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Macro and micromechanics analysis of short fiber composites stiffness: The case of old newspaper fibers–polypropylene composites

A. Serrano [b], F.X. Espinach [a,e,*], J. Tresserras [a], R. del Rey [c], N. Pellicer [d], P. Mutje [b]

[a] Design, Development and Product Innovation, Dpt. of Organization, Business Management and Product Design, Spain
[b] Laboratory of Paper Engineering and Polymer Materials, Dpt. of Chemical Engineering, University of Girona, Spain
[c] Instituto para la Gestión Integrada de las Zonas Costeras, IGIC, Escola Politècnica Superior de Gandia, Universitat Politècnica de València, Spain
[d] Dpt. of Chemical Engineering, University of Girona, Spain
[e] Escola Politecnica Superior, Avda., Lluis Santalo, s/n, 17071 Girona, Spain

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ABSTRACT

Stiffness is one of the most relevant characteristics of composite materials. Natural wood fibers have demonstrated their ability to increase the Young's moduli of composite materials, and old newspapers are a potential source of reinforcing fibers for composite materials. There are some micromechanic models to predict the Young's modulus of composite materials, and one of the input data is the intrinsic modulus of their fibers. This intrinsic modulus is a value which is difficult or impossible to measure in the case of wood fibers, due to their measures. This paper evaluates the stiffening abilities of old newspaper fibers and the possibility to back calculate the value of the intrinsic Young's modulus by means of micromechanic models. Different percentages of old newspaper fibers were compounded with polypropylene (PP). Micromechanics of the fibers were obtained using Hirsch model, Cox–Krenchel's model, Tsai–Pagano model and Halpin–Tsai equations. The most important results were the average intrinsic Young's modulus of the fibers, the mean orientation angle and the mean modulus efficiency factor.

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1. Introduction

In recent years, wood fiber and natural fibers have gained a significant interest as a reinforcing material for commercial thermoplastics [1–6]. Some advantages of natural fibers as opposed to other reinforcing materials are their high availability, their biodegradability, and their relative low cost and density. Newspapers usually contain a high percentage of recycled fibers from wood and mineral fillers, and therefore, used newspapers become a potential source of reinforcing fibers [7–11]. In spite of their advantages, the use of cellulose fibers from newspaper as reinforcement elements for thermoplastics has not been extensively investigated [7].

Virgin wood fibers and cellulosic fibers from recycled papers, when used as reinforcement with the adequate coupling agents, are able to increase both the elastic moduli and the strength of the composite materials [12–19]. The strength of natural fiber reinforced composites is highly influenced by the incompatibility between the hydrophilic fibers and the hydrophobic polymers. The most common way to improve the interface is the addition of coupling agents [10,17,19–22]. However that treatment slightly affects the stiffness of the natural fiber reinforced composites.

For structural and semi structural applications, the most relevant properties are probably stiffness and dimensional stability [23]. Lopez et al. [2] found a 1.09 ratio between the Young modulus of 30% glass fiber (GF)-polypropylene and 50% stone groundwood mechanical pulp – polypropylene composites. Old newspaper fibers (ONF), due to their good specific properties, could achieve their own space of competitiveness in semi structural applications [24].

In order to predict the elastic properties of the composite materials it is necessary to know the intrinsic Young's modulus of the fibers, defined as the average slope in the stress–strain curve in the strain interval from 0% to 0.3% [23]. Unfortunately, due to their characteristic morphology, the experimental evaluation of the Young's moduli of ONF is costly and difficult, and sometimes impossible. The mechanical properties of composites can also be predicted by means of micromechanical models. The models are based on different assumptions and experimental data. The most common one's are based on the rule of mixtures (ROM) and can be used to back calculate the intrinsic properties of the reinforcing fibers [2,25,26]. In the case of short fiber semi-aligned composites, the most commonly used models to predict the Young's modulus or the intrinsic Young's modulus, are modified rules of mixtures

* Corresponding author at: Escola Politecnica Superior, Avda., Lluis Santalo, s/n, 17071 Girona, Spain. Tel.: +34 972 418 920; fax: +34 972 418 399.
E-mail address: francisco.espinach@udg.edu (F.X. Espinach).

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(mROM) [27], Hirsch model [28], and Halpin–Tsai equations with Tsai–Pagano methods [2,29–31].

In this work composites from polypropylene (PP) and old newspaper fibers (ONF) were formulated, prepared and tested to mechanically characterize its Young's moduli. This study complements the analysis of the tensile strength of these composites carried out previously [8], and is focused on the stiffness of the composites. The moduli were analyzed from a macromechanical point of view, and their micromechanical aspects were evaluated. The values of the intrinsic Young's moduli of the fibers were obtained by means of Hirsch model [28], and also by means of Halpin–Tsai equations with Tsai–Pagano methods [29–31] and then compared, to assess the influence of the aspect ratio of the fibers on the values. By means of Cox–Krenchel equations [32,33] were studied the length efficiency factors, and also were deduced the fiber orientation factors. The mean orientation angles of the fibers, assuming a rectangular distribution of the fibers inside the matrix (square packing), were also computed.

2. Materials and methods

2.1. Materials

Old newspaper, containing 85% of wood recycled fibers and 15% of calcium carbonate mineral filler, was supplied by Punt Diari, printed by Rotimprès (Spain).

The composites were prepared using homopolymer polypropylene (PP) (Isplen PP090 G2M) which was provided by Repsol–YPF (Tarragona, Spain) as the polymer matrix. Polypropylene functionalized with maleic anhydride (MAH–PP) (Epolene G3015), which was acquired from Eastman Chemical Products (Spain) and used as coupling agent. Diethyleneglycol dimethyl ether (diglyme) was supplied by Clariant and was used as dispersing agent. Decahydronaphthalene (decalin), supplied by Fisher Scientific, was used to dissolve the PP matrix in the fiber extraction from composites process.

2.2. Disintegration of the old newspaper

The old newspapers were cut into pieces of 10 × 10 cm, approximately and were soaked in water for 3 h at 50 °C, with a 1% of NaOH, in a heated stainless steel vessel. Then the cuts were submitted to the disintegration process by means of a pilot scale pulper (Pucel Cell from Metrotech, France) equipped with an helicoidal rotor, deflectors, and with an effective volume of 20 l. The disintegration was performed at 20 rev/s rotor speed, at a temperature of 50 °C, and 10% consistency. Afterwards the pulped material was filtered and oven dried at 80 °C. Following, the fibers were dispersed in a water–diglyme (1:3) mixture. The use of diglyme in the previous step limits the formation of hydrogen bonds between the cellulosic fibers [34] . A 5% of the $CaCO_3$ was lost during the disintegration and individualization processes.

2.3. Composite preparation

PP composite materials comprising 20–50 wt% of ONF were compounded by means of a Brabender internal mixing machine. The mixing process was performed at 80 rpm rotor speed and a temperature of 180 °C for 10 min. The obtained blends were ground by means of a knives mill, dried and stored for at least 24 h before processing.

2.4. Composite processing

The samples for the tensile test were produced with a steel mould in an injection-molding machine (Meteor 40, Mateu & Solé). Ten test specimens from each obtained composite blend were used for the experiment. The processing temperatures were 175, 175, and 190 °C (the machine has three heating areas), the last corresponding to the injection nozzle. First and second pressures were 120 and 37.5 kgf/cm^2, respectively. Standard composite specimen samples (approx. 160 × 13.3 × 3.2 mm) were obtained and used to measure the tensile properties.

2.5. Mechanical characterization

The specimens were stored in a Dycometal conditioning chamber at 23 °C and 50% relative humidity for 48 h, in agreement with the ASTM: D638 standard. Afterwards, composites were assayed in a Universal testing machine (Instron™ 1122), fitted with a 5 kN load cell and operating at a rate of 2 mm/min. Young's modulus was analyzed using extensometer in dog-bone specimens. Results were obtained from the average of at least 5 samples.

2.6. Fiber extraction from composites

Reinforcing fibers were extracted from composites by matrix solubilization using a Soxhlet apparatus and decalin as solvent. Small pieces of composites were cut and placed inside a cellulose filter and set into the Soxhelt equipment. A small cotton tab was used to prevent the fibers from getting out of the filtering tube. The fiber extraction was completed after 24 h. Once the fibers were extracted, they were rinsed with acetone and then with distilled water in order to remove the solvent residue. Finally the fibers were dried in an oven at 105 °C for 24 h.

2.7. Determination of the fiber length and diameter

Fiber's length distribution and diameter of the extracted fibers were characterized by means of a MorFi Compact (Morfological fiber analyzer), from Techpap SAS (France). A minimum of two samples were analyzed.

2.8. Density measurement

The density measurement of the composite (ρ^c) was carried out using a pycnometer. Distilled water at 23 °C was used as a reference liquid. The ISO 1183-1 [35] standard was respected throughout this experiment. The density of the fiber (ρ^f) was obtained from: $\rho^c = w^c/((w^m/\rho^m) + (w^f/\rho^f))$, where w^c, w^m, and w^f are the loads in weight of the composite, matrix, and fiber and ρ^m is the density of the matrix.

2.9. Young's modulus modeling approaches

The value of the tensile modulus of natural fibers is very variable [36–39]. In some cases, as for example lignocellulosic fibers from wood or from agro-forestry wastes (corn stalks, rape stalks, hemp core fibers, etc.), the experimental measurement of their elastic modulus is practically impossible. However the intrinsic tensile or flexural modulus of the fibers is a key factor to predict the tensile or flexural modulus for the composite materials. Therefore, in these cases, the best choice is to use mathematical models to estimate the value of the intrinsic elastic modulus of the reinforcements from the tensional or flexural modulus of the composite. Hirsch model (Eq. (1)) [28], is a valid tool to estimate the intrinsic modulus of the reinforcement $\left(E_t^f\right)$ [40–42].

$$E_t^c = \beta \cdot \left(E_t^f V^f + E_t^m (1 - V^f)\right) + (1 - \beta)\frac{E_t^f \cdot E_t^m}{E_t^m \cdot V^f + E_t^f (1 - V^f)} \qquad (1)$$

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