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Properties of polypropylene composites filled with a mixture of household waste of mate-tea and wood particles

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HIGHLIGHTS

- Polymeric composite was developed using household and industrial residues.
- Effect of waste of mate-tea as filler was physically, chemically and mechanically evaluated.
- Thermal stability of PP decreased with addition of mate-tea waste and eucalypt particles.
- Use of eucalypt particles increased mechanical properties more than use of mate-tea waste.
- Insertion of eucalypt particles increased the hydrophobicity of the composites.

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ABSTRACT

This study presents the preparation of polypropylene composites filled with mixtures of household waste of mate-tea and eucalypt particles and aims to increase the economic value chain of mate-tea. Filler mixtures in proportions ranging from 0% to 60% with a fixed PP matrix at 40% were prepared in order to evaluate only the effect of the filler on thermochemical, physical and mechanical properties and on the morphology. The main findings showed that the addition of filler from natural sources decreased thermal stability of composites, but that the temperature of crystallisation increased. Composites with a higher proportion of wood particles showed higher hydrophobic character; however, only the composites with 60% and 54% of mate-tea waste particles showed significantly higher results for water absorption. The use of eucalypt particles increased more mechanical properties than that of household waste, which proves its efficiency as filler. Nevertheless, the use of household waste also increased the properties of the final product and showed that it can be a good alternative for the use of renewable materials in the production of polymeric composites.

1. Introduction

Several lignocellulosic materials are used as fillers or reinforcement in thermoplastic composites, including fibres of sugarcane, banana, jute, ramie, flax, pineapple, curauá, sisal, cotton, coir, luffa cylindrica [1], guayule biomass [2], heart-of-peach palm sheath [3], bagasse [4], sunflower stalk flour [5], and palm leaf waste [6], as well as fibres and wood particles of different tree species [7–10].

Lignocellulosic materials have become important as fillers or reinforcements in polymer or ceramic matrices due to their advantages in relation to other inorganic or synthetic materials [3]. Some advantages of these materials are that: they are fully and easily recyclable, meeting minimum recycling content requirements; they are non-abrasive to machinery; they have the same performance for lower weight and are stronger (25–30%) for the same weight; they result in non-brittle fracture on impact and are more shatter resistant; they have lower processing energy requirements and a low thermal expansion coefficient; they have a natural appearance, are easily coloured and are low cost, costing less than the base resin [1].

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The low cost of the material is its most attractive factor. According to Ayrilmis and Kaymakci [11] wood flour typically costs about 0.09–0.18 € kg^{-1}, while virgin polypropylene costs about 1.04–1.43 € kg^{-1}. The final cost of wood polymer composites (WPCs) is significantly lower than that of solid or laminated wood decking in price, performance, and processing. This cost can be further reduced if the filler or reinforcement used is from industrial, agricultural or commercial waste.

In general, the polymeric matrix used in these materials is polypropylene (PP) in its various phases, for which β-type shows the best properties [12,13]. However, other matrices used include polyvinyl chloride (PVC) and high-density polyethylene (HDPE) [2,4,14,15]. In the last ten years, a significant increase in scientific literature about the use of such matrices for the formation of composites filled/reinforced with lignocellulosic materials has been observed [5–7,16–21]. These are used for many applications such as decks, fences, floors and automotive components [1].

The mix of a synthetic polymeric matrix with fibres/residues from natural sources influences many properties of the final product, such as physical, chemical, mechanical and anatomical properties. Nevertheless, this mix implies a decrease of wettability, hydrophobicity, thermal stability and adhesion [22,23], which should be evaluated in order to understand the behaviour of the composite when in processing or recycling. On the other hand, residues such as wood particles naturally present higher stiffness and strength than the polymeric matrix, which increases the mechanical properties of the composite [24].

In this study, household wastes of mate-tea and eucalypt particles were chosen as fillers for a PP-matrix. The filler tested is from household waste of mate-tea. Mate-tea is a non-timber product resulting from plant extractivism of *Ilex paraguariensis* planting trees in southern Brazil, Argentina and Uruguay, where it is used in the preparation of a popular tea drink. Per capita consumption of mate-tea in Brazil is ~1.2 kg yr^{-1}, whereas in Argentina and Uruguay is ~5–7 kg yr^{-1} [25]. Considering the population of the three countries, 481.000 tonnes of waste is generated annually from this consumption without any target of aggregate value, which raises the possibility of using this material in the production of thermoplastic composites.

Considering the high levels of consumption of mate-tea in these Latin American countries, any attempt to increase usage of their household waste will add value to this material. Therefore, the incorporation of household waste of mate-tea into products with higher value such as WPC becomes a strategic route to explore options for developing the industrial sector and increasing the economic value of mate. In this context, the present study aims to incorporate particles of household waste of mate-tea into flat-pressed WPCs produced with wood particles of *Eucalyptus benthamii* originated from a fast-growing forest population and to evaluate the thermochemical, physical and mechanical properties and the morphology of these composites.

2. Materials and methods

2.1. Raw materials

Particles of household waste of mate-tea were collected in Paraná State, Southern Brazil, and wood chips from a fast-growing eucalypt species (*E. benthamii*) were collected from waste of a sawing process at the *Embrapa Forestry* sawmill (Fig. 1). This material showed moisture content ~70% on wet weight basis and was dried in an oven at 65 ± 2 °C until constant weight was reached. Subsequently, they were cut into small particles using a Wiley knife mill (40–60 mesh).

A matrix of polypropylene – PP H103 supplied by Braskem (Brazil) with density of 0.905 g cm^{-3} and melt flow index of 40 g min^{-1} was used. The number-average molecular weight (M_n) of the PP is 49.44 and mean weight (M_w) is 235.597.

2.2. Preparation of composites

The particles of household waste of mate-tea and eucalypt wood measuring 40–60 mesh were manually mixed in distinct concentrations of particles:matrix (total weight of 80 g) according Table 1.

The composites were moulded using a Marconi MA098/A electrically heated hydraulic press with homogenous distribution of pressure. The moulding process was carried out at 175 ± 2 °C for 20 min with a pressure of 4 MPa. Direct contact between the polypropylene powder and metal platens during the heating and pressing process occurred using wax paper.

The nominal size of the composites was 140 mm × 140 mm × 4 mm after the cooling process. Moulding of the composites for the mechanical tests were performed according to ISO and ASTM standards. Additives and coupling agents were not used in the moulding process of the composites.

2.3. Infrared spectroscopy (ATR-IR)

ATR-IR spectra of composites A, F and L were measured in a Nicolet Nexus 570 equipment by direct transmittance at a resolution of 4 cm^{-1} for 32 scans in the range of 800–4000 cm^{-1}. The alignment of the light equipment and the background spectra were collected before all the tests.

2.4. Thermogravimetric analysis (TGA)

Tests were done in a Shimadzu DTG-60H and composites A, F and L (4 mg per sample) were exposed to a temperature range between 20 and 600 °C at a constant heating rate of 10 °C min^{-1} under a constant inert atmosphere of N_2. The weight gain (TG) and the mass loss rate (DTG) were determined.

2.5. Differential scanning calorimetry (DSC)

Differential scanning calorimetry (DSC) was performed in order to analyse the temperature of crystallisation (T_c) and the melting temperature (T_m) of composites A, F and L. The temperature of crystallisation was determined by heating the samples to 180 °C and maintaining this temperature for 5 min. The samples were then cooled to 28 °C at a rate of 5 °C min^{-1}. The melting temperature was determined by heating the samples to 600 °C in order to degrade them.

2.6. Physical properties

The density of the composites was obtained according to ASTM D792-08 and the equilibrium moisture content was calculated after stabilisation of the samples' weight under controlled environment conditions (20 °C and relative humidity of 65%). Water absorption and swelling thickness tests were carried out according to ASTM D 570-98 for 2 and 24 h immersions in distilled water at room temperature. Six samples of the composites were dried in an oven (103 ± 2 °C) at constant weight. The samples were then immersed in distilled water. At the end of the immersion period, the samples were removed from the water, the wet surface was wiped off using blotting paper, and the wet weight values were determined. Water absorption (WA%) and swelling thickness (TS%) were calculated using the following Eqs. (1) and (2):

$$WA(\%) = [(WA_t - WA_0)/WA_0] * 100 \quad (1)$$

$$TS(\%) = [(TS_t - TS_0)/TS_0] * 100 \quad (2)$$

where WA_0, TS_0, WA_t, and TS_t denote the oven-dry weight and thickness, and weight and thickness after t time, respectively.

2.7. Wettability kinetics

The wettability was evaluated on four samples (stabilised at equilibrium moisture content) measuring 2 × 2 cm that were cut from each composite. Measurements of wettability were performed using a Dataphysics 0CA goniometer (sessile droplet method). The contact angle was determined by the placement of a deionized water droplet (5 μl) on three distinct points (randomly selected) of each sample (12 measurements) and in different times, after 5, 20, 35, 50 and 65 s.

2.8. Mechanical properties

The mechanical properties of the composites were evaluated through flexural strength (ISO 178), tensile strength at break (ISO 527), compressive strength (ISO 604) and adapted Janka hardness (ASTM D1037) tests for samples with small thickness. All the tests were performed in an EMIC DL30000 universal machine.