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Macro and micromechanical preliminary assessment of the tensile strength of particulate rapeseed sawdust reinforced polypropylene copolymer biocomposites for its use as building material

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HIGHLIGHTS

- Value was added to Rapeseed sawdust by using it as reinforcement.
- The composites showed competitive tensile strength.
- Composites with 50% byproduct content showed higher tensile strength than the oil-based matrix.
- Micromechanical models were used to model the behavior of the interphase of the composites.

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ABSTRACT

The growing environmental awareness promotes the research of greener and more sustainable materials for their use in building materials. Polyolefin-based wood plastic composites (WPCs) have attracted the attention of researchers in the last decades mainly due to their improved mechanical properties, low weight and low cost. On the other hand, rapeseed is one of the most extended harvests in the world and their production is expected to increase in the next years, according to FAO (Food and Agriculture Organization). Rapeseed is mainly used for the production of biodiesel, and moving from an oil-based economy to a bio-based economy will presumably involve increases of rapeseed production. Its harvest produces high amount of agroforestry residue in the shape of integral stems which can be exploited as polymer reinforcing element, although very little information is found in the literature in this respect. In this work, the viability of rapeseed sawdust reinforced polypropylene copolymer composites was analysed regarding its mechanical tensile strength. Besides, the coupling agent percentage to ensure the best performance was also studied. The tensile strength of the produced composites has been modelled using well-known micromechanical models which allow the determination of the fibre and matrix contributions to the composite strength. The results showed that competitive green composite materials can be obtained using a byproduct of rapeseed harvesting.

1. Introduction

The growing awareness towards sustainability leads to the use of greener materials by the industrial design, automotive and building industries [1–4]. In some cases, like in building industry, the use of green-materials is regulated by governments and also included in the engineers and architects curricula [5]. Under these policies, one main objective is trying to prevent a future scarcity of raw materials. Within building sector, materials must accomplish specific requirements according to their final use. While interiors household are exposed to dry environments, the materials used in exteriors must show water resistance or keep enough properties under high humidity conditions. Recently, polymer-based composites and biocomposites materials have gained attention thanks to the range of properties that they can provide. As for matrices, the

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use of biodegradable polymers is limited due to their fast degradation under humid conditions combined with temperature [6]. Therefore, the use of polyolefins reinforced with natural fibres can deliver greener materials for exterior furniture or cladding. Polyolefins like polypropylene (PP), polyethylene (PE) or polyvinylchloride (PVC) are oil-based materials, which are not compostable but can be recycled [5,7]. Moreover, polyolefin show a hydrophobic behaviour being able to work under humid conditions. These matrices have been used successfully to prepare wood plastic composites (WPC) [8,9].

Wood plastic composites (WPCs), consisting of lignocellulosic fibres embedded in an oil-polymer matrix, have been investigated and developed over the last 40 years [10,11]. Several studies have been focused on their thermal properties [12,13], degradation [14,15], rheological properties [16,17], mechanical properties [18,19], wood particle species and size [20,21], manufacture techniques [22,23], fire performance [24,25], and water resistance [26–28]. Most of these works were devoted to the analysis of WPC properties, and only a few about their ability as building material.

A literature review on product development in the building industry shows an increasing interest by WPC [29,30]. Biocomposites, such as WPCs, were used as decking and cladding [31,32] (Fig. 1). The use of natural fibres provides attracting advantages such as reduced cost, lightness, excellent balanced mechanical properties, low environmental impact [11,33]. These products need little maintenance, show high durability and are easy to install [31]. WPCs can be used outdoors (claddings like decking board and tiles or pieces for facades, railings, window frames, fences) as well as indoors (doors, window sills and boards) [10,34,35]. Nevertheless, their use in facade is still a challenge since only few aspects of WPCs have been investigated. Friedrich et al. analysed the standards and defined seven fundamental requirements for the use of WPCs in buildings facades [31,32].

The use of a renewable material as reinforcement / filler satisfies three main objectives; increase the mechanical properties, substitute an expensive material (polyolefin matrix), and decrease the amount of non-renewable phase in the composite [36,37]. Rapeseed is a worldwide produced plant for their oil, animal feeding and biodiesel production. The developed countries use to promote biodiesel, but, nowadays, it is losing support due to the high amount of energy needed for its production. The European Union has reduced the percentage of foreseen production of biofuels from 8.6% to 7%. Nonetheless, Food and Agricultural Organization of the United Nations (FAO) evaluates a 54% increase of rapeseed production up to 2023. In any case, high biomass sub-products from the rapeseed harvesting are delivered annually, which are not really well-exploited. This agroforestry residue can be used as lignocellulosic reinforcement for WPC in a more sustainable form.

In this respect, there are few published works regarding rapeseed-based composites [38–40]. These composites showed reasonable tensile strengths of 15.68, 14.72 and 11.94 MPa for PE-composites with 30, 45 and 60%w/w rapeseed contents, respectively [38]. In this case, the composites were uncoupled, and the drop of the strength when increasing the amount of reinforcement was indicative of feeble interphase. As for rapeseed sawdust-composites, the literature is even more scarce, with, to be best knowledge of the authors, only one interesting work on the water uptake of such materials [38]. The lack of information about the mechanical and thermal properties of rapeseed sawdust reinforced polypropylene composites, encouraged us to prepare a series of studies to assess the competitiveness of such materials for its use as external cladding. One relevant property for an engineering material is the analysis of its tensile strength. This property evaluates the possibilities of the material to be used for structural, semi-structural or decorative purposes. Then, if the tensile strength was not competitive with the readily available materials, conducting other tests would not make sense.

Fig. 1. WPC commercial external panels.

In this work, the compatibility of PP copolymer (PPc) with rapeseed sawdust (RS) and the use of a coupling agent was analysed. A preliminary test was carried out to assess the percentages of coupling agent that ensure good tensile strengths. Following a semi-industrial phase studied the impact of the RS contents in the tensile strength and strain of the composites. Two different mixers were used in both phases. The mixer used in the semi-industrial phase produced reinforcements with a higher aspect ratio. Finally, a micromechanical study allowed obtaining the interfacial shear strength, the orientation factor and a lower bound for the intrinsic tensile strength.

2. Materials and methods

2.1. Materials

PP–PE copolymer traded as Capilene SW 75 AV (Carmel Olefins, Haifa, Israel) was used as matrix. This polypropylene impact copolymer has an improved impact resistance compared to PP homopolymer, with a melt flow index of 65 g/10 min at 230 °C and 2.16 kg and density of 0.905 g/cm^3.

Rapeseed sawdust coming from agricultural residues nearby, provided by Cal Gall (sant Gregori, Girona, Spain) was used as reinforcing element of the PP copolymer. Rapeseed residues were was cut to 1–2 mm in a mill (Agrimsa, Sant Adrià de Besós, Spain) and grinded 3 times afterwards in a Sammic SK-5 food processor (Sammic, Barcelona, Spain). Before grinding, the fibres were dried for 24 h at 80 °C in an oven to facilitate the process. Finally, the material was screened in a sieve of 1 mm size-hole screen to ensure a regularity on the particles sizes.

Eucalyptus sawdust from Eucalyptus globulus was kindly provided by La Montañanesa (Montañana, Zaragoza, Spain). The eucalyptus sawdust is a by-product of the chemical pulp factory and was analysed for comparison purposes.

Maleated polypropylene (MAPP) from Eastman Chemical Products (Middelburg, The Netherlands), with acid number of 15 mg KOH/g and a molecular weight of 47000 g/mol, was used as coupling agent.

Decahydronaphtalene supplied by Scharlau S.L. (Sentmenat, Spain) was used to dissolve the polymer matrix during the fibre's extraction from composites materials.

2.2. Chemical composition

Lignin and hemicellulose contents were determined following the Tappi standard T211 om-93, T-222 om-88 and T223 cm-84. Cellulose amount was determined following the procedure proposed by Wise et al. [41].

2.3. Sample production and tensile characterization

The methodology followed in this work is divided in two different parts (Fig. 2). Initially, a preliminary study with different formulations of coupling agents was performed. Tensile properties of the composites at different MAPP percentages regarding the fibre content were analysed and the optimum amount of coupling agent was determined.

The composite blends were prepared using the internal mixer Brabender® plastograph (Duisburg, Germany). The rotor speed was 80 rpm and the temperature 180 °C. Once the matrix was blend in the mixing chamber, the reinforcement was incorporated and kept mixing for 10 min to ensure a proper dispersion of the fibres.