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# Effect of lubricant on mechanical and rheological properties of compatibilized PP/sawdust composites



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#### ABSTRACT

An investigation was performed regarding the effect of compatibilizer and lubricant concentrations on the mechanical and rheological properties of polypropylene/sawdust composites using two-level factorial central composite design. Maleic anhydride grafted PP (PP-g-MA) was used as compatibilizer and the lubricant used was Struktol TPW 113. Composites without these additives were also prepared. Processing was carried out in a co-rotating twin-screw extruder coupled to a Haake rheometer. Mechanical properties were assessed from tensile tests and rheological properties were evaluated by oscillatory measurements and melt flow index (MFI) test. Tensile strength improved with increasing compatibilizer concentration in the composites and reached a maximum value in the absence of lubricant. When both additives were present tensile strength decreased, suggesting a possible interaction between lubricant and compatibilizer, reducing efficiency of the latter. The general trend observed for rheological results was a decrease in complex viscosity, storage modulus and MFI when both additives were present.

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### 1. Introduction

In the last decades, there has been increasing interest in sawdust filled polymer composites. This is due to the fact that this filler shows low abrasiveness in processing equipment in comparison to synthetic fibers. Moreover, sawdust is recyclable, biodegradable and can be incorporated in high concentration in polymeric matrices, resulting in great improvements in modulus (Bettini, Uliana, & Holzschuh, 2008; Correa, Razzino, & Hage, 2007; Kazayawoko, Balatinecz, & Matuana, 1999; Zafeiropoulos, Baillie, & Hodgkinson, 2002).

Polypropylene (PP) is one of the most frequently used polymers for the development of thermoplastic based composites. Its low melt temperature allows the composite processing at temperatures below 200 °C, which makes the degradation of lignocellulosic fibers less likely to occur (Morandim-Giannetti et al., 2012). However, the non-polar characteristic of the PP matrix results in an incompatibility with the polar sawdust filler. Thus, the addition of compatibilizer (Bettini et al., 2008; Keener, Stuart, & Brown, 2004; Oksman & Clemons, 1998; Suarez, Coutinho, & Sydenstricker, 2003) or coupling agents (Bettini, Bonse, Melo, & Munoz, 2010; Costa et al., 2000; Wu, Yu, Chan, Kim, & Mai, 2000) are of fundamental importance to reduce the interfacial stress and promote adhesion between phases. The most frequently used compatibilizer for the PP and sawdust system is the polypropylene grafted with maleic anhydride, PP-g-MA (Bettini et al., 2008, 2010; Correa et al., 2007; Harper & Wolcott, 2004; Keener et al., 2004; Morandim-Giannetti et al., 2012).

The wood plastic industry, due to economical reasons, is using expressive loading levels of sawdust, as high as 60 wt%. Thus, an adverse effect is the very high viscosity and, consequently, also very high pressures required for processing, which sometimes can become impracticable (Bettini et al., 2008; Li & Wolcott, 2004, 2005, 2006).

An alternative to overcome this difficulty is the addition of lubricants. The influence of some types of lubricants (ester based or stearate), used in HDPE/sawdust composites, were assessed through rheological and mechanical properties (Adhikary, Park, Islam, & Rizvi, 2011; Harper & Wolcott, 2004; Li & Wolcott, 2004, 2005, 2006; Santi, Hage, Vlachopoulos, & Correa, 2009; Yemele et al., 2013). In these works, there was evidence that the compatibilizer, which was maleic anhydride grafted polyethylene (PE-g-MA), acted not only as a compatibilizer, but also as an internal lubricant.

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Though, depending on the nature of the lubricant, the compatibilization effect may be reduced. Li and Wolcott (2006) compared the processability of HDPE/maple composites using different lubricant systems (i.e., zinc stearate-N,N'-ethylene bis-stearamide and ester-type lubricant) and no decrease in compatibilizer efficiency was observed when an ester-based lubricant was used. On the other hand, zinc stearate lubricant reduced PE-g-MA activity.

In a previous study, Bettini et al. (2008) concluded that conventional PP-g-MA concentrations in PP/sawdust composites, when in the presence of ester-based lubricants, are insufficient to achieve the required mechanical properties.

Santi et al. (2009) also noticed that the mechanical properties of the HDPE/sawdust composites can be reduced when ester-based lubricant and PE-g-MA compatibilizer are used simultaneously. According to the authors, HDPE/sawdust composites flow properties were strongly influenced by the presence of the lubricant, which caused the reduction in consistency index but increased the power-law exponent. At low shear rates, lower than  $200 \text{ s}^{-1}$ , the presence of LLDPE-MA (compatibilizer) in HDPE/sawdust composites increased the viscosity, however, when both additives (compatibilizer and lubricant) were incorporated in these composites, the viscosity was lower.

Yemele et al. (2013) investigated the effect of the level of the lubricants (ester-based), in presence or absence of compatibilizer (polyethylene grafted with maleic anhydride), on mechanical properties of HDPE/bark composites. The authors observed higher mechanical strength and modulus, and lower toughness and strain for the composites, when lubricant content was higher (2 wt%) and the compatibilizer was present. However, the effects of the presence of lubricant on composite compatibilization were not discussed in this study.

Leu, Yang, Lo, & Yang (2012) studied the influence of material composition (compatibilizer, lubricant and mass ratio of wood and PP) on the physical and mechanical properties of polypropylene/sawdust composites and they observed that appropriate content of compatibilizer can improve the mechanical properties and significantly reduce the swelling. According to the authors, the lubricant used (fatty acid metal soap) may increase water absorption and inhibit the bonding between sawdust and polymer because the lubricant might be located mainly at the interface between these materials.

The objective of this investigation is to assess the influence of an ester-based lubricant on the compatibilization of PP/sawdust composites, through statistical analysis of mechanical, rheological, morphological properties as well as spectroscopy.

#### 2. Experimental

#### 2.1. Materials

PP powder was supplied by Quattor, under code HP550K, with melt flow index (MFI) of 4.0 g/10 min. Antioxidant and lubricant incorporated to the PP were Irganox B215 and calcium stearate, respectively. These were also incorporated to all formulations at fixed amounts of 1500 ppm and 500 ppm, respectively. Irganox B215 is a mixture of a phenolic (Irganox 1010) and a phosphite (Irgafos 168) antioxidant supplied by Ciba Especialidades Químicas.

The compatibilizer used was PP grafted with maleic anhydride (PP-g-MA), under code Polybond 3200 (MFI = 110 g/10 min at 190 °C and 2.16 kg), supplied by Crompton–Uniroyal Chemical.

The lubricant used was an ester-type lubricant agent, under code Struktol TPW 113, supplied by Parabor.

The sawdust from Pine (Mesh 80) was supplied by Pinhopó.

#### Table 1

Compositions of the PP/sawdust composites.

Test	C <sub>PP</sub> (%)	C <sub>S</sub> (%)	C <sub>C</sub> (%)	$C_{\text{TPW}}$ (%)
1	63	30	6	1
2	61	30	8	1
3	62	30	6	2
4	60	30	8	2
5	61.5	30	7	1.5
6	61.5	30	7	1.5
7	62	30	0	0
8	70	30	6	0
9	64	30	8	0
10	69	30	0	1
11	68	30	0	2
12	100	0	0	0

 $C_{PP}$ , polypropylene concentration;  $C_S$ , sawdust concentration;  $C_c$ , compatibilizer concentration (PP-g-MA) and  $C_{TPW}$ : lubricant concentration (Struktol TPW 113).

#### 2.2. Methods

#### 2.2.1. Preparation of PP/sawdust composites

Prior to extrusion the sawdust was dried in an oven at  $100 \,^{\circ}$ C for 16 h and conditioned in aluminized bags containing moisture and light barrier.

Composite components were weighed and pre-mixed in a Mecanoplast mixer. Formulations were prepared according to a central composite experimental design, where the variables compatibilizer content (PP-g-MA) and lubricant content (Struktol TPW 113) were varied. The experimental design is shown in Table 1 (formulations 1–6). For comparison, composites were also prepared without addition of lubricant (formulations 8 and 9), without the addition of compatibilizer (formulations 10 and 11), as well as without both lubricant and compatibilizer (formulation 7). A sample of neat polypropylene (formulation 12) was also prepared, as shown in Table 1.

The formulations were prepared in a co-rotating twin-screw extruder coupled to a Haake torque rheometer, at the following conditions: temperature profile: 170 °C, 175 °C, 180 °C, 180 °C, 190 °C and 185 °C; screw rotation: 250 rpm.

The composites were injection molded into tensile specimens according to ASTM D638, using a HM 60/350 Battenfeld Injection Molding GmbH molding machine at a pressure of 80 bar, using a temperature profile of 190 °C, 190 °C, 190 °C and 195 °C in zones 1, 2, 3 and nozzle, respectively.

#### 2.2.2. Mechanical properties

Tensile tests were conducted in a 5567 Instron Universal Testing Machine, according to ASTM D638, at test speed of 5 mm/min.

#### 2.2.3. Torque rheometry

In order to investigate the interaction between the compatibilizer (PP-g-MA) and lubricant (TPW), four formulations without sawdust were studied. Two formulations were control, containing PP/PP-g-MA (88/12) or PP/TPW (96/4), and two had different compatibilizer/lubricant ratios (PP/TPW/PP-g-MA, 86/12/2 and 84/12/4). The amount of compatibilizer and lubricant were higher than those used in the composite study, to allow identification by FTIR spectroscopy. These samples were analyzed by torque rheometry and FTIR spectroscopy in order to assess the kind of interaction between the two compounds. The compositions were obtained in a Haake torque rheometer, equipped with a Rheomix 600p mixing chamber, at the following process conditions: temperature 180°C, rotor frequency 50 rpm and reaction time 10 min. After processing in the rheometer, the samples were pressed into films in a hot press at 180 °C and 100 kgf/cm<sup>2</sup> and were then submitted to heat treatment at 125 °C for 24h to convert succinic acid groups into anhydride groups.

Table 2		
Tensile test and	MFI	results.

Test	C <sub>PP</sub> (%)	C <sub>S</sub> (%)	C <sub>C</sub> (%)	C <sub>TPW</sub> (%)	Tensile strength (MPa)	Elongation at break (%)	MFI (g/10 min)
1	63	30	6	1	$32.15\pm0.64$	$2.76\pm0.29$	1.14
2	61	30	8	1	$36.32\pm0.26$	$2.99\pm0.24$	1.27
3	62	30	6	2	$34.33\pm0.24$	$3.19\pm0.20$	1.30
4	60	30	8	2	$31.16\pm0.62$	$3.24\pm0.43$	1.40
5	61.5	30	7	1.5	$33.41\pm0.30$	$3.11 \pm 0.26$	1.22
6	61.5	30	7	1.5	$32.12\pm0.44$	$3.22\pm0.48$	1.24
7	62	30	0	0	$30.55 \pm 0.71$	$2.57 \pm 0.40$	1.02
8	70	30	6	0	$40.35\pm0.78$	$4.53\pm0.48$	1.01
9	64	30	8	0	$38.68 \pm 0.36$	$4.28\pm0.51$	1.10
10	69	30	0	1	$28.90\pm0.42$	$5.40\pm1.07$	1.05
11	68	30	0	2	$27.91 \pm 0.12$	$4.28\pm0.78$	2.07
12	100	0	0	0	$31.42\pm0.3$	>500	1.59

#### 2.2.4. FTIR spectroscopy

Transmittance analyses were performed in a Nicolet 6700 FTIR Spectrophotometer, with resolution of 2 cm<sup>-1</sup> and 128 scans per spectrum. Analyses were done in triplicate.

#### 2.2.5. Rheological properties

The oscillatory rheological characterization was performed in an Anton Paar rheometer with 25 mm parallel plates fixture and 1 mm gap between plates. Oscillatory dynamic measurements were carried out at constant temperature of 190 °C and within the linear viscoelastic range. A constant strain of 0.2% was determined through dynamic strain sweep test, at the frequency of 1 rad/s, and was applied for all samples. The dynamic rheological properties: storage modulus, *G'*, loss modulus, *G''* and complex viscosity,  $\eta^*$ , were recorded as a function of angular frequency in the range of 0.01–100 rad/s (dynamic frequency sweep test). Melt flow index test was performed in a DSM – MI-1 Plastometer equipment, at 190 °C and a load of 2.16 kg. This measurement was not carried out according to the temperature specified in ASTM D-1238 to avoid degradation of the lignocellulosic fibers, which, in general, occurs above 200 °C.

#### 3. Results and discussion

The results obtained from tensile testing (tensile strength and elongation at break) are presented in Table 2. It can be observed that the addition of sawdust, without the presence of compatibilizer or lubricant (comparing formulation 7–12), resulted in a decrease in both properties. This is an effect of the incompatibility between polypropylene and sawdust. Whilst the former is non-polar, the latter is polar, resulting in lack of adhesion between them.

With the introduction of lubricant in the polymer/sawdust system (formulations 10 and 11), tensile strength also reduces. Such



**Fig. 1.** SEM micrographs of the fractured surfaces in the tensile tests of the samples (a) polypropylene/30% sawdust (PP/SD) composites without compatibilizer (b) and with compatibilizer, both in absence of lubricant, (c) with lubricant and (d) with compatibilizer and lubricant.



**Fig. 2.** Effect of the variables compatibilizer (PP-g-MA) and lubricant (Struktol TPW 113) concentrations on tensile strength of the PP/sawdust composites.

behavior is expected, since the lubricant does not act as a compatibilizer. Furthermore, the remaining lubricant in the polymeric matrix after processing may contribute to the decrease in composite strength due to its low molecular weight and its incompatibility with PP matrix.

On the other hand the introduction of compatibilizer, PPg-MA, in the composites (formulations 8 and 9) results in a noticeable increase in tensile strength in comparison to the noncompatibilized composite. However, for concentrations of PP-g-MA above 6 wt%, this trend is not observed, suggesting that this is the optimum compatibilizer concentration for the system analyzed in this work. One of the most advantageous aspect of PP-g-MA as compatibilizer for polypropylene systems is that its main chain is identical to the composite matrix and, consequently, compatible. Moreover, maleic anhydride grafted chains can react or interact with the sawdust hydroxyls, achieving appropriate interfacial adhesion between these phases. The unexpected decrease in tensile strength with the increase in compatibilizer to 8 wt% might an indication that this additive has reached a limit in the system and is in excess, leading to the formation of an extra PP-g-MA phase, thus reducing its efficiency.

These results indicate that the introduction of compatibilizer in these composites is crucial for the achievement of high tensile strength. However, with the addition of lubricant to the compatibilized system, a reduction in tensile strength is noted (formulations 1–6). This suggests that the lubricant might be interacting with the compatibilizer and reduces its efficiency.

Fig. 1 presents micrographs of the PP/sawdust (PP/SD) composites without (a) and with compatibilizer (b), in absence of lubricant, and with lubricant (c) and with compatibilizer and lubricant (d). These SEM images corroborate the above analyses, in which the fibers are seen to be well adhered to the PP matrix, ruptured flush with the polymer surface (b), whereas fibers are sticking out from the PP in the noncompatibilized composites, indicating poor fiber-matrix adhesion (a). Moreover, when the lubricant is present in PP/sawdust composites, with (d) or without (c) compatibilizer, the adherence between sawdust and PP is reduced and the composites present poor sawdust distribution and the fibers are pulled out from matrix, mainly for composites with both, PP-g-MA and lubricant.

The response surface plot of the influence of the compatibilizer and lubrificant concentrations on tensile strength is presented in Fig. 2. As can be seen in Fig. 2 and Table 2, each variable affects the tensile strength. For instance, at the lower lubricant level, increasing the amount of compatibilizer improves tensile strength. However, an opposite trend is observed at the higher level of lubricant. Despite the assumption that the lubricant might reduce compatibilization efficiency, the higher its amount, the greater are the chances of anhydride or acid groups to interact with sawdust hydroxyl groups. The lowest tensile strength of the composite occurs when both additives are at their highest level (i.e., 8 wt% compatibilizer and 2 wt% lubricant). As mentioned earlier in this text, there is evidence that when at high concentration levels, the compatibilizer may form a distinct phase in the composite, decreasing compatibilization efficiency. To comfirm this behavior, SEM analyses are shown in Fig. 3, where samples a and b present PP composites containing 30 wt% sawdust and 8 wt% PPg-MA, with 1 wt% lubricant (a) and 2 wt% lubricant.

Therefore, if less compatibilizer becomes available, because of the formation of a new phase, and if lubricant concentration is at its highest level, thus increasing the chances of reaction or interaction between lubricant and compatilizer, then a decrease in compatibilization should be expected. This may be a result of the reduced availability of the anhydride and/or acid group of the PP-g-MA to interact with the sawdust hydroxyl groups, which might explain the low tensile strength shown by formulation 4.



Fig. 3. SEM micrographs of the fractured surfaces in the tensile tests of the samples: (a) polypropylene/30 wt% sawdust/8 wt% PP-g-MA/1 wt% lubricant and (b) polypropylene/30 wt% sawdust/8 wt% PP-g-MA/2 wt% lubricant.



Fig. 4. Torque curves obtained for PP/TPW/PP-g-MA with different concentrations: (88/0/12, 96/4/0, 86/12/2, 84/12/4).

#### 3.1. Analysis of the interaction between the additives

Torque rheometry was used as a tool for obtaining information about possible reactions and/or interaction between compatibilizer and lubricant. As mentioned previously, two control formulations, containing PP/PP-g-MA or PP/TPW, and two formulations presenting different compatibilizer/lubricant ratios were processed in a torque rheometer. Fig. 4 presents the torque curves obtained.

Analysis of the control samples shows that the presence of lubricant, sample (4/0) (TPW/PP-g-MA), reduces the maximum torque. The torque curve presents two maximum peaks probably due to material fusion and then lubricant dispersion in the molten matrix. The sample containing only compatibilizer, TPW/PP-g-MA (0/12), presents only one peak, related with material melting. Moreover, it was observed that when compatibilizer and lubricant are added together, the torque curves are a function of the ratio between the components. It was observed that when compatibilizer/lubricant ratio is lower (sample 2/12), the second peak related to lubricant dispersion, disappears probably due to some strong interaction between the two components. For higher ratio of components (sample 4/12) the second peak is present; however, it is less intense than in the control sample. The reduction in the peak of sample (4/12) and its disappearance in sample (2/12) are likely related to the number of succinic groups which interacted with lubricant chain. If the second peak of the lubricant is related to its dispersion as a second phase in the system, the compatibilizer may interact with the lubricant to form a single phase, thus the lubricant does not disperse in the medium. At the other hand if there are not enough succinic groups to interact with TPW functional groups the second phase and a second peak is observed during processing.

The change in the torque curve during processing, as well as the mechanical results, indicate there is some interaction between the compatibilizer and lubricant. This interaction may be physical or chemical. To evaluate the possible reactions or interactions the samples were analyzed by FTIR spectroscopy. Fig. 5 shows the FTIR spectra obtained for samples containing compatibilizer and lubricant in which only one peak is observed at 1792 cm<sup>-1</sup>, characteristic of carbonyls from five-membered cyclic anhydrides (Silverstein, Webster, & Kiemle, 2005, chap. 2), when the composition is 100/0/12, PP/TPW/PP-g-MA, another bound appears at 1740 cm<sup>-1</sup>, characteristic of carbonyls from ester groups (Silverstein et al., 2005, chap. 2), when composition is 100/4/0, PP/TPW/PP-g-MA.



Fig. 5. FTIR spectra obtained for PP/TPW/PP-g-MA with different concentrations: (88/0/12, 96/4/0, 86/12/2, 84/12/4).

When both additives, lubricant and compatibilizer, are present in the mixture, compositions 100/2/12 and 100/4/12, the peak at 1792 cm<sup>-1</sup> reduces. This band is characteristic of succinic groups presents in PP-g-MA, therefore there is evidence that the lubricant interacts with succinic groups. At the other hand, there is no change in the 1740 cm<sup>-1</sup> band, characteristic of ester groups from the lubricant. In fact there is no known reaction between organic anhydrides and esters, thus it is most likely there are some side reactions between the compatibilizer and other groups present in the lubricant structure that consume the succinic groups. As a consequence the compatibilizer capacity of PP-g-MA is reduced and this explains why the mechanical properties are reduced when TPW lubricant is added to system. Unfortunately the complete TPW structure is not known, the only information is that material is an ester-based lubricant (blend of complex, modified fatty acid ester), i.e., it may have some other functional groups along the chain that might react with the succinic groups of the compatibilizer.

In conclusion, the presence of lubricant (TWP) may lead to reduction in mechanical properties due to a reaction or interaction with succinic groups of compatibilizer (PP-g-MA). The succinic groups are responsible for the interaction between the PP matrix and the fibers, hence its number reduced the interaction is reduced and mechanical behavior is reduced. In order to reduce the influence of lubricant on final interaction of filler and matrix either, a higher amount of PP-g-MA must be added to composites or the amount of TPW must be reduced. Both options are valid, but the increase of PP-g-MA may reduce mechanical properties due to its low molar mass, and the reduction in TPW content may impair composite processing, principally when high amounts of fiber are used.

#### 3.2. Rheological analysis

The influence of compatibilizer and the lubrificant contents on the rheology of the PP/sawdust composites was also investigated in terms of complex viscosity and storage modulus (Fig. 6). The oscillatory measurements give information about both elastic and viscous properties and hence can provide a detailed characterization of the dispersion and interface than other flow measurements. It can be seen that the addition of sawdust increase both the complex viscosity and storage modulus. This is an effect of the particle–particle contact which is most likely to occur in this system containing 30 wt% of the low apparent density sawdust. Nevertheless, when lubricant is added to the PP/sawdust system, in absence of PP-g-MA, complex viscosity and storage modulus values remained close



**Fig. 6.** Effect of the variables compatibilizer content (%PP-g-MA) and the lubrificant content (%TPW) on the complex viscosity ( $\eta^*$ ) and storage modulus (G') of the PP/sawdust.

to the values of PP/sawdust composites without additives. Complex viscosity and elastic modulus are higher in the presence of compatibilizer, mainly at lower frequency. This behavior can be explained by the increased adhesion at the interface, provided by the compatibilizer.

The rheological analysis of the PP/sawdust composites with both additives is more complex. Complex viscosity and storage modulus were lower in relation to those composites, containing one or no additives. In this case, no synergistic effect of the additives in reducing the viscoelastic properties was observed, which can be explained as a consequence of the decrease in interfacial tension and interparticle contact. Similar results were also observed by Li and Wolcott (2006). Furthermore, there is no evidence of the effect of the level of these additives on these properties. However, it can be affirmed, with statistical significance of 5%, that the presence or absence of these additives do interfere with the complex viscosity and storage modulus.

In order to assess the effect of the additives on the viscosity of the composites at higher deformation (beyond the linear viscoelastic range), melt flow index (MFI) analysis was accomplished. The results are presented in Table 2. These measurements show that the presence of either additives, compatibilizer or lubricant, cause an increase in the MFI. Moreover, when both additives are present, the lubricating action is improved and an increase in the MFI (lower viscosity) is achieved. This behavior can be explained by the effect of internal lubrication of these additives in PP/sawdust composites because the compatibilizer presents a reduced molecular weight in relation to PP and TPW is a lubricant suitable for use in composites reinforced with natural fibers due to its compatibility. As can be clearly seen, the MFI results are in agreement with those observed for oscillatory rheology at higher frequency. With the increase of the concentration of either compatibilizer or lubricant, an increase on the MFI can be observed. Moreover, when both additives are present in the composite, this increase becomes even more evident.

#### 4. Conclusions

The effect of compatibilizer, PP-g-MA, and lubricant, Struktol TPW 113, contents on the mechanical and rheological properties of polypropylene/sawdust composites was assessed. PP-g-MA showed to act both as compatibilizer and internal lubricant. Increasing concentration improve tensile strength in the composites, which reached a maximum value in the absence of lubricant. When both additives are present, tensile strength decreased, suggesting a possible interaction between lubricant and compatibilizer, reducing the efficiency of the latter. The general trend observed for rheological results was a decrease in complex viscosity, storage modulus and MFI in the presence of both additives. This reduction in properties is most noticeable, probably due to the reduction in interparticle interaction and interfacial tension.

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#### References

- Adhikary, K. B., Park, C. B., Islam, M. R., & Rizvi, G. M. (2011). Effects of lubricant content on extrusion processing and mechanical properties of wood flour–highdensity polyethylene composites. *Journal of Thermoplastic Composite Materials*, 24, 155–171.
- Bettini, S. H. P., Bonse, B. C., Melo, E. A., & Munoz, P. A. R. (2010). Effect of sawdust surface treatment and compatibilizer addition on mechanical behavior, morphology, and moisture uptake of polypropylene/sawdust composites. *Polymer Engineering and Science*, 50, 978–985.
- Bettini, S. H. P., Uliana, A. T., & Holzschuh, D. (2008). Effect of process parameters and composition on mechanical, thermal, and morphological properties of polypropylene/sawdust composites. *Journal of Applied Polymer Science*, 108, 2233–2241.
- Correa, C. A., Razzino, C. A., & Hage, E. (2007). Role of maleated coupling agents on the interface adhesion of polypropylene–wood composites. *Journal of Thermoplastic Composite Materials*, 20, 323–339.
- Costa, T. H. S., Carvalho, D. L., Souza, D. C. S., Coutinho, F. M. B., Pinto, J. C., & Kokta, B. V. (2000). Statistical experimental design and modeling of polypropylene–wood fiber composites. *Polymer Testing*, 19, 419–428.
- Harper, D., & Wolcott, M. (2004). Interaction between coupling agent and lubricants in wood-polypropylene composites. Composites Part A: Applied Science and Manufacturing, 35, 385–394.
- Kazayawoko, M., Balatinecz, J. J., & Matuana, L. M. (1999). Surface modification and adhesion mechanisms in woodfiber–polypropylene composites. *Journal of Materials Science*, 34, 6189–6199.
- Keener, T. J., Stuart, R. K., & Brown, T. K. (2004). Maleated coupling agents for natural fibre composites. *Composites Part A: Applied Science and Manufacturing*, 35, 357–362.
- Leu, S.-Y., Yang, T.-H., Lo, S.-F., & Yang, T.-H. (2012). Optimized material composition to improve the physical and mechanical properties of extruded wood–plastic composites (WPCs). *Construction and Building Materials*, 29, 120–127.
- Li, T. Q., & Wolcott, M. P. (2004). Rheology of HDPE–wood composites. I. Steady state shear and extensional flow. Composites Part A: Applied Science and Manufacturing, 35, 303–311.
- Li, T. Q., & Wolcott, M. P. (2005). Rheology of wood plastics melt. Part 1. Capillary rheometry of HDPE filled with maple. *Polymer Engineering and Science*, 45, 549–559.
- Li, T. Q., & Wolcott, M. P. (2006). Rheology of wood plastics melt. Part 2. Effects of lubricating systems in HDPE/maple composites. *Polymer Engineering and Science*, 46, 464–473.
- Morandim-Giannetti, A. A., Agnelli, J. A. M., Lancas, B. Z., Magnabosco, R., Casarin, S. A., & Bettini, S. H. P. (2012). Lignin as additive in polypropylene/coir composites: Thermal, mechanical and morphological properties. *Carbohydrate Polymers*, 87, 2563–2568.

Oksman, K., & Clemons, C. (1998). Mechanical properties and morphology of impact modified polypropylene–wood flour composites. *Journal of Applied Polymer Sci*ence, 67, 1503–1513.

- Santi, C. R., Hage, E., Vlachopoulos, J., & Correa, C. A. (2009). Rheology and processing of HDPE/wood flour composites. *International Polymer Processing*, 24, 346–353.
- Silverstein, R. M., Webster, F. X., & Kiemle, D. J. (2005). Spectrometric identification of organic compounds (7th ed.). Hoboken, NJ: Wiley.
- Suarez, J. C. M., Coutinho, F. M. B., & Sydenstricker, T. H. (2003). SEM studies of tensile fracture surfaces of polypropylene-sawdust composites. *Polymer Testing*, 22, 819–824.
- Wu, J. S., Yu, D. M., Chan, C. M., Kim, J. K., & Mai, Y. M. (2000). Effect of fiber pretreatment condition on the interfacial strength and mechanical properties of wood fiber/PP composites. *Journal of Applied Polymer Science*, 76, 1000–1010.
- Yemele, M. C. N., Koubaa, A., Cloutier, A., Soulounganga, P., Stevanovic, T., & Wolcott, M. P. (2013). Effects of hot water treatment of raw bark, coupling agent, and lubricants on properties of bark/HDPE composites. *Industrial Crops and Products*, 42, 50–56.
- Zafeiropoulos, N. E., Baillie, C. A., & Hodgkinson, J. M. (2002). Engineering and characterisation of the interface in flax fibre/polypropylene composite materials. Part II. The effect of surface treatments on the interface. *Composites Part A: Applied Science and Manufacturing*, 33, 1185–1190.