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Natural Fiber-Reinforced Recycled Polypropylene: Microstructural and Mechanical Properties

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ABSTRACT: This article deals with short spruce fiber-reinforced recycled polypropylene materials. Polypropylene has been subjected to n extrusion and crushing cycles, in order to simulate the recycling process. Recycling consequences have been studied on five times and 10 times-recycled PP. Then, short spruce fibers have been added in the 10 times-recycled PP, with different fiber weight fractions: 10, 30, and 40%. For microstructural aspects, results obtained by DSC and WAXS show that added fibers lead to an increase of the polymer degree of crystallinity, and to the development of a new crystalline phase with the β form. Tensile tests have shown that fiber presence obtains high rigidity values, but embrittles materials. Fracture tests coupled with digital image correlation technique have also been performed. Results show the better accommodation of overstrains at the crack tip in the composite material. Thus, this new type of composite, made of recycled polymers and natural fibers, seems to be full of promise.

KEY WORDS: natural fiber, recycled polymer, crystallinity, crack, correlation.

INTRODUCTION

OVER THE PAST decade, the high modulus fibers such as glass, kevlar, carbon, etc., were used in numerous technical fields where high strength and stiffness are required. However, these classic fibers often cause considerable problems in terms of re-use and environmental impact [1]. Their composites end up in landfills, while some are incinerated after use and may contribute to pollution.

In recent years, manufacturing industries have shown environmental awareness in choosing materials and fiber reinforcements. The tendency to select biodegradable fibers has been a natural choice for reinforcing polymers to make them 'greener'. These fibers offer several advantages such as low cost, low weight, easy degradability, and high specific modulus [2–6]. In the study carried out by Nabi Saheb and Jog [7], the mechanical properties of some natural fibers such as jute, sisal, and flax fibers were

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Figures 1, 3–7 appear in color online: <http://jrp.sagepub.com>

compared to glass fibers. It was observed that specific moduli of these fibers are comparable to or better than those of glass fibers. This higher specific modulus is one of the advantages of using natural fibers.

Further efforts at the development of biocomposites combine natural fibers with synthetic polymers [1]. By adding the natural fibers in recycled synthetic polymers we do not only reduce waste disposal plastics but also take advantage of natural fiber properties. That is why composites made of natural fibers and recycled polymer matrix have been studied by various authors. Selke and Wichman [8] investigated the tensile and impact strength of wood and paper fiber reinforced, post-consumer, high-density polyethylene. They found that the addition of wood fibers tends to increase the impact strength but decreases elongation at break. In another study carried out by Singleton et al. [9], flax fibers were used to reinforce recycled high-density polyethylene in a composite laminate. Improvement in strength and stiffness during tensile tests and significant increase in impact toughness were obtained, depending on fiber volume fraction.

In this study, we are interested in the use of polypropylene, which represents the highest tonnage of polymer used in France, particularly for the automobile industry (except polyurethane). The consumption of polyolefin in the automobile industry rose to 325 KT in 2002. In addition, the European directive 2000/53/CE recommends the re-use of 95% of the material from vehicles out of use by 2012. Many studies have been performed on recycled polyolefin [10–12] in order to find a new way to re-use these polymers.

Polypropylene has been recycled mechanically up to 10 times in order to simulate the number of manufacturing processes. Then, spruce fibers have been incorporated in this recycled polypropylene. The consequences of the recycling process and of spruce fiber addition on polypropylene have been examined in terms of mechanical and microstructural properties. The modifications of microstructural properties have been studied by using two different experimental techniques: differential scanning calorimetry (DSC) and wide angle X-ray scattering (WAXS). In parallel, the mechanical properties have been examined in both tensile and fracture tests. In order to compare fracture mechanisms of materials, a digital image correlation method [13] has been used to obtain whole-field strain measurements around the crack tip.

MATERIALS AND SAMPLE PREPARATION

Recycling Method of Polypropylene

The polypropylene used (PP) is the APPRYL 3400 MA1 supplied by ATOFINA. It is an homopolymer polypropylene obtained by controlled rheology and presenting an isotacticity of 90% with the crystal form α . The polymer density is 0.905 g/cm^3 with a melt flow index of 42 g/10 min. Each cycle of recycling includes a crushing and a passage in the extrusion machine (Figure 1). This procedure was repeated n times with the same condition for each cycle. Then pellets obtained from the 5th and 10th cycles were introduced into the injection molding machine in order to produce two types of standard samples for tensile and fracture tests. Specimens were injected with a temperature varying from 150 to 190°C under an injection pressure of 400 bar and a holding pressure of 350 bar during 25 s. Degradations due to the recycling process were studied in the five and 10 times-recycled samples.

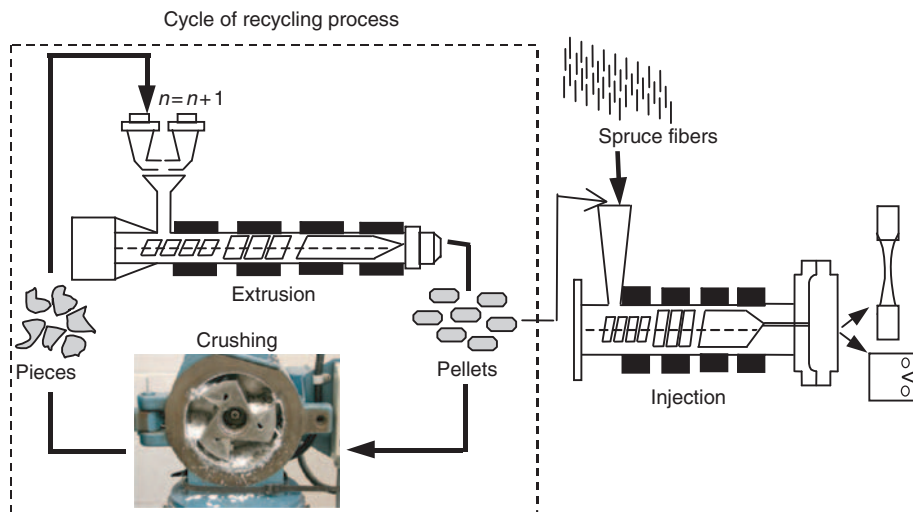


Figure 1. Recycling method of PP and processing method of PP and spruce fiber/PP samples.

Table 1. Comparison of cost and density for the studied materials.

		Relative cost	Density (calculated)
PP	Virgin PP	/	0.904
	5th cycle PP	/	0.904
	10th cycle PP	1	0.904
Composites spruce fiber/10th cycle PP	+10% spruce fibers	0.93	0.805
	+30% spruce fibers	0.80	0.658
	+40% spruce fibers	0.73	0.603

Spruce Fibers

Short spruce fibers were incorporated in the 10th cycle PP (Figure 1) with various fiber weight fractions: 10, 30, and 40%. The spruce fibers come from industrial waste and thus, are cheap. Fiber length and diameter were, respectively, 800 and 400 μm , and fiber density was 0.4 g/cm^3 , which is lower than the PP. Therefore, the obtained composites are lighter and less expensive in comparison to the virgin polypropylene (Table 1). A close inspection of fiber cross-section is shown in Figure 2. One can see on this SEM observation the complex microstructure of each spruce fiber. Each consists of cellulose fibrils wound in a matrix of hemicellulose and lignin components.

MORPHOLOGICAL ANALYSIS

Experimental Conditions

The material microstructure was characterized by two techniques: DSC and WAXS. Differential scanning calorimetry was carried out in the MDSC 2920 from TA Instruments. All samples were tested at a heating rate of 5°C/min in nitrogen atmosphere. The obtained thermograms (Figure 3(a)) show endothermic peaks of crystalline

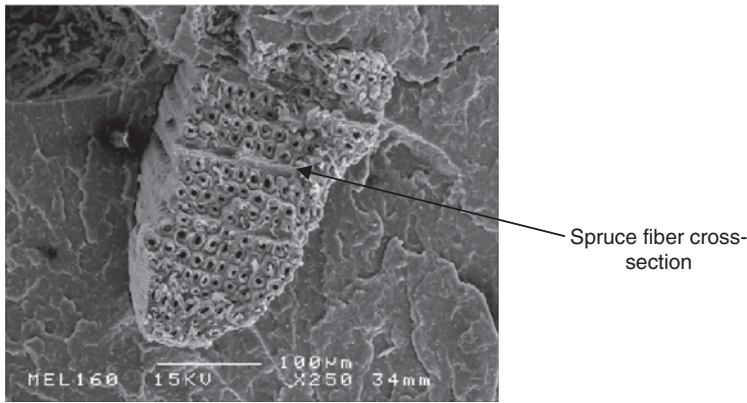


Figure 2. Section of a sample of spruce fibers/PP matrix observed by SEM.

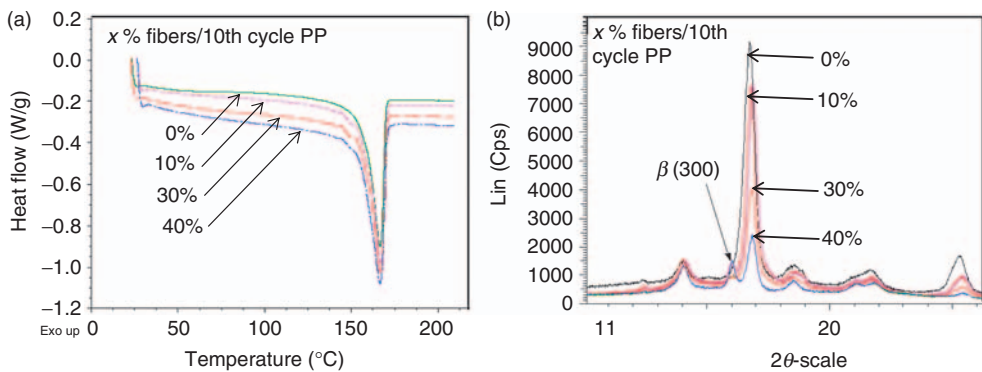


Figure 3. (a) DSC thermograms, (b) WAXS patterns, for various spruce fiber/10th cycle PP composites.

phase melting. The degree of crystallinity of polypropylene was determined by measuring peak area, and melting temperature was measured at the peak minima.

Wide-angle X-ray scattering patterns were taken by a goniometer using Cr radiation with a step of 0.015° . X-ray diffraction patterns show diffracted peaks that can be associated with crystalline planes, by using the Bragg law. Presence of β phase crystal, which has not been observed in pure PP, has been detected in WAXS scans of the composite materials. The relative content of β phase, denoted k_β , can be calculated according to Equation (1), suggested by Turner Jones [14]:

$$k_\beta = \frac{H_{\beta(300)}}{H_{\beta(300)} + (H_{\alpha(110)} + H_{\alpha(040)} + H_{\alpha(130)}}. \quad (1)$$

$H_{i(j)}$ denotes the peak height i corresponding to crystalline plane j in phase i .

Degree of Crystallinity

The thermograms obtained by DSC from 10 times-recycled PP and the PP-reinforced spruce fibers are presented in Figure 3(a). All thermograms show a large endothermic peak due to the α phase melting. In addition, a small peak located at about 147°C is detected in

Table 2. Comparison of microstructural parameters for the studied materials.

		Degree of crystallinity (%)	Proportion of β phase (%)
PP	Virgin PP	61 \pm 0.4	0
	5th cycle PP	62 \pm 0.4	0
	10th cycle PP	61 \pm 0.1	0
Composites spruce fiber/10th cycle PP	+10% spruce fibers	64 \pm 1	2 \pm 3
	+30% spruce fibers	68 \pm 2	13 \pm 3
	+40% spruce fibers	71 \pm 2	11 \pm 1

thermograms of the composites. This second endothermic peak corresponds to the melting of the β phase in polypropylene [15,16].

The measured values (Table 2) show a constant degree of crystallinity in the PP with the recycling process. Otherwise, this value was found to increase with the proportion of spruce fibers in the case of the reinforced PP. This observation can be explained by an assumption that the cellulose fibers act as efficient nucleating agents for the crystallization of PP. In fact, the fiber surfaces play the role of nucleation sites for the crystallization [17,18]. This higher degree of crystallinity will contribute to the improvement of the composite stiffness.

Crystalline Phases

The analysis of the diffractograms obtained by WAXS (Figure 3(b)) shows that the highest peaks corresponding to the α phase can be found at scattering angles of 14° (110), 17° (040), 18.5° (130), 21° (111), and 22° (131 and 041). The β phase can be detected thanks to the presence of a peak located at 16° (300) [15]. The results show that the dominating crystalline phase in PP-reinforced composite remains the monoclinic α phase. The proportion of β phase was estimated using Equation (1) and was found to increase with the fiber weight fraction: from 2% for 10% of fibers to 11% for 40% fibers (Table 2). This new phase, which has an important crack resistance [19], will contribute to improving the toughness of the composites.

MECHANICAL ANALYSIS

Tensile Behavior

The tensile tests were carried out at ambient temperature at a constant cross-head displacement rate of 20 mm/min. Experimental stress–strain curves for pure PP and spruce fiber-reinforced PP composites are plotted in Figure 4. In order to facilitate the legibility of this figure, the curve of pure PP was truncated.

The elastic modulus has been obtained from the initial slope of the true stress–strain plot measured by a clip-on extensometer. Experimental values of tensile properties (Young's modulus, elongation at break, yield stress) are summed up in Table 3. It has been observed that after 10 reprocessing cycles, the tensile modulus was reduced by 8.5% from its initial value, the yield stress was decreased by 7%, and the elongation at break was significantly reduced by 50%. Results obtained for composite materials show that spruce

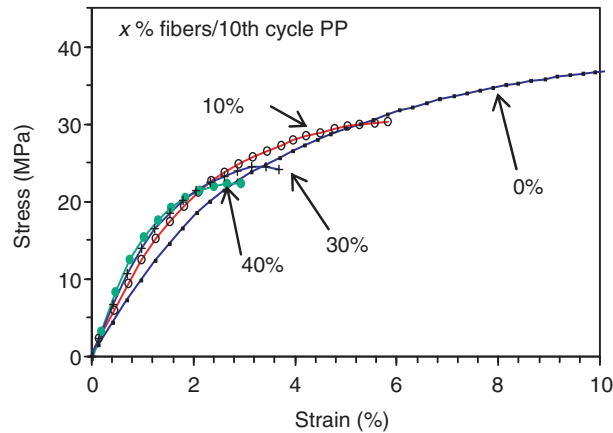


Figure 4. Tensile behavior of various spruce fiber/recycled PP composites.

Table 3. Comparison of mechanical characteristics for the studied materials.

		Young's modulus (MPa)	Elongation at break (%)	Yield stress (MPa)	Toughness (MPa \sqrt{m})
PP	Virgin PP	1880 \pm 51	120	41.5 \pm 0.1	2.2 \pm 0.1
	5th cycle PP	1820 \pm 23	82	40.7 \pm 0.2	–
	10th cycle PP	1720 \pm 03	63 \pm 14	38.6 \pm 0.6	2.5 \pm 0.1
Composites spruce fiber/10th cycle PP	+10% spruce fibers	1864 \pm 24	6.1 \pm 0.3	30.0 \pm 0.1	–
	+30% spruce fibers	2315 \pm 42	3.6 \pm 0.3	24.0 \pm 0.1	2.7 \pm 0.1
	+40% spruce fibers	2575 \pm 26	3.2 \pm 0.3	22.5 \pm 0.3	–

fiber addition allows us to compensate the degraded tensile stiffness. For fiber weight fraction of 30 and 40%, fiber presence enhances the tensile stiffness even further than in virgin PP. Otherwise, spruce fiber addition contributes to making the material considerably more brittle. It leads to a sharp drop in elongation at break and a decrease in yield stress.

Fracture Mechanical Behavior

Fracture behavior has been studied by testing a CT specimen in mode I crack propagation. Tests have been performed at a 1 mm/min cross-head velocity with normalized specimens [20,21] made of 10 times-recycled PP and composite of 30% spruce fiber/10th cycle PP. The load–displacement curves recorded during the test are shown in Figure 5. It can be noted that the fracture behavior of the composite material was considerably modified compared to the pure recycled polymer: the initial slope of the curve is higher, and that can be connected to the higher composite Young's modulus; the maximal applied load is far superior to the value obtained with the 10 times-recycled PP; and the crack propagation is stable in the composite, unlike a sudden failure occurring in non-reinforced PP.

The measured toughness is listed in Table 3. Fracture toughness increases 8% in the composite compared to the 10 times-recycled PP, and 20% compared to the virgin PP. In order to understand the fracture mechanisms in the two materials, fracture tests

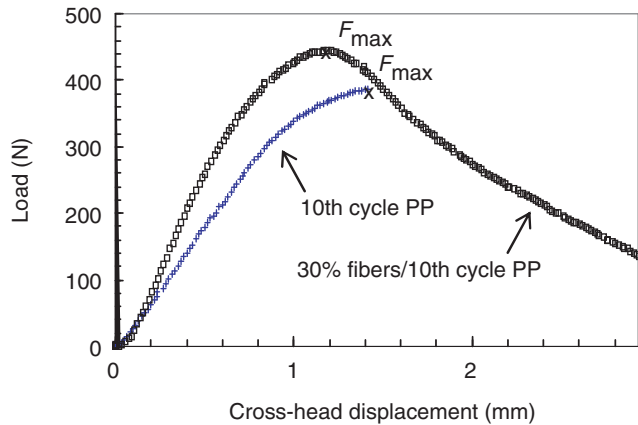


Figure 5. Fracture behavior of recycled PP with or without spruce fibers.

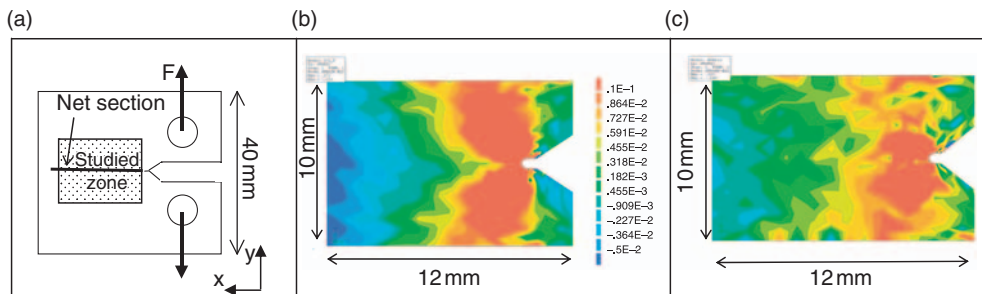


Figure 6. Digital image correlation measurements: (a) specimen geometry, (b) and (c) strain maps measured at F_{max} respectively for the 10th cycle PP, and for the spruce fiber/10th cycle PP composite.

were coupled with a digital image correlation method to obtain the whole-field strain measurement [13–22]. The principle of this technique is to record the random grainy pattern created on the specimen surface before and after loading the sample. In this case, the first image was recorded before loading and the second image was recorded at a maximal load. The correlation of these images was carried out using the GRANU.exe software, developed at the LPM laboratory. This technique allows us to measure the displacements at each ‘point’ on the sample surface with a precision of about 1 μm , each ‘point’ covering a small square of approximately $360 \times 360 \mu\text{m}^2$. Measurements were carried out in 561 ‘points’ inside a zone of $12 \times 10 \text{mm}^2$. The nearest point from the crack tip is located at a distance of about 270 μm from it. Experimental longitudinal strain maps measured at the maximal applied loads are presented in Figure 6 for the 10 times-recycled PP and the spruce fiber-reinforced recycled PP. Comparison of the two maps highlights the differences in strain distribution at the crack tip. In fact, the zone of overstrain around the crack tip shows a butterfly wing shape in the 10 times-recycled PP. On the contrary, this zone seems much more disturbed and scattered in the composite of spruce fiber/10th cycle PP.

In order to compare more precisely the longitudinal strain values measured at the crack tip in the two materials, we have also plotted the measured longitudinal strain ϵ_{yy} values along the net section in Figure 7.

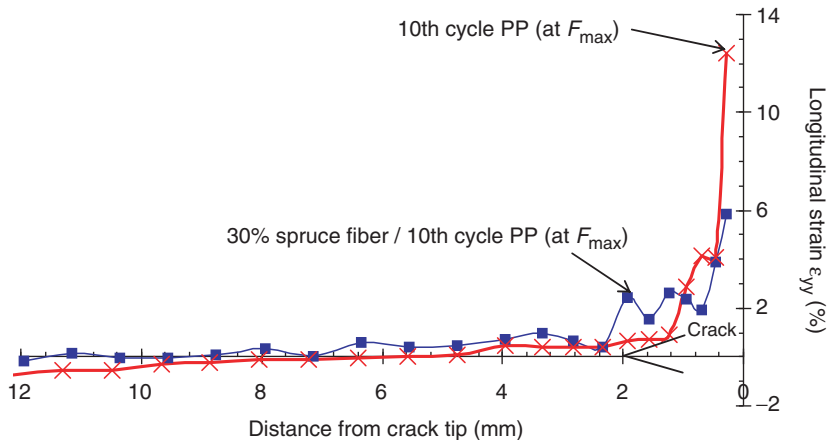


Figure 7. ε_{yy} strains measured by digital image correlation technique on the CT specimen net section at F_{max} for the recycled PP with and without spruce fibers.

The comparison of the strain values measured on the net section shows that the zone of overstrain value in the pure PP is concentrated near the crack tip with very high values: the highest strain value reaches up to 12.5% at maximal load. Otherwise this zone is larger in the composite (covering up to 2 mm from the crack tip) and the highest strain value obtained is two times lower (about 6%). All these results show the influence of spruce fibers on the overstrain accommodation around the crack tip: fiber presence enables a better distribution of the overstrain zone and thus reduces the strain value reached at the crack tip. This explains the higher toughness of the composite material.

CONCLUSIONS

The addition of spruce fibers in recycled polypropylene was studied in terms of microstructural and mechanical properties. It has been shown that the presence of spruce fibers in recycled polymer matrix causes the appearance of crystalline β phase and increases the polymer crystallinity. It leads to a stiffer but more brittle material in tension and it increases recycled polymer toughness, modifying the distribution of the high-strain zone in front of the crack tip. Thus, this new composite material generation made of recycled polymer and natural fibers seems to be full of promise: compared with the recycled PP, the obtained materials are less expensive, lighter, and have higher stiffness and toughness.

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