# Investigation on the pultrusion of thermoplastic preimpregnated polypropylene-glass tapes

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Keywords: Pultrusion, Thermoplastic, Polypropylene, Prepregs, Tapes

Abstract. Pultrusion processes have been widely used and developed to produce composite profiles in fiber-reinforced thermoset. Recently industry and research community are investigating the adoption of thermoplastic polymers in the production of pultruded components exhibiting structural stability at higher temperatures and improved sustainability. Aiming to study the thermoplastic pultrusion process and how the process parameters settings affect the composite profiles produced, a laboratory-scale pultrusion line has been designed and produced. The pultrusion die is fed by polypropylene pre-impregnated tapes reinforced by unidirectional continuous glass fibers. It consists of a tapered converging cavity heated by electrical plates governed by PID controllers and a straight cooling die. This work aims to study the feasibility of the process, the consolidation of the tapes, and the interactions between the processed materials and the cavity walls by using embedded traveling thermocouples and load cells. The experimental test presented in this paper highlights that the preimpregnated tapes processed have been wellconsolidated with the parameters adopted and the pultruded profile presents good quality at a visual inspection of the external surfaces and cross-section, indicating a good melting and hardening of the polypropylene matrix. The interactions between the processed materials and the die walls are evaluated by means of a cross-analysis of thermal and load data.

# Introduction

Pultrusion is a widely consolidated process for the manufacturing of constant cross-section profiles in continuous fibers reinforced thermoset [1]. The success of this process is related to several factors: i) the high tensile properties due to the orientation and the volume fraction of the reinforcement; ii) the continuous nature of the process, which in turn guarantee high productivity, repeatability, and automation; iii) the low energy and human intervention required; iv) low or null waste of raw materials [2,3]. Due to their properties, pultruded profiles are widely employed in many industrial or civil sectors [4,5]. Traditionally, thermoset systems have been preferred for their low viscosity and their easy manufacturability. Nevertheless, generally, thermosets presents several drawbacks if compared to thermoplastics, namely lower in-service temperature, lower mechanical properties, production of dangerous volatile compounds due to the polymerization during the manufacturing or process related defects and distortions [6,7]. Moreover, the thermoplastic polymers can be remelted and re-used, promoting a more sustainable production [8].

Of course, the adoption of a different matrix system requires a different pultrusion line, equipped with features designed to process thermoplastics. The major difference stands in the

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presence of a heating die and a successive cooling die. The former is responsible for the matrix melting and flowing; the latter give place to the matrix cooling and hardening [9]. In the thermoplastic pultrusion the reinforcement impregnation can be done either online or offline. In the first case, the polymeric system is heated up, in order to minimize the viscosity and injected through the advancing fibers within the heating die. The offline impregnation is based on the usage of preimpregnated tissues or tapes [10]. The pultrusion of preimpregnated tapes guarantees a better distribution of the matrix and the absence of dry zones, but it requires longer crossing time to ensure the tapes consolidation in a unique composite profile, and therefore, a lower processing speed. In the case of pultrusion of preimpregnated tapes, the heating die presents a highly sloped tapered chamber, converging to the final cross-section shape. Such slope constrains the advancing tapes and determines an increase in the pressure, which in turn confers the shape of the profile, promotes the transversal flow of the melted matrix and consolidates the tapes. The cooling die presents a straight cavity with the shape of the final cross-section which holds the materials in their positions while the matrix is hardening [11,12].

Therefore, the main factors affecting the thermoplastic pultrusion are the heating ramp, determining the thermoplastic softening and melting, the transversal compression, determining the transversal flow of the polymer and the compaction of the tapes, and the cooling ramp, hardening the thermoplastic. The main challenges in the monitoring of the process and the measurement of the variables of interest in pultrusion are related to the fact that all the transitions and shaping phases are included into a closed non-transparent die. The method of the traveling thermocouple is the most commonly adopted to collect the internal temperature data [7]. It is carried out by connecting a thermocouple to the advancing materials at the die entrance and measure the temperature during the time necessary to cross the pultrusion die. The data collected in the time domain can be transferred to the position domain since the velocity of the process is constant and known and it corresponds to the pulling speed. The evaluation of the pressure can be achieved by means of pressure sensors [13], nevertheless these sensors cannot be fixed easily in a pultrusion die without altering its geometry. Moreover, the measurement achieved is representative of the localized pressure at the sensor location, while the pressure in a tapered cavity exhibits a marked variability along with the die [14]. More pieces of information on the advancing materials transitions and on their interaction with the die cavity, can be indirectly inferred from the measurement of the pulling force, since it strictly depends on the nature of the interaction between processed materials and the die cavity walls [15].

This study aims to analyze the pultrusion of tapes in glass fiber reinforced polypropylene pultruded in a laboratory scale line. The internal temperature and the pulling force data have been analyzed to detect, describe, and quantify the main behaviors occurring along with the pultrusion die.

### **Materials and Methods**

The pultrusion line adopted in the present experimental activity consists of a 300 mm heating die followed by a 300 mm cooling die. The line adopted is illustrated in Fig. 1a.

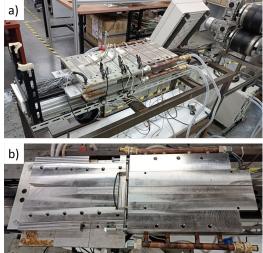


Fig. 1. a) Thermoplastic pultrusion setup; b) view of the disassembled lower sides of the heating die and cooling die.

In the thermoplastic pultrusion setup shown in Fig. 1a, the advancing direction is oriented from the left towards the right. The preimpregnated tapes approach the pultrusion line presents from the heating die side, where three couple of electric platens are provide heating energy to the system. The platens' powers are governed by three PID controllers, receiving the feedback signals from three sensors fixed on each of the upper side platens. The cooling die is mechanically fixed downstream the heating die. The cooling system is based on water circulation through a copper pipes grid embedded within milled cavities. The water cooling and circulation is governed by an industrial chiller, with temperature setpoint of 13°C. Fig. 1b shows the internal cavities of the disassembled lower sides. The heating die presents a tapered converging zone to favor the tapes entrance and to compact them, followed by a straight cavity with constant cross-section. The tapered zone has a length of 160 mm. The cavity of the cooling die is straight with constant rectangular cross-section 25 mm wide and 4 mm thick.

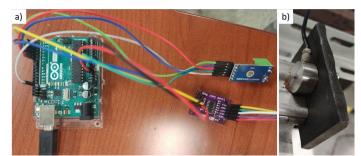
This setup has been used to pultrude preimpregnated tapes provided by CompTape BV (Delft, Netherlands) in glass roving reinforced polypropylene. Each tape has dimensions of 6.35 mm in width and 0.6 mm in thickness and contains the 60% in volume of reinforcement. The tapes have been arranged in 6 layers of 4 tapes per layer. With this arrangement, 24 preimpregnated tapes have been pultruded, to achieve a profile with a fiber volume fraction of 55%. Considering the polypropylene matrix, a temperature of 200°C has been set for the platens. The dies have been mounted on supports free to slide on a longitudinal guide. At the limit of the guide a compressive button load cell is fixed to measure the pulling force (Fig. 2b). The pulling system, consisting of counter-rotating caterpillars, moves the tapes at a velocity of 170 mm/min.

The internal temperature of the advancing tapes within the pultrusion dies has been measured using the traveling thermocouple method: Once the process is stationary (the pulling force are stable), the bulb of a wire thermocouple is connected to one of the tapes, in such a way to have the bulb between the third and the fourth layer and at the middle of the pultruded width. The aim is to measure the temperature of a point as close as possible to the barycenter of the cross-section. The wire must be as thin as possible to have a low invasive measure. In this case, the two wires for the two conductor alloys of the k-type thermocouple (chromel-alumel) have diameter of 0.2 mm. Once the bulb crosses the entire length of the two dies, the wires have been cut.

During pultrusion, the load cell measures the overall force resisting to the pulling. Such force is related to the interactions arising at the contact between dies' walls and pultruded materials. The

local interaction between die and advancing materials has been evaluated cutting the tapes at the heating die inlet and analyzing the unloading curve, as described in [16].

The collection of the data acquired by the load cell and the thermocouple has been managed using an Arduino board equipped with signals' amplifiers (Fig 2a) and connected to a computer.



*Fig. 2. a)* Data collection management board equipped with signals' amplifiers; b) compressive button load cell fixed at the guide limit.

### **Results and Discussion**

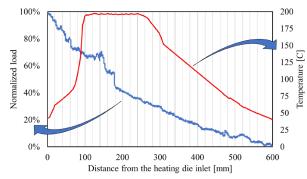
The described setup for thermoplastic tapes pultrusion has produced well compacted and continuous pultruded composites.



*Fig. 3. a)* Pultrusion setup while processing glass-polypropylene tapes; b) obtained pultruded profile.

Fig. 3a shows the pultrusion dies during the process. It is worth noting the tapes approaching to the heating die inlet. Each tape is separated with each other, and all of them are converging driven by the shape of the heating die cavity. At the cooling die outlet, the pultruded profile appears continuous and compacted, as demonstrated in the close up in Fig. 3b.

The internal temperature of the pultruded materials acquired by the traveling thermocouple and the load discharge curve measured by the load cell is reported in Fig. 4.



*Fig. 4. Total load discharge curve and internal temperature of the advancing materials along with the heating and the cooling dies.* 

The tapes reach the heating die entrance with a temperature slightly superior to the environmental conditions, due to the diffusion in the air of the heat from the heating die. The temperature increase is slight along with the initial 80 mm. Indeed, at the heating die entrance, the cavity is wider, and the tapes are not in direct contact with each other and with the die. After this point the compaction and compression of the tapes driven by the converging walls determines a sharp increase in the temperature, which reaches approximately 195°C. This temperature is kept constant until 250 mm of distance from the heating die inlet. Even if the cooling die starts from 300 mm from the inlet, the heat dissipation of the cooling fluid affects also the temperature of the final portion of the heating die. The transition effects are visible in a continuous decrease in the slope of the temperature curve until the end of the heating die. Along with the last 300 mm, the heat dissipation gives place to a linear decrease in the temperature.

The polypropylene transitions are driven by the temperature and affect the interaction with the dies' walls. The earliest 150 mm are characterized by an irregular decrease of the total load with large fluctuations. Indeed, in this zone, the tapes are not compressed against the die walls. The contact is furthermore irregular if compared to the stationary process because of the absence of the pretension due to the cut of the tapes. The tape compaction can be recognized by the drop in load between 140 mm and 160 mm of distance from the die inlet. The melting temperature is reached and overcome starting from 100 mm from the heating die inlet. The effects of melting and rheological evolution of polypropylene can be observed clearly between 160 mm and 180 mm by a marked drop in the resisting load. Drops in the cumulated load indicate peaks in the local force. Indeed the thermoplastic passes from a solid state to a gel state, characterized by an adhesive local interaction typical of the high viscosity fluids, and finally reaches the melt state. The low viscosity reached by the polypropylene after 180 mm from the die inlet gives place to a lubricant effect promoting the sliding of the materials. The main resistance between 180 mm and 350 mm from the heating die inlet is related to the viscous drag. The linear decrease in the total load curve indicates a constant resistant force along with this zone. The polypropylene solidifies again in the last 250 mm from the inlet. The transition to a solid compact composite determines a solid friction interaction between the die walls and the pultruded profile surface. This interaction can be recognized by the fluctuations visible from 350 mm from the heating die inlet and becoming larger in the last 150 mm of the die.

### Summary

This work presents an experimental setup for the pultrusion of glass/polypropylene preimpregnated tapes. The measurement of the internal temperature evidenced that the polypropylene melts in the heating die, where it is compacted and shaped. The heat dissipation in the cooling die gives place to a fast solidification of the matrix. The compaction of the tapes gives place to the most prominent local loads peaks, while the melted polypropylene acts as a lubricant, promoting a regular and smooth sliding of the advancing taps.

# Acknowledgement

Authors gratefully acknowledge CompTape BV (Delft, Netherlands) for its precious support in the choice of materials and for the tapes supply.

# References

[1] P. Boisse, R. Akkerman, P. Carlone, L. Kärger, S. V. Lomov, J.A. Sherwood, Advances in composite forming through 25 years of ESAFORM, Int. J. Mater. Form. 15 (2022). https://doi.org/10.1007/s12289-022-01682-8

[2] L. Nickels, The future of pultrusion, Reinf. Plast. 63 (2019) 132-135. https://doi.org/10.1016/j.repl.2019.01.003 Materials Research Proceedings 28 (2023) 393-398

[3] A.M. Fairuz, S.M. Sapuan, E.S. Zainudin, C.N.A. Jaafar, Polymer composite manufacturing using a pultrusion process: A review, Am. J. Appl. Sci. 11 (2014) 1798-1810. https://doi.org/10.3844/ajassp.2014.1798.1810

[4] A. Vedernikov, A. Safonov, F. Tucci, P. Carlone, I. Akhatov, Pultruded materials and structures: A review, J. Compos. Mater. 54 (2020) 4081-4117. https://doi.org/10.1177/0021998320922894

[5] P. Zhou, C. Li, Y. Bai, S. Dong, G. Xian, A. Vedernikov, I. Akhatov, A. Safonov, Q. Yue, Durability study on the interlaminar shear behavior of glass-fibre reinforced polypropylene (GFRPP) bars for marine applications, Constr. Build. Mater. 349 (2022) 128694. https://doi.org/10.1016/j.conbuildmat.2022.128694

[6] A. Vedernikov, A. Safonov, F. Tucci, P. Carlone, I. Akhatov, Modeling Spring-In of L-Shaped Structural Profiles Pultruded at Different Pulling Speeds, Polym. 13 (2021). https://doi.org/10.3390/polym13162748

[7] A. Vedernikov, F. Tucci, P. Carlone, S. Gusev, S. Konev, D. Firsov, I. Akhatov, A. Safonov, Effects of pulling speed on structural performance of L-shaped pultruded profiles, Compos. Struct. 255 (2021) 112967. https://doi.org/10.1016/j.compstruct.2020.112967

[8] R. Bernatas, S. Dagreou, A. Despax-Ferreres, A. Barasinski, Recycling of fiber reinforced composites with a focus on thermoplastic composites, Clean. Eng. Technol. 5 (2021) 100272. https://doi.org/10.1016/j.clet.2021.100272

[9] K. Minchenkov, A. Vedernikov, Y. Kuzminova, S. Gusev, A. Sulimov, A. Gulyaev, A. Kreslavskaya, I. Prosyanoy, G. Xian, I. Akhatov, A. Safonov, Effects of the quality of preconsolidated materials on the mechanical properties and morphology of thermoplastic pultruded flat laminates, Compos. Commun. 35 (2022) 101281. https://doi.org/10.1016/j.coco.2022.101281 [10] P. Simacek, S.G. Advani, Simulating tape resin infiltration during thermoset pultrusion Compos. Part Appl. Sci. Manuf. 72 (2015) process. А 115-126. https://doi.org/10.1016/j.compositesa.2015.01.020

[11] K. Minchenkov, A. Vedernikov, A. Safonov, I. Akhatov, Thermoplastic pultrusion: A review, Polym. 13 (2021) 1-36. https://doi.org/10.3390/polym13020180

[12] A. Vedernikov, K. Minchenkov, S. Gusev, A. Sulimov, P. Zhou, C. Li, G. Xian, I. Akhatov, A. Safonov, Effects of the Pre-Consolidated Materials Manufacturing Method on the Mechanical Properties of Pultruded Thermoplastic Composites, Polym. 14 (2022) 1-15. https://doi.org/10.3390/polym14112246

[13] F. Tucci, R. Bezerra, F. Rubino, P. Carlone, Multiphase flow simulation in injection pultrusion with variable properties, Mater. Manuf. Process. 35 (2020) 152-162. https://doi.org/10.1080/10426914.2020.1711928

[14] S.S. Rahatekar, J.A. Roux, Numerical simulation of pressure variation and resin flow in injection pultrusion, J. Compos. Mater. 37 (2003) 1067-1082. https://doi.org/10.1177/0021998303037012005

[15] A.A. Safonov, P. Carlone, I. Akhatov, Mathematical simulation of pultrusion processes: A review, Compos. Struct. 184 (2018) 153-177. https://doi.org/10.1016/j.compstruct.2017.09.093

[16] F. Tucci, D. Larrea-Wachtendorff, G. Ferrari, P. Carlone, Pulling force analysis in injection pultrusion of glass/epoxy composites, Mater. Manuf. Process. 00 (2022) 1-12. https://doi.org/10.1080/10426914.2022.2049296