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Innovating routes for the reused of PP-flax and PP-glass non woven composites: A comparative study

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ABSTRACT

The significant industrial development of non-woven biocomposites requires the implementation of environmentally and economically coherent end-of-life recycling solutions. In this study, we studied the recycling of a non-woven poly-(propylene)-flax composite by injection but also by thermo compression. For comparison, a material with the same architecture but reinforced by glass fibres was studied. Both recycling methods showed strong specificities. Injection recycling leads to efficiently homogenised microstructures of the parts but also to drastically reduced lengths of the fibres, up to 10 times lower than with compression moulding. This method globally promotes high failure strengths while compression moulding, by preserving the length of the fibrous reinforcements, guarantees higher stiffness. This work also highlights the impacts of the length and division of the fibre elements on the microstructure of the injected parts; thus, after a series of compression recycling cycles, injected parts exhibit an important skin-core effect larger than after initial injection recycling cycles, whether in terms of orientation or local fibre volume fraction. As a consequence, after a series of recycling by compression, a new injection cycle has for effect to improve the tensile mechanical performances. For example, the strength and modulus of PP-flax composites are increased by 103% and 75%, respectively. These results highlight the technical feasibility and relevance of implementing these two recycling methods, depending on the volumes or equipment available and the final properties to promote, as they enable the production of new high-performance parts.

1. Introduction

The end of life of composites is a major issue for this industry and the depletion of fossil resources requires producers to promote the recycling of parts after use. Some sectors, such as the automobile, have implemented drastic regulations to comply with standards and European directives [1] that promote end-of-life treatment through the recyclability of materials [2–4]. In response, the design of mechanically recyclable composites involves the development of thermoplastic matrices instead of thermoset resins [5].

Currently, many researches and industrial developments dealing with the use of natural fibres to substitute glass fibres in some applications are conducted. Indeed, they are produced from renewable resources, need 5 to 10 times less non-renewable energy, have a low density inducing interesting specific mechanical performances [6] and are able to store carbon dioxide thanks to photosynthesis [7,8]. These factors contribute to a great reduction of the environmental impact during car manufacturing and use [9]. In Europe, flax and hemp are the most frequently used natural fibres due to their high specific mechanical properties [6,10,11] and moderate cost. European production land reaches 114.000 ha per year and France grows 75% of the European flax fibre (2001–2008), being thus the first worldwide producer. When these plant fibres are associated with bio-based or biodegradable matrix such as PLLA/PHB alternative end-of life routes such as composting [12,13]

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can be carry out. However, those matrices are not easily usable in automotive applications due to their low thermal stability, low tenacity and high price. For these reasons PP is often preferred because of its good mechanical property and chemical stability [14,15].

In this context, the use of compression moulded non-woven PP-flax biocomposites is greatly appreciated in vehicle manufacturing (mainly in car interior parts); these materials offer a short time process cycle, limited raw material cost [16] and they can potentially offer both sound absorption (thanks to porosities) and good mechanical properties [17]. It is possible to vary their porosity in a range from 5 to 60%, so that to obtain configurable materials, covering a large panel of applications. If a 60% porosity content appears to be ideal to reach good absorption performance, in this case, the mechanical properties, for a fibre fraction of 40%-vol, dropped drastically from $E_{0\%} = 6$ GPa to $E_{60\%} = 1$ GPa and $\sigma_{0\%} = 40$ MPa to $\sigma_{60\%} = 10$ MPa for the tensile modulus and maximal stress, respectively. In the automotive sector, non-woven use is divided fairly between glass and plant fibres. Mechanically speaking, literature shows that non-woven composite moduli are generally included between 5 and 7 GPa for both low porosity PP glass or flax non wovens [18–20]. The strength at break is generally higher when glass fibre is used [18,20]. Interestingly, when maleic anhydrid grafted PP (PP/PPgMA) is used modulus and strength at break of flax composites are highly improved reaching 9.5 Gpa and 95 MPa [18,19,21], respectively.

The main drawback of non woven composite manufacturing is the waste production because ~25%wt of the used semi-product is thrown away. Due to the previously described advantages of plant fibres and the necessary need to value the non wovens wastes, recycling is a preferable alternative to incineration and landfilling because it helps to dispense with all or part of production phase [22], save raw material and a level of performance can be maintained. A viable solution could be the reutilisation of theses wastes for injection moulding which is the most common way to recycle such products. Biocomposites showed that they may exhibit an interesting recycling behaviour characterised by successive process cycles inducing only few modifications in tensile modulus and strength values for vegetal fibres whereas for glass fibres the decrease was very substantial, particularly with a stable PP matrix [23–25]. This phenomenon is mainly explained by the evolution of fibre length but also by the poor adhesion between glass fibres and matrix after several cycles.

In addition to fibre length or microscopic analysis, it is possible to deeper investigate the composite by using tomography or micro-tomography which are powerful tools to explore microstructure of plant fibre bundles [26,27] but also to investigate structure or damage in plant fibres composites. Thus, the fibre-bundle structure within a composite can be studied through tomography for modelling investigations [28]. Moreover, Almansour et al. [29] and Rask et al. [30] used tomography to investigate damages within flax/basalt vinyl ester hybrid composites and PP-flax UD composites, respectively, without any alteration of the composite structure as it is the case for SEM observations, for example. Tomography is also commonly used to better know the fibre orientation and porosity content within injected composite as demonstrated by Martin et al. [31] on PP-flax or Albrecht et al. [32] on PP-sisal materials.

In a previous study [33], two innovative routes for flax non wovens recycling were explored; a first one consists in introducing ground wastes in injectable compounds and the second one consists in the reintroduction of these shreds in new-non wovens. It was proved that this incorporation was structurally and mechanically successful until a reincorporation rate of 40%-wt. The aim of the present paper is to explore a new recycling possibilities, representing an important issue for these kind of products.

The recycling of PP/flax or glass non wovens was firstly investigated by carrying out four successive injection cycles without any compounding stage. The evolution of the fibre length as well as the evolution of the rheology, the mechanical properties and the microstructure of injected samples were monitored all along the cycles. Both tomographic and SEM analysis were performed to analyse the microstructures of the successively recycled parts. The results were compared to another way of recycling inspired from the manufacturing of chopped tapes by compression moulding. The interest of this process is to combine the advantages of using centimetre fibre length (on the contrary to injection moulding that uses millimetric fibre length) and the possibility to manufacture complex shapes [34–41].

However, instead of using chopped tapes, this work proposes to use composite manufactured flat parts as initial raw material to mimic the use of composite production scraps or composites at the end of their life. The process consisting of compressing centimetric piece of an initial composite (with a defined or a random shape) is used in this context as a recycling technique. The main challenge here consists of using pieces of composite initial materials that are initially in a hard form on the contrary to the chopped tapes. Moreover, these pieces of composites possess initial thicknesses that are much higher than the chopped tapes and one can expect that the flow and the rearrangement of the pieces of composites arranged randomly in the mould with a defined covering factor do not behave in the same manner as with chopped tapes. It is therefore proposed in this work to compare this innovative recycling route using centimetric pieces of composites (with therefore centimetric fibre lengths) to a direct injection moulding recycling route (without any addition of fresh polymer) during four successive recycling steps.

More precisely, after compression moulding, non-woven plates were specifically cut and successively hot pressed. Three compression cycles were carried out before a final injection cycle. The two recycling routes were compared through mechanical and structural investigations.

2. Experimental section

2.1. Materials

Flax tows (Linum usitatissimum) were used as reinforcement fibres and cultivated in Normandy area (France) in 2014. They were combined with black polypropylene fibres to manufacture commingled nonwoven according to the carding/cross lapping/needle punching technology developed by EcoTechnilin® SAS (Valliquerville – France). Recycled E-glass fibres were also combined with polypropylene to manufacture a second nonwoven. The fibre-matrix ratio is 50-50%wt and areal weights were 2290 g/m^2 and 2000 g/m^2, respectively for PP/flax and PP/glass nonwovens. Moreover, the nonwoven reinforcement is considered quasi-isotropic [42]. The melt flow index (MFI) of the non-woven PP is 25 g/10 min (at 190 °C and under a load of 2.16 kg).

2.2. Nonwoven plates compression moulding for injection moulding and compression moulding recycling cycles

Commingled PP/flax and PP/glass nonwovens were produced by hot compression moulding for injection moulding recycling cycles. A LabTech Scientific 50 T hydraulic press was used to manufacture plates with a porosity content as close as possible to zero. They were obtained by setting the plate thickness to a constant value (3.5 mm) and stacking two nonwoven plies. The plies were hot pressed at 200 °C and 20 bars during 8 min. The product was then