MODELING AND EXPERIMENTS IN THERMOPLASTIC COMPOSITE PULTRUSION

Khongor Jaamiyana1 and Uday Vaidya2,*
Department of Materials Science and Engineering
The University of Alabama at Birmingham, Birmingham, AL 35294-4461
1Master’s Student, 2Professor and Director, Composites Center

Abstract

Thermoplastic pultrusion is an emerging process and has number of applications as structural elements for trucks, buses and transportation components. The present work focuses on using a modeling approach for thermoplastic pultrusion. Computer Aided Design, Finite Element Analysis, and Computational Fluid Dynamic software as well as analytical methods were used to model the pultrusion process. A pultrusion die was designed based on the amount of time required for E-glass/polypropylene hot-melt impregnated tapes to fully melt and consolidate in the die. A fluid simulation study was conducted to characterize how the processing parameter of die temperature and pulling speed affects the amount of force required to pull the material through the die. The results show that the pulling force increases as the pull speed increases. Both the fluid model and the experimental data show excellent correlation. The developed approaches can be extended to a variety of thermoplastic composites.

Introduction

Pultrusion is a type of processing technique to make constant cross-section composites of almost any length. The materials are typically continuous fiber reinforced polymers that are pulled through a heated forming die where the material being processed is molded into the shape of the die cavity. There are wide range of structural applications that pultrusion can be used for such as transportation, construction, and marine structures just to name a few (Chotard, 1998, (Gilby, 1998). Both thermoset and thermoplastic polymers can be pultruded but their processing techniques differ from one another.

In thermoset pultrusion, virgin fibers are guided through a resin bath where the fibers get impregnated with the thermoset resin. After the fibers are fully wet, the saturated fibers are pulled through a heated forming die and the cross-linking takes place. By allowing the resin to cure inside the die, the composite takes form of the die cross-section. After the matrix has cured, the material is cut at the desired length (Gadam, 2000). Thermoset pultrusion is also a much more mature process that has been well established and is readily available commercially.
Thermoplastic pultrusion in general is relatively new process and there is limited work that has been done. The process is complicated by the high resin viscosity of thermoplastics that are usually two or three orders of magnitude higher than thermoset resins (Akovali, 2001). This results in only partial fiber wet out when pulled through a resin bath. Thus the fibers are pre-impregnated (prepreg) with the matrix before being pultruded. The prepreg fibers may come in a form of commingled tows, hot-melt impregnated fibers, or powered impregnated tows. In place of a resin bath, the prepreg materials are pulled through a pre-heating section (Hudson, 2009) (Carlsson, 1998, Sala, 1997). The pre-heater is used to shorten the processing time by increasing the temperature of the composite almost to its melting point. After the pre-heater the material enters the heated forming die where the matrix melts and consolidation takes place. Unlike thermoset pultrusion, the thermoplastic matrix is cooled while still inside the die to maintain die shape. It is not uncommon to have two or more separate dies where one is used to melt and the other to cool the composite (Hudson, 2009).

The high resin viscosity, non-Newtonian resin flow, resin solidification and melting, and possibility of more than one die make thermoplastics much more difficult to process. But thermoplastic matrix offer higher toughness and damage tolerance than their thermoset counter parts. They are also recyclable, can be locally welded, and rapidly produced when compared to thermoset resins that make them more desirable for various applications such as construction, automotive, marine structures, and military equipment (Mallick, 1993) (Hudson, 2009) (Sala, 1997) (Miller, 1998) (Haffner, 1998) (Ma, 1990) (Nejhad, 1997).

Technical Approach

Generate analytical heat transfer model

For the present study the material form used was hot melt impregnated E-Glass/Polypropylene (E-glass/PP) 0.5 inch tapes with 59.3% fiber weight fraction. The die profile was chosen to process rectangular bars that are 25.4 wide and 4.3m thick and the length d was designed based on the thermal conductive properties of the pultruded material. The length of the die was determined based on the amount of time required to melt the pultruding material through its entire thickness. It was assumed that once the entire thickness has reached beyond its melt temperature the material has fully consolidated.

With this approach, the pultrusion process was simplified as a steady state heat transfer problem where the thermoplastic composite was model as plane wall of thickness \( L \) in contact with the constant die surface temperature \( T_s \). Since the width of the pultrusion profile was much greater than the thickness, the heat transfer along the width of the profile was ignored while axial heat transfer was thought to be negligible, making the model into a one-dimensional analysis. Pantaleao et al. concluded that at speeds around 0.0015 m/s (similar speeds achieved in this research) the anisotropic conduction rates can be neglected as his model showed very little difference when axial conduction through the fibers was considered versus when axial conduction was not considered (Pantaleao, 2002). Figure 1 illustrates this approach.
It was assumed that the pultruded material was entering the die at a uniform temperature of \( T_i \) and since it is heated from the top and bottom symmetrically, only one half the thickness was considered. As the material enters the die, the top surface is instantly brought up to the die temperature of \( T_s \), which is at a higher temperature than the material melting point \( T_m \).

The die length \( d \) was determined by first calculating the amount of time it took the surface temperature \( T_s \) to transfer through the material thickness and melt the center by using (Equation 1) (Schneider, 1957):

\[
\frac{T_x - T_s}{T_i - T_s} = \sum_{n=1}^{4} \left[ C_n \exp\left( -\zeta_n^2 \cdot Fo \right) \cdot \cos\left( \zeta_n \cdot L \right) \right] \tag{Equation 1}
\]

where \( T_x \) is the temperature at the location of interest and the coefficient \( C_n \) is expressed in (Equation 2),

\[
C_n = \frac{4 \sin(\zeta_n)}{2 \cdot \zeta_n + \sin(2 \zeta_n)} \tag{Equation 2}
\]

the eigenvalues \( \zeta \) are positive roots of the transcendental equation, shown in Equation 3.

\[
\zeta_n \cdot \tan(\zeta_n) = Bi \tag{Equation 3}
\]

and Biot number \( Bi \), which relates the material’s thermal convection \( h \) and conduction \( k \) terms shown in Equation 4.

\[
Bi = \frac{h \cdot L}{k} \tag{Equation 4}
\]

The Fourier number \( Fo \) in (Equation 1 was solved using Equation 5.

\[
Fo = \frac{\alpha \cdot t}{L^2} \tag{Equation 5}
\]

where \( \alpha \) there is the thermal diffusivity,
where the material’s thermal conductivity $k$, density $\rho$, and the heat capacity $c_p$ are taken into account. These thermal properties for the composite are calculated using mass fraction $W_f$ and volume fraction $V_f$ methods shown below (Roux, 1998) (Pantaleao, 2002). The values for the parameter are gathered and are tabulated in Table 1 for E-glass/polypropylene (Tadmor, 2006).

$$k := \frac{\frac{1}{W_f} \left( \frac{k_g}{1 - W_f} \right) + \frac{k_{pp}}{1 - W_f}}{\frac{k_g}{1 - W_f} + \frac{k_{pp}}{1 - W_f}}$$  \hspace{1cm} \text{(Equation 7)}$$

$$\rho := \rho_f V_f + \rho_r (1 - V_f)$$  \hspace{1cm} \text{(Equation 8)}$$

$$c_p := c_{p_g} V_f + c_{p_{pp}} (1 - V_f)$$  \hspace{1cm} \text{(Equation 9)}$$

Once the time $t$ is solved from Equation 1, the length $d$ was be calculated by

$$d = t \cdot V$$  \hspace{1cm} \text{(Equation 10)}$$

where $V$ is the operating pull speed of the pultruder. This length is the minimum length required for the die to melt the pultruding material to melt through its entire thickness at the given speed.

<table>
<thead>
<tr>
<th>Material</th>
<th>Thermal Conductivity $k$</th>
<th>Density $\rho$</th>
<th>Heat Capacity $c_p$</th>
<th>Melt Temperature $T_m$</th>
<th>Weight Fraction</th>
</tr>
</thead>
<tbody>
<tr>
<td>Polypropylene</td>
<td>$0.2 \frac{W}{mK}$</td>
<td>$900 \frac{kg}{m^3}$</td>
<td>$1950 \frac{J}{kg K}$</td>
<td>$167^\circ C$</td>
<td>0.407</td>
</tr>
<tr>
<td>E-Glass</td>
<td>$1.0 \frac{W}{mK}$</td>
<td>$2540 \frac{kg}{m^3}$</td>
<td>$840 \frac{J}{kg K}$</td>
<td>-</td>
<td>0.593</td>
</tr>
<tr>
<td>E-Glass/PP</td>
<td>$0.381 \frac{W}{mK}$</td>
<td>$1458 \frac{kg}{m^3}$</td>
<td>$1572 \frac{J}{kg K}$</td>
<td>$167^\circ C$</td>
<td>1</td>
</tr>
</tbody>
</table>

For the current study, it was found that die length needed to be 14.4 cm of constant surface temperature of 185$^\circ C$ in order for the center of the material to reach its melt temperature. This length was considered at the highest pull speed capable by the machine of $0.0144 \frac{m}{s}$. If the operating speed was slowed, then the required length to melt the material through would likewise decrease since the material will be in the die for longer time. By considering the maximum pull speed available gives it a certain safety factor since the normal operating speed is not at maximum speed.

Once the material has fully melted and consolidated, it is cooled inside the die to maintain the die shape and initiate crystallization. A reverse calculation was made to find how long the die needs to be in order for the center of the material to cool from melt temperature to reach the polymer crystallization temperature $T_c$ of a 100$^\circ C$. Using the same maximum speed, the required length was found to be 12.9 cm. The final length of the pultrusion die was 40.64 cm.
The overall die length increased slightly, to properly secure the die onto the fixed frame. The die is clamped into 12.7 mm pin that is attached to a steel frame and is allowed to pivot and is simply supported on another steel frame to help support the weight.

**Design of Pultrusion Die**

The die geometry was generated using CAD software Creo Element/Pro. The die was made of two halves bolted together by eighteen 5/16 inch (7.94 mm) socket head cap screw bolts. It features a 5° taper for 95.25 mm into the die to allow sufficient heat transfer. The rationale behind this was to efficiently transfer heat through the material. A larger taper angle would create big air gaps between the tape layers, creating a large resistance to transfer heat through the gaps. Past the tapered section, the die consists of constant cross-section.

The top and the bottom half of the die were each heated with three 300 W 76.2 mm long 12.7 mm diameter cartridge heaters which are positioned perpendicular to the pull direction. The cartridge heaters were inserted into an aluminum block that is 25.4 thick, 228.6 inches long, and has the same width as the die. This aluminum block was then tightly clamped to the die. By having separate heaters it gives the user the option, should it be needed, to control the temperatures of each heater individually and have different temperature zones in the die. This option also allows the user to remove a set of heaters if a more heat conductive material, such as carbon fiber is being pultruded.

To understand the temperature profile of the die when the is heated, Finite Element Analysis software ANSYS Work Bench was used to create a mesh and a steady state heat transfer model, as shown in Figure 2. The model simulates the conductive heat transfer from the cartridge heaters and creates a temperature profile for the entire die apparatus. Thermal contact resistance, convection among the surfaces, and the heat load required to change temperature of the material were all taken into account. Since the die is symmetrical along the top and the bottom halves, only a ½ scale model was created to save simulation time. An iterative modeling process was taken to find the appropriate positions for the cartridge heaters to achieve the desired temperature profile along inner walls of the die. The cooling portion of the die, which consists of constant cross-section, was cooled by a chiller.

![Figure 2](image-url)  
*(Left) Steady state heat transfer analysis. Only ½ scale model was used due to symmetry. The pultrusion die, along with the heating and the cooling unit, rests on two steel frame members. Natural convection was applied to these surfaces, shown in yellow. (Right) Heat transfer analysis using ANSYS WB.*
Model the die cavity with accurate die temperature profile

Computational Fluid Dynamics (CFD) is a powerful tool used to simulate fluid flow. It can used to generate velocity vectors, temperature profiles, predict pressure, and capture shear stress fields to name a few. In thermoplastic pultrusion processing the matrix is heated beyond its melt temperature and turned into molten fluid. This molten fluid is pulled and thus generates fluid dynamic motion inside the die. However, the phase changes in the process, the non-Newtonian fluid flow, and the reinforcement fibers make CFD difficult to characterize thermoplastic pultrusion processing and very few studies have taken an advantage of this tool.

In this study, a CFD program Star CCM+ was used to simulate the molten fluid flow inside the die and the overall goal was to predict the force it requires to pull the material with respect to processing temperature and pull speed. The temperature ranges that were examined were 175°C, 185°C, 195°C, 205°C, and 215°C, and the speeds that was studied were 0.00182 m/s, 0.00407 m/s, 0.00681 m/s, 0.001068 m/s, and 0.0144 m/s. This was done by modeling the die cavity (surface which the material is in contact with) and assigning the same temperature profile found from the pultrusion heat transfer model to the cavity walls, shown in Figure 3. By giving the fluid the same thermal properties as the composite calculated in Table 1, an accurate CFD model for melt and solidification model was made. The fluid flow type, laminar or turbulent, and the viscosity were also identified to improve the model.

To identify the fluid flow regime, the Reynolds number was calculated using Equation 11.

\[
Re = \frac{\rho \cdot V \cdot D_h}{\mu}
\]

(Equation 11)

The \(D_h\) term is known as the hydraulic diameter which is a characteristic length used for irregular shapes such as rectangles and discs, and the \(\mu\) is the dynamic viscosity of the fluid (F. P. Incropera, 2007). The hydraulic diameter was calculated using Equation 12.

\[
D_h = \frac{4 \cdot A_c}{P}
\]

(Equation 12)

where \(A_c\) is the cross-sectional area, and the term \(P\) is the wetted perimeter. From the above equation, the Reynolds number was calculated to be far below the turbulent range, mostly due to the high viscosity of the fluid and slow pull speed.

Mapping the resin viscosity

Modeling the fluid dynamic for a thermoplastic material however is not trivial and a full understanding of the material rheology behavior must be made before modeling. Thermoplastics are different in that they have a non-Newtonian flow behavior. Newtonian fluids, such as water, have a linear applied stress and shear strain rate relation, and their viscosity is only dependent temperature and pressure (Askeland, 2006) (Chhabra, 1999). The viscosity relation for Newtonian fluids is defined in Equation 13.

\[
\tau = \eta \cdot \gamma
\]

(Equation 13)
where \( \tau \) is the shear stress, \( \eta \) is the viscosity, and \( \gamma \) is the shear strain rate.

Thermoplastics on the other hand have a shear thinning behavior, where the shear stress and shear strain relation is nonlinear. The shear thinning effect lowers the viscosity when the rate of deformation increases. The ratio between shear stress and shear rate decreases as either the shear stress or the shear rate increases (Wilkinson, 1960) (Sperling, 2006). This phenomenon occurs because at lower shear rates, the entanglements in the polymer chains are much higher than it is at higher shear rates. At high shear rates, the polymer chains become disentangled and the mobility of the chains increases, therefore lowering the bulk melt viscosity (Osswald, 2011).

Ostwald and de Waale formed a simple model to represent the shear thinning region in the viscosity versus strain rate curve known as the power-law, as expressed in Equation 14.

\[
\eta = m(T) \gamma^n \quad \text{(Equation 14)}
\]

where \( m \) is known as the consistency index and \( n \) as the power law index. However, at lower shear rates near the Newtonian plateau region, rates typically witnessed in pultrusion processing, the model overshoots and loses its accuracy (Oswald, 2011) (Ostwald, 1925) (Waale, 1923). For this research the expected shear rate were 0.42 s\(^{-1}\) to 3.4 s\(^{-1}\).

To accurately predict the resin viscosity, a rheometer with parallel-plate oscillating motion was used. Figure 3 maps out the resin viscosity at various temperatures and shear rates of interest. Even though the test was conducted with neat resin, a study by Thomasset et al suggests that the shear viscosity of PP did not change much when compared to PP with 40% long fiber by volume (Thomasset, 2005). The values from the graph were inputted into the fluid simulation in an effort to simulate a realistic molten resin flow. The fluid was also assigned the same thermal properties, from Table 1, as the composite.
Once the thermal property and the rheology of the fluid are inputted into the software, a study was done to simulate phase change of the matrix, from solid to liquid to solid again as seen in thermoplastic pultrusion process. This was done at the most extreme operating conditions where it is least likely to melt; i.e. at the highest speed, when the material spends the least amount of time in the die, and at the lowest operating temperature, where there is the least amount of energy in the system. At all other operating parameter, the chances of melting and consolidating would increase. Figure 4 shows the solid to liquid volume fraction at the center plane of die where it is furthest away from the heating source. It can be seen that the resin enters the die at solid state, melts towards center, and solidifies again before exiting, see Figure 5. This study corresponds directly with the analytical approach where the material was said to have melted through its center and solidified before leaving the die.
Effect of processing parameter die shear stress and pull force

Quantifying the viscosity also allows the user to predict the shear stress along the walls imposed by the resin flow. This can be used to study the structural stress within the die but more importantly to find how much force is required to pull the material through the particular die profile and study how the processing parameter affects the pull force required. Ultimately, knowing this can be used to design the power transplant of the pultrusion system. Figure 6 shows the stress witnessed among the die surfaces at highest shear rate and when the viscosity was at its highest. The results indicate that the stress on the steel die wall is far less than the yield stress of the material.

Furthermore, a study was done on how the pultrusion speed affects the pull force. This was examined by keeping the die temperature at a constant 175°C while incrementally increasing the pull speed. The results of which can be found in Figure 7. The graph indicates that the pull force increases as the pull speed increases. This is a consequence of increased shear stress on the cavity walls. Even though the viscosity decreases slightly from the increased shear rate, the shear rate also causes the shear stress to increase at a high rate. As a result, the pull force required to pull the material increases when the pull speed increases.

![Image of shear stress along die cavity walls](image)

**Figure 6 - Shear stress along the die cavity walls. Pull direction left to right.**

![Graph showing prediction of pull speed versus pull force](image)

**Figure 7 - Prediction of pull speed versus pull force using CFD model. The pull force increases from the increased shear rate.**
The next phase of the study is to examine the effect of increased temperature on the pull force. This experiment will be done similar to the study above but the speed, not the temperature, will be held constant. The temperature will be increased incrementally. More FEA analysis will be done to predict the die temperature profile at the increased temperatures and the predicted die profile temperature will be assigned to the die cavity walls in the CFD simulation. The prediction for this case is that the pull force will decrease if the die temperature rises. The rationale behind this theory is based on the fact that the viscosity will be lowered at higher temperatures.

**Summary**

Thermoplastic pultrusion is a new and developing technique to process composite materials. Reinforcement fiber is pre-impregnated with the thermoplastic matrix and is pulled through a heated forming die to produce a constant cross-sectioned structural member of almost any desired length. It is a very versatile process that is used in various different applications such as construction to automotive. This research details a method to design die for thermoplastic pultrusion by analytical heat transfer analysis and the use of computational fluid dynamic model to aid the design process.

In the thermoplastic pultrusion process, the matrix is heated to the point where it melts inside the die and consolidates. Further down the die, the material is cooled and freezes inside the die to take shape of the die cavity. This melting-consolidating-freezing process was modeled with a steady state one dimensional heat transfer analysis. The model was used to design the die length based on the amount of time it takes to complete the phases changing process.

A fluid dynamic software was used to model the molten resin inside the die. The model was used to characterize the fluid dynamic forces imposed by the molten resin on the die cavity walls. It was used to predict the amount of force required to pull the material through the die. As of now, the die has been machined based on the thermal model, fluid models has been created to predict how the pull speed and the die temperature affect the pull force. The model will be validated by taking force measurements during the process and microscopy analysis will be made to check for voids and validate the consolidation.

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**References**