



# Structural, mechanical and thermal properties of bio-based hybrid composites from waste coir residues: Fibers and shell particles

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## ABSTRACT

Polypropylene hybrid composites were produced by using coir fibers and coir shell particles at a total weight content of 20%. The effect of hybridization and coupling agent on the morphological, structural, thermal and mechanical properties was investigated. The morphological and structural results show that the coupling agent increased filler/matrix interfacial adhesion. Thermogravimetric analysis indicates that thermal degradation of shell particles (285/465 °C) is lower than coir fibers (331/474 °C). The combination of coir fibers and particles has a positive effect on the Young's modulus with hybrid composites showing intermediate values between non-hybrid composites (improvement between 35 and 50%). The modeling of the mechanical behavior of the manufactured composites was done by physical models (Voigt, Reuss, and Tsai–Pagano models).

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## 1. Introduction

Processing of plastic composites using bio-filler as reinforcement has engineering and commercial interest to produce novel classes of composites (Kazemi Najafi, 2013; Thakur et al., 2014a; Awal et al., 2015; Boujmal et al., 2014). Developments in the production of natural polymers/composites is focused on mechanical improvement and reduction of structural defects to ensure durability, reliability, cost reduction and increased production rates (Abdellaoui et al., 2015; Thakur and Thakur, 2014b, 2014c). For polymer strengthening, natural fillers have been used because of their significant specific mechanical properties and advantages including low density, recyclability, low cost, high specific strength, good thermal properties, and reduced tool

wear (Thakur et al., 2014a; Essabir et al., 2015a). These bio-fillers have found increased applications due to their availability as renewable materials and ecological concern (Qaiss et al., 2014, 2015; Thakur et al., 2013a). Polymer composites based on natural fibers have also attractive advantages such as low cost, low density, good thermal and mechanical resistance (Qaiss et al., 2014, 2015; Ashori and Nourbakhsh, 2010; Akil et al., 2014), degradable characteristics and environmental friendliness (Essabir et al., 2013a, 2013b; Thakur et al., 2013b). Also, biocomposites have been shown to reduce costs and improved mechanical, thermal and rheological properties (Unterweger et al., 2014; Malha et al., 2013; Arrakhiz et al., 2013a; Essabir et al., 2013c).

To further improve on the properties of polymer composites, mixing in a single matrix particles and short fibers led to the development of hybrid composites (Arrakhiz et al., 2013b; Shao-Yun and Bernd, 1998). Particles and short fibers can be directly incorporated into thermoplastics by conventional methods such as extrusion compounding and injection molding (Arrakhiz et al., 2013b).

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A better understanding of the chemical composition and natural fillers surface is necessary to develop natural fillers-reinforced composites. Lignocellulosic fillers are mostly composed of cellulose (Arrakhiz et al., 2012a; El Mechtali et al., 2015) which is a semi-crystalline polysaccharide. But the large amount of hydroxyl groups in cellulose gives natural fillers hydrophilic properties which, when used to reinforce hydrophobic matrices, produces interfacial incompatibility between the fillers and the matrix leading to poor moisture absorption resistance (Xue et al., 2007) and low mechanical properties (Belgacem et al., 1995). However, improving compatibility can be achieved by physical (Belgacem et al., 1995, 1996) or chemical modification (Arrakhiz et al., 2012a, El Mechtali et al., 2015) of the bio-fillers. Nevertheless, addition of a coupling agent like grafted copolymers is mostly done (Essabir et al., 2013a). Maleic anhydride-grafted poly(styrene-ethylene/butyl-diene styrene) (SEBS-g-MA) can be used as a coupling agent as well as an impact modifier. The chemical groups of maleic anhydride are able to react with the hydroxyls of the bio-fillers, while the SEBS copolymer is compatible with most polyolefin matrices. These reactions can give rise to strong links between the fillers and the matrix, making the composite mechanically viable. The addition of coupling agent has a direct effect on all the physical and thermal properties of the resulting composites (Arrakhiz et al., 2012a, 2012b).

Research is moving forward in order to find ways to use bio-fillers in place of conventional fillers as reinforcement in thermoplastic polymers. Arrakhiz et al. (2012b) manufactured chemically treated Alfa fibers reinforced PP composites using a heated two-roll mill. El Khaoulani et al. (2013) showed the potential of hemp fibers in reinforcing thermoplastic polymers, while Malha et al. (2013) prepared stratified doum fibers filled PP using compression molding.

From the literature, coir fibers are one of the most used natural fibers as reinforcement in polymer matrices due to their availability combined with high structural and mechanical properties (Arrakhiz et al., 2012a). Mir et al. (2013) reported that coir fibers have high failure strain, providing better strain compatibility between the fiber and the matrix in short fiber reinforced composites. In addition, coir fibers can be stretched beyond their elastic limit without rupture due to some helical arrangement ( $45^\circ$ ) of the micro-fibrils (Mir et al., 2013). Generally, when producing the coir fruits, tons of coir residues are produced, especially coir shells. Some research reported that the fruit shells can be used as a potential alternative fuel for diesel engine, or an energy source in an open core gasifier (Wever et al., 2012; Tiryaki et al., 2014). But in general these residues are used for heating. Few researches investigated the use of coir shell particles as reinforcement/filler in polymers, but this material can give an added-value to the polymer. So, the fruit shell particles can be used as an alternative to mineral fillers such as calcium carbonate and talc in the plastics industry (Essabir et al., 2014).

Hybridization of two kinds of fillers having different morphological, structural, chemical and physical properties offers some advantages over the use of either filler used alone in a polymer matrix (John and Venkata Naidu, 2004; Jawaid et al., 2014). Hybrid composites received attention from several researchers as a way to enhance the mechanical prop-

**Table 1**  
Coir fibers and shells particle size distribution.

Coir	Diameter ( $\mu\text{m}$ )	Length ( $\mu\text{m}$ )
Fibers	138	718.8
Shells particles	17.7	–

erties of composites (John and Venkata Naidu, 2004; Jawaid et al., 2014). In this case, apart from the conventional uses of coir residues (coir fibers) as mentioned above, our research investigates ways to use fruit shell residues, in particular both coir fibers and coir nut shell particles as reinforcement in hybrid polymer composites. This work provides an overview of a method to develop polypropylene (PP) hybrid composites based on these residues. The coir was obtained from coconut trees found in tropical countries which fruits (fibers and shell particles) can be used since they are abundant in nature and have a negligible effect on the environment due to their biodegradable properties (Arrakhiz et al., 2012a). Adhesion between the fiber and the polymer is one of the factors affecting the manufactured composites. In order to increase adhesion, it is proposed to first chemically treat with NaOH and then compatibilize by the addition of a coupling agent (SEBS-g-MA). The effects of coupling agent and filler ratio (fibers and particles) on the thermal and mechanical properties of the hybrid composites are reported. From the results obtained, the Tsai-Pagano model is used to determine the mechanical properties of the composites.

## 2. Materials and methods

### 2.1. Materials

All reagents used in this work were of analytical/reagent grade. The matrix used was polypropylene (PP) with a density of  $0.9 \text{ g/cm}^3$  and a melting temperature of  $165^\circ\text{C}$ . This polymer was supplied by ExxonMobil Chemicals. Sodium hydroxide (NaOH, 98%) was obtained from Sigma-Aldrich and acetic acid ( $\text{CH}_3\text{COOH}$ , 99–100%) was supplied by Riedel-Haën. The coupling agent is styrene-(ethylene-butene)-styrene triblock copolymer grafted with 1.4 to 2 wt. % maleic anhydride (SEBS-g-MA), supplied by Shell (Kraton FG-1901X). The coconut fibers and shells were supplied by Fibrivoire (Ivory Coast). Before use, the fibers and shells were grinded in a precision grinder (FRITSCH Pulverisette 19) equipped with a 1 mm sieve opening. Table 1 presents the particle size distribution of the grinded fibers and shell particles.

### 2.2. Treatment of coir fibers

The crushed coir fibers were first washed with water and then kept for 48 h in a 1.6 mol/L sodium hydroxide solution. The obtained fibers were then removed from the caustic solution and treated with acetic acid (100 mL) to neutralize the remaining caustic (Arrakhiz et al., 2012b). The materials were finally air dried for 24 h. This treatment was applied to remove lignins, waxes and resins (oils) covering the external surface of the fiber walls and to expose (better access) the hydroxyl groups.

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