PRODUCTION OF EPOXY/E-GLASS COMPOSITES USING “CLEAN PULTRUSION”

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ABSTRACT

The primary aim of the work reported in this thesis was to develop an environmentally friendly pultrusion technique for the production of the fibre reinforced composites. The resin bath used in the conventional pultrusion was replaced with two custom-designed resin impregnators (prototypes-I and prototypes-II).

The primary aspect of the resin impregnators was to reduce the time for through-thickness impregnation by spreading the fibre bundles. Two rigs were designed and evaluated to spread the rovings: an automated fibre spreading rig and a serpentine fibre spreading unit. The serpentine fibre spreading unit was seen to give the best results. In a separate set of experiments it was shown that the through-thickness impregnation of the fibre bundles was improved by spreading.

The composites were produced using the resin impregnators (prototypes-I and prototypes-II) at three pultrusion speeds. Reference samples were also obtained from dip-type resin bath. The physical and mechanical properties of the pultruded composites were compared. It was shown that the properties of the composites were enhanced whilst using “Clean” pultrusion as compared to the conventional resin-bath system.

In conclusion it was shown that the “Clean” pultrusion could be used as an alternative to the conventional resin bath pultrusion.

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1. INTRODUCTION

1.1. BACKGROUND

Pultrusion is a cost-effective and high-speed process for the production of fibre reinforced composites. This process produces continuous lengths of reinforced polymer composite profiles with constant cross-sections. The fibre reinforced composites produced have high performance characteristics (high specific strength and stiffness, light-weight and corrosion resistance). Pultruded composites are used in wide areas of applications and industries including aerospace, automotive, marine and civil (Vaughan, 2003).

The pultrusion process involves pulling impregnated fibre bundles through a heated steel forming die. The profile is cured and pulled to a cutting station where it is cut to the required length. Both thermoplastic and thermosetting matrices are used in pultrusion. In a conventional pultrusion process, with thermoset resins, the reinforcements are impregnated with resin and are passed through a die where resin undergoes a non-reversible chemical reaction (curing), forming a hard and infusible product. The most common resins used in pultrusion are unsaturated polyester, epoxy, vinyl ester and phenolics. Unsaturated polyesters and vinyl esters are most commonly used due to lower price and good properties. Epoxy resins on the other hand have superior properties however are more expensive. The most common types of reinforcements used are E-glass and carbon.

Generally, an open dip-type resin bath is used to impregnate the fibre bundles where the fibre bundles are passed through a resin bath. Pultrusion using an open resin bath system has environmental issues, for example, emission of environmentally harmful vapours such as styrene in case of unsaturated polyester resin systems, limitations in using fast-cure resin systems, and requirements for the resin bath and ancillary equipment to be cleaned with large volumes of solvents. To rectify the issues associated with the open bath pultrusion systems, resin-injection pultrusion has been developed. Here resin is injected into the fibre bundles at high pressure. Difficulty in the complete impregnation of the fibre bundles and higher costs of the high-pressure resin dispensing equipment are the prohibitive factors in wider use of the resin-injection pultrusion.

1.2. AIMS AND OBJECTIVES

The focus of this project is on developing an energy-efficient pultrusion process. The specific aims of this project are as follows: (i) To undertake pultrusion trials using a custom-built resin impregnator in conjunction with a fibre spreading device. (ii) To develop an appreciation of the techniques and models associated with fibre spreading and impregnation. (iii) To undertake experiments to monitor the rate of axial and transverse impregnation. (iv) To determine the physical and mechanical properties of pultruded composites manufactured using the conventional bath and "Clean" pultrusion. The physical and mechanical properties to be determined include density, fibre volume fraction, flexural strength, flexural modulus, inter-laminar shear strength, tensile strength and tensile modulus. This will be carried out to assess the relative merits of the "Clean" and conventional pultrusion methods.

1.3. STRUCTURE OF THE THESIS

Chapter 2 discusses the literature review regarding pultrusion, fibre spreading, resin impregnation and manufacturing methods.

Chapter 3 discusses the experimental methods associated with pultrusion, fibre spreading; impregnation monitoring and the testing of composites.

Chapter 4 discusses the results of fibre spreading with two different spreading rigs. Also discussed are the multi-tow spreading and the twists in the fibre bundles. Impregnation results and testing of the composites are also discussed.

Chapters 5 and 6 discuss the key conclusions reached and recommendations for future research respectively.

2. LITERATURE REVIEW

2.1. FIBRE REINFORCED COMPOSITES

Fibre composites are renowned for their high specific stiffness, strength and toughness (Donnet et al., 1977). Composites are made from two or more constituent materials with different physical or chemical properties (Hull and Clyne, 1996). Most composites that are used in the industry are based on thermosets or thermoplastics, which are reinforced with fibres such as carbon, glass or aramid. Carbon fibres are relatively expensive and therefore, less expensive fibres such as glass are generally used in a number of industrial sectors (Hwang et al., 1999).

2.1.1. Reinforcements

The role of reinforcements in composite materials is to carry the majority of the load that is imposed on the structure. Fibres such as carbon and aramid possess high tensile strengths because of the nature of the bonding and the orientation of molecules along the fibre direction (Srindhar, 1993). More than 95% reinforcements used in composites are glass fibres. Glass fibres have an amorphous structure and are isotropic in comparison to carbon and aramid fibres (Ehrenstein, 2001). Glass fibres also have good thermal and electrical insulation, good fire resistance as well as being relatively inexpensive (Ehrenstein, 2001).

Carbon fibres used for reinforcement of high-performance composites have reached a high level of quality. Around 95% of today's carbon fibres are based on PAN (polyacrylonitrile) fibres, a requirement of carbon fibres is to use fibres with a strain to failure of at least 1.5%; and stiffness values above 250 GPa (Fitzer and Frohs, 1990). Carbon fibres based on mesophase pitch are also available commercially.

2.1.2. Resin systems

The resin systems used in composites can be split into two general groups, thermoplastic and thermosetting polymers. Thermoplastic polymers soften with heating; the inter-chain forces are weak which allows the segments in the polymer chain to 'rotate' about the bonds (Morley, 1987).

In thermosetting polymers a three-dimensional network is produced by cross-links being formed by chemical reactions between the resin and hardener. Once cured, thermosets will not become liquid again if re-heated. However the mechanical properties change over a certain temperature range and this is called the glass transition temperature (Morley, 1987). The advantage of using thermosets is that these possesses viscosity values less than 100 Pa·s; thermoplastics have viscosities in the range 500-5000 Pa·s. The higher the viscosity of the resin system, the greater the problems in manufacturing composites (Chang et al., 1988). In thermoplastics, the dispersion of fibres can be poor and the quality of impregnation can be a concern (Klinkmuller et al., 1992).

The main types of thermoset resins used are the conventional resin systems, such as polyesters, epoxy and vinyl esters (Hull and Clyne, 1996). Polyurethane is still a relatively new resin system as far as pultrusion is concerned. Table 1 shows the advantages and disadvantages of conventional resin systems. Polyester is the most used resin and is the cheapest. Epoxy resins are used to produce composites with tougher properties and are superior to other alternative thermosetting systems; however they are much more expensive. Hull and Clyne (1996) showed that epoxy resin has good resistance to heat distortion and they exhibit lower shrinkage during curing than polyesters and vinyl esters.

Table 1 A summary of the advantages and disadvantages of the common classes of thermosetting resins.

Resin	Advantages	Disadvantages
Polyester	• Easy to use • Lower costs for these resins available	• High shrinkage cure • Moderate mechanical properties • High styrene emissions in open moulds • Limited range of working times
Vinyl ester	• High chemical/environmental resistance • Higher-mechanical properties than polyesters	• High styrene content • High cure shrinkage • Higher cost than polyesters • Post cure generally required for high properties
Epoxy	• High mechanical and thermal properties • High water resistance • Long working times available • Low cure shrinkage	• Most expensive • Corrosive • Critical handling

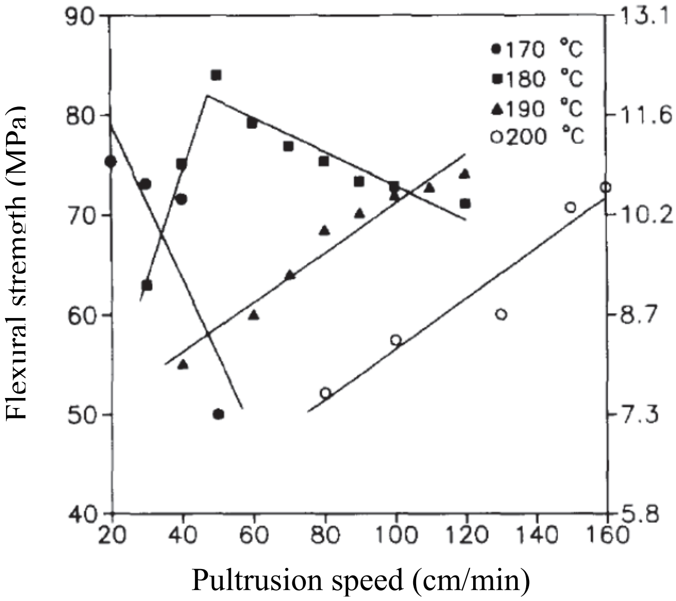
2.2. PULTRUSION

Pultrusion is a process which produces composites profiles, with a constant cross-section in continuous lengths (Tattersall, 1981). It involves drawing the fibres from creels and passing them through a resin bath to impregnate the bundles. Usually centre-pull creels are used in pultrusion. The impregnated fibre bundles are pulled through a heated die here the resin cures and the shape of the profile is formed. The composite is pulled from the die towards a cut-off saw where it is cut to the required length (Hull and Clyne, 1996). Complex composite profiles can be produced by pultrusion. The main parameters that have to be taken into account whilst pultruding are: pultrusion speed, die temperature, post-curing temperature, the degree of impregnation, pull-force and the tension of the fibres.

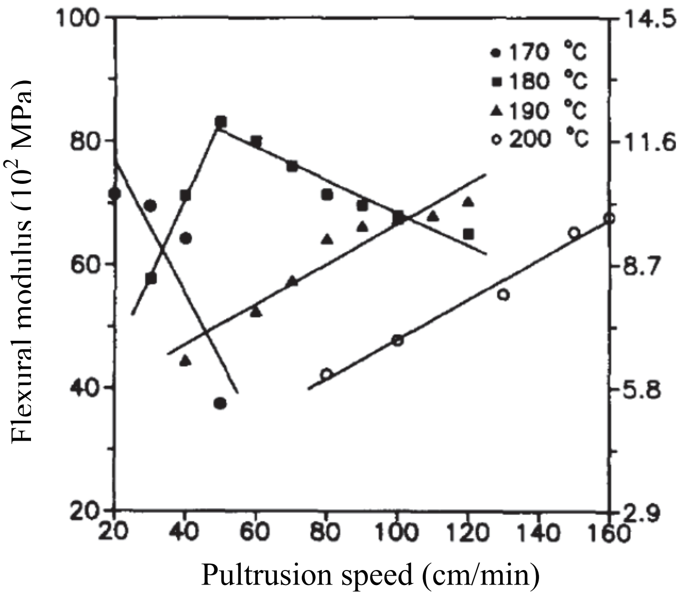
Within the composites industry, pultrusion is one of the fastest growing processes. However, there have been environmental concerns over the years over the emissions given off from open resin baths. Although, the resin-injection pultrusion addresses this problem but, the cost associated with the resin-injection equipment and specially designed dies are prohibitive factors in wide spread acceptance of this technique (Bannister, 2001).

Thorsted (1987) used an epoxy resin system to produce continuous lengths of cured fibre epoxy composites; epoxy was used as it has a lower value of resin-shrinkage as compared to unsaturated polyester and vinyl ester resin systems. This process was carried out in an open resin bath. A resin system was used that had a long pot life and a low viscosity enough to wet-out the fibres, resulting in low cost production of composites having good mechanical properties.

Chen et al. (1992) investigated a range of processing parameters during pultrusion using a polyurethane resin. The effects of the processing conditions on the mechanical (flexural strength and modulus) and thermal properties of fibre reinforced polyurethane composites were analysed. They used pultrusion speeds between 20 and 160 cm min^{-1} and die temperatures between 170-200 °C.



(a)



(b)

Figure 1 Flexural properties of pultruded polyurethane/E-glass composite as a function of pultrusion speed at various die temperatures. (a) flexural strength; and (b) flexural modulus. Reproduced from Chen et al., (1992).

Their results showed that the composites with the highest flexural strength were produced when they are hauled-off between 40-60cm min^{-1} at a temperature of 180 $^{\circ}$C. When a lower pulling rate was used, it allows the residence time of the resin in the die to be longer. Therefore it was proposed that the degree of polymerisation would be higher, and the wet-out of fibres will be better.

Chen et al. (1992) also reported that at a low pultrusion speed and a temperature of 170 $^{\circ}$C, the thermal and mechanical properties were found to be higher, however when the die temperature was increased to 180 $^{\circ}$C and the pulling rate was maintained below 50 cm min^{-1}, the properties were observed to decrease as the polyurethane started to degrade.

2.2.1. Conventional dip-type resin bath-based pultrusion

Conventional dip-type resin bath-based pultrusion is shown schematically in Figure 2. This technique has been used for many years; however there are a number of problems the conventional manufacturing process faces. The first concern is the emission of harmful volatiles such as styrene from the resin bath. Dube et al. (1995) stated that the processing of thermoset resins has concerns during fibre wetting and cure, and this is linked to the 'pot life' of the resin. With thermosetting resins, the viscosity increases with time and this will have an adverse effect on wetting and it can also lead to premature gelation of resin outside the die.

The limited pot-life of the resin in the resin bath means that the resin in the bath can set into a solid; this resin will have to be removed prior to the resumption of production. Having a limited pot life also means that fast-curing resins cannot be used in resin bath-based pultrusion.

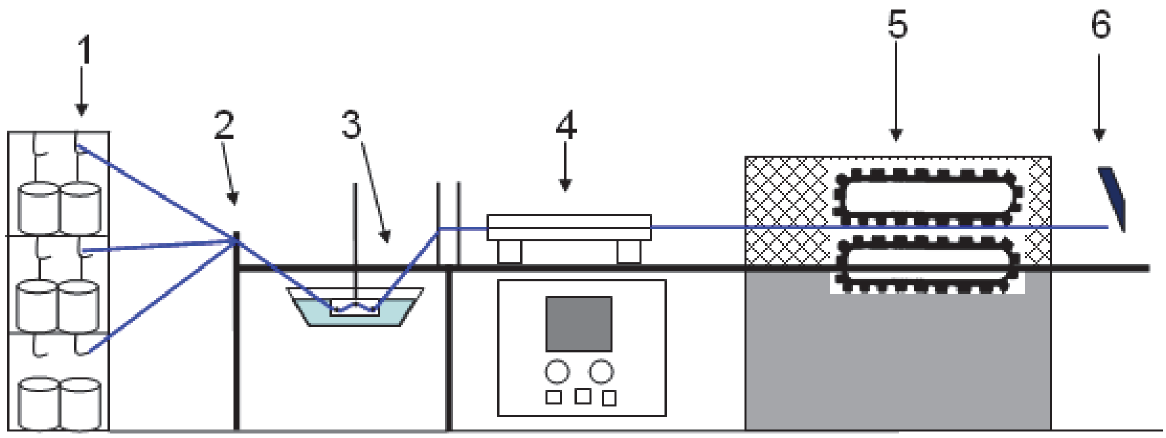


Figure 2 Schematic illustration of conventional dip-type resin bath-based pultrusion. The details of the coded items are: (1) creel stand: (2) back plate; (3) resin bath; (4) die; (5) cleat-type caterpillar; (6) cut-off saw.

Another major issue associated with conventional pultrusion is the need for the equipment to be cleaned thoroughly at the end of each production. Due to the fact that large volumes of solvents have to be used for the cleaning operation, this will contribute the overall costs of production. Furthermore it is also necessary for the solvents to be recovered prior to disposal (Pandita et al, 2012).

At the end of production in conventional pultrusion, the excess resin present in the resin bath has to be disposed. Precautions must be taken as the excess resin can exotherm. Therefore when large resin baths are used the waste resin has to be split into smaller volumes prior to curing.

2.2.2. Resin-injection pultrusion

A significant improvement in the pultrusion process is the replacement of the traditional resin bath with a resin-injection system, as shown in Figure 3. The resin-injection system is enclosed; therefore the loss of solvents is reduced significantly. This reduces the emissions from the process also resulting in a more stable and consistent resin supply (Strong, 1996).

The resin-injection system allows the time it takes from resin mixing to curing to be much shorter than conventional pultrusion, therefore fast curing resin systems can be used (Ding et al, 2000). The disadvantages of using the resin-injection system are that it is more expensive than the other pultrusion systems. The added cost is due to high-pressure resin dispensing equipment and specially designed dies. The fibre bundles are in a packed state at the resin-injection point and very high pressure is required for complete impregnation.

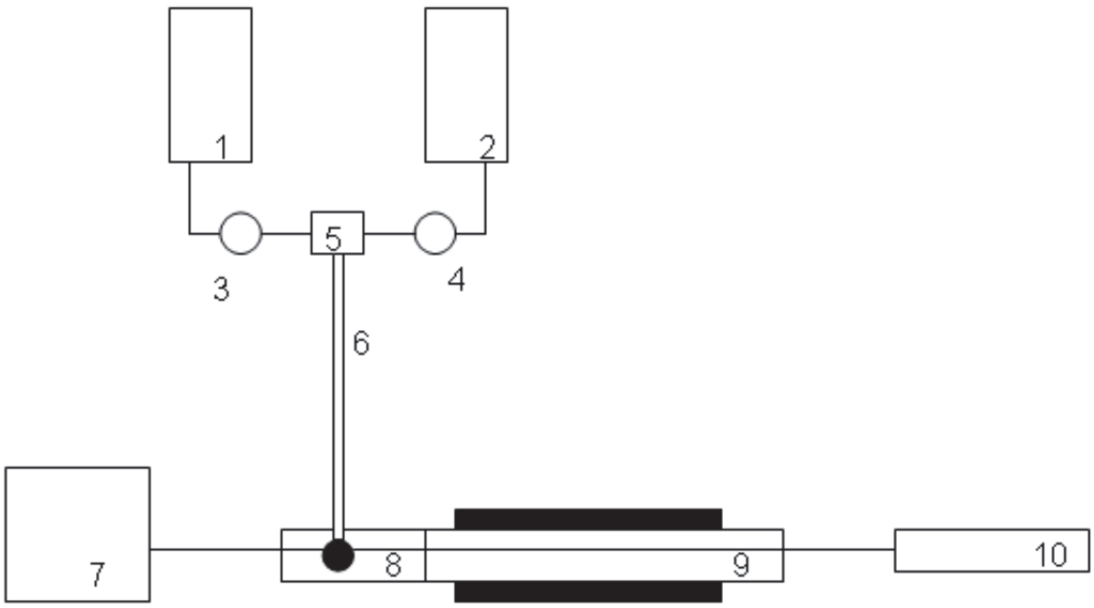


Figure 3 Schematic illustration of the resin-injection pultrusion process. The coded items re as follows: (1) resin container; (2) hardener container; (3,4) pumps; (5) manifold; (6) static mixer; (7) creels; (8) injection point; (9) die with heating element; and (10) puller.

Brown et al. (2010) presented a modified resin-injection pultrusion method which included a chamber for coating the external surfaces of the fibre bundles and a second chamber for coating the fibres within the bundles. They used a resin-injection resin system (as shown in Figure 4) which is intended to provide the resulting composite material with greater mechanical and physical properties in contrast to using the conventional resin impregnation system.

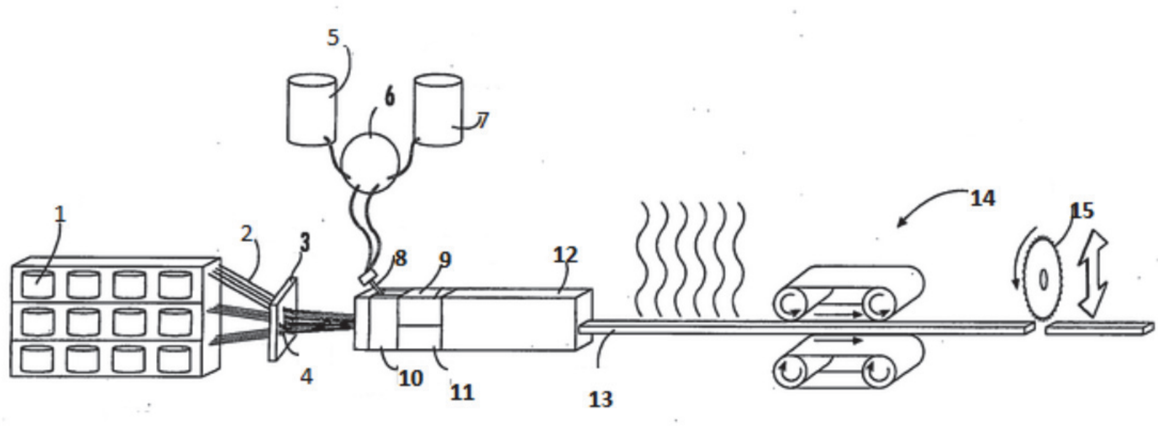


Figure 4 Pultrusion system used by Brown et al. (2010). The details of coding are as follows: (1) fibre creels; (2) fibre tows; (3) supporting card; (4) holes; (5) container 1; (6) metering device; (7) container 2; (8) disposable static mixer; (9) impregnation die; (10) chamber 1; (11) chamber 2; (12) die; (13) cooling; (14) puller; and (15) cut-off saw.

2.2.3. "Clean" pultrusion

Clean pultrusion is being developed by the Sensors and Composites group at the University of Birmingham (2012) and the author was a member of this team. A schematic of the "Clean" pultrusion process is shown in Figure 5.

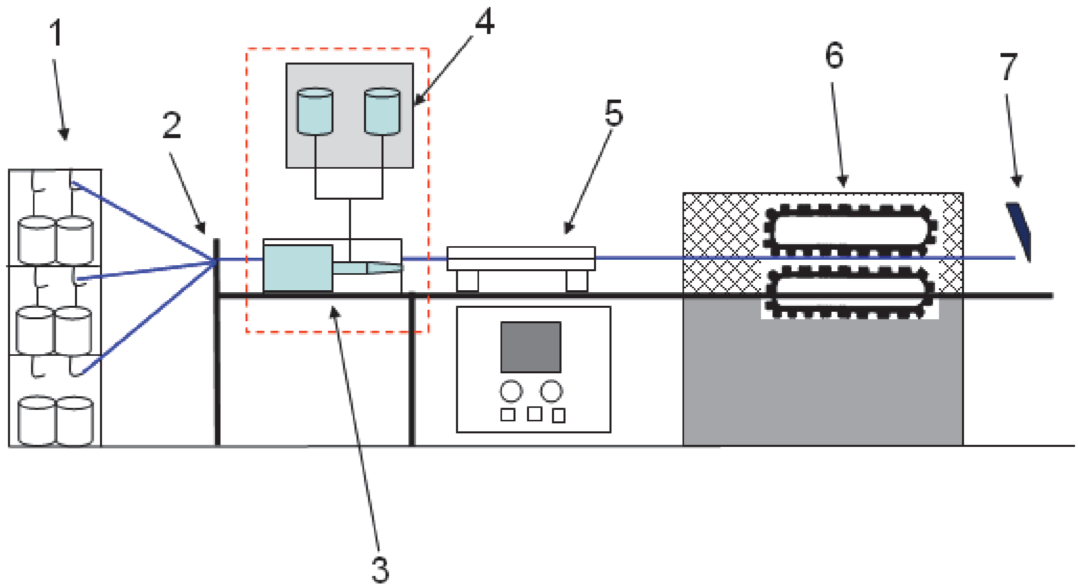


Figure 5 Schematic illustration of the 'Clean' pultrusion process. The coded items are as follows: (1) roving racks; (2) guide plates; (3) fibre spreader and resin impregnator; (4) resin dispenser; (5) die; (6) puller; and (7) cut-off saw.

Here the open resin bath was replaced with a custom-built resin impregnation unit. The resin and the hardener were located in separate containers and pumped on demand into a manifold. The liquid streams were then directed into a static mixer where the resin and hardener were mixed. Prior to impregnation the fibres were spread, which enhanced the transverse impregnation of the mixed resin into the fibres. A custom-built fibre spreading rig was used to spread the fibre bundles. The spread ribbon of the fibre bundles was impregnated in the impregnator which was designed on the principles of pin-, injection- and capillary-impregnation. The impregnation unit also contains a feedback control system, which controls the throughput of the resin and hardener to the pultrusion speed.

2.3. FIBRE SPREADING

2.3.1. Models for fibre spreading

An important stage of the clean pultrusion process is the impregnation of a bundle of fibres. If the fibre bundles used in pultrusion can be spread prior to impregnation, the thickness of the bundle will be decreased and thus it would take less time for resin to penetrate through the fibre bundle. The process of fibre spreading is shown in Figure 6.

(a) (b)

Figure 6 Schematic illustration of the fibre spreading: (a) as-received bundle, and (b) spread bundle.

Wilson (1997) proposed following equation to predict the width of the spread fibre after it has been reciprocated over a bar:

$$\omega = \left(12AL\cos a\right)^{1/3} \qquad \text{Equation 1}$$

where is the width of the spread fibre, A is the cross sectional area of the fibre bundle, L is the distance between bars and α is the angle between vertical and fibre bundle.

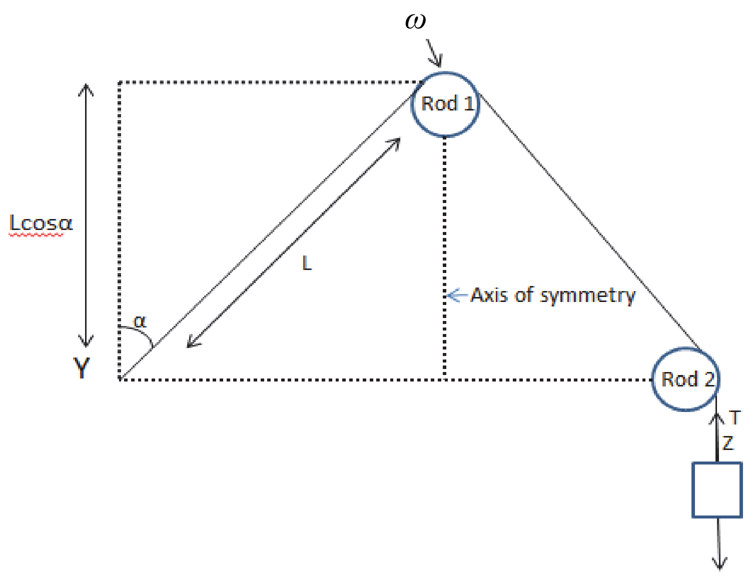


Figure 7 Schematic illustration of side view of the experimental set up adopted by Wilson (1997) studying the lateral spreading in a single fibre bundle.

2.3.2. Fibre spreading methods

Many techniques have been used to induce fibre spreading including: mechanical, electrostatic, pneumatic, vibration and acoustic-based methods.

Mechanical-based fibre spreading techniques use rollers, pins and guide tubes to spread the filaments in a bundle. It has been reported that if the tension in the mechanical based spreading is high; it can cause damage to the fibres. Van den Hoven (1981) used a cylindrical guide containing a range of grooves, which rotate affecting the width of the fibre bundles. Kiss et al. (2002) constructed a system which enabled them to separate fibres without 'fuzz' formation or any fibre breakage. Smooth grooved guide bars and splitter bars were used.

Electrostatic fibre spreading uses a high-voltage to establish an electric field through which the filament passes. Uchiyama et al. (1972) spread fibres by passing them continuously through a narrow passage with two sets of electrodes. The potential was at least 500 Volts. The second electrode was in alignment with the first electrode but it was spaced out from the narrow passage spreading the individual filaments. This method of electrostatic spreading has safety implications but it results in lower fibre damage.

Pneumatic-based fibre spreading involves passing the fibre bundles through a jet device and subjecting it to high velocity stream of gaseous liquid (Pryor, 1985). The spread fibre bundles via this method are volumised and not suitable for the resin impregnators used in this study.

Vibration-based fibre spreading generally uses a vibrating roller. Yamamoto et al. (1988) spread fibre bundles by passing a bundle of fibres, under tension, around one or two or more freely rotatable rollers whilst vibrating at least one of the rollers to spread the fibres. The main disadvantage of this method is that the fibre bundle are subjected to abrasive forces from surface of spreading station, causing filaments to break generating 'fuzz' (Tanaka., et al, 2004). Iyer et al (1991) presented a method where the tow zigzags over rods adjacent to a loud speaker. Here the acoustic energy from the speaker was used to spread the fibres.

Irfan et al. (2013) developed an automated fibre spreading rig where a combination of mechanical and vibration was used to spread the fibre bundles. This rig was used in the current study.

2.4. RESIN IMPREGNATION

The techniques to impregnate the rovings in the pultrusion process have been described in Sections 2.2.1., 2.2.2. and 2.2.3. Efficient resin impregnation is a major part in the formation of a good quality reinforced composites. The reinforcement fibres must be fully impregnated (wetted-out) by the resin. If the fibres are not fully impregnated, then the mechanical and physical properties of the composite produced will be poor. Therefore it is imperative to appreciate the process of impregnation.

2.4.1. Techniques for monitoring resin impregnation

Ahn et al. (1995) reviewed three different techniques to measure the impregnation of three-dimensional permeability of fibre performs, and compared their results. The first method is the pressure drop technique. In this method rectangular or circular thin plates are cut out of the preform. If the rectangular sections are used, the sections are placed into a mould consisting of two parallel plates. If the circular section is used, it is placed inside a cylindrical mould. Liquid of a known viscosity is introduced into the mould. The inlet pressure and flow rates are measured as the liquid penetrates the sample.

The second method investigated by Ahn et al. (1995) is the flow visualisation technique. A thin rectangular plate is taken from the preform and placed between two transparent plates. A liquid of a known viscosity is introduced at the centre of the plane. The inlet pressure is kept constant; the position of the liquid front is observed and is recorded as a function of time.

The third method reported was the fibre optic technique. This technique was used to measure preforms with a more complex fibre arrangement where the pressure drop and flow visualisation techniques could not be used. Short sections of cladding were removed from the optical fibre, and then embedded in the preform. A laser light was then used to transmit light through the optical fibre and the light intensity at the end of the optical fibre was recorded. As the liquid penetrates the preform is passes over the cladded areas causing a sudden decrease in the light intensity transmitted. It was proposed that the changes in light intensity can be used to infer the rate of movement of the liquid front.

3. EXPERIMENTAL

3.1. MATERIALS

The materials used in this study are summarised below:

3.1.1 Resin

The epoxy/amine resin system used in this study was supplied by Huntsman Advanced Materials. The details of the resin formulation are presented in Table 2.

Table 2 Details of the resin system used in this study

Components	Supplier	Part by weight
Araldite LY 556 (Epoxy resin)	Huntsman Advanced Materials	100
Hardener HY 917 (Anhydride hardener)	Huntsman Advanced Materials	90
Accelerator DY 070 (Imidazole accelerator)	Huntsman Advanced Materials	2
OP wax (Internal mould release)	BASF	4

3.1.2 Fibre

The fibres used in this study were donated by PPG industries UK. The E-glass fibre type used was Hybon® 2026. Hybon® 2026 fibre glass is a continuous filament; single-end roving designed to reinforce polyester, epoxy and vinyl ester resin systems. The detailed specifications of the fibres are provided in Table 3.

Table 3 Details of the Hybon® 2026 E-glass fibres used in this study

Characteristic	Value
Type of sizing	Silane
Per cent of sizing, nominal (by wt. of glass)	0.70%
Nominal filament diameter (microns)	17
Roving tex (g/Km)	2400

3.2. FIBRE SPREADING

3.2.1. Automated fibre spreading rig

In the first set of fibre spreading experiments an automated fibre spreading rig was used to spread the fibre bundles. This rig was designed on the principles of tension and tension-release as described by Irfan et al. (2012). A schematic illustration and a photograph of the automated fibre spreading rig are shown in Figures 8 and 9 respectively.

The fibre bundle is pulled through a pre-tension release system which sets the level of pre-tension of the fibre. Rollers containing motorised disks are then used to spread the fibre bundles via the action of tension and tension-release. The spread fibre is then passed onto the exit roller, which maintains the degree of spreading.

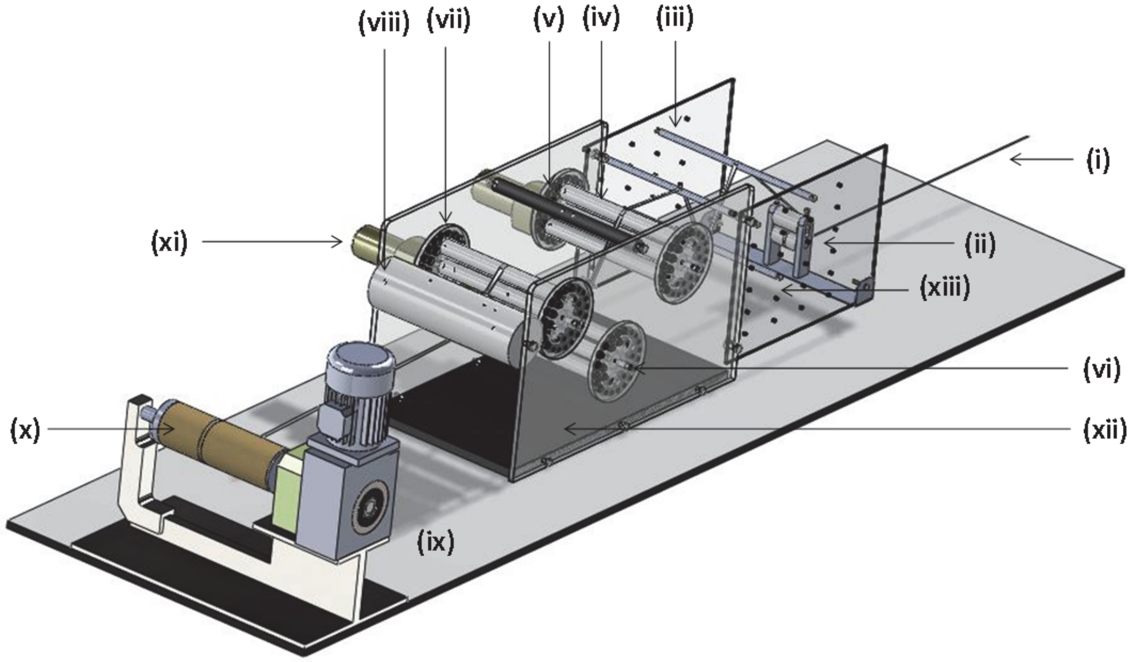


Figure 8 Schematic illustration of the tension-release rig. The coded items are: (i) input roving from the creel; (ii) pre-tensioning device; (iii) secondary pre-tensioning bar; (iv) acetal rods; (v) first roller-disk; (vi) second roller-disk; (vii) third roller-disk (measuring point); (viii) exit roller; (ix) haul-off motor; (x) mandrel; (xi) motor for the roller-disk; (xii) perspex frame; and (xiii) perspex frame with a facility to control the secondary tension. Illustration by Francisco Nieves, University of Birmingham, 2013. Rig design by VR Machavaram and construction by Mark Paget, University of Birmingham, 2011.

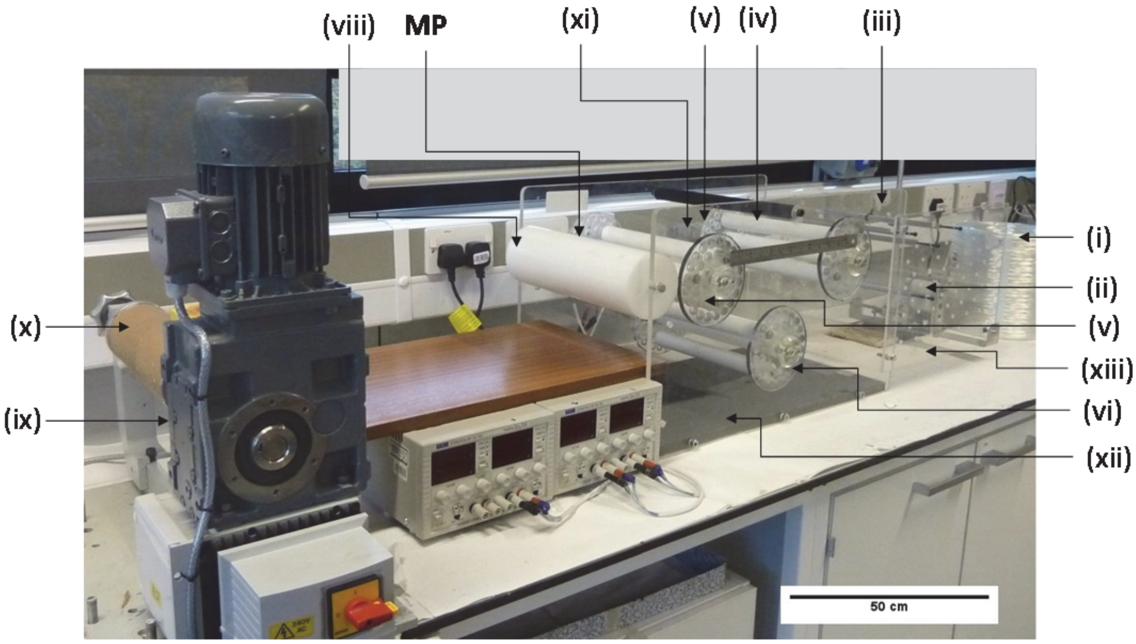


Figure 9 Photograph of the tension-release fibre spreading rig. The position where the widths of the spread rovings were measured is indicated as MP.

The rig consisted of three motorised discs which had between one and six rollers attached to it. The experiment started with an as- received fibre bundle which was 4.18 mm in width. The bundle was marked and then guided through a guiding device and passed through the rollers and onto the rotating discs. The fibre bundle is pulled over the first disc, under the second disc and over the third disc and then it is passed under the exit roller. The final step involved the fibre wound onto haul-off unit. Vernier calipers were used to measure the width of the spread fibre and the measurement was recorded. To stop measuring the same length of fibre twice in a given experiment, the marked part of the fibre has to be passed throughout the whole system before the next test run starts. The haul-off winder is used to wind the glass fibres at specific speeds.

The number of experiments undertaken using the tension-release system to spread the fibres was based on a Taguchi-based experimental matrix. The different parameters considered are detailed in Table 4.

Table 4 Factors and levels selected in the experiment matrix.

Factors	Factor description	Level 1	Level 2	Level 3	Level 4	Level 5	Level 6
A	Configuration*	1	2	3	4	5	6
B	Pre-tension (N)	2	6	10	-	-	-
C	Pull speed (m/min)	1	3	5	-	-	-
D	Disk rotation (rpm)	25	50	100	-	-	-

* See Figure 10 for the details of each of the rotating roller-disk configurations.

Minitab software was used to derive the L18 experimental matrix as shown ion Table 5. These parameters were chosen as they were thought to have an effect on the degree of fibre spreading that could be achieved.

Table 5 L_{18} experimental matrix for the Taguchi-based investigation.

Roller configuration	Tension (N)	Haul-off speed (m/min)	Disk rotation (RPM)
A	2	1	25
A	6	3	50
A	10	5	100
B	2	1	50
B	6	3	100
B	10	5	25
C	2	3	25
C	6	5	50
C	10	1	100
D	2	5	100
D	6	1	25
D	10	3	50
E	2	3	100
E	6	5	25
E	10	1	50
F	2	5	50
F	6	1	100
F	10	3	25

Step 1: Calibration of the linear speed

Three different haul-off speeds were used; 1 m/min, 3 m/min and 5 m/min. The haul-off unit was used to control the winding speed. The tension-release equipment was calibrated against the linear winding speed. This was done by setting the haul-off unit at a specified setting, then marking sections of the fibre and then recording the time taken for the marked sections to cross a set point.

Step 2: Calibration of the disk rotation of the roller configuration

Three different disk rotations were used; 25 rpm, 50 rpm and 100 rpm. A tachometer was used to determine the number of rotations the disks made per minute. The three disk rollers were connected to three power supplies. Each power supply consisted of screens in which the voltage can be changed to change the speed of the disks to achieve the required rotation rates.

Step 3: Calibration of the initial tension

Three levels of tensions were used; 2 N, 6 N and 10 N. The tension was measured by using a tension meter; the pins were adjusted on the tension-release system to give the three different tensions needed to carry out the experiment.

Step 4: Spreading experiments

There were four points where the tow width (mm) was measured: pinch rollers. 1^{st} rotation disk; 3^{rd} rotation disk; and exit roller

Each experiment for all the configurations was repeated ten times. Six roller configurations of the roller disk were used to find the best configuration for the spreading of glass fibre. Six configurations were used to determine the optimum positions of the rollers to achieve the highest degree of fibre spreading. Figure 10 shows the six different configurations.

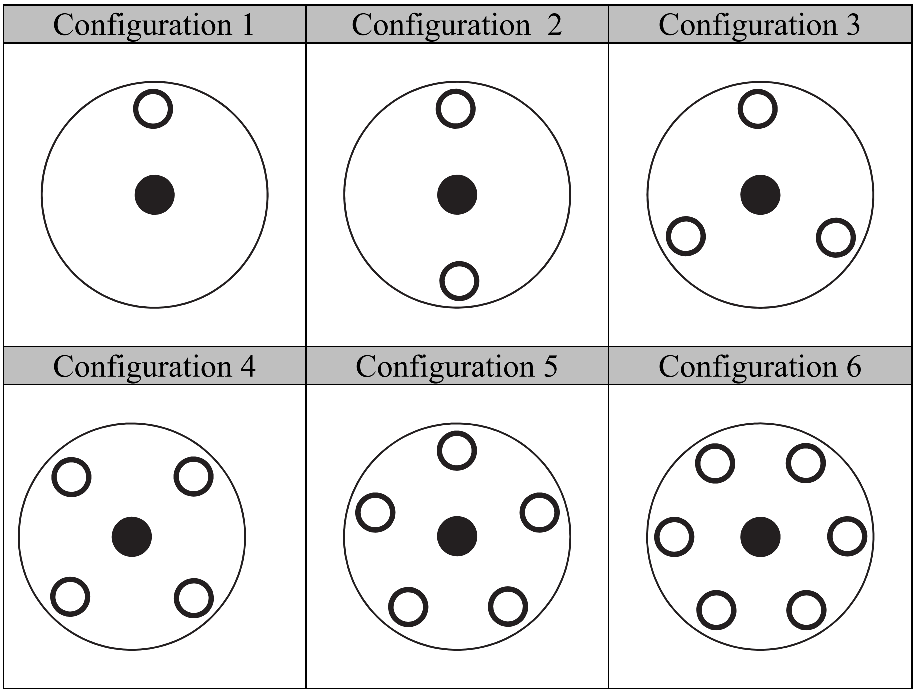


Figure 10 Roller configurations considered for the tension release system (Irfan et al, 2013).

3.2.2. Fibre spreading using serpentine fibre spreading rig

During the course of the experiments that were carried out using the tension-release rig, it was observed that the degree of fibre spreading at the serpentine rollers was significant, hence simple versions of the tension-release system, named as “serpentine rig” was considered. The serpentine rig, as shown in Figure 11, was much smaller than the tension-release system used for pultrusion. A schematic illustration of the serpentine rig is shown in Figure 12.

It consisted of a pinch roller and three rollers and/or profiled rollers. Profiled rollers were rollers covered by a sheet of ball bearings as shown in Figure 14. The ball bearings help in breaking up the binder on the fibre bundles and increase the spreading. There was a total of seven configurations, as shown in Figure 13, in which two of them were the extremes; configuration-1 were all plain rollers and configuration-6 were all profiled rollers.

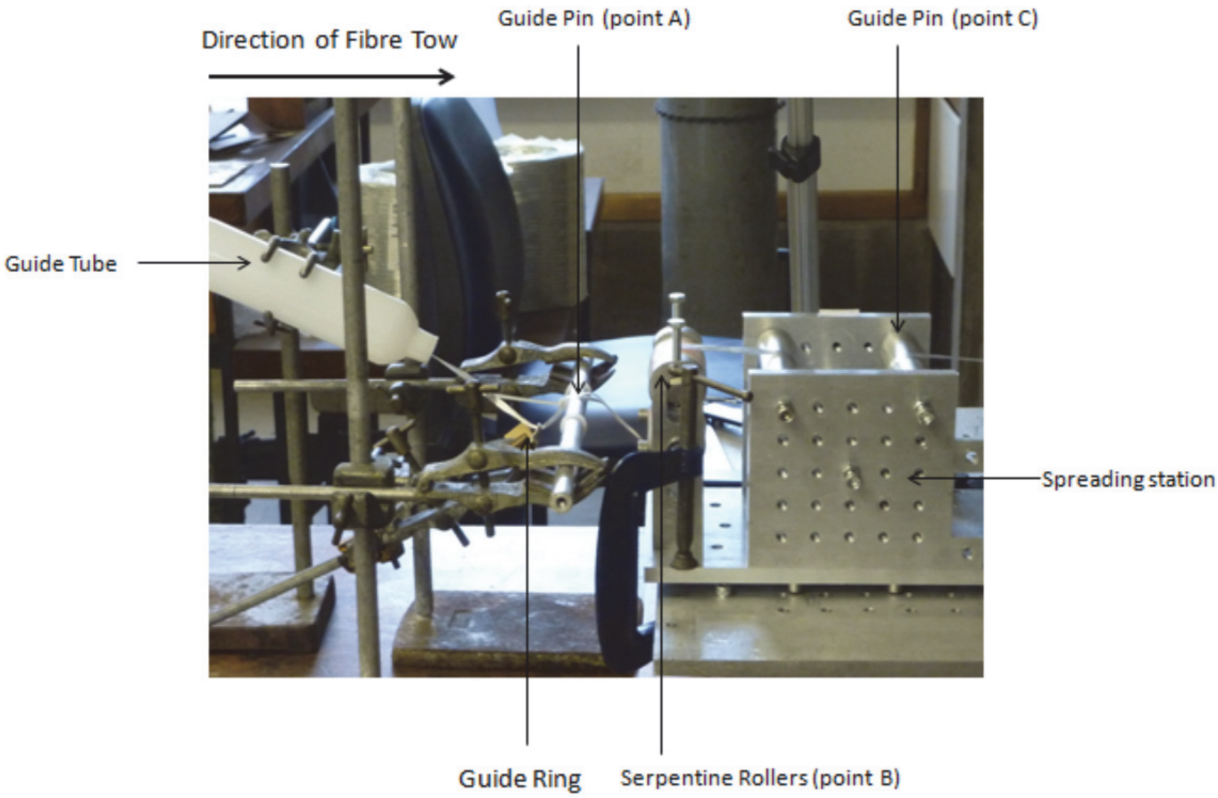


Figure 11 Photograph showing serpentine fibre spreading unit set-up, measuring points and key features.

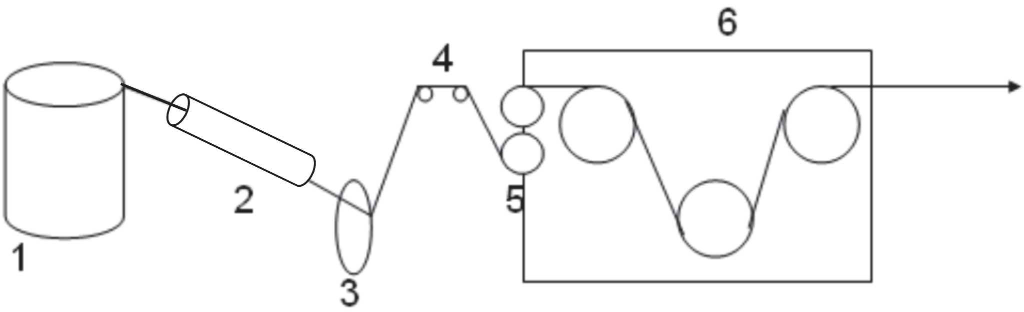


Figure 12 Schematic illustration of the serpentine fibre spreading unit. The coded items are: (1) creel; (2) guide tube; (3) guide ring; (4) guide point; (5) serpentine rollers; and (6) spreading station.

All other roller configurations involved were a combination of plain and profiled rollers. Figure 13 shows the combinations of profiled and plain rollers, the profiled rollers are spotted black and plain rollers are in white.

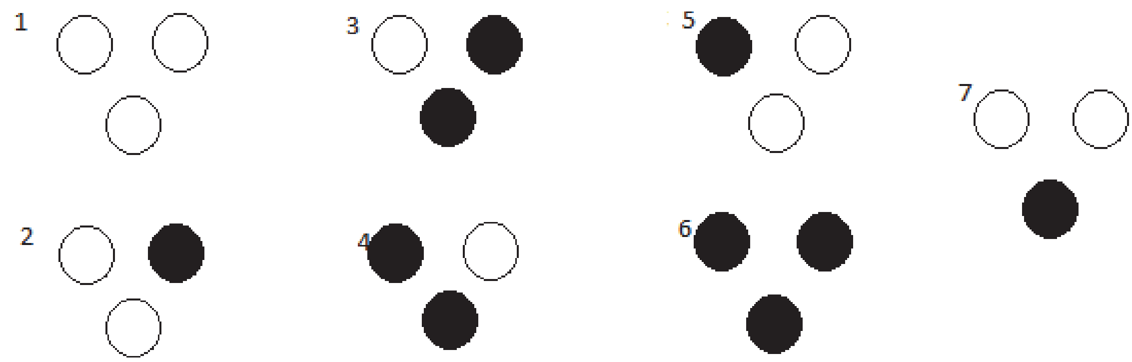


Figure 13 The configurations used in the serpentine fibre spreading set up using both plain (open circle) and profiled rollers (filled circle).

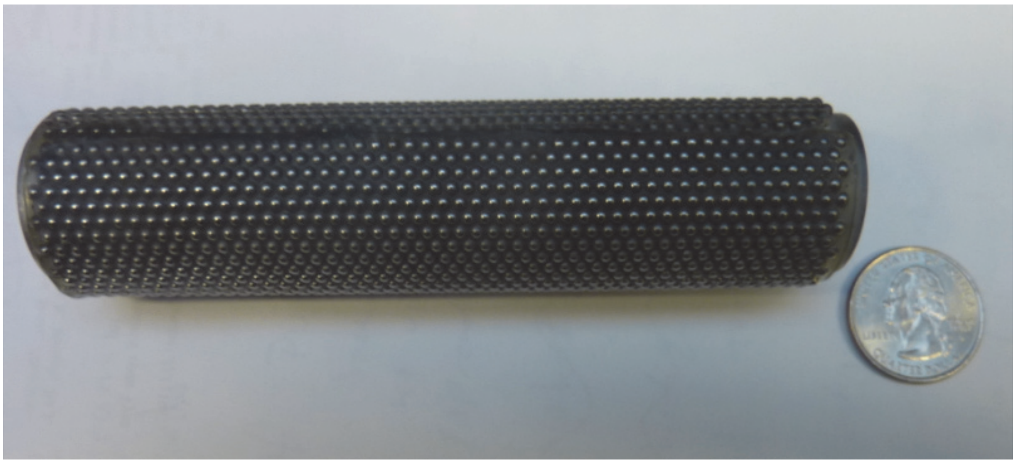


Figure 14 Photograph of a profiled roller. The roller was manufactured by Ramani Mahendaran, Sensors and Composites Group, University of Birmingham, 2012.

The profiled rollers were produced by the Sensors and Composites Group at the University of Birmingham. The profiled rollers consisted of aligned ball bearings that were surface-mounted on an aluminium rod. A silicone mould was made to cast a thermosetting reason. The resin was then part-cured and cooled. This 'sheet' was then wrapped around an aluminium rod and the ball bearings were pressed into the soft resin. This assembly was put in the oven to cure the resin.

The optimum values achieved previously from the Taguchi analysis in the tension-release experiments were used in the serpentine fibre spreading unit. Only one pull-speed and tension was used, the other parameters were fixed. The pull-speed was 1 mm/min and the tension was

2 N. With reference to Figure 11, the measurements were taken at: The guide pin (point-A), pinch roller (point-B), and the exit roller (point-C). The precautions were taken at the start of the experiment to ensure that no length of fibre was measured more than once, similar to that stated previously for the tension release rig. Figure 11 shows the set-up of the serpentine fibre spreading unit and its relevant key features and the measuring points for the fibre widths. The glass fibre was passed through the guide tube to control the trajectory of the fibre bundle from the creel to the serpentine spreading station; this was also the case with the guide ring to keep the glass fibre straight and pointing at the spreading station. The glass fibre was then passed over the guide pin (first measuring point) and through the serpentine rollers (second measuring point). From the serpentine rollers the glass fibre tow was pulled onto the spreading station where there were the 3-roller pins (profiled or plain). The final measurement of the spread fibre was taken at the exit roller (point c).

3.2.3. Multi-tow spreading

With reference to the serpentine fibre spreading unit configuration-2 was chosen as this was the best combination as obtained experimentally. Configurations-1and -6 were also used in conjunction with configuration two to measure the spreading of multiple tows. The tows were measured at three points: guide pin (point A); pinch roller (point B); and the exit roller (point C). All the other parameters were kept the same. The number of tows used was 3, 6, 9 and 12. Wider-pinch rollers were made to accommodate the increased number of tows. The wider pinch rollers were set up in the same way as described previously.

3.3. TWIST IN THE FIBRE BUNDLE

A short experiment was carried out to investigate the effect of twisted fibres on the extent of spreading of the neighbouring glass fibres. The serpentine fibre spreading unit was used and configuration-2 was for the reasons stated previously. The fibres were attached to the haul-off machine and the machine was switched on. When a twist was observed the haul of machine was stopped when the twisted fibre reached the serpentine rollers. The centre of the twist was then marked and the extent of spreading at the final roller was measured; the distance between the serpentine rollers and the final roller was 150 mm. These tests were repeated twenty times.

A second batch of experiments was carried out on three tows to observe if the twist of fibres has an effect on the neighbouring fibres. As before, the system was allowed to run until a twist was seen going through the pinch rollers, at this point the haul-off winder was stopped and the tows were marked and measured. The haul-off unit was then switched on until the marked tows reached the top of the pinch rollers when the winder was stopped and the widths of the tows were measured. This test was repeated twenty times.

3.4. IMPREGNATION MONITORING

A summary of the materials and equipment used in the experiments to monitor the impregnation of E-glass is summarised in Table 6.

Table 6 Materials and equipment used for the impregnation monitoring of E-glass fibres

Materials and Equipment
2400 tex E-glass fibres
Norland optical adhesive 63 (NOA 63)
Two Veho USB microscopes
Observation rig
Spreading station of the serpentine fibre spreading unit
Haul of winder provided by Pultrex LTD
Thorlabs UV75 light gun

A single fibre bundle was traversed through the serpentine fibre spreading unit, and was tied to the haul-off winder, making sure no twists in the fibre were present. The haul-off rig was set to operate at 1 m/min until a section of spread fibre was 8 mm or over at the observation section. The two cameras were set below and above the glass fibre to view impregnation from both sides; both cameras were focussed on a graticule and set to record the video. A syringe dispenser was used to drop a controlled amount of adhesive onto the centre of the fibre bundle. A UV curable resin (NOA 63) was used in these studies. As soon as the drop was dispensed onto the surface of the fibre bundle, a stopwatch is started and the cameras were allowed to record. The times used to analyse the progression of the impregnation were; 10, 20, 30, 40, 50, 60, 120, 180, 240, 300 and 600 seconds. After the impregnation for a specific time, the UV-gun was used to cure the resin. The fibre bundle was then cut and both ends of the fibre were taped to secure the ends of the bundle. Videos and images were also saved for analysis. Food colouring was used to create a contrast between the cured resin and the un-impregnated sections of the fibres.

3.5. PULTRUSION

3.5.1. Pultrusion machine

A pultrusion machine from Pultrex Ltd (model PX100C-3T) was used to manufacture glass fibre composites. The pultrusion machine had a maximum pull-force of 3 tons and used cleat-type tractor puller system for hauling off the composite.

3.5.2. Conventional resin bath-based pultrusion

Conventional resin bath-based experiments were performed to obtain the reference samples. Here a dip-type resin bath with a capacity of five litres was used. The fibre bundles were threaded through the back plate, plunger, pre-shaping guide plate and die one at a time. Initially the bundles were pulled through the die manually but after twenty rovings had been threaded the puller was used. The rovings were taped with a cello-tape before passing through the puller.

Once all the rovings were made to pass through the die, the heaters were set to the 160 °C. The components of the reins were mixed in a container manually and were poured in the resin bath. The pullers were started to commence the pultrusion experiment. Once the cured composites passed through the puller, these were labelled and cut into 1.5 m for testing. The resin bath, plunger, pre-shaping guide plates and resin feed-back tray were cleaned with acetone in a fume cupboard. The pultrusion speeds used were 0.3 m/s, 0.4 m/s and 0.5 m/s.

3.5.3. Clean pultrusion

Two custom-built resin impregnators (Prototype-I and Prototype-II) were used to impregnate the rovings in the clean pultrusion experiments. These resin impregnators were designed by the Sensor and Composites Group, University of Birmingham.

The fibre bundles were threaded through the back-plate, fibre spreader, resin impregnator, die and puller as described for the resin bath. The resin was deposited onto the spread fibre bundles via a resin injector as shown in Figure 15.

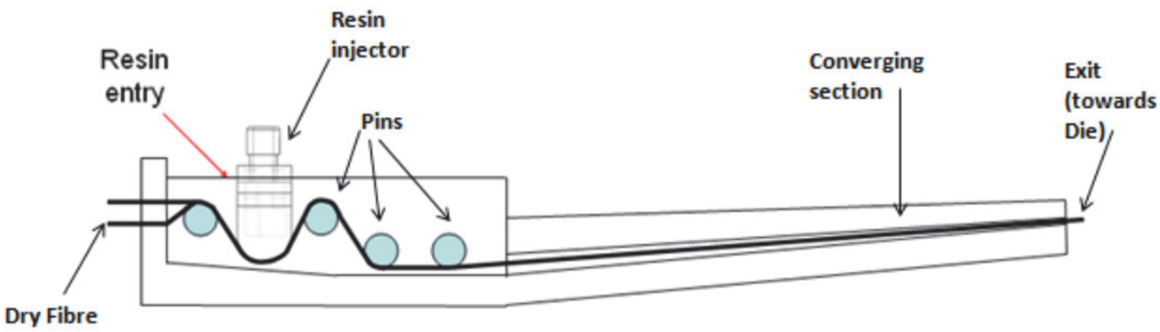


Figure 15 Schematic illustration of the trajectory through the resin impregnator Prototype-I. Illustration by Mark Paget, University of Birmingham, UK.

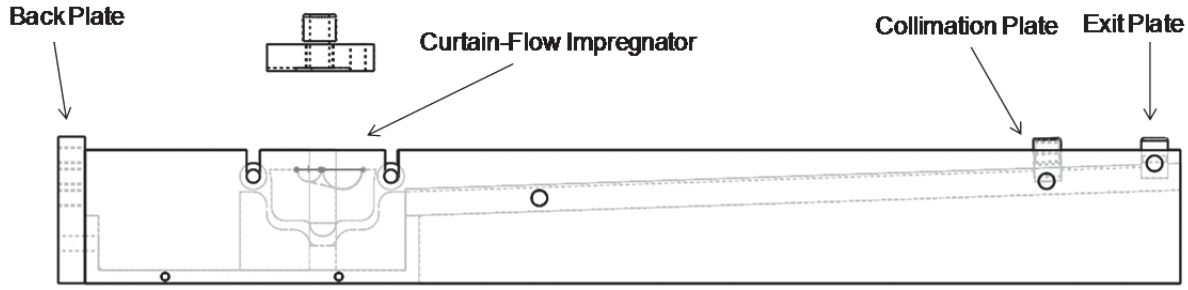


Figure 16 Schematic illustration of the trajectory through the resin impregnator Prototype-II. Illustration by Mark Paget, University of Birmingham, UK.

The fibres were then drawn through a heated die which is set at 160 ^{o}C. The cure composites from the die were pulled, labelled and cut as described previously for the resin bath. The composites were produced at three pultrusion speeds (0.3 m/s, 0.4 m/s and 0.5 m/s).

3.5.4. Post- curing of the pultruded samples

After samples were labelled and cut, three samples from each set were post-cured in the oven at 70 ^{o}C for 4 hours. These post-cured samples were then compared to the standard samples with regard to mechanical properties and image analysis.

3.6. EVALUATION OF PULTRUDED COMPOSITES

3.6.1. Density

Five Samples for each specimen were cut and edges were polished with 400-grit emery paper to remove sharp edges caused by cutting (ASTM D2734). The density was measured using a density determination kit AP250D. The weight and buoyancy of the samples were measured; also the water temperature was taken. Equation six was used to determine the density of each sample (ASTM D792-00). Figure 17 shows the equipment used to measure density.

Procedure for measuring the density was as follows:

(i) Measure and record the temperature, then weigh the specimen in air.

(ii) Mount the immersion vessel on the support and immerse completely the suspended specimen in water. The vessel must not touch the sample holder or specimen. Then record the mass.

(iii) Weigh sample holder in water and record this weight.

The procedure was repeated a minimum of two times per sample.

The following formula was used to calculate the densities (ρ) of the neat resin and the pultruded composites:

$$\rho = \frac{W_{air}}{W_{air} - W_{water}} \times \rho_{water} \qquad \textbf{Equation 2}$$

where W_{air} is the weight of the sample in air, W_{water} is the weight of the sample in the water and ρ_{water} is the density of the water at 22 °C. An average of five samples was used for the density measurements.

Figure 17 Photograph showing the equipment used to measure the density of the pultruded composites.

3.6.2. Fibre volume fraction

The fibre volume fraction was determined using ASTM D2584. A ceramic crucible was used for each sample. The crucible was first weighed then the sample was placed in the crucible and weighed again. The crucible with the sample was then placed in a furnace at a temperature of 565 °C and heated for six hours. Five samples were placed in the furnace at one time. After the six hours at 565 °C, the furnace was switched off and the samples were left in the furnace overnight to cool to ambient. The samples were then weighed again, and the mass-loss was recorded.

The fibre volume fraction of the composite (V_f) was calculated using following relationship:

$$V_f = \frac{\rho_m W_f}{\rho_f W_m - \rho_m W_f} \qquad \textbf{Equation 3}$$

where W_f is the weight of the fibres, W_m is the weight of the matrix, ρ_f and ρ_m are the densities of the fibres and the matrix respectively.

3.6.3. Void fractions

The void fraction (V_v) was determined in accordance with ASTM D 2734-03 using the following relationship:

$$V_v = \frac{100(\rho_T - \rho_c)}{\rho_T} \qquad \textbf{Equation 4}$$

where ρ_T and ρ_c are the theoretical and measured densities of the composite respectively. The theoretical density of the composite (ρ_T) was calculated using:

$$\rho_T = \frac{100}{\left(\frac{M}{\rho_m} + \frac{R}{\rho_f}\right)} \qquad \textbf{Equation 5}$$

where M and R are the percentage weights of the matrix and the fibres respectively in the composite.

3.6.4. Image analysis

An optical microscope (Axioskope 2) is used to view the surface of the glass fibre composites. The pultruded composites were cut, mounted and polished using conventional procedures. The polished samples were inspected to determine the fibre alignment and to infer the void content.

3.6.5. Instron machine

An Instron machine (Model 5566) capacity of 10 kN was used to obtain the tensile strength, flexural strength and modulus, interlaminar shear strength of the pultruded sample.

3.6.6. Inter-laminar shear strength

The inter-laminar shear strength (ILSS) was calculated according to tests were carried out in accordance with ASTM D2344. The main objective of the short-beam test is to maximise the induced shear stress by reducing the support-span-length-to-specimen-thickness ratio to 4:1.

Ten samples were tested for each type of pultruded composite. The length, width and thickness of the pultruded samples were 20 ± 0.2 mm, 10 ± 0.2 mm 2 ± 0.2 mm respectively. The sample was loaded in 3-point bending to measure the interlaminar shear strength. The load was applied by means of a loading 'nose' on the centre of the midpoint of the test samples. The span length of the two supports were fixed and were set 12.0 mm apart and the loading nose above was set at 6.0 mm. The speed of testing was set at a rate of cross-head movement of 1.0 mm/min. The inter-laminar shear strength (ILSS) was determined as follows:

$$ILSS = 0.75 \times \frac{Pm}{b \times h}$$ **Equation 6**

where P_m is the maximum load observed during the test, b is the specimen width and h is the measured specimen thickness.

3.6.7. Flexural testing (four-point bending)

The flexural tests (four-point bending) were carried pout according to the method stated in ASTM D6272-02. Seven samples were cut for each batch of pultruded sample using the water cooled diamond wheel cutter. The dimension of the test specimen was of 50 mm × 12.7 mm × 2.2 mm. A strain rate of 0.01 mm/min was used with a support span-to-depth ratio of 16:1. The experimental set up with regard to the position of the loading configuration on the sample is shown in Figure 17.

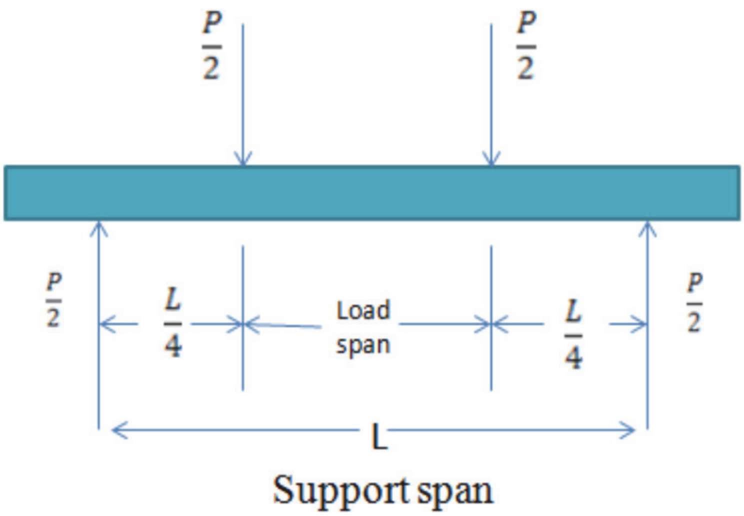


Figure 18 Schematic Illustration of the four point bending set up loading points.

Following equations were used for calculating the flexural strength and stiffness:

$$S = 3PL / 4bd^2$$ **Equation 7**

where S is the stress, P is the load, L is the support span, B is the width, D is the depth.

$$E_B = 0.17L^3 m / bd^3$$ **Equation 8**

E_B is the modulus of elasticity in bending, L is the support span, b is the width, d is the depth, m is the slope of the tangent to the initial straight line.

3.6.8. Tensile testing

The tensile tests were carried pout according to the method stated in ASTM D3039M.The dimensions of the composites specimen were 250 mm × 25 mm × 2.5 mm. Aluminium end-tabs were used to grip the composite and the aluminium end-tabs were sand blasted and then cleaned using IPA. The resin and hardener (Scotch-weld 9323) were weighed in the required ratio mix and then applied onto the tabs. This assembly was then clamped at each end to hold the tabs and placed in the oven for two hours at 66 ^{o}C. The sample was loaded at a cross-head displacement rate of 2 mm/min. Electrical resistance strain gages were attached onto the surface of the composite.

The tensile strength was calculated using the relationship:

$$\sigma = \frac{F}{A}$$ **Equation 9**

where σ is the tensile stress, F is the peak force, A is the initial cross-sectional area of the specimen.

The strain was calculated using the relationship:

$$\varepsilon = \frac{\Delta L_o}{L_o}$$ **Equation 10**

where ε is the strain, L_o is the gauge length, ΔL_o is the increase in specimen length between the gauge marks.

4. RESULTS AND DISCUSSION

4.1 FIBRE SPREADING EXPERIMENTS WITH FIBRE SPREADING RIG

The first set of results was gathered from the fibre spreading rig, where 18 experiments were carried out. These experiments were carried out using the experimental matrix given in Table 5. The results showed that maximum fibre spreading achieved at point 1 (pinch rollers) and point 3 (exit roller). Figure 19 shows the average values from the two points for each of the 18 experiments.

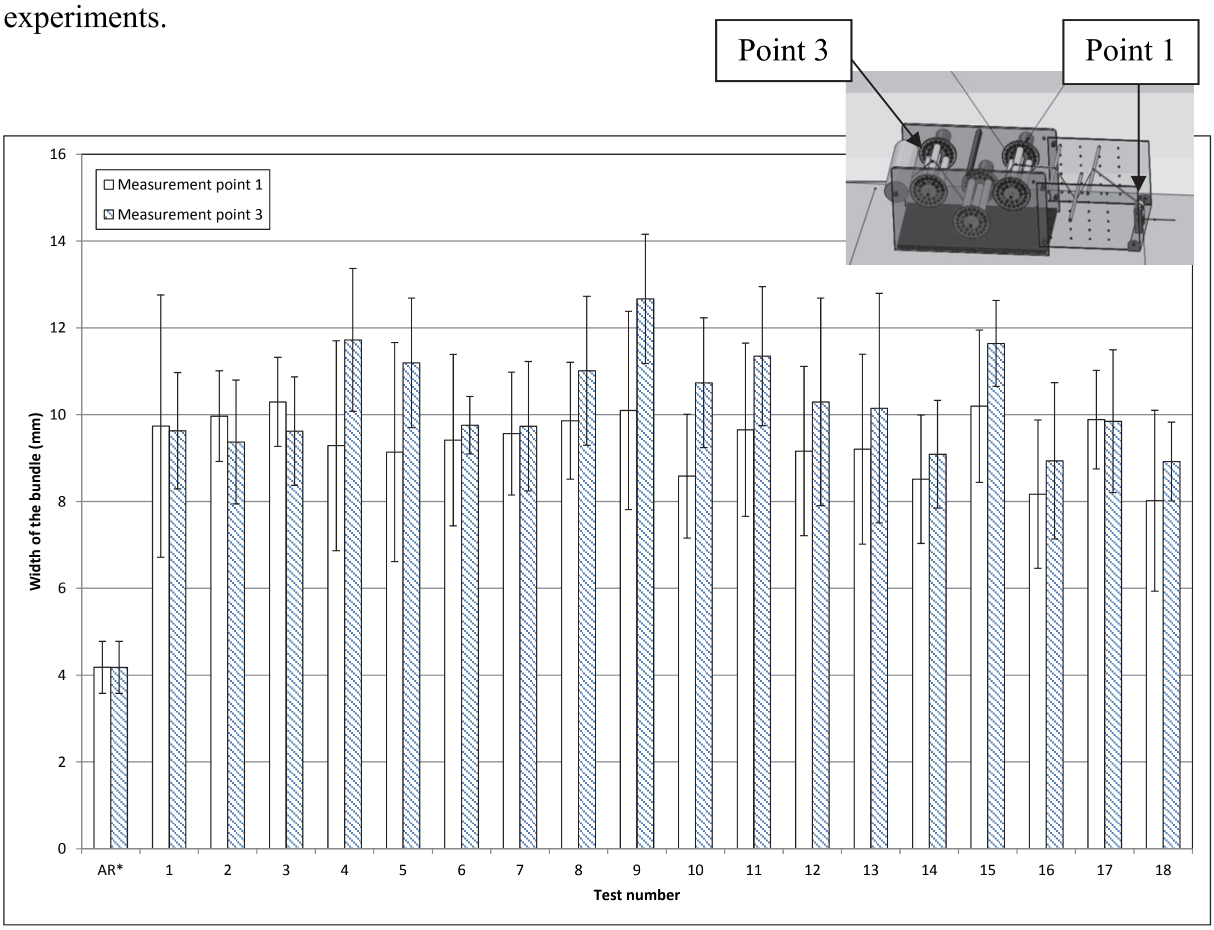


Figure 19 Graph showing the average fibre widths at point 1 and point 3 for each of the 18 experiments enlisted in Table 5.

The highest values achieved during the fibre spreading experiments were found at the exit roller and was measured at 12.7 mm. This corresponds to experiments number 9 as shown in Figure 19. With regard to the degree of spreading, configuration-3 (Figure 10) was observed to give the best results along with a disk rotational speed of 100 rpm, pre-tension value of 2 N and a winding speed of 1 m/min.

The configuration of the roller-disk assembly with three rollers (Figure 10) provided the highest value of fibre spreading. For three roller setup, the configuration is repeated after a disk-rotational angle of 120°. Here both tension and tension-release acts uniformly on a unit length of the fibre roving which results in improvement of fibre spreading. The value of the pre-tension is important in the continuous production process such as pultrusion. If the pre-tension is on lower side, the rovings may meander and entangle. On the other hand, at high values the rovings may get damaged. Higher pre-tensions can also be detrimental for impregnation due to the compactness of the roving, thus restricting the flow of the resin. It was also noted that a lower fibre haul-off speed was required for better fibre spreading. This is because more residence time is available to the roving to experience the tension-release action. Finally, it was also observed that a higher rotational speed of the roller-disk improves fibre spreading. This is related to the increase in the tension-release action per unit length of the roving per unit time.

At the pinch roller, the disk rotation and the configurations does not have an effect as the pinch rollers are positioned ahead of any of the parameters in the tension-release system. The tension-release system requires a great amount of space and it is time consuming when it is necessary to change configurations according to the Taguchi matrix. Moreover, since each disk is powered electrically, independent power supplies are required. On the other hand, just having the pinch rollers would be cost-effective; hence the decision was taken to evacuate a simple pinch-roller system as a means of spreading the fibres.

4.2 FIBRE SPREADING EXPERIMENTS WITH THE SERPENTINE FIBRE SPREADING RIG

The serpentine fibre spreading unit was used for the second set of spreading experiments. This is a smaller system than the tension-release fibre spreading unit. The serpentine fibre spreading unit consisted of two different types of rollers; plain and profiled. A total of seven configurations were used, which included two extremes; configuration-1, which had all plain rollers and configuration-6, which were all profiled rollers. The optimum values for tension and winding speed selected were similar to those in the Taguchi method.

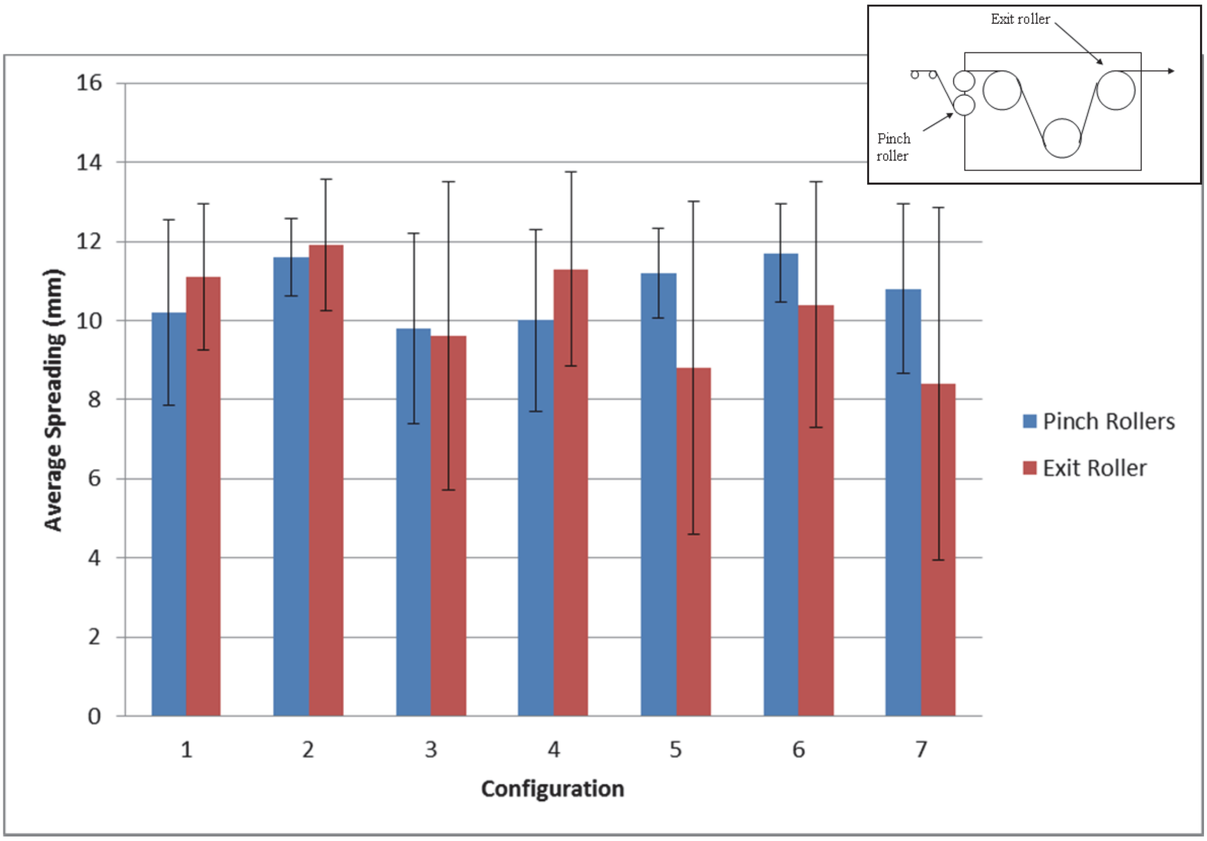


Figure 20 Summary of the average fibre spreading measurements at the pinch and exit rollers when using the serpentine fibre spreading unit.

It can be seen from Figure 20 that configuration-2 achieved the best fibre spreading results, followed closely by configuration-1 which were all plain rollers. The average spreading at the pinch rollers was 9.64 mm whereas the average measurement at the exit roller was 8.92 mm. These averages can be compared to the results obtained from the tension release-system and the same question can be asked; if a series of pinch rollers would be more effective than the present methods for the spreading of a single fibre bundle. Although it can be seen that the exit roller in configuration-2 achieved the highest spreading measurement, it is apparent that 3 of the 7 configurations recorded below 100% fibre spreading. However, it was observed over the course of this study, the pinch rollers regularly produced 100% spreading on average. This consistency of spreading from the pinch rollers is also apparent in the smaller standard deviation values shown in comparison to the exit rollers. The exit rollers show a greater variation in spreading (configurations-3, -5 and -7). This variation can be due to the twists in the fibre bundle.

4.3 MULTI-TOW SPREADING

The serpentine fibre spreading rig was used as the spreading station to determine the degree of spreading when using multiple tows. Three configurations were used (configurations-1, -2 and -6). Configuration-2 was also used as this gave the best spreading measurements during the spreading of a single fibre tow in the serpentine fibre spreading set up. Initially the experiment was meant to be first conducted with 3 tows leading up to 6, 9 and then 12-tows. The first problem encountered during this experiment was that the rollers being used in the spreading station were too small to fit and spread more than 6 tows, therefore the spreading experiments of 9 and 12 tows could not be carried out.

Three tows were first pulled through the station; it was seen that the average fibre spreading was less than spreading with one tow. Then the number of tows was increased to 6, and it was seen that the baseline tension increased, which resulted in a decrease in average spreading. Therefore it is speculated that if 9 and 12-tows were able to be carried out in this experiment, then the average spreading would be less than using 3 or 6 tows. The effectiveness of the spreading when multiple tows are used diminishes if the thickness of the tow is different. The effects of randomly occurring twists in the fibre will have an adverse effect per tow on the average spreading. Results of the fibre spreading at the pinch roller and exit roller were analysed. Figure 21 shows the results at the pinch roller. The results show that the fibre width decreases from 1 tow to 6 tows; this is part-caused by the increased tension at the pinch rollers. It also shows that configuration-6 was the best out of the three. The standard error bars show that 100% spreading is still achievable for most configurations and tows from the as-received position.

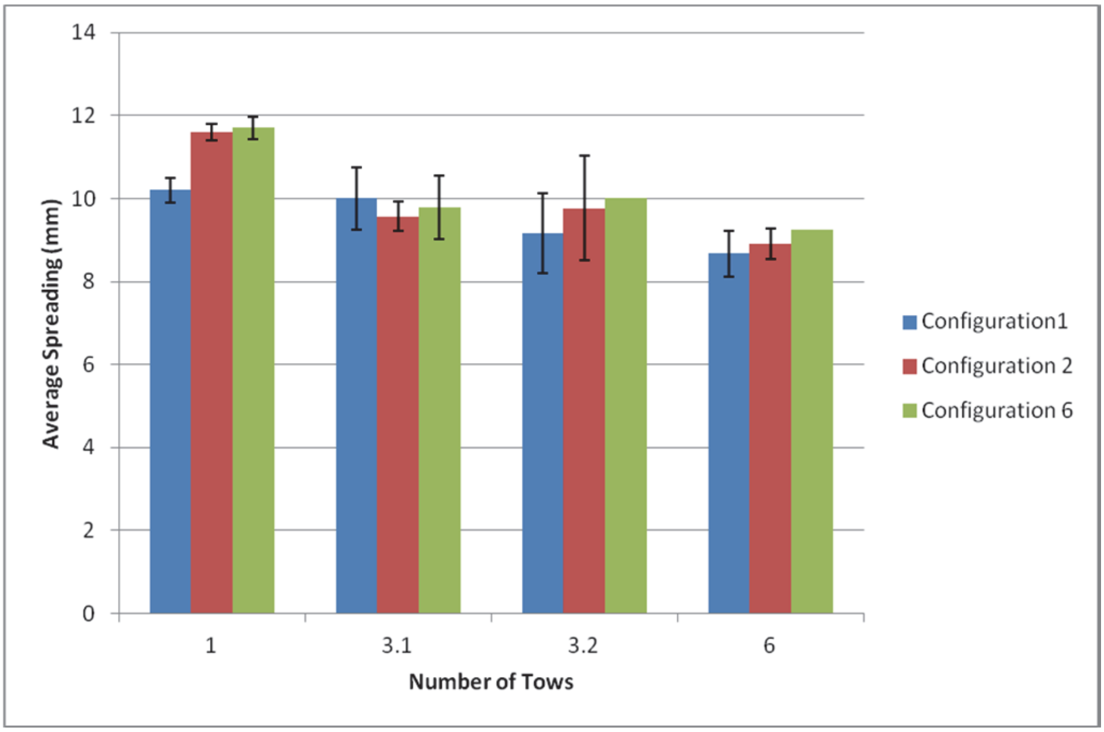


Figure 21 Measured extent of fibre spreading at the pinch rollers, for configurations-1, 2 and 6 for trials with 1, 3 and 6 tows.

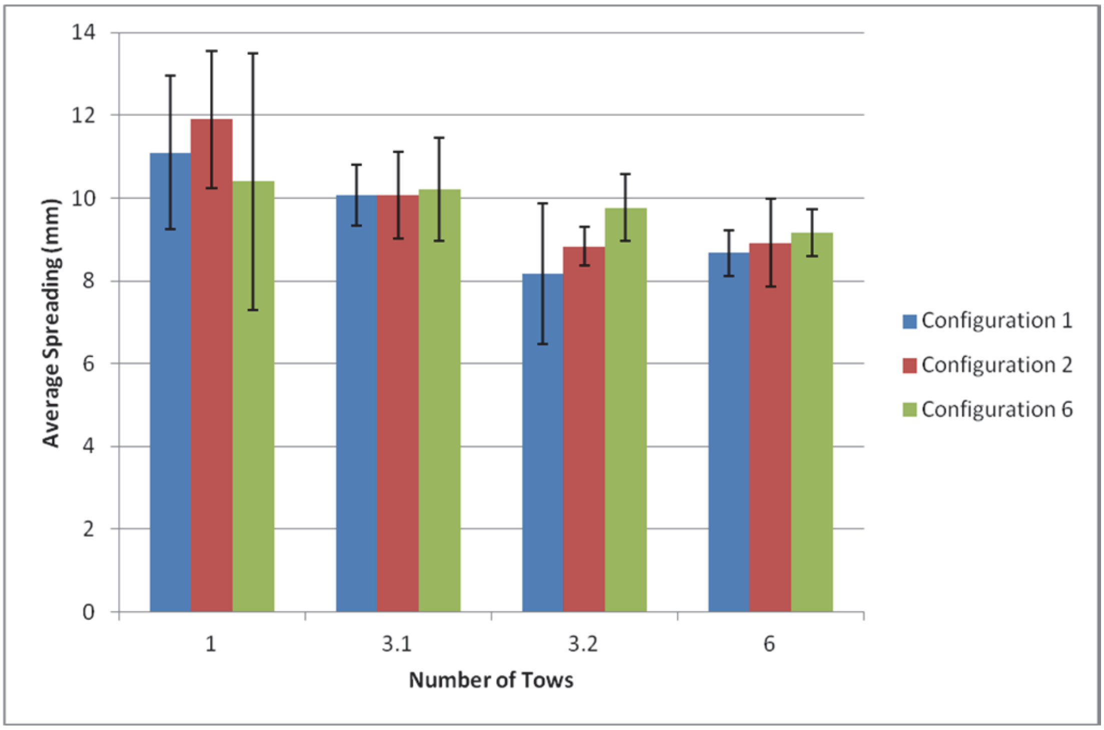


Figure 22 Spreading at the exit rollers, for configurations-1, 2 and 6 for trials with 1, 3 and 6 tows.

Figure 22 shows the spreading at the exit roller. It can again be seen that the spreading at the exit rollers decrease due to the increased tension as more tows are added. The tension appeared to be greater at the exit rollers because of the spreading station and pinch rollers experiencing an increased tension. It is seen from Figure 22 that configuration-2 resulted in the greatest spreading; however as the number of tows increased, configuration-6 (profiled rollers) became the most favourable configuration. Also seen with an increase in tows there was a decrease in the standard deviation.

It can be seen that the measurements from the pinch rollers are very similar to the data obtained from the exit rollers; this was also observed for data associated with the tension-release system. The variance from the pinch rollers is much smaller than the variance at the exit rollers and gives the idea of testing the spreading of fibre tows using a series of pinch rollers to achieve the similar if not better data.

4.4 TWIST IN THE TOW

Twists in the fibre tow can occur randomly as the fibre tow is pulled from centre of the creel. It is difficult to predict to as when a twist will occur, and so the degree to which a twist in a tow affects the fibre spreading was analysed.

Figure 23 shows the spreading at the pinch rollers and the length of the fibre tow affected by the twists. The spreading at the roller show 100% spreading and the average spreading was 11.15 mm. The distance of a twisted fibre tow was not consistent and the values varied from 75 mm to 130 mm.

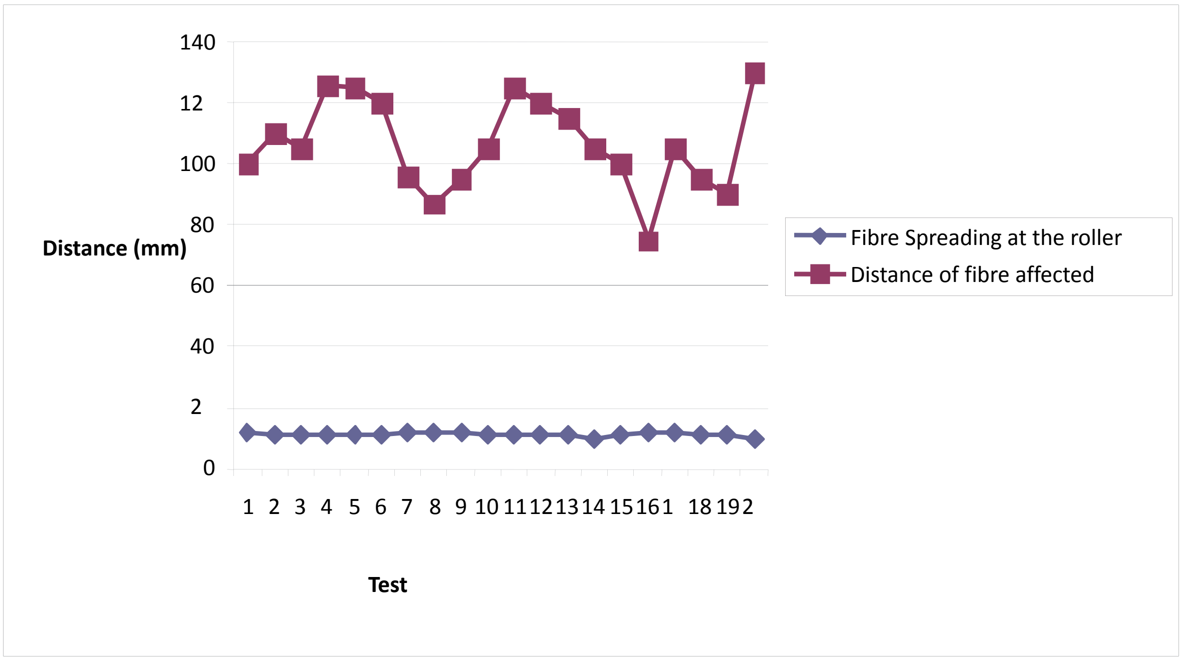
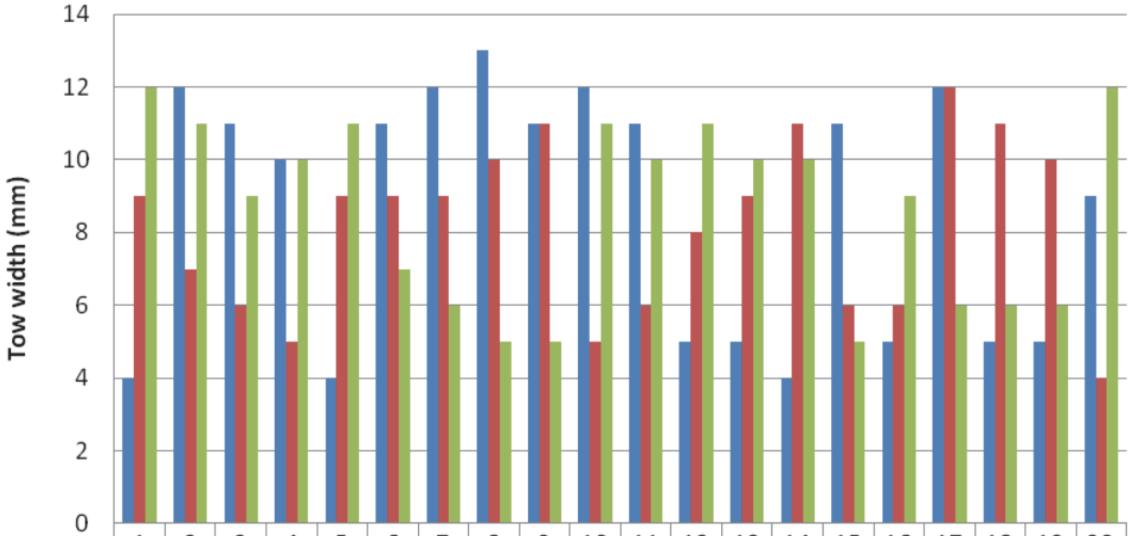


Figure 23 Fibre spreading at the pinch roller and the distance of fibre affected by a twist.



	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20
Tow 1	4	12	11	10	4	11	12	13	11	12	11	5	5	4	11	5	12	5	5	9
Tow 2	9	7	6	5	9	9	9	10	11	5	6	8	9	11	6	6	12	11	10	4
Tow 3	12	11	9	10	11	7	6	5	5	11	10	11	10	10	5	9	6	6	6	12

Figure 24 The effect of twists on multi-tow spreading

Figure 24 shows that as a twist occur in one tow, the neighbouring fibre tow still reached or were very close to reaching 100% spreading. During the experiment when multiple tows showed twists, the untwisted fibre was not affected by the neighbouring twisted fibres. It can be seen that from the results that the neighbouring fibre tows of a twisted fibre are not hugely or not affected at all by the twists. Sample 1 show that tow-1 has a twist however tows-2 and 3 still reached 100 % spreading. Samples 15 and 16 show two tows that are twisted but this has not affected the neighbouring tow.

4.5 IMPREGNATION STUDIES

Images from the top and bottom of the samples were used and measurements were taken in the longitudinal, transverse and through-thickness direction. Resin flow in the transverse and longitudinal directions was calculated using the top image; the through-thickness impregnation was calculated using the bottom image. The experiments carried out using 2400 tex Hybon 2026 E-glass fibre bundle. Both of these types were used in there as-received state and were also spread using the serpentine fibre spreading unit

Results show that with an increase in impregnation time, there was an increase in longitudinal impregnation; however the transverse impregnation remained relatively constant. During the impregnation of as-received fibres there was no observation of through thickness impregnation up to 30-45 seconds, however it weren't until 50 seconds that the fibres showed through thickness impregnation. During the analysis, it was decided to concentrate on the results obtained for impregnation times of 0-120 seconds.

Following the experiments with as-received fibres, spread fibres were obtained from the serpentine fibre-spreading unit. The same parameters were used to spread the fibres that were used in the serpentine fibre spreading experiments. Samples were spread to a width of 8 mm. The samples were then impregnated and analysed. It was found that there was an increase in longitudinal impregnation and through-thickness impregnation with increasing impregnation time. However, the transverse impregnation remained constant. The results show that spreading the fibres shows a better rate of impregnation when compared to the as-received fibres. Through-thickness impregnation for spread fibres and as-received fibres is 30 and 50 seconds respectively. Wilson (1997) supports this as he states that by spreading the fibre tow, the tow width increases, whilst decreasing tow thickness which aids impregnation. As through-thickness impregnation increases with the spread fibres there was a decrease in longitudinal impregnation in comparison to the as-received fibres. This is understandable as the drop of the resin that is placed on the surface of a spread bundle will permeate it faster. Thus the driving force for the axial impregnation will decrease.

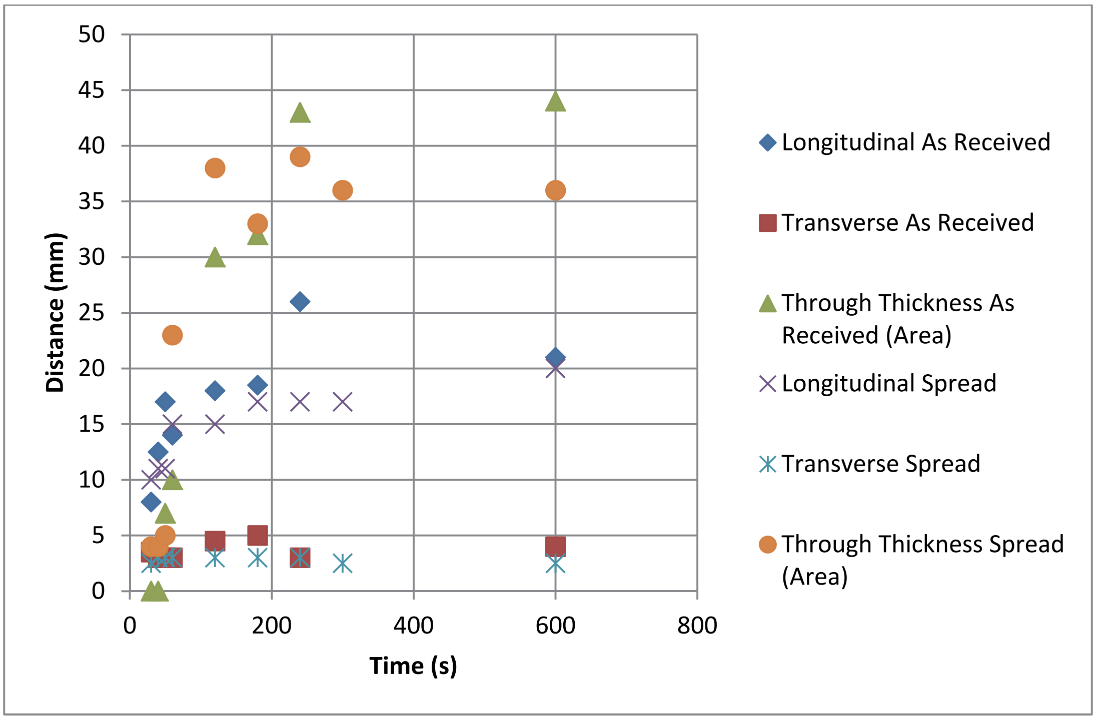


Figure 25 The distance of resin flow-front as a function of time using PPG fibres as-received and fibre spread

4.6 EVALUATION OF THE PULTRUDED COMPOSITES

The physical (density, fibre volume fraction) and mechanical (flexural strength/modulus, inter-laminar shear strength, tensile strength) properties of the composites produced from the clean and conventional pultrusion were evaluated to compare two processes.

4.6.1. Density and fibre volume fraction

Figure 26 shows the density and fibre volume fraction data for the resin bath, impregnator Prototype-I and II. Three different pultrusion speeds were used 0.3, 0.4 and 0.5 m/min. The densities of the samples range between 2053 to 2228 kg/m^3.

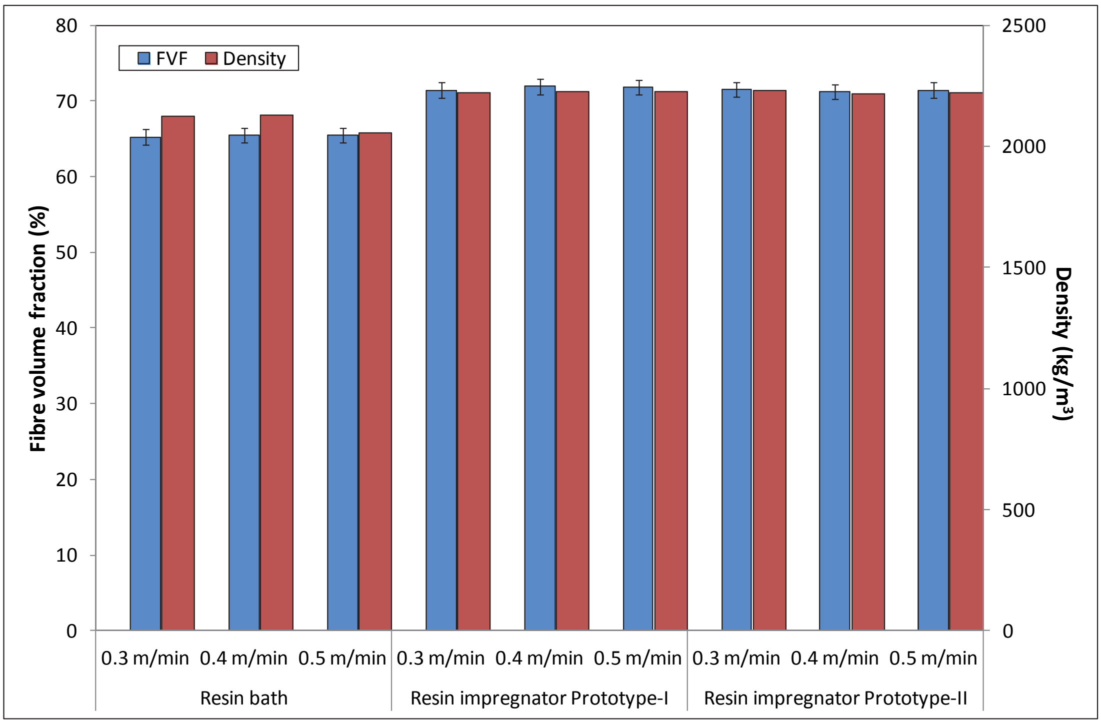


Figure 26 Fibre volume fractions and densities of the pultruded samples obtained from resin bath, I and II impregnators at three pultrusion speeds.

The percentage fibre volume fraction varies between 65 and 71 % fibre volume fraction. The resin bath shows the lower fibre volume fraction and the lowest density results, this is because a lower number of fibre tows (50 tows) were used during the pultrusion process as compared to Prototype-I and II which use 52 tows. Prototype-I and II show similar density ranges throughout. The practical limit of the fibre volume fraction in polymer composites is around 70 % (Hull and Clyne, 1996). It can be seen that this limit is reached when resin impregnator Prototypes-I and II are employed.

4.6.2. **Void fraction**

The presence of voids in a composite will decrease the mechanical properties (Judd and Wright, 1978). Figure 27 shows the percentage void fractions of the three impregnators used alongside three different pultrusion speeds.

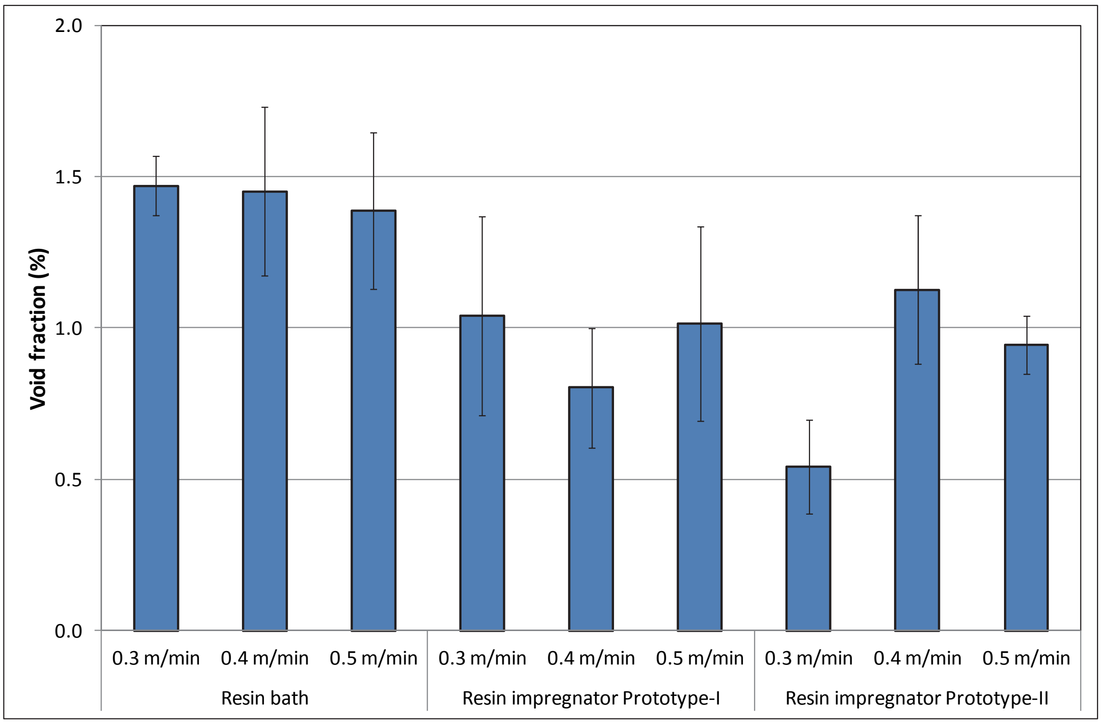


Figure 27 Void fractions of the pultruded samples obtained from resin bath, I and II impregnators at three pultrusion speeds.

Prototype-I and II impregnators show lower void fractions compared to the resin bath. However there is no conclusive relationship for the voids against the pultrusion speed.

The resin bath shows a higher void content across all speeds. There was a noticeable reduction in void fraction when the fibre spreading rig was used prior in impregnation. The reduction in voids in the “Clean” pultrusion can be attributed to breaking-up of the binder in the fibre bundles due to fibre spreading process. The binder in the fibre bundles can exist in the form of lumps and can impede the resin impregnation process.

4.6.3. Image analysis

Figure 28 show images taken from the resin bath samples and the resin impregnators. With reference to Figure 28, the following conclusions were reached after inspecting each individual section.

In general, the voids content in the resin bath samples were observed to be higher when compared to those pultruded using the resin impregnator.

The void contents in the samples pultruded using resin impregnator Prototype-I was seen to have the lowest void content when compared to the other two techniques.

Increasing the pultrusion speed was seen to enhance the relative void content for the three impregnation methods. This is due to the reduction in the residence time of the impregnators.

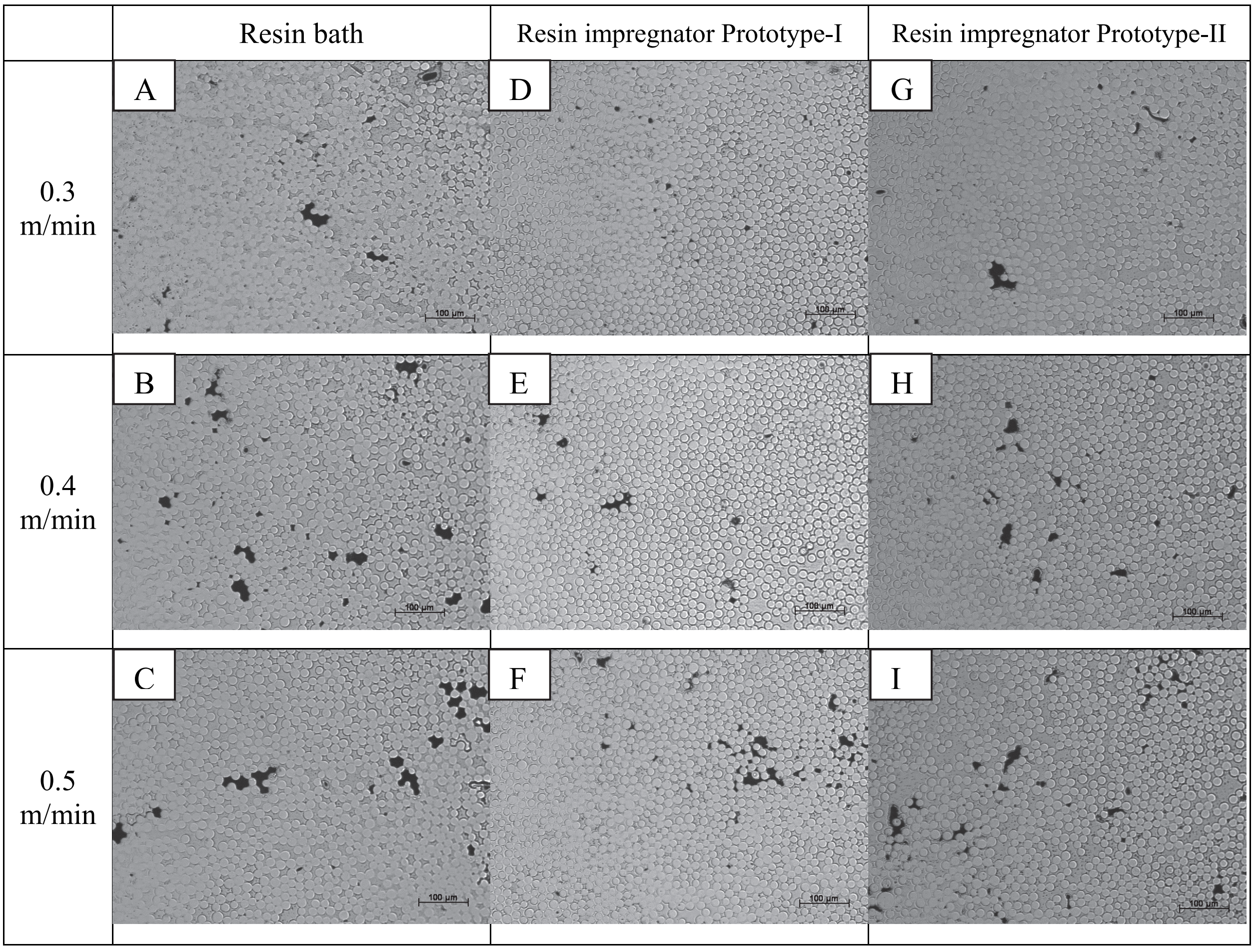


Figure 28 Optical micrographs for pultruded samples manufactured sing the resin bath, resin impregnator Prototype-I and resin impregnator Prototype-II.

4.6.4. Inter-laminar shear strength (ILSS)

The inter-laminar shear strengths (ILSSs) for the conventional and clean composite are shown in Figure 29. The normalised values (at 60% fibre volume fraction) are also shown. The results show an increase in the inter-laminar shear strength from the resin bath to impregnators-I and II. The results range from the lowest 70 MPa to 85 MPa. The resin bath shows the lowest ILSS, the composites produced from the resin bath indicated a larger number of voids compared to the other two impregnators. The presence of defects such as voids can cause considerable scatter in ILSSs (Wisnom et al, 1996).

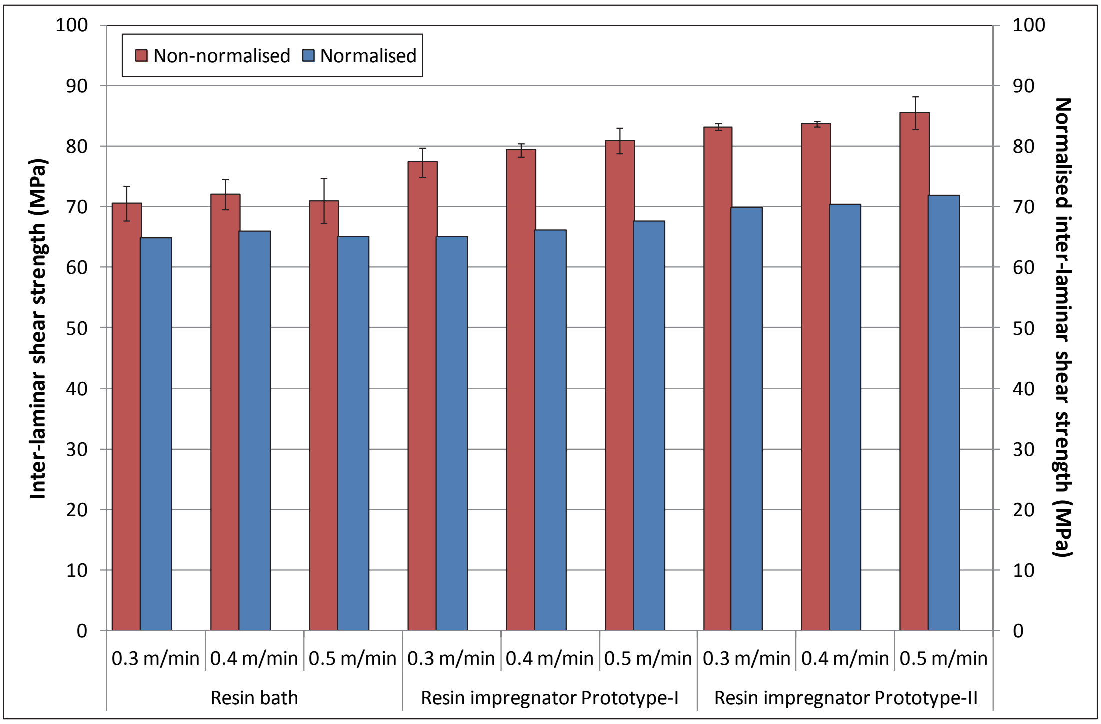


Figure 29 Inter-laminar shear strengths of the pultruded samples obtained from resin bath, I and II impregnators at three pultrusion speeds.

Fewer voids are produced with impregnators-I and II therefore the ILSS is improved compared to the composites produced from the resin bath. The higher ILSS can also be due to improved impregnation, as the fibres are spread the binder on the fibres breaks improving the impregnation of the glass fibre bundle.

4.6.5. Flexural properties

The flexural properties were determined by four-point bending in accordance with ASTM standard: D6272-02. Dimensions of the specimens were 50mm (L) × 12.7mm (W) × 2.3mm (H).

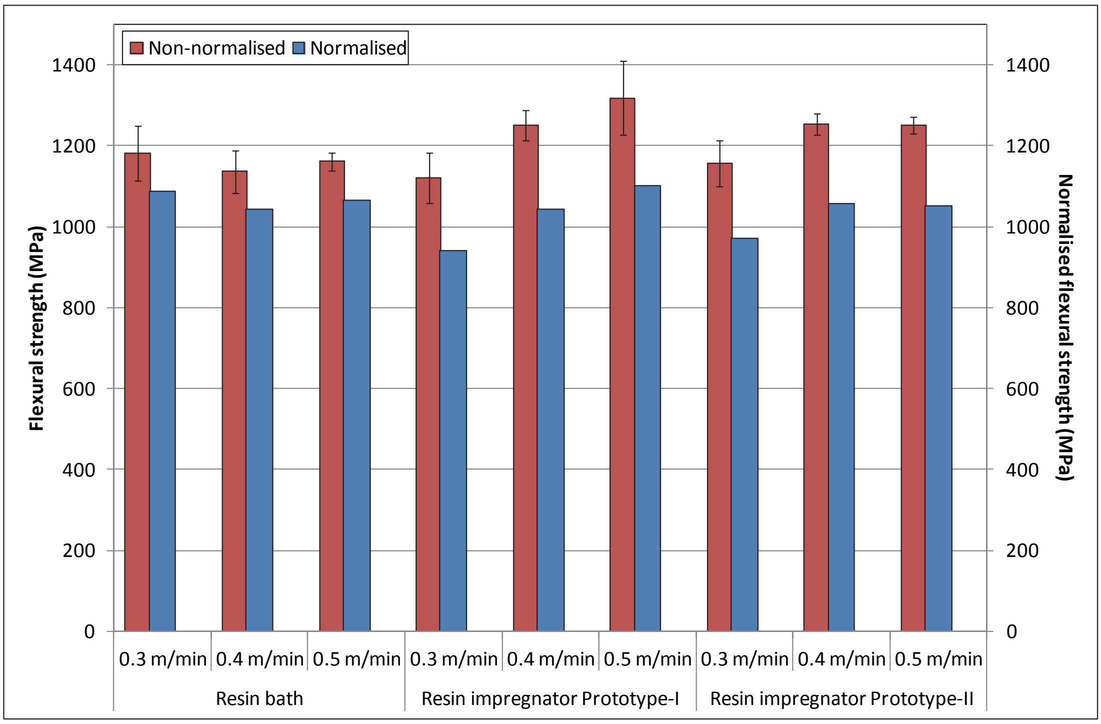


Figure 30 Flexural strengths of the pultruded samples obtained from resin bath, I and II impregnators at three pultrusion speeds.

It can be seen that the non-normalised flexural strength values for the composites produced using "Clean" pultrusion are higher than conventional composites. There is not much difference in the normalised values. It can also be seen that the properties are retained even at faster pultrusion speeds.

A comparison of the flexural moduli for "Clean" and conventional composites is shown in Figure 30. It can be seen that both non-normalised and normalised values for the "Clean" composites are higher than the conventionally produced composites. These values are on the higher side of those previously reported in literature (Bogner et al, 2000).

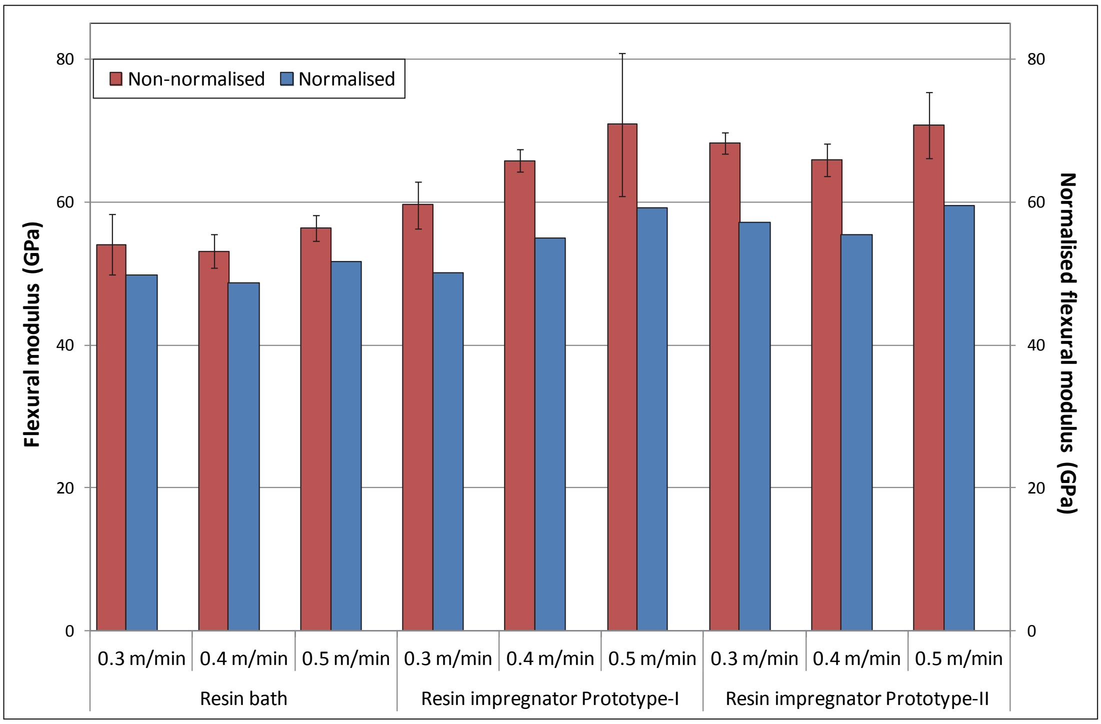


Figure 31 Flexural moduli of the pultruded samples obtained from resin bath, I and II impregnators at three pultrusion speeds.

The samples were also cured to study the effect of curing on the flexural properties. The realists for flexural strengths and moduli for the post-cured samples are shown in Figures 32 and 33. Most of the results of both standard and post-cured samples are equal. Post-curing the samples usually complete any incomplete cross-linking. The flexural strength results show that the composite samples are not increasing; this must show that the reinforced composites have completed cured. Post-curing higher than 60°C does not give any remarkable rise in flexural strength (Aruniit et al 2012).

The samples that have been post-cured show a higher flexural modulus for the resin bath, impregnators Prototype-I and Prototype-II at all three speeds. This increase in flexural modulus shows that the stiffness of the composite increases.

The flexural properties could have been influenced by a number of factors, this could be the fibre alignment, fibre volume fraction, void content or the twisting of the fibres through the impregnation unit, however spreading E-glass fibres improves the degree of impregnation because it breaks down the binder. The trajectory of the fibres was seen to be more controlled when using the 'clean' pultrusion technique.

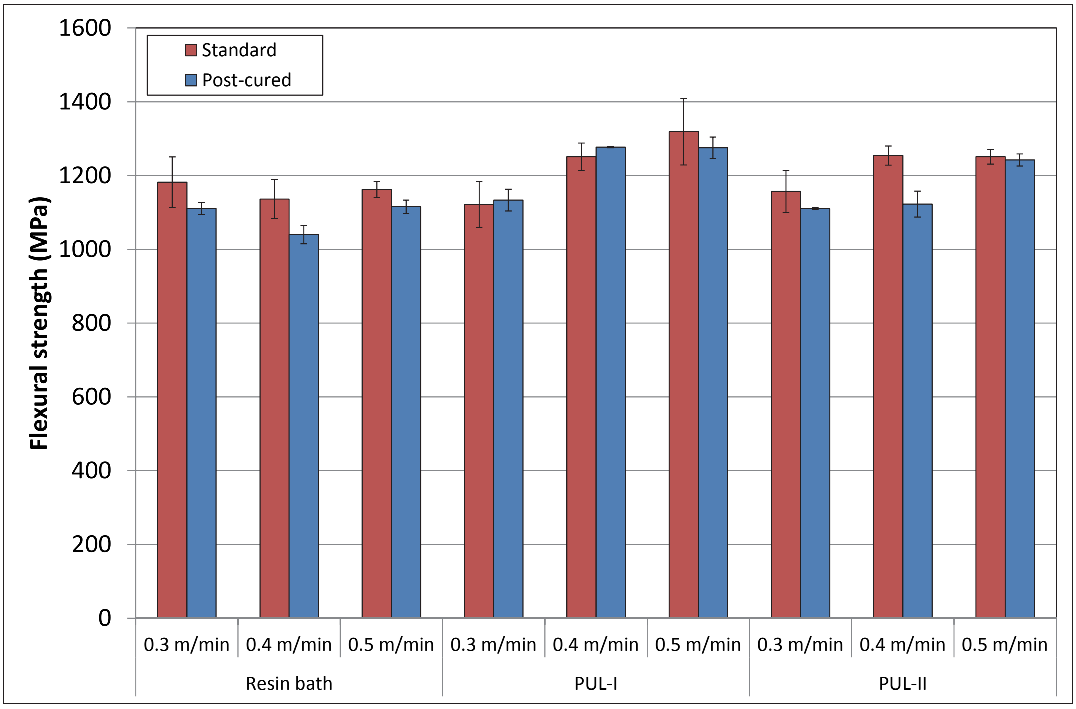


Figure 32 Flexural strength of the standard and post-cured pultruded samples obtained from resin bath, I and II impregnators at three pultrusion speeds.

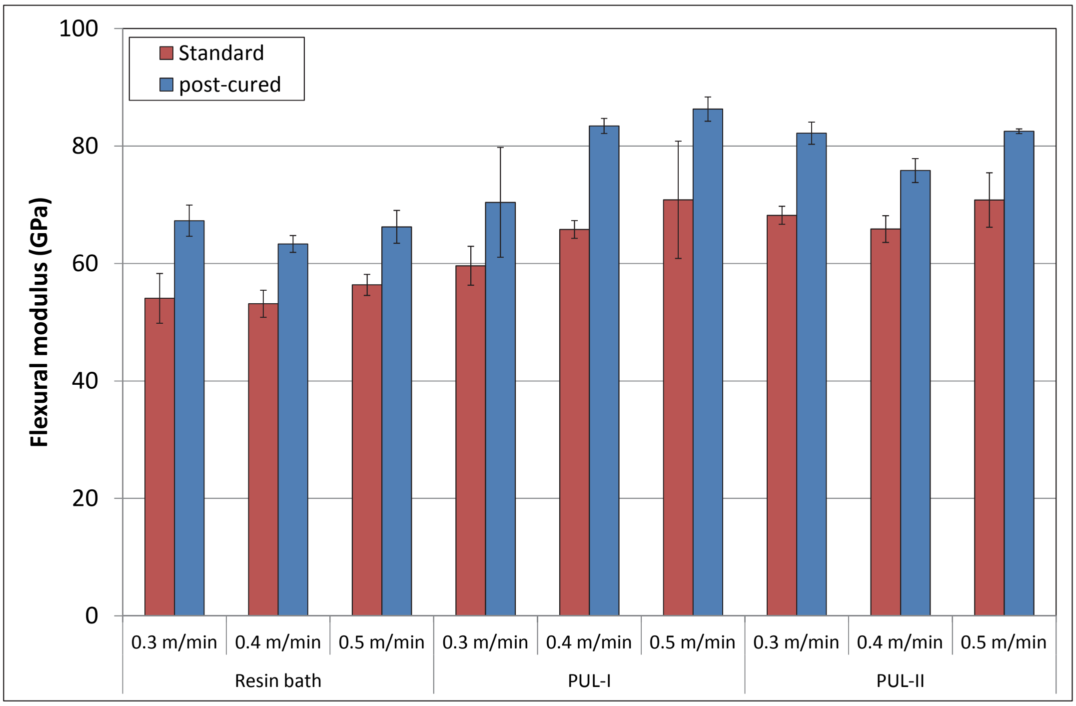


Figure 33 Flexural moduli of the standard and post-cured pultruded samples obtained from resin bath, I and II impregnators at three pultrusion speeds.

4.6.6. Tensile properties

The tensile strengths and moduli for the conventional and clean composite are shown in Figures 34 and 35. It can be seen that although the non-normalised tensile strength are higher for "clean" pultrusion but normalised value lie within the same range. There is not much difference in the tensile moduli values for the values of tensile moduli for the "Clean" and conventional composites.

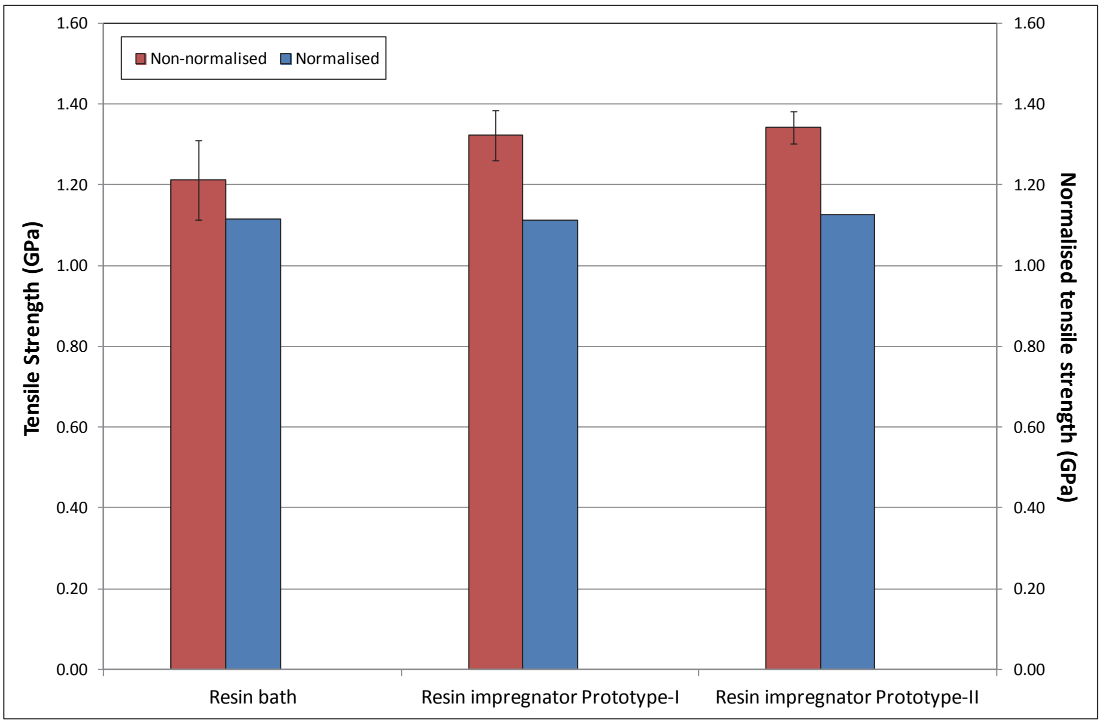


Figure 34 Tensile strength of the pultruded samples obtained from resin bath, I and II impregnators at three pultrusion speeds.

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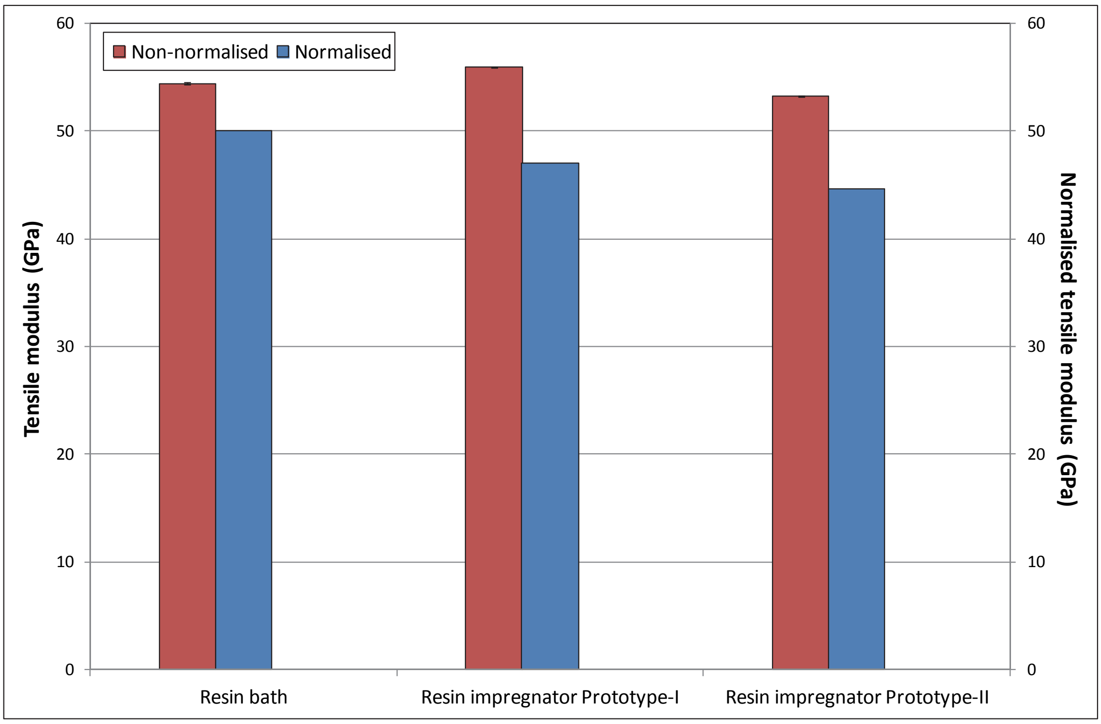

Figure 35 Young's modulus of the pultruded samples obtained from resin bath, I and II impregnators at three pultrusion speeds.

5. CONCULUSIONS AND RECOMMENDATIONS FOR FUTURE WORK

5.1. CONCLUSIONS

Conventional resin bath-based pultrusion has been used for a number of years. In this day and age the focus is on producing profiles in an eco-friendly and in an energy efficient manner. The primary aim of this study was to analyse and develop an environmentally-friendly pultrusion technique for the production of fibre-reinforced composites. Conventional pultrusion has its problems such as, waste resin being produced. Large volumes of cleaning solvents are used to clean the system, which takes up time and increases overall costs. The emissions of VOCs such as styrene in the polyester resin system are a serious problem with the conventional system and also the resin systems with a limited pot life cannot be used with the conventional resin bath system. However there is an alternative, which is the use of 'Clean' pultrusion. Resin impregnators Prototypes-I and II show that polymer reinforced composites can be produced and can provide better results by replacing the conventional resin bath. The emission of VOCs is minimised and the impregnation of fibres is enhanced.

Polymer reinforced composites were produced using both conventional and "Clean" pultrusion. It was shown that the mechanical properties of the composites produced from "Clean" pultrusion were equivalent or marginally better than those produced via conventional resin bath, for example the void fraction was reduced, and there were increases in both flexural and ILSS properties. The increase in properties could be due to the better control of fibre trajectory and alignment, but can also be due to the fewer amount of voids produced from Prototype-I and II impregnators because of the improved impregnation then the binder is broken down due to the spreading of the fibres compared to the composites produced from the resin bath. Increasing the pultrusion speed does not affect the properties much and results are positioned in similar ranges and do not show any significant differences between 0.3 and 0.5m/min. Micrographs of the pultruded composites show that at the lower speeds there are not as many voids present due to the fibres moving slower improving the fibre wet-out of the composites therefore enhancing impregnation. Poor impregnation can cause voids and a poor interface between the fibres and resin, which will have a detrimental effect of the mechanical properties.

Fibre spreading is an important part prior to the production of polymer-reinforced composites. It decreases the thickness of the fibre bundle therefore it takes less time for the resin to penetrate through the fibre bundle. Two fibre-spreading rigs were used; automated spreading rig and the serpentine fibre spreading rig. Both systems allowed the fibres to reach 100% spreading however the serpentine rig was used in the pultrusion experiments as it was more compact less complicated and easier to use and could accommodate more rovings than the automated spreading rig.

The impregnation of as-received and spread glass fibres was also studied, it was seen that spreading the fibres shows a better through thickness impregnation however a decrease in the longitudinal direction.

In conclusion it was shown that the “Clean” pultrusion could be used as an alternative to the conventional resin bath pultrusion.

5.2.RECOMMENDATIONS FOR FUTURE WORK

The following list is recommendations for configuring the work started in this study:

1. The “Clean” pultrusion can be used with other types of resin systems including more environmentally friendly resins such as polyurethane, which offers a styrene-free resin solution and has been shown in previous studies to improve the properties of composites over tradition resin types.

2. Research a more improved mechanical technique to spread the glass fibres more consistently. The automated spreading rig can be looked into further to see if there is a way in making it more compact and user friendly.

3. A detailed life cycle analysis (LCA) should be carried out to assess in the true green credentials of the clean pultrusion technique, as it has proven to enhance enhanced reinforced composites compared to the conventional system which showcases a number of problems with emissions amongst one of a few disadvantages.

4. The “Clean” pultrusion technique should be evaluated under industrial environments to see if the process is viable.

5. The feasibility of using resin systems with significantly fast cure kinetics should be investigated, which could reduce the time and energy used on the pultrusion process.

6. REFERENCES

A.B. Strong. Pultrusion. High productivity now, getting even better. 1-12.

A. Carlsson and B.T. Astrom. . (1998). Experimental investigation of pultrusion of glass fibre reinforced polypropylene composites. Composites A. 29 (A), 585-593.

A. Aruniit, J. Kers, A. Krumme, T. Poltimae, K. Tall. (2012). Preliminary Study of the Influence of Post Curing Parameters to the Particle Reinforced Composite's Mechanical and Physical Properties. Materials science. 18 (3), 1392-1320.

J.R. Brown, S. Kharchenko, H.D. Coffee, I. Huang (2010), System for producing pultruded parts, publication number: Ep1960184B1

Chen-Chi M. Ma, Mong-Song Yn, Chin-Hsing Chen and Chin-Lung Chiang. (1990). Processing and properties of pultruded thermoplastic composites (I). Composites Manufacturing. 1 (3), 191-196.

Chin-Hsing Chen & Chen-Chi M. Ma. (1992). Puitruded fibre reinforced polyurethane composites II. Effect of processing parameters on mechanical and thermal properties. Composites Science and Technology. 45, 345-352.

D.Hull and T.W. CLyne (1996). An introduction to composite materials. Cambridge: Cambridge University press.

Department of mechanical and manufacturing. C.P. Bhat

K. Devi, P.S. Kalla, Dhanasekaran, B. Zhang and A. Ramazan. (2011). Sustainability of fibre reinforced composites: status and vision for future. Proceedings of the ASME 2011 International Mechanical Engineering Congress & Exposition, Denver, Colorado, USA.

G.W Ehrenstein (2000). Polymeric materials . Germany: Hanser Gardner.

Dr. J. G. Vaughan, Dr. E. Lackey, H. D. Coffee. N. Barksby, J.L. Lambach. (2003). Pultrusion of Fast-Gel Thermoset Polyurethanes: Processing Considerations and Mechanical Properties. Composites 2003 Convention and Trade Show Composites Fabricators Association Anaheim, California, USA.

D. Troutman. Polyurethane Pultrusion Application Successes with large profiles. Director of Marketing and Product Development Creative Pultrusions, Inc. Alum Bank, PA 15521, United States of America.

R.J Gaymans and E. Wevers (1998). Impregnation of a glass fibre roving with a polypropylene melt in a pin assisted process. Composites A; 29: 663–670.

D. Hall. (1981). An introduction to composite materials. Cambridge: Cambridge university press.

V.S. Hota, G. Rao, R.Ritchey. (1992). Response of structural systems made of pultruded fibre reinforced plastics. 47th annual conference, composites institute. The society of the plastics institute, Inc.

J. B. Donnet and P. Ehrburger. (1977). Carbon fibre in polymer polymer reinforcement. Carbon. 15, 143-152.

J.G Morley (1987). High performance fibre composites. London: academic press limited.

L. Ye, V. Klinkmuller and K. Friedrich. (1992). Impregnation and Consolidation in Composites Made of GF/PP Powder Impregnated Bundles. Journal of Thermoplastic composite materials. 5, 32-48.

N.C.W. Judd and W.W. Wright, (1978). Voids and their effects on the mechanical properties of composites -an Appraisal. SAMPE Journal. (2), 10-14.

M. G. Dube, G. L. Batch, J. H. Vogel, C. W. Macosko. (1995). Reaction Injection Pultrusion of Thermoplastic and Thermoset Composites. POLYMER COMPOSITES. 16 (5), 378-385.

M. S. Irfan, V. R. Machavaram, R. S. Mahendran, N. Shotton-Gale, C. F. Wait, M. A. Paget, M. Hudson and G. F.Fernando. (2011). Lateral spreading of a fiber bundle via mechanical means. Journal of Composite Materials. 46 (3), 311-330.

M.K. Sridhar. (1993). Fibre reinforcements for composites. Defence Science Journal. 43 (4), 365-368.

M. Bannister. (2001). Challenges for composites into the next millennium, a reinforcement perspective. Composites. 32 (a), 901-910.

M. Connolly, J. King, T. Shidaker and A. Duncan. (1). Processing and Characterization of Pultruded Polyurethane Composites. Huntsman International LLC 2190 Executive Hills Boulevard, Auburn Hills, Michigan 48326, USA. 1-16.

M. Connolly, J. King, T. Shidaker and A. Duncan. (2006). characterization of pultruded polyuretahne composites: Environmental exposure and component assembly testing. COMPOSITES Convention and Trade Show American Composites Manufacturers Association St. Louis, MO USA. Huntsman Polyurethanes, Auburn Hills, MI.

P.K. Mallick (2007). Fiber-reinforced Composites, Materials, Manufacturing, and Design. CRC Press

P.Tattersall (editor: N.L. Hancox) (1981). Fibre composite hybrid materials. London: Applied science publishers LTD.

R. R. Joshi, E. H. Cheolas, E. F. Cassidy, W. J. Karoly*, and H. D. Coffee. (2001). Polyurethane based pultruded resins improve the environmental image of composite materials. COMPOSITES Convention and Trade Show Composites Fabricators Association. Tampa, FL USA.

R.A. Cageao, J. Hayes, C. Snyder, Polyurethane resins for pultrusion. . Bayer MaterialScience LLC, Pittsburgh PA 15201.

S.D.R. Wilson. (1997). Lateral spreading of fibre tows. Journal of Engineering Mathematics. 32, 19-26.

S.U.K. Gadam, J.A. Roux, T.A. McCarty, J.G. Vaughan. (2000). The impact of pultrusion processing parameters on resin pressure rise inside a tapered cylindrical die for glass-fibre/epoxy composites. Composites science and technology. 60, 945-958.

S.D. Pandita, M.S. Irfan, V.R. Machavaram, N. Shotton-Gale, R.S. Mahendran, C.F. Wait, M.A. Paget, D. Harris, C. Leek and G.F. Fernando. (2012). Clean wet-filament winding – Part 1: design concept and simulations. Journal of Composite Materials, 1-12.

Seminar report on pultrusion process. Manipal Institute of Technology SP systems. (2001). Resin Systems for Use in Fibre-Reinforced Composite Materials. Available: http://www.azom.com/article.aspx?ArticleID=986. Last accessed 20th august 2012.

T.F. Star. (2000). Pultrusion for engineers. Cambridge: woodhead publishing.

S.H. Ahn, W.I. Lee, G.S. Springer . (2005). Measure of the three-dimensional permeability of fiber preforms using embeded fibre optic sensors. journal of composite materials. 29 (6), 714-733.

J.L. Thomason (1995). The interface region in glass fibre reinforced epoxy resin composites: 2. Water absorption, voids and the interface. composites. 26 (7), 477-485.

T.H. Thorsted (1987), Epoxy resin system and pultrusion processemploying same. Patent number: 4,842,667

M.R. Wisnom, T. Reynolds, G.N. William. (1996). Reduction in interlaminar shear strength by discrete and distributed voids. composites science and technology. 56 (1), 93-101.

D. Zhongman, L. Shoujie, Y. Huan, and L.J. LEE. (2000). Numerical and Experimental Analysis of Resin Flow and Cure in Resin Injection Pultrusion (RIP). Polymer composites. 21 (5), 762-778.