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Semichemical fibres of *Leucaena collinsii* reinforced polypropylene composites: Young's modulus analysis and fibre diameter effect on the stiffness

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ABSTRACT

Leucaena genus offers environmental benefits related with soil recovering. The use of natural fibres as reinforcement in composites materials is a current technology for strengthening and stiffening polymeric materials. Besides, some authors suggest the idea that the diameter has a strong influence on the material stiffness. The material's stiffness is one of the most important design parameters. This work studies the *Leucaena collinsii* influence on the polypropylene composite materials stiffness. A tensile test was performed for measuring the Young's moduli from composite materials reinforced with 20–50% of fibre. Furthermore, the diameter influence on that property has been studied through modelling. The mechanical results show a high *L. collinsii* stiffness contribution, increasing its stiffness a 400%. The fibre intrinsic Young modulus was located in the high range of natural fibre's intrinsic Young's modulus. The fibre diameter rendered little influence on the fibre intrinsic stiffness.

1. Introduction

One of the most relevant material properties when designing structural applications are the dimensional stability and the stiffness [1], as it is frequently pretended that the designs suffer low deformation during its application. The Young's modulus measures the average slope in the elastic region [1], which is related with the stiffness and is experimentally measured between the strain interval of 0.05–0.25%.

In a composite material, the resulting Young's modulus is mainly related with the volumetric fraction of the fibre in the composite material, the matrix Young's modulus and the intrinsic Young's modulus of the fibre. Despite it is difficult or in some cases impossible to measure the intrinsic Young's modulus of short fibres nowadays, it is easy to calculate it by the application of different models. The most frequently used are the modified rule of mixtures (RoM), the Hirsch model and Halpin–Tsai equations.

The combined properties between matrix and reinforcement in polymer composite materials offer different advantages such as good specific properties which led them, especially when reinforced with mineral fibres (glass fibres), to have a strong presence in aerospacial, automotive, construction and sporting industry [2].

The addition of wood fibres is an industrial mechanism to increase the stiffness as well as the strength by combining the properties of each phase. The main application sectors are construction and automotive [3]. Nevertheless, it has been proved that they can also be used in structural applications like in pump producing [4], or in rope making [5].

A great number of studies about using wood fibres for reinforcing thermoplastic polymeric matrixes can be found in the literature [6–11]. Although natural fibres perform a reasonably good enhancement on the composite material mechanical properties, they show lower reinforcement effect than mineral fibres such as glass fibres (GF) [12]. However, the higher flexibility of natural fibres when compared with glass fibres leads them to a higher recyclability [13], fact that is gaining importance as an environmental benefit.

Regarding the matrix, the use of thermoplastic polymers has a clear environmental advantage when compared with thermosets. The non-existing chemical bonds between different polymeric chains allow the material to be melted and recycled [2]. Nevertheless, thermoset polymers allow an easy fibre orientation,

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alignment and to obtain different textile structures when working with bast fibres [14,15]. It is also noteworthy that natural fibres have been also studied as reinforcement in ceramic matrixes composites, especially destined to building and constructive applications [16,17].

In the literature are different works that evaluate the fibre diameter influence on the fibre intrinsic Young's modulus. Apparently, the fibre diameter is strongly linked with the intrinsic tensile stiffness of the fibre [18–21]. However, other studies, which analyse the Young's moduli of fibres with different diameters, show that the fibre stiffness is not influenced by the fibre diameter [22], due to variation in the cellulose content for the location randomness, besides the crack defects along every fibre.

Although the wide range of fibres studied for reinforcing composite materials, regarding *Leucaena collinsii* (LCN), no previous works have been published using it in composites materials production. Despite not being studied in that field, the fast growing characteristics on poor soils of *Leucaena* genus [23], together with the ability of legumes to fix nitrogen in the ground, have proved that they show good soil recovering characteristics [24–26]. This big environmental potential has led some authors to investigate the use of *Leucaena* genus in different fields such as papermaking and biomass production [27].

In this work, LCN semichemical fibres coming from pruning residues was used as reinforcement in thermoplastic composite production. Besides, composite's Young's moduli were measured through tensile test in order to analyse the stiffness evolution when increasing the LCN content. Furthermore, the intrinsic Young's modulus of the fibre composite reinforced with a 30% w/w was studied, as well as the orientation angle of the fibres in the bosom of the matrix. Additionally, and in order to corroborate the fibre diameter influence on that property, a diameter modelling was performed.

2. Materials and methods

2.1. Materials

Polypropylene PP090 K2M, used as polymeric matrix, was kindly supplied by Repsol-YPF (Tarragona, Spain).

The reinforcement was obtained from subjecting a *L. collinsii* fibre supplied by University of Huelva (Huelva, Spain) to a chemical and thermal treatment.

In order to improve the interface quality of the composite material, a maleic anhydride grafted polypropylene was used. It was an Epolene G3015 supplied by Eastman Chemical Products (San Roque, Spain). It had a 24,800 Da Mn and a 15 mg KOH/g acid number.

Decahydronaphthalene, supplied by Scharlau, S.L. (Sentmenat, Spain) was used as solvent in the polypropylene extraction for recovering the fibres from the samples.

2.2. Composite and sample obtaining

A kinetic mixer was used for obtaining the composite material. The materials were introduced through the machine's feeder at 300 rpm and, after the fibre and matrix were inside the mixing room, the speed was enhanced to 2500 rpm. When the blend temperature exceeded 190 °C, the composite was discharged and turned to pellet in a knives mill. Composite materials with fibre contents from 20 to 50% were obtained. All the composites added a 6% of MAPP w/w_{fibre}. From now on the materials will be referred as PP + XLCN + 6M, were PP is the matrix, *X* is the weight percentage of LCN fibres, and 6M the 6% of MAPP.

Afterwards, the composite materials were injected according to ISO 527-1:2000 shape specimens standard with a Meteor-40 injection machine.

The samples were stabilized for 48 h in a climatic chamber at 23 °C and 50% of relative humidity.

2.3. Tensile test

The standard ISO 527-1:2000 was followed in this study. For every material, ten samples were tested to tensile in a DSC-10 dynamometer supplied by IDMtest (San Sebastián, Spain) for determining the Young's modulus. An MFA 2 extensometer was used for a more precise deformation measurement.

2.4. Fibre morphology determination

The fibre diameter and length were determined after injection moulding. The fibres were recovered from the composite material samples by extracting the polypropylene matrix with decahydronaphthalene. The lengths and diameters were measured following the standard ISO/FDIS 160652 by a MorFi equipment.

3. Results and discussion

3.1. Young's modulus analysis

As it is found in the literature, the addition of more rigid and not soluble components to a polymer matrix, increases the composite material stiffness [8,28–30].

Table 1 shows the measured Young's moduli and the strain when the stress is maximum obtained by tensile test. The standard deviations are given inside brackets.

Table 1 shows that the incorporation of LCN increases the stiffness of the material, performing an enhancement of 400% when a 50% of fibre was added. This phenomenon was due to the addition of a more rigid phase than the matrix [31]. Some authors also suggest that the reinforcement impedes the mobility of the polymers, resulting in an enhancement on the composite material stiffness [8].

Although the reasonably high improvement on the stiffness that LCN confers to the composite when compared with soft ground wood fibres [32], or with old newspaper fibres reinforced composites [33], GF composites render higher Young's modulus values. For example, tensile samples of PP reinforced with a 30% w/w of GF performed a Young's modulus of 5.6 GPa [4], a slightly higher result than the one obtained in this study with a 40% w/w of LCN.

Fig. 1 shows the lineal evolution of the stiffness when increasing contents of LCN were added.

It is foreseeable that the flexural properties of the material will follow a lineal tendency between the same studied percentages [34], although the flexural moduli are expected to be lower than the ones found by tensile test [8].

The lineal evolution of that property is related to a lineal contribution of each component that forms the composite material.

Table 1
Fibre volumetric fractions, Young's modulus and deformation experimental results.

	V^F (%)	E_t^c (GPa)	ε_t^c (%)
PP	0	1.5 (0.1)	9.5 (0.2)
PP + 20LCN + 6M	14.5	3.6 (0.2)	5.7 (0.5)
PP + 30LCN + 6M	22.6	4.3 (0.1)	4.3 (0.3)
PP + 40LCN + 6M	31.2	5.4 (0.2)	3.8 (0.3)
PP + 50LCN + 6M	40.5	6.0 (0.2)	3.3 (0.3)