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Observation of the structure of a composite polypropylene/flax and damage mechanisms under stress

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ABSTRACT

The knowledge of the microstructure and of the damage mechanism under mechanical solicitation of polypropylene (PP)/flax fiber composites is a key point of their development. The aim of this work is to study the fibers orientation and dispersion as well as the damage development of an injected PP/flax fiber composite. First, we studied the flax fiber orientation in the skin and core layers, and, to quantify the impact of fiber bundles, we evidenced their presence in injected specimens, and we focused on their effects on the tensile properties. Then, we correlated this morphology with the tensile properties of the various areas. Finally, we investigated the initiation and propagation of cracks into the PP/flax composite in tensile mode by using in situ SEM observations. Despite some differences, our results highlighted many similarities between the flax fiber and glass fiber composites, and evidenced some improvements to carry out in order to increase the part of industrial PP/vegetal fibers composites.

1. Introduction

In order to improve the weak mechanical properties of numbers of polymers, short fibers are commonly used without any modification of the processing technique, such as injection molding or extrusion. These fibers improve the mechanical properties of the part, all the more so as their orientation is close to the main mechanical solicitations (Godara and Raabe, 2007). Due to their important mechanical anisotropy (Bourmaud and Baley, 2009; Cichocki Jr. and Thomason, 2002), this phenomenon is deepened with vegetal fibers (Herrera-Franco and Valadez-González, 2004; Joseph et al., 1999). Some particular characteristics of these composites are the flow-induced fiber orientation and the difficulty for the tool designer to get an optimal fiber orientation (Vincent et al., 2005). Nevertheless, adding short fibers to a polymer is relatively easy and could be very effective in reinforcing the polymer, inducing an increase of the stiffness and the strength, but as the same time, a decrease in the elongation at break.

The most common composites are thermoplastics, such as polyolefins or polyamides, reinforced with glass fibers. These composites are consumed in large volume, especially in the automotive industry. In recent years, we have noticed an expanding demand from manufacturers to turn to incorporating bio based materials in auto parts (PSA, 2010). This use is justified by ecological and economic interests. Plant fibers used in this study come from flax stem, and exhibit specific mechanical properties that are equivalent to those of glass fibers (Baley, 2002; Bourmaud et al., 2010; Pillin et al., 2011); due to this interesting mechanical behavior, flax fibers have become a credible alternative to glass fibers. Fiber-reinforced composite materials have a lower environmental impact than fiberglass reinforced composite materials (Bodros et al., 2007) and exhibit an interesting recyclability (Bourmaud and Baley, 2007; Bourmaud et al., 2011). Moreover, the plant fibers are renewable and require less energy in their development compared to glass fibers (Le Duigou et al., 2011).

The introduction of a new material requires knowledge of its microstructure – especially of the fibers' orientation, dispersion and distribution – and its mechanical behavior. If the microstructure and the orientation of fiberglass reinforced injected composites are well known, it is not the case for biocomposites. Indeed, the orientation of short glass fibers in a center-gated disk and the existence of a skin-core effect on injected parts have been demonstrated in numerous publications. By experimental works on composite disks, Bay and Tucker (1992a,b) showed that the injected part can be divided into several layers of preferential orientation of fibers, symmetrical about the mid-thickness of the sample. They noticed, from the edge toward the center a layer of biaxially oriented fibers, a layer of radially oriented fibers in the flow, a transition layer with random orientation and finally, a core layer where the fibers are oriented perpendicular to the flow. In addition to methods based on

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microscopic observation, some authors used ultrasonic backscattering to estimate fiber orientation in injection molded specimens (Bechtold et al., 1998) or X-ray tomography (Bernasconi et al., 2008; Schell et al., 2006). Tomography is a promising way which enables to visualize in situ orientation without any alteration or treatment of the composite and could be used on vegetal-fibers-reinforced polymers. Thus, Ragoubi et al. (2012) used X-ray tomography to estimate orientation of miscanthus fibers in a polylactic (PLA) or polypropylene (PP) matrix.

Some authors studied the orientation of vegetal fibers in thermoplastics composites in more usual ways. Peltola et al. (2011) investigated the orientation of flax and hemp fibers in starch acetate matrix injected composites through optical microscopy of polished cross-sections of composites; they shown that fibers were predominantly orientated in the melt flow direction. Bledzki and Faruk (2006) studied the elaboration of injection molded microcellular wood fiber-PP composites. They highlighted a three layer sandwich structure with various fibers orientations similar to a skin core effect. Correa et al. (1996) investigated the anisotropy of injected cellulose fibers composites and evidenced, by microscopic observations, that fiber alignment varies, depending on the position inside the mold, influenced by the flow induced at the injection point. A paper by Vilaseca et al. (2010) is dedicated to the evaluation of the structure and mechanical properties of injected abaca/PP composites. They proceeded to orientation measurements by through optical microphotograph analysis, and showed the existence of a skin core effect with perpendicular orientation at the flow direction of the injected part.

The mechanical properties of composites vary according to fiber orientation (Godara and Raabe, 2007; Herrera-Franco and Valadez-González, 2004; Joseph et al., 1999). In the injected part, the skin core effect has an influence on the mechanical properties of the different layers. Huang et al. (1999) used a mathematical model developed by Takao et al. (1982) to predict the values of Young's modulus for the skin and core layers in glass/PP composites. In the skin layer, the fibers tend to align parallel to the melt flow direction inducing superior mechanical properties than those of the core layer, where the fibers take up a more random orientation.

The mechanical behavior and fracture of a composite material can be studied by observing the damage mechanisms in the composite material. Micro-damage can be analyzed by in situ microscopic observations under stress. For example, Sato et al. (1991a) studied the damage mechanisms of composite polycarbonate/glass fibers. These damage mechanisms have been little studied in composites reinforced with vegetal fibers. We can consider one study by Beckermann and Pickering (2009) about the estimation of strength prediction modeling; they calculated the failure probability of PP-hemp composite and shown that the reinforcing fibers need to be equal to – or longer than – $4L_c$ (L_c = critical length) (Kelly and Tyson, 1965) to achieve the maximum theoretical composite strength. Gironès et al. (2011) investigated the impact properties of pp-abaca composites; they noticed that the incorporation of maleated PP coupling agent provides a higher resistance to crack formation but has a moderate influence on crack propagation. In the case of polymer reinforcement through the use of vegetal fibers, we can highlight the poor interfacial strength of these fibers; this point could be a restraint to the use of vegetal fibers. Consequently, theoretical determination of the critical length could have some limits, and it could be better to analyze the damage mechanisms by an observation way.

The aim of our study is, first, to analyze the microstructure of injected flax fiber reinforced PP. In a first step, after a morphological and mechanical study of the flax fibers that were used, we studied the core and skin samples microstructure thanks to optical observations. In a second step, through a mechanical and microscopic study, a relationship was shown to exist between the presence of fiber flax bundles flax and rupture of the specimens. The composites' microstructure was correlated to their mechanical properties. Then, finally, initiation and propagation of cracks in the composite were investigated by carrying out some in situ SEM tensile experiment.

2. Experimental

2.1. Materials

Flax fibers (Marylin variety) were obtained from the CTLN® Company (Le Neubourg, France). They were harvested in the North-West of France in the beginning of September 2003, being sowed at the beginning of April and pulled in August. After pulling, plants were laid over the field for drying for 4 weeks to allow dew-retting, that is the development of fungi within the stem, which degrades their cortical tissues and further facilitates the extraction of the fibers. The fibers were scutched, carded and cut to a 2 mm length. The moisture content of the fibers has been checked by using TGA measurement and found to be 6.3%. The composites contain 21% by weight of fibers (13.7 vol.%).

The polymer used as a matrix is a poly-(propylene) PPC 10642 from Total Petrochemicals®. The melt flow index of this PP is 44 g/10 min at 230 °C (under a load of 2.16 kg). In order to improve the compatibility between the fibers and the PP matrix, we used a maleic anhydride grafted PP (PP-g-MA), OREVAC CA 100, from Arkema®. The MFI of this compatibilizer is 10 g/10 min (190 °C – 0.325 kg). PP-g-MA is widely used as a coupling agent for vegetal fibers reinforced PP (Godara et al., 2009). PP with 4 wt% of PP-g-MA was used. For comparison, a short glass fibers (30 wt% or 13.7 vol.%) reinforced PP (A Schulmann®) was used.

2.2. Compounding and processing

Flax fibers were dried under vacuum at 60 °C during 12 h and PP was then extruded with PP-g-MA and flax fibers. Compounding was achieved in a single screw extruder at 20 rpm and with the following temperature profile: 175/180/190 and 190 °C in the nozzle. Compounded pellets were also dried under vacuum at 60 °C for 48 h.

Injection molding was then carried out on a Battenfeld machine. All parameters were kept constant during the injection molding process. The temperature profile was kept as follows: 165/170/175/180 and 180 °C in the nozzle. The mold temperature was maintained at 30 °C. Virgin PP, PP 13.7 vol.% flax fibers and PP 13.7 vol.% glass fibers were injected in a mold designed to produce ISO-527 normalized specimens.

To be sure of the fiber volume rate in our samples, density measurements have been carried out after injection molding. We calculated the fiber rate by a classical mixing rule, by knowing the fiber and matrix density. We found $13.9 \pm 0.4\%$ for the PP/flax composite and 13.8 ± 0.1 for the PP/glass one. The good correlation between the theoretical and experimental values could be explained by a suitable fiber proportioning and by the density measurement done on a significant sample of around 2 g.

2.3. Tensile tests on single fibers

Tensile tests on single fibers were carried out according to the AFNOR XPT25-501-2 norm, at a controlled temperature (23 °C) and relative humidity (48%), and longitudinal mechanical properties (Young's modulus, ultimate strength and failure strain) of elementary flax fibers were determined. Due to the short fiber length (about 20–30 mm), a gauge length of 10 mm was chosen. The fiber was clamped on a universal MTS type tensile testing machine equipped with a 2 N capacity load cell, and loaded at a constant