proofed due to the powdery nature of most wood). However, the cost can be high since the wood needs to be heated twice, first during drying and then in the processing step; the equipment occupies a large space; and it is also very difficult to store the dried wood. In a second approach, the drying is done together with processing in a twin-screw extruder (M. Hotchkiss, Farrel, Cincinnati), in which the wood is fed in first and the plastic part-way down the barrel, or it can be done in a continuous mixer. This gives the advantage of tremendous free volume and surface renewal capabilities but low output. Recently, Davis Standard has introduced the Woodtruder into the market, which is a parallel, 28/1 L/D counter-rotating twin-screw extruder into which the wood is fed, and which has a single screw, side-injection extruder to feed melt plastics. No material preparation is needed, with moisture removal via atmospheric and vacuum vents. The advantages of this system include the low speed, low shear action, the separate melting process of polymer prior to addition of fibers, and the elimination of the cost of drying, size reduction, and blending operations but it complicates the process and requires high investment.

An alternative approach using chemical principles to resolve the drying problem and improve the wood composite performance was developed by Got et al. [17]. In general, the humidity and wood acidity are absorbed/neutralized with/by basic oxides, such as CaO, during processing. As a result no drying of wood is required and degradation is limited. However, according to the authors the amount of calcium oxide must be carefully calculated so that the water of the wood cellulose filler becomes finally 2-5% by weight, otherwise the composites will become very sensitive to humidity. Since humidity in wood varies from one location to another, the method disclosed in this patent is difficult as the correct humidity of the wood is not always easy to determine. In addition, the use of CaO requires surface treatment of the CaO. This paper presents a method to overcome wood humidity and improve the mechanical properties of the wood composites in both dry and wet conditions in which the basic oxide-based filler is used without surface treatment. The method was also validated for banana and flax fibers.

EXPERIMENTAL

Materials

Reground recycled PP1 and PP2 were obtained from Novoplas (Quebec, Canada) in the form of an injection grade with MI = 5 and 8 g/min, respectively. Virgin PP6100 SM was supplied by Montell (PP-a). High melt flow index PP (PP-b) was provided by Unicolor (Quebec, Canada). Coupling agents (CA) based on maleic anhydride (MA) grafted polypropylene, such as Epolene-43 (E43) (acid number AN = 45; Mn = 9,100; ~4.81 wt% of MA), Epolene-3015 (E3015) (AN = 15; Mn = 47,000; ~1.31 wt% of MA), and Epolene-3003 (E3003) (AN = 8; Mn = 52,000; ~0.71 wt% of MA), all from Eastman Chemicals, were used in this study. An impact modifier, ethylene-propylene rubber DSM Sarlink 419 (EPR), was also used in some formulations.

Different spruce sawdusts (> 2% moisture) were provided by JER Envirotech (Vancouver, Canada) (Fig. 1). The particle distribution of different wood sawdust sources is shown in Fig. 2. The sawdust was screened to obtain an average particle size of 0.5 mm and

homogenized prior to processing. Banana fiber and flax fiber were kindly supplied by JER Environtech and Biolin (Alberta, Canada), respectively.

Calcium oxide CaO was supplied by The C.P. Hall Company with max 92 wt% CaO. Aluminum oxide Al₂O₃ was obtained from Malakof Industries Inc with min 95 wt% Al₂O₃.

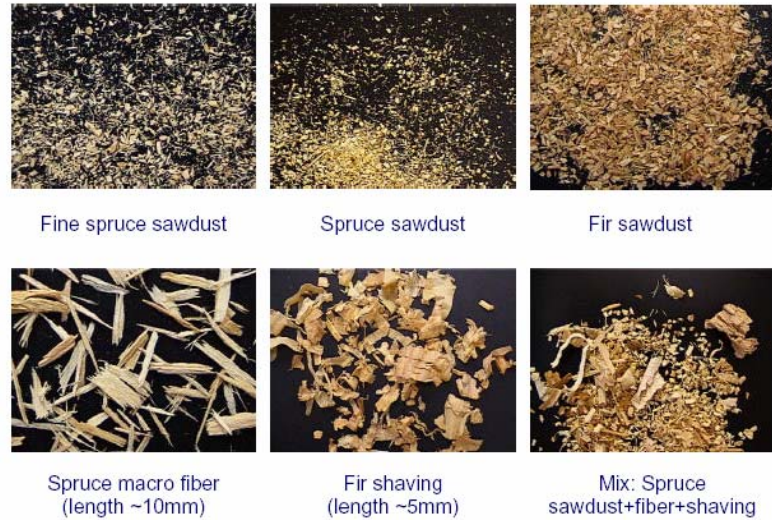


Figure 1. Different recycled wood sources.

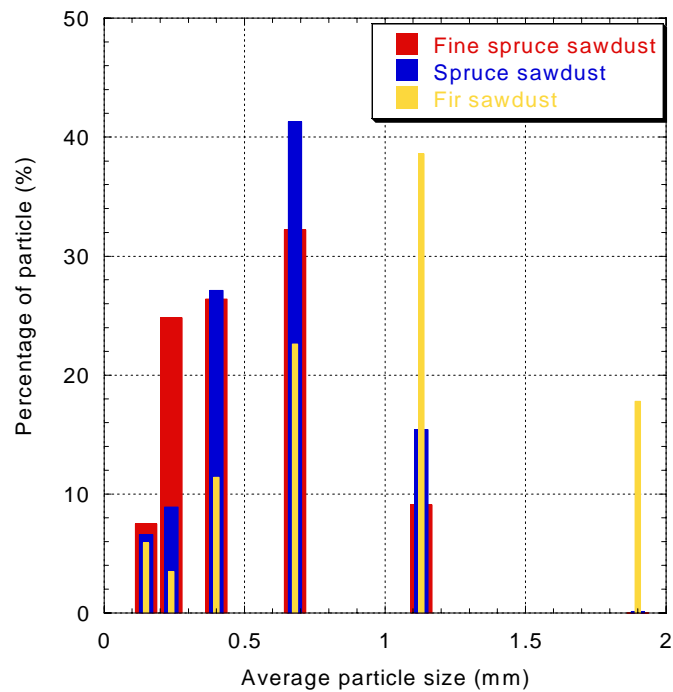


Figure 2. Size distribution of the wood sources.

Composite Preparation

The sawdust was dry-blended with the polymer pellets then fed into a twin-screw extruder Extrusion Spec W&P 30 mm having L/D = 40; speed = 150-175 rpm; T_{max} = 185°C. For testing, samples were molded by injection (BOY 30A injection machine at T = 200°C). The wood sources were dried in a vacuum oven at 100°C for more than 6 h prior to processing. All samples contain 40wt% wood particles and 2wt% coupling agent.

Characterization

Density measurements were made with a pycnometer AccuPyc 1330. Water absorption was determined by measuring the weight gain or loss after immersing the sample in water for 1 to 7 days.

Microstructural observations of the composites were made using a JEOL JSM-6100 scanning electron microscope (SEM). Observations of the dispersion of the cellulosic filler in the polyolefin matrix were made using a Leitz Dialux 20 optical polarised microscope (OM), and of their interface using SEM. Interaction between the basic reactive filler and the graft polyolefin was studied by means of transmission infrared spectroscopy (FTIR) at room temperature on a Nicolet Magna 860 Fourier transform instrument at a resolution of 4 cm⁻¹.

Tensile properties were measured according to ASTM D638. Impact resistance (ASTM D256) was evaluated using an un-notched IZOD impact test. All tests were conducted at ambient temperature. To evaluate the resistance to humidity, samples were immersed in water for different periods of time between 1 and 7 days and then tested.

Results and Discussion

Wood particles are very well dispersed in the PP matrix and the SEM image in Figure 3 demonstrates a good coverage of the matrix on the particles. The hollow structure of the wood particles was also observed. The tensile properties of the composites are given in Fig. 4. The strength and modulus of the composites are superior to those of the pure matrix. The use of fillers in the formulation improves both strength and modulus. The increase of filler amount improves the both properties for samples with basic fillers but reduces the strength in the case of the inert fillers. Among them, CaO exhibits the best reinforcing effect providing highest strength and modulus. In order to understand the role of basic filler in the formulation, FTIR of different blends of the pure PP matrix with CaO, E43, CaO and E43 have been studied. Figure 5 illustrates the raw spectra of those samples along with the difference spectra obtained when the PP spectrum is subtracted. The results demonstrate that the reaction between CaO and the anhydride and acid groups of the E43 took place easily during processing, forming a calcium carboxylate salt (peak at 1560 cm⁻¹).

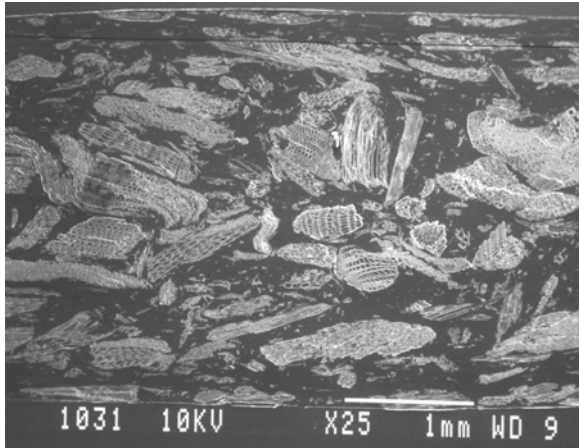


Figure 3. SEM of the wood composite.

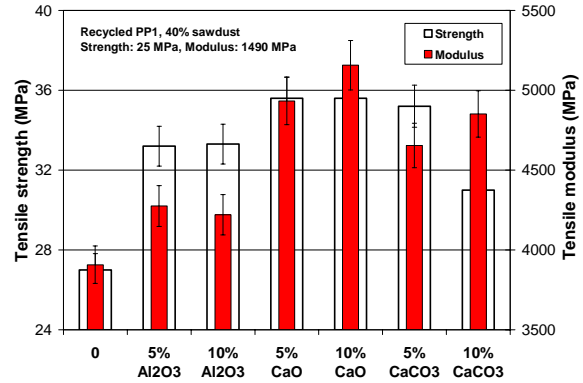


Figure 4. Tensile properties of the wood composites

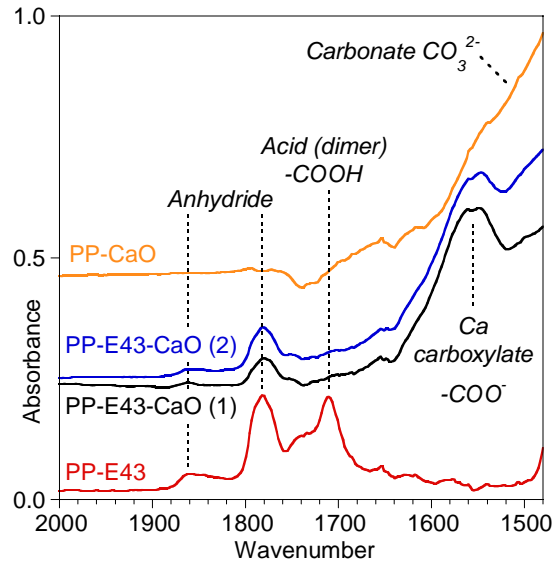
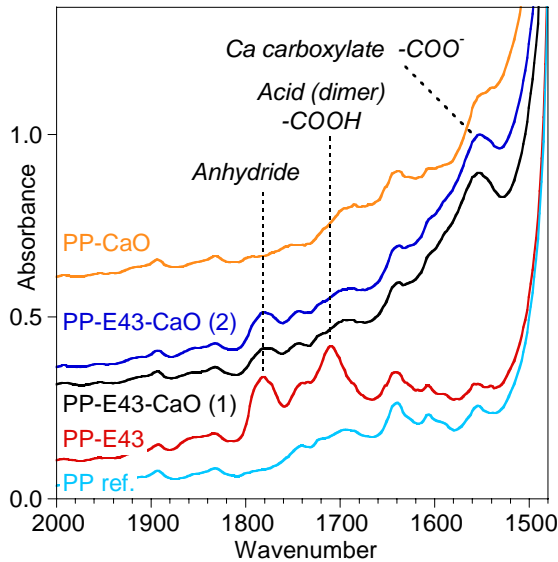


Figure 5. FTIR spectra of the composites: a) raw spectra and b) difference spectra with PP spectrum subtracted.

The effect of the coupling agent chemistry (molecular weight and grafting amount) on the formulation was also examined. As indicated in Figure 6, in the absence of CaO, E3015 provides a greater strength but lower modulus. This can be explained by the fact that E3015 has higher molecular weight, which allows a better compatibility with the PP matrix. In the presence of CaO, E43 gives better performance than E3015. E43 which has a high amount of

grafting, should allow better interaction with the wood particles and the CaO. The negative effect of the low molecular weight of E43 can be significantly reduced by the reaction between CaO and E43. Thus, all lead to a better improvement in interface and dispersion. Figure 7 shows that E3015 provides a better ductility and toughness as a result of its higher molecular weight compared to E43. However, with the incorporation of CaO, ductility and toughness of samples formulated with E3015 were reduced, while those of samples formulated with E43 increased (due to a better chance for chemical reaction between E43 and CaO). At 10wt% CaO, there is no difference between the two formulations in the ductility and toughness. In addition, the TGA study also demonstrates that the presence of CaO has significantly improved the thermal stability of the composites as shown in Table 1. As a result of decreased degradation during processing, the mechanical performance should be improved.

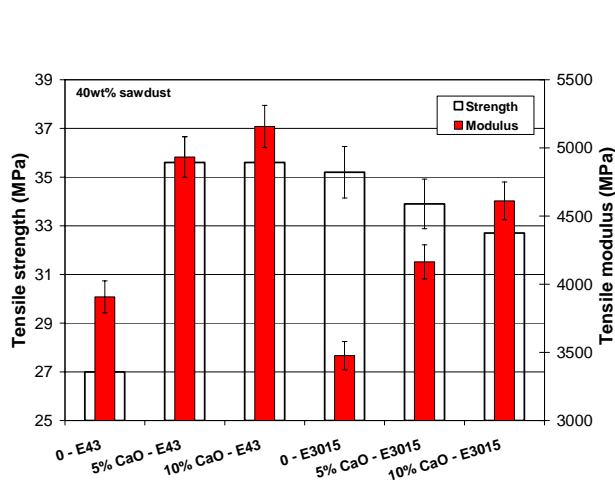


Figure 6. Tensile properties of the wood composites with different compatibilizers

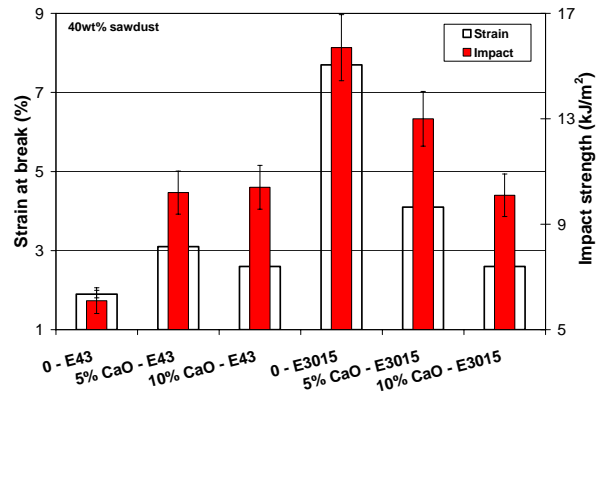


Figure 7. Ductility and impact of the wood composites with different compatibilizers.

Table 1. Thermo-stability of the composites.

No	T _{10%} (°C)	T _{20%} (°C)	Weight loss at 500°C (wt%)
No CaO	334	364	91
10% CaO	346	398	73

Table 2. Water absorption of the composites.

Sample	Water Absorption (%)		
	24 h in water	24 h in water, 24 h in air	7 days in water
No CaO	0.35	0.2	0.9
5% CaO	0.55	0.25	1.25
10% CaO	0.55	0.3	1.45

The effect of the type of PP on the formulation was also studied. Figure 8 shows the tensile properties of the composites prepared from recycled PP1, recycled PP2, and pure PP Profax 1274. The positive effect of CaO in the mechanical performance of the wood composites was again confirmed in all three PP matrices.

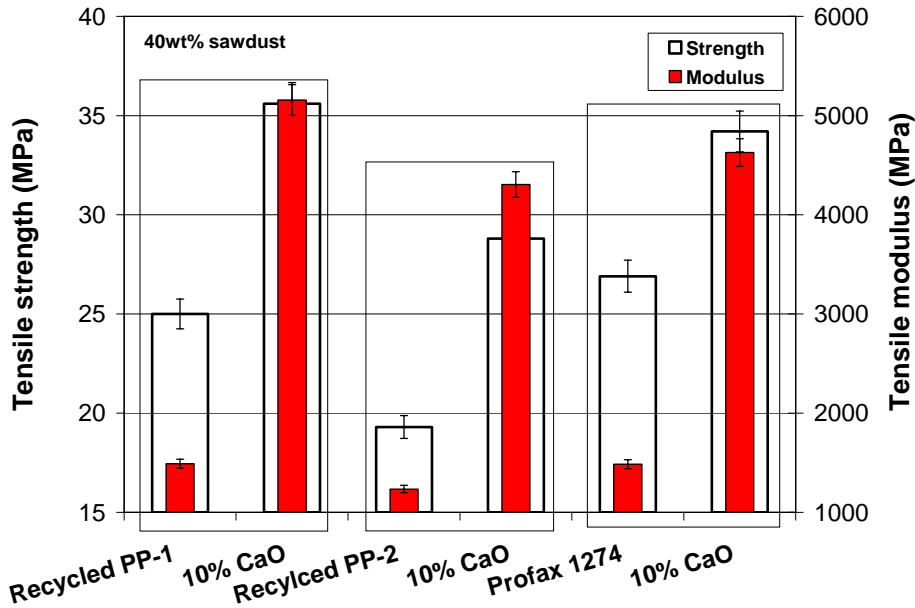


Fig. 8. Tensile properties of the wood composites with different PP matrices.

The effect of CaO on the formulation was also examined using compression molding. The matrix was extruded to form a film and the banana and flax fibers were prepared in the form of mat as shown in Figure 9. The concentration of fiber was kept at 20 wt%. Preliminary results show that the presence of CaO improves the strength by more than 20% of and the modulus by over 40% as shown in Figure 10.



Fig. 9. Tensile properties of the wood composites

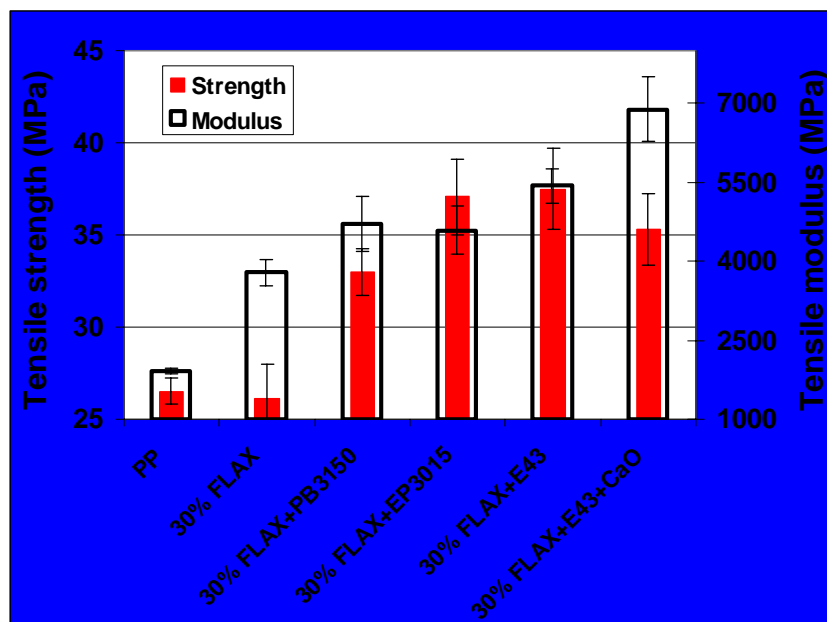


Fig. 10. Tensile properties of the wood composites

Table 2 addresses concerns about the effect of CaO on the water absorption of the composites. There is some increase in the water uptake for sample containing CaO. However, it should be stressed here that the mechanical properties of all samples remain unchanged after conditioning in water for 24 h and 7 days.

Conclusions

The results show that the presence of basic oxides like CaO and Al₂O₃ has significantly improved the mechanical performance of the wood composites. These oxides absorb the humidity and neutralize the acidity of the wood reinforcement, thus limiting the degradation during compounding. The short-chain coupling agent MAgPP E43 provides a better advantage since it has a higher mobility to impregnate the wood particle surface. The loss in mechanical properties resulting from the short-chain molecules of E43 is limited by the presence of basic oxide fillers. Besides the reinforcing effect, these fillers can react with the MA group of E43 to increase the effective molecular weight of the E43, thus improving the composite performance. The presence of basic oxide fillers should improve the flame resistance and decay resistance. Further experiments involving these aspects are under consideration.

Acknowledgements

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