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Impact response of recycled polypropylene-based composites under a wide range of temperature: Effect of filler content and recycling

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ABSTRACT

This work aimed at investigating the thermal mechanical behavior of recycled polypropylene (PP)-based composites under dynamic loading. PP was blended by extrusion with different fractions of ethylene octene copolymer (EOC) as soft rubber toughening agent and talc as rigid reinforcing agent. To simulate mechanical recycling, the composites were grinded and re-extruded up to 6 times. The dynamic behavior was studied by means of a split Hopkinson pressure bar at various strain rates and temperatures. We found that neat PP and PP/talc composites presented a brittle behavior at low temperatures. The addition of EOC inclusions markedly improved the impact resistance of PP and PP/talc. The results also indicated that the impact resistance of PP/talc was improved with the recycling numbers due to a fragmentation of the talc particles during the reprocessing inducing a self-reinforcement. However, the impact resistance of PP/EOC decreased with the recycling due to chain scission mechanisms. Concerning PP/EOC/talc composite, its dynamic behavior was almost constant with the recycling number possibly induced by equilibrium between self-reinforcement and chain scission mechanisms. Complementary information about the dynamic behavior of the materials was deduced from optical microscopy investigation of the morphology after dynamic compression testing.

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1. Introduction

Polypropylene (PP)-based composites are increasingly becoming preferred engineering materials in the automotive application and in particular for both exterior and interior parts. However, the increasing use of PP-based composites results in huge plastic wastes from end-of-life vehicles (ELV). For both environmental protection and waste management considerations, it is of great importance to drastically increase the re-use and recovery rate of these plastic wastes. Among different recycling methods of plastics, the mechanical recycling is the simplest and the more ecological one where the post-used ELV plastics are mechanically grinded and reprocessed to produce new structural parts [1,2].

The mechanical reprocessing at high temperature, intensive shearing conditions, and with the presence of oxygen and impurities could lead to thermal, thermo-oxidative and thermo-mechanical degradations of the material and consequently, can affect the properties of the resulting recycled material [3–8]. The impact of filler content and mechanical recycling on the properties of PP-based composite were studied in our previous work where attention was focused on the evolution of rheological, chemical, physical and quasi-static tensile properties with the reprocessing number. It was found that recycling induced a reduction of PP molecular weight that was attributed to chain scission mechanism. The decreased molecular weight was accompanied with a decreased viscosity of the material. However, the reduction of the length of PP chains increased their mobility and hence, facilitated their rearrangement during the crystallization step. In addition, no significant oxidation was observed in the case of the recycled materials, due to the presence of anti-oxidants in neat PP [9,10].

To the best of our knowledge, the influence of the filler content and recycling number on the dynamic behavior of PP-based composites has received little to no attention. In the automotive industry, PP-based composites are generally used for the manufacturing of bumpers. For this application, if an accident occurs, composite structure could be subjected during the impact to a single loading mode as bending, tension, and compression, or to a combination of them. In this context, dynamic compression testing is generally considered as simple and practical way to investigate

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Table 1
Main properties for the virgin and recycled materials [9,10].

Materials	Recycling number	Weight-average molecular weight of PP[a]	Crystallinity index of PP (%)[a]	Glass transition temperature of PP (°C)[a,b]	Mean particle diameter of EOC (μm)	Mean particle length of talc (μm)	Mean particle thickness of talc (μm)	Mean particle aspect ratio (μm)
Neat PP	0P	214000 ± 5230	49.2 ± 0.2	12.2 ± 0.2	–	–	–	–
	6P	192300 ± 2550	50.6 ± 0.1	13.0 ± 0.4	–	–	–	–
PP/EOC	0P	189550 ± 500	48.9 ± 0.2	12.2 ± 0.6	0.27	–	–	1.42
80/20	6P	157300 ± 420	50.3 ± 0.1	13.1 ± 0.5	0.22	–	–	1.28
PP/Talc	0P	206900 ± 3540	51.2 ± 0.1	11.3 ± 0.6	–	1.91	0.66	3.06
80/20	6P	203900 ± 2400	52.2 ± 0.2	10.5 ± 0.3	–	1.84	0.54	3.77

[a] Average value with standard deviation.
[b] Glass transition temperature of EOC is about −43 °C.

the mechanical response of the materials for high strain rate impact. A detailed study of high strain rate and temperature sensitivities of non-recycled and recycled PP-based composites would be of high scientific interest for the development of a robust constitutive model. In particular, such a model could be used as a fully three-dimensional user subroutine (VUMAT) for commercial Finite Element code such as ABAQUS. The aim of this work was to study the impact of the content of ethylene octene copolymer (EOC) as toughening agent and talc as reinforcing agent on the dynamic compression behavior of recycled PP-based composites, prior and after mechanical recycling. The dynamic compression behavior was investigated by using split hopkinson pressure bars (SHPB) under a wide range of temperature and strain rate. The macroscopic mechanical properties were correlated with the morphology of the studied composites by optical microscopy (OM) observations after dynamic compression testing.

2. Experiments

2.1. Materials and processing

A highly isotactic polypropylene (PP) supplied by Lyondellbasell was selected for this study (reference Moplen HP500 N, Frankfurt am Main, Germany). This PP had a melt flow index (MFI) of 12 g/10 min and a density of 0.9 g/cm^3 [11]. The impact modifier of PP was a metallocene ethylene octene copolymer (EOC) supplied by Exxonmobil (reference Exact TM 8230, Brussels, Belgium). This EOC had a MFI of 30 g/10 min and a density of 0.882 g/cm^3, and it contained 72 wt.% of ethylene and 28 wt.% of octene. The talc powder was kindly offered by Luzenac (referenced Steamic T1 CF, Toulouse, France), and had a density of 2.78 g/cm^3 and a median diameter (D50) of 1.9 μm.

PP blends containing 0 wt.%, 10 wt.% and 20 wt.% of EOC or talc (PP/EOC, 100/0, 90/10, 80/20, or PP/talc, 100/0, 90/10, 80/20, respectively) were prepared by extrusion. PP with 20 wt.% of EOC and 10 wt.% of talc (PP/EOC/Talc 70/20/10) was also processed. These compositions are similar to available commercial PP composites for car bumpers [2]. The talc powder was dried for 12 h at 80 °C before mixing. The mixing of the materials was conducted with a single screw BUSS Kneader extruder model PR46 (Pratteln, Switzerland), with a screw diameter (D) of 46.5 mm and a length/diameter ratio L/D of 11, at 200 °C and 50 rpm and under air conditions. The obtained molten filaments of PP-based composites were quenched by a cold water bath and were subsequently pelletized using a rapid granulator. The pellets were dried in an air-circulating oven for 60 min to minimize the moisture for the subsequent step [12]. To simulate the procedures of mechanical recycling, neat PP and PP-based composites were subjected to multiple extrusion procedures using the same extruder and the same processing conditions, up to 6 recycling procedures. We considered that 6 processing passes were enough to identify the degradation mechanisms [2]. In this paper the results obtained for the recycling numbers of 0, 3 and 6 are shown.

Finally, tensile samples for all the materials were injected by means of a Billion 90 tons injection molding machine (Bellignat, France) with temperature profile ranging from 190 °C to 220 °C and a screw rotation speed of 180 rpm. Note that neat PP and PP-based composites were denoted 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. Concerning the recycling number, it was denoted as 0P, 3P and 6P.

Table 1 lists some main properties of the virgin and the recycled materials for which detailed information about the material characterization has been reported elsewhere [9,10].

2.2. Mechanical tests at high strain rates

A homemade split Hopkinson pressure bars (SHPB) was used to perform the uniaxial compressive high strain rate testing (Fig. 1). This apparatus consisted of three parts, a striker, an input bar and an output bar made of 316L steel, with lengths of 500 mm, 2903 mm and 2890 mm, respectively, and the same diameter of 22 mm. The strain gages were glued on the middle of the incident and the transmitted bars with a distance 1500 mm from the interface of the specimen and bars. The cylindrical samples with a diameter of 8 mm and a thickness of 3 mm were cut from the injected tensile samples and placed between the input bar and the output bar. It is to be noted that thin samples are more acceptable than thick samples to minimize the effects of the non-uniform stress field and inertia of samples in SHPB approach [13,14]. Petroleum jelly was used as lubricant to limit the friction between the bars and the sample. A furnace with two symmetrical resistance heaters was installed for high temperature testing. For the low temperature testing, liquid nitrogen was mixed with different contents of absolute ethyl alcohol depending on the required testing temperature. Four thermocouples were inserted into the furnace to measure and control the temperature and hence, to ensure a homogeneous testing temperature. At each change of temperature, the furnace was heated/cooled without samples for 30 min to obtain a uniform temperature field, and then the sample was placed between the two bars for 15 min to allow for thermal equilibration.

The striker was used to generate a longitudinal compressive wave. Once this compressive wave reached the incident bar, strain gages cemented on this bar recorded an incident wave $\varepsilon_I(t)$. The difference in the mechanical impedances at the interface between the incident bar and the specimen resulted in the fact that a part of the incident wave reflected back along the incident bar while the other part transmitted through the specimen and then within the transmitted bar. The reflected wave $\varepsilon_R(t)$ was measured by the same strain gages cemented on the incident bar. The transmitted wave $\varepsilon_T(t)$ can be obtained by the same type of strain gages glued on the transmitted bar. In addition, two speed sensors

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