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Biocomposites based on Argan nut shell and a polymer matrix: Effect of filler content and coupling agent

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ABSTRACT

This study aimed at developing a biocomposite using polypropylene (PP) as the matrix and Argan nut shell (ANS) as reinforcement. Also, styrene–(ethylene–butene)–styrene triblock copolymer grafted with maleic anhydride (SEBS-g-MA) was used as a coupling agent. The samples were prepared by using extrusion compounding followed by injection molding to determine the effect of filler and coupling agent content on the morphological, thermal, mechanical, and hygroscopic properties of the biocomposites. SEM micrographs revealed that good ANS dispersion/distribution into PP was achieved with an important reduction of fiber pull-out, micro-spaces, and voids with coupling agent addition. This led to substantial improvement intension, torsion, and water absorption reduction due to improved interfacial adhesion. Although ANS particles did not significantly modify the thermal stability of PP, the use of a coupling agent increased it. The experimental data were compared with several theoretical models such Voigt, Reuss, Hirsch, and Tsai–Pagano to characterize the interfacial adhesion quality and to determine the elastic modulus of a single ANS particle. Finally, all the results show that Argan waste to produce PP biocomposites is an interesting avenue to effectively deal with agricultural wastes and develop valuable industrial and practical applications.

1. Introduction

Over the last decade, concepts such as sustainable development, industrial ecology, and green chemistry led to the development of new generations of composite materials (Awal, Rana, & Sain, 2015; Bai, Sun, Xu, Dong, & Liu, 2015; Beckermann & Pickering, 2008; Kazemi Najafi, 2013). These composites are now constantly evolving toward more efficient and less expensive products in response to environmental constraints and regulatory requirements such as recycling (Adhikary, Pang, & Staiger, 2008; Karsli, Yesil, & Aytac, 2013; Khanam & AlMaadeed, 2014). The most commonly used inorganic fine powders for several industrial products such as paper, paints, rubbers, and polymers applications is calcium carbonate (Jia, Li, Ma, Sun, & Zhu, 2012) or talc (Castillo et al., 2013; Wang et al., 2013). They are not only added to reduce costs (fillers), but also to improve thermal, mechanical, and dimensional stability (Ishak et al., 2013; Thakur & Manju, 2014; Wang et al., 2013). However, another important property for the plastics industry is low density, which is not the case for $CaCO_3$ or talc (El Mechtali et al., 2015; Essabir, Bensalah, Bouhfid, & Qaiss, 2014; Nekhlaoui et al., 2015). This is why other reinforcements having lower density are investigated to reduce weight while keeping rigidity. This led to the use of different plants or their residues as biofillers in synthetic polymer matrices to develop new fillers/plastics innovative composites with good overall properties (Fiore, Scalici, & Valenza, 2014; Malha et al., 2013; Qaiss, Bouhfid, & Essabir, 2014; Zhang, 2014). In this context, almond shells, apricot, coco, olive, pistachio, and date palm earned a great deal of interest for applied research and industrial applications due to their high specific properties and availability (Tiryaki, Yagmur, Banford, & Aktas, 2014). Recently, Naghmouchi, Espinach, Mutjé, and Boufi (2014) incorporated solid waste from the olive oil industry in polypropylene to produce high performance composites with improved properties. Similarly, AlMaadeed, Nógellová, Mičušík, Novák, and Krupa

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(2014) developed composites based on low density polyethylene filled with date palm powder. From their results, the mechanical properties of the composites were substantially improved for biofiller addition up to a maximum content of 70 wt%. Thus, transforming polymers into materials with high structural and mechanical properties that can be competitive with fossil derivatives led to investigation of different biocomposites (Abdellaoui, Bensalah, Echaabi, Bouhfid, & Qaiss, 2015; Boujmal et al., 2014; Elkhaoulani, Arrakhiz, Benmoussa, Bouhfid, & Qaiss, 2013). Moreover, biofillers offer many advantages due to their unique features such as low cost, low density, ease of implementation, high specific modulus, low energy consumption, zero CO_2 emissions in production, low abrasive properties, biodegradability, non-toxicity and continuous availability (Arrakhiz et al., 2012a; Qaiss et al., 2014; Qaiss, Bouhfid, & Essabir, 2015a; Qaiss, Bouhfid, & Essabir, 2015b).

Biofillers are hydrophilic since they are composed of cellulose, lignin, pectin, and hemicelluloses, which contain hydroxyl groups with high reactive functionality (Arrakhiz et al., 2012b, 2012c; Müller, Renner, Móczó, Fekete, & Pukánszky, 2014). These biofillers are therefore incompatible with hydrophobic polyolefin matrices and also have low moisture resistance. The lack of compatibility between hydrophobic matrices and hydrophilic fillers leads to poor interfacial adhesion and the formation of aggregates during processing, all leading to low mechanical properties (Arrakhiz et al., 2012b, 2012c). It is then necessary to improve this interfacial adhesion as to improve water absorption and mechanical properties of the resulting composites (Huu Nam, Ogihara, Huy Tung, & Kobayashi, 2011; Mir, Nafsin, Hasan, Hasan, & Hassan, 2013). Arrakhiz et al. (2012b) and Arrakhiz, Benmoussa, Bouhfid, and Qaiss (2013) reported that an appropriate treatment applied to the biofiller or polymer matrix may overcome the main drawbacks of using biofillers, such as poor interfacial bonding with the polymer matrix, bad filler distribution/dispersion, and low thermal stability compared with inorganic fillers.

So the aim of this work is to develop a new biocomposite based on polypropylene reinforced with Argan nut shells (ANS). The idea of using ANS in a thermoplastic matrix is not only to minimize the environmental impact of ANS as residues, but also to maximize performance, functionality, sustainability, and profitability of these natural resources. So this work consists of ANS valorization since it is an abundant waste resulting from Argan oil extraction. It is proposed to use these residues as reinforcement in a polymer matrix to improve the biocomposite properties (Essabir et al., 2013b; Essabir, Achaby, Hilali, Bouhfid, & Qaiss, 2015; Essabir, Hilali, et al., 2015). Biofillers have a promising future as reinforcement in polymer matrices because they produce high specific mechanical properties (Essabir et al., 2013b, 2015a, 2015b).

2. Materials and experimental detail

2.1. Materials

The matrix used was polypropylene PPH03BPM (TASNEE, Saudi Arabia) which is an extrusion/co-extrusion grade with a melt flow index of 3.0 g/10 min (ASTM 1238), a density of 0.9 g/cm^3 (ISO 1183), and a Vicat softening temperature of 156 °C (ISO 306). Styrene–(ethylene–butene)–styrene triblock copolymer (with a polystyrene content of 30%) grafted with 1.4–2 wt.% of maleic anhydride (SEBS-*g*-MA) was used as the coupling agent: Kraton FG-1901X supplied by ExxonMobil Chemical. Argan nut shell (ANS) was selected as the biofiller (ρ = 1.29 g/cm^3, and average diameter of 50 μm) and provided from rural areas of south-western Morocco (Souss Massa, Agadir).

2.2. Bio-composite manufacturing

2.2.1. Argan nuts shell preparation

The Argan nut shells were collected as residues after extraction of the almond (fruit core) by a traditional hand-press method (Essabir et al., 2013b, 2015a). The stone, called Argan nut shell, represented about 86% of the Argan weight and is mainly composed of cellulose, hemicelluloses and lignin. The biofiller used in this study consisted of Argan nut shell (stones). Before further uses the Argan nut shells were ground in a precision grinder (FRITSCH Pulverisette 19) equipped with a 250 μm sieve size. This fraction was then sifted on a 100 μm sieve size to remove the presence of any waxes and to obtain finer particles (between 10 and 100 μm).

2.2.2. Biocomposite preparation

The neat polypropylene and the modified polypropylene were used as polymer matrix; the polypropylene was modified by adding a coupling agent (SEBS-*g*-MA) at 8 wt.% (Fig. 1). Before use, ANS particles were dried in an oven under vacuum at 80 °C to reduce their humidity content. Then, compounding was performed in a twin-screw extruder. First, PP and SEBS-*g*-MA were introduced together in the main feed hopper, then ANS was added (0, 5, 10, 15, 20, 25, and 30 wt.%) via a side stuffer in a Leistritz ZSE-18 (D = 18 mm, L/D = 40) twin-screw extruder (Leistritz Extrusions Technik GmbH, Germany) (Fig. 2). Mixing was performed at 125 rpm for the main screws and a 40 rpm side-feeder screw speed for particle feeding. The temperature profile (7 zones) was fixed at 170, 170, 175, 180,

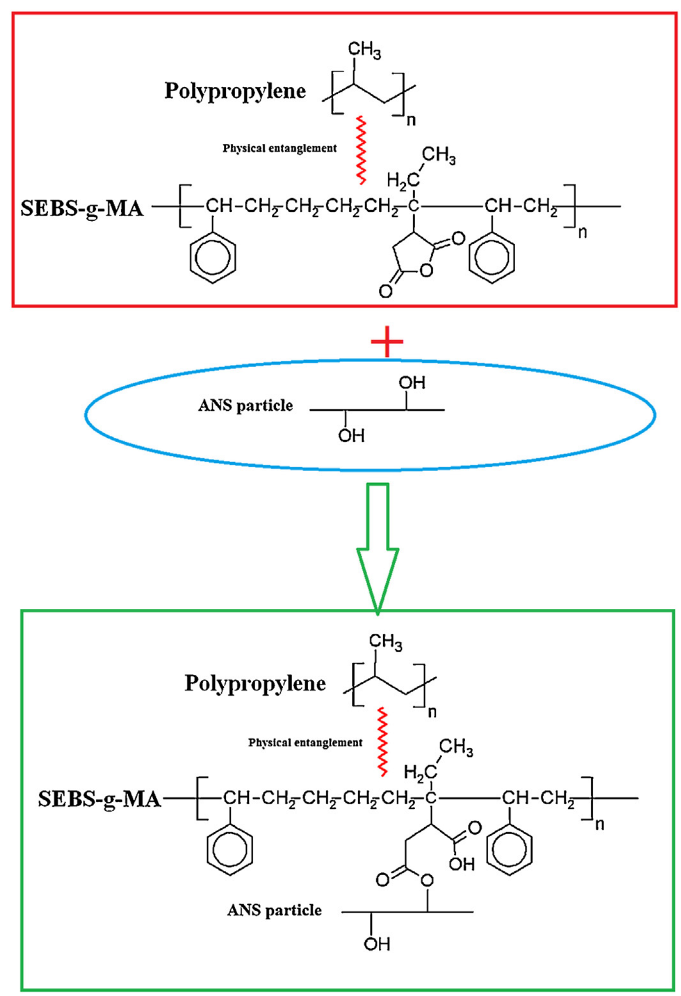

Fig. 1. Mechanism synthesis scheme of biocomposite material.