Thermoplastic pultrusion of recycled PET matrix composites

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Abstract. Recently, many industry sectors are investigating safe ways to replace conventional materials by adopting thermoplastic matrix composites. Indeed, the adoption of this class of polymeric matrices enables other post-process operations, such as forming and welding. Moreover, the diffusion and the improvement of thermoplastic matrix composites can promote the usage of recycled polymers, which would dramatically improve the environmental sustainability of the production. The aim of this work is the assessment of the thermoplastic pultrusion of preimpregnated tapes made of glass fibers and recycled polyethylene terephthalate (PET) matrix. A thermoplastic pultrusion line consisting of a heating-forming die and a cooling die has been used to manufacture a rectangular cross-section profile having dimensions of 25 mm in width and 4 mm in thickness. The internal temperature has been measured during the process by using a wire thermocouple. The composite produced has been assessed by interlaminar shear strength testing, and its cross-section has been analyzed by optical microscopy to assess the continuity of the matrix, the internal structure, and the distribution of the fibrous reinforcement.

Introduction

Generally, thermoplastic matrix composites present several relevant advantages in comparison with fiber reinforced thermosets, in terms of mechanical and physical properties, and, due to this reason, they collected the interest of industry sectors requiring high-performance materials. Moreover, they are post-formable and weldable [1,2]. Nevertheless, their high viscosity did not favor their spreading as a matrix for composite. Indeed, high viscosity determines issues in reinforcement-matrix combination and increases the risk of defects and manufacturing costs. Even if recently many researchers are actively working on thermoplastics viscosity reduction [3], a viable solution for the adoption of these matrices is based on the usage of pre-impregnated reinforcement [4]. In this case, the raw materials are generally more expensive, but, on the other hand, they are easier to be processed and the defects related to the matrix-reinforcement combination are excluded since the impregnation occurs off-line.

Moreover, thermoplastic polymers are remarkably interesting from the point of view of sustainability. Indeed, these materials can be remelted and reused several times, even if contamination and degradation could complicate repetitive recycling. It is noteworthy considering that the remelting and reusing strategy is easily applicable in unreinforced polymers, which do not contain reinforcing phases. The presence of fibers excludes the possibility of an easy separation of the phases. Nevertheless, the formability of the composite when heated opens new possibilities of re-forming the components at its end of life. Another option for more sustainable manufacturing is related to the use of recycled thermoplastics as a matrix for composites [5]. In this case, polyethylene terephthalate and polypropylene are the most available materials. Of course, the adoption of recycled polymers involves possible issues in the quality of the composite products due to the larger deviations from the average physical and mechanical properties. Therefore, highly stable and controllable processes are preferable [6].

Pultrusion can respond to this stability requirement. Indeed, it is a continuous process for manufacturing constant cross-section elements in composite material [7]. Pultrusion has been designed and developed considering thermoset matrices, nevertheless, recently, a growing number of studies have focused on the revision of the pultrusion line for the manufacturing of fiber reinforced thermoplastics. The most used approaches are based on the usage of low-viscosity thermoplastics or the pultrusion of pre-impregnated reinforcement [8].

The aim of this work is the study of the pultrusion of preimpregnated tapes in glass/recycled polyethylene terephthalate. The composite profile cross-section has been analyzed by optical microscopy to assess the continuity of the matrix and the reinforcement distribution, and interlaminar shear strength testing has been carried out to evaluate its mechanical performance.

Materials and Methods

Preconsolidated tapes in glass fiber reinforced thermoplastic provided by CompTape BV (Delft, Netherlands) have been adopted for the present investigation. The reinforcement consists of glass fiber linear rovings having a tex number of 2400, while the matrix is composed of 95% recycled polyethylene terephthalate (PET) and 5% other polymeric components improving the compatibility with the fibrous reinforcement. The tapes have a width of 6 mm and a thickness of about 0.65 mm, with a linear mass of 4.36 ± 0.35 g/m and they appear of brownish color (Fig. 1).

Figure 1. Preconsolidated tapes in glass fiber reinforced recycled PET.

Aiming to achieve a composite profile with a volume fraction of reinforcement of about 30%, 31 tapes have been pultruded using a laboratory-scale thermoplastic pultrusion line used to produce rectangular cross-section profiles having a width of 25 mm and thickness of 4 mm. It is worth mentioning that in the case of thermoplastic pultrusion, the fiber volume fraction is sensibly lower than the values achieved using thermoset resins. The value of 30% is comparable with the one previously achieved by other authors [9–12].

The pultrusion line adopted is moved by a continuous counter-rotating caterpillar system, moving the processed materials along with two dies, mechanically connected with each other, as represented in Fig. 2.

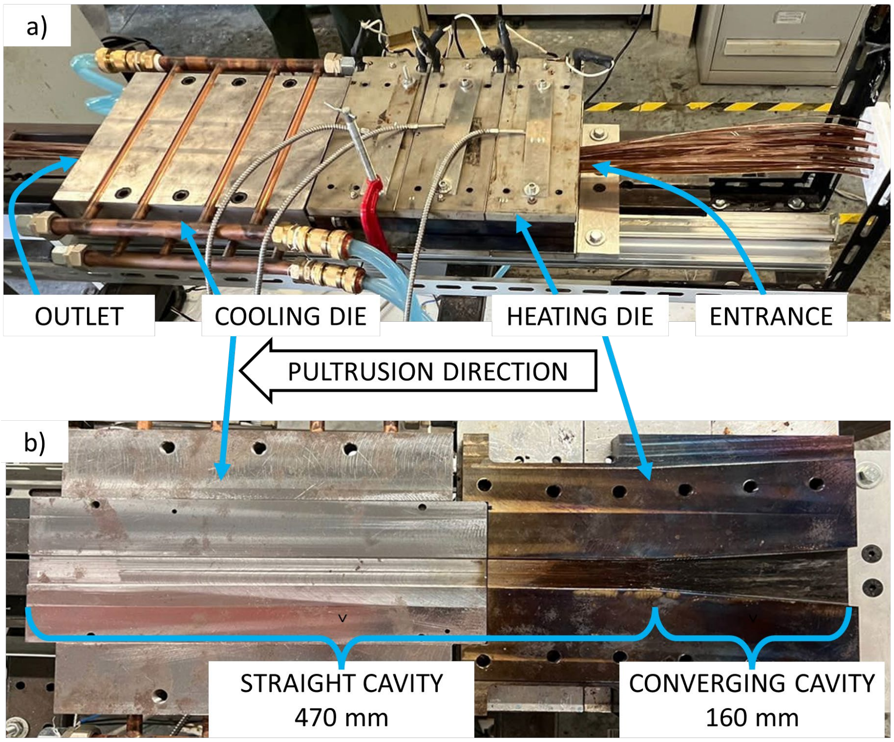

Figure 2. a) Thermoplastic pultrusion dies; b) Cavities of the pultrusion dies.

The first die is responsible for the tapes' compaction and composite forming. It is heated by electrical platens governed by PID controllers. The temperature set point of 240°C has been chosen to overcome the melting point of the adopted matrix. The heating die is 280 mm long and, as visible in Fig. 2b, the cavity is convergent in the earliest 160 mm, with a taper angle of 4.35°, then it presents a constant cross-section having dimensions of the final profile. The increase in temperature softens and melts the thermoplastic, while the geometrical convergence compacts the tapes and promotes the matrix transversal flow. The cooling die cavity is 350 mm long and it has a constant section geometry. A chiller is responsible for the absorption of the thermal energy via the cooling fluid (a mixture of water and glycol) flowing through the copper pipes visible in Fig. 2. The temperature setpoint for the cooling fluid is 13°C. The advancing velocity of 200 mm/min is controlled by the caterpillar pulling system.

During the process, in steady state conditions, the internal temperature was measured using the traveling thermocouple method [13,14]. The thermocouple has been fixed to a central tape in order to evaluate the core temperature. Being the pultrusion line equipped with a load cell, the process has been considered in steady state conditions based on the stabilization of the pulling force, which is highly variable during the initial transient.

The cross-section of the produced composite profile has been analyzed by optical microscopy using a stereomicroscope Motic SMZ171 and an optical microscope Nikon Eclipse L150. The mechanical properties have been evaluated by interlaminar shear strength test performed in the short beam shear mode according to the ASTM D2344. Three samples having a width of 18 mm, and thickness of 4 mm were collected from the produced profile and tested using a span length of 20 mm.

Results and Discussion

At a visual inspection, the produced profile appeared well compacted and with constant cross-section, even if the external surface presented marked irregularities, as visible in Fig. 3.

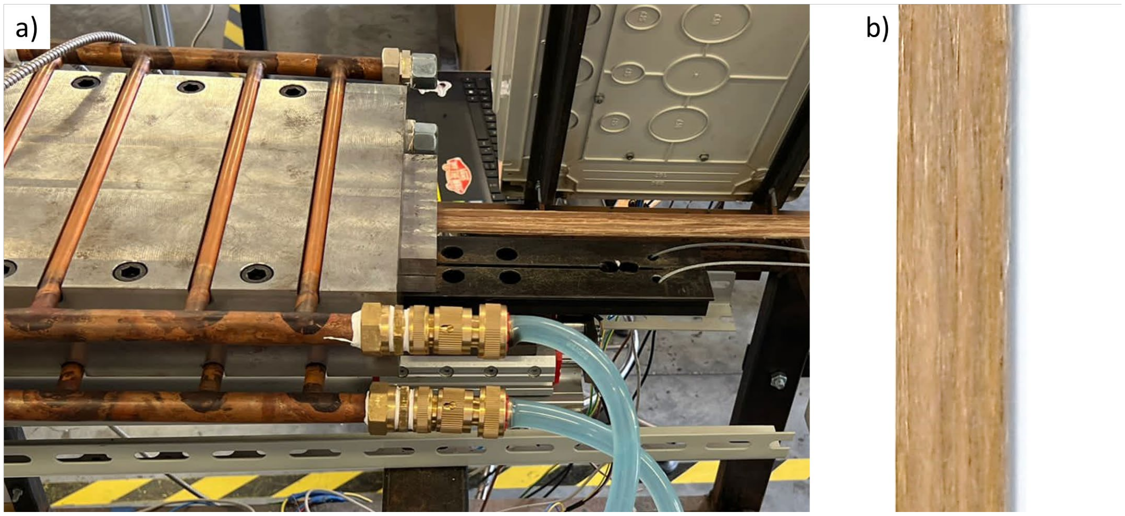

Figure 3. a) Pultruded composite at the cooling die outlet; b) Surface of the pultruded composite.

The surface unevenness has been already detected in previous studies on thermoplastic pultrusion with different materials [15,16]. The microscope analysis of the internal structure is necessary to assess the quality of the composite.

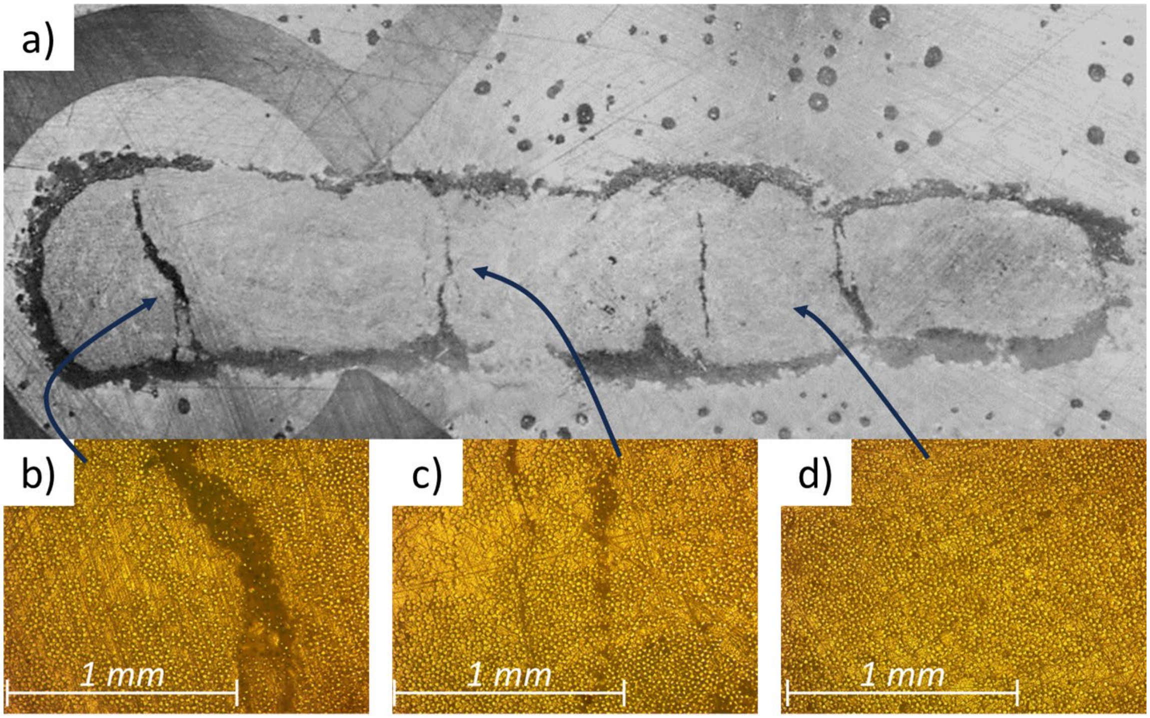

Figure 4. a) Micrographic view of the profile cross-section acquired by stereomicroscope; b),c), and d) cross-section details acquired by optical microscope.

Fig. 4 (a) reports the overall view of the cross-section of the produced composite acquired by stereomicroscope. On average, the profile presents good compaction, nevertheless, there are several marked transversal cracks crossing the thickness of the profile. The cracks can be analyzed in higher detail by the microscope images Fig 4 (b-c). Cracks do not appear at the boundary of the tapes, but, on the contrary, they cross the tapes. This suggests that the compaction phase in the heating die has been successfully completed, and the cracks formed and propagated successively. The cooling phase could be responsible for an excessive heat flow provoking fast internal contractions of the materials and the consequent cracking. It is worth mentioning that in the zones not affected by cracks, the matrix generally appears continuous, and the reinforcement is homogeneously distributed Fig. 4 (d). The unevenness of the external surface is evident in the micrography of the whole section. In thicker zones, a concentration of tapes is visible, probably due to the uncontrolled mobility of the tapes while crossing the converging cavity of the heating die. A stiffer constraint of the tapes upstream of the entrance in the heating dies could provide a preliminary arrangement that could mitigate this effect.

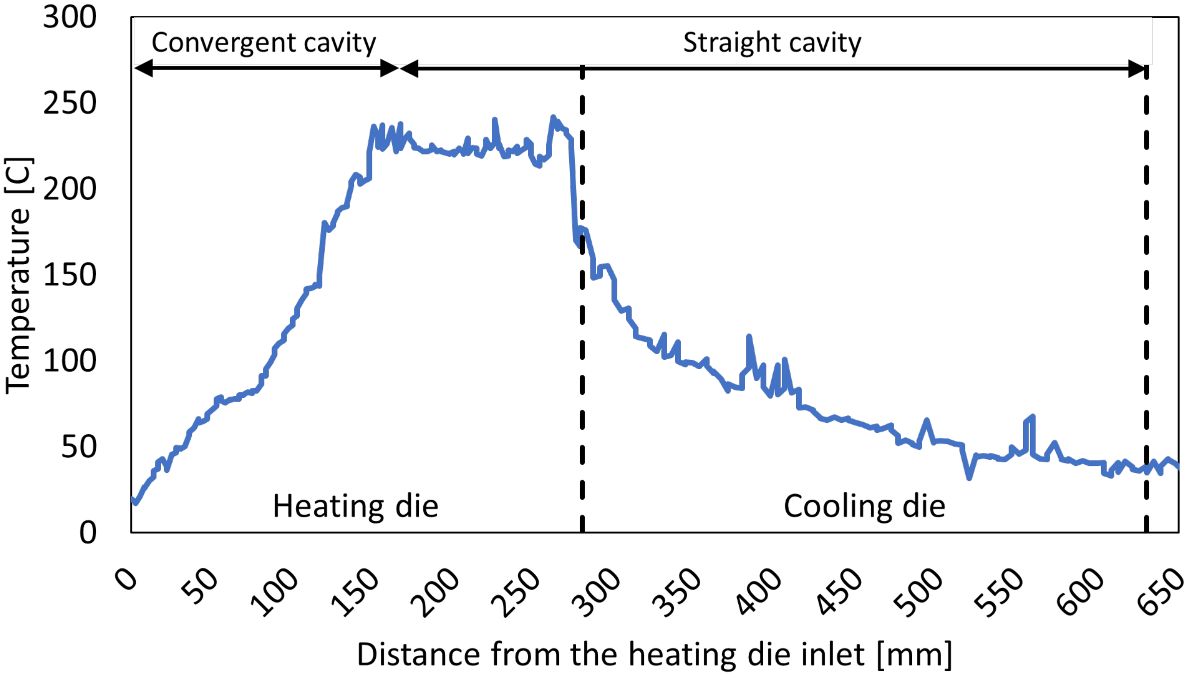

Figure 5. Internal temperature during pultrusion.

The temperature data acquired using the traveling thermocouple method are reported in Fig. 5. The tapes pass through the heating die inlet at environmental temperature, since, in this experiment, no preheating systems have been used. The temperature increases along with the convergent zone of the cavity. Considering the taper angle and the dimensions of the tapes, at 120 mm from the die inlet, the compaction of the material starts, and the sensitive bulb detects a sharp increase in temperature. The temperature is sufficient to start the matrix softening and melting. The presence of a viscous liquid phase and the tapering shape of the cavity provoke an increase in pressure and transversal flow of thermoplastic, which, in turn, generates noise in the thermal signal detected by the thermocouple. The pressure increase in tapered pultrusion cavities has been predicted and demonstrated in several literature works [17–20]. The temperature along with the straight portion of the heating die oscillates around 235°C. The decrease in temperature starts in the last millimeters of the heating die and continues along with the entire cooling die. At the crossover between the two dies, the most relevant temperature drop can be observed. Such gradient could be responsible for the formation and propagation of the cracks, nevertheless, further repetitions of the thermal measurement and deeper analyses are necessary to confirm this hypothesis.

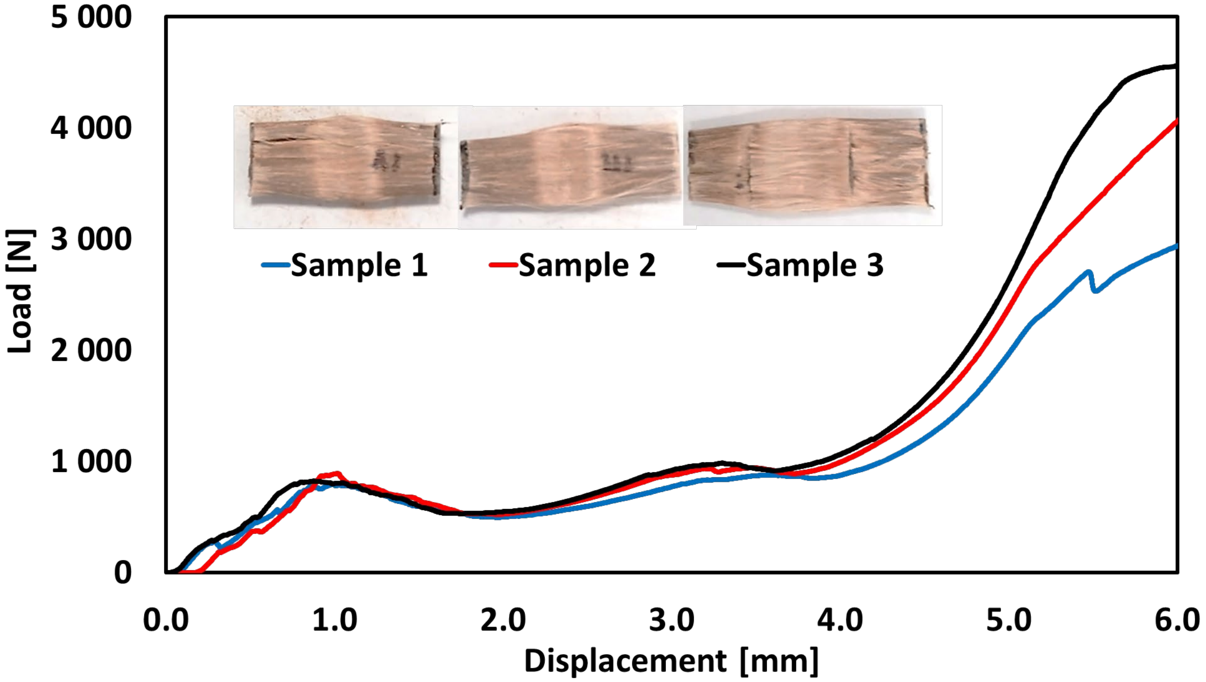

Figure 6. Interlaminar shear strength of the pultruded profiles.

Fig. 6 depicts the results of the interlaminar shear strength test. The three curves present initially a linear behavior until a displacement of 1 mm. After that, the samples exhibited large plastic deformations. The maximum load observed is about 4550 N. A longitudinal crack, visible in the failed sample 1, formed during testing at a displacement of about 5.5. According to the ASTM D2344 standard, the maximum interlaminar shear strength of the pultruded composite has a value of 45 MPa.

Conclusions

Tapes in fiber reinforced recycled PET have been pultruded using a laboratory-scale thermoplastic pultrusion line. The composite profile produced appeared continuous and well-compacted, but the external surface appeared irregular at a visual inspection, evidencing the necessity for a stiffer pre-tensioning of the tapes. The microscope observation of the cross-section revealed a good consolidation of the tapes but the presence of through-the-thickness cracks. This undesired effect could be mitigated by setting a slower cooling process, which will be investigated in future research. Despite the defects detected, the samples collected from the pultruded profile exhibited a relevant value of interlaminar shear strength.

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