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Use of residual agricultural plastics and cellulose fibers for obtaining sustainable eco-composites prevents waste generation

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ABSTRACT

Crop protection residual plastic films are a growing environmental problem which requires efficient solutions. Their suitability as matrices for obtaining sustainable eco-composites reinforced with industrially-sourced residual natural fibers was investigated in order to boost their recovery and prevent waste generation. The analysis of the studied residual agricultural plastics revealed that they are low density polyethylene still containing significant amounts of ethylene-vinyl acetate (2.5–4.5 wt%). A pilot–plant extrusion-compounding technology was applied to a selected recycled plastic from residual agricultural films and the residual cellulose fibers for obtaining the eco-composites. The effects of cellulose-fiber content and a selected maleic anhydride-modified polyethylene coupling agent on the properties and interfacial adhesion of the eco-composites were investigated. By using micromechanical models, scientific data of the intrinsic modulus and strength of the *Eucalyptus Globulus* residual fibers, hitherto scarcely available in literature, were found to be 16.4 GPa and 180 MPa, respectively, thus revealing their suitability as cost-effective reinforcement. Tensile modulus and strength of the eco-composites were up to 667% and 70% greater than those of the neat agricultural recycled plastic, the latter due to the enhanced compatibility provided by the ethylene-vinyl acetate found. When the coupling agent was added, tensile and flexural strengths increased up to a maximum of 20.26 MPa and 23.96 MPa, respectively. Property variations were found to be due to the fiber length reduction and the interfacial adhesion improvement caused by the coupling agent as well as to its plasticizing effect. The properties achieved revealed the suitability of the eco-composites for their immediate application in the production of numerous environmentally sustainable and cost-effective end-products from the aforementioned wastes.

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1. Introduction

Plastic films used in modern intensive agriculture are turning into a growing environmental issue at the end of their useful life. It would then be very interesting to develop efficient solutions that pay off for the costs associated to their recovery, thus preventing waste generation. Due to the important role of plastic films in the production of a host of vegetables and fruits, the area covered with greenhouse, tunnel and mulch plastic films has been increasing since they were first used (by the late 50's) (Dept. of Agriculture, Food and Environment of Spain, 2012). Thus, by the year 2009, only in China, the country with the greatest greenhouse area in the world, 1,000,000 ha were covered with greenhouse and tunnel plastic films (Cajaraville et al., 2010). In Europe, the greatest greenhouse area is located in Spain, with more than 60,800 ha of greenhouses by 2012 (Dept. of Agriculture, Food and Environment of Spain, 2012).

At the end of the season or after a few seasons those plastic films must be replaced by new ones. According to the most reliable data available, the European production of virgin agricultural plastic films in 2011 was more than 1.7 Mt, most of them being low density

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List of acronyms: AF200, ALFATEN 200; DSC, Differential Scanning Calorimetry; EVA, Ethylene-Vinyl-Acetate Copolymer; FTIR, Fourier Transform Infrared; LDPE, Low-Density Polyethylene; MAPE, Maleic Anhydride-Modified Polyethylene Copolymer; MFI, Melt Flow Index; OIT, Oxidation Induction Time; RAPF, Residual Agricultural Plastic Films; RCF, Residual Cellulose Fibers; SEM, Scanning Electron Microscopy; UV, Ultraviolet; VA, Vinyl Acetate.

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polyethylene (LDPE). However, the recovery rate of agricultural plastics was only about 50% and the mechanical recycling rate was just around 23% (Plastics Europe, 2011, 2012).

The high volume of residual agricultural plastic films (RAPF) generated, along with their disposal by the farmers through on-site land filling and subsequent burning, turn them into a challenging environmental issue. Moreover, it must be taken into account that many of these films are heavily contaminated with soil, fertilizers and pesticides (Hussain and Hamid, 2003).

One of the ways tried for minimizing the environmental problem associated to the RAPF has been their incineration under controlled conditions. However, from an environmental and sustainability point of view, the mechanical recycling of agricultural plastic film wastes would be a superior solution to incineration.

Thus, it has been reported that it is possible to recycle waste greenhouse films for the same application, producing films with a layer of virgin polyethylene and another one consisting of a blend of recycled polyethylene modified with virgin thermoplastics and other stabilizers (Abdel-Bary et al., 1998). Also, it was shown that the use of additives enables the re-building of polyethylene chains, thus improving mechanical properties of residual plastic films (Scaffaro et al., 2006).

Agricultural plastic waste can be also used as matrix for composite materials, mainly reinforced with cellulose fibers. RAPF can show several advantages in this application. As they are designed to withstand intensive sun exposure during long periods of time, agricultural plastic films contain relevant amounts of several additives, such as antioxidants and Ultra-Violet (UV) stabilizers, which might still be present in the RAPF (Hussain and Hamid, 2003). On the other hand, they might also contain ethylene-vinyl-acetate copolymer (EVA) (Abdelmouleh et al., 2007). The presence of the polar vinyl acetate moiety in the waste plastic could be interesting, as it would enhance the compatibility between the non-polar polyethylene matrix and the polar cellulose, thus improving the polyethylene-cellulose interfacial adhesion and hence the composite properties. It would therefore be interesting to study if the RAPF still contain noticeable amounts of EVA, antioxidants and stabilizers, as they could give additional value to the recycled agricultural plastic pellets obtained thereof for being used as matrices in cellulose-reinforced eco-composites.

Different cellulosic materials (such as wood flour and cellulose fibers) have been used for enhancing the tensile, flexural and thermal properties of several polymeric matrices, thus giving rise to composite materials with important final applications (Abdelmouleh et al., 2007; Bledzki and Gassan, 1999; de la Orden et al., 2007; González-Sánchez et al., 2008). Among the works done, some recently published ones used softwood cellulose pulp fibers (Sdrobiş et al., 2012) and Doum fibers (Arrakhiz et al., 2013) for reinforcing virgin LDPE. Composites were prepared by batch melt mixing or twin-screw extrusion and the composites tensile modulus and strength achieved were about 0.25–0.48 GPa and 10–11 MPa, respectively.

In addition to the low cost, density and energy required for production, the main advantages of the use of cellulose fibers are the worldwide availability of renewable sources from which they can be easily obtained, their biodegradability, the lower abrasion of processing equipment, the lower risk for operators if inhaled and, finally, their CO_2 neutrality should they be incinerated (Bledzki and Gassan, 1999). Moreover, they may enhance the biodegradability of LDPE, thus making more environmentally sustainable composite materials (Sunilkumar et al., 2012).

There are large amounts of residual cellulose fibers (RCF) that can be used as reinforcement. For instance, in the world's main process used for the production of cellulose pulp (the Kraft process), a waste stream containing cellulose fibers is generated especially at the washing stage after the wood cooking. For a big pulp mill (*ca.* 500,000 t/year), around 12 t/day of residual cellulose fibers (with a moisture content of 50 wt%) may be generated, which are difficult to dispose off for this industry.

Then, the chance of using these residual cellulose fibers (RCF) as renewable reinforcement for polymeric-matrix eco-composites is of great interest from the environmental and economic points of view. Furthermore, the production of composites from RCF and RAPF may induce social benefits in terms of new jobs.

Therefore, this work aims to study the suitability of recycled plastic pellets obtained from residual agricultural plastic films and cellulose fibers as raw materials for composites. For this purpose, the residual materials and the recycled agricultural plastics were characterized. The chemical nature and the presence of additives and EVA copolymer in both RAPF and the recycled plastic pellets obtained thereof were studied by Ultra-Violet (UV) and Fourier Transform Infrared (FTIR) spectroscopic techniques. Melt Flow Indices (MFI), tensile properties, crystallinity degrees and Oxidation Induction Times (OIT), measured by Differential Scanning Calorimetry (DSC), were also determined.

An extrusion-compounding technology at pilot–plant scale was applied to the selected recycled agricultural plastic and the RCF in order to obtain cellulose-reinforced composites. The effects of cellulose content (25–35 wt% of fibers) on the tensile and flexural properties of composites were determined. 35 wt% was selected as the upper limit because in a previous study it was determined that cellulose contents higher than 35 wt% make very difficult the extrusion and injection molding processing of these composites. Also, the effect of the addition of two selected percentages (1.5 and 3 wt%) of a selected maleic anhydride-modified polyethylene (MAPE) coupling agent on the composite properties and the fiber-matrix interfacial adhesion was investigated. In order to achieve a further understanding of composite properties and possibilities of use of the RCF, the reinforcing capability of the residual fibers was estimated through the fitting of experimental results to micromechanical models.

2. Materials and methods

The materials and reagents, the characterization methods of the raw materials, the method used for preparing the composites and the testing methods used through the work are described in this section.

2.1. Materials and reagents

Three residual agricultural plastic films (RAPF) from greenhouses and other uses, two recycled agricultural plastic pellets obtained from different mixtures of RAPF and one commercial virgin plastic (commonly used in the manufacturing of greenhouse films) were characterized. The three RAPF, supplied in shredded films form by BEFESA Plásticos (Spain), were a whitish translucent 0.08 mm thick film (referred to as R1), a transparent 0.04 mm thick film (referred to as R2) and a yellowish translucent 0.19 mm thick film (referred to as R3). The two recycled agricultural plastic pellets were ALFATEN200$^{\mathbf{TM}}$ (referred to as AF200), supplied by BEFESA Plásticos (Spain), and PEBD-IB, supplied by IBACPLAST (Spain). The commercial virgin plastic, supplied in pellet form by REPSOL (Spain), was CA2131A, an LDPE with different additives. The main properties of the recycled plastic pellets, given by the suppliers, are shown in Table 1.

Residual cellulose fibers (RCF), a by-product obtained from the manufacturing of Kraft cellulose pulp, mainly consisting of unbleached *Eucaliptus Globulus* cellulose, were supplied by ENCE-Navia (Spain). They were provided as flakes suitable for their

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