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Mechanical and thermal properties of bio-composites based on polypropylene reinforced with Nut-shells of Argan particles

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ABSTRACT

This study treats the combined effects of both particle sizes and particle loading on the mechanical and thermal properties of polypropylene (PP) composites reinforced with Nut-shells of Argan (NA) particles. Three range sizes of particles were used in the presence of a polypropylene matrix grafted with 8 wt.% of a linear block copolymer based on styrene and butadiene coupling agent, to improve adhesion between the particles and the matrix. The composites were prepared through melt-blending using an internal mixer and the tensile specimens were prepared using a hot press molding machine. Scanning Electron Microscopy (SEM), Fourier Transform Infrared Spectroscopy (FT-IR), Thermo Gravimetric Analysis (TGA), Differential Thermal Analysis (DTA) and tensile tests were employed to characterize the composites at 10, 15, 20 and 25 wt.% particle contents. Results show a clear improvement in Young's modulus from the use of particles when compared to the neat PP, a gain of 42.65%, 26.7% and 2.9% at 20 wt.% particle loading, for particle range 1, 2 and 3, respectively. In addition a notable increase in the Young's modulus was observed when decrease the particle size. The thermal stability of composites exhibits a slight decrease (256–230 °C) with particles loading from 10 to 25 wt.%, against neat PP (258 °C).

1. Introduction

Manufacturing of high performance engineering materials from renewable resources is one ambitious goal currently being pursued by researchers across the world [1–5]. The environmental constraints and the new regulations on the recycling of composite materials have pushed manufacturers to develop new materials from renewable resources. The introduction of natural fillers in polymer matrix can provide significant advantages compared to traditional fillers used in composites such as glass and carbon fibers [1]. The polymer based natural filler composites have increased worldwide due to their low cost, low density, renewable and biodegradable character, good mechanical properties [2] and to their environmental friendliness [3,4]. Lot of researches [5–10]; have been done to take advantages of the wide range of natural fillers properties such as mechanical and thermal properties [11–13]. These studies are mainly focused on varying filler content [14], and their chemical treatment and the choice of the polymer matrix [15]. The mechanical properties of composites reinforced by particles depend strongly on the dispersion /distribution state, interfacial adhesion, morphology and particle loading [16]. Particle size has an obvious effect on these mechanical properties [17]. For example, smaller calcium carbonate particles provide higher strength of filled polypropylene composites at a given particle loading [9]. However, the hydrophilic character of natural fillers leads to low compatibility with hydrophobic polymer matrix and also to a poor dimensional stability as water uptake swells fillers [18]. Therefore, a lack of compatibility between natural fillers and polymeric matrix such as polypropylene (PP) leads to poor adhesion between natural particles and matrix. In addition, the thermal stability of natural fillers limits the use of its composites in the processing at a high temperature [19], especially in the extrusion and hot compression processes [8]. Polymer modification improves the interfacial adhesion when one end of the molecule is tethered to the reinforced surface, while the function of the other end reacts with the polymer phase [20]. In order to provide good adhesion and increase the shock resistance and ductility of the composite, the polymer modification and chemical treatments of fillers are the most used [20].

Various natural fillers such as alfa [6], coir [7], banana [8], calcium carbonate [9], clay [10], graphene [21], have been tested as new reinforcement agents in polymer-based composite materials. Among these natural fillers, the Argan Nut-shells could have a promising future as new reinforcement in polymer composites. The forest of Argan occupies about 830,000 ha in south-west of Morocco. It is one of the most common trees in the Moroccan area

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[22]. All parts of the Argan tree are used by local people: wood and woody shell of the fruit for heating, the almond fruit for production of Argan oil. While, fruit pulp and seed cake residue from the production of Argan oil is used for cattle [22]. The importance of the Argan tree in the rural economy of the semi-arid region is therefore considerable. The majority of the Argan forest land is state-owned, with a right to use a very important for local populations: collecting dead wood, harvesting fruit, and career development culture between Argan. The Nut-shells of Argan (NA) have an oval cross through a slot lengthwise contains small fibers. The NA was characterized by large specific mechanical properties such as stiffness and strength allowing high value to this material by finding new applications.

The aim of this study is to characterize the mechanical and thermal properties of Nut-shells of Argan (NA) particles used as reinforcement in polypropylene matrix. The polypropylene matrix was grafted with 8 wt.% of a linear block copolymer based on styrene and butadiene coupling agent. The composites were prepared using an internal mixer and the tensile specimens pressed by a hot press molding machine. The Nut-shells of Argan and their composites were characterized by SEM microscopy, FT-IR, TGA/TDA analysis and mechanical tests.

2. Material and experimental details

2.1. Material

Polypropylene (PP) was used to prepare a reinforced composite (ExxonMobil chemical, a density of 0.9 g/cm^3, and melting temperature of 165 °C). Nut-shells of Argan (NA) were collected from rural areas of south-west of Morocco (Souss Massa). The compatibilizing agent is a linear block copolymer based on styrene and butadiene, with 29.5 wt.% of styrene, supplied by shell (Kraton D1152 E polymer).

2.2. Experimental procedure

2.2.1. Preparation of the granulometry of particles

Particle size analysis allowed us to separate the NA particles in three ranges size depending on the diameter (D) of the particles. Table 1 illustrates the size distribution of particles. Three series of samples (Polypropylene (PP) reinforced by NA particles) were prepared:

- First series with 10, 15, 20 and 25 wt.% NA particles belonging to a particles size range 1.
- Second series with 10, 15, 20 and 25 wt.% NA particles belonging to a particles size range 2.
- Third series with 10, 15, 20 and 25 wt.% NA particles belonging to a particles size range 3.

2.2.2. Matrix preparation

The polypropylene was modified by adding a compatibilizer, 8 wt.% of a linear block copolymer based on styrene and butadiene. The resulting composites are denoted PP-D1152.

The compatibilized PP was extruded in a twin screw extruder (LEISTRITZ EXTRUSIONS TECHNIK GMBH, Germany), with the main screws rotating at 125 rpm while the side-feeding screw used for compatibilizer was set at 40 rpm. The extruder barrel was heated with the following profile of 200, 200, 200, 200, 180, 180, 180, and 180 °C [23] from hopper to the die, respectively.

2.2.3. Compounding and casting of test specimen

The compatibilized polypropylene matrix was compounded with NA particles at various fibers content, using a two roll mill (Thermo Haak Rheomix, Germany), the PP matrix and the NA particles build a batch of 30 g. The mixing conditions in terms of temperature, cross speed rate and mixing time were set at 220 °C, 60 rpm and 2 min 30 s, respectively. The mixing time was optimized to homogenize the dispersion and distribution of particles in the PP matrix, at that time the torque measured was constant. The rolls are then stopped and the composite removed from the heated rolls before being cut into small pieces for hot press.

Hot press molding was performed using an automatic press (CARVER INC, USA) with two heated platens; samples were pressed at 200 °C for 2 min and cooled to room temperature. The used mold was shaped according to ISO 527-1 (2012) [24] for mechanical testing.

2.3. Characterization

2.3.1. Scanning electron microscopy

Scanning electron microscopy (SEM) was recorded using EDAX Quanta 200, manufactured by FEI Company. SEM was used to analyze the morphology of the NA particles and dispersion of particles into the polypropylene matrix. The samples were cryofractured.

2.3.2. Fourier transforms infrared (FTIR) spectroscopy analysis

Fourier transform infrared spectra was recorded using a VERTEX 70 FTIR spectrophotometer. FTIR spectral analysis was performed within the wave number range of 500–4000 cm^{-1} and a resolution of 4 cm^{-1}.

2.3.3. Thermal analysis

The thermo gravimetric analysis (TGA) and differential thermal analysis (DTA) was carried out using a DTG-60 analyzer supplied by Shimadzu Company (Japan). TGA and DTA thermograms of NA particles and composites containing particles were obtained from ambient temperature to 600 °C at a heating rate of 10 °C min^{-1}.

2.3.4. Mechanical testing

The mechanical properties of the composites, namely their tensile properties were characterized to evaluate the improvement achieved by the various (AN) particles content in the polymer. Tensile tests were in accordance with ISO 527-01 (2012) [24], at a room temperature, using an Instron 8821S Universal Testing Machine. The cross-head speed was fixed at 3 mm/min, and the load was applied until the specimen was failed. The resulting load was measured using a 5 KN load cell. The mechanical Analyses were repeated five times per material, and the averages were used as representative values.

Table 1
NA particles distribution.

Range 1		Range 2		Range 3	
Sizes (μm)	Loading (wt.%)	Sizes (μm)	Loading (wt.%)	Sizes (μm)	Loading (wt.%)
D<125	20	125 < D < 160	20	160 < D < 250	20
125 < D < 160	60	160 < D < 250	60	250 < D < 315	60
160 < D < 250	20	250 < D < 315	20	315 < D < 360	20