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Elastic and yield behaviors of recycled polypropylene-based composites: Experimental and modeling study

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ABSTRACT

In this work, stiffness and yield behaviors of recycled polypropylene (PP) based composites have been investigated by means of dynamic mechanical analysis (DMA) and split Hopkinson pressure bar (SHPB). It was found that the mechanical behaviors of non-recycled and recycled PP composites depend on temperature and frequency/strain-rate as well as on filler content and recycling cycle. For modeling the elastic modulus and yield stress, two new approaches were proposed. We extended a statistical stiffness model for neat polymers with temperature and frequency/strain-rate dependences by incorporating a Mori-Tanaka based approach and a two-population model to predict the elastic modulus of PP composites. By considering the initial modulus of neat PP and filler aspect ratio with reprocessing dependences, the predicted elastic modulus not only depended on the test temperature and frequency/strain-rate but also depended on the filler content and recycling number. To predict the yield behavior, we extended the modified cooperative model with temperature and strain rate dependences for neat polymers by incorporating a three-phase approach and a two-population model. In particular, the internal stress for neat PP and the interphase parameter B for PP-based composites were considered with reprocessing dependences. Predicted yield stresses by this new approach not only depended on the strain rate and temperature but also depended on the filler content and reprocessing number. A good agreement was found between experimental results and predictions.

1. Introduction

For automotive application, polypropylene is currently replacing other materials because of its low density, high resistance to aging and low price. In order to increase the toughness of neat PP, impact modifiers such as ethylene propylene rubber (EPR) and ethylene propylene diene monomer (EPDM) are frequently used [1,2]. Recently, ethylene octene copolymer (EOC) tends to be used to improve the toughness of PP due to its higher compatibility and processability than EPR and EPDM [3]. However, the strength and the stiffness of PP decrease with the addition of these soft rubber inclusions. To compensate the effect of these soft particles, the addition of mineral rigid fillers such as talc is recommended [4].

The increasing use of PP-based composite leads to huge plastic wastes from end-of-life vehicles and consequently, represents a threat to our environment. Therefore, it is of high importance to increase the re-use and recovery rate of these plastic wastes. Although different recycling options exist, the mechanical recycling by grinding and reprocessing is the easier and most ecological end-of-life scenario concerning the re-use of plastic wastes [5–7].

The composite reprocessing at high temperature, under important shearing stress, and with the possible presence of oxygen and impurities could lead to degradation and hence, to change of the properties compared to non-recycled composites [8]. In our previous work, we studied the impact of filler content and mechanical recycling on the properties of PP-based composites [9,10]. We found that recycling induced a reduction in PP molecular weight. This reduction was attributed to chain scission mechanism resulting from the reprocessing. The lower molecular weight of PP increased the polymeric chain mobility and hence, facilitated their rearrangement during the crystallization process. It was found that the decomposition temperature was quite constant with the number of recycling (reprocessing) cycles. Therefore, the recycled materials could be used under the same temperature range than

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the non-recycled ones. In addition, we found that the EOC inclusions stabilized the quasi-static tensile elongation-at-break up to 3 cycles due to a decrease of their size and a homogenization of their shape during the reprocessing. At the same time, the talc fillers increased the quasi-static tensile Young's modulus and the tensile yield stress of the composites with the recycling number due to a decrease of their size and an increase of their aspect ratio [9,10].

To the best of our knowledge, the modeling of mechanical responses for recycled PP-based composites was little investigated, especially in the case of dynamic behavior modeling. For automotive application, PP-based composites are generally used to manufacture bumpers involving compressive impact loading under a wide range of temperatures. For modeling the effective elastic stiffness, some existing works have reported the prediction of stiffness for polymer-based composites with a good agreement in comparison to the experimental results [11–14]. However, these models accounted only for the filler volume fraction, aspect ratio (AR) and orientation of the particles. They did not consider any temperature and strain rate dependences, although neat polymers and polymer-based composites mechanical behavior is well-known to be sensitive to these factors. By considering the effect of temperature and strain rate, Richeton et al. [15] proposed a unified model for predicting the stiffness of amorphous polymers based on the equation of Mahieux and Reifsnider [16,17]. Recently, we extended Richeton model [15] to describe the dependence of the elastic modulus of PP-based nanocomposites on the testing temperature, frequency/strain-rate, nanofiller volume fraction and on the extent of exfoliation [18]. For modeling the yield behavior with the stain rate and temperature effects, Richeton et al. [19] have also proposed a new formulation of the cooperative model for amorphous polymers. Gueguen et al. [20] extended this cooperative model to predict the yield stress of semi-crystalline polymers by considering the composition of semi-crystalline polymer as two phases (amorphous and crystalline phases). Recently, Matadi et al. [21] incorporated a three-phase approach into Gueguen model to estimate the effective yield stress of PP-based nanocomposites under a wide range of strain rates and temperatures. In this new approach, the effect of nanoparticles and their distribution were considered to predict yield stress of the nanocomposites.

In this study, we proposed to improve the statistical stiffness model of Richeton for neat polymers with temperature and frequency/strain-rate dependences [15] by incorporating the Mori-Tanaka model (Tandon and Weng [22]) and Chow [23] single filler model to predict the effective elastic behavior of PP/EOC and PP/Talc composites. For the material that contains the two fillers (EOC and talc), we incorporated a "multiplicative" model [24,25] into the Richeton model to predict the effective modulus. For modeling the yield stress of the materials, we suggested extending the Gueguen model for neat polymers with temperature and frequency/strain-rate dependences [20] by incorporating the Pukanszhy model [26] and the "multiplicative" model to estimate the effective yield stress for PP-based composite with single filler and two fillers, respectively. In order to take into account the impact of the recycling on the stiffness and yield behaviors of the materials, we incorporated the reprocessing strengthening coefficients into the new models. In our new developed approaches, the stiffness and yield behaviors not only depended on the frequency/strain-rate and temperature, but also depended on the filler volume fraction and on the number of reprocessing cycle. These models were validated on PP-based composites with different weight percentages of fillers and with different recycling numbers. The experimental investigation consisted of dynamic mechanical analysis (DMA) testing and dynamic compressive testing by the use of the split Hopkinson pressure bar (SHPB).

2. Experiments

2.1. Materials and processing

An isotactic polypropylene (PP) referenced as Moplen HP500N from Lyondellbasell was used for this study. The melt flow index (MFI) of this PP was 12 g/10 min, while its density was 0.9 g/cm^3. For the impact modifier, we selected a metallocene ethylene octene copolymer (EOC) (Exact TM 8230, Exxonmobil). The MFI of this EOC was 30 g/10 min and its density was 0.882 g/cm^3. The talc powder, referenced as Steamic T1 CF, was supplied by Luzenac. The density of this talc powder was 2.78 g/cm^3.

PP-based composites with 10 wt % and 20 wt % of EOC or talc (PP/EOC, 90/10, 80/20, or PP/Talc, 90/10, 80/20, respectively) were prepared by a single screw BUSS Kneader extruder model PR46 at 200 °C and 50 rpm and under air. We also prepared the PP with 20 wt % of EOC and 10 wt % of talc (PP/EOC/Talc 70/20/10). It is worth noticing that these compositions are similar to the commercial ones for car bumpers [27]. To simulate the mechanical recycling, PP-based composites were subjected to multiple extrusion procedures using the same extruder and the same processing conditions, up to 6 recycling cycles. We believed that 6 processing passes were enough to generate significant degradation [27]. We presented in this paper the results obtained for the recycling numbers of 0, 3 and 6. Tensile samples for all the materials were injected by means of a Billion 90 tons injection molding machine. Note that neat PP and PP-based composites were labeled as PP neat, PP/EOC x/y, PP/Talc x/z and PP/EOC/Talc x/y/z, respectively, where x, y and z are the weight percent of PP, EOC and talc, respectively. The recycling numbers were labeled as 0P, 3P and 6P. The tested materials are summarized in Table 1.

2.2. Dynamic mechanical analysis

Dynamic mechanical analysis (DMA) was conducted on a Netzsch DMA 242C instrument. Specimens of dimensions of 16 mm × 10 mm × 3 mm were cut from the injected tensile specimens. In order to study the frequency sensitivity of the non-recycled and recycled PP-based composites, materials were subjected to deformation at the frequencies of 1 Hz and 10 Hz, at temperatures ranging from −100 °C to 140 °C and with a heating rate of 2 K/min. For DMA, we reported averaged values and representative curves resulting from 3 experiments per tested material formulation.

Table 1
Composition of the tested materials with the number of reprocessing.

Materials	EOC wt%	Talc wt%	Number of reprocessing
PP neat 0P	0	0	0
PP neat 3P	0	0	3
PP neat 6P	0	0	6
PP/EOC 90/10 0P	10	0	0
PP/EOC 90/10 3P	10	0	3
PP/EOC 90/10 6P	10	0	6
PP/EOC 80/20 0P	20	0	0
PP/EOC 80/20 3P	20	0	3
PP/EOC 80/20 6P	20	0	6
PP/EOC/Talc 70/20/10 0P	20	10	0
PP/EOC/Talc 70/20/10 3P	20	10	3
PP/EOC/Talc 70/20/10 6P	20	10	6
PP/Talc 90/10 0P	0	10	0
PP/Talc 90/10 3P	0	10	3
PP/Talc 90/10 6P	0	10	6
PP/Talc 80/20 0P	0	20	0
PP/Talc 80/20 3P	0	20	3
PP/Talc 80/20 6P	0	20	6