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Mihai, Mihaela; Stoeffler, Karen

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LOWER COST, LOWER WEIGHT, AND GREENER POLYPROPYLENE BIOCOMPOSITES FOR AUTOMOTIVE APPLICATIONS

M. Mihai, K. Stoeffler

Polymer Bioproducts, Automotive and Surface Transportation,
National Research Council Canada, Boucherville, Qc., Canada
Mihaela.Mihai@cnrc-nrc.gc.ca, Karen.Stoeffler@cnrc-nrc.gc.ca

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Summary: *This paper discloses methods to produce sustainable blends and composites based on polypropylene (PP) as viable eco-solutions for automotive interior applications. Different biomaterials based on PP were prepared, containing up to 50 wt.% of renewable content (cellulosic fibers and/or polylactide). These biomaterials were evaluated in terms of morphology, mechanical, and thermal properties, as well as for cost and weight reductions. The tensile strength, tensile modulus, and heat deflection temperature presented at least equivalent values comparing to neat PP and to commercial PP compounds currently used in automotive interior parts. Foamed parts, obtained from these biocomposites through foam injection molding process, presented at least similar properties as unfoamed and commercial grades while being up to 25 wt.% lighter, 33 % less expensive, and 50 wt.% greener.*

1 INTRODUCTION

The current worldwide automotive industry orientates toward the use of environmental friendly raw materials and fabrication processings to produce greener parts having lower weight and lower cost while reducing the cars fuel consumption and maintaining their safety. In this spirit, the automotive manufacturers look at the implementation of lighter and less expensive thermoplastic polymer parts and of lower-cost thermoplastic biocomposites containing bio-sourced materials. Thermoplastic bioblends and biocomposites seem to have high potential to act as partial or complete substitutes of petroleum thermoplastics and composites or other heavy and expensive current used automotive parts [1].

About 180 kg of thermoplastics are used in one car, which represents 12 % of the car weight and 50 % of the car volume. Most of these thermoplastics are actually fiberglass-reinforced composites or mineral-filled compounds [1, 2]. Amongst them, polypropylene (PP), nylons (PA6, PA66) and styrenics (such as ABS) are the most exploited polymers in the fabrication of thermoplastic automotive parts. PP, the most commonly used resin for injection-molding resin in 2014, has a global demand estimated at 33 million tons per year [3]. PP utilization in automotive manufacture, it accounts for 80 kg per vehicle and is mostly used to manufacture dashboards, central consoles, headliners, front/end modules, IP carriers, tubs in floor, and many other parts. There are several studies on PP / cellulosic biocomposites in the literature. It was proven that the mechanical properties of PP / cellulosic biocomposites

depend on the type of cellulosic source [4-8], fiber treatment [9, 10], fiber / polymer affinity [8, 11, 12] and processing conditions [13]. On the other hand, improvements in mechanical properties of PP / PLA blends and PP / PLA / cellulosics biocomposites were obtained when adequate coupling agents and processing parameters were considered [14-16]. A recent study, that gives priority to the environmental factors and sustainability, shows that PP / hemp fiber biocomposites gained the higher rank in the selection process and can be recommended to automotive component manufacturers to support greener technologies [17]. On another hand, the use of kenaf fibers as alternative reinforcement material in the polymer composites, proved to reduce the environmental impact throughout the product life cycle towards achieving better sustainable performance of the product compared to similar component using glass fiber composites [18, 19].

In this paper, we use the following strategies to improve the environmental performance of PP based automotive interior parts:

- Replacement of mineral-filled PP or glass fiber reinforced PP by PP biocomposites containing cellulosic fibers, to allow weight and cost reductions;
- Substitution of a part of PP matrix by a bioplastic, to further increase the renewable content;
- Use of injection foaming process, to further reduce weight and cost of PP bioblends or biocomposites;
- Combination of these strategies.

Using these strategies, sizeable cost and weight reductions were achieved using PP biomaterials. When cellulosic fibers replaced up to 40 wt.% of PP, around 33 % cost reduction was reached. Up to 25 % lighter parts can be produced by adapting the injection foaming process to PP biocomposite behavior. Combinations of PP with cellulosic fibers and PLA further allow the fabrication of biomaterials with up to 50 wt.% renewable content while preserving significant cost and weight reductions. These environmental friendly PP biomaterials present at least equivalent performances comparing to currently used PP compounds and PP composites and should be considered as eco-solutions in the manufacturing of new automotive interior parts.

2 EXPERIMENTAL PART

2.1 Materials

The PP used in this work was Pro-fax 6323 from Lyondell Basell, a general-purpose homopolymer for injection molding applications. The properties of four PP automotive commercial grades were used as references as follows:

- Two mineral-filled PP from ACLO Compounders Inc.: Accutech 20L (PP filled with 20 wt.% talc) and Accutech 40L (PP filled with 40 wt.% talc).
- Two glass fibers (GF) reinforced PP from Plastics Group of America: Polifil GFPP-20 (PP reinforced with 20 wt.% GF) and Polifil GFPP-40 (PP reinforced with 40 wt.% GF).

PLA 8302D, an amorphous grade from Nature Works, was selected as the bio-sourced minor phase polymer for the production of PP / PLA bioblends and biocomposites. The cellulosic fibers used for biocomposites preparation were: short flax fibers supplied by Schweitzer Mauduit Canada, thermo-mechanical pulp (TMP) supplied by Papier Masson / White Birch, and wood fibers in form of dices (WoodForce - WF) supplied by Sonae Industria. The flax fibers and the TMP fibers have very low bulk densities and were pelletized before compounding to ensure a consistent flow rate during the extrusion. The cellulosic fiber concentrations used to produce the biocomposites were 20 wt.% and 40 wt.%.

The physical aspects of the cellulosic fibers used in this work are presented in Figure 1. Glass fibers of 6.5 mm in length were also used to produce hybrid composites with the purpose to asset the reinforcement potential of the cellulosic fibers. Appropriate coupling agents were used in PP / cellulosics, PP / PLA bioblend and PP / PLA / cellulosic formulations. All raw materials were dried for 24 hours before extrusion at appropriate temperatures.

Figure 1. Physical aspects of A) Flax, B) TMP, C) WF fibers, and D) short glass fibers as used in the compounding process.

2.2 Processing

2.2.1 Extrusion Process

The extrusion line operated to compound the PP biomaterials was a Leistritz 34 mm co-rotating twin-screw extruder with 12 mixing zones and L/D ratio of 40. Two feeding locations were available: polymers and additives were fed in the first zone while cellulosic fibers were incorporated at mid-extruder. The screw configuration was specifically designed in our laboratory with the purpose to avoid as much as possible the attrition of the fibers during the compounding while warranting a very good dispersion and distribution through the polymer melt. At the extrusion line exit, a capillary die of 2 mm in diameter was used. The temperature was set at 180 °C all along the extrusion line.

2.2.2 Injection molding and injection foaming

The compounded pellets were dried at 80 °C for 24 hours and then injection molded using a 34 tons BOY injection molding press. The injection barrel and mold temperatures were around 190-200°C. Standard specimens were molded according to ASTM D638 and ASTM D259 for tensile and Izod impact properties evaluation, respectively. 1 wt.% of Hydrocerol 1514 from Clariant was used as chemical blowing agent (CBA).

2.3 Characterization

2.3.1 Morphology

Scanning electron microscopy (SEM) was carried out on impact fractured biocomposite surfaces. A coating of gold / palladium alloy was applied on the specimens prior to the observation. A JEOL JSM-6100 SEM at a voltage of 10 kV was used.

2.3.2 Mechanical Properties

Tensile testing was carried out according to ASTM D638 at a rate of 5 mm/min on standard type I dog-bone specimen. The tensile modulus (TM) and the tensile strength (TS) were measured. A video extensometer was used to determine the tensile modulus. The Izod impact strength (IS) was measured according to ASTM D256 using notched specimens and a 2 kg hammer. All reported values are the average of at least five tests.

2.3.3 Heat Deflection Temperature

The heat deflection temperature (HDT) was measured using an Instron Ceast HDT-3-Vicat device according to ASTM D648. A rectangular bar was tested in the edgewise position as a simple beam with a 0.455 MPa load applied at its center. The loaded specimen was immersed in a heat-transfer medium which temperature rose by 2 °C/min. The HDT under flexural load was recorded as the temperature of the medium at which the test bars deflected by 0.25 mm.

3 RESULTS AND DISCUSSIONS

3.1 Morphology of PP biocomposites

Figure 2 discloses the morphology of transversal fractured surfaces of biocomposite specimens resulted after the Izod-impact testing. The micrographs correspond to PP biocomposites containing 20 wt.% flax, without and with coupling agent at three different magnifications (x100, x500 and x1000). All other PP biocomposites obtained in this work presented similar morphological aspects of their fractured surfaces and, for straightforwardness, were not presented here.

The micrographs at lower magnification disclosed cellulosic fibers uniformly distributed within the corresponding matrices. This indicated an excellent dispersion and distribution of the cellulosic fiber pellets in the polymer matrix due to the dynamics created by the screw configuration specially designed for this work in our laboratory. The fractured surfaces of the biocomposites without coupling agents (left column) displayed significant pullout of cellulosic fibres. Moreover, the fractures were produced at the cellulosic fiber / matrix interfaces, which indicate poor adhesion due to the absence of the coupling agent. Less cellulosic fiber pullout (right column) was observed when a coupling agent was incorporated in the biocomposite formulation. In this case, the cracks were propagated predominantly through the matrix and through cellulosic fibers themselves because of the enhanced fiber-matrix interfacial strength. These observations confirmed an upgraded polymer / cellulosic fibers adhesion in the compatibilized system.

3.2 Low cost biocomposites based on PP

Replacement of a part of thermoplastic matrix by cellulosic fibers

Figure 3 discloses a comparison of mechanical properties and relative costs of neat PP, PP / 20 wt.% cellulosic biocomposites developed in this work and a commercial PP / 20 wt.% talc. The tensile strengths of PP / 20 wt.% flax, PP / 20 wt.% TMP and PP / 20 wt.% WF had values between 34 and 38 MPa which were up to 15-30 % higher than the neat PP (30 MPa) and at least equal comparing at PP / 20 wt.% talc commercial grade (35 MPa). The tensile modulus of biocomposites presented up to 200 % higher values (3000-3400 MPa) compared to virgin PP (1600 MPa) and PP commercial grade (1800 MPa). The Izod impact strength of

biocomposites was also increased, while the elongation at break decreased as anticipated due to the incorporation of cellulosic fibers. Impact modifiers are recommended strategies to be applied further to increase the impact properties of those biocomposites.

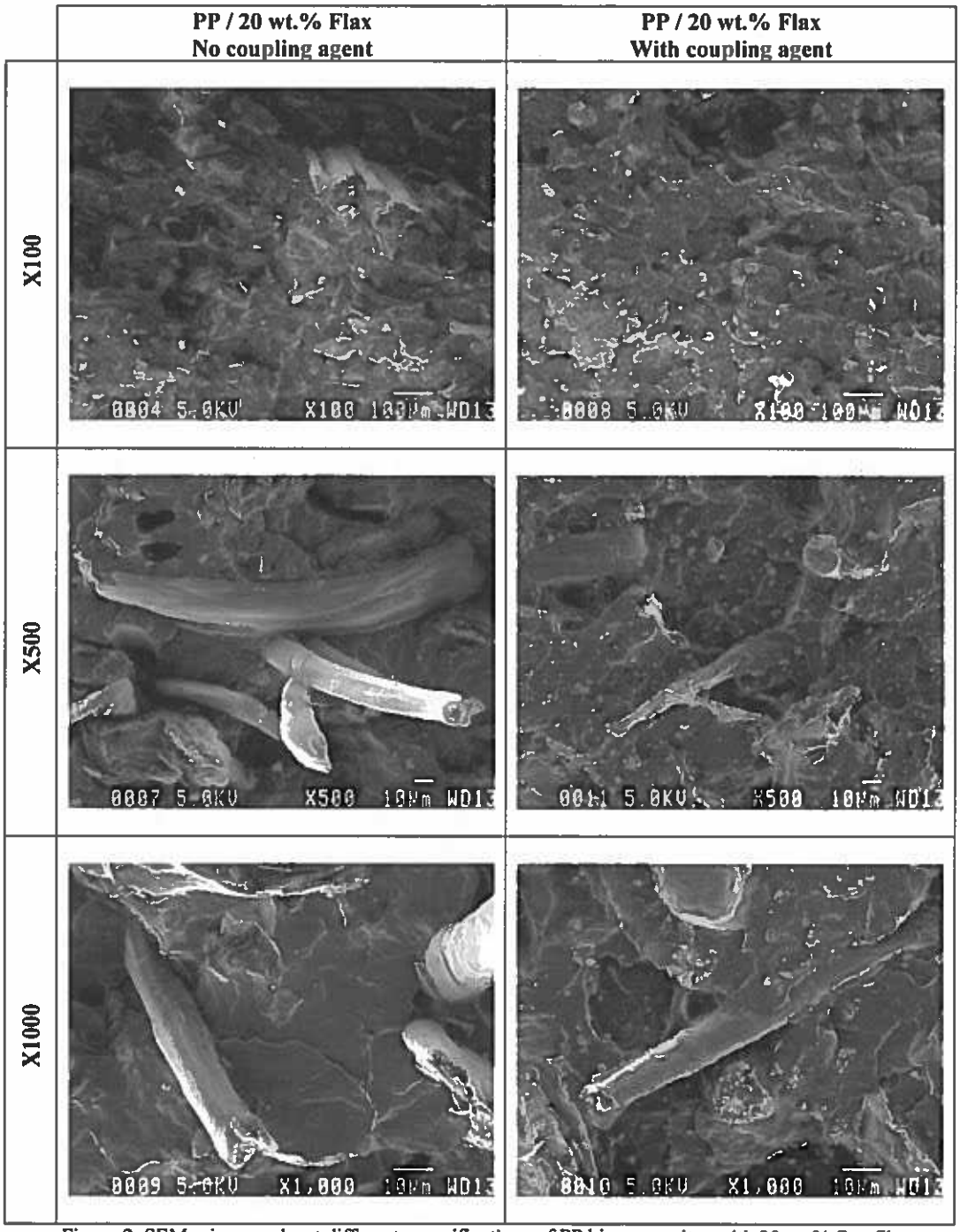

Figure 2. SEM micrographs at different magnifications of PP biocomposites with 20 wt.% flax fibers, without and with coupling agent.

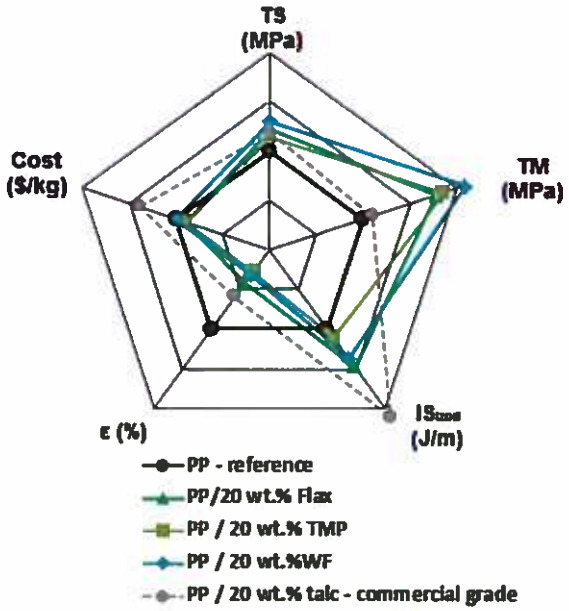

Figure 3. Comparison of mechanical properties and costs of: reference PP, PP / 20 wt.% cellulosics and PP / 20 wt.% talc commercial grade.

The market prices of neat PP, of the three types of cellulosic fibers, and of commercial PP references (Table 1a) were considered in the calculation of the material costs of PP biocomposites formulated, processed and characterized in this work (Table 1b). As a function of cellulosic fiber type and content, the PP biocomposites presented cost reductions from 3 % to 33 % compared to the current PP automotive grades while maintaining adequate mechanical performances comparing at current PP commercial grades.

PP ($/kg)	Co-PP ($/kg)	Flax ($/kg)	TMP ($/kg)	WF ($/kg)	PP / 20% talc ($/kg)	PP / 20% GF ($/kg)
1.8	3.8	1.0	0.3	1.5	2.5	2.5

Table 1a. Market prices of raw materials.

	Cellulosic content			
	20 wt.%		40 wt.%	
	Cost ($/kg)	Cost reduction (%)	Cost ($/kg)	Cost reduction (%)
PP / Flax	1.6	9.0	1.4	18.0
PP / TMP	1.6	17.0	1.3	33.4
PP / WF	1.8	3.4	1.7	6.8

Table 1b. Calculated material cost and cost reductions of obtained PP biocomposites.

3.3 Lightweight biocomposites based on PP

In our experimental trials, lightweight biocomposites were obtained using two strategies:

- Formulation strategy: replacement of a fraction of GF usually used as reinforcements in commercial automotive composites by cellulosic fibers to obtain hybrid composites.

- Processing strategy: foaming of biocomposites in injection molding using chemical blowing agents (CBAs).

Formulation strategy: lightweighting through replacement of GF by cellulosic fibers

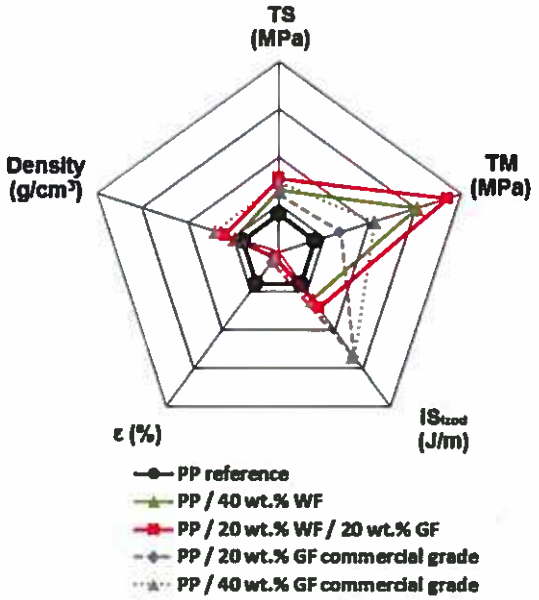

Figure 4. Comparison of properties for: neat PP, PP biocomposites and hybrids from this work, and commercial grades of PP / 20 wt.% and 40 wt.% GF.

The density of cellulosic fibers, depending on fiber type and fiber source, varies from 1.3 to 1.5 g/cm^3 while the density of GF, function of its grade, is almost two times higher, i.e. 2.55-2.6 g/cm^3. As per our study, the price of cellulosic fibers can vary from 0.3 to 1.5 $/kg and is lower than the price of GF, i.e. 3-4 $/kg. Replacing totally or partially the glass fibers by cellulosic fibers is a weight and cost dual strategy that encourages the replacement of GF composites by biocomposites. Figures 4 compares the properties of neat PP, PP biocomposites containing 40 wt.% WF, PP hybrids with 20 wt.% WF / 20 wt. GF and PP commercial automotive grades containing 20 and 40 wt.% GF. The hybrid composite PP / 20 wt.% WF / 20 wt.% GF obtained in this work presented a 93 % higher tensile strength (57 MPa) compared to neat PP (30 MPa) and a similar tensile strength as PP / 40 wt.% GF commercial grade (54 MPa). Its tensile modulus (7600 MPa) was evidently higher than the two commercial PP references (2800 and 4400 MPa respectively). As expected, its impact strength and elongation at break were lower than the commercial references because the presence of flexible cellulosic fibers comparing to the rigid glass fibers and because the 50 % lesser content in rigid glass fibers. As specified before, our biomaterials are basic formulations that could be upgraded straightforwardly using impact modifiers as per application needs. The density of PP / 20 wt.% WF / 20 wt.% GF hybrid (1.33 g/cm^3) was around 15 % lower than that of PP / 40 wt.% GF commercial reference (1.58 g/cm^3). Therefore, our current set of experiments and data seems to indicate that the partial replacement of GF reinforcements from PP composites by cellulosic fibers conducts to hybrid composites having equivalent or higher mechanical performances than PP/GF

commercial grades, while having around 15 % density reductions, up to 28 % material cost reduction and 20 wt.% renewable content.

Process strategy: lightweighting by injection foaming

In thermoplastic foam injection molding process, a chemical or physical blowing agent is introduced into the polymer melt initiating the formation of a single-phase polymer / gas solution, which will expand after injection into the mold cavity. This process leads to the formation of a structure with foamed core and compact skin layers. The thickness of the core and the skin layers depends on polymer type, additives present in the compound, type of blowing agent, and parameters used in the process (melt temperature, mold temperature, cooling time, injection speed etc.). The most important advantage of the foaming in injection molding is the reduction of the weight of the final injected part.

In this work, PP biocomposites containing 40 wt.% cellulosic fibers and PP hybrids containing 20 wt.% cellulosics / 20 wt.% GF were foamed during the injection molding process using 2 wt.% of Hydrocerol 1514 as chemical blowing agent. These biocomposites were injection foamed in rectangular parts for Izod impact tests and dogbones for tensile tests. The purpose was to use the foamed samples directly in impact and tensile evaluations without cutting them from bigger parts to avoid defects and induction of premature ruptures during mechanical testing.

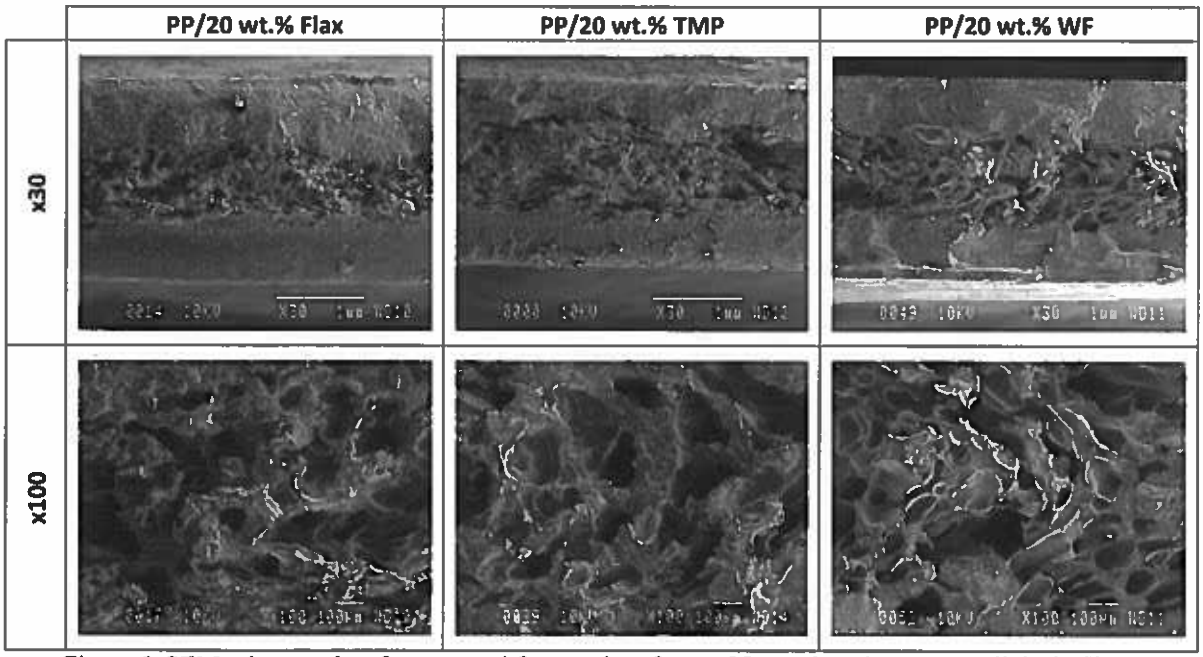

Figure 5. SEM micrographs of transversal fractured surfaces of foamed PP / 20 wt.% cellulosic fibers biocomposite samples.

Figure 5 shows transversal sections of PP / 20 wt.% Flax, PP / 20 wt.% TMP and PP / 20 wt.% WF biocomposite foamed parts at low magnification (first row) and high magnification (second row). The first row discloses the sectional structure of injected parts where the foamed core and the compact skin layers can easily be differentiated. The micrographs on the second row focus on the central foamed area and unveil the voids formed in biocomposite

matrix by the gas liberated during the decomposition of the chemical blowing agent. The formation of those voids helped at the part weight reduction while the unfoamed skin, due to its integrity, helped to preserve as much as possible the mechanical performances.

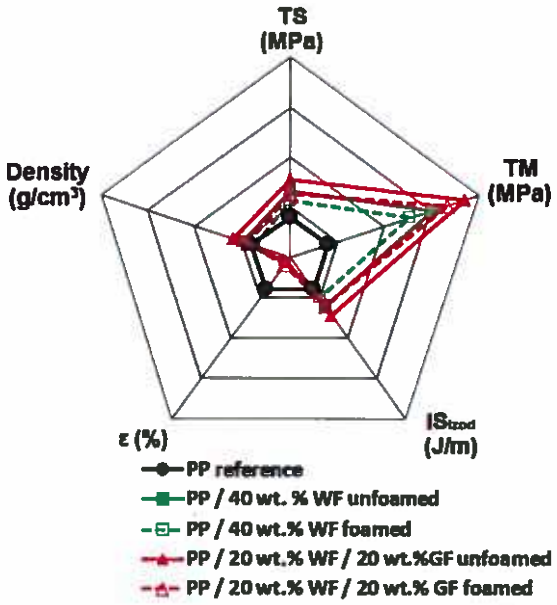

Figure 6. Properties comparison of neat PP, unfoamed and foamed PP / 40 wt.% WF biocomposites and, unfoamed and foamed corresponding hybrid.

Figure 6 unveils a comparision of tensile strength, tensile modulus, impact streght, elasticity, and density of neat PP, PP / 40 wt.% WF biocomposite unfoamed and foamed, and PP / 20 wt.% WF / 20 wt.% GF hybrid unfoamed and foamed. The foamed PP / 40 wt.% WF exhibit a slight decrease in mechanical properties compared to their unfoamed counterparts: the tensile strength decreased from 48 to 41 MPa, the tensile modulus from 6200 MPa to 5220 MPa, the impact strength from 29 to 24 J/m whilst the elongation remained unchanged. These were around 15 % reduction in properties compared to the unfoamed biocomposite. However, the properties of foamed parts remained essentially higher compared to neat PP. The most important benefit of foamed PP biocomposite was clearly the density. The foamed PP / 40 wt.% WF presented a density of 0.86 g/cm^3 which is 20 % lower compared to unfoamed PP / 40 wt.% WF, i.e. 1.1 g/cm^3. The 15 % lost in mechanical performances is waged by a 20 % lower density. In the case of the hybrids, the foamed PP / 20 wt.% WF / 20 wt.% GF presented a decrease of the tensile strength from 57 MPa to 47 MPa, of the tensile modulus from 7600 MPa to 6850 MPa, of the impact streght from 34 to 29 J/m. As the biocomposites, the hybrids presented a loss of around 15 % in performace because the foaming, fact greatly compensated by a density reduction of 20 % (from 1.32 g/cm^3 to 1.06 g/cm^3). Therefore, the current set of data seems to indicate that the PP biocomposites and hybrids foamed in this work disclosed almost similar mechanical perfomances compared to unfoamed parts while beeing 20 % ligther and, as consistent outcome, having a 20 % cost reduction.

3.4 Greener bioblends and biocomposites based on PP

The substitution of a part of PP matrix by a bioplastic was used to increase the renewable content. In this work, PLA was used as bioplastic and to replace 30 wt.% of the PP matrix. The compatibilized PP / PLA bioblend was further reinforced with cellulosic fibers to obtain PP / PLA biocomposites.

Figure 7 reveals mechanical properties and HDT values of neat PP, PP / PLA 30 wt.% bioblend, their biocomposites with 20 wt.% WF, and a PP / 20 wt.% talc commercial grade. As presented in Figure 3, the biocomposites PP / 20 wt.% WF performed better than neat PP. PP / 30 wt.% PLA, which incorporated in the formulation an adequate coupling agent, disclosed a 20 % higher tensile strength (33 vs. 29 MPa), 25 % higher tensile modulus (2100 vs. 1600 MPa) and 10 % higher impact strength (21 vs. 19 J/m) compared to neat PP. PP / PLA biocomposite with 20 wt.% WF presents 12 % higher tensile strength (40 vs. 35 MPa), 30 % higher tensile modulus (3800 vs. 2900 MPa) and 25 % higher Izod impact strength (35 vs. 28 J/m) compared to PP / 20 wt.% WF biocomposites. Moreover, the PP / 20 wt.% WF biocomposite had a HDT of 110 °C and the PP / 30 wt.% PLA / 20 wt.% WF biocomposite had a HDT of 126 °C, values being similar or higher than the one for neat PP (80 °C) and for commercial reference PP / 20 wt.% talc (115 °C). It should be noted also that PP / 30 wt.% PLA / 20 wt.% WF biocomposite presented similar or higher performances compared to commercial PP / 20 wt.% talc. Obviously, the replacement of a part of PP with PLA leads not only to a bioplastic and biocomposite that are more performant from mechanical and thermal points of view, but also greener. The PP / PLA bioblends and biocomposites can be formulated to contain at least up to 50 wt.% renewable content while having upgraded performances. Furthermore, PP bioblends and biocomposites can easily be adapted to injection and injection-foaming processes without changing the process settings. Figure 8 shows an injected PP / PLA and PP/PLA biocomposite parts fabricated by injection molding in our semi-industrial scale laboratory.

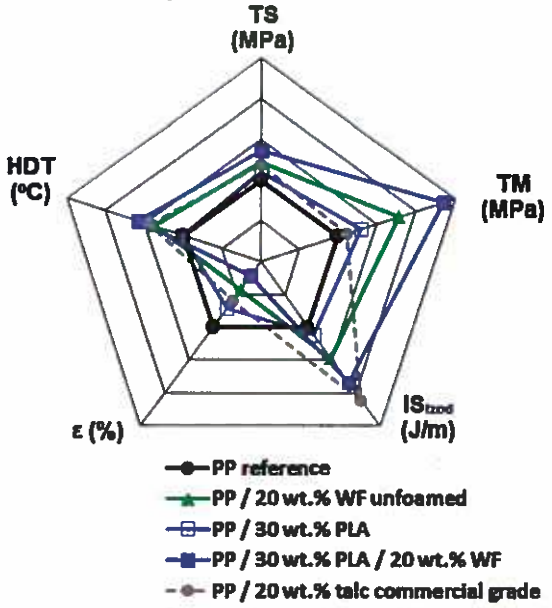

Figure 7. Properties comparison of: reference PP, PP bioblend and biocomposites from this work and the commercial grade PP / 20 wt.% talc.

Figure 8. Injection molded samples of PP / PLA bioblend and its corresponding biocomposite.

4 CONCLUSIONS

This paper disclosed formulation and processing solutions towards lighter, lower cost PP sustainable blends and composites that have high potential applicability to be used in automotive interior parts fabrication.

Biocomposites containing up to 40 % cellulosic fibers based on PP and PP / PLA blends presented equivalent or higher mechanical and thermal properties comparing to conventional thermoplastic compounds and composites currently used in automotive industry, while having:

- Up to 25 % lower material cost due to the incorporation of up to 40 wt.% of low cost cellulosic fibers;
- Up to 33 % lower weight due to the partial or complete replacement of glass fibers by cellulosic fibers and/or by using an adequate injection foaming process;
- Up to 50 % renewable content when besides cellulosic fibers, PP/PLA bioblends were used as matrix.

This work proved the feasibility of novel PP bioblends and biocomposites as eco-solutions for automotive applications. Beyond this feasibility, the lower-cost, lower-weight and the greener content are capital characteristics of these biomaterials. Although impact properties, moisture sensitivity and odor still need to be improved, those innovative bioblends and biocomposites enable designing engineering thermoplastics compounds containing up to 50 wt.% renewable content.

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