OPTIMIZATION OF THERMOPLASTIC PULTRUSION PROCESS USING COMMINGLED FIBERS

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ABSTRACT

This work focuses on experimental studies and process modeling pertaining to pultrusion of glass reinforced thermoplastic composites. The experimental work focuses on optimizing the process and investigating process variables which are relevant to the processing of commingled glass polypropylene (PP) fibers. Several parameters affecting the quality of the pultruded parts were identified. A laboratory scale thermoplastic pultrusion machine was set up and glass/PP pultruded composite profiles were fabricated using commingled yarns for different processing variables. The pultrusion process includes tows of commingled fibers, preheater, heating die, chiller and a pulling mechanism. The work describes the process conditions that govern the pultrusion of PP reinforced glass fibers and their behavior on the final properties. The process variables investigated were the number of tows, die temperature, pulling force and pulling speed. Optical microscopy and mechanical testing including flexural strength and modulus have been conducted to evaluate the structure-process-property relationships. Finite element modeling (FEM) study showed that the temperature of the composite in the pultrusion process correlated well with the experiment, hence validating the process.
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1 INTRODUCTION

The pultrusion process which was conceptualized in 1950 by Brandt Goldsworthy is used for making fiber reinforced polymer composites of constant cross section and continuous length. Pultrusion finds applications in electrical, corrosion, building and consumer markets [1, 2]. In the pultrusion process, reinforcing fibers, impregnated with matrix, are pulled through a heated die where they acquire the shape of the die cavity and consolidate before they are cut into required lengths [2-5]. The pultrusion process provides an economical way to produce thermoplastic as well as thermoset materials and promises to deliver superior quality product [1]. Processing parameters play an important role with regards to the appearance and quality of a pultruded part. The process parameters which affect the final properties of the pultruded part are; pulling speed, preheating condition, heating temperature in the die, and cooling rate [6, 7]. A recent study conducted by Composites Fabricators Association (CFA) reported that the annual United States (US) output of pultruded materials is approximately 8.6 million kg of 5% of the total 1.7 billion kg of FRP materials manufactured in US’s $24 billion composites industry. The true growth prospect for this technology is in the development of structural profiles, particularly for bridge construction, offshore structures and large overhead infrastructure applications.

In recent years, pultruded profiles are replacing conventional construction materials such as wood, aluminum and steel in infrastructure, construction,
transportation, consumer, corrosion and electrical markets because it provides high strength structural composite parts and complex profiles. The process is suitable for high volume fabrication of high fiber content linear profiles with a constant cross section such as beams, channels, tubing and rod stock.

Pultruded products are advantageous over structures made from conventional materials like steel and aluminum because of their superior mechanical properties, ease of installation, weight saving and cost effectiveness. Pultruded profiles weigh 50% less than aluminum and 75% less than steel for equivalent strength. Pultruded structures can be pre-assembled into sections and readily brought to the installation site for construction of sea walls, bridges and other structural profiles. A retrofit installation on a bridge made of glass fiber reinforced pultruded tubes filled with concrete and epoxy coated steel cables in Ohio took only 14 days as compared to 32 days it would had taken using conventional materials [8]. In the bridge upgrade in Blacksburg, Virginia, steel flange beams were replaced by pultruded carbon fiber reinforced plastics (FRP) beams which contributed to increased stiffness to magnitude of $6 \times 10^6$ psi [8].

Pultruded beams can achieve better performance over all long term than steel and concrete because it can get rid of the corrosion elements. Despite the fact that initial cost of pultruded structures would be higher than steel, they are replacing steel in sea walls and off shore oil exploration members owing to their weight savings, durability, and resistance to salt and salt air. Pultruded structures are far less expensive over the long run, because structures can last much longer.

Pultrusion has significant market potential in the construction of trim, gutters, doors and windows. The profiles provide resistance to moisture; they do not warp, rot or
corrode. FRP materials maintain dimensional stability over changing temperatures. Pultrusion also finds market in electrical applications such as pultruded power transmission poles and towers. The products are lighter, enable faster installation, are much safer, required reduced maintainance and longer lasting than structures made of conventional materials. Creative Pultrusions in collaboration with General Motors (GM) has developed a prototype pultruded tailgate assembly for a pickup truck [8]. It offers 30% weight savings and 90% reduction in investment costs over metallic counterpart. Pultruded panels have also been used in fire trucks and buses, roll up doors, corner sections, side posts and other structural components for automotive applications [8]. GM has developed a light weight drive shaft made up of glass and carbon fiber reinforced vinyl ester pultruded over an aluminum tube for pickup trucks. These shafts have superior performance based on two years of testing simulating 20 years of vehicle life with an average weight savings of 9 kg [8].

Thermoplastics in powder form have been used to produce the frame and chassis of an all composite passenger sedan developed by Automotive Design and Composites Ltd. [8]. The vehicle featured pultruded carbon fiber reinforced frame rails, overhead supports, and other structural members that were post formed using heat and pressure to bend them into required shapes.

In pultrusion, thermoset resins have been historically preferred over thermoplastic resins because of the ease of impregnation. Thermoset resins are materials that cannot be melted once they are cured. The curing takes place through a chemical reaction which involves a cross linking process. On reheating, thermosets undergo decomposition before they reach the melting point. A thermoset resin cannot be melted and reused, implying
that they cannot be recycled, except when used as a filler material. On the other hand, thermoplastic resins melt to a liquid state when heated and freeze to a brittle, glassy state when sufficiently cooled. Unlike that of thermoset, a thermoplastic resin does not undergo any curing reaction. On reheating, the polymeric chains break easily because they are linked by weak van der Waals forces causing the thermoplastic to melt. Therefore, thermoplastic resins can be recycled [10].

In recent years, pultrusion of thermoplastic composites has grown because it presents significant advantages over thermosets including improved toughness and durability, enhanced damage resistance, environmental resistance, cost-effective processing, post-process formability and recyclability [6, 11, 12]. Thermoplastic materials provide ease of processing which is limited to melting the matrix, compaction under pressure and finally cooling [4]. Thermoplastic are easy to join using plastic welding processes instead of adhesives and fasteners.

In previous years, pultrusion with thermoplastic materials had been difficult and challenging, primarily due to the higher viscosity of thermoplastic resins as compared to thermoset resins, resulting in poor fiber impregnation [4, 10, 13]. But this drawback has been overcome due to the development of new processing technologies and number of intermediate material forms such as commingled fibers, powder impregnated bundles, hot melt impregnated tows and in-situ polymerization.

In 1999, Dow plastics developed Fulcrum, a low viscosity resin technology, especially for thermoplastic pultrusion process. Fulcrum provided a solution to overcome poor fiber impregnation [5, 8]. In this technology thermoplastic polyurethane resin was pultruded by reversing the polymerization process in its melt stage. Thermoplastic
composites fabricated using this technology demonstrated the same mechanical properties as that of the thermoset composites, in addition to providing improved toughness and damage tolerance with greater processing versatility; since the profiles were thermoformable.

Commingled tows with partial impregnation capabilities have been developed to improve processability of thermoplastics [11, 12]. In commingled tows, the fibers as well as the matrix (PP) are in fiber form. The matrix fibers are commingled with reinforcing fibers essentially of similar diameter in order to achieve adequate distribution of the respective constituents [11]. Commingled tows provide a good balance between versatility of performance, ease of processing and price. PP and polyethylene (PE) are commonly used since they can be processed easily and are less expensive than other resins. A number of semi finished products have been developed where the matrices and the reinforcing fibers are commingled, but there still remain difficulties to achieve good wetting of fibers with the resin [12, 13]. However, pultrusion of thermoplastic matrices is becoming increasingly popular with the development of commingled tows which facilitates impregnation [14].
CHAPTER 2 LITERATURE REVIEW

2.1 Thermoplastic Pultrusion

Thermoplastic pultrusion is a manufacturing process in which thermoplastic materials which might be in pre impregnated form, powder impregnated bundles, hybrid yarns, melt form etc. are pulled through the heating die where they impregnate the reinforcing fibers before cooling sufficiently to consolidate and are then cut to required lengths [15-19].

Figure 1. Schematic of pultrusion process

A typical set up for thermoplastic pultrusion is shown in Fig. 1. The thermoplastic material stored on the creel is guided into the preheating chamber, where it is heated to the melt temperature of the polymer in order to facilitate fast processing.
After preheating, the commingled tows are pulled through the electrically heated die where the resin melts and impregnates the reinforcing fibers, before in the cooling die. The profile is then pulled through the die with the help of a pulling device placed at the end of the pultrusion line. The profile is cut to required shapes.

A typical pultrusion machine consists of the following parts:

1. Preheater

2. Die assembly
   A. Heating die
   B. Cooling die

3. Pulling Mechanism

1. Preheater

The preheater facilitates rapid processing of the material since it heats the polymer close to its melt temperature and initiates wet out of the reinforcing fibers. It is essential to preheat the material before it enters the heated die, otherwise the required process temperature in the core section of the material is not attained and the composite does not fully consolidate. During heating the material must be:

(1) in no contact with the inner area of the preheater to prevent it from melting and sticking to the preheater.

(2) continuous to avoid localized overheating and degradation; and

(3) uniform to minimize temperature difference through the thickness of composite.
The length of the preheater can be decreased by short preheating times, and increased pultrusion speed. The preheater zone is equipped with strip heaters or infrared heaters to heat the material, and thermocouples to monitor and control the material temperature.

2. Die assembly

The die assembly comprises of heating and cooling dies separated by a narrow gap and are perfectly assembled, ensuring alignment. In the heating die the material gets heated to its melt temperature. The PP polymer flows, impregnating the glass fiber bundles. The exiting material from the heating die cools and consolidates in the cooling die. The die assembly has a cavity which is tapered at the entrance to allow slight overfill in order to assist build up of consolidation pressure. The heating dies are provided with heaters which ensure heating of the material in the die and thermocouples to monitor the temperature. The cooling dies are generally equipped with chillers. The cooling die extracts heat from the material aiding consolidation.

The die assembly consists of a lower and an upper part that are assembled by screws. A graphite gasket is placed between the upper and lower die to along the length to increase the pressure inside the die. It also avoids heat loss and prevents material of the sides of the die.

The dies are machined from tool steel and the cavities are highly polished. The die cavity is chrome plated in order to reduce friction between the moving material and the cavity walls.
3. Pulling Mechanism

The pultrusion line is equipped with a pulling mechanism that enables to pull the material through the die at different speeds and then it is cut into required lengths. The pulling mechanism consists of polyurethane coated metal plates of constant cross section that aid in gripping the material while pulling. The pulling mechanism is operated via a variable speed direct current (DC) motor. The DC motor drives polyurethane plates through a pneumatic transmission. Each machine is designed for a maximum linear speed and pulling force that can be changed by varying the motor speed.

2.2 Processing Cycle

Bernet et al. [20] described the processing cycle for a thermoplastic pultruded part made up of commingled fibers assuming that the mechanism of consolidation is fiber impregnation which is validated using the work done by Phillips et al. [21] on consolidation of carbon fiber (CF)/polyetherimide (PEI) prepregs. The processing cycle consists of raising the heating temperature to the melting point of the resin and applying pressure for a fixed period of time for appropriate consolidation and finally cooling at constant pressure. Three main mechanisms take place during the consolidation stage, (1) close contact of fibers due to compression; (2) autohesion (resistance to separate two bonded identical film); and (3) fiber impregnation. From these three processes, close fiber contact and autohesion processes contribute to only 1% of total consolidation time, the rest is consumed by impregnation.

2.3 Thermoplastic Pultrusion Methods

The thermoplastic pultrusion can be carried out in two ways as shown in Fig. 2,
1. Reactive pultrusion

2. Non-reactive pultrusion

Figure 2. Various thermoplastic pultrusion methods

Note: Adapted from “Reaction injection pultrusion of PA12 composites: process and modeling”, A. Luisier, P.E. Bourban, J.E. Manson, 2003, Composites: Part A 34, p. 583-595.

2.3.1. Reactive Pultrusion

The reactive pultrusion process comprises of both thermoset as well as thermoplastic pultrusion processes. It features an injection unit to pump low viscosity thermoplastic resin to bring about the chemical reaction necessary for in-situ polymerization. As shown in Fig. 3, reactive pultrusion process is similar to that of
reactive thermoset pultrusion but differs in the design of the injection unit used for in-situ polymerization. In this process, the injection unit comprises of two small storage tanks to store molten lactum above its melting point and a liquid system (nitrogen) at room temperature and is designed in such a way that both materials inject into the mold with an accurate ratio, under high pressure and short residence time.

![Schematic of reactive thermoplastic pultrusion process](image)

Figure 3. Schematic of reactive thermoplastic pultrusion process

Note: Adapted from “Reaction injection pultrusion of PA12 composites: process and modeling”, A. Luisier, P.E. Bourban, J.E. Manson, 2003, Composites: Part A 34, p. 583-595.

2.3.2 Non- Reactive Pultrusion

In non-reactive pultrusion process there is no chemical reaction involved. The material passes through the heating die, melts in the heating die and consolidates in the cooling die as shown in Fig. 1.

2.4 Limitations of Thermoplastic pultrusion process

Though thermoplastic materials are replacing thermoset materials in several applications, they have limitations summarized as follows:
1. The process parameters for thermoplastics are not fully developed which results in pre-production cost and quality limitations.

2. Thermoplastic materials have high melt viscosity which results in improper impregnation of reinforcing fibers.

3. They are susceptible to deformation while consolidation.

4. There are pressure limitations in the die resulting in inadequate pressure for impregnation.

5. High line speeds are difficult to achieve since it detorirates the mechanical properties, because of improper impregnation.

2.5 Technology Development

Several researchers have investigated the pultrusion technology for thermoplastics as well as thermoset materials with respect to choice of material used, optimization, parametric effects on mechanical properties and heat transfer modeling.

With respect to technology development in case of thermoplastic pultrusion Michaeli et al [22] carried out braided fiber layup pultrusion using PP. They employed a core spinning process for one of the material systems using PP/glass hybrid, commingled yarn of carbon (C)/liquid crystalline polymer (LPC) and carbon (C)/poly ether ether ketone (PEEK).

The basic setup of a pultrusion braider is similar to that of the thermoset pultrusion line with the only difference that the coil magazine is replaced by a textile braiding machine. In this process the preliminary material used is filaments of dry hybrid yarns used as reinforcements.
According to Michaeli et al [22] the processing of hybrid yarns provides advantages over other thermoplastic fiber reinforced materials as starting products being:

1. Limitless combinations of materials
2. Ease of transport and handling
3. Potential for textile processing, and
4. Easy process technology, only a little different as compared to traditional pultrusion.

Sala et al [23] used fiber impregnation technique using poly butylene terephthalate (PBT) thermoplastic resin. They demonstrated manufacturing rates 5-10 times higher than thermoset pultrusion, better diffusion of the matrix in the fiber network and higher toughness of the resulting part.

Luiser et al [24] developed a reactive thermoplastic pultrusion process by using PA12 and took into account in-situ polymerization of low viscosity thermoplastic lauryllactum monomer. This technology includes principles of thermosets as well as thermoplastic pultrusion. It differs from conventional thermoplastic pultrusion because it involves complex chemical reactions of thermoset resin cure.

2.6 Processing parameters and its effect on other variables and mechanical properties

As in any other processes, processing parameters play an important role on the outcome of the product.

In case of pultrusion process the processing parameters that are varied for obtaining a good quality part are; preheating temperature, heating die temperature, pulling speed and pulling force. Carlson et al [4] describe the processing conditions for
melt impregnated glass/PP prepeg and the influence of processing conditions on the resulting mechanical properties. They investigated process variables with respect to preheating temperature, die temperature, cooled die temperature and pulling speed and assessed the effects of these variables by characterizing flexural modulus and strength, surface finish, and pulling force. They showed that with increase in line speed, good surface finish can be obtained, but the fiber distribution over the cross section is not properly obtained. With increase in pulling speed from 2 to 6 mm/s, they observed the surface to become glossy and smoother as shown in Fig. 4 and 5. They observed the appearance of smaller spherulites at the surface of the composite that creates many nucleation sites resulting in further crystallization. However higher pulling speeds adversely affect mechanical properties because it does not provide enough time for the fibers to distribute uniformly throughout the cross section resulting in improper impregnation.

![Gloss versus pulling speed](image)

Figure 4. Gloss versus pulling speed
Carlson et al [4] also studied the effect of preheat temperature on the appearance and mechanical properties of the material. They reported that preheat temperature controls the shine of the surface. They found that at higher preheat temperature (175°C), the prepeg resin melts, resulting in fiber relaxation and upsetting the fiber distribution, resulting in small decrease in the flexural modulus (around 2 GPa) along with loss of gloss and surface finish.

![Graph showing roughness versus pulling speed](image)

**Figure 5.** Roughness versus pulling speed

With variation in the heating die temperature they found a variety of fiber distribution patterns. The pultruded profiles manufactured using high die (225°C)
temperature showed a fairly uniform distribution of fibers over the cross section. Some of the samples demonstrated a distinctive ply structure which slowly disappeared at the surface giving a fairly uniform fiber distribution. On the other hand, samples manufactured using low die temperature (205°C) showed distinctive ply structures which did not disappear at the surface, therefore lacking uniform distribution. The temperature did not reach the center of the composite resulting in low level of consolidation and fiber distribution.

Sala et al [23] evaluated the influence of parameters including pultrusion velocity, temperature and die length using PBT powder impregnated glass fiber bundles. They studied the effect of die length on pulling force, speed on pulling force and die length on pressure. They developed various models including a pulling force model.

Figure 6. Pulling force versus die length

Sala et al [23] reported that there is a reduction in pulling force with increase in die length due to unchanged dimensions of the die at the inlet and outlet, and also taper of the die decreases along the length resulting in drop in back flow component with drag flow remaining unchanged (Fig. 6).

Sala et al [23] also reported that when the pulling velocity is increased, the pulling force is increased as shown in Fig. 7. As the velocity is increased, shear stresses acting in the thin layer between the flowing material and the die wall give rise to viscous force that increases with increasing velocity. The viscous force is estimated as:

\[
F_{\text{vis.}} = 2(b + h) \int_0^L \left( \eta \frac{U}{\delta} \right) dx \quad [1]
\]

where \(b\) is breadth of the die, \(h\) is height of the die, \(L\) is length of the die, \(\eta\) is viscosity of the resin, \(U\) is the line speed and \(\delta\) is the thickness of the resin layer.
Based on the experimental work and numerical modeling, the pulling force decreased gradually with increase in temperature as shown in Fig. 8 [23]. Their reasoning was that with increase in temperature, the resin flows freely and provides less resistance to pulling, which results in decreased pulling force.
Figure 8. Pulling force versus temperature: comparison between the numerical models


Michaeli et al [22] studied different processing parameters in which pultrusion was carried out using braided fibers and a thermoplastic PP matrix. Pultrusion trials were carried out using different processing variables. The quality of the pultruded profile was found to depend on the kind of material used during processing and on the amount of resin and fiber content [22]. The optical properties show marked improvement when parallel hybrid materials such as commingled fibers were used. This is due to improved fiber-matrix wet out and homogeneous fiber distribution.

Angelov et al [13] studied the influence of line speed on pultruded profiles and evaluated the quality by performing a three point bend test, as well as Charpy impact test
on flax/PP material. Flax fibers are natural fibers which are environmentally friendly, recyclable and exhibit superior specific properties alike to the properties of glass fiber reinforced composites. The authors found that at higher pulling speeds flexural strength of the pultruded profile decreased as compared to lower pulling speed. Additionally, they found that the flexural strength gets affected with variation in fiber content. When they increased the fiber content from 30% to 50% the flexural properties increased approximately by 30%. They reported the same magnitude of impact energy for samples pultruded using 30% and 50% fiber content respectively.

Figure 9. Flexural strength of pultruded and compression molded samples


Angelov et al [13] also compared the mechanical properties of the composites manufactured using a pultrusion process and a compression molding process. They found that the samples made using compression molding process demonstrated higher
flexural properties than the pultruded samples as shown in Fig. 9. This can be attributed to higher impregnation attained in compression molded samples than the pultruded samples. This shows that there is strong dependency of processing parameters including temperature and pressure on the impregnation quality, and hence on the mechanical properties of the composite.

Miller et al [11] developed a correlation between experiments and a model for commingled glass/PP fibers and dry powder impregnated PA 12/glass fiber reinforced towpregs. They found that although high pultrusion speeds were not successfully achieved the mechanical properties, surface and impregnation qualities were adequate. The mechanical properties deteriorate due to inadequate consolidation. They reported minimal void content at high line speeds as high as 10 m/min.

Miller et al [11] evaluated the mechanical properties using three point bend flexural test for pultruded profiles of PA12/GF laminates manufactured from prepegs. The pultrusion line speeds varied between 1-10 m/min. Figure 10 shows decrease in flexural strength with increase in the line speed. Cavities appeared inside the pultruded part because of void formation with increase in line speed. The values of flexural strength changed between 600 to 700 MPa with the final void content values ranging from 1 to 4 %. Mechanical properties were adequate and are not notably influenced by void content with the increase in line speed.
The mechanical properties relative to line speed and impregnation quality for commingled profiles were measured in terms of flexural strength. Figures 11 and 12 illustrate that the flexural properties were reduced to some extent as the line speed was increased, due to the decrease in impregnation time causing improper fiber wetout and resulting in void formation. For the speeds varying from 1-5 m/min the void content was measured to from 0 to 2 %. For the speed varying from 5-10 m/min, the void content was
1 to 4%. The flexural strengths was 600 to 700 MPa for both the line speeds and void contents.

![Figure 11. Flexural strength versus line speed for pultruded commingled PP/GF 20 X 2 mm strip](image)


Creighton et al [25] studied the compressive strength of carbon fiber/epoxy composite pultruded profiles using. The failure occurs parallel to the fiber axis. The main reason for the decrease in compressive strength was attributed to the void aspect ratio. In the case of elliptical shaped voids, peak shear stress develops adjacent to the void, and the shear stress decays at a distance away from the void. Hence they predicted
that these changes in the stress state influenced by the aspect ratio significantly affect the mechanical properties resulting in reduced strength.

Figure 12. Flexural strength versus line speed for a pultruded commingled PP/GF 2 mm diameter rod


2.7 Impregnation models and heat transfer simulation

The heating system in a pultrusion line is comprised of a heating die where melt impregnation takes place, and a cooling die where consolidation takes place [12, 26, 27]. Most of the impregnation takes place at the tapered end of the die where the temperature of the composite is in steady state. In commingled tows, the molten resin does not
disperse uniformly among the reinforcing fibers. This leads to fiber agglomeration and formation of voids due to incomplete impregnation, which causes deterioration of mechanical properties as can be seen in Fig. 13.

The major problem of optimizing the thermoplastic pultrusion process is to find a way by which maximum impregnation can be achieved under given processing conditions. This can be achieved through modeling which can help in predicting the temperature profiles in both the pultruded part and pultrusion die. But the real problem is the impregnation of the fibers by the polymer in thermoplastic materials.

![Diagram of cross section of commingled yarn](image)

**Figure 13.** Simplification of the cross section of the commingled yarn as an array of agglomeration


Several authors conducted numerical modeling to predict impregnation of the system and the heating mechanism taking place in the die. In developing the model for a die, many authors [28-31] assumed temperature and pressure to be the dominant factors
involved in proper wet out, and hence built their models to evaluate the temperature and pressure profiles in the die and the composite, and the subsequent influence of these parameters on the final pultruded part.

The impregnation and consolidation of commingled fibers has been studied by several researchers [32-37]. Some authors have developed microscopic models of the impregnation process. Gibson et al [38] and van West et al [39] established a model by employing a semi empirical Darcy’s law. Van West et al further developed the model by implementing the work done by Kozeny-Carman [40] and Gutowski [41] which describes variation in permeability in the fibrous bed related to extremely viscous resin flow of a thermoplastic matrix.

The work published thus far differs in the intricacies present in the computer model, the variety of materials used, and whether a microscopic or a macroscopic path is followed. Because the pultrusion process and its mechanisms involve heat transfer and flow of the material through the die, the problem of heat transfer is divided into a heat transfer problem and flow problem pertaining to impregnation. To address these problems, well-known equations including the energy equation, equation of motion and Darcy’s law among others are used.

Kim et al [14] studied the pultrusion process using a set of thermoplastic commingled yarns (Twintex® RPP60 and RPP75 respectively) and implemented macroscopic and microscopic sub models to develop an impregnation model. They examined and studied the development of the resin pressure inside the heating die in order to effectively model the impregnation process. Their model describes macroscopic
axial flow of resin along the arrangement of fiber agglomerations and resin flow impregnation through fiber agglomerations respectively.

Figure 14. Microphotographs of a typical cross section of the pultruded parts (a) poorly impregnated part using RPP75 yarns and (b) fully impregnated part form RPP60 yarns with same processing conditions


They performed analysis and experiments and found that the fiber agglomeration size is the most significant factor in improving the degree of impregnation. Figure 14 shows a pultruded part from Twintex® RPP75 and RPP60 yarns respectively. As the
agglomeration size of RPP75 is large, it exhibits incomplete impregnation as compared to RPP60 yarns for the same processing conditions. This suggests that for rapid and adequate impregnation of the fibers, the agglomeration size has to be small because it will induce high macroscopic pressure resulting in good impregnation.

Figure 15. Degree of impregnation of pultruded parts as functions of pulling speed for different processing temperatures and die inlet areas for RPP75. Comparisons between experimental data and predictions by the model


According to Kim et al [14], the taper geometry also plays an important role in affecting the degree of impregnation but not as significant as the agglomeration size. To accomplish better impregnation a small taper entrance and long taper length is considered...
necessary. This helps in building high macroscopic pressure inside the die resulting in good impregnation. However, the drawback of using a die with a small taper entrance is it may obstruct the entry of raw material into the die because of high shear rate caused by backward resin flow. It may also lead to high pulling force due to increase in macroscopic pressure. However the data they generated for smaller die entrance area demonstrated lower degree of impregnation than estimated at high pulling speed as shown in Fig. 15. They attributed this to shear thinning of the resin at high shear rates of induced pressure with decrease in resin velocity.

Michaeli et al [22] investigated the pressure in the die and they assumed that impregnation is a function of temperature, time and resin viscosity for commingled fibers. Their emphasis was on the temperature difference in the die and its effect on the die pressure. They offered two reasons for pressure built up in the die. Pressure built up in the die cavity is due to the thermal expansion which is caused by a temperature difference at the entrance and end of the die. Pressure also builds up at the entrance of the die because of resin back flow. They measured the pressure inside the die using strain gauges and calculated the pressure using a thermal expansion model. The pressure in the die plotted versus the temperature difference for measured average values and calculated value is illustrated in Fig. 16. The difference in the pressure shows that both values are independent of the type of semi-finished product used during processing.
Seo et al [42] examined the impregnation of continuous thermoplastic PEEK 150P/graphite composite. They explained the degree of impregnation as a function of time for different processing variables including pressure, temperature and tow size.

Seo et al [42] discussed two cases for impregnation i.e. an ideal and a real case. They reported that permeability of the fiber bundle is a function of the applied pressure. In an ideal condition, each single fiber is perfectly straight and parallel with respect to each other. When the pressure is applied, the fiber bundle moves closer to each other leaving no space for resin to penetrate between them. In the real case, however, every
fiber is not perfectly straight and parallel to each other, but is somewhat wavy in nature. This makes fiber bundles to come into physical contact with each other at some points when the pressure is applied. Between these contact points there is some space for the resin to pass through and this space is directly associated with the permeability of the fiber bundle which provides resistance to the flow of resin. With increase in pressure the space gets smaller, leaving less space for the resin to penetrate, and decreases the resistance to the resin flow providing adequate impregnation.

Haffner et al [1] studied the pressure and velocity profile. They observed that at the entrance of the die the pressure is not evenly distributed because of high viscosity of the polymer. However, as the material passes through the die, pressure starts to stabilize gradually as the polymer becomes more fluid and is well distributed towards the end. In the velocity profile they found the resin near the walls of the die has low viscosity, while the viscosity closer to the center of the composite is high. In the center section of the die, the matrix is evenly distributed and the polymer flows in a forward direction, while at the end of the heating die, the matrix accelerates due to decrease in pressure resulting in steadily impregnating moving fiber bundles.

They reported that the fiber bundle which is nearer to the heating die wall goes beyond the melting temperature of the matrix, while the fiber bundle closer to the center of the die is in the process of reaching this critical temperature at some distance in the die. In case the fiber bundle does not reach the melting temperature, the grouped polymer particles get squeezed into the interstitial regions between the single glass fibers. This results in improper wetting and hence voids appear.
Michaeli et al [22] applied improved heat transfer and conduction theory for a system under high pressure and found that with high pressure and temperature the resin tends to flow back and decreases viscosity; resulting in improper impregnation. In addition, they measured the temperature profile on the length of the pultrusion line to calculate the definite temperature profiles in different zones for different fiber/resin combinations and stated that the material gets heated by conduction, convection and radiation.

They concluded that hot air heating (convection) is more suitable for heating, since it protects the material from overheating due to variation in the pultrusion speed caused due to gripping and releasing of the material by the pullers. For cooling systems and fiber/matrix combinations, the highest cooling rate was achieved with water, and a system with high specific heat capacity has lower cooling rate as in case of the system with PP matrix as compared to poly ethylene terephthalate (PET) system. According to them these two factors contribute to the consolidation of the material in the cooling zone.

Astrom et al [43] developed a set of models to explain the distribution of temperature and pressure inside a carbon (C)/polyether ether ketone (PEEK) thermoplastic composite when it is moving through the pultrusion die. In addition to this, a model to describe the build up of pulling resistance in the pultrusion die was also developed. The models presented in their paper relates to an idealized and steady state pultrusion process.

The temperature model put forth by Astrom et al [43] accounts for one dimensional heat transfer taking place in an infinite composite slab with a set boundary temperature or heat flux from the surfaces. In developing the temperature model they
considered the material to be transversely isotropic in nature, and ignored the heat transfer along the width.

Haffner et al [1] developed a three dimensional model to simulate the temperature profile and macroscopic flow in the die for a thermoplastic composite material employing both non Newtonian model and enthalpy model. Their study concentrated on various process parameters using thermoplastic prepegs such as powder impregnated sheathed yarns (glass fibers and PBT). They presented an approach focusing on the finite element simulation of the conditions in the heated die. Their model explains the anisotropic fluid flow and thermal properties inside the die section. In addition to this the effect of shear thinning behavior is presented and its effect on the flow of the matrix profile is shown.

Chachad et al [9] reported three dimensional numerical investigations of the thermochemical aspects of the design and manufacturing of a pultruded composite. They formulated a numerical model based on the Patankar’s finite difference technique [44] and developed a three dimensional model accounting for cure and for the anisotropic material in the heating die. They developed temperature profiles for each of the heating components including metal platens, heating platens, die and composite, eliminating the need for predetermined die wall temperatures as a boundary condition.

Liu [45] described a numerical simulation of the pultrusion process using fiber-vinyl ester composite I-beam using. The procedure is capable of predicting temperature profiles in both the pultruded part and the pultrusion tool, as well as the curing profile in the part under different process conditions. Liu et al [3] formulated a numerical model to obtain temperature and curing profiles at different temperature settings and pull speeds.
In most studies [3, 19, 46], the conservation of energy principle is used in the governing equation for heat transfer in the die. This is represented by:

$$\rho c_p \frac{\partial T}{\partial t} = \frac{\partial}{\partial x} \left( k_x \frac{\partial T}{\partial x} \right) + \frac{\partial}{\partial y} \left( k_y \frac{\partial T}{\partial y} \right) + \frac{\partial}{\partial z} \left( k_z \frac{\partial T}{\partial z} \right) + q \quad [4]$$

where, \( T \) is the temperature in degree Kelvin; \( \rho \) is the density of the die material; \( c_p \) is the specific heat of the tooling material; \( k \) is the thermal conductivity of the die; and \( q \) is the rate of energy transfer at the boundary.

In pultrusion, the fibers are impregnated with the resin before entering the heated die, so it can be assumed that the resin does not flow. The pultruded part is moving in the pull direction at the pull speed [3, 9, 24, 45]. The energy equation for the part can be written as

$$\bar{\rho} c_p \left( \frac{\partial T}{\partial t} + u \frac{\partial T}{\partial x} \right) = \frac{\partial}{\partial x} \left( \bar{k}_x \frac{\partial T}{\partial x} \right) + \frac{\partial}{\partial y} \left( \bar{k}_y \frac{\partial T}{\partial y} \right) + \frac{\partial}{\partial z} \left( \bar{k}_z \frac{\partial T}{\partial z} \right) + V_r Q \quad [5]$$

where \( V_r \) is the volume fraction of resin; \( u \) is the pulling speed; \( \bar{\rho} \) is the lumped density; \( c_p \) is the specific heat for the composite material; \( \bar{k}_x, \bar{k}_y, \bar{k}_z \) are the lumped thermal conductivities in \( x, y \) and \( z \) directions respectively; and \( Q \) is the rate of internal heat generation caused by the resin reaction.

The lumped properties are evaluated by the following equations:

$$\bar{\rho} = (1 - V_r) \rho_f + V_r \rho_r, \quad [6]$$

$$\bar{c}_p = \frac{(1 - V_r) \rho_f c_{pf} + \rho_r V_r c_{pr}}{\bar{\rho}}, \quad [7]$$
\[
\bar{k} = \frac{k_f k_r \bar{\rho}}{(1-V_r) \rho_f k_r + V_r \rho_r k_r}
\]  

Also, for the two dimensional process where the influence of pressure on the heat of reaction is neglected is given by the following global heat transfer model. Here the solution is approached by using an iterative technique of two sub models.

\[
\rho c u \frac{\partial T}{\partial x} - \rho \left( k_x \frac{\partial T}{\partial x} \right) - \rho \left( k_y \frac{\partial T}{\partial y} \right) - H_r \rho m_m \frac{\partial \alpha}{\partial t} = 0
\]

where \( T \) is the temperature of the material; \( k \) is the thermal conductivity of the material; \( c \) is the heat capacity of the material; \( u \) is the line speed; \( H_r \) is ultimate heat of reaction; \( m_m \) is the mass fraction of matrix; \( \alpha \) is degree of cure; and \( \rho \) is density of the material

Sunil et al. [47] investigated the cure optimization of thermoset pultrusion process by varying preheating and cooling die temperatures to minimize overshooting of the temperature in the heating die which could cause degradation of the material system. The work carried out in [48-50] reported that within the heating die, the temperature of the composite goes beyond its melting temperature, therefore degrading the composite and affecting the desired degree of cure. Thus the maximum permissible temperature within the composite is a limiting factor during optimization, and once the permissible temperature is achieved, further optimization cannot be achieved without causing degradation. This can be minimized by choosing a proper pre heat as well as cooling die temperature for the composite.

Sunil et al [47] also employed a three dimensional finite elements/nodal control volume approach and analyzed different cases by changing temperature of a glass/epoxy
system at the preheat die inlet and cooling die. The results showed that if the preheating and cooling die temperature is carefully chosen the overshooting of the temperature can be minimized, and the polymer prevented from degradation. They also stated that the cooling die temperature has more influence on the curing than the preheat temperature of the die. Sunil et al [47] showed the effect of preheat and cooling temperatures on the material, and in the same way Michaeli et al [22] extended the work by conducting post analysis of the material of the cooling section and suggested optimal temperature of the material when it exits the cooling die. They stated that the material exiting the cooling die is below its melting temperature. In this scenario there is a possibility of the material developing tensile forces due to the action of alternating pullers. These tensile forces can be minimized by maintaining the temperature of the pultruded part below the glass transition temperature of the polymer.

2.8 Pulling Force Measurement

The prediction of pulling force of the material generated inside the die is of utmost importance in designing the die as well as in determining the process conditions [51]. The measurement of pulling force is necessary because there is resistance to the flow of material mainly due to the frictional forces arising due to the friction between the passing material and the die walls. To minimize these frictional forces, the inside cavity of the die is chrome plated [13, 24]. The heated die cavity is tapered at the entrance to facilitate the fiber supply and to build up pressure which is caused from backward resin flow [14, 51]. This back flow of resin causes obstacle for the material to pass freely through the die cavity and hence increases the pulling force required to pull the material.
Yun et al. [51] carried out an analysis to predict the pulling force during the pultrusion of phenolic foam composites with methylene chloride (CH₂Cl₂) as the blowing agent. They developed a model to explain the interaction between the pulling force and the process variables such as die temperature, amount of blowing agent and pulling speed. They found that the pulling force in pultrusion of foamed composites is significantly higher than composites with no foam due to pressure exerted by expanding bubbles. In their modeling approach to predict the pulling force, they studied the pressure built in the die by applying an one-dimensional porous medium flow for both tapered and flat regions of the heated die wall and this was validated using load cells. They reported that the pulling force is generated inside the pultrusion die when the fiber bundles pass through the tapered entrance after impregnation. This causes excess resin to squeeze out in the opposite direction of material movement. The excess resin gets squeezed out in the opposite direction of the pulling speed. These fiber bundles cause resistance to flow, hence cause the pressure in the resin to increase towards the taper end, hence increasing pulling force in this region. After passing the tapered region, the pressure decreases due to viscous shearing of the resin along the die wall. Once the composite is introduced into the straight section of the die after passing the tapered region, viscous shear takes place between the die wall and the fiber bundle which causes loss of pressure, and hence decrease in pulling force.

Srinivasagupta et al [5] reported steady state and dynamic analysis of a bench scale injected pultrusion process using a DERAKANE 441-400 epoxy vinyl ester resin system. The main objective of their research was to set up a cost-effective process that helps in achieving maximum production without breaching the process parameters such
as the pulling speed, temperature and pressure. According to them, maintaining

temperature profiles in the die at different points guarantees melting of polymer, but

adequate impregnation and consolidation depends on the operating variables such as

polymer/fiber properties, pressure and pull speed.

They developed a pulling force model on the assumption that the resin back flow

and the viscous drag of the material on the die wall are the contributing factors for the

pulling force. In their pulling force model they divided the die into two sections i.e. the
tapered section and the heating section. According to them, the tapered section of the die
is the chief contributing factor for the force acting in the die. This force is due to the

compaction force from viscous drag of resin, the taper angle and the fiber elastic forces.

In the heating section, the primary contributing factor for the pulling force is the viscous
drag arising when a thin layer of resin is sheared between the fibers and the die wall.

Carlsson et al [4] reported that the pulling force is mainly influenced by only one

process variable i.e. pulling speed. With increase in the pulling speed higher pulling

force is achieved. Their results showed a concave upward dependency of pulling force

with variation in the pulling speed and a large standard deviation for the maximum

pulling speed of 6 mm/s. They reported that at this speed the process was interrupted for

some process variables and in these cases the pulling force constantly increased until the

fibers failed. To evaluate the pulling force at the time of failure, so that fiber failure can

be avoided, they established a mechanism that aided them in measuring pulling force.

They designed a die assembly that was free to move in the pulling direction. The die

assembly was equipped with two round headed screws at the exit. When the material was

pulled through the die assembly, the assembly moved towards a vertical lever, which was
hinged at one end and contacted a load cell at the other end. As the vertical lever comes into contact with load cell, it exerts pressure on the cell, which is recorded using a data acquisition software.

Sala et al. [23] explained that in a steady state process the resin flow inside the die is entirely directed along the fibers and the matrix powder fills the total space between the fibers. The resin back flows in the die due to the fiber motion in the forward direction. This back flow is opposite to the pulling direction and also opposite to the pressure gradient which is caused due to the tapering of the die, and also because of the high viscosity and low velocity.

Sala et al. [23] attempted to evaluate the pressure in the die to calculate the pulling force. They attributed the pulling force to two reasons i.e. the forces due to the back flow of the resin, and due to the viscous forces originating in the thin layer formed between the composite and the die walls.

In their pulling force model, they divided the model in two parts. In the first part they calculated the back flow forces and in the second part, the viscous forces. In the first part they neglected the effect of wall and the small tapering angle of the die and only considered the forces originating from resin back flow which starts off due to stresses exerted by the resin when it moves in the opposite direction to the fiber movement. When the wall effect and tapering angle was neglected, the force equilibrium acting on a volume element of the die is written as:

$$\frac{1}{A} \frac{dF_{b,f}}{dx} = \frac{dP}{dx}$$

[10]
Therefore the back force along the length of the die is given by:

\[
F_{b, fl.} = \int_0^L A \frac{dP}{dx} \, dx
\]  

[11]

where \( A \) is the cross sectional area of the die, \( F_{b, fl.} \) are the forces due to the back flow. In the second part of model development viscous forces were measured by considering parameters including moving velocity, thickness of the resin layer etc, since these forces are created by shear stresses acting between the thin resin layer of the composite and die wall moving at velocity \( U \).

Both Newtonian and power-law models are employed to describe the resin viscosity. In the case of the Newtonian model, the resin viscosity is given by:

\[
F_{vis.} = 2(b + h) \int_0^L \left( \eta \frac{U}{\delta} \right) \, dx
\]  

[12]

where \( b \) and \( h \) are the width and height of the composite, \( L \) the length, \( \eta \) is resin viscosity, \( U \) the composite velocity and \( \delta \) is thickness between thin resin layer and die wall evaluated experimentally.

In the power law model, the equation for stress distribution is expressed as:

\[
\tau_{rz} = \frac{1}{2} \frac{dP}{dZ} \left( r - \frac{R^2}{r} \right)
\]  

[13]

where \( \tau_{rz} \) is stress distribution in the thin layer of resin, \( r \) is fiber radius and \( R \) is external radius of the annulus.
Equation 13 describes the shear stress distribution in the thin layer of resin. The integration between R and thickness of thin layer along the die length gives the value of pulling force required to overcome the viscous friction.
CHAPTER 3 OBJECTIVES

The major objectives of this research are:

(1) To develop a thermoplastic pultrusion process using commingled fibers.

(2) To study the effect of different processing variables in thermoplastic pultrusion.

(3) To evaluate mechanical and physical properties of pultruded thermoplastic composite and compare them with relevant published work.

(4) To develop a finite element analysis model to describe the heat transfer of the composite material in the die.
CHAPTER 4 MATERIALS AND METHODS

4.1 Material

Glass/PP Twintex® [6] RPP75, shown in Fig. 17 was used in the present work. The Twintex® material was provided by Saint-Gobain Vetrotex. Vetrotex has patented the procedure of manufacturing commingled tows with a uniform distribution of glass and PP fiber bundles, with different glass fiber volume fractions. The commingled tows manufactured by Vetrotex [7], under the trade name Twintex® [6] comprise 25% matrix and 75% glass by weight (48% by volume). The diameters of PP and glass fibers are 14 and 20 μm respectively [7]. The PP matrix has a melting temperature, T_m, of around 165 °C (329°F). Twintex® provides many advantages including excellent fatigue, stiffness and temperature resistance comparable to long fiber thermoplastics, aluminum and/or steel.
Figure 17. RPP75 commingled tows provided by Vetrutex [7]

Figure 18. Set up of thermoplastic pultrusion line
4.2 Experimental Method

The experimental set of thermoplastic pultrusion process used in the present research is shown in Fig. 18. The material was fed to the preheater through four separate square slots of approximately 1 cm$^2$ each. These slots provide some level of pre-tension to the tows as they are fed into the pre heater, in order to minimize their tangling and sagging. The slots help in maintaining constant thickness of the resulting composite. The number of tows varied between 80 to 90 counts. The commingled tows were guided and pulled through the preheating chamber with the help of a pneumatic pulling device placed at the end of the pultrusion line at a speed varying from 0.38 m/min to 1.4 m/min.

The pre heater was maintained at 149 °C (300°F) which was slightly lower than the melting temperature of the polymer (165 °C) to avoid dripping of the polymer while it entered into the heating die. After preheating, the commingled tows were automatically pulled through the heated die. The PP melted in the heated die causing its flow and impregnation of the glass fibers. The die temperature ranged from 177 °C to 220 °C (350 °F to 429 °F) to evaluate an optimum processing window for pultrusion, such that the material would not decompose, and at the same time melting is achieved. The optimum temperature window was established between 199 °C to 216 °C (390 °F to 420 °F). The temperature of the die was controlled by a thermocouple placed near the die cavity. To avoid heat loss and maintain constant temperature, the heating die was insulated with refractory material. Once the material passed through the heating die, it entered the cooling die for consolidation. Heat was extracted from the material instantaneously by chillers containing anti-freeze liquid circulating through the pipes in the chiller.
The effect of processing variables and their interrelationships were investigated.

The parameters investigated were:

1. *Effect of die temperature on pulling force*

   In these experiments, the pulling force was measured for a die temperature of 199, 204, 210 and 216 °C (390, 400, 410 and 420 °F). The line speed was kept constant at 0.38 m/min.

2. *Effect of die temperature on line speed*

   In these experiments, the die temperature was varied as 199, 204, 210 and 216 °C (390, 400, 410 and 420 °F) and variations in the line speed were measured manually using a measuring tape after 1 min travel. In this manner three readings were taken and averaged. The initial line speed was maintained at 0.38 m/min for a temperature of 199 °C (390 °F) and then the change in line speed was measured as temperature increased.

3. *Effect of number of tows on pulling force*

   In this experiment pulling force was measured with respect to the increased number of tows from 80 to 90. Here, the pulling speed and die temperature were kept constant at 0.38 m/min and 204 °C (400 °F) respectively.

4. *Effect of line speed on pulling force*
In this experiment the pulling force was measured by varying the line speed. The line speeds were 0.38, 0.58 and 0.77 m/min. The die temperature was kept constant at 204 °C (400 °F).

4.3 Pulling force measurement

The pultrusion line is equipped with a pulling mechanism that consists of two pairs of polyurethane coated metal plates. In each pair, the plates are driven by a variable speed DC motor through a pneumatic transmission. The machine has been designed for a maximum pulling force of 2000N and a maximum linear speed of 1.7 m/s. To enable measurement of the pulling force, a floating die mechanism was adapted from Carlsson et al [4] as illustrated in Fig. 19.

![Floating die mechanism for measuring force](image)

Figure 19. Floating die mechanism for measuring force
The floating die assembly mounted on a rectangular bar is free to move in the pulling direction. The other rectangular frame is fixed to which two load cells are attached on either side of the tool. When the pullers pull the material, the floating die moves in the pulling direction until it comes into contact with the load cells and stops. This exerts pressure on the load cells whose readings are acquired using Personal DaqViewXL software. The load cells used have sensitivity of 2mV/V and supply voltage of 10V DC.

4.4 Mechanical Testing

Flexural testing was carried out in accordance with ASTM D 790M [26]. The flexural tests were conducted to evaluate mechanical properties of the pultruded part with respect to varying die temperature, line speed and number of tows. The flexural properties were determined by performing a three point bending test. The test was performed on 4 samples of each varying parameter and an average was taken of the four flexural strength values in order to eliminate any error and to make sure the readings are correct. The span length used was 75 mm, and the loading rate was 2 mm/min. The test specimens used were 95 mm long 22.75 mm wide and on average 4.5 mm thick.

4.5 Microscopy and Void Content Measurement

Digital microscopic analysis was performed to evaluate the void content in the pultruded part. Microscopy was performed along the transverse cross section of the composite manufactured using different number of tows.

The percentage of void content was measured using a point fraction method. The test method is based on stereologic principle in which a 9 point grid over a two
dimensional microstructure was placed. Six different placements of the grid on different fields of microstructure were made. Mean and standard deviation of the measurement series was computed. The confidence interval was compared and the results were obtained using a point fraction formula.

4.6 FEM Modeling

Thermal analysis was carried out using a computational scheme to simulate the pultrusion process by implementing suitable boundary conditions. Considering heat transfer from upper and lower platens of the die, convective heat transfer as boundary condition was assumed. The cooling die and composite anisotropy was also taken into account. The model was build using Pro/Engineer Wildfire 3.0 software and was imported to the ANSYS 10.0 software for finite element analysis.
CHAPTER 5 RESULTS AND DISCUSSIONS

5.1 Objective # 1. To develop a thermoplastic pultrusion process using commingled fibers.

The goal of this component was to develop a thermoplastic pultrusion process by using commingled fibers to understand the effectiveness of fiber wetting and consolidation.

Section 2 outlined a typical experimental set up for a pultrusion process. The pultrusion line at UAB was developed along similar lines and Fig. 1 provides a schematic of the components of the pultrusion line. The experiments in the present work were carried out with respect to the following processing variables, namely (a) die temperature (b) line speed (c) number of tows and (d) pulling force.

In the present experimental study, the commingled tows were guided into the preheating chamber at a line speed varying from 0.38 – 1.38 in/min, where they were heated near the melt temperature of 149 ºC (300 ºF) in order to facilitate fast processing. The numbers of tows were varied from 80 to 90 and were pulled through an electrically heated die with the help of a pulling device placed at the end of the pultrusion line. In the heating die, the material was heated above its melting point between the processing temperature of 199 - 216 ºC (390 - 420 ºF) [25] where the PP melted and impregnated the glass fibers, before being consolidated in the cooling die.
Figure 20. Processing cycle for thermoplastic PP/glass fibers

Figure 20 shows the processing cycle for thermoplastic PP/glass commingled fibers. The figure shows that the temperature increases in the preheating zone with increasing time and reaches the melting point of PP, while the pressure remains constant. In the impregnation zone as the temperature reaches the melt temperature, the pressure increases up to certain point along the length of the heating die and remains constant thereafter. The material enters the cooling die; its temperature decreases gradually with time but the pressure remains the same as is in heating die. The pressure decreases rapidly as it nears the exit of the die, hence cooling and consolidating material.

The heating and cooling die used in the experiment is made from tool steel having highly polished cavities. The die cavities are chrome plated in order to reduce friction between the moving material and the cavity walls. Three cartridge heaters are inserted into the die ensuring heating of the material and one thermocouple is placed close to the
die cavity to monitor the temperature. The cooling die is attached separately to the heating die and is perfectly assembled using four steel strips placed at top and bottom of the dies holding them together, ensuring alignment. The cooling die could be detached from the heating die to permit a change in length of the constant cross section of the die. The dimensions of the heating die measures 101.6 x 25.4 x 324 mm and the cooling die measures 101.6 x 25.4 x 324 mm respectively. The cavity of the cooled die has a constant cross section of 4.5 x 22.75 mm. The cooling die is cooled by chillers made from aluminum alloy.

5.2 Objective # 2. To study the effect of different processing variables in thermoplastic pultrusion.

The second objective of the research was to study the processing variables with respect to each other. The processing variables play an important role in the outcome of the product and optimizing these is critical.

The interdependency of the variables which result in optimizing the process are:

1. Effect of die temperature on pulling force: It is necessary to study the pulling force with respect to the die temperature. The pulling force-temperature relationship is necessary to establish the optimum pressure inside the die, which aids in proper wetting and impregnation of the fibers.

2. Effect of die temperature on line speed: It is necessary to study the interdependency of die temperature on line speed because optimum line speed must be maintained during the process to properly consolidate the material, and
also avoid burning of polymer if the line speed happens to slow down with decrease in temperature.

3. Effect of number of tows on pulling force: This interdependency establishes the limits of the die geometry and material strength. High pulling force causes high pressure to build up in the die slot with increased material flow. The die slot area limited to certain square inches, offers high resistance resulting in snapping of the material. Material breakage causes discontinuity in the process, which is not desirable in a manufacturing scenario.

4. Effect of line speed on pulling force: This is necessary to maintain a line speed so that the pulling force does not increase to an undesirable value whereby material breakage inside the die can occur, and bring the process to a halt.

5.2.1 Effect of die temperature on pulling force

Effect of die temperature on pulling force was studied by varying temperature between 199 ºC to 216 ºC (390 ºF to 420 ºF) and keeping number of tows and line speed constant. In this case 90 tows were used and line speed was kept at 0.38 m/min. Figure 21 illustrates that the pulling force increased gradually and then gradually decreased with increase in die temperature over a range of 199 ºC to 216 ºC (390 ºF to 420 ºF). It can be seen that the pulling force tends to increase between 199 ºC to 216 ºC (390 ºF to 420 ºF). In this temperature range the material is not yet in a flow state and has maximum density because the voids are less, and hence requires high force for pulling. Eventually, the material melts and tends to flow back towards the entrance inducing resistance to the material pull resulting in increased pulling force. Once this condition is overcome by increasing the temperature beyond 204 ºC (400 ºF), the material reaches an easy flow
state, the resin viscosity starts lowering initiating free resin flow in the die and providing less resistance to the pulling of material which results in decreased pulling force [24]. Also, with the increase in temperature, pulling forces becomes less dominant as the resin starts to flow freely and also there is no limitation on the polymerization rate [24], and the line is only limited by frictional forces. The major contribution to the pull force is the viscous drag arising when a thin layer of polymer is sheared between the fibers and the die wall. With increase in temperature, the viscous drag with the wall decreases, hence decreasing the pulling force [5]. This is in agreement with the study conducted by Sala et al [23].

Figure 21. Effect of die temperature on pulling force for commingled PP/glass fiber composite
5.2.2 Effect of die temperature on line speed

This study was carried out by changing die temperature and keeping number of
tows constant i.e. 90 in this case. For the die temperature of 199, 204, 210 and 216 ºC
(390, 400, 410 and 420 ºF) for the 90 tow configuration, the values of line speed obtained
were 0.381, 0.386, 0.441 and 0.456 m/min respectively. This suggests that the line speed
increases as the die temperature increases, which is illustrated in Fig. 22. At low
temperatures, the polymer viscosity is high compared to that at higher temperatures. At
low temperature there is increased resistance within the die for material flow, while the
material flows readily at higher temperatures [6, 11]. This suggests that the effect of
temperature on resin viscosity overcomes the effect of polymerization due to decrease in
viscosity in the impregnation zone, hence increasing the line speed. At low temperatures
the line speed is limited by the polymerization time, hence at low temperatures, line
speed is low [24]. This data is comparable to the study conducted by Luisier et al [24] in
which they defined processing in terms of pultrusion line speed and die temperature.
5.2.3 Effect of number of tows on pulling force

The effect of number of tows on pulling force was evaluated for 80, 85 and 90 tows respectively. During this study, die temperature and line speed were maintained constant. The die temperature and line speed were maintained at 204 °C (400 °F) and 0.38 m/min respectively. From Fig. 23, it can be seen that the pulling force increases with an increase in the number of tows. With increase in number of tows, resistance to pulling increases as the die fills out. The density of the material increases since the compaction pressure increases with each tow resulting in filling of the gaps between the tows. The other reason for the increased pulling force is the resin back flow which resists the further movement of the material through the die. This resin back flow is increased with gradual increase in the number of tows fed to the die. With gradual increase in the number of tows, the resin back flow increases and the pulling mechanism needs to
generate more power to overcome this resin back flow pressure. This increases the pulling force. Higher pulling force is required to overcome the resin back flow so that the material can move in the pulling direction. However, preliminary data shows that the pulling force is similar for 85 and 90 tows. With 85 tows the back flow of the resin is higher due to increased heat transfer through the fiber bed because less dense material passes through the die. However in case of 90 tows, the density of the material inside the die is higher. Hence, the heat flow in the material near the die walls is lesser. This causes less resin back flow as compared to the 85 tows and the pulling force is limited by increased frictional resistance between the material and the die wall. It might appears that the pulling force reaches a limiting value for 85 to 90 tows and they exhibit similar pulling force.

Figure 23. Effect of number of tows on pulling force for commingled PP/glass fiber composite
5.2.4 Effect of line speed on pulling force

The study of effect of line speed on pulling force was carried out by keeping die temperature and number of tows constant. For this study, die temperature was maintained at 204 °C (400 °F) and the number of tows used were 90. Figure 24 shows that the pulling force increases with the line speed. Line speed was varied as 0.381, 0.584, 0.774 m/min which resulted in a pulling force of 779.48, 827.23, 941.93 N respectively. With the increase in line speed, shear stresses acting in the thin layer between the material flowing through the die cavity and the die wall generates viscous forces. These viscous forces act in the direction opposite to the pulling direction, hence providing obstacle for material flow. This gives rise to the increased pulling force because the obstacle to pull the material forward in the pulling direction has to be overcome. Hence with increase in line speed, the pulling force is increased. This data agrees with the study conducted by Sala et al [23]. With increase in line speed, the frictional forces between the material and die wall increase, which resists flow of material through the die [24]. The back flow increases, resulting in pressure increase inside the die.
5.3 Objective #3. To evaluate mechanical and physical properties of pultruded thermoplastic composite and compare them with relevant published work.

Flexural testing was carried out in accordance with ASTM D 790M [25]. The flexural tests were conducted to evaluate mechanical properties of the pultruded part. The flexural properties were determined by performing a three point bending test. The span length used was 75 mm, and the loading rate was 2 mm/min. The test specimens were 95 mm long 22.75 mm wide and on average 4.5 mm thick.

5.3.1 Effect of line speed on flexural strength

Figures 25 and 26 illustrate the effect of line speed on flexural strength and modulus for 90 tows. Figure 8 shows that the flexural strength reduced by 57 % as a function of increase in line speed from 0.38 to 1.38 m/min. This is attributed to the reduction in consolidation time and an increase in void content due to improper
impregnation at higher line speeds. At higher line speeds the tows do not attain heat fully at the center of the composite, which results in insufficient melting of the polymer at the center, hence resulting in decreased impregnation time. This causes improper fiber wet-out resulting in void formation affecting mechanical properties adversely. The study conducted by Miller et al [11] shows a similar trend for commingled fibers.

Figure 25. Effect of line speed on flexural strength for commingled PP/glass fiber composite
5.3.2 Effect of die temperature on flexural strength

The effect of die temperature on flexural strength was studied for temperatures of 199, 204, 210 and 216 ºC (390, 400, 410 and 420 ºF) respectively, by using 90 tows in each scenario. The results show the flexural strength varying from 200 to 230 MPa for the above temperatures. A maximum value of flexural strength is attained for material processed at a temperature of 204 and 216 ºC (400 and 420 ºF). Considering that the material is in a non-flow state till 204 ºC (400 ºF), the result is valid because the density of the material is high due to fewer voids in the composite. The material shows decrease in flexural strength at 210 ºC (410 ºF). At this temperature the molten resin is in its initial flow state or not dispersed uniformly in the reinforcing fibers leading to fiber agglomeration and formation of voids causing deterioration of flexural properties. When temperature increased to 216 ºC (420 ºF) the flexural strength showed the highest value.

Figure 26. Flexural stress versus strain curve showing maximum modulus at minimum line speed 0.38 m/min
This can be attributed to an increase in resin flow, which in turn results in maximum impregnation due to a lower matrix viscosity with increasing temperature [12]. The results shown in Fig. 27 and 28 provide an indication of optimum temperature for processing.

Figure 27. Effect of temperature on flexural strength for commingled PP/glass fiber composite
5.3.3 Effect of number of tows on flexural strength

Figures 29 and 30 illustrate the effects of number of tows (80, 85 and 90 respectively) on flexural strength and flexural modulus respectively, with die temperature and line speed being constant at 204 ºC (400 ºF) and 0.38 m/min respectively. The flexural strength is observed to increase with increasing number of tows as illustrated in Fig. 29 and it increases by 17% when increased from 80 to 90 tows. This is attributed to an increase in volume fraction of the fibers. As the fibers are the primary load bearing
elements, an increase in their volume fraction increases the mechanical strength of the composite, provided there is adequate impregnation.

Figure 29. Effect of number of tows of flexural strength for commingled PP/ glass fiber composite

Figure 30. Effect of number of tows of flexural modulus
5.3.4 Microstructural analysis

Figure 31 shows typical microphotographs of the cross section of pultruded glass/PP bars for 80, 85 and 90 tows respectively. The effect of fiber agglomeration on the impregnation state was evaluated. As seen from the figure, there are several voids due to incomplete impregnation. As the number of tows are increased from 80 to 90 counts, the void content reduces. This is because the increase in number of tows fills the free spaces in the mold. This leads to increase in compaction pressure required for impregnation, and allows resin to flow readily that enables increased wet-out of the tows. A study conducted by Kim et al [14] shows similar trends of impregnation on pultruded composites manufactured using commingled tows. According to their study, the fiber agglomeration size is the most important parameter in enhancing the degree of impregnation. Smaller agglomerates enable faster processing with proper impregnation, because they induce high pressure. The taper geometry also affects degree of impregnation. In general, it has been found that a smaller taper angle and a longer taper zone, the part attains better impregnation. The other reason for improper impregnation is the resin back flow from the die which causes void formation due to lack of sufficient resin to wet the fibers [5].

Since the main problem with a thermoplastic matrix is the difficulty of impregnation due to its high viscosity, it becomes imperative to study wetting of the fibers by measuring void volume fraction. The void volume fraction is given by [5, 26]:

\[ V_f + V_m + V_v = 1 \]  \[7\]
where, $V_f$ is the fiber volume fraction; $V_m$ is the matrix volume fraction; and $V_v$ is the void volume fraction.

![Microphotographs](image1.png) ![Microphotographs](image2.png) ![Microphotographs](image3.png)

(a) (b) (c)

Figure 31. Microphotographs of impregnated pultruded product using RPP75 (a) with 80 number of tows (b) with 85 number of tows (c) with 90 number of tows

The volume void fraction was calculated using a point fraction method as described in Chapter 3. The point fraction method provides a range of percentage of voids in the microstructure and gives information about the degree of impregnation. With increase in number of tows from 80 to 90, the void content decreased from 54% to 21% for the upper limit, and 24% to 12% for the lower limit as listed in Table 1.

The void results indicate that additional tows pulled through the die may further reduce the air pockets; and hence voids in the composite. However, the present process offers limitations in terms of die geometry. A tapered die enables packing of additional tows to minimize void content.

The density for the pultruded profile made from 80, 85 and 90 tows respectively was calculated to estimate the void content in the composite. The samples used for this were 95 mm long 22.75 mm wide and on average 4.5 mm thick.
The density values as stated in Table 2 show that there is increase in density as the number of tows are increased from 80 to 90. When the density of the composite made of 80 tows was compared to the density of composite made of 90 tows, it was found that there is increase in the density of 90 tows by 10.23%. This increase in the density can be attributed to the decrease in void content due to increased packing factor of the material inside the die. This is also clear from the microphotographs shown in Fig. 31 and void content results stated in Table 1.

Table 1. Void fraction values

<table>
<thead>
<tr>
<th>Quantities</th>
<th>80</th>
<th>85</th>
<th>90</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean, $\bar{X}$</td>
<td>3.6</td>
<td>2.5</td>
<td>1.5</td>
</tr>
<tr>
<td>Standard Deviation, S</td>
<td>1.63</td>
<td>1.04</td>
<td>0.54</td>
</tr>
<tr>
<td>Standard Error, S.E</td>
<td>0.66</td>
<td>0.42</td>
<td>0.22</td>
</tr>
<tr>
<td>95% C.I of P.F</td>
<td>3.6 ± 1.32</td>
<td>2.5 ± 0.84</td>
<td>1.5 ± 0.44</td>
</tr>
<tr>
<td>95% C.I for void fraction</td>
<td>0.25 – 0.54</td>
<td>0.18 – 0.36</td>
<td>0.11 – 0.21</td>
</tr>
<tr>
<td>% Void volume fraction</td>
<td>25 – 54</td>
<td>18 – 36</td>
<td>11 – 21</td>
</tr>
</tbody>
</table>

Table 2. Density Measurement

<table>
<thead>
<tr>
<th>Number of tows</th>
<th>Weight (gm)</th>
<th>Volume (mm³)</th>
<th>Density (g/mm³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>80</td>
<td>10.98</td>
<td>9725.6</td>
<td>0.001128</td>
</tr>
</tbody>
</table>
### Number of tows

<table>
<thead>
<tr>
<th>Number of tows</th>
<th>Weight (gm)</th>
<th>Volume (mm³)</th>
<th>Density (g/mm³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>85</td>
<td>11.56</td>
<td>9725.6</td>
<td>0.001188</td>
</tr>
<tr>
<td>90</td>
<td>12.20</td>
<td>9725.6</td>
<td>0.001254</td>
</tr>
</tbody>
</table>

5.4 Objective # 4. To develop finite element analysis model to describe the heat transfer of the material in the die.

Computer modeling was carried out for evaluating the one dimensional temperature distribution and to validate the experimental process by comparing the output temperature of the composite. This also provides an insight to the optimum processing window in terms of temperature.

The modeling work for commingled fibers reported in many papers [6, 9, 27, and 28] has emphasized on the numerical modeling pertaining to impregnation and consolidation of the material. There is not much focus in the heat transfer modeling of commingled fibers using FEM. Several modeling studies of pultruded profiles have focused on thermoset resins, but not much work has been done using thermoplastic resins, particularly commingled fibers [33, 34, 38, 39].

In this study, three dimensional FEM based steady state heat transfer modeling using ANSYS 10.0 has been conducted. This model helped in predicting the temperature profiles of various zones of the die and the final pultruded part.

The geometry of the die is constructed using Pro/Engineer Wildfire 3.0. The solid model is imported to ANSYS 10.0 for further analysis. The dimension of the heating die measures 101.6 x 25.4 x 324 mm and of the cooling die measures 101.6 x 25.4 x 171.5
mm and both dies are fastened together by four metal strips. The die is equipped with two rectangular slots for material passage having a width of 23 mm and thickness of 4.5 mm approximately which runs throughout the length of the die on either side of the center line of the die. The die also shows four rounded slots for the heaters of diameter 6.35 mm and length of 76.2 mm as shown in Fig. 32. During the experimental study the die was equipped with a graphite gasket between the upper and lower die which was approximately 1 mm thick in order to eliminate any pressure loss and to avoid material squeezing out from the side walls of the die. However, the model does not take the gasket into account. The die was considered as whole (perfect seal) since it was assumed that the gasket does not have much influence on the resulting product.

Figure 32. Finite element model of the die
The die is assigned with properties of thermal conductivity of 25.57 W/m-k (177.3 BTU-in/hr-ft²-°F), density of 7750.37 Kg/m³ (0.280 lb/in³) and specific heat of 461.8 J/kg-k (0.1103 BTU/lb °F). The material is assigned a thermal conductivity of 0.346 W/m-k (2.4 BTU-in/hr-ft²-°F) and density of 1107.19 Kg/m³ (0.04 lb/in³) [52].

A fine mesh was adopted comprising 240373 nodes and 92330 elements as shown in Fig. 33. The heating die was meshed considering SOLID 87 elements. The SOLID 87 is a 10 noded element and rounded edges and curved geometry can be accounted for in the model. The cooling die and the material are meshed using SOLID 90 elements. The SOLID 90 element is a 20 noded element with mid edge node. It has 3-D thermal conduction capability and provides compatible temperature shapes that are well suited to model curved boundaries. For simulating contact surfaces between the material and die walls, an 8 noded CONTA 175 element has been used.
The initial boundary condition (temperatures) considered ambient temperature (24 °C i.e. 75 °F) for convective mode (radiation neglected) and the final temperature of the die which was measured during the experiment with a thermocouple placed at the left hand side corner of the vertical face of the heating die is 204 °C (400 °F). This is achieved by 3 cartridge heaters each of 76.2mm in length and 6.35mm in diameter. But this is not a true reading of the temperature because it is measured far from the heating zone. In order to get the actual temperature at the heating zone, the heat flux generated from each of the 240V and 300W was considered which was estimated to be 51W. Also, heat flow from the cooling die is estimated as -22W on basis of the experimental output temperature of the composite.
The initial boundary condition for the material is taken to be 149 °C (300 °F) because the material is preheating, and enters directly into the die through preheater.

Figure 34 shows heat distribution along the length of the die. The analysis shows that the actual temperature at the entrance is 212 °C (414 °F) as compared to the set temperature of 204 °F (400 °F) measured at left vertical face of the die. Hence, it can be ascertained that the actual processing has been occurred at 212 °C (414 °F). So the actual length of heating zone temperature of 212 °C (414 °F) through which the material passes is only 81mm for a period of 12 s with a line speed of 0.38 m/min. This suggests that the material spends less time in the actual processing zone temperature. After it enters the temperature zone which is below the minimum temperature required for processing which is 199 °C (390 °F) and continues to travel till it reaches cooling zone.

The longitudinal core section of the composite is as shown in Fig. 35. Figure 35 shows that the material when traveling through different sections of the die is conducting heat adequately through its core. However, the actual problem lies in the heat loss through the die which is unable to maintain the required processing temperature through its length. The die could maintain the processing temperature up to 1/4th of its length. For adequate melt and impregnation, the processing zone has to be at least 3/4th of the die length, which is not the case. So the material when enters the die melts, but fails to impregnate properly throughout the length of the die hence creating voids in the core.
Figure 34. Heat transfer distribution through the die
Figure 35. Longitudinal section of the die through the material

Figure 36 shows the distribution of heat along the length of the material. It shows the material entering the die at 168 ºC (335 ºF), while the preheating temperature was 149 ºC (300 ºF). This increase in the material temperature before entering the die may be due to the reverse heat transfer taking place from heating die to the material as is seen in Fig. 34 thereby increasing the temperature of the material. The output temperature of the composite is 14 ºC (58 ºF) as compared to the experimental temperature range of 15 ºC to 23 ºC (59 ºF to 73 ºF).
Figure 36. Heat transfer distribution through the composite
CHAPTER 6 SUMMARY

1. The present research on thermoplastic pultrusion has focused on finding major processing parameters and appropriate process windows for PP/glass thermoplastic commingled fibers.

2. The effect of line speed on flexural strength and modulus accounted for 57% reduction with increased line speed from 0.38 to 1.38 m/min. However, to achieve high flexural strength the speed has to be maintained at 0.38 m/min, which is too slow for commercial manufacturing.

3. The process parameter relationship was studied to understand the interdependency of variables on each other. Effect on pulling force was studied by varying die temperature, pulling speed and number of tows respectively. It was found out that the pulling force increased with increase in line speed and number of tows and decreased with increase in temperature. With increase in line speed from 0.381 to 0.774 m/min pulling force increased from 779.48N to 941.93N. It shows that there is increase of 18% which is attributed to increase in frictional forces between die wall and material. With increase in number of tows from 80 to 90, the pulling force increased due to increase in compaction pressure with each introducing tow resulting in filling the gaps. When temperature was increased from 199 to 216 °C (390 to 420 °F), the pulling force increased between 199 to 216 °C (390 to 400 °F) and then decreased. This is attributed to the maximum
density of the material in the heating zone in the undecomposed state, before reaching 216 °C (420 °F). It was found out that the line speed increased from 0.381 to 0.456 m/min with increase in temperature from 199 to 216 °C (390 to 420 °F) because of the high viscosity of the polymer at low temperature, as compared to high temperature.

4. A 13% increase in flexural properties between the lower and upper limits of the temperatures, with regard to the die temperature.

5. When the number of tows increased from 80 to 90, the flexural strength increases by 17%. The increase in fiber content contributes effectively in increasing mechanical properties.

6. Three dimensional FEM modeling has been carried out to predict the melting and impregnation of the fibers simulating heat distribution along the length of the die and composite. It simulates temperature distribution along the length of the die and composite as it travels through the pultrusion line. The model takes into account the prescribed boundary conditions. The model helped in analyzing temperature distributions and illustrates how the composite temperature varies at any location in the die.

7. Very little efforts have been made in designing the die. By optimizing the die that higher pulling speeds and packing of more tows could be possible. The present die has limitations in terms of higher pulling speeds, and fractures the material if the pulling speed is increased beyond 1.38 m/min.
CHAPTER 7 CONCLUSIONS

1. The pultrusion process for RPP75 PP/glass commingled fibers was successfully optimized.

2. Die design was not modified, but with a modified design, higher speeds can be obtained. Modifications can be made at the taper entrance and along the taper length. Smaller taper entrance and longer taper length could bring about improved impregnation. On the other hand, it may obstruct feeding of the material as it will create high shear rate flow in the direction opposite to pulling.

3. The presence of void content can be linked to lack of proper impregnation due to fiber agglomeration size. RPP60 provides better impregnation and ease of processing due to small agglomeration size as compared to RPP75. In addition to voids, sloughing is often observed.

4. Changes in flexural strength values show that the flexural properties are very susceptible to variations in the process parameters within the investigated ranges. A suitable combination of all these process parameters can provide a desirable result.

5. The comparison between the experimental work and simulation shows that the process shows similar output temperature that of the part. Hence, it can be concluded that the FEM modeling achieved a reliable quantitative estimate of temperature distribution in both the die and the pultruded part.
6. The FEM modeling can be of use to minimize experiments, thus saving time and money while optimizing the thermoplastic pultrusion process for ultimate performance. It could be helpful in providing essential information on tooling and process design for the pultrusion.

7. The FEM modeling could be further used for investigating the transient phase, which will be helpful in studying variable void content during manufacturing.
LIST OF REFERENCES


1980.


