Study of the Flexural Modulus and the Micromechanics of Old Newspaper Reinforced Polypropylene Composites

Francesc X. Espinach,[a] Miquel A. Chamorro-Trenado,[b] Joan Llorens,[b] Josep Tresserras,[a] Neus Pellicer,[c] Fabiola Vilaseca,[d,]* and Angels Pelach [c]

The inclusion of old newspaper as reinforcement in composites promotes the use of environmentally friendly materials, reduces landfilling, decreases chemical treatments to bleach the recycled paper, and provides alternatives to glass fiber. While there are studies on the tensile strength and stiffness of old newspaper reinforced materials, there is a lack of analysis of their flexural properties. Considering that bending loads are very common, the flexural stiffness of the materials must be examined prior to industrial use. In this study, the flexural moduli of composites made from old newspaper fibers and polypropylene were compared with other composites. The composites showed moduli ranging from 2.1 to 4.1 GPa, and a composite with 50% newspaper content had a flexural modulus comparable to 10% glass fiber composite. These values allow an industrial use of the composites for semi-structural purposes. A method to assess the intrinsic flexural modulus of the reinforcements was presented and evaluated against other micromechanics models. The value of the efficiency factors and a mean orientation angle were also obtained.

Keywords: Old newspaper; Polypropylene; Recycling; Flexural modulus, Micromechanics

*Contact information: a: Design, Development, and Product Innovation, Department of Organization, Business Management, and Product Design, Escola Politècnica Superior, 17003 Girona, Spain; b: Construction Advanced Technologies and Sustainability, Department of Architecture and Construction Engineering, University of Girona, Escola Politècnica Superior, 17003 Girona, Spain; c: Laboratory of Paper Engineering and Polymer Materials, Department of Chemical and Agricultural Engineering and Agrifood Technology, Escola Politècnica Superior, 17003 Girona, Spain; d: Advanced Biomaterials and Nanotechnology, Department of Chemical and Agricultural Engineering and Agrifood Technology, Escola Politècnica Superior, 17003 Girona, Spain; *Corresponding author: fabiola.vilaseca@udg.edu*

INTRODUCTION

Composites are usually formulated to obtain new materials with enhanced properties, compared to its phases. These properties are usually related to the mechanical performance or the lightness of the composite constituents. In the case of carbon fiber reinforced resins, it is possible to obtain materials with higher stiffness, strength, and less weight (Espinach *et al.* 2017a). Economic and environmental concerns also justify the formulation of composite materials (Oliver-Ortega *et al.* 2018b). The use of fillers has been a common solution to avoid using 100% oil-based matrices. Nonetheless, adding a cheap phase to a composite decreases the material cost, but it can also reduce its mechanical performance. The use of reinforcements from renewable sources allow formulating more environmentally friendly products. The formulation of greener materials with enhanced mechanical properties is a common goal for the composite materials researchers, society, and industry (Al-Oqla and Sapuan 2014; Gu *et al.* 2016).

The use of natural fibers such as annual plants, wood fibers, or agroforestry wastes in enhanced composite materials has advantages and disadvantages (Joshi *et al.* 2004; Koronis *et al.* 2013; Summerscales *et al.* 2013). Natural fiber reinforced composites have proven to be competitive in front of glass fiber reinforced polyolefin (Kong *et al.* 2016; Pickering *et al.* 2016; Pil *et al.* 2016). Natural fibers have less specific weight than glass fibers and consequently its composites have higher specific properties than glass fiber-based ones (Granda *et al.* 2016a,b). Wood fiber reinforced composites are used to manufacture building materials and automotive parts (Singh *et al.* 2011; Kong *et al.* 2016; Siengchin 2017). Nonetheless, the utilization of agroforestry waste or industry byproducts is a good strategy to add value to such materials while promoting a circular economy (Gu *et al.* 2016; Jiménez *et al.* 2017). In this sense, textile waste, agroforestry straws or cotton flocks can be sources for composite reinforcements (Araújo *et al.* 2017; Serra *et al.* 2017). Another possible industrial byproduct available as reinforcement is used paper (Serrano *et al.* 2013; Sahin *et al*. 2017). Waste paper causes environmental concerns because it is a wood-based material and has implications in deforestation (Lykidis *et al.* 2012). Newspapers are low lifespan products that are usually recycled as pulp but also show a high rate of incineration or landfilling (Serrano *et al.* 2014b). Its recycling as papermaking pulp involves deinking processes and the use of reagents and energy, thus polluting the environment. On the other hand energy recovering by incineration means releasing CO_2 and increasing the greenhouse effect. Thus, recovering higher rates of old newspaper can be seen as an environmentally friendly strategy. This goal can be reached by adding value to old newspapers and its use as composite reinforcement is proposed by the authors.

The literature shows that there is interest in old newspaper as composite reinforcement. Composite materials based on old newspaper (ONP) reinforced polypropylene exhibited noticeable tensile strengths and Young's moduli (Serrano *et al.* 2013, 2014a). Compared with glass fiber-based materials, old newspaper-based composites showed higher specific properties, low costs of the reinforcing fibers, and lower equipment abrasion (Ali *et al.* 2011; Nedjma *et al.* 2013). In fact, the literature supports that a pressure pump body can be manufactured with a composite with ONP 40 wt% replacing a 20 wt% glass fiber reinforced polypropylene (Serrano *et al.* 2014b). However, old newspaper-based composites present poor interphases with polyolefin matrices (El Mansouri *et al.* 2012), which is expected for hydrophilic reinforcements (Reixach *et al.* 2013b). Coupling agents such as anhydride-grafted polypropylene improve composite interphases (Serrano *et al.* 2013). The literature about the flexural modulus of ONP-based composites is scarce, showing flexural modulus in the order of 3.1 GPa for materials containing ONP 50 wt% (Yuan *et at.* 1999). This value can be considered low in comparison to other natural fiber reinforces composites. Another study obtains a notable flexural modulus of 5.1 GPa with 30 wt% ONP contents and the presence of talc (Huda *et al*. 2005). Nonetheless, these composites showed low flexural strengths, probably due to the lack of coupling agents in the composite formulation.

Composites reinforced by semi-aligned short fibers show noticeable anisotropy on properties like strength and stiffness. While the flexural strength has values usually above the tensile strength, the bending modulus tends to be lower than the Young's modulus (Espinach *et al.* 2015, 2017b). Due to the fact that many industrial components are subjected to bending stresses, it is necessary to know how they deform under these loads. Thus, a key value is the flexural modulus.

In this work, ONP-reinforced polypropylene composites were formulated, manufactured, and tested under three point test bending conditions. The main objective of this study was to evaluate the flexural modulus of these composites at different reinforcement contents. Usually, the modulus of composites is not influenced by the quality of the matrix reinforcement interphase (Espinach *et al.* 2013). However, good fiber-matrix interphases ensure higher strength values and higher strains at breaking (Hashemi 2008; Franco-Marques *et al.* 2011; Serra *et al.* 2017). Therefore, materials with and without coupling agents were assayed. The experimental flexural moduli were compared with other natural fibers and glass fiber reinforced polypropylene composites. To understand the contribution of the composite phases to the flexural moduli of the composites, a micromechanical analysis was proposed. In a first stage, a fiber flexural modulus factor was used to evaluate the neat contribution of the matrix. In a second stage, Hirsch's equation was used to evaluate the intrinsic flexural modulus of the matrix (Hirsch 1962). This modulus was used to solve a modified rule of mixtures and obtain the efficiency factor value. Cox and Krenchel's equation was used to obtain the length efficiency factor to evaluate the impact of the fiber morphology on the flexural modulus (Cox 1952; Krenchel 1964). The orientation efficiency factor was also obtained along with the value of a theoretical mean orientation angle. Finally, another micromechanics model was used to compute the intrinsic flexural modulus as well as the efficiency factors to triangulate such results. The proposed model was the Tsai and Pagano model and the Halpin and Tsai equations (Reixach *et al.* 2013a). This model implicitly added the morphologic properties of the reinforcements to the equations while Hirsch's equation does it explicitly. The obtained composites showed competitive flexural moduli comparable to glass fiber or lignocellulosic-based composites at lower reinforcement contents. The use of old newspaper as a source of reinforcements avoids its incineration, dumping, or deinking.

EXPERIMENTAL

Materials

Old newspapers were kindly provided by Punt Diary (Girona, Spain). The paper was composed of thermo-mechanical pulp from hardwood and calcium carbonate fillers, 85% and 15%, respectively.

The matrix was a polypropylene homopolymer (PP) ISPLEN PP090 from REPSOL (Tarragona, Spain), with a density of 0.905 g/cm^3. To increase the compatibility of the interphase, a maleic anhydride-grafted polypropylene (MAPP) coupling agent was included in the formulations of some composites. The coupling agent was Epolene® G3015 from Eastman Chemical Company (Madrid, Spain). This coupling agent had an acid number of 15 mg KOH/g and a molecular weight of 47000 g/mol.

Diethyleneglycol dimethyl ether (diglyme) supplied by Clariant (Vila-seca, Spain) was used as a dispersing agent (Rodriguez *et al.* 2010).

Decahydronaphthalene (decalin) supplied by Fisher Scientific (Madrid, Spain) was used to dissolve the PP matrix and recover the reinforcements from the composite prior to their morphological analysis.

Recovering of Old Newspaper Fibers

The old newspapers were cut into 10 x 10 cm pieces and soaked in a sodium hydroxide bath before starting the disintegration process. Old newspaper fibers were disintegrated using a lab pulper by Pulcel Cell from Metrotech (Madrid, Spain), and the thermo-mechanical pulp was recovered. The equipment had a 20 L volume capacity and operated at 20 rev/s speed and 50 °C. The obtained pulp was dried at 80 °C for 24 h in a Dycometal oven (Viladecans, Spain). Afterwards, the pulp was dispersed in a 1:3 water-diglyme suspension. Approximately 5% of the initial $CaCO_3$ was lost during the disintegration and individualization processes. This method was previously used to obtain composites submitted to tensile testing (Serrano *et al.* 2013). The obtained old newspaper fibers (ONP) were used as reinforcement. The density of the ONP was evaluated at 1.350 g/cm^3.

Composite Preparation and Sample Injection Molding

Composites with ONP contents ranging from 20 to 50 wt% were formulated and prepared in a Brabender® GmbH plastograph mixer (Duisburg, Germany). The equipment worked at 80 rpm and 180 °C for 10 min to obtain the composite materials. Two separate batches were prepared, one including 6% of MAPP, with respect to the fiber content, and another without MAPP. The blends were extracted from the mixer and pelletized to a granular diameter of 3 mm in a blade mill from Agrimsa (Barcelona, Spain). Finally, the pellets were dehumidified for 24 h in an oven at 80 °C.

The test specimens (126 x 12.5 x 3mm) were mold injected in a Meteor-40 injection machine from Mateu & Solé (Barcelona, Spain). The injection temperature was 180 °C with a peak of 190 °C at the injection nozzle. At least ten specimens of each formulation were obtained.

Mechanical Characterization

Before any test, the specimens were stored in a conditioning chamber for 48 h, at 23 °C and 50% relative humidity, in agreement with the ASTM D618-13 (2013) standard.

The specimens were submitted to a three point bending test following ASTM D790-17 (2017) using the 1122 universal testing machine by Instron® (Barcelona, Spain). The experimental results were the mean of at least 5 experiments.

The strain at flexural (ε_f^C) was evaluated as: $\varepsilon_f^C=(6 \cdot D \cdot d)/L^2$, where D (mm) is the maximum deflection at the center of the specimen, d (mm) the specimen depth, and L (mm) the support span (52.6 mm).

Morphology of the Reinforcements

The reinforcing ONP fibers were recovered by matrix solubilization in a Soxhlet apparatus. Decalin was used as a PP solvent. The recovered fibers were rinsed with acetone and distilled water to remove any decalin residue.

The morphology of the fibers was measured in a MorFi Compact by Techpap SAS (Grenoble, France). The equipment returned information on the mean length and width of the reinforcements as well as the length and width percentage distributions. The ONP fiber length distribution histogram can be consult in the literature (Serrano *et al*. 2013).

Micromechanics Models

To examine the contribution of the different phases to the flexural modulus of the composites, a micromechanics study was proposed. The equations and models used to perform the study are presented below.

Rule of mixtures

The modified rule of mixtures for the flexural modulus (mRoM) of a two-phase composite was derived from a modified rule of mixtures for the Young's modulus for such composites (Serrano *et al.* 2014a; Granda *et al.* 2016b),

$$E_f^C = \eta_{f,e} \cdot E_f^F \cdot V^F + (1 - V^F) \cdot E_f^M \qquad (1)$$

where E_f^C, E_f^F, and E_f^M are the flexural modulus (GPa) of the composite, the reinforcement and the matrix, respectively. The flexural modulus efficiency factor ($\eta_{f,e}$) was added to the equation to take into account the mean orientation of the fibers and its morphology, and to balance the contribution of the fibers to the flexural modulus of the composite. This factor ranges from 0 to 1. The volume fraction of the reinforcement (V^F) was deduced from the weight fractions and the density of the phases.

The mRoM was used to compute the value of the intrinsic flexural modulus of the fibers from the experimental data.

The flexural modulus efficiency factor can be decomposed as the product of different efficiency factors in order to account for the orientation ($\eta_{f,o}$) and the length ($\eta_{f,l}$), as represented in Eq. 2.

$$\eta_{f,e} = \eta_{f,l} \cdot \eta_{f,o} \qquad (2)$$

Other authors propose other factors to consider the shape of reinforcements, but its inclusion was discarded for the present research (Virk *et al.* 2012).

Fiber flexural modulus factor

One of the main weaknesses of the modified rule of mixtures of semi-aligned fiber reinforced composites is related with the presence of $\eta_{\mathrm{f,e}}$ in the equation. The mRoM was used to predict the flexural modulus of a composite for a certain formulation. However, determining the intrinsic flexural modulus of the fibers was quite complex. Because the $\eta_{\mathrm{f,e}}$ was unknown, the number of unknowns raised up to two, so the equation could not be resolved. Anyway, the contribution of the fibers to the flexural modulus of the matrix was $\eta_{\mathrm{f,e}} \cdot E_{\mathrm{f}}^{\mathrm{F}}$, and the mRoM could be rearranged to account for the contribution of the reinforcements against its volume fraction as shown in Eq. 3.

$$\eta_{\mathrm{f,e}} \cdot E_{\mathrm{f}}^{\mathrm{F}} \cdot (V^{\mathrm{F}}) = \frac{(1-V^{\mathrm{F}}) \cdot E_{\mathrm{f}}^{\mathrm{M}}}{E_{\mathrm{f}}^{\mathrm{C}}} \tag{3}$$

Therefore, if the contribution of the reinforcement was plotted against its volume fraction, the slope of the regression line, passing through (0,0), was defined as the Fiber Flexural Modulus Factor (*FFMF*) (Espinach *et al.* 2015, 2017b).

Hirsch equation

Another option to obtain the intrinsic modulus of reinforcement was the use of micromechanics models like Hirsch's equation. Equation 4 is based on the Voigt and Reus models.

$$E_{\mathrm{f}}^{\mathrm{C}} = \beta \left(E_{\mathrm{f}}^{\mathrm{F}} \cdot V^{\mathrm{F}} + E_{\mathrm{f}}^{\mathrm{M}} (1 - V^{\mathrm{F}}) \right) + (1-\beta) \frac{E_{\mathrm{f}}^{\mathrm{F}} \cdot E_{\mathrm{f}}^{\mathrm{M}}}{E_{\mathrm{f}}^{\mathrm{M}} \cdot V^{\mathrm{F}} + E_{\mathrm{f}}^{\mathrm{F}} \cdot (1-V^{\mathrm{F}})} \tag{4}$$

In Eq. 4, the β factor determined the weight of the contributions of the fibers virtually aligned with the loads and those perpendiculars to such loads. Thus, the factor was equivalent to a mean orientation factor. The model fit with the experimental values for $\beta = 0.4$ (Delgado-Aguilar *et al.* 2017).

Cox and Krenchel model

In the presentation of the mRoM (Eq. 1), a flexural efficiency modulus factor ($\eta_{\mathrm{f,e}}$) was introduced. Once the intrinsic flexural modulus of the reinforcements was obtained using Hirsch's equation (Eq. 4), it was possible to compute the value of $\eta_{\mathrm{f,e}}$. This factor related with the length and orientation factors according to Eq. 2. The values of the length and orientation factors informed the study about the contribution of the mean length and orientation of the reinforcements in their contribution to the flexural modulus. In this line, the Cox and Krenchel (Cox 1952; Krenchel 1964) model allowed for the calculation of the value of the length efficiency factor ($\eta_{\mathrm{f,l}}$),

$$\eta_{\mathrm{f,l}} = 1 - \frac{tanh(\chi \cdot L^{\mathrm{F}}/2)}{(\chi \cdot L^{\mathrm{F}}/2)} \tag{5}$$

with

$$\chi = \frac{1}{R^{\mathrm{F}}} \sqrt{\frac{E_{\mathrm{f}}^{\mathrm{M}}}{E_{\mathrm{f}}^{\mathrm{F}} \cdot (1-\upsilon) \cdot Ln\sqrt{\pi/4 \cdot V^{\mathrm{F}}}}} \tag{6}$$

where L^{F} and R^{F} refer to the mean lengths (µm) and radius (µm) (width) of the reinforcements, respectively, and υ is the Poison's ratio of the matrix. Once the length efficiency factor was known it was possible to obtain the value of the orientation efficiency factor by using Eq. 2.

Mean orientation of the fibers

Fukuda and Kawata presented a method that linked the orientation efficiency factor (η_o) with a theoretical limit orientation angle of the fibers (α_o) (Fukuda and Kawata 1974). The authors also defined a connection between the limit angle and different fiber distributions. Semi-aligned short fiber reinforced composites did not show a particular distribution of the fibers, but the literature shows that rectangular and triangular distributions are the most common (Jiménez *et al.* 2017).

Rectangular distribution:

$$\eta_o = \frac{\sin(\alpha_o)}{\alpha_o}\left(\frac{3-\nu}{4}\cdot\frac{\sin(\alpha_o)}{\alpha_o} + \frac{1-\nu}{4}\cdot\frac{\sin(\alpha_o)}{\alpha_o}\right) \tag{7}$$

Triangular distribution:

$$\eta_o = 4\cdot\frac{1-\cos(\alpha_o)}{\alpha_o^2}\left(\frac{3-\nu}{4}\cdot\frac{1-\cos(\alpha_o)}{\alpha_o^2} + \frac{1+\nu}{4}\cdot\frac{1-\cos(3\alpha_o)}{9\cdot\alpha_o^2}\right) \tag{8}$$

Sanomura and Kawamura (2003) defined an orientation factor (f_p) that established a correlation between the limit orientation angle and the theoretical mean orientation angle of the fibers (α), as shown in Eq. 9.

$$f_p = \frac{\sin(2\alpha_o)}{2\alpha_o} = 2\cdot cos^2(\alpha)\text{-}1 \tag{9}$$

Tsai and Pagano model

The Hirsch equation (Eq. 4) has proven its utility to predict the intrinsic flexural modulus of the reinforcements. Nevertheless, this equation did not include explicitly any morphologic property of the reinforcements. Thus, the Tsai and Pagano model and the Halpin and Tsai equations are proposed as a way to obtain the intrinsic flexural modulus of the reinforcements.

The Tsai and Pagano model defined the flexural modulus of a composite from its longitudinal (E^{11}) and transversal (E^{22}) moduli according to Eq. 10:

$$E_f^C = \frac{3}{8}E^{11} + \frac{5}{8}E^{22} \tag{10}$$

The Halpin and Tsai equations were used to define E^{11} and E^{22} (Espinach *et al.* 2013).

$$E^{11} = \frac{1+2\left(L^F/2R^F\right)\cdot\frac{\left(E_t^F/E_t^M\right)-1}{\left(E_t^F/E_t^M\right)+2\left(L^F/2R^F\right)}\cdot V^F}{1-\frac{\left(E_t^F/E_t^M\right)-1}{\left(E_t^F/E_t^M\right)+2\left(L^F/2R^F\right)}\cdot V^F}E_t^M \tag{11}$$

$$E^{22} = \frac{1+2\cdot\frac{\left(E_t^F/E_t^M\right)-1}{\left(E_t^F/E_t^M\right)+2}\cdot V^F}{1-\frac{\left(E_t^F/E_t^M\right)-1}{\left(E_t^F/E_t^M\right)+2}\cdot V^F}E_t^M \tag{12}$$

In these equations, the aspect ratio of the fibers (ratio between the length and the diameter) was used to determine the value of the longitudinal modulus.

RESULTS AND DISCUSSION

The inclusion of a stiffer phase in the formulation of a composite usually delivers a new material with a higher modulus than the matrix, and also with lower strains at the break (Oliver-Ortega *et al.* 2018a). Therefore, as expected, the flexural modulus of ONP reinforced composites increased with the increasing percentages of reinforcement (Table 1).

Table 1. Experimental Flexural Modulus and Strain at Break for All Composites

Composite	V^F (v/v)	E_f^C (GPa)	ε_f^C (%)
PP	0	1.1 ± 0.12	6.5 ± 0.25
PP20ONP0MAPP	0.144	2.1 ± 0.16	3.7 ± 0.16
PP30ONP0MAPP	0.223	2.7 ± 0.22	2.7 ± 0.11
PP40ONP0MAPP	0.309	3.1 ± 0.19	2.6 ± 0.13
PP50ONP0MAPP	0.402	4.0 ± 0.23	1.6 ± 0.12
PP20ONP6MAPP	0.144	2.2 ± 0.14	4.2 ± 0.18
PP30ONP6MAPP	0.223	2.5 ± 0.18	4.0 ± 0.15
PP40ONP6MAPP	0.309	3.2 ± 0.24	2.6 ± 0.13
PP50ONP6MAPP	0.402	4.1 ± 0.21	2.5 ± 0.14

The presence of coupling agents in the formulations of the composites showed little impact on the flexural moduli, and the composites with or without MAPP gave a very similar bending moduli. This was expected because the interphase has little influence on the Young's modulus of the composites (Osman *et al.* 2010; Reixach *et al.* 2013a). Some authors relate this limited effect with the low strains where the modulus was measured (Granda *et al.* 2016b).

The flexural modulus of the composite containing 50 wt% of ONP was by 3.7 times the flexural modulus of the matrix. This increment was slightly lower than that for 50 wt% stone groundwood (SGW) reinforced PP composites, where the increase was of 4.9 times (Lopez *et al.* 2012b). However, 30 wt% glass fiber (GF) reinforced PP composites rendered flexural modulus 5.54 times higher than the matrix (Girones *et al.* 2011). Considering that GF reinforced PP are commodity materials for the industry, it is worth mentioning that the composite with 50 wt% ONP provided the same flexural modulus than the composite with 10 wt% GF content. Such composites are normally used for product bodies, ornamental parts, or semi-structural parts under comparatively low bending loads (Serrano *et al.* 2014b). Therefore, in terms of the flexural modulus, the ONP-based composites could be beneficial for the industry in cases were environmental concerns are involved.

The evolution of the flexural modulus of the composites with the reinforcement volume fraction was linear (Fig. 1). The regression lines for the coupled and uncoupled composites are almost the same, demonstrating the little impact of the coupling agents on the flexural modulus of the composites. Some authors connect this linear behavior with a good dispersion of the fibers inside the composite (Granda *et al.* 2016b).

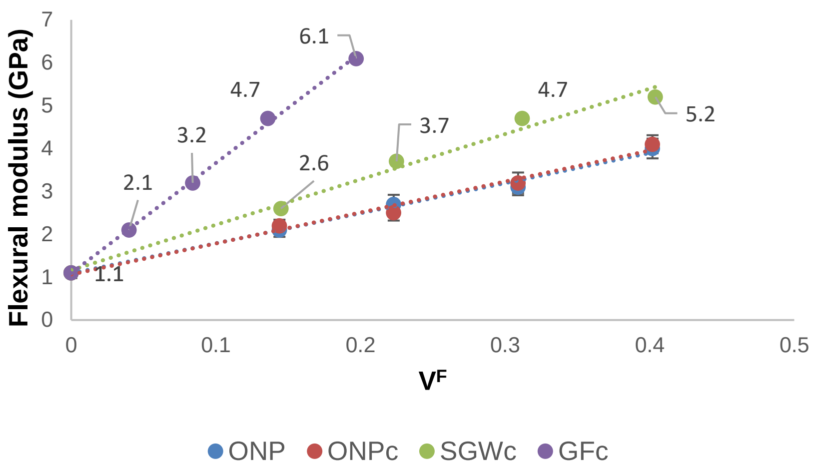

Fig. 1. Evolution of the flexural modulus of the coupled and uncoupled ONP, coupled SGW and coupled GF reinforced PP composites against the reinforcement contents (Girones *et al.* 2011; Lopez *et al.* 2012b)

The inclusion of a stiffer, but also more fragile phase into the composite formulation decreased the strains at the break (Table 1). The results showed remarkable decreases of the strain at the break with the ONP content. For this property, however, the coupling agent had a visible influence. Coupled materials showed higher strains at break than uncoupled materials. Uncoupled and coupled 50 wt% ONP reinforced composites showed strains at the break of 24.6% and 38.5% times the strain of the matrix, respectively. The ONP based composites showed flexural moduli slightly lower that SGWc and noticeably lower than GFc. The influence of the coupling agent was related with the better load stress transfer for the coupled system, combined with Hooke's law. Figure 2 shows the evolution of the strain at the break of some composites against the reinforcement content, where ONP, ONPc, GFc, and SGWc were uncoupled old newspaper, coupled old newspaper, coupled glass fibers, and coupled stone ground wood reinforced PP composites, respectively.

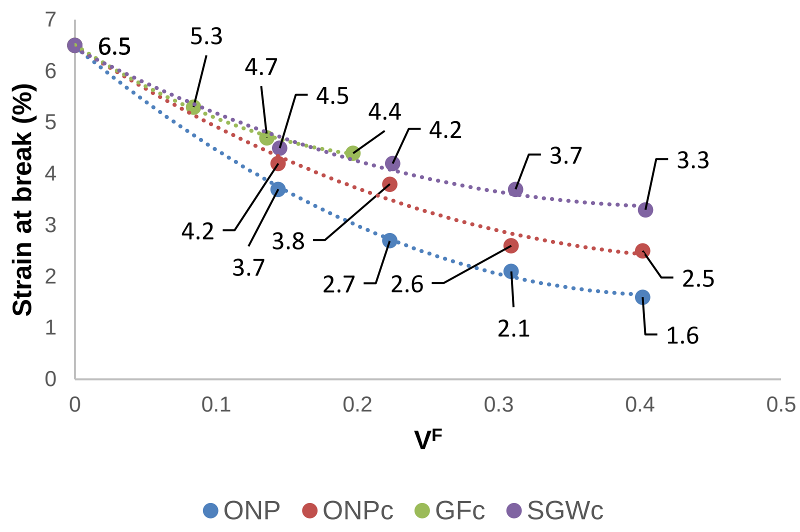

Fig. 2. Evolution of the strain at the break of the composites against several reinforcement contents (Girones *et al.* 2011; Lopez *et al.* 2012b)

The figure shows a non-linear evolution of the strains at the break for the all of the composites, with a quasi-asymptotic behavior towards the lowest values. The figure clearly points out the differences between the coupled and uncoupled ONP–composites. Anyhow, the strains at the break of the ONP-based composites were noticeably lower than those of SGW and GF-based materials. This has implications on the possible uses of the ONP-based materials that will need stiffening by geometry (*i.e.* use of ribs) to prevent failures.

The chemical composition of the ONP and the SGW fibers was fairly similar (Lopez *et al.* 2011, 2013). Thus, the different behavior for the ONP and SGW-based composites could be due to the presence of $CaCO_3$ in the newspaper sheets, the presence of inks, or the reduction of the aspect ratio of the ONP fibers due to recycling processes. In the case of the fiber morphology, SGW fibers used had aspect ratios between 23.2 and 18.4 (Lopez *et al.* 2012a) while ONP fibers showed aspect ratios between 19.1 and 15.7. Therefore, the influence could not be fully attributed to any of the mentioned possible causes, and more research is needed to isolate the referred causes. In any case, deinking or eliminating all the $CaCO_3$ involves the use of reactants and energy.

Contributions of the Reinforcement to the Flexural Modulus of Composites

The concept of the Fiber Flexural Modulus Factor (*FFMF*) is introduced in the Experimental section. Figure 3 depicts the obtained *FFMF* values for the current ONP-composites compared with other reinforcements. The slope corresponding to the ONP-based composites was inferior to that of other reinforcements. The *FFMF* for the current ONP composites were 30% below the *FFMF* of the SGW composites. The possible causes include the newspaper formulation itself (charges, inks), rather than in the intrinsic fiber morphology, as discussed in the previous section. The slope of the regression curve indicated a positive and continuous contribution of the ONP to the flexural modulus of the composites.

The GFc exhibited a 3.2 times higher contribution for V^F than ONP. This roughly means that 3.2 more times of ONP volume fractions were needed to attain the same bending modulus than with the GFc composites. The density of the GF was higher than that of ONP, thus the weight percentages of ONP were also noticeably higher (Table 1).

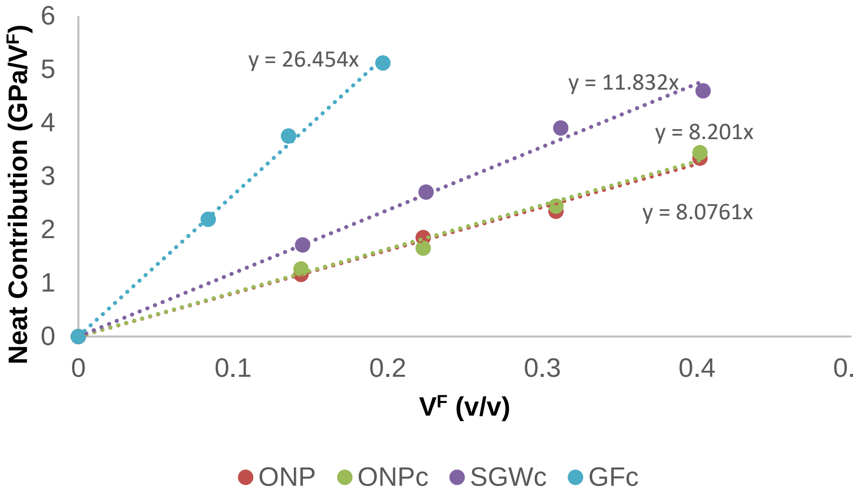

Fig. 3. Neat contribution of the reinforcements to the flexural modulus of polypropylene-based composites

The ratio between a fiber tensile strength factor (*FTSF*) and a fiber flexural strength factor (*FFSF*) has been proposed to compute the value of the intrinsic flexural strength of the reinforcements (Lopez *et al.* 2012a). The *FFMF* was a result of a rearrangement of the mRoM (Eq. 3). Some researchers define a fiber tensile modulus factor (*FTMF*) by rearranging the mRoM for the Young's modulus. The ratio between the contributions of the fibers to the flexural and the Young's modulus was proposed as a way to predict the value of the intrinsic flexural modulus of the reinforcements, as shown in Eq. 13.

$$\frac{FFMF}{FTMF} = \frac{E_f^F \cdot \eta_{f,e} \cdot V^F}{E_t^F \cdot \eta_{t,e} \cdot V^F} \tag{13}$$

The equation assumes that the efficiency factors for the Young's and the flexural modulus are equivalent ($\eta_{t,e} = \eta_{f,e}$); therefore, it was possible to compute the intrinsic flexural modulus if intrinsic Young's modulus, the *FFMF*, and the *FTMF* were known. For the case of ONP, the literature indicated that ONP had an intrinsic Young's modulus of 22.8 GPa and a *FTMF* of 11.32 (Serrano *et al.* 2014a). Therefore, according to Eq. 13, the value of the intrinsic flexural modulus of the ONP was around 16.3 GPa. Nonetheless, Eq. 13 presented a theoretical analysis and needed to be confirmed by using more established models.

Micromechanics of the Flexural Modulus

The first model used to compute a theoretical intrinsic flexural modulus was the Hirsch equation (Eq. 4). Table 2 presents the intrinsic flexural modulus, with the efficiency factor, as well as the length and orientation factors. The mean intrinsic flexural moduli for the uncoupled and coupled composites were 16.40 ± 0.86 GPa and 16.63 ± 1.57 GPa, respectively. Both values were the same, considering a 95% confidence rate. These intrinsic flexural moduli were noticeably lower than the intrinsic bending modulus of the SGW as PP reinforcement, of 24.7 GPa (Lopez *et al.* 2013). The obtained values were very similar to the theoretical value from Eq. 13. The efficiency factors were also very similar, despite the presence of the coupling agents or the volume fraction of reinforcement, and the factors were in the range from 0.44 to 0.53. From previous works, the efficiency factor of the semi-aligned short fibers reinforced composites rendered values around 0.5 supporting the relevance of the results (Serrano *et al.* 2014a; Granda *et al.* 2016b; Delgado-Aguilar *et al.* 2017).

Table 2. Intrinsic Flexural Modulus of the ONP Obtained from Hirsch Equation and Micromechanics of the Flexural Modulus of the Composites

Composite	E_f^F (GPa)	η_e	η_l	η_o
PP20ONP0MAPP	16.72	0.491	0.805	0.578
PP30ONP0MAPP	17.06	0.504	0.870	0.579
PP40ONP0MAPP	15.13	0.462	0.864	0.534
PP50ONP0MAPP	16.68	0.507	0.886	0.572
PP20ONP6MAPP	18.45	0.527	0.850	0.620
PP30ONP6MAPP	14.84	0.443	0.869	0.510
PP40ONP6MAPP	15.92	0.475	0.864	0.550
PP50ONP6MAPP	17.29	0.514	0.886	0.582

To apply Cox and Krenchel's model (Eq. 5 and 6), the morphology of the reinforcements was determined. In all formulations, the average diameter (width) of the reinforcement was 21.6 µm, whereas the mean length for the composites at 20, 30, 40, and 50 wt% of ONP were 411, 406, 336, and 340 µm, respectively. Some of the authors discussed in depth the morphology of the ONP fibers and its changes during composite preparation (Serrano *et al.* 2013). The variations were attributed to attrition during composite processing (Serrano *et al.* 2013; Granda *et al.* 2016b). The length efficiency factors are also presented in Table 2, with average values of 0.86 ± 0.03 and 0.87 ± 0.01 for uncoupled and coupled composites, respectively. The mean orientation efficiency factors deduced from Eq. 2 gave average values around 0.57 ± 0.02 and 0.57 ± 0.04, for the same respective formulations. The length efficiency factors showed higher values than the orientation efficiency factors, revealing its higher impact on the flexural modulus. Considering that the orientation efficiency factor was related to the mean orientation angle of the fibers inside the composite, it was worth determining the theoretical mean orientation angle, to evaluate the degree of alignment of the ONP fibers inside the composites. Equations 7, 8, and 9 were used to compute the limit and theoretical mean orientation angles, for rectangular and triangular distributions, of which the results are presented in Table 3.

Table 3. Limit Angle and Mean Orientation Angle of the ONP for the Rectangular and Triangular Distributions

Composite	Rectangular		Triangular	
	α_0	α	α_0	α
PP20ONP0MAPP	49.0	27.3	72.5	38.4
PP30ONP0MAPP	48.9	27.3	72.3	38.4
PP40ONP0MAPP	52.7	29.2	78.5	40.9
PP50ONP0MAPP	49.5	27.6	73.3	38.8
PP20ONP6MAPP	45.6	25.5	66.9	36.0
PP30ONP6MAPP	54.7	30.2	82.0	42.3
PP40ONP6MAPP	51.3	28.5	76.3	40.0
PP50ONP6MAPP	48.7	27.1	71.9	38.2

The theoretical mean values of orientation angles for the rectangular and triangular distributions were of 27.4 ± 0.5º and 39.1 ± 0.7º, respectively. In previous works (Serrano *et al.* 2014a), the analysis of the orientation angle from the tensile strength results was found at 43º; therefore, the triangular distribution seemed more consistent with this previous analysis. Nonetheless, the orientation factors for the strength and the modulus can deliver different mean theoretical orientation angles (Reixach *et al.* 2013a; Granda *et al.* 2016b). A triangular distribution only approximates the semi-random distribution of fibers inside the composite.

As mentioned in the Experimental section, the mRoM does not include explicitly the morphology of the reinforcements in its formulation. Thus, in order to evaluate the impact of the morphology and triangulate the obtained intrinsic micromechanics parameters, the Tsai and Pagano model and the Halpin and Tsai equations were applied (Table 4).

Table 4. Intrinsic Flexural Modulus of the ONP Obtained from Halpin and Tsai Equation and Micromechanics of the Flexural Modulus of the Composites

Composite	E_f^F (GPa)	η_e	η_l	η_o
PP20ONP0MAPP	19.13	0.458	0.845	0.542
PP30ONP0MAPP	18.83	0.470	0.865	0.543
PP40ONP0MAPP	15.55	0.430	0.860	0.501
PP50ONP0MAPP	16.83	0.473	0.882	0.536
PP20ONP6MAPP	22.44	0.485	0.843	0.575
PP30ONP6MAPP	15.29	0.407	0.863	0.472
PP40ONP6MAPP	16.80	0.437	0.860	0.509
PP50ONP6MAPP	17.75	0.474	0.881	0.538

The mean value for the intrinsic flexural modulus was 17.8 ± 2.3 GPa. This value differed from the one obtained from the Hirsh equation, 16.5 ± 1.2 GPa. However, an independent two sample t-test revealed that there was no evidence, at a 95% confidence rate, of the two values being different. The same was valid for the rest of the micromechanics parameters.

CONCLUSIONS

1. A method to compute the intrinsic flexural modulus of the reinforcements based on the fiber tensile modulus factor and the fiber flexural modulus factor was proposed. The values delivered by this method were compared with those obtained with the Hirsch equation and Tsai and Pagano model and the Halpin and Tsai equations. The results corroborated the usefulness of the method for ONP reinforced PP composites. An intrinsic flexural modulus for ONP fibers was established at 17.8 GPa by the Halpin-Tsai equations.
2. The ONP reinforced PP composites yielded lower flexural modulus than those of SGW or GF reinforced materials. Still, ONP composites with 50 wt% fiber content had comparable flexural modulus to 10 wt% GF reinforced composites. The use of 50 wt% of a renewable phase reduced the amount of oil-based matter.
3. The intrinsic flexural modulus of ONP fibers was inferior to that of mechanical pulp used for papermaking. The main differences for these types of fibers were the presence of $CaCO_3$ and inks, as well as the lower mean fiber lengths. However, more research is needed to isolate these factors and evaluate its impact on the flexural modulus of these composites.
4. The orientation factors rendered values around 0.5 and the length factor around 0.86, in line with the literature. It was possible evaluating a theoretical 39.1º mean orientation angle of the fibers, assuming a triangular distribution.

ACKNOWLEDGMENTS

The authors are grateful for the support of the University of Girona and its researcher progress program.

REFERENCES CITED

Al-Oqla, F. M., and Sapuan, S. (2014). "Natural fiber reinforced polymer composites in industrial applications: Feasibility of date palm fibers for sustainable automotive industry," *J. Clean. Prod.* 66, 347-354. DOI: 10.1016/j.jclepro.2013.10.050

Ali, M. A., El-Nemr, K. F., and Hassan, M. M. (2011). "Waste newsprint fibers for reinforcement of radiation-cured styrene butadiene rubber-based composites - Part I: Mechanical and physical properties," *J. Reinf. Plast. Comp.* 30(8), 721-737. DOI: 10.1177/0731684411407949

Araújo, R. S., Rezende, C. C., Marques, M. F. V., Ferreira, L. C., Russo, P., Emanuela, M. E., Avolio, R., Avella, M., and Gentile, G. (2017). "Polypropylene-based composites reinforced with textile wastes," *J. Appl. Polym. Sci.* 134(28), 45060-n/a. DOI: 10.1002/app.45060

Cox, H. L. (1952). "The elasticity and strength of paper and other fibrous materials," *British J. Appl. Phys.* 3(MAR), 72-79. DOI: 10.1088/0508-3443/3/3/302

Delgado-Aguilar, M., Julián, F., Tarrés, Q., Méndez, J. A., Mutjé, P., and Espinach, F. X. (2017). "Bio composite from bleached pine fibers reinforced polylactic acid as a replacement of glass fiber reinforced polypropylene, macro and micro-mechanics of the Young's modulus," *Compos. Part B-Eng.* 125, 203-210. DOI: 10.1016/j.compositesb.2017.05.058

El Mansouri, N.-E., Espinach, F. X., Julian, F., Verdaguer, N., Torres, L., Llop, M. F., and

Mutje, P. (2012). “Research on the suitability of organosolv semi-chemical triticale fibers as reinforcement for recycled HDPE composites,” *BioResources* 7(4), 5032-5047. DOI: 10.15376/biores.7.4.5032-5047
Espinach, F., Delgado-Aguilar, M., Puig, J., Julian, F., Boufi, S., and Mutjé, P. (2015). “Flexural properties of fully biodegradable alpha-grass fibers reinforced starch-based thermoplastics,” *Compos. Part B-Eng.* 81, 98-106. DOI: 10.1016/j.compositesb.2015.07.004
Espinach, F. X., Granda, L. A., Tarrés, Q., Duran, J., Fullana-i-Palmer, P., and Mutjé, P. (2017a). “Mechanical and micromechanical tensile strength of eucalyptus bleached fibers reinforced polyoxymethylene composites,” *Compos. Part B-Eng.* 116, 333-339. DOI: 10.1016/j.compositesb.2016.10.073
Espinach, F. X., Julian, F., Verdaguer, N., Torres, L., Pelach, M. A., Vilaseca, F., and Mutje, P. (2013). “Analysis of tensile and flexural modulus in hemp strands/polypropylene composites,” *Compos. Part B-Eng.* 47, 339-343. DOI: 10.1016/j.compositesb.2012.11.021
Espinach, F. X., Mendez, J. A., Granda, L. A., Pelach, M. A., Delgado-Aguilar, M., and Mutje, P. (2017b). “Bleached kraft softwood fibers reinforced polylactic acid composites, tensile and flexural strengths,” in: *Natural Fibre-reinforced Biodegradable and Bioresorbable Polymer Composites*, A. Lau (ed.), Elsevier, UK.
Franco-Marques, E., Mendez, J. A., Pelach, M. A., Vilaseca, F., Bayer, J., and Mutje, P. (2011). “Influence of coupling agents in the preparation of polypropylene composites reinforced with recycled fibers,” *Chem. Eng. J.* 166(3), 1170-1178. DOI: 10.1016/j.cej.2010.12.031
Fukuda, H., and Kawata, K. (1974). “On Young’s modulus of short fibre composites,” *Fibre Sci. Technol.* 7(3), 207-222. DOI: 10.1016/0015-0568(74)90018-9
Girones, J., Lopez, J. P., Vilaseca, F., Bayer R, J., Herrera-Franco, P. J., and Mutje, P. (2011). “Biocomposites from Musa textilis and polypropylene: Evaluation of flexural properties and impact strength,” *Compos. Sci. Technol.* 71(2), 122-128. DOI: 10.1016/j.compscitech.2010.10.012
Granda, L. A., Espinach, F. X., Lopez, F., Garcia, J. C., Delgado-Aguilar, M., and Mutje, P. (2016a). “Semichemical fibres of *Leucaena collinsii* reinforced polypropylene: Macromechanical and micromechanical analysis,” *Compos. Part B-Eng.* 91, 384-391. DOI: 10.1016/j.compositesb.2016.01.035
Granda, L. A., Espinach, F. X., Mendez, J. A., Tresserras, J., Delgado-Aguilar, M., and Mutje, P. (2016b). “Semichemical fibres of *Leucaena collinsii* reinforced polypropylene composites: Young’s modulus analysis and fibre diameter effect on the stiffness,” *Compos. Part B-Eng.* 92, 332-337. DOI: 10.1016/j.compositesb.2016.02.023
Gu, F., Hall, P., and Miles, N. J. (2016). “Performance evaluation for composites based on recycled polypropylene using principal component analysis and cluster analysis,” *J. Clean. Prod.* 115, 343-353. DOI: 10.1016/j.jclepro.2015.12.062
Hashemi, S. (2008). “Tensile and flexural properties of injection-moulded short glass fibre and glass bead ABS composites in the presence of weldlines,” *J. Mater. Sci.* 43(2), 721-731. DOI: 10.1007/s10853-007-2187-4
Hirsch, T. (1962). “Modulus of elasticity of concrete affected by elastic moduli of cement paste matrix and aggregate,” *Journal of American Concrete Institute* 59(3), 427-451. DOI: 10.14359/7924
Huda, M. S., Drzal, L. T., Misra, M., Mohanty, A. K., Williams, K., and Mielewsky, D. F. (2005). “A study on biocomposites from recycled newspaper fiber and poly(lactic acid)," *Industrial & Engineering Chemistry Research* 44(15), 5593-5601. DOI: 10.1021/ie0488849
International, A. (2013). “Standard Practice for Conditioning Plastics for Testing,” Vol.

ASTM D618-13, ASTM International, West Conshohocken, PA.
International, A. (2017). "Standard test methods for flexural properties of unreinforced and reinforced plastics and electrical insulating materials," Vol. ASTM D790 - 17, ASTM International, West Conshohocken, PA.
Jiménez, A. M., Delgado-Aguilar, M., Tarrés, Q., Quintana, G., Fullana-i-Palmer, P., Mutjé, P., and Espinach, F. X. (2017). "Sugarcane bagasse reinforced composites: Studies on the Young's modulus and macro and micro-mechanics," *BioResources* 12(2), 3618-3629. DOI: 10.15376/biores.12.2.3618-3629
Joshi, S. V., Drzal, L. T., Mohanty, A. K., and Arora, S. (2004). "Are natural fiber composites environmentally superior to glass fiber reinforced composites?," *Compos. Part A-Appl. S.* 35(3), 371-376. DOI: 10.1016/j.compositesa.2003.09.016
Kong, C., Lee, H., and Park, H. (2016). "Design and manufacturing of automobile hood using natural composite structure," *Compos. Part B-Eng.* 91, 18-26. DOI: 10.1016/j.compositesb.2015.12.033
Koronis, G., Silva, A., and Fontul, M. (2013). "Green composites: A review of adequate materials for automotive applications," *Compos. Part B-Eng.* 44(1), 120-127. DOI: 10.1016/j.compositesb.2012.07.004
Krenchel, H. (1964). *Fibre Reinforcement*, Akademisk Forlag, Copenhagen.
Lopez, J. P., Girones, J., Mendez, J. A., Pelach, M. A., Vilaseca, F., and Mutje, P. (2013). "Impact and flexural properties of stone-ground wood pulp-reinforced polypropylene composites," *Polym. Composite.* 34(6), 842-848. DOI: 10.1002/pc.22486
Lopez, J. P., Mendez, J. A., El Mansouri, N. E., Mutje, P., and Vilaseca, F. (2011). "Mean intrinsic tensile properties of stone groundwood fibers from softwood," *BioResources* 6(4), 5037-5049. DOI: 10.15376/biores.6.4.5037-5049
Lopez, J. P., Mendez, J. A., Espinach, F. X., Julian, F., Mutje, P., and Vilaseca, F. (2012a). "Tensile strength characteristics of polypropylene composites reinforced with stone groundwood fibers from softwood," *BioResources* 7(3), 3188-3200. DOI: 10.15376/biores.7.3.3188-3200
Lopez, J. P., Mutje, P., Pelach, M. A., El Mansouri, N. E., Boufi, S., and Vilaseca, F. (2012b). "Analysis of the tensile modulus of PP composites reinforced with Stone grounwood fibers from softwood," *BioResources* 7(1), 1310-1323. DOI: 10.15376/biores.7.1.1310-1323
Lykidis, C., Parnavela, C., Goulounis, N., and Grigoriou, A. (2012). "Potential for utilizing waste corrugated paper containers into wood composites using UF and PMDI resin systems," *Eur. J. Wood. Wood Prod.* 70(6), 811-818. DOI: 10.1007/s00107-012-0618-7
Nedjma, S., Djidjelli, H., Boukerrou, A., Benachour, D., and Chibani, N. (2013). "Deinked and acetylated fiber of newspapers," *J. Appl. Polym. Sci.* 127(6), 4795-4801. DOI: 10.1002/app.38048
Oliver-Ortega, H., Llop, M., Espinach, F. X., Tarrés, Q., Ardanuy, M., and Mutjé, P. (2018a). "Study of the flexural modulus of lignocellulosic fibers reinforced bio-based polyamide11 green composites," *Compos. Part B-Eng.* 152, 126-132. DOI: 10.1016/j.compositesb.2018.07.001
Oliver-Ortega, H., Méndez, J. A., Reixach, R., Espinach, F. X., Ardanuy, M., and Mutjé, P. (2018b). "Towards more sustainable material formulations: A comparative assessment of PA11-SGW flexural performance versus oil-based composites," *Polymers-Basel* 10(4), 440. DOI: 10.3390/polym10040440
Osman, H., Ismail, H., and Mustapha, M. (2010). "Effects of maleic anhydride polypropylene on tensile, water absorption, and morphological properties of recycled newspaper filled polypropylene/natural rubber composites," *J. Compos. Mater.* 44(12), 1477-1491. DOI: 10.1177/0021998309359212
Pickering, K. L., Efendy, M. A., and Le, T. M. (2016). "A review of recent developments in

natural fibre composites and their mechanical performance," *Compos. Part A-Appl. S.* 83, 98-112. DOI:
Pil, L., Bensadoun, F., Pariset, J., and Verpoest, I. (2016). "Why are designers fascinated by flax and hemp fibre composites?," *Compos. Part A-Appl. S.* 83, 193-205. DOI: 10.1016/j.compositesa.2015.11.004
Reixach, R., Espinach, F. X., Franco-Marquès, E., Ramirez de Cartagena, F., Pellicer, N., Tresserras, J., and Mutjé, P. (2013a). "Modeling of the tensile moduli of mechanical, thermomechanical, and chemi-thermomechanical pulps from orange tree pruning," *Polym. Composite.* 34(11), 1840-1846. DOI: 10.1002/pc.22589
Reixach, R., Franco-Marquès, E., El Mansouri, N.-E., de Cartagena, F. R., Arbat, G., Espinach, F. X., and Mutjé, P. (2013b). "Micromechanics of mechanical, thermomechanical, and chemi-thermomechanical pulp from orange tree pruning as polypropylene reinforcement: A comparative study," *BioResources* 8(3), 3231-3246. DOI: 10.15376/biores.8.3.3231-3246
Rodriguez, M., Rodriguez, A., Bayer R. J., Vilaseca, F., Girones, J., and Mutje, P. (2010). "Determination of corn stalk fibers' strength through modeling of the mechanical properties of its composites," *BioResources* 5(4), 2535-2546. DOI: 10.15376/biores.5.4.2535-2546
Sahin, A., Tasdemir, H. M., Karabulut, A. F., and Gürü, M. (2017). "Mechanical and thermal properties of particleboard manufactured from waste peachnut shell with glass powder," *Arabian Journal forScience and Engineering* 5(4), 2535-2543. DOI: 10.1007/s13369-017-2427-0
Sanomura, Y., and Kawamura, M. (2003). "Fiber orientation control of short-fiber reinforced thermoplastics by ram extrusion," *Polym. Composite.* 24(5), 587-596. DOI: 10.1002/pc.10055
Serra, A., Tarrés, Q., Claramunt, J., Mutjé, P., Ardanuy, M., and Espinach, F. (2017). "Behavior of the interphase of dyed cotton residue flocks reinforced polypropylene composites," *Compos. Part B-Eng.* 128, 200-207. DOI: 10.1016/j.compositesb.2017.07.015
Serrano, A., Espinach, F. X., Julian, F., del Rey, R., Mendez, J. A., and Mutje, P. (2013). "Estimation of the interfacial shears strength, orientation factor and mean equivalent intrinsic tensile strength in old newspaper fiber/polypropylene composites," *Compos. Part B-Eng.* (50), 232-238. DOI: 10.1016/j.compositesb.2013.02.018
Serrano, A., Espinach, F. X., Tresserras, J., del Rey, R., Pellicer, N., and Mutje, P. (2014a). "Macro and micromechanics analysis of short fiber composites stiffness: The case of old newspaper fibers-polypropylene composites," *Mater. Design* 55, 319-324. DOI: 10.1016/j.matdes.2013.10.011
Serrano, A., Espinach, F. X., Tresserras, J., Pellicer, N., Alcala, M., and Mutje, P. (2014b). "Study on the technical feasibility of replacing glass fibers by old newspaper recycled fibers as polypropylene reinforcement," *J. Clean. Prod.* 65, 489-496. DOI: 10.1016/j.jclepro.2013.10.003
Siengchin, S. (2017). "Potential use of 'green' composites in automotive applications," Budapest Univ. Technol. & Econ. Dept. Polymer Eng., Muegyetem RKP 3, Budapest, H-1111, Hungary.
Singh, B., Gupta, M., Tarannum, H., and Randhawa, A. (2011). "Natural fiber-based composite building materials," in: *Cellulose Fibers: Bio-and Nano-Polymer Composites*, Springer, pp. 701-720.
Summerscales, J., Virk, A., and Hall, W. (2013). "A review of bast fibres and their composites: Part 3-Modelling," *Compos. Part A – Appl. S.* 44, 132-139. DOI: 10.1016/j.compositesa.2012.08.018
Virk, A. S., Hall, W., and Summerscales, J. (2012). "Modulus and strength prediction for

natural fibre composites," *Mater. Sci. Tech.* 28(7), 864-871. DOI: 10.1179/1743284712y.0000000022

Yuan, X. H., Zhang, Y. X., and Zhang, X. F. (1999). "Maleated polypropylene as a coupling agent for polypropylene-waste newspaper flour composites," *Journal of Applied Polymer Science*. 71(2), 333-337. DOI: 10.1002/(sici)1097-4628(19990110)71:2<333::AID-APP17>3.0.CO;2-Q

Article submitted: October 19, 2018; Peer review completed: November 26, 2018; Revisions accepted: February 12, 2019. Published: March 13, 2019.
DOI: 10.15376/biores.14.2.3578-3593