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Pre-impregnated natural fibre-thermoplastic composite tape manufacture using a novel process

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ABSTRACT

Pre-impregnated flax and thermoplastic poly(amide) composite tapes have been produced using a novel process. The manufacturing method uses an impregnation unit with a siphon system to impregnate continuous flax yarns with the polymer in the form of a slurry. After water evaporation, the powder is sintered and the coated yarns are compressed by passing them through a pair of heated rollers. Using a parametric study of the process, tape quality has been assured using the key outcome criteria of tensile strength/stiffness, surface roughness, fibre weight fraction, width and thickness. The temperature of the air heater placed before the roller has the biggest influence on tape quality. A heating model was developed using finite element software LS-DYNA. The research novelty comes from producing composite tapes with good tensile properties and surface finish using aligned *natural* fibres; the feasibility of automated tape placement and winding has also been demonstrated.

1. Introduction

An increased awareness of the environmental impact of non-recyclable and non-biodegradable plastics and other materials during composites manufacturing and their end-of-life disposal has led to strong interest in finding nature-based alternatives to synthetic materials. Composites that utilise natural fibres, which are derived from plant materials, and recyclable or biodegradable polymeric matrices make both manufacture and disposal more environment friendly. At present, the use of natural fibre composites is mainly restricted to non-structural components due to limitations in mechanical properties. The typical performance of a natural fibre composite is inferior in comparison to those of synthetic composites. However, the specific properties of some natural fibres are close to or even better than those of E-glass fibres, which indicates that their composites' performance can be improved. The primary focus of this paper is to demonstrate a new manufacturing process to fully realise the potential of natural fibre composites in the form of thermoplastic impregnated tapes. The challenge with unidirectional natural fibre composites is to align the fibres in one direction, as the fibres are only available in a discontinuous form. While the manual alignment of individual fibres is feasible on the laboratory scale, it would be difficult and time consuming to use this method to produce even moderate-sized real parts on a commercial basis. Arranging the fibres into a yarn holds individual fibres together by applying twist. This overcomes some of the difficulties of aligning discontinuous natural fibres within a composite, although the twist means that the fibres will not be straight [1]. Thus the strength and stiffness of the composite will be lower than those in the case of continuous, fully aligned fibres.

In general, the strength and stiffness of a composite increase as the fibre content increases. This occurs until a maximum value is reached, at which point the matrix can no longer properly wet out the fibres, and both strength and stiffness begin to decrease [2]. If natural fibres are to be used in structural applications, more emphasis needs to be placed on producing composites with a fibre volume fraction between 0.5 and 0.7. In areas where high quality composites are required, pre-impregnated ('prepreg') materials are generally used. They allow high quality composite parts to be produced, with low void content and high fibre volume fractions. Creating natural fibre composites using prepregging technologies should therefore enable parts to be produced with the same advantages. One elementary form of a prepreg material is thermoplastic composite tape. It consists of aligned fibres embedded in a thermoplastic polymer and is typically available in widths ranging from 5

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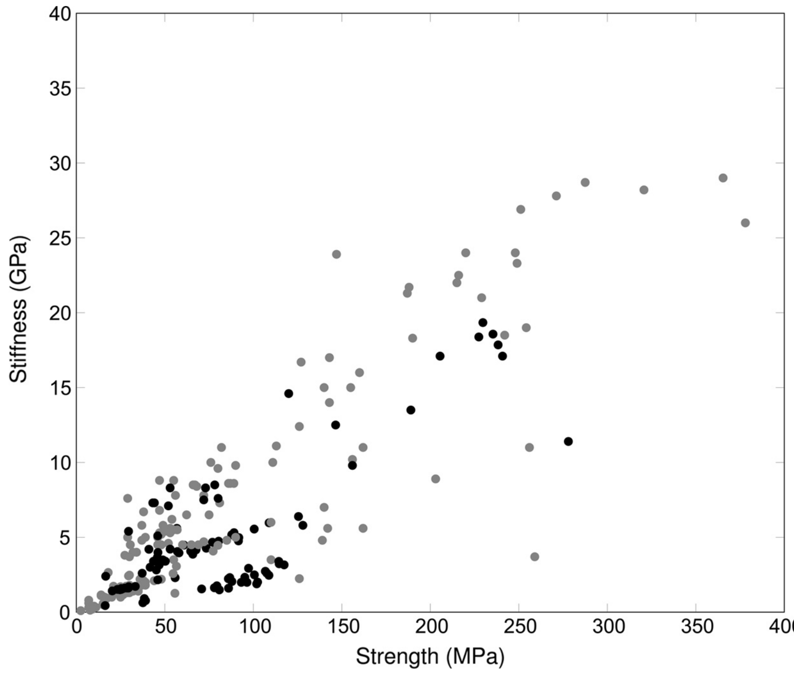

Fig. 1. Tensile strength and stiffness of natural fibre composites. Black markers represent biodegradable polymeric matrices, while other matrices are shown in grey.

to 300 mm. Tapes are mainly produced by powder or melt impregnation [3–7], using synthetic materials.

Fig. 1 shows the tensile strengths and moduli of natural fibre composites, as found from a review of the literature [1,2,8–42]. Composites which use biodegradable polymeric matrices are indicated by black markers while those using other types of polymers are represented in grey. It can be seen that the majority of natural fibre composites have tensile strengths and moduli below 150 MPa and 20 GPa, respectively. Some of the best performing biodegradable polyester based natural fibre composites have been achieved with poly(lactic acid), PLA [18]. There are very few cases reported in the literature where natural fibre composites have greater than 300 MPa tensile strength and 25 GPa tensile stiffness. Among these, Hepworth et al. [17] achieved a tensile strength and stiffness of 378 MPa and 26 GPa, respectively, using bundles of combed decorticated flax fibres that were pre-treated with a 50% mixture of poly(vinyl alcohol), PVA, and water. A very high final fibre volume fraction of 0.8 was claimed to have been achieved. Ochi [35] tested Manila hemp and other natural fibres, to make composites using a starch-based emulsion resin. The best tensile properties of 365 MPa strength and 29 GPa stiffness were obtained at the highest fibre volume fraction of 0.7. Madsen [2] achieved properties of up to 320.7 MPa tensile strength and 28.7 GPa tensile stiffness with flax and poly(propylene). In this case, flax yarn was wound onto metal frames, producing fibre assemblies with high yarn alignment and controlled uniform thickness. Poly(propylene) matrix foils were then combined with the fibre assemblies using a film stacking technique and the entire composite stack was then processed by heating and vacuum consolidation. These results indicate that with the right materials, composite structure and manufacturing techniques, high mechanical properties can indeed be achieved with natural fibres.

With respect to the desired composite structure and its manufacturability, a natural fibre-thermoplastic tape manufacturing process would have a number of advantages. The tape making process can be semi or fully continuous, improving process speed and the ease of scalability for large volume production. Producing a tape material as a precursor would allow fibres in the final product to be highly aligned in the direction of loading, enabling the production of a well-designed and efficient part. The flexibility and width-wise scalability of a tape manufacturing process would also allow the same process to be used to manufacture unidirectional sheets, which are in effect wide tapes. The work presented in this paper details the development of a continuous natural fibre-thermoplastic composite tape manufacturing process using consolidation rollers. The process has been evaluated using the experimental design methodology proposed by Taguchi [43]. This allows the effects of many different factors on the outcome criteria to be examined by a condensed set of experiments. The manufacture of natural fibre tapes may be seen as an essential step towards improving the mechanical properties of natural fibre composites, allowing a move towards a structural range of product capabilities.

2. Manufacture of pre-impregnated natural fibre thermoplastic composite tape

This section describes the manufacturing of pre-impregnated natural fibre-thermoplastic composite tapes using an innovative processing methodology. A Belgian variety of flax fibre in the form of the smallest available 3-yarn size of 82.7 tex was selected, and supplied by Jaya Shree Textiles, India. Flax fibre has superior tensile properties (800–1500 MPa strength and 60–80 GPa modulus) [44,45]. Each yarn was approximately 0.27 mm in diameter, while the individual fibres themselves were 20–30 μm in diameter (as per manufacturer supplied data), and had lengths of several millimetres. The yarn had a specific strength of 0.29–0.33 N/tex, and a density of 1.49 g/cm^3. Yarns spun from bleached flax fibres were selected to avoid fat, wax, pectin and other impurities, which could cause mechanical and chemical adhesion problems between fibres and the matrix. To explore the possibility of producing truly biocomposite tapes, a biodegradable polymer, poly(lactic) acid (PLA) was initially chosen as the matrix material. Polymer 4042D, an extrusion/thermoforming grade of PLA from NatureWorks LLC, USA, has a strength and stiffness of 60 MPa and 3.5 GPa, respectively (manufacturer datasheet). The PLA was dissolved in Tetrahydrofuran (THF) solvent, and a solution impregnation method was used to coat the PLA onto the flax yarn. A thermoplastic composite tape making process using continuous compression moulding of