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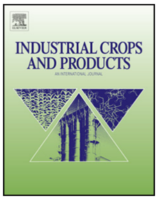

Direct impregnation of thermoplastic melt into flax textile reinforcement for semi-structural composite parts

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ABSTRACT

We performed an experimental study to manufacture flax fiber textile reinforced thermoplastic composites by direct impregnation method in a closed mold. To reduce the process time, we adopted compression resin transfer molding resin because of its short flow path during the highly viscous resin impregnation process. Fiber preheating process was introduced before the impregnation process in order to avoid the resin cooling down when contacting the flax fibers. Optimal process temperature conditions such as mold temperature, resin temperature and fiber temperature were experimentally determined while minimizing the effect of thermal degradation of flax fiber and obtaining good impregnation quality. In particular, a sequential pressure cycle was proposed to improve the impregnation quality and to obtain the uniform fiber volume fraction in the final part. The impregnation quality was assessed by the density measurement and by the micrographic image analysis. The mechanical properties of the final parts were comparable with the literature values obtained by the compression molding of matrix-fiber premixed materials such as commingle yarns or film stacking, whereas the process cycle time of was much shorter than the other conventional methods in the literature.

1. Introduction

Recently, the strict environmental regulations and the oil price volatility have led to growing interest in bio-based materials which are from renewable resources (Carus et al., 2015). In particular, natural fibers have been attracting great attention from many industrial sectors because of their low cost, low density, high specific mechanical properties and recyclability (Saheb and Jog, 1999). Among the natural fibers, flax fiber has a great specific modulus and thus it is regarded as one of candidates to replace E-glass fibers which are the most common synthetic fiber reinforcements (Bodros et al., 2007).

Indeed, many automotive parts have already been made from flax fiber reinforced polymers mainly based on discontinuous fiber reinforced polyester or polypropylene (PP). Nevertheless, due to the short fiber length and low fiber volume fraction, the mechanical properties of those materials are relatively low. Therefore, flax fiber composites have been used mainly for non-structural automotive parts such as door panel, car roof and parcel shelves (Summerscales et al., 2010). Currently, a principal reason for the adoption of flax fiber composites in automotive industry is eco-friendly marketing rather than technical demand.

Conversely, the use of continuous fiber reinforcement can be an effective way to enhance the mechanical performance of composites. Natural fiber textile reinforced thermoset composites have been fabricated by resin transfer molding (Oksman, 2001) or vacuum infusion process (Rodriguez et al., 2005), and the simulations of the resin flow during liquid composite molding (LCM) process (Nguyen et al., 2015) were performed. These manufacturing routes using thermoset resin are not economically viable for automotive applications, however, due to the long curing time and the difficulty in recycling. On the other hand, thermoplastic polymers have great potential to fabricate structural composites at a low cost by virtue of no extra process time for curing reaction, long storage time, and ease of recycling (Svensson et al., 1998). Because of the high viscosity of thermoplastic matrix, however, it is a big technical challenge to develop cost-effective process technologies by inducing fast impregnation of fiber reinforcement with thermoplastic resin. The issue of long impregnation time is very critical in the automotive industry where the acceptable process cycle time is only several minutes. The resin viscosity can be decreased by

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Abbreviations: PP, polypropylene; LCM, liquid composite molding; CRTM, compression resin transfer molding; TGA, thermogravimetric analysis; TG, thermogravimetric; DTG, differential thermogravimetric; MAPP, maleic anhydride grafted PP; LGF/PP, long glass fiber reinforced polypropylene.

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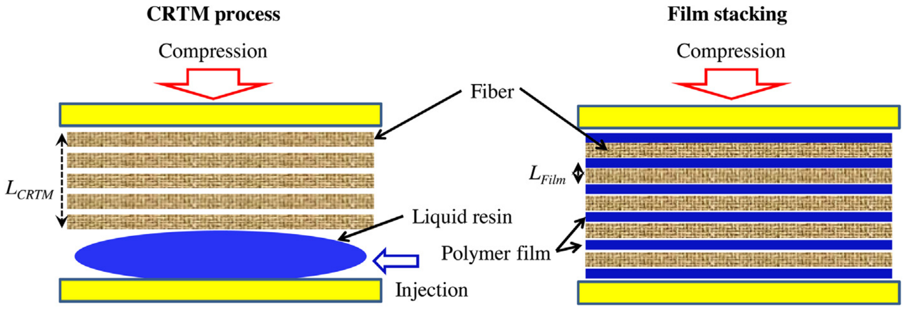

Fig. 1. Comparison of the length of flow path between CRTM process and film stacking process.

increasing the resin temperature. Natural fibers may be degraded at elevated temperature, however, although this behavior is a function of time at temperature (Van de velde and Kikekens, 2002). In general, the thermal decomposition temperature of cellulose fibers is around 200 °C which becomes a maximal processing temperature of natural fiber composites in practice (Lilholt and Lawther, 2000). Therefore, the reduction of resin flow path would be an alternative way to decrease the impregnation time. The common approach for this purpose is the thermo-consolidation of semi-products, such as commingled yarns, powder impregnated fabrics and film stacking, which are premixed forms of solid thermoplastic polymer material and fiber reinforcement (Svensson et al., 1998). For these processing methods, however, matrix-reinforcement premixed materials should be prepared prior to final composites fabrication and the total manufacturing cost is subsequently increased.

Nowadays, LCM processes are regarded as cost effective manufacturing techniques for structural composites because semi-products are not required and dry fiber preform can be directly impregnated by polymer resin in a closed mold. Especially, compression resin transfer molding (CRTM) process is regarded as a promising manufacturing technique to obtain structural parts within a short cycle time for automotive applications. During the CRTM process, the resin is introduced in the open gap between the mold platen and the dry fiber preform preplaced inside a partially open mold cavity. Then, the mold is closed to induce the resin flow in the thickness direction of the fiber preform (Kang and Lee, 1999). Consequently, the resin flow path in the CRTM process becomes shorter than in the conventional resin transfer molding process where the principal resin flow takes place in the planar directions of a thin part. The resin flow path in the CRTM process (L_{CRTM} in Fig. 1) is still longer than that in the thermo-consolidation of film stacking (L_{Film} in Fig. 1), however, as shown in Fig. 1. Hence, the resin temperature in the CRTM process should be higher than that in the film stacking process to decrease the resin viscosity. Meanwhile, this higher resin temperature can induce thermal degradation of natural fiber reinforcement. As a result, an optimal process window should be determined with respect to the thermal degradation of natural fibers and the impregnation quality, as shown in Fig. 2 (Ouagne et al., 2010).

We investigated the effects of process temperature and compression pressure on the impregnation quality during the CRTM process to fabricate continuous flax fiber reinforced thermoplastic composites. The impregnation quality was assessed in terms of void content in the final parts which were evaluated by a density measurement method and an image analysis method. Thermogravimetric analysis (TGA) was performed to characterize the thermal degradation of flax fiber used in this study. The mechanical properties such as flexural modulus and flexural strength were measured by three point bending tests. In particular, we analyzed the inter-relations among the process conditions, the void content and distribution, and the mechanical properties. Finally, we compared the process cycle time and the mechanical properties of the final parts obtained by the current manufacturing techniques with those reported in the literature.

2. Experimental procedure

2.1. Materials

As thermoplastic matrix, we used commercial PP for injection molding, PPC 13442 with a melt flow index of 100 g/10 min (230 °C, 2.16 kg), produced by Total petrochemical. To measure the viscosity of PP, the rheometer (Thermo fisher scientific, HAAKE MARS III) was used. The oscillatory measurement was performed at 10 Hz frequency, while the temperature was increased at the rate of 0.1 °C/min. The density of PP is 0.905 g/cm^3 and the melting point is 165 °C according to the supplier's technical datasheet. We chose Nattex 600 (Dehondt Technologies, France) which is flax 3 × 3 twill weave fabric whose areal density is 600 g/m^2 before drying and is used generally for LCM process such as lay-up, infusion and resin transfer molding. The surface of flax fiber was not treated.

2.2. Manufacturing process

Flax/PP composite plates were fabricated in a rectangular mold with dimensions of 340 × 270 × 2 mm^3. Prior to the manufacturing process, the flax fabrics were dried under vacuum at 0.2 bar and 80 °C for four hours and the PP was dried under vacuum at 80 °C for 40 min.

The schematic of the manufacturing process is illustrated in Fig. 3. If hot resin is in contact with cold fibers or mold wall during the impregnation process, the resin temperature will be decreased instantly and the resin viscosity will increase fast. Therefore, the mold and the flax fiber reinforcement were pre-heated to an elevated temperature above the melting point of the resin. Consequently, we applied an isothermal mold filling condition where the fiber, the resin and the mold were submitted to a same fixed

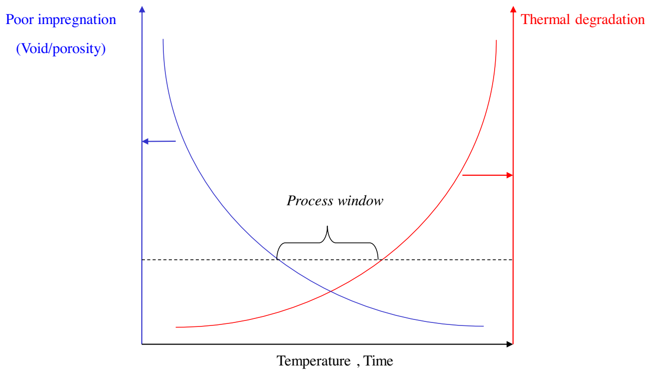

Fig. 2. Presentation of the optimal process window by considering the resin impregnation quality and the thermal degradation of natural fibers.

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