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# Designing with glass reinforced engineering thermoplastics

*P.J. Bates, V.T. Bui  
Department of Chemistry and Chemical Engineering  
Royal Military College of Canada  
P.O. Box 17000 STN Forces  
Kingston, ON K7K 7B4  
Canada*

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## 1. ABSTRACT

Glass reinforced thermoplastic materials are used in a wide variety of civilian and military applications. They are often chosen over metals and glass reinforced thermosetting materials for reasons of lower cost combined with equal or superior part performance. The higher production rates and part function integration capability associated with injection moulding, allow these thermoplastic based compounds to deliver a lower part cost than other often less expensive metallic or thermosetting materials. Prior to profiting from these cost advantages, the designer of glass reinforced injection moulded parts must first confront a number of issues : orientation of the glass fibre reinforcement causing anisotropy of all material properties, weldlines leading to areas of low mechanical strength, temperature limitations of engineering thermoplastics, as well as high equipment costs due to the high pressures required to manufacture large parts. Recently, a number of new glass reinforced engineering thermoplastic materials have been developed to address some of these problems. Semi-aromatic polyamides, long fibre reinforced compounds, and glass mat reinforced thermoplastic are several examples.

## 2. INTRODUCTION

The ability to produce a correctly functioning part using glass reinforced thermoplastics will depend largely on the nature of the thermoplastic polymer matrix, the form of the glass reinforcement and the processing technique used to manufacture the part. Each of these aspects is discussed in the following sections with emphasis on several developments within that particular area.

## 3. THERMOPLASTIC POLYMER

### 3.1 Thermoplastic Background

From their humble beginnings following the second World War, glass reinforced thermoplastics have grown into a 700 million pound market [1]. Although the total volume of glass reinforced thermoplastics is still smaller than that of glass reinforced thermosetting resins, the former present many unique advantages.



Thermoplastic polymers differ from thermosetting resins in that thermoplastics are processable in a relatively high temperature melt state and then will solidify without a chemical reaction upon cooling below a characteristic temperature. Versus thermosetting resins, this generally allows thermoplastic polymers to have :

- **lower processing cycle time** : The major component of most thermoset resin or thermoplastic polymer processing cycles is the time required for material solidification. The solidification of thermoplastic polymer is predominantly linked to the rate of cooling of the material which is measured in seconds. The solidification of thermoset resin is related to the rate of the cure or cross-linking reaction which is often measured in minutes.
- **increased ductility** : In general, thermoplastic polymers have a higher elongation at break than thermoset resins. This is often attributed to the absence of chemical cross-linking within the thermoplastic matrix.
- **unlimited shelf life** : The absence of chemical cross-linking in thermoplastic polymers allows these materials to be stored for very long periods of time without a change in molecular weight. The shelf life of uncured thermoset resins is limited.
- **reprocessing of scrap** : Although often of secondary importance, clean thermoplastic scrap, resulting from industrially rejected parts or trimming, can be reground and recycled in many processing operations. This is often performed at regrind levels of the order of 20 weight % without significant changes in mechanical or physical properties of the manufactured part [2]. Recycling of industrial thermoset scrap is more difficult due to the chemical cross-links which prevent the cured thermoset resin from flowing.

It is possible to reinforce most thermoplastic polymers. Semi-crystalline polymers have however achieved the most industrial success due, in large part, to their superior thermomechanical properties versus those of purely amorphous polymers.

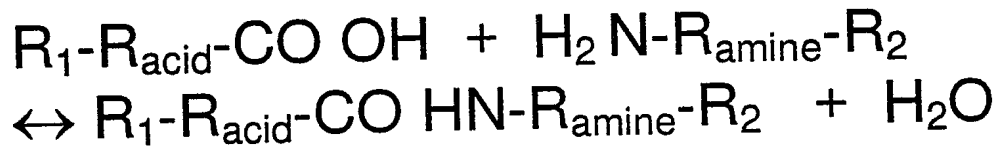
Amorphous polymers exhibit one principal characteristic temperature called the glass transition temperature. The glass transition temperature is associated with movement of various segments of the polymer chain. Above the glass transition temperature, amorphous polymers display little mechanical strength and are, under these conditions, considered to be in the melt state. Semi-crystalline polymers exhibit both a glass transition temperature and a melting temperature. The latter is associated with the breakup of the ordered crystalline regions. Under certain short term loading, reinforced semi-crystalline polymers may be used at temperatures almost up to their crystalline melting point.

Two of the most common semi-crystalline thermoplastic polymers used industrially with reinforcements are polypropylenes and polyamides (nylons). Polypropylene is chosen for its low cost, high fluidity and chemical inertness. Nylon is used for good adhesion to the reinforcement and its relatively high glass transition and melting temperatures. It is impossible to cover the axes of development of these two common thermoplastic polymers in this short article. Instead, it is interesting to focus on recent developments in the polyamide family.

### 3.2 Polyamide - axes of development

Polyamide was discovered in the 1930's by Dupont. This family of polymers is characterized by a relatively high strength and stiffness which makes it well suited to many engineering applications [3]. This is mainly due to the hydrogen bonding that occurs between amide linkages on adjacent polymer chains. These amide linkages also improve the mechanical properties of reinforced polyamides by facilitating the chemical coupling of the polymer to the surface of the reinforcement.

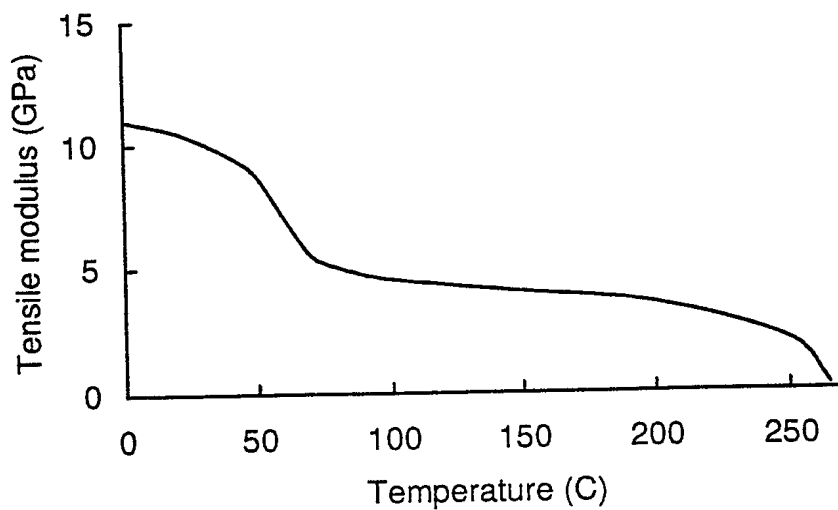
Polyamide is polymerized by a condensation reaction between diacid and diamine monomer species. **Figure 1** shows the stepwise condensation of two polyamide chains resulting in water being liberated :



**Figure 1** - Condensation reaction between a diacid and diamine monomer.

Of the different types of polyamides that exist, polyamide 66 is one of the most common. The 66 denotes the presence of 6 carbon atoms on both the diacid and diamine monomer species. Polyamide 66 is referred to as an aliphatic polyamide as the carbons are in a linear chain.

Polyamide 66 has a glass transition temperature of 50°C and a crystalline melting temperature of 260°C [4]. When reinforced with 30% glass fibre, a typical elastic modulus versus temperature profile is given in **Figure 2** [5]. The initial large drop in modulus occurs near the glass transition temperature of 50°C. The modulus drops off to near zero at the crystalline melting temperature of 260°C:



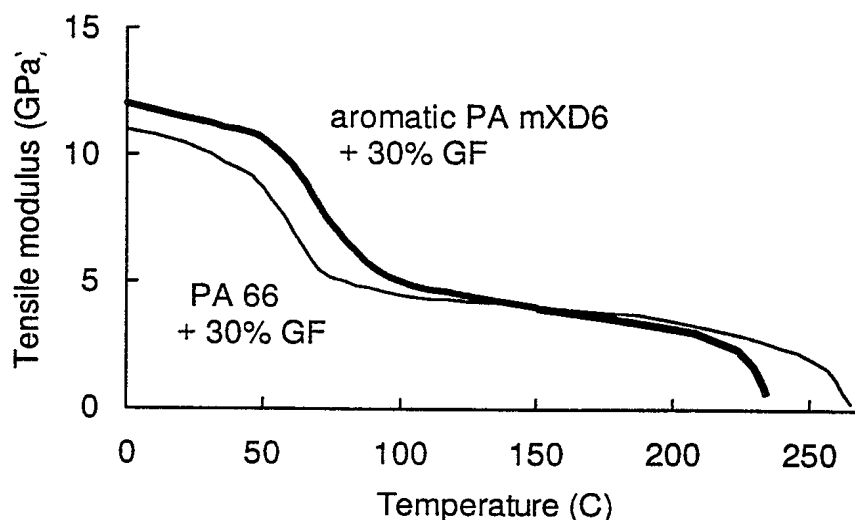
**Figure 2** - Tensile modulus as a function of temperature of a 30% glass reinforced polyamide 66 compound [5].

In order to expand the market of aliphatic polyamides towards higher temperature applications, it is necessary to increase the glass transition temperature and/or the crystalline melting temperature of the polymer. In the last 15 years a number of new polyamide polymers have been developed by incorporating either aromatic diamine or diacid monomers (Table 1). These semi-aromatic polyamides (as opposed to fully aromatic polyamides such as Kevlar®, Nomex® ...) exhibit higher glass transition temperatures, and higher or lower melting temperatures depending on the crystalline structure and amide/amide interactions [3].

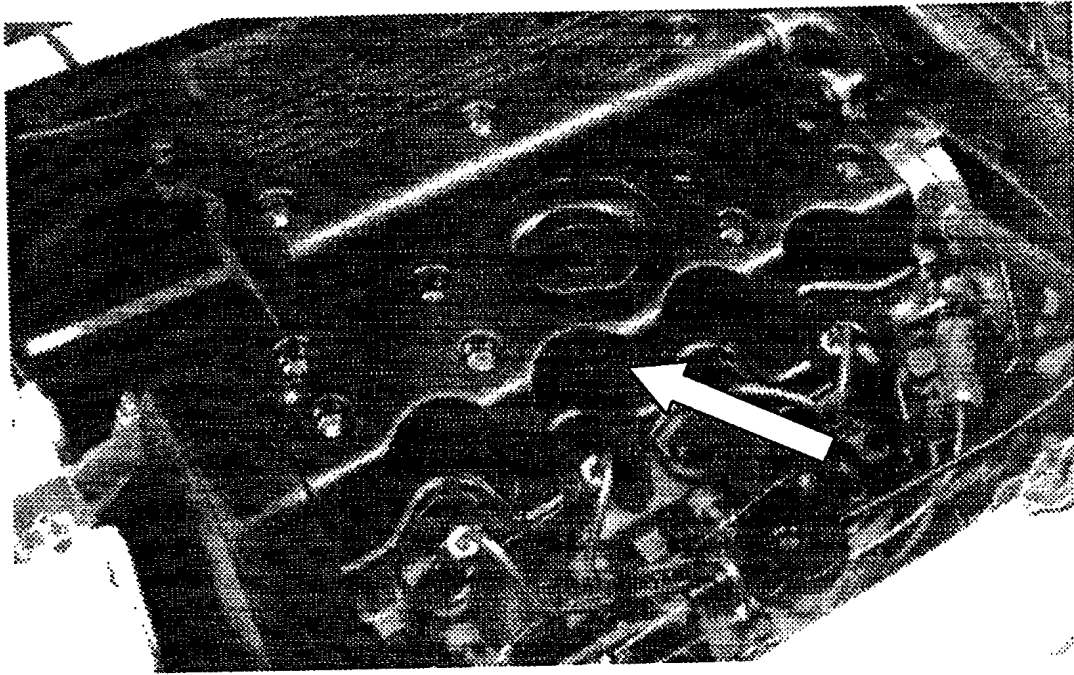
Commercial trade name	Polymer producer
AMODEL®	Amoco (USA)
GRIVORY®	EMS Chemie (CH)
IXEF®	Solvay (B)
RENY®	Mitsubishi Gas Chemical (J)
ULTRAMID® T	BASF (D)
ZYTEL® HTN	Dupont (USA)

**Table 1** : Partial list of commercialized semi-aromatic polyamides

An example of such a semi-aromatic polyamide is polyamide mXD6 which is polymerized using an aromatic diamine. A comparison of the elastic modulus of comparably reinforced polyamide 66 and mXD6 is shown in Figure 3 [2]. The semi-aromatic polyamide's higher elastic modulus in the temperature region below 125°C is due to its higher glass transition temperature. The superior stiffness of the polyamide mXD6 in this temperature region has allowed it to displace aliphatic polyamide 66 in many applications such as the diesel engine valve cover shown in Figure 4. The central bolting of this part to the engine block necessitated excellent high temperature stiffness along the cover edge to prevent oil leakage.



**Figure 3** - Tensile modulus as a function of temperature for a 30% glass reinforced polyamide 66 compound and a 30% glass reinforced semi-aromatic polyamide mXD6 compound [2].



**Figure 4** - Peugeot TUD3 motor valve cover moulded by Mecaplast (F) using semi-aromatic polyamide mXD6 reinforced with 50% glass fibre (IXEF<sup>®</sup> 1022 from Solvay).

#### 4. GLASS FIBRES

Reinforcing fibres used in the thermoplastic industry can be divided into inorganic (glass, boron ....) and organic (carbon, aramid ....) groups. On a mass basis, inorganic glass fibres make up over 99% of the reinforcement used currently in thermoplastics [1].

##### 4.1 Glass fibre parameters

Glass fibres, like any reinforcement, play a predominant role in determining the mechanical and physical properties of the composite material and of the finished part. They also influence the processing technique that can be used. The glass fibre parameters affecting properties are :

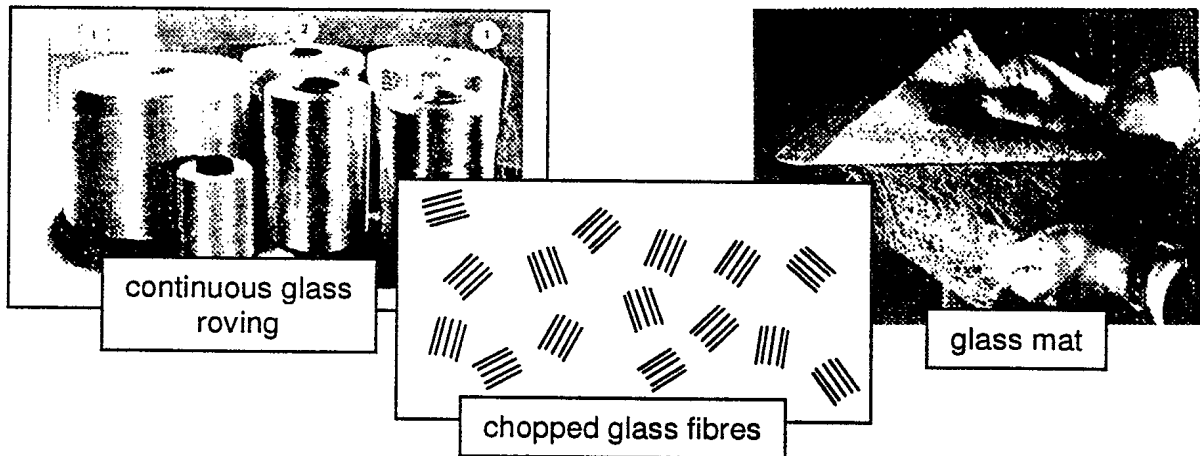
- **glass composition** : There exist several different glass compositions denoted by letters : E - general purpose; D - improved dielectric properties; R or S-improved mechanical properties. E type glass is used almost exclusively with reinforced thermoplastics [6].
- **glass fibre sizing** : Sizing is the chemical coating applied to the individual glass monofilaments immediately after they are formed. Sizing chemistry is tailored for the polymer and the processing technique. Sizing permits coupling between the polymer matrix and the reinforcement, adhesion among the individual glass monofilaments prior to processing, and lubrication between the monofilaments and any solid surfaces encountered during handling or processing [6]. The sizing composition is generally a closely guarded secret of glass fibre manufacturers as it is one of the major methods of product differentiation.



- **glass monofilament diameter** : The diameter of the individual monofilaments used in glass fibres is usually in the range of 10 to 24  $\mu\text{m}$  [6, 7]. The actual diameter of a given glass fibre product is a compromise between mechanical properties, price, and ability to produce the glass product. To a certain point, smaller diameter monofilaments improve the strength of reinforced thermoplastics by increasing the surface area for a given reinforcement level [7]. Conversely, larger diameter monofilaments are generally lower in cost due to the higher production throughput during the fibre manufacture.
- **reinforcement form** : Glass fibres are available in several different forms (**Figure 5**) : *rovings* composed of several hundred to several thousand near parallel continuous monofilaments, *chopped fibres* composed bundles of parallel monofilaments of length 5 mm to 30 mm, and *glass mat*. Glass mat can be composed of randomly oriented fibre (shown in Figure 5), woven rovings or dispersed chopped fibre.

The reinforcement form will dictate, to a large extent, the type of processing that can be used to make the finished part. In addition, the reinforcement form will also determine how the reinforcement should be combined with the polymer prior to final processing. This necessary combination step is often referred to as compounding.

Currently most of the glass fibre used with thermoplastic polymer is of the chopped fibre form. The compounding of the polymer with the chopped fibre is done predominantly using a single or twin screw extruder (section 4.2). The resulting compound is then usually processed using the injection moulding process (section 5.2).



**Figure 5** - Illustration of the various forms of glass fibre that can be used with glass reinforced thermoplastics.

## 4.2 Thermoplastic-Glass Fibre Compounding

Compounding of the glass fibre and the thermoplastic polymer is a necessary step prior to final part processing. The compounding of thermoplastic polymers with glass fibres is more difficult than compounding similar reinforcement with thermosetting resins. This is due to the lower viscosity of the thermoset resins prior to curing than that of most thermoplastic polymers in the melt state.

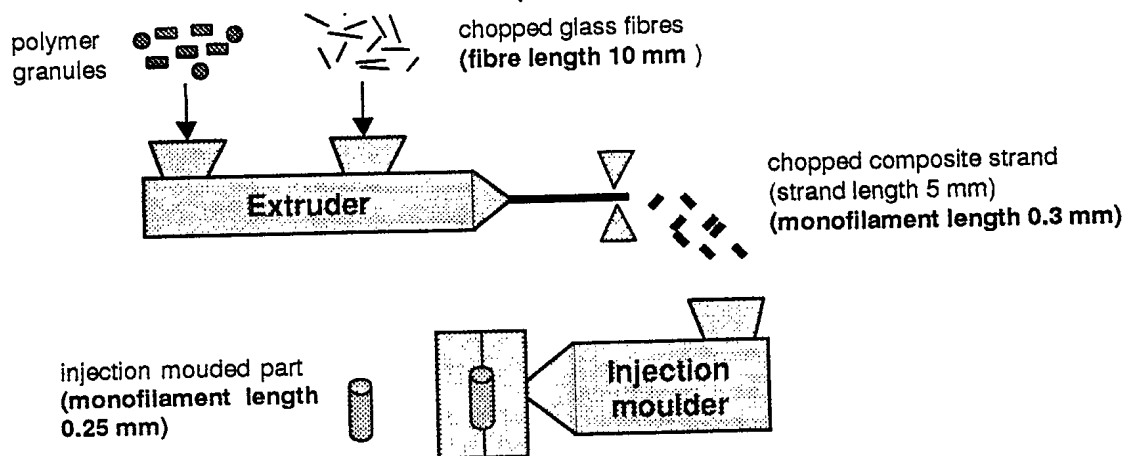
Many different techniques are used to compound thermoplastic polymers and glass fibres. They are often inspired from those used with thermosetting resins. Some of these techniques and their axes of development are discussed in the following sections.

#### 4.2.1 Compounding of thermoplastic polymer in the melt state

##### 4.2.1.1 Extrusion compounding

The most common technique used to compound glass fibres and thermoplastic polymer is extrusion compounding [8]. The process is shown schematically in **Figure 6**. It involves using the shearing action of an extruder or static mixer to distribute and disperse glass mono filaments throughout a polymer melt. The fibres are introduced into the extruder either in chopped or roving form. A *discontinuously reinforced composite strand* is extruded and chopped to form pellets which are then used in downstream processing such as injection moulding.

Extrusion compounding is an economical means of mixing most polymers with glass fibres as well as other additives. Its major disadvantage is the degradation of glass monofilament length during the extrusion process. For this reason, extrusion compounding is often referred to as short fibre compounding and the resulting material is called short fibre compound. The average residual monofilament length and length profile in the composite strand depend on extrusion conditions [9]. However, a number average value of the order 300  $\mu\text{m}$  is typical. The glass monofilaments will undergo further degradation during the final processing.

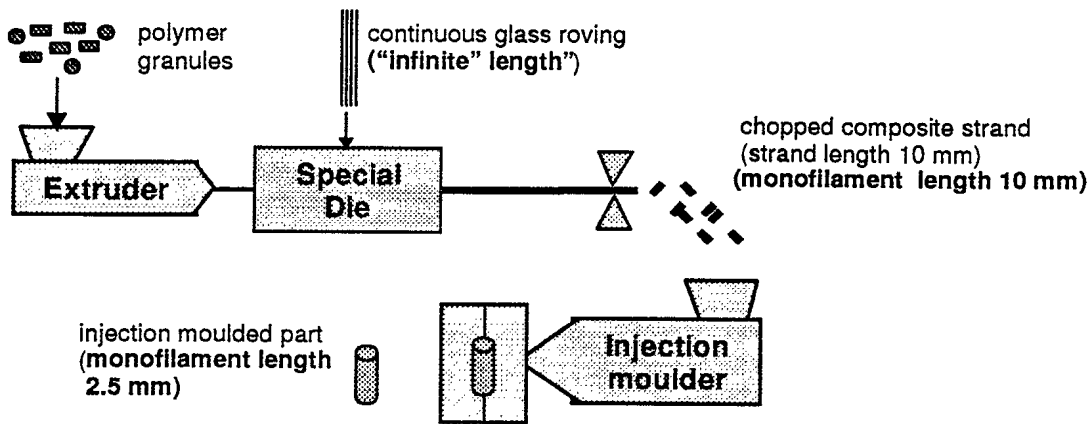


**Figure 6** - Schematic diagram of a typical extrusion compounding operation using chopped glass fibres. The compounding is followed by injection moulding processing.

##### 4.2.1.2 Pultrusion compounding

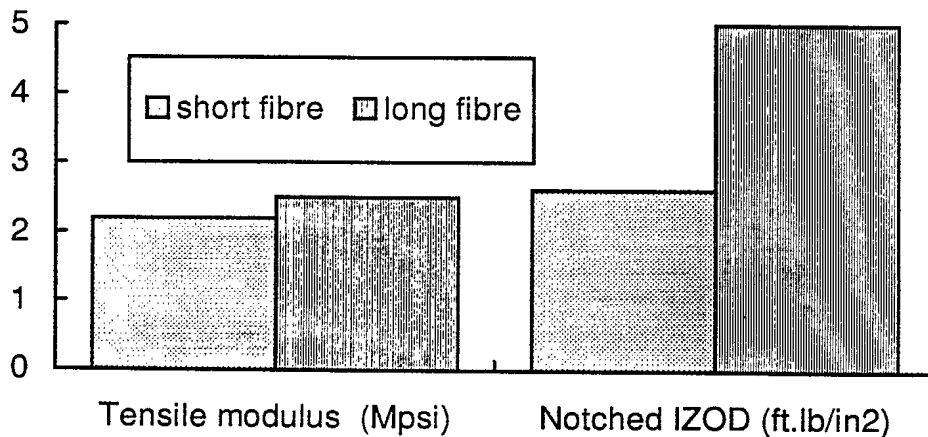
A number of alternative compounding techniques have been developed to minimize the fibre length degradation during compounding [10, 11, 12]. Most of these techniques use glass fibres in roving form. The roving is often impregnated by molten polymer in a specially designed die (**Figure 7**). A *continuously reinforced strand* is then pulled from the die and then chopped to form pellets for downstream injection moulding processing.

Although monofilament length degradation will still occur during the injection moulding process, the average monofilament length in the finished part is generally higher than that of parts made using short fibre compounds. For this reason, this pultrusion compounded material is often referred to as long-fibre compound.



**Figure 7** - Schematic diagram of a typical pultrusion compounding operation using glass roving. The compounding is followed by injection moulding processing.

The ability to achieve higher residual glass monofilament lengths in the injection moulded parts translates to improved mechanical properties. **Figure 8** shows that notched IZOD impact resistance of 50% glass reinforced polyamide 66 is improved dramatically when long fibre compounds are used [13]. This increase in impact strength is obtained without a drop in modulus. This latter point is important as it is also possible to improve the impact performance of glass reinforced compound via rubber addition. This results, however, in a decrease in modulus.



**Figure 8** - Comparison of the room temperature tensile modulus and notched IZOD impact properties of polyamide 66 reinforced with 50% (wt) glass short and long fibres [13].

Several long fibre materials are currently commercially available : Verton (LNP) and Celstran (Hoechst Celanese).

#### 4.2.1.3 Compression/lamination

Extrusion and pultrusion compounding are used to manufacture compounds containing either discontinuous or continuous monofilaments. In order to manufacture reinforced thermoplastic polymer compound in sheet form, a technique similar to that used in making thermoset sheet moulding compound (SMC) [4, 14] is often used. In such a process, glass mat is combined with several molten polymer sheets. The composite mixture passes through a lamination process where an applied compressive stress forces the molten polymer to penetrate the mat. The impregnated mat is then cooled and cut into blanks. This type of composite material is generally processed into the final part in a compression moulding operation [15].

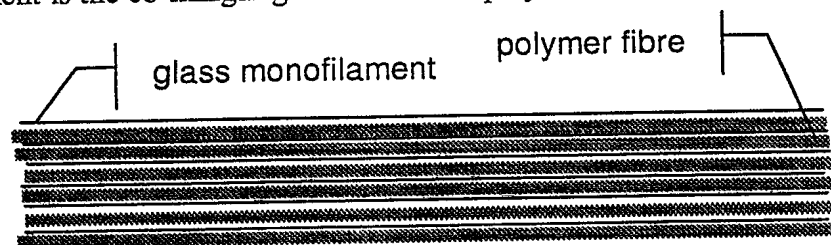
AZDEL<sup>®</sup> is the trade name of such a polypropylene based glass reinforced mat. It is manufactured by a joint venture of GE and PPG.

#### 4.2.2 Compounding of thermoplastic polymer in the solid state

The techniques described in section 4.2.1 all involve combining a molten thermoplastic polymer with reinforcement and applying pressure to force the two components into intimate contact. It is also possible to combine polymer in the solid state with the reinforcement, and then use the heat and pressure during final part processing to achieve intimate contact of the two materials.

##### 4.2.2.1 Co-mingling

One of the most recent techniques for compounding of thermoplastic polymer and glass reinforcement is the co-mingling of continuous polymer and glass fibres (**Figure 9**).



**Figure 9** : Schematic representation of a co-mingled polymer and glass fibre composite.

A new example of such a co-mingled composite material is manufactured by Vetrotex under the trade name TWINTEX<sup>®</sup> [16]. One advantage of co-mingled composite strands versus pultrusion compounded strands is flexibility. Co-mingled material can be woven or assembled as unidirectional reinforcement prior to final part processing. A number different processing operations are under evaluation with this material. They include compression moulding, vacuum forming and filament winding.

#### 4.2.2.2 Powder impregnation

Prior to the introduction of co-mingled composite fibres, an alternative technique was often used to obtain flexible composite strands. It involved incorporating polymer powder between reinforcing monofilaments. This compounding operation was often performed in a specially designed fluidized bed [17, 18]. This material is frequently filament wound over metal pressure vessels to increase their burst pressure [19].

## 5. PROCESSING TECHNIQUE

### 5.1 Introduction

The processing technique used to manufacture the finished part will depend largely on the form of the glass fibre as well as the thermoplastic-glass compounding technique. These combinations have been briefly discussed in the previous sections and are summarized in **Table 2**.

Glass fibre form	Compounding technique	Typical final processing
chopped fibre	extrusion	<ul style="list-style-type: none"><li>• injection moulding</li></ul>
continuous roving	pultrusion	<ul style="list-style-type: none"><li>• injection moulding</li></ul>
	co-mingling	<ul style="list-style-type: none"><li>• filament winding</li><li>• compression moulding</li><li>• vacuum forming</li></ul>
	powder impregnation	<ul style="list-style-type: none"><li>• filament winding</li></ul>
mat	compression/lamination	<ul style="list-style-type: none"><li>• compression moulding</li><li>• vacuum forming</li></ul>

**Table 2** : Typical final processing as a function of the glass fibre form and compounding.

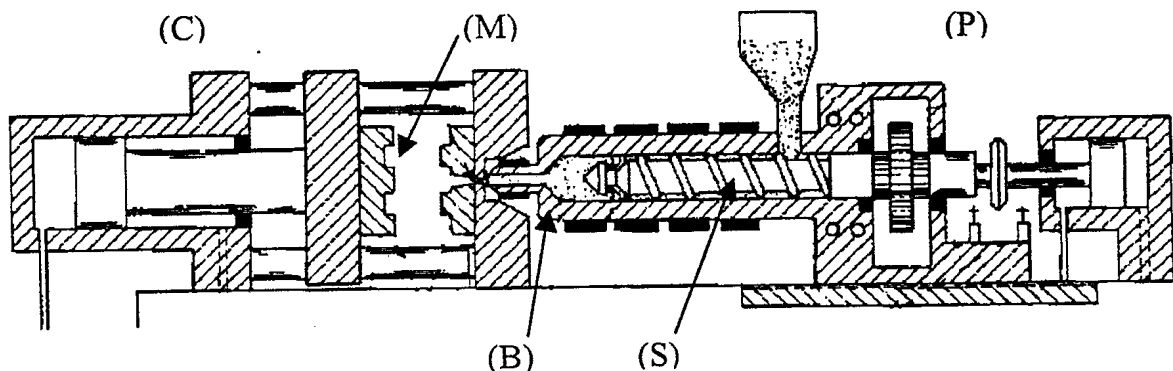
Although a wide variety of fibre forms, compounding and processing techniques exist, injection moulding of extrusion compounded chopped glass fibre and thermoplastic polymer is today the most common combination [20]. Injection moulding's popularity is the result of its ability to manufacture complex small to medium sized parts at high production rates in an automated process. In spite of these strengths, this process presents a number of important weaknesses that need to be considered when designing a glass reinforced thermoplastic part for injection moulding or when selecting the appropriate processing technique.

### 5.2 Injection moulding process

In order to better understand the issues relating to injection moulding, it is important to first understand the process itself. The process is divided into a number of sequential operations :

- **Plastification** - the reinforced thermoplastic pellets are introduced into the plastification unit (**Figure 10**) which consists of a rotating screw inside a cylindrical barrel. The polymer matrix is melted and the composite mixture accumulates in front of the screw until the required volume for injection is available.
- **Injection** - the mould is closed and the screw quickly moves forward to push the molten material through a series of channels into the mould cavity(ies).
- **Holding** - the screw then applies pressure to the melt for a specified time. This creates a slow flow of material from the plastification unit to the mould cavity(ies). This additional material compensates for material shrinkage inside the cavity(ies).
- **Cooling/Ejection** - the material is left inside the cooled mould until sufficient solidification has occurred for the part(s) to be ejected without significant deformation.

The plastification, injection and holding phases cause a number of process limitations. These issues are described in the following sections.



**Figure 10** - Schematic diagram of an injection moulding machine showing : (P) plastification unit, (B) barrel, (S) screw, (C) clamping unit and (M) mould in the open position.

### 5.3 Injection moulding issues

#### 5.3.1 Fibre length degradation

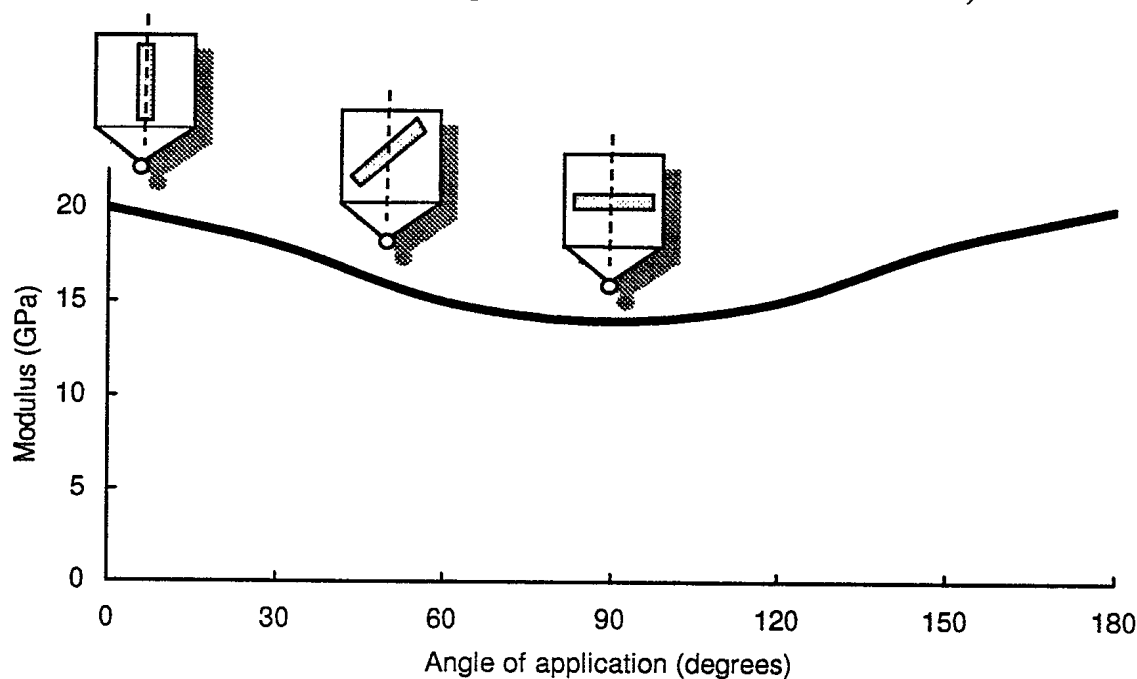
The shearing action of the plastification unit's screw will cause a degradation of the glass monofilament length [20]. Decreases in length will lead to a drop in mechanical properties of the part. As discussed in sections 4.2.1.1 and 4.2.1.2, degradation during injection moulding is critical if pultrusion compounded material is used, but is generally less important when using extrusion compounded material.

The monofilament degradation during the injection moulding step may be attenuated, but not eliminated, by the judicious choice of low shear injection moulding conditions [21].

### 5.3.2 Monofilament orientation

The glass monofilaments injected with the molten polymer will be oriented in the cavity by the shear stresses created during mould filling. The estimation of the orientation from mould geometry and injection moulding conditions is the subject of much research [22, 23]. In the absence of converging and diverging flows, monofilaments will be oriented preferentially in the direction of flow. The mechanical properties of the part will therefore be a maximum in this direction.

To illustrate the effect of glass monofilament orientation on mechanical properties, flexural specimens were cut at various angles from plates moulded using a 50% glass reinforced nylon compound [2]. The flexural modulus of these samples as a function of the angle is given in Figure 11. It is observed that the modulus is a maximum when the sample is cut from the plate in the direction of flow (direction of preferential monofilament orientation).

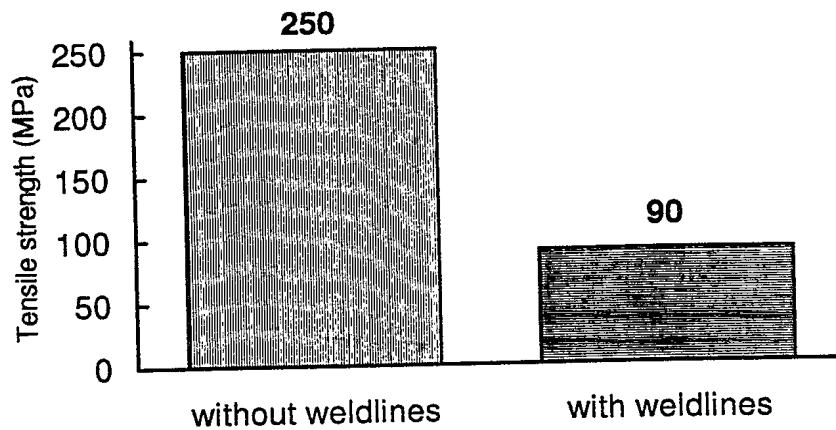


**Figure 11 :** Flexural modulus as a function of application angle for a plate injection moulded using a 50% glass reinforced PA mXD6 [2]

The monofilament orientation in injection moulded parts must be accounted for during part design as well as during the interpretation of property data sheets. Most data sheet properties of glass reinforced materials are measured in the direction of flow and hence in the direction of preferential glass orientation.

A second orientation related weakness of glass reinforced injection moulded thermoplastics occurs at weldlines. Weldlines are the interface created when two fronts of molten material meet during the injection phase. This can occur if multiple gates are used or if the cavity contains cores or inserts around which the molten material must flow. The flow induced shear stresses at the weldline are small as little flow occurs across this interface. Few glass monofilaments will therefore be oriented across the weldline and the mechanical properties perpendicular to the weldline plane are reduced to near those of the unreinforced polymer.

**Figure 12** compares the tensile strength of injection moulded samples which contain no weldlines and those containing a central weldline. The central weldline sample was injection moulded using a cavity containing two end gates. Both materials are 50% glass reinforced PA mXD6 [2]. The tensile strength of the sample containing the weldline is comparable to that of the unreinforced polymer - 100 MPa [24].



**Figure 12 :** Tensile strength of 50% glass reinforced PA mXD6 using samples with and without central weldlines.

Although weldlines are difficult to eliminate in most injection moulded parts, the weldline strength of the fibre reinforced thermoplastic polymer can be maximized by :

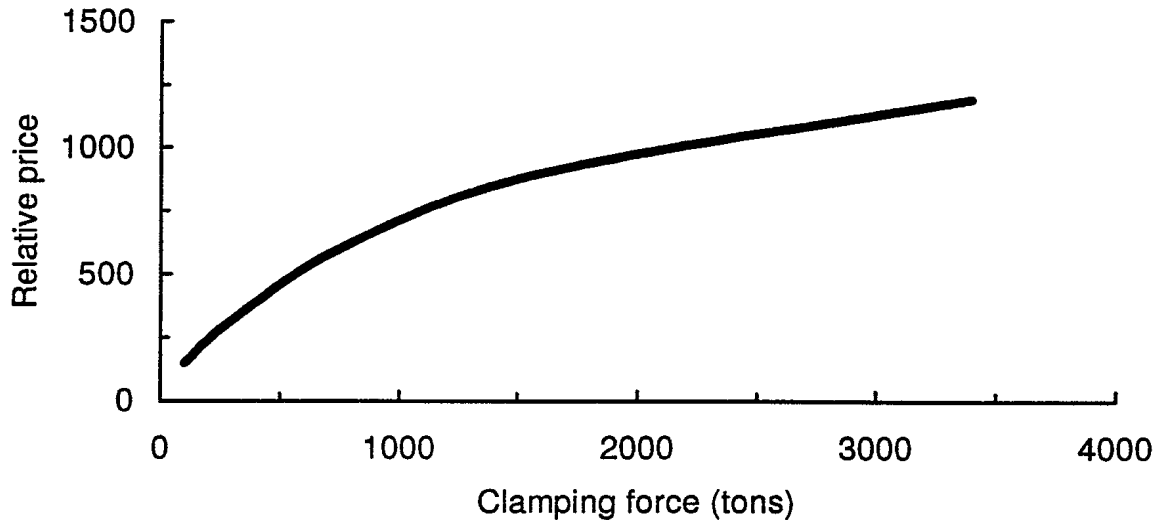
- **correct gating** : In general, the designer should minimize the number of gates to minimize the number of weldlines. The gate should also be positioned to move the weldlines to locations in the part which are under low stress.
- **correct venting** : It is necessary to allow evacuation of the air trapped between the melt fronts just prior to the formation of the weldline. This air can become trapped in the material upon solidification and can reduce the strength of the weldline. Air evacuation occurs through passages called vents that are machined into the mould usually along its parting line.
- **correct processing** : For maximum weldline strength, it is important to inject the hot material quickly to minimize cooling of the material prior to weldline formation.

### 5.3.3 High pressures during the injection moulding cycle

The injection moulding process requires high melt pressures to inject the material into the mould and to hold the pressure. Large clamping forces are thus needed to keep the mould closed. The injection moulding machine clamping force is an important factor in determining machine cost. **Figure 13** shows the relative price of injection moulding machines as a function of the clamping force in tons. [2]



Although high pressures are required for most injection moulding materials, the pressure (hence clamping force) is often higher for glass reinforced polymers versus the non-reinforced variety due to their higher melt viscosity.



**Figure 12** : Relative price versus clamping force for injection moulding machines [2].

Minor reductions in injection moulding pressure can be made by using several gates or centrally positioning the gate to minimize the flow length. Both these options have, however, implications on weldlines and glass monofilament orientation in the part.

Modified injection moulding techniques such as gas-assist injection moulding can also reduce both the injection and holding pressures [25, 26]. Gas-assist involves injecting an inert gas into the molten polymer to complete the injection and holding phases. The gas forms a hollow channel in the centre of the part.

#### 5.4 Alternative processes

Although minor improvements can be made to the injection moulding process to address the various issues outlined in section 5.3, eliminating them involves using alternative processing techniques such as compression moulding, vacuum forming and filament winding. These techniques are commonly used for reinforced thermoset resins.

These alternative processes generally use continuous reinforcement (see table 2) and involve a more controlled placement of the reinforcement during processing. Although compression moulding involves high pressures, the pressure is generally lower than that observed during injection moulding due to the lower flow length and absence of a gate and runner system [15]. The lower pressure of these processes therefore reduces the equipment costs compared with injection moulding.

In spite of these advantages, there remain many challenges in using these alternative processes. They can be broadly grouped into two categories :

- **compounding** : The compounding cost associated with manufacturing thermoplastic polymer with either continuous roving or glass mat is significantly higher than classic extrusion compounding. New techniques must be developed and older ones improved if these costs are to decrease.
- **processing** : Many of the techniques used to process continuously reinforced thermoplastics are relatively new or have been adapted from thermoset processing. More work is needed to optimize these processes for thermoplastic polymers.

The Royal Military College of Canada (RMC) is actively involved in continuous glass reinforced thermoplastic research. Two research projects are currently underway :

- **Experiments and modeling of a pultrusion compounding process** : The objective of this research is to model the thermoplastic melt impregnation of continuous glass roving in a pultrusion compounding process as non-newtonian fluid flow through porous media. Bench scale experiments are performed to estimate the model parameters. Model results are compared with existing pilot scale pultrusion compounding data.
- **Low pressure forming of glass fibre reinforced thermoplastic mat** : Co-mingled glass and polymer fibres can be woven to form mats which can be processed using a variety of techniques. The objective of this research is to study the suitability of this type of material to low pressure forming of large panels to be used as part of a ballistic protection system. The experimental and modeling work will involve all stages of the part manufacture process: heating, vacuum forming, composite consolidation and part cooling.

For further information on any of these subjects, please contact the authors.

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