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Innovative flax tapes reinforced Acrodur biocomposites: A new alternative for automotive applications

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ABSTRACT

Flax Acrodur biocomposites are elaborated with an innovative flax reinforcement consisting of long technical fibers unidirectionally arranged without any weft and twist. The fibers cohesion is performed by using a new process consisting by reactivating the pectin cement. A polyester thermoset matrix (Acrodur) is used to impregnate the flax reinforcement and to produce unidirectional (UD) laminates. The relationship between the main process variables (drying, fibers content, densification and curing parameters) and the properties of the biocomposites is investigated. The optimized biocomposites have an elastic modulus of 18 ± 1 GPa with 55% wt.% flax fiber content and a low density of 0.93 g/cm^3. The thermal stability of the developed biocomposites is also investigated by Differential Scanning Calorimetry (DSC), Thermogravimetric Analysis (TGA) and Dynamic Mechanical Analysis (DMA). DMA results show a slight change of the storage modulus in a range of temperature from 23 °C to 160 °C. The appropriate processing parameters for the biocomposites are established. The developed flax tapes reinforced Acrodur biocomposites have a potential to be integrated for automotive applications thanks to their high stiffness/weight ratio and environmental advantages.

1. Introduction

To reduce the carbon dioxide amount released into the atmosphere, the car makers are actually looking for extremely lightweight materials in order to decrease the fuel consumption. Whereas glass fibers in composite parts become critical due to their weight and recycling difficulty, incorporating renewable resources such as long flax fibers allows creating biocomposites with high stiffness/weight ratio [1–3]. Flax fibers present a high strength, low density and better environmental impact. Their derived bio-based composites have an advantage over the glass fiber reinforced composites in terms of specific mechanical properties [4–8]. Furthermore, flax fibers are cheap and biodegradable materials, coming from a bio-sourced agriculture and widely available over the world.

Baets et al. [9] have highlighted the impact of flax reinforcement architecture on the resulting properties of derived epoxy composites. They explained that the unidirectional flax reinforcement without any twist gives to the epoxy composite the best mechanical properties compared to the woven, crimp and knitted flax reinforcements. The introduction of twist decreases the fibers bundles performance. A higher amount of twist and crimp leads to a lower stiffness and less effective impregnation due to the dense structure of yarns. Furthermore, the literature has shown that the characteristics of elementary flax fibers depend on their morphology, biochemical composition and on the presence and location of defects in the stem. Charlet et al. [10] investigated the impact of biochemical and morphological characteristics of flax fibers on their mechanical properties. They have shown that the scattering tensile mechanical properties of flax fibers are dependent on their lengths and on the stem diameter. Charlet et al. [7] have observed that there are three different types of behavior during a tensile test associated to the structural organization, such as the hypothesis of a progressive alignment of the cellulose microfibrils in the thickest cell wall following the tensile axis. Lamy et al. [11] and Hughes [12] have studied the influence of flax fiber defects on the stiffness properties and found that the mechanical properties of laminates depend on the defects (dislocations, kinks or micro-compression) and their dispersion in

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the fibers. Koronis et al. [1] reported that a significant variation in flax fibers properties can be observed and explained by the different harvesting season and regions and by the sun, rain and soil conditions.

Automotive parts industry is highly selective in terms of the matrix characteristics. Automotive specifications require using matrices with good viscoelastic properties, high thermal stability and a short processing time. The most common system used today is polypropylene in nonstructural trims. This matrix is favored for its low density, excellent processability, mechanical properties and low costs [2,3,13,14]. However, it is not well suited for developing structural systems. Manufacturing skins of sandwich panels requires using a matrix with a good thermal stability and having high thermomechanical properties for bearing heavy loads from ambient temperature to 80 °C. Furthermore, natural fibers have a strong polar character which creates many problems of incompatibility with most thermoplastics especially polyolefins [15,16]. A number of conventional thermoplastics and thermoset matrices have the ability to reinforce natural fibers [4,8,17,18]. Muralidhar et al. [19] investigated the thermal and viscoelastic properties of flax preform reinforced epoxy composites and showed that the storage modulus of the virgin epoxy resin and its flax derived composite were drastically affected by temperatures over 70 °C. Oksman et al. [18] compared the thermomechanical behavior of two flax reinforced thermoplastic composites. They found that at ambient temperature, the mechanical properties of flax/Polylactic acid (PLA) composites are promising and superior to those of flax/Polypropylene (PP) composites, but these properties decrease drastically as the temperature exceeds their glass transition temperature. John et al. [20] have characterized the viscoelastic properties of flax reinforced polypropylene composites. Their results show that the thermal stability and the viscoelastic properties increase with the incorporation of nonwoven flax at temperatures lower than 40 °C. But, when the temperature increases the storage modulus of the composite material decreases dramatically.

Despite the great potential of thermoplastics at room temperature, they have limited applications at high temperatures. All these considerations have been taken into account to choose a matrix with promising characteristics fulfilling the automotive specifications. The choice was done on a thermoset acrylic polyester resin (Acrodur DS3530 of BASF). Liang et al. [21] have studied kenaf composite based on the same resin. The Fourier Transform Infrared Spectrometer (FTIR) spectra of the biocomposites showed the formation of ester linkages between the carboxyl groups of the Acrodur resin and the hydroxyl groups of the cellulosic fiber during hot pressing. The Acrodur resin allows reducing the hot compression time of the biocomposites. The swelling thickness and water absorption of the composites were reduced by 55% and 64%, respectively. Further, the bending strength and modulus were improved by 72% and 188%, respectively. Medina et al. [22] investigated the dependence of the mechanical properties of natural fibers reinforced Acrodur composites on the process pressure. They concluded that the properties of the composite display clearly the dependence on the molding parameters, especially on the pressure and pressing time. They showed the importance of Acrodur prepregs drying under vacuum and the negative effect of the moisture content on the composite visual aspect and mechanical properties. They reported also that Acrodur exhibits a good adhesion with natural fibers. In automotive industry, the hot compression time has not only a mechanical effect but also a significant economic impact: automotive parts should be produced in less than 3 min.

This work aims to develop new flax reinforced polyesters composite which will be used to manufacture automotive structural systems supporting heavy loads (up to 100 kg). To get high mechanical properties, a new nonwoven flax reinforcement has been developed by considering its optimal architectural characteristics. It has unidirectional architecture and does not contain any twist and crimp. It is based on long aligned technical flax fibers without any load carrying thermoplastic fibers. This paper also presents the optimization of the processing parameters and the results of the thermomechanical behavior of flax tapes reinforced Acrodur composites.

2. Materials and processing

2.1. Dry flax tapes reinforcement

Long flax fibers are used to develop weight-controlled nonwoven flax tapes [23]. The fibers are unidirectionally arranged without any twist. To maintain the cohesion of the parallel fibers, a new process based on the reactivation of the pectin cement is adopted by spraying a water mist on the surface of fibers bundles. The processing of flax tapes begins by decorticating and separating the bundles from the flax stems. The tapes are then cleaned by scutching to remove the remaining shives and woody core of the stems. The flax bundles are aligned to the input of the manufacturing machine in order to be stretched, moistened by a water mist and continuously dried through an infrared oven (Fig. 1).

This process holds together the fibers to obtain unidirectional nonwoven flax fiber tapes of 50–200 g/m^2 weight and 40 cm width (Fig. 2).

2.2. Acrodur resin (DS 3530)

Acrodur resin DS 3530 (BASF Company) was chosen to manufacture flax composite materials for automotive industry. It allows good mechanical properties, short crosslinking time and easy handling and cleaning. It is a dispersion of a modified polycarboxylic acid and a polyol (cross-linking agent) creating a thermoset material (Fig. 3). This formaldehyde-free binder is adapted to wood and vegetable fibers such as flax, sisal or jute. Acrodur enables to overcome adhesion problems with natural fibers and reduces the processing costs by avoiding surface treatments. For other polymer matrices, treatment of fiber or/and matrix is needed to optimize the adhesion. Physical and chemical methods of surface modification for natural fibers are reviewed in [24]. Curing in presence of moisture activates bonding by an esterification reaction between the acid group of Acrodur and hydroxyl groups of the flax tape constituents. Acrodur resin has the ability to form hydrogen and covalent bonds with the hydroxyl groups present in cellulose through ester groups and the composite produced is more hydrophobic. When a good fiber–matrix adhesion is achieved, the path of wicking water molecules is tortuous due to better fiber–matrix integrity.

The initial water content is about 50% but it can be diluted for an easier impregnation. The cross-linking needs less than 3 min curing time at 160–180 °C. Comparatively the cross-linking of epoxy resin needs more than 1 h at 150–200 °C, which considerably increases the process cost.

2.3. Manufacturing of flax tapes reinforced composites

For the manufacturing of composite laminates, dry flax tape reinforcement of 70 g/m^2, 120 g/m^2 and 200 g/m^2 were used. However, we will only present the results for the flax tape reinforcement of 120 g/m^2. In fact, the flax tape of 200 g/m^2 does not allow a good impregnation and the flax tape of 70 g/m^2 requires more plies to obtain the same stiffness as the 120 g/m^2 (3 plies of 120 g/m^2 vs 5 plies of 70 g/m^2) leading to a loss of productivity.

The manufacturing process consists of four steps as shown in Fig. 4: (1) Impregnation, (2) Drying under vacuum, (3) Stacking, (4) Curing. The dry flax tape of 120 g/m^2 was impregnated manually by using roll coating method. Before drying, the weight