COMPARATIVE STUDY OF MECHANICAL, RHEOLOGICAL AND MORPHOLOGICAL PROPERTIES OF DIFFERENT CELLULOSE-REINFORCED HIGH DENSITY POLYETHYLENE COMPOSITES UPON MECHANICAL RECYCLING

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Abstract

Injection molded testing specimens of original composites with different cellulose fibers composition were reprocessed up to five cycles under the same molding conditions. Original and five-cycle reprocessed specimens finally obtained were tested for determining their melt flow index, tensile properties and their morphology for comparison.
In this work quantitative effects of cellulosic fibers content, type of coupling agent and the number of reprocessing cycles onto the properties of those composites were determined.

Keywords: *Cellulose fibers, mechanical recycling, coupling agent.*

1. Introduction

Plastic-matrix composites reinforced with several kinds of cellulosic fibers are being object of a growing interest and intensive research for replacing other composite materials containing inorganic fillers and/or reinforcements. This is due to the inherent advantages of the former ones over the latter ones, such as lower density, lower abrasion of processing equipment, higher specific mechanical properties, biodegradability and lower cost, among others.

On the other hand, there is also a growing social concern regarding the environmental impact of polymeric materials and their residues, and the sustainability of the developed products. Therefore, the introduction of new raw materials for manufacturing end-products is subjected to specific analysis in order to analyze their viability to be recycled. On this regard, mechanical recycling of polymeric materials offers significant opportunities for their waste reduction while maintaining both materials and energy.

2. Materials and Methods

Cellulosic fibers used as reinforcement in composites were supplied by ENCE-Navia, a kraft pulp mill located in Navia, Asturias (Spain). They were a by-product obtained in the manufacture of Kraft cellulose pulp, consisting of unbleached cellulose with a cellulose content higher than 95%, and quantities of unreacted wood. The commercial high density polyethylene (HDPE) used as polymeric matrix for composites consisted of Eraclene MQ-74 homopolymer pellets (Polimeri Europa, Milano, Italy) with a density of 952 kg/m^3 (ASTM D1505) and a melt flow index of 11 g/10 min measured at 190 ºC and 2.16 kg, according to ASTM D1238.

Two commercially available maleic anhydride-modified polypropylene polymers (Epolene E-43 and G-3002) were used as coupling agents for the reference composite materials (Eastman Chemical Products, Kingsport, TN, USA). Epolene E-43 has an acid number of 45, a weight average molecular weight of 9100 g/mol, and a number average molecular weight of 3900 g/mol. Epolene G-3002 has an acid number of 60, a weight average molecular weight of 60000 g/mol, and a number average molecular weight of 20000 g/mol.

Melt flow indexes of original and five-cycle reprocessed composites were measured at 190 ºC and 2.16 kg, according to ASTM D1238 standard, using a DAVENPORT melt flow indexer, mod. 2233 (Lloyd Instruments, Farehand, UK).

Tensile modulus of elasticity of composites were determined according to ISO R527 standard at a nominal strain rate of 1 mm/min and the other tensile properties were determined at 50 mm/min using a Hounsfield Universal Testing Machine, mod. JJ T5501 (Salfords, England). A minimum of six specimens were measured for each trial.

The morphology of the composites was examined using an XL30 ESEM PHILIPS electron microscope at 25 kV. Samples of injection molded test specimens were cryogenically fractured in liquid nitrogen and sputter-coated with gold before viewing.

3. Results and Discussion

The variation of the melt flow indexes of cellulosic reinforced/HDPE composites mainly depends on the fiber content and the number of reprocessing cycles. The relative melt flow index increase of the five-cycle recycled composites over the original ones is higher as the cellulosic fiber content of the composites increases.

Upon reprocessing, the composites containing the E-43 maleated polypropylene showed a smoother decrease of the tensile modulus than that containing the G-3002 maleated polypropylene, with a bigger acidity and molecular weight than the E-43.

However, the choice of G-3002 as coupling agent derives in a superior increase of the elongation at yield and a lesser decrease in tensile strength at yield by reprocessing, in comparison with the E-43 coupling agent.

Moreover, the influence of recycling process on the elongation at tensile strength at break is more noticeable for composites containing the G-3002 coupling agent, showing a higher increase in comparison with the E-43 ones.

Composites without coupling agent and those containing E-43 coupling agent carried out a lesser degradation by reprocessing by injection than G-3002 coupling agent composites, so indicating a possible degradation of the later agent by mechanical recycling.

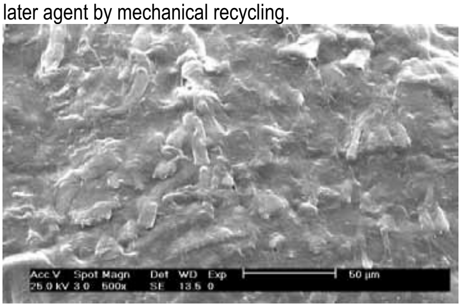

Fig. 1. Microphotography of 40 wt.% fibers and 1.5 wt% MAPP E-43 composite.

Figure 1 shows the microphotography of 40 wt% fibers and 1.5 wt.% MAPP E-43 composite. The fibers are observed homogeneously dispersed into the matrix. There is not showed practically holes between fibers and matrix, indicating that the fibers are embedded in the HDPE matrix due to an increase in the adhesion between fibers and matrix because of the effect of the coupling agent MAPP E-43, in comparison with the composites without coupling agent.

On the other hand, Figure 2 shows the 40 wt.% fibers and 1.5 wt% MAPP E-43 recycling counterpart composite. The difficult to observe fibers is a proof of their breakage. But some fibers and fiber residues can be observe embedded in the matrix, with good adhesion with the polymer in mostly of them, although there is also some holes, indicating fiber extraction and fibers coming out of the matrix by mechanical recycling effect.

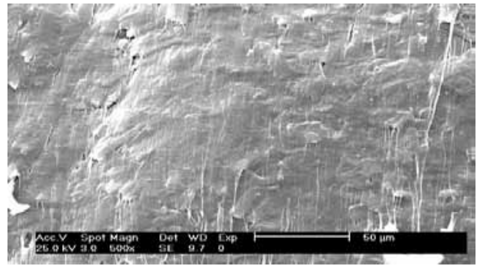

Fig. 2. Microphotography of 40 wt.% fibers and 1.5 wt% MAPP E-43 recycling composite

4. Conclusions

Although, there has been demonstrated the advantage of using coupling agent in order to improve the mechanical properties of the original composites, it is necessary, from a recyclability point of view, to be very careful in choosing the type of coupling agent. Otherwise, it would be much convenient not to use coupling agent in the formulations.

Despite it is interesting the use of high content of fibers from the economical point of view, considering recycling, a high proportion of fibers produces higher degradations by shear effect on the composite materials. As a whole, the formulation that carried out the highest degradation was the 48 wt.% cellulose content one.

The best results, considering good properties for the original composites and viability of recycling, were obtained for 40 wt.% cellulose content PE composites using the Epolene E-43 as coupling agent.

The stress-strain curves of the 40 wt.% original and recycled composites either with or without coupling agent, show the different behavior of these materials in tensile tests, from a hard material for the former ones to a deformable material for the last ones.

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Acknowledgements

The authors are grateful for the financial support of MICINN (project MAT2010-19883). J. Arranz-Andrés is grateful to the CSIC JAE-Doc Program for his financial support.