

# METHOD FOR IMPROVING PROCESSING AND PROPERTIES OF CELLULOSE FIBRE THERMOPLASTIC COMPOSITES

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**SUMMARY:** Cellulose fibre composites have been used extensively in the construction industry in North America and have begun to attract much attention for the automotive sector in Europe, South America, and Asia. In order to increase its competitiveness, it is important for the composites industry to reduce energy consumption and costs as well as to improve the output and the performance. To facilitate the extrusion process as well as to reduce the processing cost and energy and the complications related to wet cellulose sources, different types of cheap reactive additives have been incorporated in the process in different ways. This new approach allows elimination of the cellulose drying step and the safety challenges related to the high flammability of the dried cellulose source, thus reducing sufficiently the overall processing energy and cost, and also facilitating the flow in the extruder. In addition, it can also significantly improve the mechanical and thermal properties and the flame resistance of the composites. The investigation has been performed on polypropylene composites with wood fibres and two types of flax fibres. The first one was obtained by hammer-milling the field-retted flax fibres while the second one was produced by bio-scouring (using a genetically modified pectate lyase at 37°C and pH 8.5) the flax fibres obtained from a mechanical decortication process.

**KEYWORDS:** natural fibre, polyolefin, formulation, processing, mechanical properties, fire resistance

## INTRODUCTION

Although the pioneering work in natural fibre thermoplastic composites started in the 1970s, they are still undergoing substantial development even today. Wood fibre thermoplastic composites are gaining market share because of their advantages over wood and metal in terms of longevity,

appearance, life-cycle cost, and value (1, 2). They are attractive, insect- and rot-resistant, and paintable, while they can be made to have the look of wood. In addition, they are stiffer and cheaper than plastic products while at the same time they can be worked, cut, glued and fastened with the same screws or nails as wood. On the other hand vegetable fibre thermoplastic composites are attractive to the automotive industry because of their low density and ecological advantages over conventional composites (3-5). As a result, wood fibre composites have been used extensively in the construction industry in North America while vegetable fibre composites have been introduced into personal vehicles in Europe, South America, and Asia. In order to increase its competitiveness, it is important for the composites industry to reduce energy consumption and cost as well as to improve output and material performance.

At this stage in the technology, the processing of natural fibre thermoplastic composites has been well explored (6, 7). However it is essential that the natural fibre reinforcement must be dried well before compounding with the polymer matrix in order to avoid degradation and formation of bubbles or voids. The drying process consumes a significant amount of energy and also exhibits a high safety risk owing to the highly flammable cellulose source. Several different approaches have been developed to overcome these problems, but each of them poses different challenges.

To overcome the problem of cellulose humidity and improve the mechanical properties and fire resistance of cellulose composites this paper presents a new method in which basic oxide filler is used. In principle, the humidity and wood acidity are absorbed or neutralized by adding basic oxides, such as calcium oxide, during processing. As a result, the drying of cellulose is not required and degradation is limited. Wood fibres and two different types of flax fibres were used in this work. One was obtained by hammer-milling the field-retted flax fibres, while the other was produced by bio-scouring the flax fibres after a mechanical decortication process.

## EXPERIMENTAL

Polypropylene Profax 1274 (PP) was purchased from Basell. Two coupling agents (CA) based on maleic anhydride (MA) grafted polypropylene, namely Epolene 43 (E43) ( $M_n = 9,100$ ; ~ 4.8 wt% of MA) from Eastman Chemicals and Polybond 3200 (PB3200) ( $M_n = 84,000$ ; ~ 1.0 wt% of MA) from Chemtura Corp., were used in this study. Spruce saw dust was provided by JER Envirotech (Canada). Two flax fibres were used throughout this study : (a) hammer-milled field-retted flax fibres (F1) were kindly provided by Schweitzer-Mauduit Canada and (b) mechanically decorticated flax fibres (F2) were kindly provided by Biolin Research Inc. However, the latter were treated further, that is the surface of these fibres was scoured enzymatically at 37°C and pH 8.5 using an engineered pectate lyase (8). Calcium oxide (CaO) and aluminum oxide ( $Al_2O_3$ ) were supplied by C. P. Hall Company and Malakof Industries Inc., respectively.

The wood composites were prepared with a twin-screw extruder Extrusion Spec W&P 30 mm having  $L/D = 40$  operated at a speed = 150-175 rpm and  $T_{max} = 185^\circ C$ . Samples were molded by injection at  $T = 200^\circ C$ . The flax fibre composites were fabricated on a Brabender Plasticorder at a speed of 60 rpm and  $T_{max} = 185^\circ C$ . All composite samples contained 40 wt% cellulose fibres. In addition, 2 wt% E43 was added in the wood composites while 2 wt% PB3200 was added in the flax fibre composites. The inorganic fillers were used at concentrations of 5 and 10 wt%. Transmission infrared spectra were measured on thin hot-pressed films using a Nicolet Magna 860 Fourier transform instrument at a resolution of  $4\text{ cm}^{-1}$ . A JEOL JSM-6100 scanning electron

microscope (SEM) at a voltage of 10 kV was utilized to observe the dispersion of the cellulose fibres in the PP matrix by examining polished surfaces. The cellulose fibre-PP matrix interface was examined on cryogenically fractured specimens using SEM. The limiting oxygen index (LOI) was measured using a Qualitest LOI analyzer according to ASTM D2863-97. Tensile properties were tested at room temperature on an Instron machine according to ASTM D638.

## RESULTS AND DISCUSSION

The effects of the basic fillers on the formulation were examined. The mechanical properties of the wood composites formulated with the same E43 coupling agent in the presence and absence of reactive fillers CaO and Al<sub>2</sub>O<sub>3</sub> are reported in Fig. 1. It is seen that the modulus and the strength of the wood composites increase significantly with the presence of reactive fillers. This apparent improvement could be due to the fact that during extrusion the reactive fillers interact with moisture and acid-like impurities in the wood, thus reducing the degradation/oxidation of the composites. In addition, the reactive fillers can react with the maleic anhydride group of the coupling agent, thus improving their interface with the PP matrix. As a result the reactive fillers can also be effective reinforcements for the PP matrix. CaO has a greater reinforcing effect on the composite properties than Al<sub>2</sub>O<sub>3</sub>; this may be due to its greater alkalinity and hence better reactivity with the coupling agent.

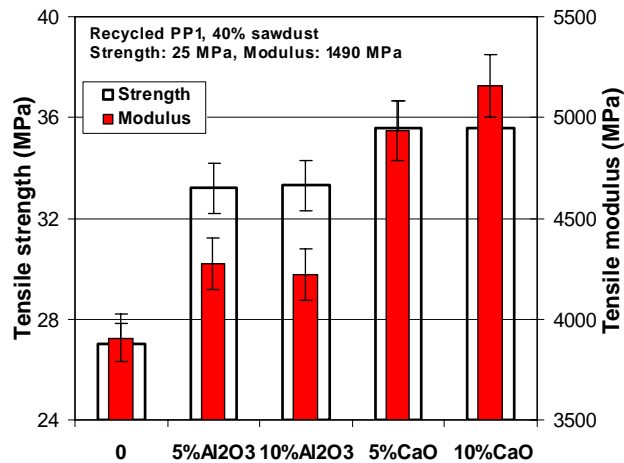


Fig. 1 Tensile properties of the wood composites fabricated with different reactive fillers.

The interaction between CaO and coupling agent was studied by means of FT-IR spectroscopy. Fig. 2a shows the spectra of thin films of PP, a blend of PP and E43, a blend of PP and CaO, and a blend of PP, E43, and CaO. Fig. 2b shows the difference spectra obtained from these by subtracting the PP spectrum. The difference spectrum of the CaO-E43-PP blend indicates that the peak at 1710 cm<sup>-1</sup>, assigned to hydrogen-bonded dimeric carboxylic acid groups of the maleic acid in the coupling agent E43, has disappeared and been replaced by a new peak at 1560 cm<sup>-1</sup>, assigned to calcium carboxylate. This proves that the maleic acid groups of the coupling agent can chemically react with the CaO particles.

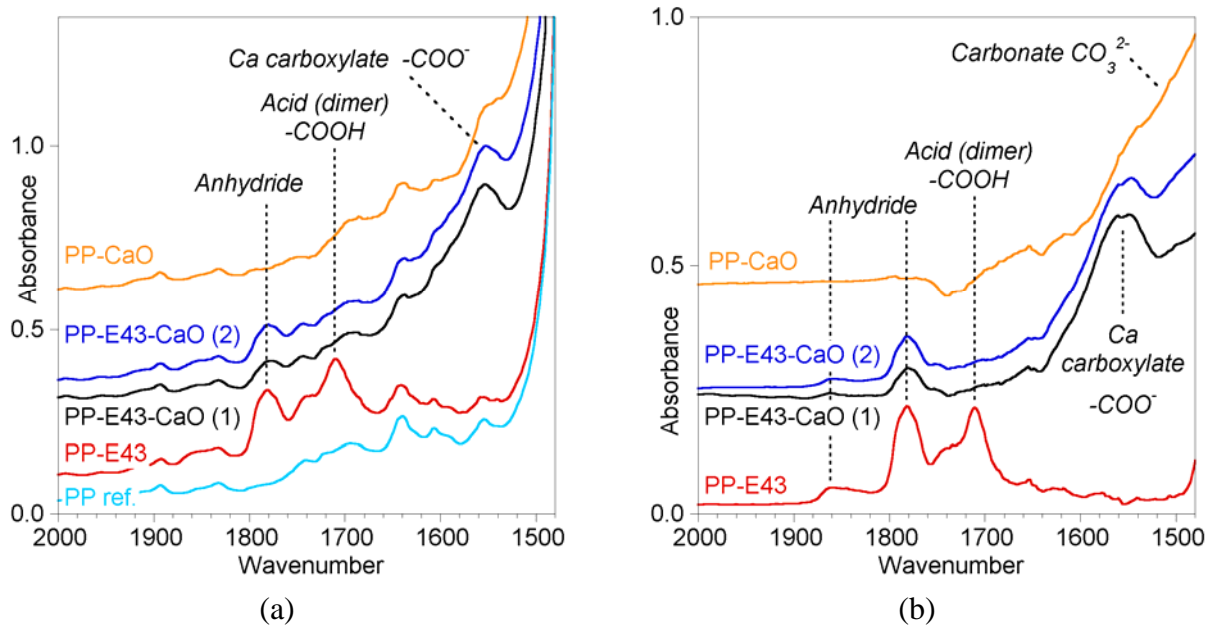


Fig. 2 FT-IR spectra of PP, a blend of PP and E43, a blend of PP and CaO, and a blend of PP, E43 and CaO: (a) original spectra as recorded; (b) difference spectra obtained by subtracting the PP spectrum.

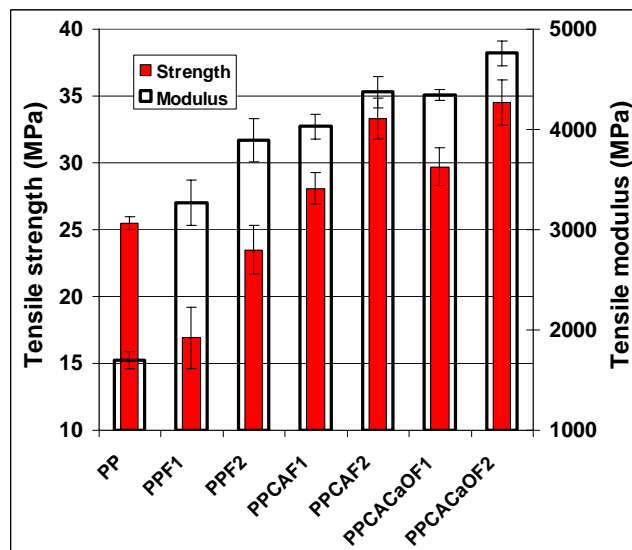


Fig. 3 Tensile properties of the flax PP composites.

The positive effect of CaO on the mechanical properties of flax fibre composites was also observed. Fig. 3 demonstrates the tensile strength and modulus of the composites fabricated from both types of flax fibres F1 and F2 in the presence and absence of coupling agent (CA) and CaO (10 wt%). The composites fabricated without CA have only greater modulus as compared to the PP matrix. However, the presence of CA results in significant further improvement in both the strength and modulus of the composites. Addition of CaO in the formulation results in the greatest strength and modulus. The enzyme-treated fibre (F2) exhibits the best reinforcing effect in all cases. This may be explained by the fact that the F2 with better retting (separation of fibre filaments) facilitated the fibre dispersion while its cleaner surface allows better interfacial

interaction between the fibre and the PP matrix. These factors determine the composite mechanical performance.

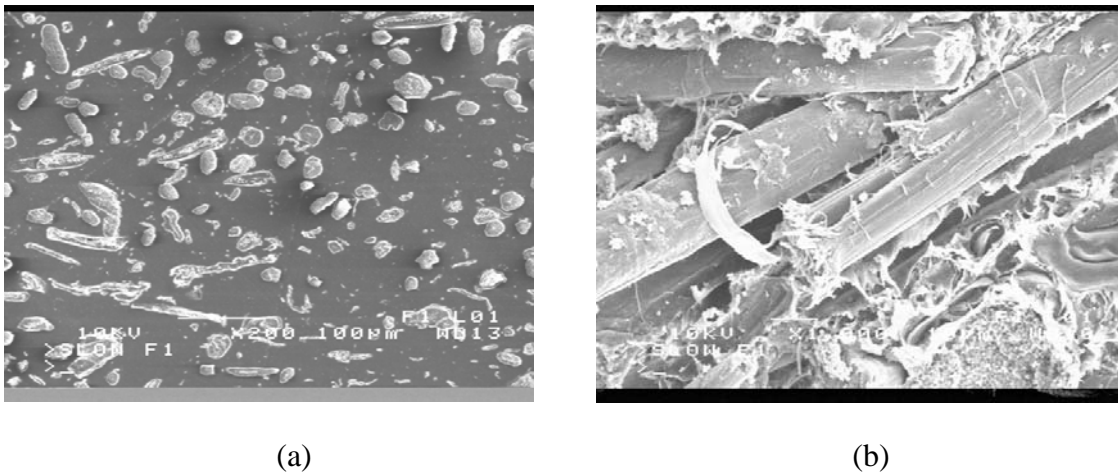


Fig. 4 SEM images of the flax fibre composites: (a) dispersion of enzyme treated fibres F2 (polished specimen) and (b) dispersion of CaO particles (fractured specimen).

The quality of dispersion of the flax fibres F2 in the PP matrix is very good. Fig. 4a illustrates the uniform distribution of the flax fibres F2 and shows that the fibres are well encapsulated by the matrix, without evidence of holes or voids. However, it was difficult to locate the CaO particles in this photo, so observations were performed on a cryogenically fractured specimen. Fig. 4b reveals the presence of fine CaO particles in the matrix and also a good fibre-matrix interface.

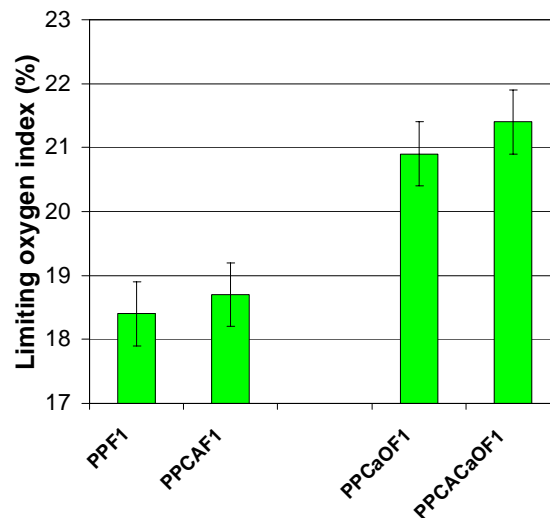


Fig. 5. Limiting oxygen index of flax PP composites.

The presence of CaO improved not only the mechanical properties but also the thermal stability and fire resistance of the cellulose composites. Fig. 5 illustrates the limiting oxygen index of the flax fibre - PP composites in the absence and presence of CA. The presence of CaO in the formulation significantly improves the fire resistance of the composites. This could be due to the fact that CaO can disperse the energy during burning. It is also possible that CaO reacted with impurities in the fibres, which often have low molecular weights and are thus more flammable.

## CONCLUSIONS

The results show that the presence of basic oxides combined with the use of coupling agent significantly improves the mechanical performance and fire resistance of the cellulose-based composites. These oxides absorb the humidity and neutralize the acidity of the cellulose reinforcement, thus limiting the degradation during compounding. It was also found that CaO is one of the best fillers for cellulose composites due to its high reactivity and low cost. The aid of the enzyme treatment (bio-scouring) also brings further improvement in composite performance.

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