Macromolecular Symposia
www.ms-journal.de

Evaluation of the Effect of Recycling of Carbon Fibers for Polypropylene Reinforcement

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The recycling of carbon fibers (CFs) from composites is becoming a necessity, with a gain both from economic and environmental point of views because carbon fibers are a high value-added material and the energy requirement for their production is pretty high. The use of such recycled fibers needs to find reliable applications to fully close the circular economic loop and here it is demonstrated that their applications as short fiber reinforcement in thermoplastics are not only convenient but as reliable as prostin fibers. This work wants to evaluate the effect of the recycling and reprocessing recycled carbon fibers versus pristine fibers, when inserted in polyprolylene (PP), in the presence of different amounts of compatibilizer to promote a better fiber/matrix interaction.

1. Introduction

The future of carbon fiber-reinforced polymers (CFRPs) looks promising. Although the global market of composites has been subjected to a quite significant reduction caused by the pandemic issues in 2019, in the last years it started to increase again reaching, in 2021, a global market of $94.3 billion, thus overcoming the 2018 values. The huge boost in their production involves the crucial drawback of CFRPs recover and carbon fibers (CFs) recycling. Turning CFRPs waste into a valuable resource and closing the loop in their life cycle is vital for the continuous use of the material in several applications[1] with their recycling important in both economic and environmental terms. One of the most promising recycle technologies is the *pyrogasification* of composite material, to obtain high-quality and high value-added CFs.[2] Considering the still relatively high prices of virgin fibers both in economic (30–40€)[3] and energetic terms to produce it (183–286 MJ kg^{-1}),[4] *pyrogasification* appears economically and environmentally attractive. The first step of *pyrogasification* is *pyrolysis* of waste composites, which leads to a degradation of the matrix and recovery of the solid fraction composed of CFs„ covered by a thin layer of char. The second step is the *gasification* which, via a partial oxidation of the CFs surface, provides char removal. Industrial reports showed that the energy needed for carbon fibers recycle is only 5–10% of the energy required to produce virgin CFs.[5] Moreover, pyrogasification changes the surface chemistry of the recovered fibers thus impacting significantly on the reinforcement/matrix interface adhesion.[6] Carbon fibers/thermoplastic composites exhibit attractive behaviors: high specific strength, environmental resistance, damage tolerance, superior impact resistance, and especially the ability to be remelted and reprocessed.[7] One of the most recent applications in this field uses polypropylene (PP) as a matrix for CFRPs.[8] Since, as previously stated, *r*CFs from the *pyrogasification* process show more polar and reactive groups on their surface,[9] with respect to pristine CFs their interaction with an apolar matrix such as PP might not be highly effective, hence the use of a compatibilizer might be took into consideration. One of the most suitable compatibilizers to increase the interfacial adhesion of PP-based composites is maleic anhydride-grafted-polypropylene (MAPP), which is obtained by the grafting of PP chains with maleic anhydride which in turns provide hydrophilic and reactive functional groups. This compatibilizer is not expensive and is able to significantly increase compatibility increasing the composite proprieties as well.[10] The aim of this work is to prove that recycled carbon fibers can replace pristine one in short fibers CFRPs with PP as a matrix, to evaluate the effects of the manufacturing-mixing process on the fibers' dimensions and the effect of different amount of MAPP as compatibilizer. So, blends containing 10 wt% of *r*CFs, without any additional treatment after the recycling process and variable amount of MAPP, have been manufactured. Moreover, for the sake of comparison, analogous samples

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DOI: 10.1002/masy.202400013

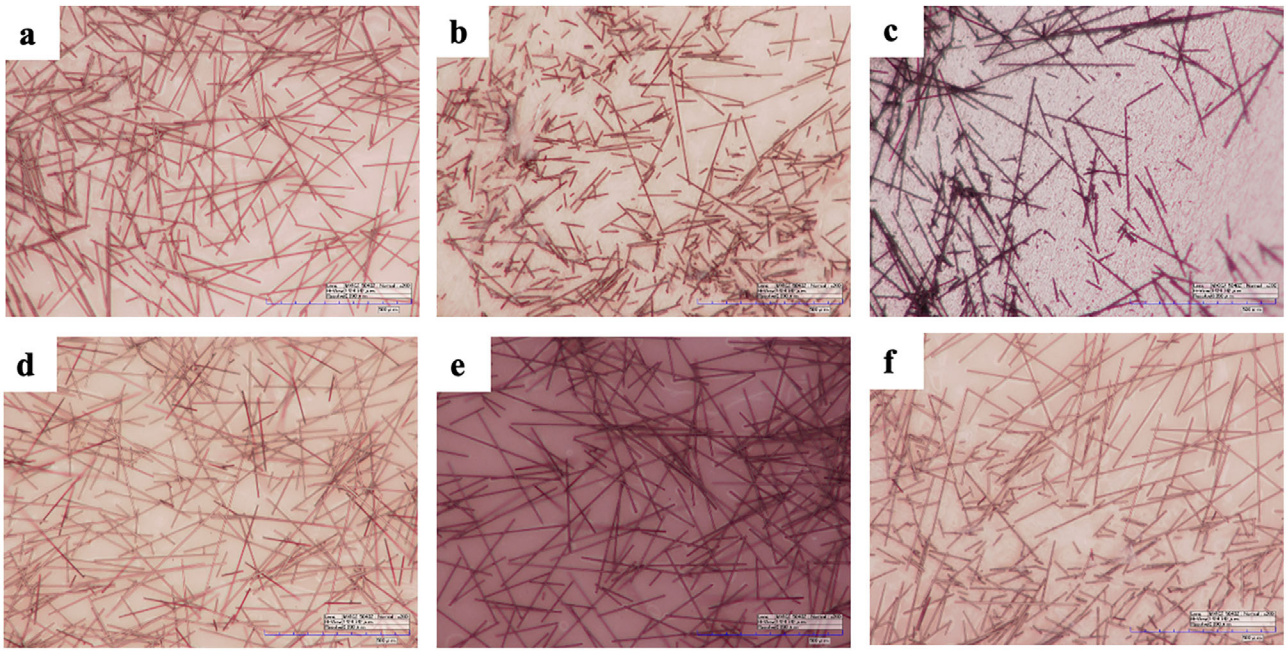

Figure 1. Optical microscope images of CFs deriving from thermal degradation of PP matrix PP*v*CF1a), PP*v*CF2 b), PP*v*CF3 c), PP*r*CF1 d), PP*r*CF2 e), PP*r*CF3 f). The scale bar indicates 500 µm.

Table 1. Composition of produced composites.

Composites	Compatibilizer [wt%]	Fibers [%wt%]	Type of fibers
PP*r*CF1	2.5	10	Recycled
PP*r*CF2	5.0	10	Recycled
PP*r*CF3	7.5	10	Recycled
PP*v*CF1	2.5	10	Virgin
PP*v*CF2	5.0	10	Virgin
PP*v*CF3	7.5	10	Virgin

Table 2. CFs content and average length of formulated composites.

Composite samples	W_r [a] [wt%]	CFs length [µm]
PP*v*CF1	9.47±0.06	233±142
PP*v*CF2	9.54±0.08	183±113
PP*v*CF3	10.08±0.42	179±112
PP*r*CF1	10.34±0.39	238±140
PP*r*CF2	10.04±0.02	251±122
PP*r*CF3	9.93±0.07	201±140

[a] CFs content measured by TGA.

with washed virgin fibers (*v*CFs) were obtained, in order to evaluate the effect of the partially oxidized surface of rCFs. All composites were characterized via thermogravimetric analysis (TGA) and optical microscopy for fiber/polymer composition and fiber length distribution evaluation.

2. Experimental Section

Recycled carbon fibers (*r*CFs) were supplied by curti SpA (Castelbolognese, Italy) and derive from pyrogasification of consolidated CFRP waste epoxy composites based on T700 carbon fibers in a semi-industrial pilot plant.[9] Virgin carbon fibers (*v*CFs), were unidirectional fabric UC 301 based on Toray T700S 12 K dry fabrics kindly provided by Bucci Composites (Faenza, Italy). All fibers were cut to 7 mm length, then *v*CFs were washed twice with acetone at reflux for 2 days, in order to remove the sizing agent. Acetone was purchased by Sigma Aldrich and used without further purification. Six composite materials were produced with different amount of compatibilizer and the same amount of carbon fiber. **Table 1** shows the composition of the six composites produced. All of the composites are prepared in the same condition: PP, compatibilizer, and CFs are mixed in a twin screw Brabender at 175 °C for 7 min with a screw speed of 75 rpm to guarantee the complete melting of the thermoplastic material. Thermal stability and CFs content of composites were investigated by TGA carried out on a TA Instrument SDT Q600 at 20 °C min^{-1} from room temperature to 500 °C under nitrogen flow (20 mL min^{-1}) and leaving them in isotherm for 20 min. TGA runs intended for CFs content determination were carried out in triplicate for each sample, on approximately 20 mg of sample. The evaluation of the CFs length was carried out on the residues from the thermogravimetric analysis using a digital microscope (HIROX), with a magnification of 200×.

3. Results and Discussion

During the compounding, thermal and mechanical stresses are applied on the composite, these allow to obtain the final material, but can damage the fibers. The evaluation of fiber dimension is showed in **Figure 1** and in **Table 2** and clearly demonstrates that

fibers, starting at 7 mm, are downsized during the process down to about 200 μm.

While the length decrease was expected because of the mechanical stresses due to the twin screw mixing operation, what was surprisingly found was that the length of *r*CFs seems to be generally higher than the one of *v*CFs. Indeed, *r*CFs are thought to be more fragile than prisitine fibers and were thus expected to undergo a significant fragmentation during the processing, to a higher, or at least comparable extent with respect to *v*CFs. These same results, however, well compared with previously obtained data[11] based on PLA reinforced with *r*CFs and *v*CFs, where *r*CFs are obtained with the same pyrogasification process though in different batches. This fact thus support the ability of the recycling method to provide fibers which are not significantly embrittled upon recycling, thus being at least comparable, if not even better, than pristine fibers when mixed with thermoplastic matrices.

4. Conclusion

This work aims to evaluate the effect of the recycle process and reprocessing on their cycled carbon fibers, by producing carbon fiber-reinforced thermoplastic composites. The study of the fiber length distribution demonstrated that recycled carbon fibers after compounding operation better preserve their dimension, showing a high potential for application in fields where the fiber dimension is not crucial, and thus the lack of control of fiber size that happens upon recycling is not detrimental.

Acknowledgements

The authors acknowledge European Union – NextGenerationEU for funding via National Sustainable Mobility Center CN00000023, Italian Ministry of University and Research Decree n. 1033–17/06/2022, Spoke 11–Innovative Materials & Lightweighting and via Project Ecosyster—Ecosystem for Sustainable Transition in Emilia-Romagna, project funded under the National Recovery and Resilience Plan (NRRP), Mission 04 Component 2 Investment 1.5—NextGenerationEU, Grant Number: J33C22001240001 and via Project Infrastructure for ENergy TRAnsition aNd Circular Economy @ EuroNanoLab″ IR0000027, iENTRANCE@ENL for the support.

Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

carbon fibers, CFRPs, polypropylene, recycling

Received: January 5, 2024

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