Procedure for making flat thermoplastic composite plates by Automated Fiber Placement and their mechanical properties

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A Thesis
In
The Department
Of
Mechanical and Industrial Engineering

Presented in Partial Fulfillment of the Requirements
For the Degree of
Master of Applied Science (Mechanical Engineering) at
Concordia University
Montreal, Quebec, Canada

April 2015

CONCORDIA UNIVERSITY

School of Graduate Studies

This is to certify that the thesis prepared

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Entitled: **Procedure for making flat thermoplastic composite plates by Automated Fiber Placement and their mechanical properties**

and submitted in partial fulfillment of the requirements for the degree of

Master of Applied Science (Mechanical Engineering)

Complies with the regulations of the university and meets the accepted standards with respect to originality and quality.

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Abstract

Procedure for making flat thermoplastic composite plates by Automated Fiber Placement and their mechanical properties

Automated Fiber Placement (AFP) is one of the new manufacturing processes for composites that has many advantages such as fast rate of material deposition, repeatability, and reduction of material waste. Thermoplastic composites compared with thermoset composites show a lot of advantages such as higher impact resistance, recyclability etc. and they are very suitable for primary structures of an aircraft. However, manufacturing of thermoplastic composites requires more efforts because of the very high viscosity of thermoplastic resin.

This study proposed a manufacturing procedure to make flat thermoplastic laminates with different layup sequences by using a heated mandrel during layup process with AFP. It is shown that using a mandrel heated to a temperature higher than glass transition can make flat laminates. Processing parameters which consist of hot gas temperature, compaction force, layup speed, mandrel temperature, were found for different laminates. Moreover, repass after material deposition helps to reduce void content inside the laminates.

The obtained flat laminates show the crystallized thermoplastic resin. The high degree of crystallinity is recommended to have good mechanical properties. Tensile, compression and in-plane shear tests were carried out to determine mechanical properties of the laminates made by AFP. Results are compared with those made by autoclave. Test results show that laminates made by AFP have higher tensile modulus, higher compression modulus, similar tensile strength, and almost equal of shear modulus. However, a reduction in compression strength is observed.

Keywords: thermoplastic composites, PEEK, APC-2/AS4, automated fiber placement, manufacturing procedure, characterization, process parameters.

Acknowledgement

First of all, I would like to express my gratitude to my supervisor, Dr. Suong Van Hoa, for his unconditional support and comprehensiveness throughout this project.

I am grateful to the late Dr. Ming Xie, Dr. Daniel Rosca, Mr. Heng Wang, Mr. Dainius Juras for their considerable help regarding my experimental work. I would like to thank Jeffrey Fortin Simpson for his time spent operating the AFP machine and giving me many valuable advices.

Thanks to all my colleagues at Concordia Center for Composites for their great support and friendship.

I would like to thank the examining committee for spending time reading this thesis and providing critical comments.

Last but not least, I would like to express my appreciation to my family for their endless sacrifice during my study.

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CHAPTER 1 Introduction

Nowadays, composite materials are widely used in various applications because of their light weight and good mechanical properties. Especially for aerospace industry, composite is a material which manufacturers focus to develop in order to reduce airplane weight and fuel consumption. Several decades before, composite materials were used only to make secondary structures and interior parts. However, development of new materials and manufacturing techniques advanced the application of composites on primary structures such as fuselage, airframe. For instance, the Boeing's 787 Dreamliner contains 50% weight of composite and Airbus new airplane A350 XWB has the frame containing 52% composite [1].

This chapter gives the basic definition of composite materials as well as manufacturing techniques of thermoplastic composite products.

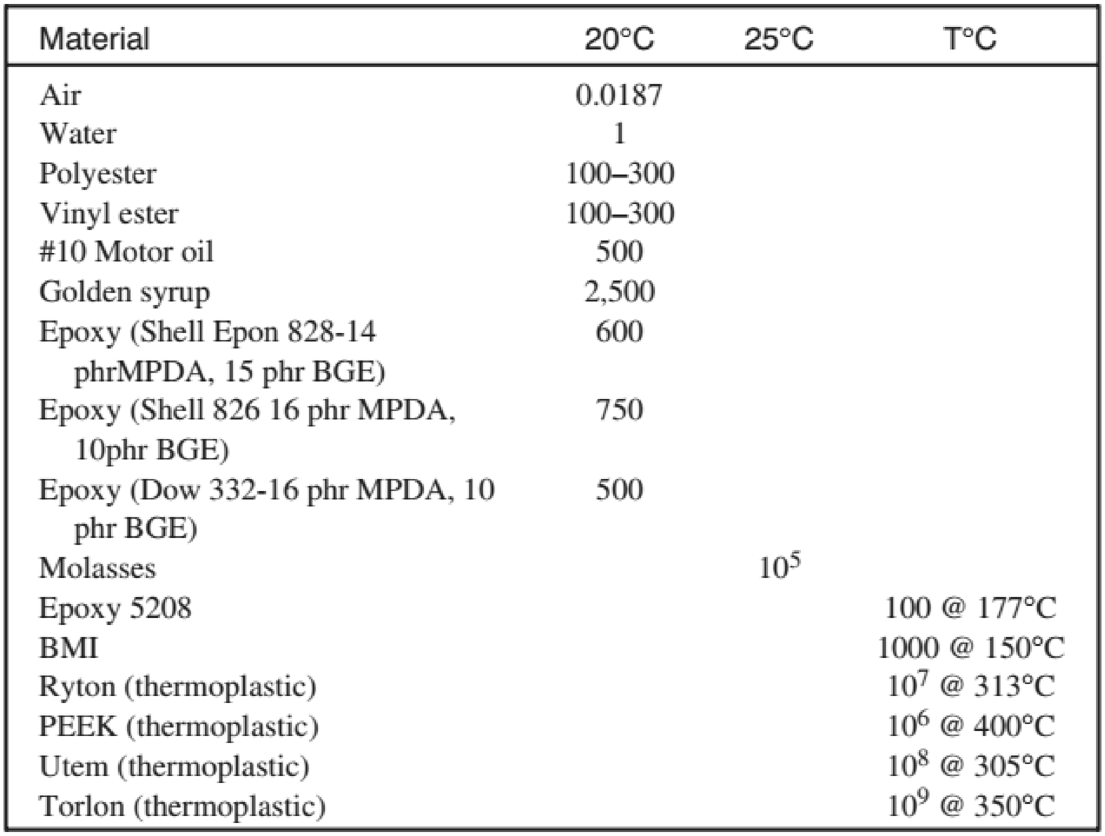
1.1 Composite material definition

Composite is a material which is formed by combining two or more different materials, which is usually called matrix and reinforcement. The matrix can be thermoset resin, thermoplastic resin or metal. Its function is to hold the reinforcement together. Reinforcement which normally gives strength to composite can be continuous fibers, short fibers, fabrics or metals. The combination creates a unique material which has orthotropic properties in most cases. This means composites can be very stiff in one direction but less stiff in others.

Classical Lamination Theory (CLT) is usually used to calculate properties of composite laminate based on properties of one layer called lamina (Ex, Ey, Gxy…). Lamina is a thin layer of composite; in case of continuous fiber, all the fibers have the same orientation.

Based on resins, one can divide composites into 2 types: thermoset (thermosetting) composites and thermoplastic composites. Thermoset composites use thermoset resins such as polyester resins, vinyl ester resins and epoxy. Thermoset resins are common because they are in liquid state at room temperature so that it is convenient for impregnate reinforcing fibers to make composite. The most advantage of thermoset composite is it is simple for the manufacturing because of its low viscosity so that it is easy to impregnate resin into fibers to form composite. However, thermoset resins show disadvantages. First of all, formulated thermoset resins have a limited shelf life and pot life. The shelf life varies from 1 month to over 2 years and depends on ambient temperature. The pot life is the available time that the mixture of resin and hardener can be used for manufacturing before they become hard or too viscous. To avoid thermoset resin cure before impregnation, the process has to be well prepared. If prepregs are used, they are usually stored in a refrigerator, and are taken out for some time before layup. Process to cure thermoset composites usually has 2 steps. First step is to heat up thermoset resin to certain temperature at which viscosity is low enough to impregnate fibers. Second step is to cure all resins in the laminate. Vacuum and/or pressure are normally used to remove voids inside laminate. After curing, the composite will have the fixed shape and cannot be re-melted and so that it is hard to be recycled. Although there are techniques to recycle thermoset composites known as mechanical recycling and thermal processes [2], this issue is still under-development.

Thermoplastic composites use thermoplastic resins such as polypropylene, polycarbonate, poly vinyl chloride, PEEK, PEKK etc. Different from thermoset resin, thermoplastic resins do not need hardener to become solid. In fact, thermoplastic resins melt at a certain temperature and return to a solid state due to cooling. Compared with thermoset composites, thermoplastic composites show advantages. For instance, thermoplastic composites usually have higher impact strength, and higher strain for failure. Moreover, thermoplastic resin has no shelf life, no pot life and high recyclability because it just needs to be re-melted to re-manufacture. However, thermoplastic composites have difficulties in manufacturing because of very high viscosity of thermoplastic resin (Table 1.1) and are mostly manufactured at higher temperature than thermoset composites.



Material	20°C	25°C	T°C
Air	0.0187		
Water	1		
Polyester	100–300		
Vinyl ester	100–300		
#10 Motor oil	500		
Golden syrup	2,500		
Epoxy (Shell Epon 828-14 phrMPDA, 15 phr BGE)	600		
Epoxy (Shell 826 16 phr MPDA, 10phr BGE)	750		
Epoxy (Dow 332-16 phr MPDA, 10 phr BGE)	500		
Molasses		10^5	
Epoxy 5208			100 @ 177°C
BMI			1000 @ 150°C
Ryton (thermoplastic)			10^7 @ 313°C
PEEK (thermoplastic)			10^6 @ 400°C
Utem (thermoplastic)			10^8 @ 305°C
Torlon (thermoplastic)			10^9 @ 350°C

Table 1.1 Viscosity (in centipoise) of a few Thermoset and Thermoplastic Materials [3]

1.2 Manufacturing techniques for thermoplastic composites

Due to the fact that properties of thermoplastic and thermoset resins are different, manufacturing parameters for them are different. Thermoset resin has low viscosity which makes it easier to impregnate into fiber to form composite. Thermoplastic resin has very high viscosity so it needs a different strategy to make thermoplastic laminate. This strategy usually consists of two steps. First step is creating of preliminary combination of matrix and fiber which can be prepreg tape, reinforcing fibers with filaments made from thermoplastic matrix, matrix or fabric film sandwich [3]. Second step uses these preliminary combinations to form final product. In this section, several techniques which can be used to manufacture thermoplastic composite are presented including Autoclave Consolidation, Compression Molding, Pultrusion, Filament Winding, Fiber Tape Placement, and Fiber Tow Placement.

Autoclave consolidation

Autoclave consolidation for thermoplastic composite is similar to Hand Layup (Dry Layup) technique for thermoset. This is the simplest manufacturing technique and is usually used for parts with small dimensions. In most cases, prepreg is used for thermoplastic autoclave consolidation technique. Figure 1.1 shows the layup procedure for autoclave consolidation process. First of all, a caul plate (optional) and a high temperature released film is placed on the mold. Prepreg tapes are then cut into desired shape and placed on to the mold in designed orientation. Spot welding can be used to attach 2 adjacent layers together. Afterward, another treated released film is put on top of the final layer. Pressure plate and several style glass layers are then placed consequently. Finally mold with laminate is vacuum bagged and the assembly is put inside autoclave. Autoclave applies heat and pressure in specific cure cycle (Figure 1.2) to melt thermoplastic resin, bond layers together and consolidate the laminate.

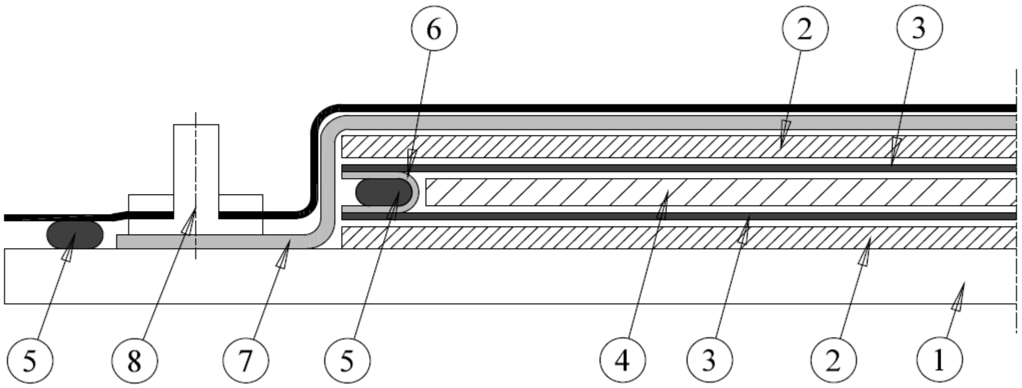


Figure 1.1 Typical layup for Autoclave Consolidation

(1) Tool plate; (2) Caul plate; (3) Bagging film; (4) Laminate; (5) High temperature sealant tape; (6) plain woven fiber glass cloth; (7) breather; (8) Vacuum valve

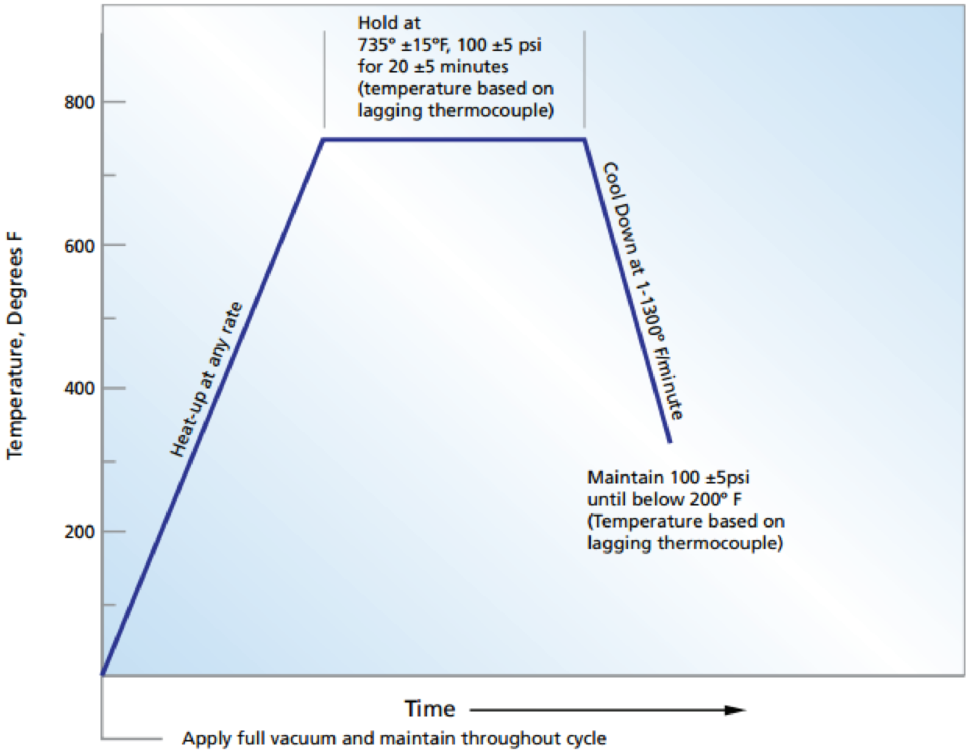


Figure 1.2 Typical cure cycle for thermoplastic composite by Autoclave Consolidation [4]

Compression molding

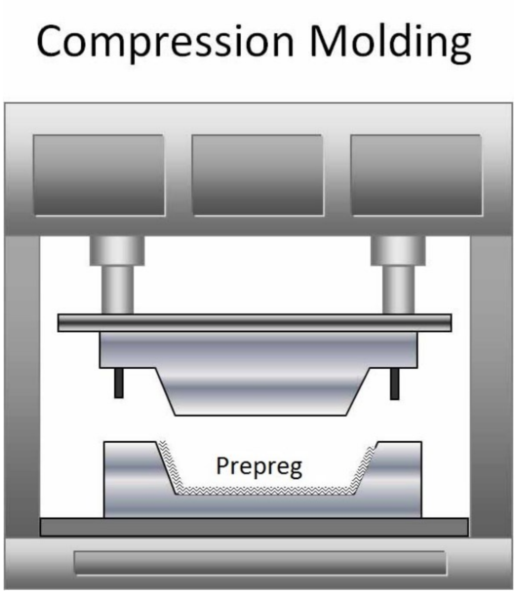


Figure 1.3 Compression molding for composites [5]

Compression molding is an efficient process to manufacture both thermoset and thermoplastic composites and is widely used in automotive industry. Raw material for thermoplastic compression molding is thermoplastic composite prepregs. Figure 1.3 illustrates procedure of compression molding technique for thermoplastic composite. Prepreg thermoplastic tapes are firstly placed in the lower mold based on the designed layup sequence. Two parts of the mold and also the raw material can be pre-heated. Afterward, the upper mold moves down, apply heat and pressure to the material to form the final product.

Filament Winding

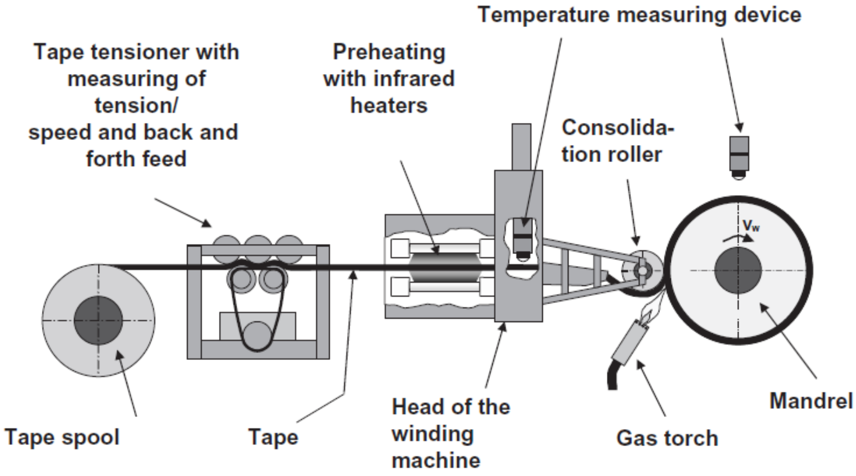


Figure 1.4 Filament winding for thermoplastic composite [6]

Filament winding is usually used to manufacture composite pressure pipes and vessels of both thermoset and thermoplastic composites. It is one of the oldest techniques which is automated. Procedure for this technique is as shown in Figure 1.4. Thermoplastic tape from tape spool is fed to preheating unit then is passed to roller. Hot gas torch then heats the tape and substrate to melt resin. Heated thermoplastic tape is wounded onto a rotating mandrel which has cylindrical or convex shape. Force applied by roller bonds tape to substrate to form laminate.

Pultrusion

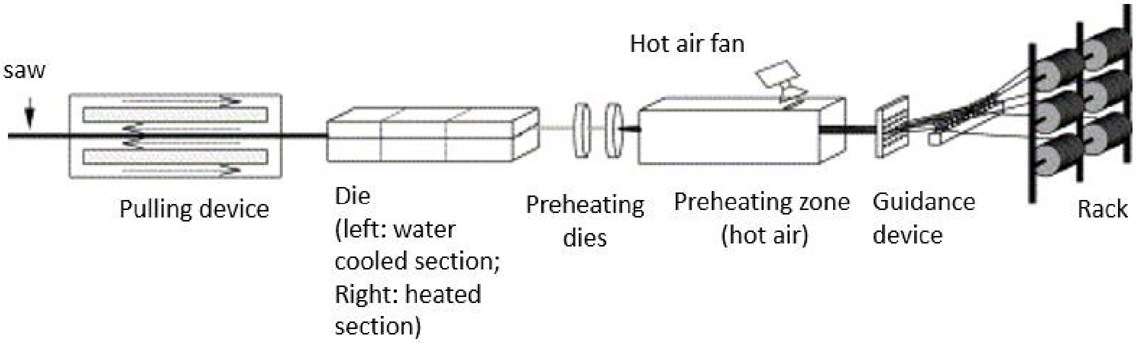


Figure 1.5 Pultrusion for thermoplastic composite[7]

Pultrusion is already widely used for thermoset resin composite because of its high productivity and also its continuous manufacturing. Since the viscosity of thermoplastic resin is much higher than that of thermoset resin, pultrusion process also needs some modification. Figure 1.5 demonstrates pultrusion procedure for thermoplastic composite. Fibers with comingled thermoplastic resin from bundles go through guidance devices toward a preheating zone. Preheating zone is added to heat fibers in order to enhance impregnation of resin into fiber bundles. Fibers are then passed through a die with heated section on right side. At this point, thermoplastic resin melts and wets the fibers to form composite products. The cooled section on left side of the die helps final products stay in form because at high temperature, thermoplastic composite can be deformed.

Automated Tape Laying (ATL)

Automated Tape Laying (Figure 1.6) is a fast manufacturing method in which prepreg tapes are laid down to mandrel to make part. In both cases for thermoplastic prepreg or thermoset prepreg, after laying down process finishes, part is put into autoclave for curing (thermoset) or consolidating (thermoplastic). Head is very flexible in controlling lay down angle, starting and ending point of individual tape. Preheat system is usually required in case to thermoset tape but is critical in case of thermoplastic material.

Figure 1.6 Automated Tape Laying (Photo Courtesy from Mtorres website) [8]

Automated Fiber Placement (AFP)

AFP is a manufacturing technique that can be used for both thermoset and thermoplastic composites. The principle of Automated Fiber Placement is quite similar to that of ATL. The difference is instead of laying the tape, AFP lay down one or multiple tows. This seems to reduce productivity but AFP is better in making more complex parts. More details of AFP process will be described in chapter 2.

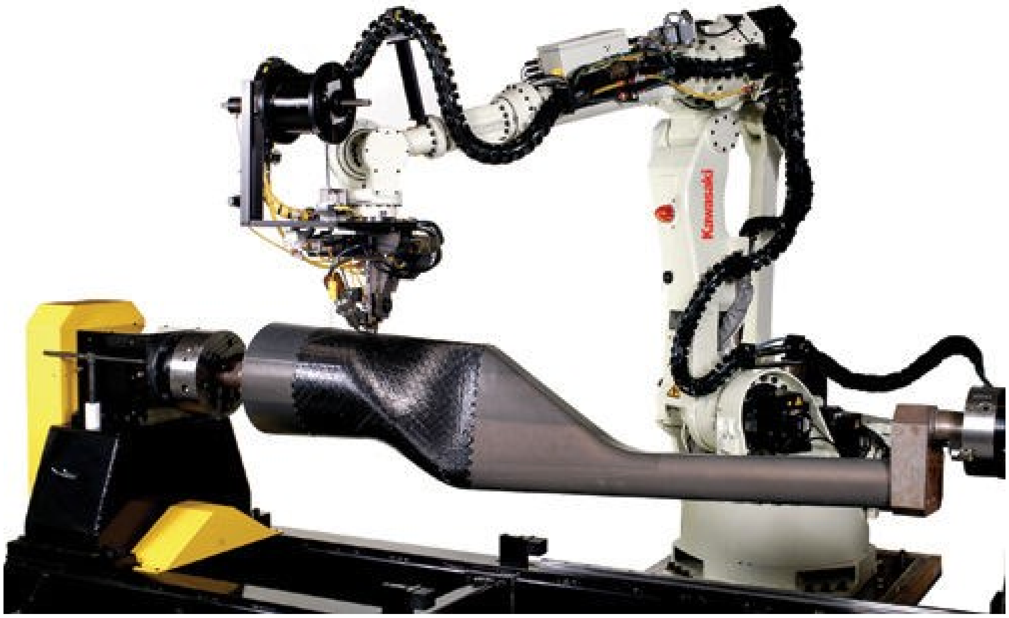


Figure 1.7. Automated Fiber Placement (Photo Courtesy from Atomated Dynamics website)[9]

CHAPTER 2 Literature review and research objectives

Summary: This chapter presents the literature review and motivation for this thesis. A survey of research works done by other researchers and the existing problems of AFP with in situ consolidation process to make flat thermoplastic laminates. Finally, research objectives are presented at the end of this chapter.

2.1 Literature review

2.1.1 Introduction of Automated Fiber Placement with in situ consolidation

As a brief introduction from previous chapter, AFP can help in automating composites manufacturing. It is also a solution for traditional technique such as Hand layup which is usually slow and is based on the skill of the technician. Hand layup technique increases the labor cost and variability of the product. Automated Tape Placement and Automated Fiber Placement helps to automate the process, reduces labor cost and material waste.

The heart of Automated Fiber Placement machine is the tape deposition head which consists of a tow feeding unit, one or multiple rollers, and a tow heating system. Figure 2.1 shows the thermoplastic tape deposition head from Automated Dynamics which has one roller. It uses nitrogen gas to heat the tow and its compaction force is generated by roller. The head from Accudyne (Figure 2.2) is more complex which has the capability of placing one 75 mm prepreg tape or twelve 6.35 mm tows. Moreover, it has also four active compactors to provide normal compaction force rather than compaction force from only rollers.

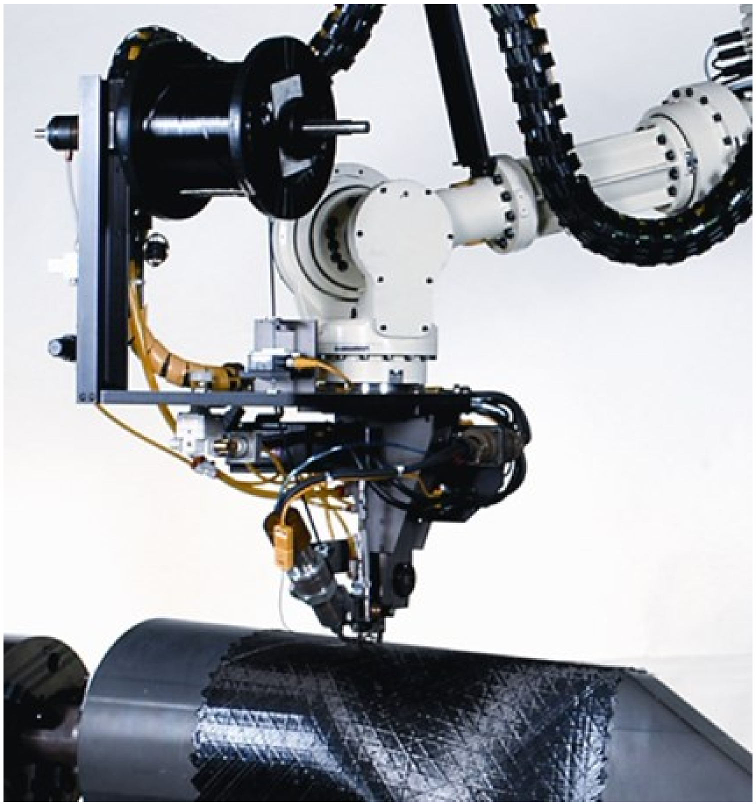


Figure 2.1 Automated dynamics thermoplastic tow deposition head (photo courtesy from Automated Dynamics website)[9]

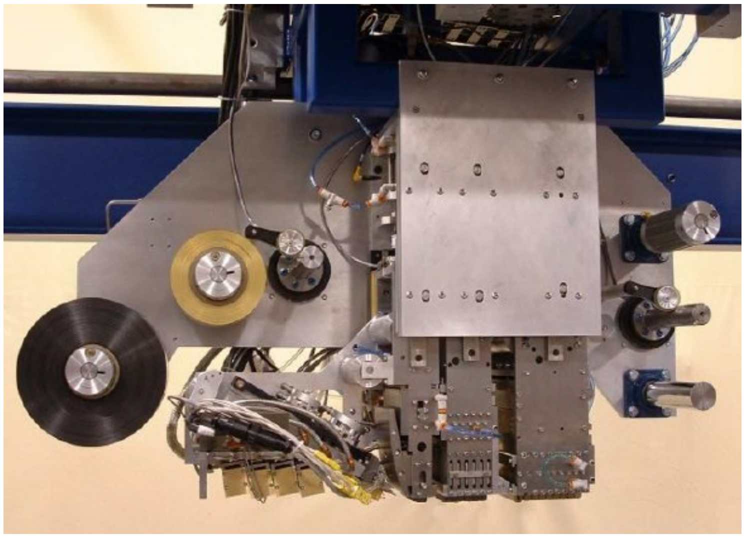


Figure 2.2 Accudyne thermoplastic in-situ consolidation tape deposition head [10]

In spite of the fact that there are various types of AFP machines, the main manufacturing principle is quite similar. Figure 2.3 illustrates the in-situ consolidation tow placement process. Prepreg tow is fed to roller and is heated by a hot gas torch or by a laser system up to melting temperature. For hot gas torch, the inert gas is usually nitrogen to avoid any explosion. Another role of this inert gas is to protect tows from oxidative degradation during layup process [11] [12]. Roller with or without its own heating system then lays down the tow to the substrate and applies compaction force to bond it to the substrate. Consolidation of the tows happens under transient high temperature condition. The tow is then cut to finish one tow. Afterward, the robot brings the head to a new position and starts laying the next tow. This process repeats until the designed part is built.

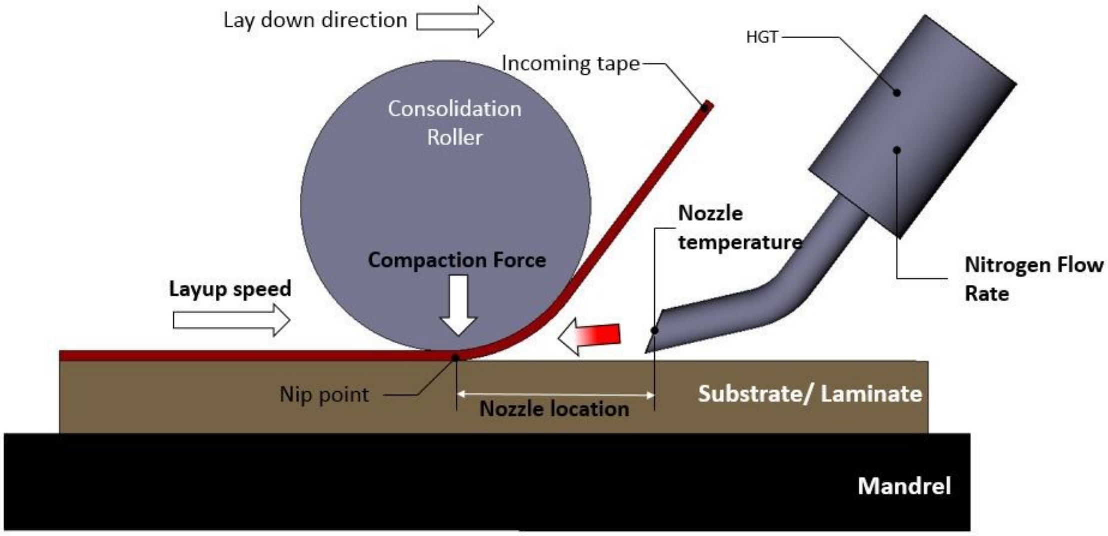


Figure 2.3 Schematic drawing of fiber placement Process

2.1.2 Parameters affecting the manufacturing process.

During manufacturing process, there are a few factors affect the quality of the final product. These are material properties, hot gas temperature, layup speed, compaction force of the roller, mandrel’s temperature etc. The effects of individual variable or combination of some of them have been presented in many papers [10, 13-27].

Materials

First of all is the effect of thermoplastic material properties. Different thermoplastic resins have different properties such as glass transition temperature (T_g) and melting temperature (T_m). Therefore, depending on material used for manufacturing, process window will be changed. Gruber et al. [10] concluded that material, process and head parameters have a big influence on intraply and interply void content inside laminate made by AFP with in situ consolidation. Furthermore, this paper showed that by using commercial grade thermoplastic tapes, there is not enough time at manufacturing temperatures to fully consolidate and bond tapes to substrate during in situ consolidation process. Also, some tapes lacking surface resin may not generate full layer-to-layer weld strength. Finally, by improving tape quality, laminate properties are improved. For example, Short Beam Shear Strength (SBSS) increases from 76% to 105% of the SBSS of an autoclave laminate by changing from commercial APC-2 AS4 for still-developing APC-2 AS4.

APC2-AS4 prepeg which is also used in this study contains PEEK resin which is considered as semi-crystalline material [13] and the degree of crystallinity affects the properties of the final laminate [14, 15]. These papers show that the higher crystallinity laminate provides higher tensile, compression and shear modulus and strength. However, the mode I fracture toughness decreases when the degree of crystallinity increases [15]. The degree of crystallinity of thermoplastic composite depends mainly on the cooling rate [14, 16]; it decreases when the cooling rate increases. Moreover, non-uniform crystallinity in a part influences residual stress in the laminate[17].

Processing parameters

Cai [18], proposed a window of processing parameters for the manufacturing of thermoplastic composite cones using AFP. Nitrogen flow rate was kept constant at 70 Standard Liter Per Minute (SLPM), nozzle temperature varied from 900^0C to 950^0C, process rate changed between 1 in/s and 3 in/s, compaction force is from 30 kgf to 50 kgf, and nozzle location to the roller is in between 0.45 in and 0.85 in. Moreover, using Taguchi’s method, an optimal set of parameters including nozzle temperature, process rate, compaction force and nozzle location to the roller was introduced. However, manufacturing a cone which has no free edge is different with manufacturing a plate which has four free edges. Cones can keep their shape because of the constraints inside the structure while laminates with free edges can be deformed after manufacturing.

During manufacturing process, laminate is subjected to several heating and cooling cycles which will result in residual stresses inside final laminates. These residual stresses can cause matrix cracking which can reduce the stiffness up to 35% [19] [20]. Heating and cooling are affected by many parameters such as layup speed, temperature of hot gas torch, layup sequences, ambient temperature etc. Tierney and Gillespie [21] studied the crystallization behavior of PEEK composites in different heating and cooling rate conditions. Hot gas temperature was kept at 850^0C, layup speed and the distance from the torch to prepreg were varied. They found that the crystallinity along cross section of a single ply is uneven and it is affected by both speed and the distance between torch and prepreg. The low layup speed provided higher crystallinity but higher speed resulted in more even crystallinity along cross section.

Another parameter affecting residual stresses of composite laminate is tape tension. Lu et al. [6] found that the increment of tape tension has a big effect on the final residual stresses when doing filament winding. If the tape tensions are constant, radial residual stresses are tensile. In case of increasing tape tension, radial stresses are compressive. In case of decreasing tape tension, absolute value of radial and circumferential residual stresses are higher than those with constant tape tensions.

Brzeski and Schledjewski [22] showed the effect of tool temperature on laminate properties when manufacturing composite laminates by AFP with in situ consolidation. By increasing tool temperature, interlaminar shear strength increases. However, if temperature of the tool is too high, temperature of the laminate after consolidation is also too high so that deconsolidation will happen and interply adhesion is decreased. Another conclusion from this paper is the heat input ratio between incoming tape and substrate influence residual stresses of the laminate.

De-consolidation and re-consolidation of thermoplastic laminate was also presented in the research of Ye et al. [23]. Material used in this paper is CF/PPS thermoplastic laminates. The paper concluded that if the applied pressure is less than a critical value de-consolidation will occur and if it is higher than critical value, re consolidation will happen. It also showed that de-compaction behavior of the fiber network and applied external pressure determine final de-consolidation state while resin melt viscosity which depends on processing temperature affects the time for de-consolidation to reach a stable state.

As heat transfer is the main problem which involves most variables during the manufacturing process, heat transfer models for tape placement have been introduced in a numbers of papers [24-27]. These studies used 2D model to simulate heat transfer from nitrogen gas torch through the thickness of the composite. Grove [25] showed that for all laminates which have more than 2 plies, the highest cooling rates are at the top surface. Moreover, this cooling rate is independent of laminates thickness. Sonnez et al. [27] also combined heat transfer with its effect on the crystallization. They also suggested that preheating substrate is necessary for better bonding between incoming tape and substrate but preheating the tape or roller does not improve bonding condition.

2.2 Previous works done and research objectives.

2.2.1 Previous works and problems definition

Even though AFP has a lot of advantages in manufacturing thermoplastic composites, if the processing parameters are not correct, quality of laminates made by AFP with in-situ consolidation still cannot reach the level of quality in laminates made by autoclave consolidation [28-30]. The reduction of mechanical properties can be explained firstly by the poor bonding between layers deposited by AFP. Figure 2.4 shows the delamination at outer layer of the thick composite tube made at Concordia Center for Composites (CONCOM) using Cetex TC1200 PEEK AS-4 from TenCate Advanced Composites Company. Laminate configuration is $[90/(25/-25)_{45}]$, inner diameter is 1.5 inches (18.1 mm), and outer diameter is 2.405 inches (61.1 mm). Processing parameters are: HGT=875^{0}C, Nitrogen flow rate = 75 SLPM, layup speed = 2.75 inches/s (70 mm/s), and Compaction force = 75 lbf (333.6N). Another reason to explain the reduction in mechanical properties is void fraction of laminates made by AFP is in most cases higher than those made by autoclave consolidation [18]. Figure 2.5 shows a microscopic picture of a laminate made by AFP with defects as voids, resin rich areas and delamination.

Figure 2.4 Poor bonding between layers of composite tube made by AFP

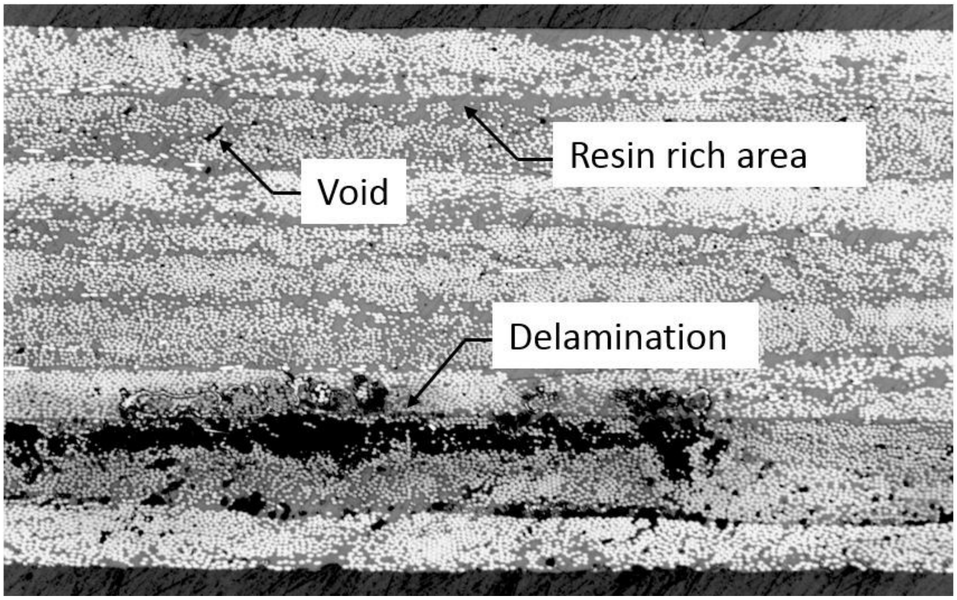


Figure 2.5 Microscopic picture for a laminate made by AFP

Another problem found during manufacturing thermoplastic plates by AFP is the deformations of the final products. Cai [18] found that the laminates manufactured by AFP were warped and separated from the mold during lay-down process (Figure 2.6). This laminate was manufactured on a preheated tool at 120^0C. However, manufacturing parameters were not specified.

Figure 2.6 Illustration of warped laminate made by AFP[18]

Accudyne succeeded in making flat laminates and providing a few mechanical properties [28]. In their thermoplastic in-situ consolidation tape deposition head (Figure 2.2) there are two torches, one heated shoe, one heated roller, one chilled roller and one chilled shoe. By using this system, laminate has more time at high temperature to create good bond before cooling down by chilled roller and chilled shoe. Moreover, this head helps to maintain the uniformity of temperature on a larger area. However their system is very complex, expensive, and it is not convenient to manufacturing on a curve mandrel.

Tape deposition head of AFP machine at Concordia is simpler with only one roller and the main heat source is a hot gas torch heated by nitrogen. The lack of heated shoe and chilled shoe creates higher gradient temperature in the same area. Therefore, higher residual stresses are built up in final laminate and cause deformation.

To solve the problem of warpage during manufacturing with simple AFP as one at Concordia University is a challenge. Several trials were done by Cai and Simpson but warpage of final laminate was still observed [18, 31].For the first trial, they used a vacuum pallet to hold a Siliglas G7 laminate which had been drilled many holes to make vacuum ports. Siliglas G7 laminate is a glass fabric reinforcement in a high temperature resistant silicon resin [32].A porous Teflon Tape was attached on top of the G7 laminate. Finally, AFP laid down

$[0]_8$ laminate on this Teflon Tape. The idea was to have the first ply bond well to the tool to keep laminate flat during the lay-down process. Layup sequence of the laminate is $[0]_8$. HGT temperature was 760^0C, nitrogen flow rate was 75 SLPM, layup speed was 2 inch/s (50.8 mm/s), and compaction force was 33.7 lbf (15kgf). The obtained laminate (Figure 2.8) was flatter than in their previous study [18] but it still did not meet requirements for testing. They noticed that vacuum could not hold the work down on the tool when laying the third ply.

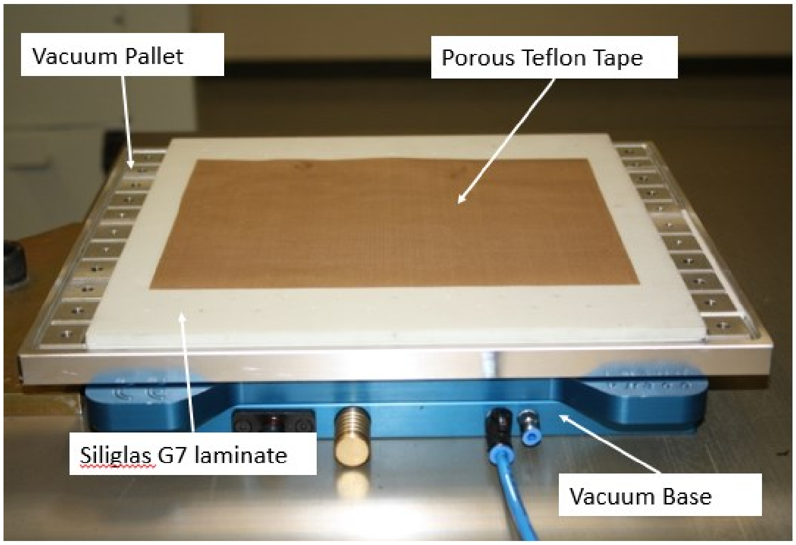


Figure 2.7 Vacuum tool setup for first attempt[31]

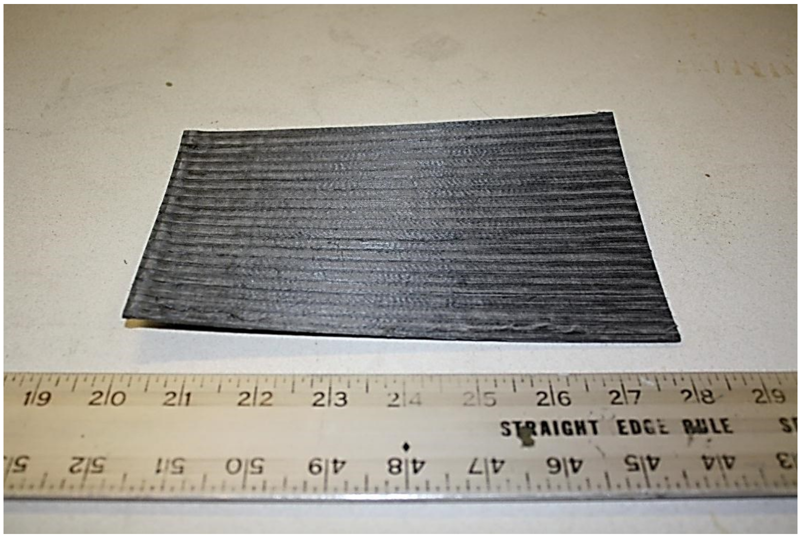


Figure 2.8 Fiber placed uni-directional laminate of $[0]_8$ after removing from the vacuum pallet (Attempt 1)[31]

Second attempt still used the vacuum pallet to hold a G7 laminate. The difference is the first ply was bonded manually to composite tool to improve the bonding between first ply and tool. All parameters were kept the same as previous trial. Figure 2.10 shows the laminate after removing from composite tool. Compared with previous attempt, this laminate is flatter. However, it is still slightly curved up in both direction.

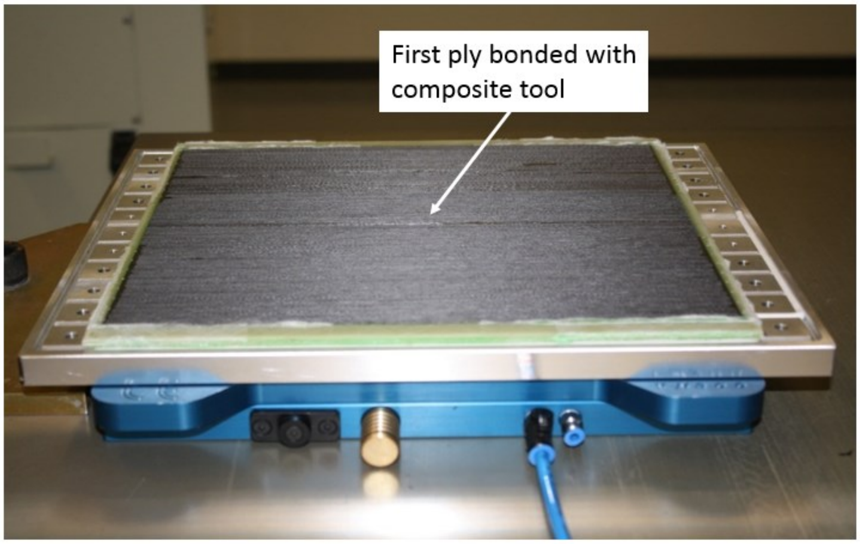


Figure 2.9 Vacuum tool setup for first attempt[31]

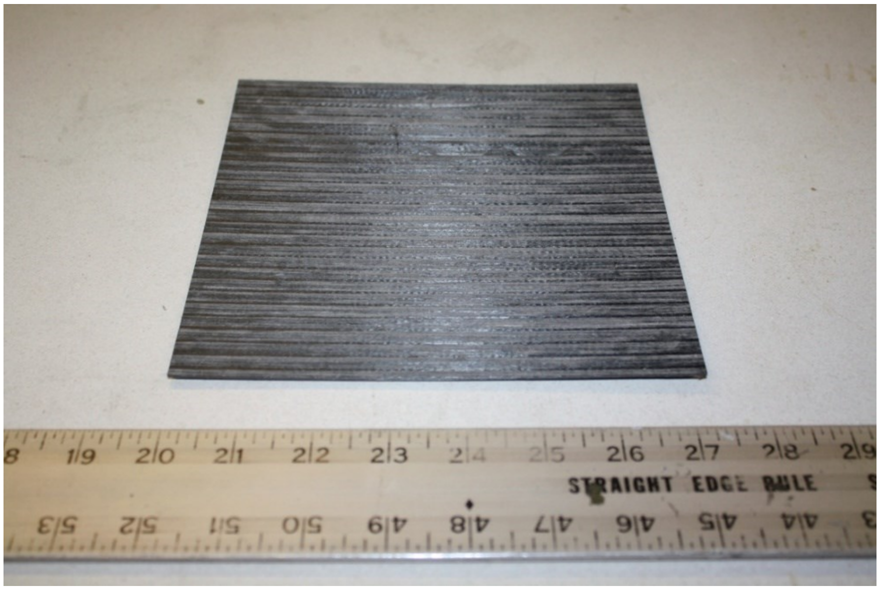


Figure 2.10 Fiber placed uni-directional laminate of $[0]_8$ after removing from the vacuum pallet (Attempt 2)[31]

2.2.2 Characterization of thermoplastic composites

Yoon and Sun [33] carried out uniaxial tension tests to characterize elastic-viscoplastic properties of AS4/PEEK composite. Unidirectional off-axis coupon specimens were cut from 0_{10} panel and tested while varying the monotonic strain rates under 2 different temperatures which are 24^{0}C (75^{0}F) and 121^{0}C (250^{0}F) consequently. They proved that overstress viscoplasticity model and modified Bodner and Partom's viscoplasticity model can be used to describe the viscoplasic behavior of the AS4/PEEK composite. Furthermore, comparison between these 2 models showed that in monotonic loading case, viscoplasticity model gives more accurate results than Bodner and Partom's viscoplasticity.

Ong and Liou [34] investigated the effect of impact damage on mechanical behaviors of thermoplastic composites. Initially, drop-weight impact was applied on 16 ply quasi-isotropic samples which are made of Gr/PEEK (AS4/APC-2 and IM7/APC-2). After the impact, these specimens were then passed to compression, tension and fatigue tests (tension-tension cyclic loading). Their results showed that both compressive and tensile strengths reduce after the impact damage but retention strengths for compression and tension are not the same. Moreover, for fatigue mechanical behavior, impact damages decrease the fatigue life but do not affect the mechanical behavior of laminates.

A recent work of Comer et al. [29] characterized some other properties of laminate made by laser-assisted ATP. The roller is made by silicone, roller pressure is 1.2 bar (17.4 psi); incoming tape and substrate is heated by 3kW diode laser heat source. Nominal target temperature is kept at 420^{0}C. Tool temperature is varied depending on layup sequences. However, the obtained laminate seems to have high void fraction. Open Hole Compression (OHC), flexural, Interlaminar Shear Strength (ILSS) tests were performed. Both modulus and strength are less than those made by autoclave.

2.2.3 Research objectives

This study has two objectives. The first objective is to propose a process in order to manufacture flat thermoplastic laminates by AFP with in situ consolidation which meet flatness requirement for testing standards. These thermoplastic composite laminates should also have acceptable quality in terms of void content, degree of bonding and crystallinity. The second objective is to characterize the materials made by the proposed process. This section combines several tests based on ASTM standards such as tensile, compression, and shear.

CHAPTER 3 Development of thermoplastic composites manufacturing process to make flat laminates by AFP

Summary: This chapter proposes a procedure to manufacture thermoplastic flat laminates with different layup sequences using AFP with in-situ consolidation. The outline for this chapter is as follows:

- Examination of raw materials
- Process to manufacture thermoplastic flat plates made by AFP with in-situ consolidation
- Preliminary quality verification for thermoplastic laminates made by AFP
    - Void content
    - Crystallinity

3.1 Examination of raw materials

There are different suppliers for thermoplastic material. In aerospace industry, the two biggest are CYTEC and TENCATE. In this study, carbon fiber-polyether ether ketone (APC2-AS4) from CYTEC was selected which has standard width of 0.25 inch (6.35 mm) and standard thickness of 0.05 inch (0.127 mm). As mentioned in data sheet of APC-2 PEEK from CYTEC website [4] this prepreg has 60% fiber volume, its glass transition temperature T_g is 143^0C.

3.1.1 Microscopic observation

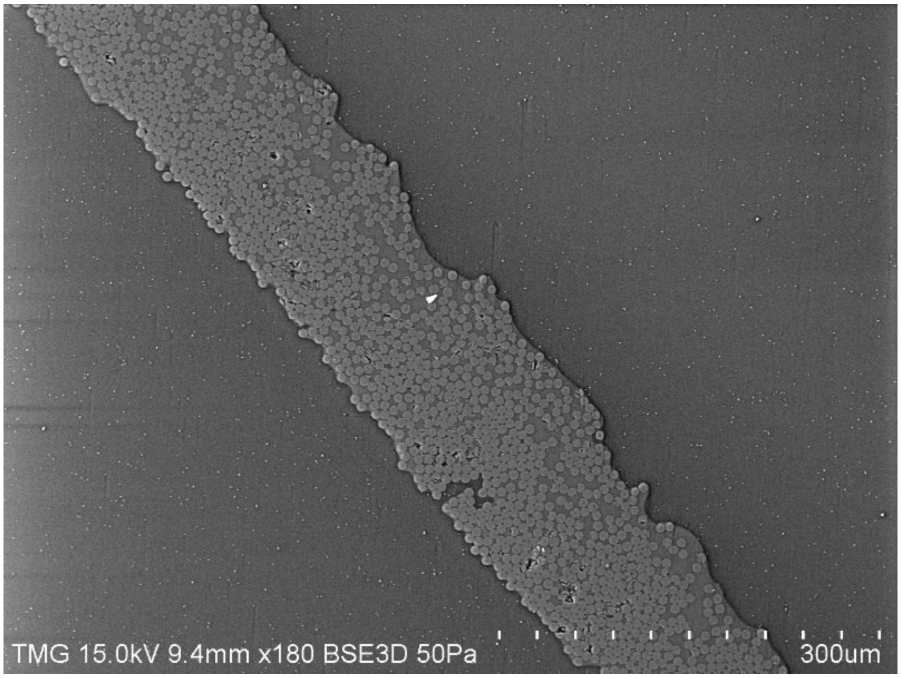
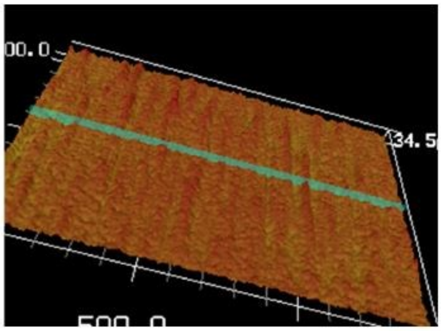
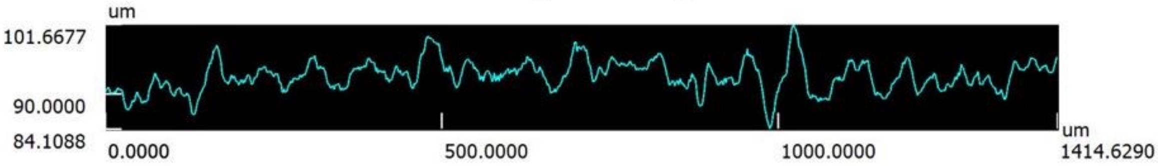


Figure 3.1 Micrograph of CYTEC prepreg



a) 3D view image



b) Roughness measurement

Figure 3.2 3D image and roughness measurement of CYTEC APC-2 PEEK Prepreg

Figure 3.1 shows a cross section of a tow of PEEK APC-2/AS4 from CYTEC. The average thickness of prepreg tape is 0.005 in (0.127 mm). It shows clearly the variation of surface roughness along tow width. Moreover, it is noted that the roughness on two sides are different, one with high surface roughness value and the other is smoother. The smoother side has higher fiber fraction and less resin near its surface than the opposite side of the prepreg. During layup process, the rougher side of the previous ply will be in contact with the smoother side of the current ply. By flipping the prepreg tape, the smoother side can be on top and will be in contact with the rougher side of the incoming tape. In reality, which side is up does not affect the quality of the final laminate. From the 3D view in Figure 3.2, it is noticed that on the surface of the prepreg, some fibers are exposed creating channels along fiber direction but not in transverse direction. These channels may affect the bonding between layers when fiber orientation between layers are not the same. Furthermore, as seen in Figure 3.1, there are some resin rich areas, some places where fibers are in contact with others and some small amount of void. These defects may have influence on the quality of the final products.

3.1.2 Crystallinity examination

During the process, thermoplastic prepregs undergo several heating and cooling cycles with high temperature gradient. Furthermore, these temperature cycles are not identical at all positions of the laminate so that it may not only create difference of crystallinity between raw material and final product but also degree of crystallinity (DOC) at different places of the laminates. In this section, we examined the DOC of APC2-AS4 prepreg to have a reference to compare with DOC of laminate after manufacturing process.

Differential Scanning Calorimetry (DSC) is used to determine DOC of thermoplastic composite by measuring the amount of heat going in and out of the sample. First of all, a small piece from the prepreg was cut and its weight was measured. After that, it was put into a pan and a lid was closed. Sample was then placed into test chamber with reference sample. Sample was then ramped up for 10^0C per minute up to 400^0C which is above melting temperature of PEEK. Heat flow was recorded and plotted versus time during the process (Figure 3.3).

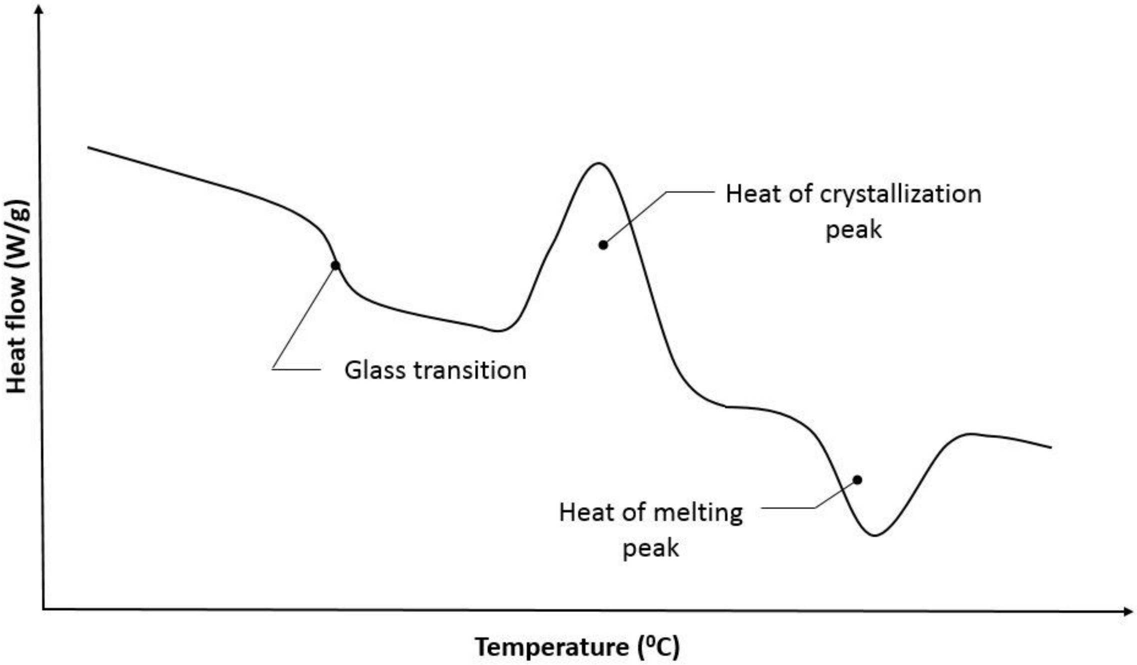


Figure 3.3 Typical Heat flow curve for semi-crystalline polymer.

On heating, the first peak represents crystallization process which is exothermic reaction ($\Delta H < 0$) and the second peak is when crystals melt which is endothermic reaction ($\Delta H > 0$). By integrating this curve, the amount of heat absorbed or released from sample is calculated and then it is used to calculate DOC by the following equation:

$$DOC = \frac{|\Delta H_m| - |\Delta H_c|}{\Delta H_f(1 - w_f)} \times 100\%$$

Where:

H_m: heat of fusion

H_c: heat of crystallization

ΔH_f: heat of fusion for a 100% crystalline material. $\Delta H_f = 130\, J/g$ for 100% crystalline PEEK [35].

w_f: weight fraction of fiber content in a composite. $w_f = 0.68\, for\, APC2/AS4$ from CYTEC

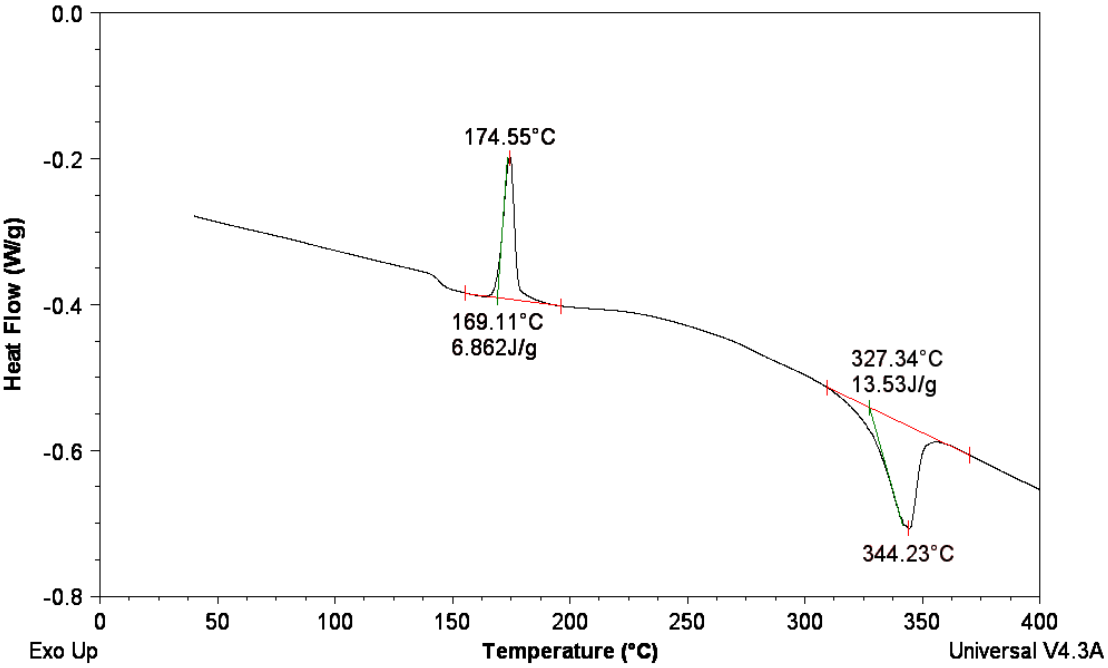


Figure 3.4 Heat Flow cuvre of APC-2/AS4 prepreg

Figure 3.4 illustrates the "heat flow" of APC-2/AS4 prepreg during heating process. It is clearly seen that the glass transition temperature is around 143^0C, heat of crystallization is 6.862 J/g, heat of fusion is 13.53 J/g and the DOC can be calculated as follow:

$$DOC_{prepreg} = \frac{13.53 - 6.862}{130(1 - 0.68)} = 16.47\%$$

3.2 Process to produce thermoplastic flat plate made by AFP and heated mandrel

3.2.1 Equipment

Automated Fiber Placement Machine

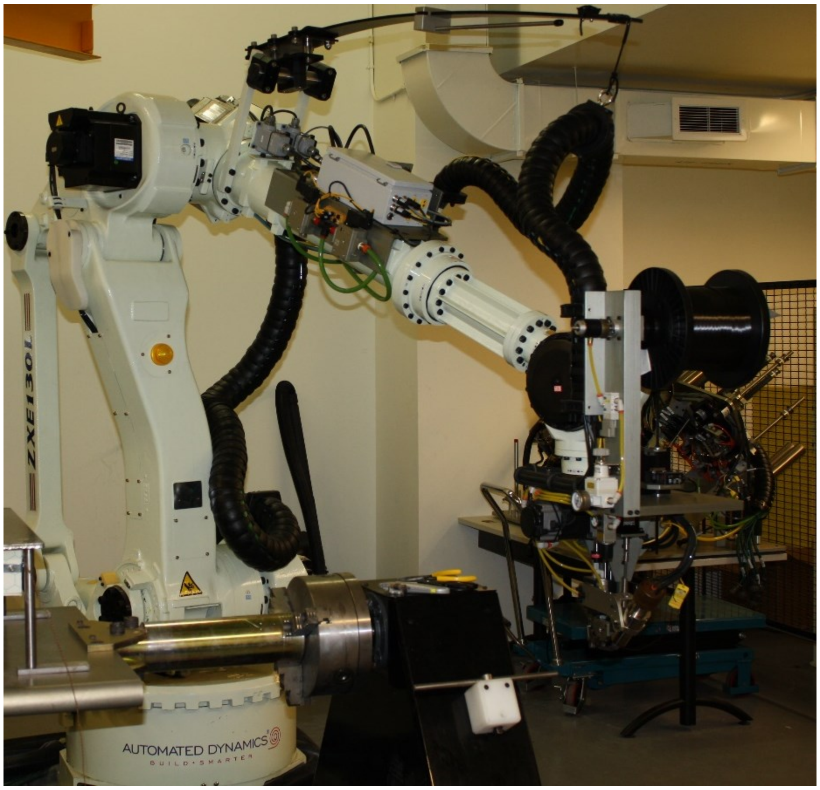


Figure 3.5 Automated Fiber Placement from Automated Dynamics

AFP machine used for manufacturing throughout this work is XTP-500 supplied by Automated Dynamics. The robot arm has 6 degrees of freedom controlled by 6 servo motors. The robot and control units are made by Kawasaki and the head which is used to lay down material is made by Automated Dynamics.

The head for thermoplastic composite composes principally a hot gas torch which is heated by nitrogen, roller, and material feeding unit. Therefore, main controllable parameters of AFP robot are nitrogen flow rate, hot gas temperature at the outlet of the torch, compaction force from roller applying to substrate and material feeding rate.

Hot gas torch is responsible to heat up incoming material and substrate to bring their surface temperature to be higher than melting temperature so that layers can be bonded together to form laminate under compaction force from roller. Position of this torch can be controlled manually in order to change the distance to roller center line and angle with horizontal line. Changing this parameter will vary the heat contribution from torch to roller and substrate and will affect the manufacturing process. A thermocouple is put inside the nozzle to measure temperature at the outlet of the nozzle and give feedback signal to controller. However, this thermocouple blocks the gas flow and makes uneven temperature distribution at the outlet and on the roller (Figure 3.7).

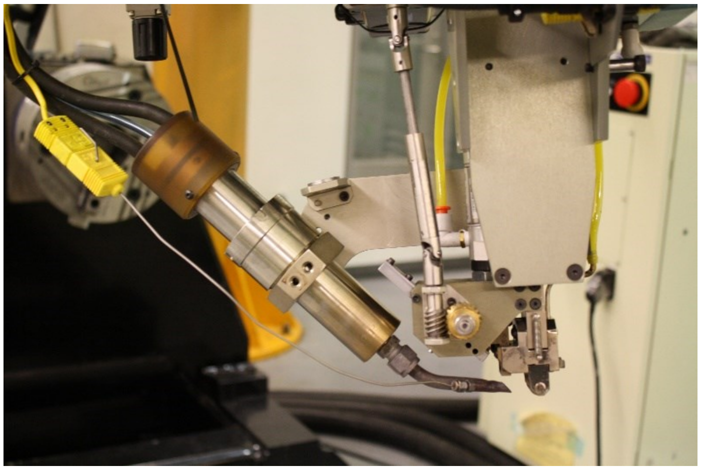


Figure 3.6 Gas Torch of the AFP

Roller is used to lay down material on to mandrel. Because geometry of the laminate can be flat or curved, different rollers are used for different geometries and different tow widths. In this study, roller with 0.5 inch (12.7 mm) diameter and 0.7 inch (17.8 mm) of width is chosen. Despite this roller is mostly used for 0.5 inch (12.7 mm) width band, it is noticed that using wider roller helps to have better surface finish and reduce impact on two sides of laid down tape. Roller is made from stainless steel, it plays a role of heat transfer element from gas flow to the incoming tape by conduction. Figure 3.7 shows variation of temperature along the width of 0.7 inch (17.8 mm) width roller when HGT was 900^0C. It is clearly seen that temperature profile is not symmetric with respect to mid-point. As mentioned above, the thermocouple blocks the outlet of the nozzle and causes this problem. However, the temperature is above 400^0C which is sufficient to melt the prepreg.

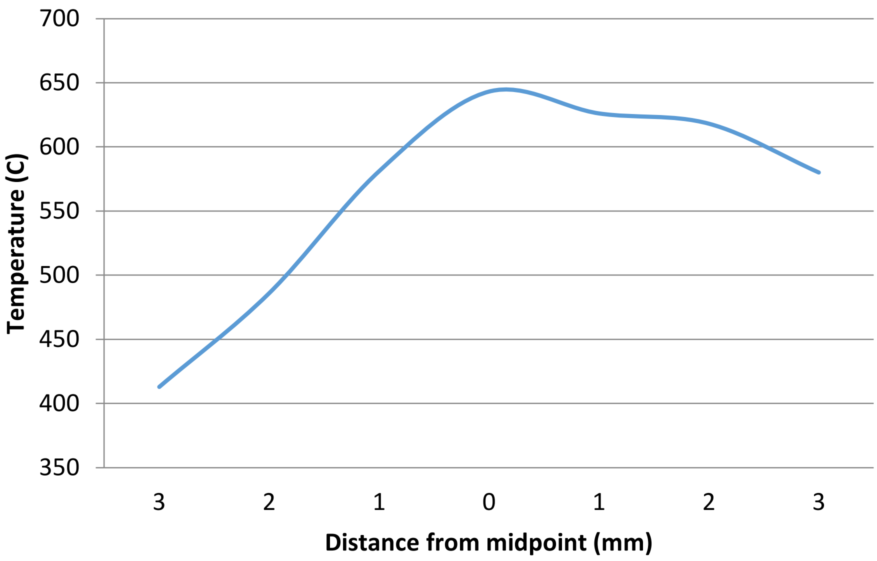


Figure 3.7 Temperature along the width of the roller when HGT=900^{0}C

Radiant heating panel

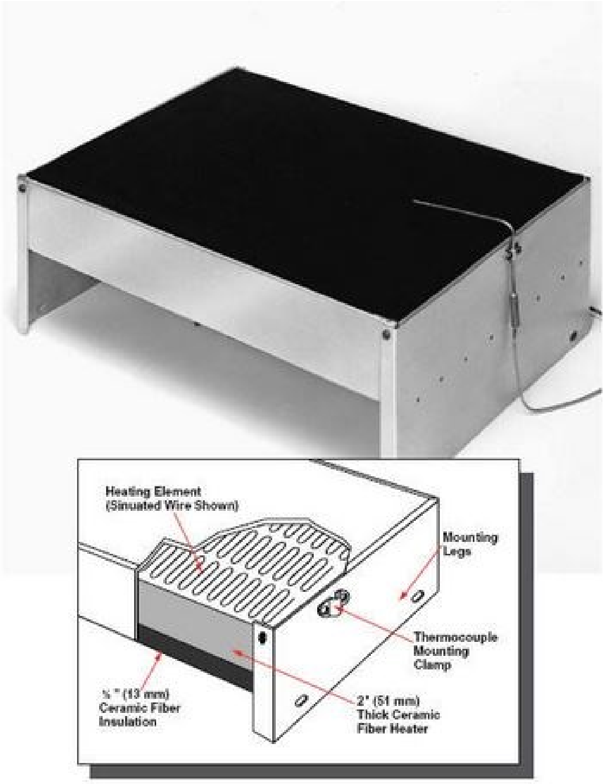


Figure 3.8 RAYMAX Radiant heating panel (Photo courtesy from Watlow website [36])

Heating panel used is RAYMAX 2030 made by Watlow. This is a non-contact heating equipment using sinuated wire as heating element. The advantage of radiant heater is its heating time which is typically three times faster than that of conventional convection heater. The heater selected for this study has the power of 5700 Watt, watt density of 30W/in^2 using electrical current of 600V; surface temperature is up to 1095^{0}C (2000^{0}F). Dimensions of this radiant heater are 24 inches (609.6 mm) long and 16 inches (406.4 mm) wide and the height is 8 inches (203.2 mm). At the bottom of the heater is placed a ½ inch (12.7 mm) thick ceramic fiber insulation to reduce heat loss.

Radiant heater is used to heat up steel table surface. It was placed underneath bottom surface of the steel table with a distance of 3 inches (76.2 mm).

Steel Table

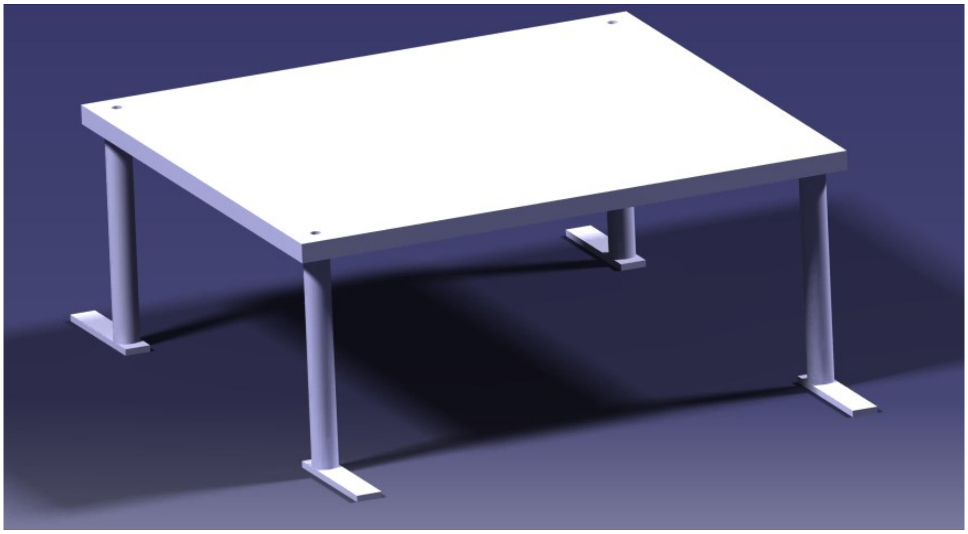


Figure 3.9 Steel table

Steel table is used as mandrel and material will be laid down on top surface. The reason to choose steel for the table is its small thermal expansion in comparison with aluminum, and also because of its higher stiffness. The small CTE helps to reduce mismatch between mandrel and laminate, and higher stiffness is to avoid deformation during manufacturing at high temperature. Dimension of the steel table is 30 inches (762 mm) long, 22 inches (558.8 mm) wide and 9.25

inches (235 mm) high. Surface thickness is 3/8 inch (9.5 mm). The bottom side of the table is coated with black paint in order to absorb more heat from the radiant heater.

	Steel
Thermal expansion coefficient ($\mathbf{10^{-6}\frac{m}{mK}}$)	12.0
Young modulus at room temperature (GPa)	200

Table 3.1 Thermal expansion and Young modulus of Steel [37]

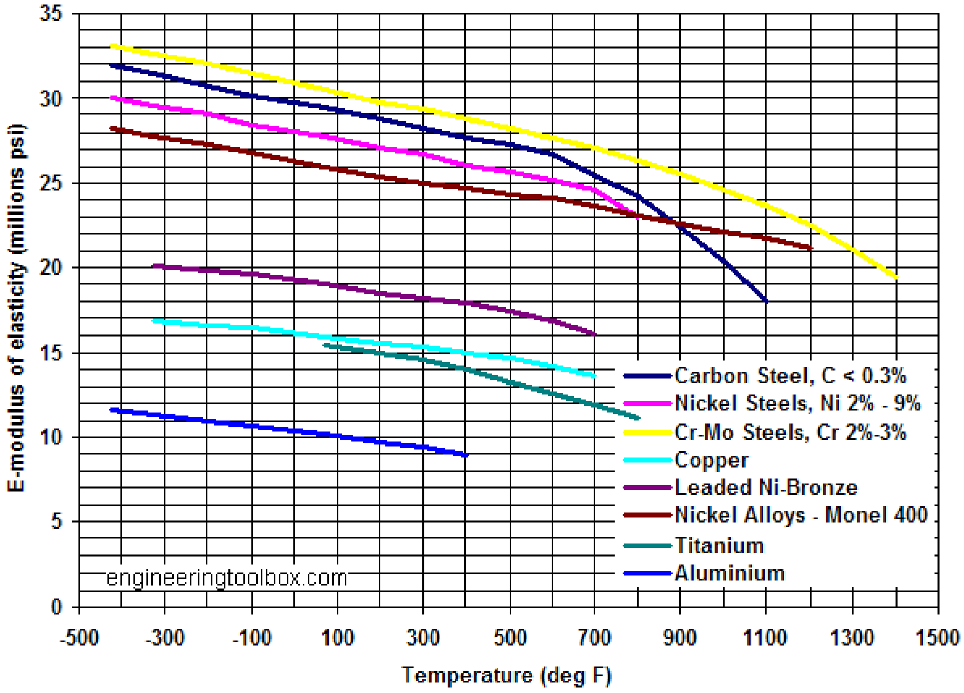


Figure 3.10 Young modulus versus temperature of different materials [37]

Temperature measurement equipment

Infrared camera and thermocouples were used to measure temperature during the process.

Infrared camera is FLIR camera which has a range from -120^{0}C to 1200^{0}C (248^{0}F to 2192 ^{0}F) but is divided into 3 smaller ranges which is -120^{0}C to 200^{0}C (248 ^{0}F to 392^{0}F), 200^{0}C to 650^{0}C (392 ^{0}F to 1202 ^{0}F), 650^{0}C to 1200^{0}C (1202 ^{0}F to 2192 ^{0}F).

Thermal couples type K are used to measure point temperature at several control points on the surface.

3.2.1 Setup and manufacturing

Heated table setup

Steel table was placed above radiant heater; distance from top surface of the heater to bottom surface of the table is 3 inches (76.2 mm). Due to dimensions of heater and table, distance from edge of the heater to edge of the mandrel in x direction is 3 inches (76.2 mm), and in y direction, that distance is also 3 inches (76.2 mm). Four legs of the table were clamped to the base to fix the table during manufacturing process. To avoid heat loss to environment, surrounding space between heater and steel table was covered by insolation panel. A thermocouple is placed at the middle of the bottom side of the mandrel and is connected to the controller for the feedback signal. In other words, the temperature of the bottom surface but not the top surface is controlled. In order to have information about surface temperature, infrared camera is used. The working area where material is laid down, is at the middle of the mandrel because this area has the most uniform temperature distribution.

Figure 3.11 Heated mandrel setup

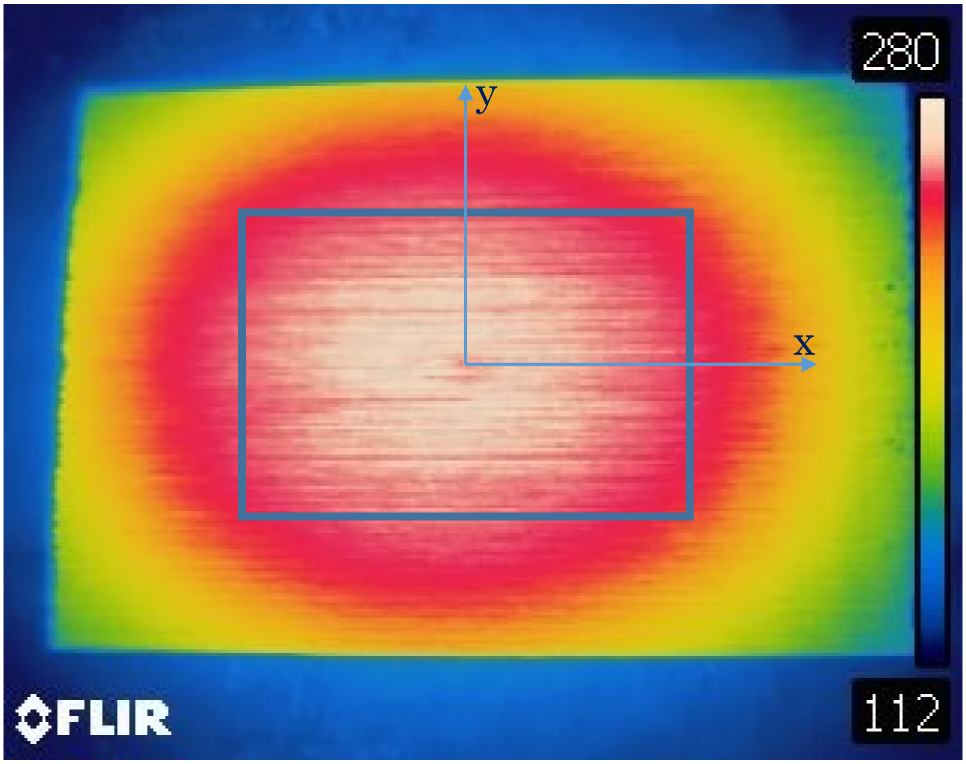


Figure 3.12 Temperature distribution on top surface of first layer.

Figure 3.12 shows temperature distribution on the heating table after laying the first ply. Heating table was set at 250^0C. Temperature drops about 40^0C from x=0 to x=±6 inch (152.4 mm) in x direction. In y direction, it drops about 40^0C from y=0 at the center to y= ±5 inch (127 mm). From this temperature distribution, the size of the laminate was decided to minimize stress due to temperature difference. The chosen dimension is 12 inch x 6 inch (304.8 mm x 152.4 mm) which is good enough to make samples for testing.

Measurement

Beside the thermocouple on the bottom side of the heated table, four others type K thermocouples are placed at different locations and are connected to a data acquisition system. These thermocouples are not to control the temperature but to verify the temperature distribution inside the working area during the process. Another independent source to measure temperature on the surface is using infrared camera.

Processing parameters

As mentioned in chapter 2, main controllable parameters during the manufacturing are Hot Gas Temperature (HGT), feed rate of the roller or layup speed, compaction force, nitrogen flow rate, and temperature of mandrel bottom surface.

3.2.2 Unidirectional laminates manufacturing

The very basic laminates to be manufactured are unidirectional laminates. Furthermore, in order to satisfy ASTM standards requirements for tensile and compression tests, laminates with 2 layup sequences $[0]_8$ and$[0]_{12}$ were made.

3.2.2.1 $[\mathbf{0}]_8$ *laminates*

Dimensions of $[0]_8$ laminates are 11 inches (279.4 mm) long and 6 inches (152.4 mm) wide. In order to make this laminate, the size for the first ply is 11 inches x 6 inches (279.4 mm x 152.4 mm). Layup direction is as shown in Figure 3.13. For all plies, prepreg was laid down from left to right. Since sliding may happen when laying down material directly on top of the substrate, for subsequent layers, it was decided to extend 0.25 inch (6.4 mm) outside the edge of previous layer at the beginning and at the end. This means the prepreg will be placed down and bond to the metal substrate at the starting point. To avoid building up weak contact at band boundary, the plies are offset by half of a band.

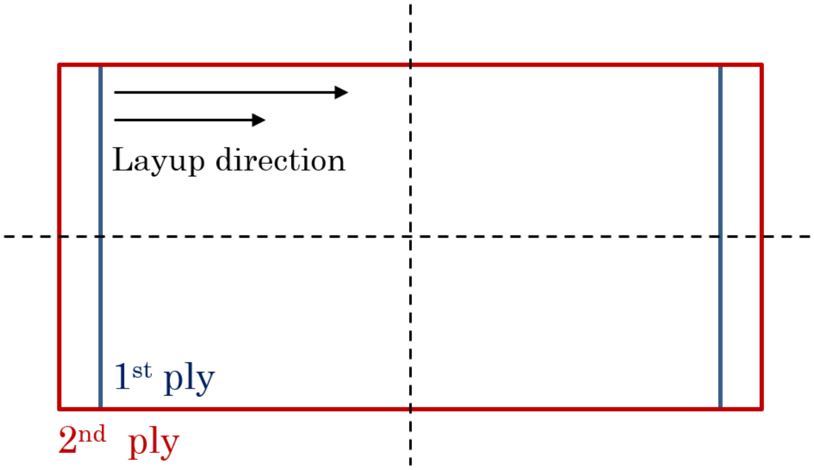


Figure 3.13 Layup direction for $[0]_8$ laminate

Processing parameters were set as in Table 3.2. Temperature at the bottom of the mandrel was kept at 200^{0}C during layup process. Temperature of Hot Gas Torch was 825^{0}C, nitrogen flow rate was 60 SLPM, layup speed was 0.5 in/s (0.76 m/minute) for the first ply and was increased to 1 in/s (1.52 m/minute) for plies two to eight. Compaction force was 100 lbf. In reality, during manufacturing, compaction forced increased gradually and could only be obtained this value after a certain time, around 1 second. Subsequently, one inch of each side of the plate was trimmed to get final laminate for testing.

Parameter	**Value**
Hot Gas Temperature	825^{0}C
Layup speed	0.5 in/s (0.76 m/minute) for 1st ply, 1 in/s (1.52 m/minute) for plies 2nd to 8th
Compaction force	100 lbf
Steel plate temperature	200^{0}C
Nitrogen Flow rate	60 SLPM

Table 3.2 Processing parameters for [0]$_8$ laminates

Once layup process is finished, laminate was left for natural cooling after manufacturing by turning off the heater. After cooling, the laminate was automatically released partly from mandrel. The laminate is flat, and only a small part at one edge was popped up from the surface (Figure 3.14). Furthermore, the surface finish on the top is good. In comparison with previous samples, this is the flattest laminate obtained, and it is good enough for tensile test.

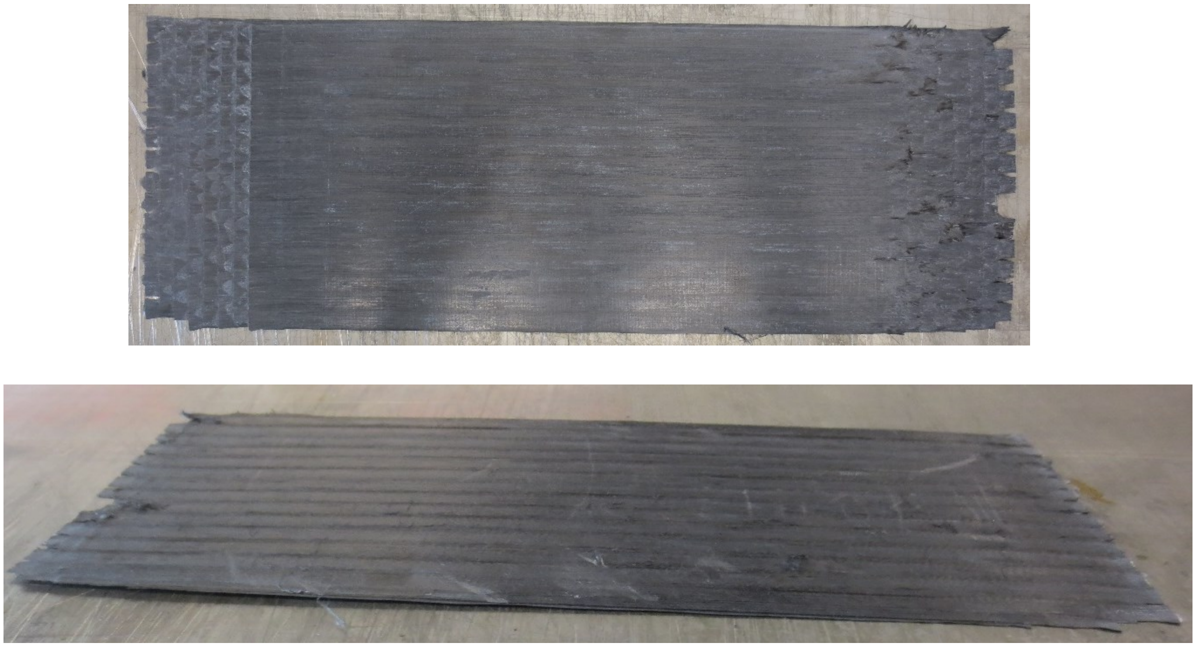


Figure 3.14[0]$_8$ laminate after removing from mandrel

3.2.2.2 $[\mathbf{0}]_{\mathbf{12}}$ *laminates*

Unidirectional composite laminates with 12 layers were made very similar to the one with 8 layers. This laminate was used to characterize compressive properties of material. Based on standard size of testing samples, dimensions for the first ply are 7 inches x 6 inches (177.8 mm x 152.4 mm). Layup direction was same as in $[0]_8$ laminate from left to right and a 0.25 inch offset on both sides after each layer was activated (Figure 3.13).

Processing parameters are presented in Table 3.3. Temperature at the bottom of the mandrel was kept at 200^0C during layup process. Temperature of Hot Gas Torch was 825^0C, nitrogen flow rate was 60 SLPM, layup speed was 0.5 in/s (0.76 m/minute) for the first ply and was increased to 1 in/s (1.52m/minute) for the plies two to twelve. Compaction force was 100 lbf.

There is a difference between the process for the 12 layer laminate and the 8 layer laminate. It was noticed that the temperature of the 9^{th} layer was too hot and the material was too soft to apply the 10^{th} layer. Therefore, compressed air was used to reduce the surface temperature. This helps to reduce the softness of the previously laid material and to improve bonding. The reduction of surface temperature also helped to obtain better surface finish.

Parameter	Value
Hot Gas Temperature	825^0C
Layup speed	0.5 in/s (0.76 m/minute) for 1st ply, 1 in/s (1.52 m/minute) for plies 2nd to 12th
Compaction force	100 lbf
Steel plate temperature	200^0C
Nitrogen Flow rate	60 SLPM

Table 3.3 Processing parameters for $[0]_{12}$ laminate

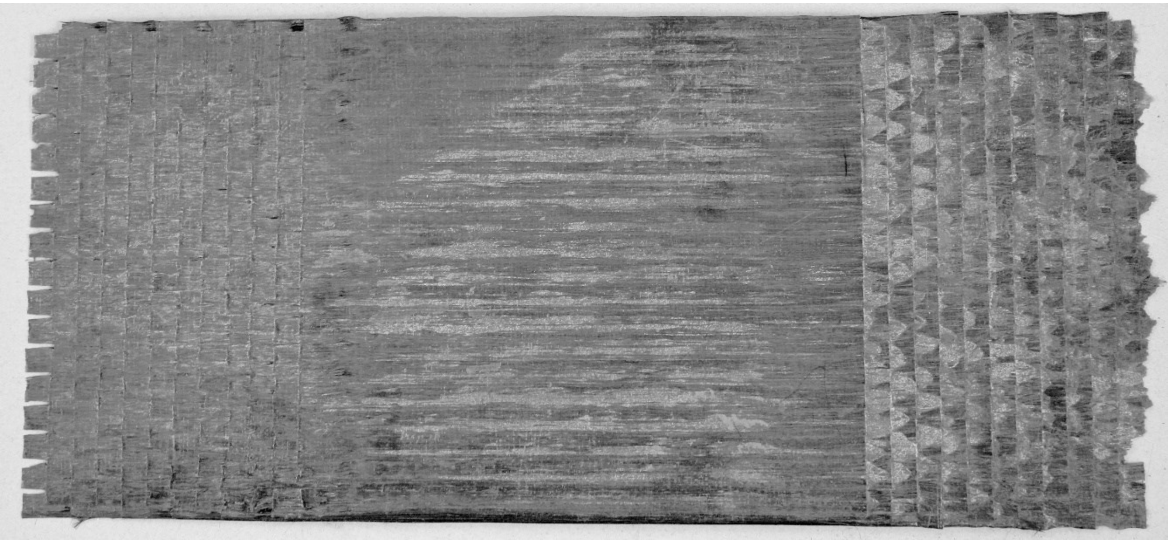


Figure 3.15 $[0]_{12}$ laminate after removing from mandrel

Once layup process is finished, laminate was left for natural cooling after manufacturing by turning off the heater. After cooling, the laminate was automatically released partly from mandrel. The laminate is flat, and only a small part at one edge was popped up from the surface (Figure 3.15).

3.2.3 $[45/-45]_{2S}$ laminates manufacturing

3.2.3.1 Manufacturing $[45/-45]_{2S}$ laminates without a caul plate

$[45/-45]_{2S}$ laminates were made in order to characterize shear modulus and shear strength of the material. The dimensions of these laminates are 11 inches x 7 inches (279.4 mm x 177.8 mm). First ply was laid down with size of 14 inches x 10 inches (355.6 mm x 254.0 mm) but the following plies have the same dimensions of 11 inches x 7 inches (177.8 mm x 152.4 mm). In other words, there was no offset of every layer to have the start point on metal mandrel. Otherwise, from the second ply of $[45/-45]_{2S}$ laminate, the starting point was right on the composite substrate. The reason of having start point on the previously laid substrate is incoming tape did not bond well to metal mandrel if the temperature of mandrel was too low. However, keeping the mandrel temperature at 200^0C makes the substrate become too soft and laying down material on this soft substrate will destroy the laminate. Layup direction is as shown in Figure 3.16. Layup direction for 45^0 layers is in negative x and negative y direction; layup direction for -45^0 layers is in positive x and negative y direction.

Processing parameters are presented in Table 3.4. Mandrel temperature is set to 200^0C during layup of the first two plies and then was reduced to 155^0C during the layup of 3rd to 8th ply. Layup speed is 0.5 in/s (0.76 m/minute) for the first ply and is increased to 1 in/s (1.52 m/minute) for the remaining process. HGT is 825^0C and compaction force is 100 lbs. Moreover, starting from the 3rd ply, compressed air was used to cool down the surface simultaneously in order to keep the top surface not too soft. After finishing the 8th ply, the mandrel temperature was set to 170^0C and was held for 10 hours. Afterward, the mandrel temperature was reduced with the rate of 1^0C/min to 140^0C and then the mandrel was cooled down naturally by turning off the heater.

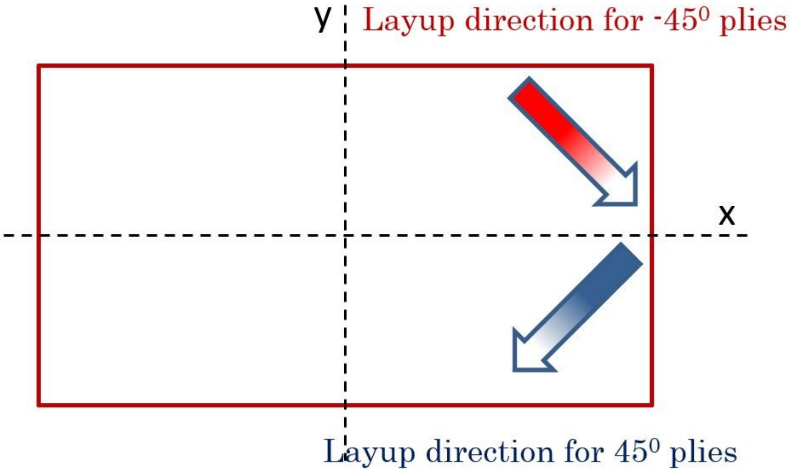


Figure 3.16 Layup direction for $[45/-45]_{2S}$ laminate

Parameter	Value
Hot Gas Temperature	825^0C
Feed rate of the Roller	0.5 in/s (0.76 m/minute) for 1st ply, 1 in/s (1.52 m/minute) for plies 2nd to 8th .
Compaction force	100 lbs
Tool surface temperature	200^0C for 1st and 2nd ply, 155^0C for 3rd to 8th ply
Nitrogen Flow rate	60 SLPM

Table 3.4 Processing parameters for $[45/-45]_{2S}$ laminate without caul plate

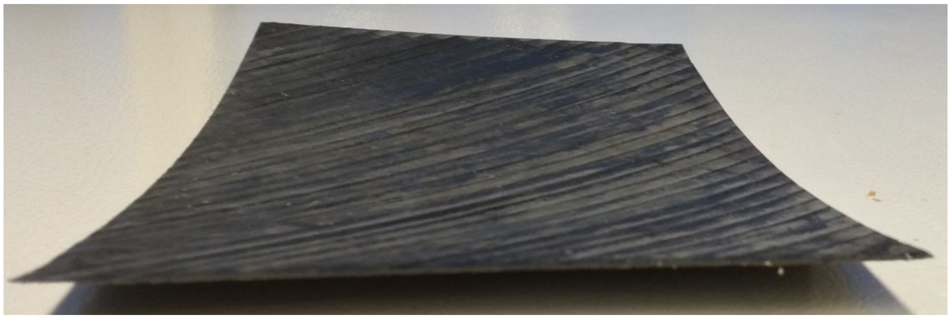


Figure 3.17 $[45/-45]_{2S}$ laminate after removing from mandrel (without caul plate)

Figure 3.17 shows the $[45/-45]_{2S}$ laminate after trimming the edges. It is clearly seen that the laminate is not as flat as the unidirectional $[0]_8$ and $[0]_{12}$ laminates. Even when the temperature was kept at 170^0C for long time to release residual stresses, deformation still happened. It can be explained by the uneven cooling rate on through the thickness of the laminate during cooling process combined with the difference of thermal expansion in different directions. The bottom surface attached to the heated mandrel and top surface is in contact with air. After turning off the heater, top surface is cooled down much faster than bottom surface.

3.2.3.1 Manufacturing $[45/-45]_{2S}$ laminate with a caul plate

In order to make cooling rate through the thickness of the laminate to be uniform, it is required to have same temperature conditions on both sides. The bottom side was stuck to the mandrel and could not be removed when the laminate was hot. Therefore, a caul plate with the same material and same dimensions as the mold was used to put on top the laminate.

Parameter	Value
Hot Gas Temperature	825^0C
Feed rate of the Roller	0.5 in/s (0.76 m/minute) for 1st ply, 1 in/s (1.52 m/minute) for plies 2nd to 8th .
Compaction force	100 lbs
Tool surface temperature	200^0C for 1st and 2nd ply, 155^0C for 3rd to 8th ply
Nitrogen Flow rate	60 SLPM

Table 3.5 Processing parameters for $[45/-45]_{2S}$ laminate with caul plate

The dimensions of these laminates are 11 inches x 7 inches (279.4 mm x 177.8 mm). First ply was laid down with size of 14 inches x 10 inches (355.6 mm x 254.0 mm) but the following plies have the same dimensions of 11 inches x 7 inches (279.4 mm x 177.8 mm). In other words, there is no offset of every layer to have the start point on metal mandrel. Otherwise, from the second ply of $[45/-45]_{2S}$ laminate, the starting point is right on the composite substrate. Layup direction is as shown in Figure 3.16. Layup direction for 45^0 layers is in negative x and negative y direction; layup direction for -45^0 layers is in positive x and negative y direction.

Processing parameters are presented in Table 3.5. Mandrel temperature was kept at 200^0C during of the first two plies and then was reduced to 155^0C during the layup of 3rd to 8th ply. Layup speed is 0.5 in/s (0.76 m/minute) for the first ply and is increased to 1 in/s (1.52 m/minute) for the remaining process. HGT is 825^0C, nitrogen flow rate is 60 SLPM and compaction force is 100 lbs. Moreover, starting from the 3rd ply, compressed air was used to cool down the surface simultaneously in order to keep the top surface not too soft. After finishing the 8th ply, a caul plate was placed on top of the laminate. Insulation panel was used to cover the top of the caul plate. Heated mandrel temperature was set at 200^0C. Heater was left for one hour to bring temperature of the whole assembly to 200^0C. Finally, heater was turned off and laminate was cooled down naturally between two caul plates.

a) Top view

b) Isometric view

Figure 3.18 $[45/-45]_{2S}$ laminate after removing from mandrel (with caul plate)

((a) Top view (b) Isometric view)

Figure 3.18 shows the $[45/-45]_{2S}$ laminate after trimming the edges. The laminate is flat and there is no warpage. It is clearly seen that using a caul plate with same temperature as mandrel placing on top made cooling rate through thickness uniform. Furthermore, hot caul plate helped to reduce cooling rate of the whole laminate so it had more time at temperature higher than T_g to release residual stresses due to cooling. The obtained laminate met requirement for testing.

3. 3 Preliminary quality verification

3.3.1 Microscopic observation

$[0]_8$ *laminates*

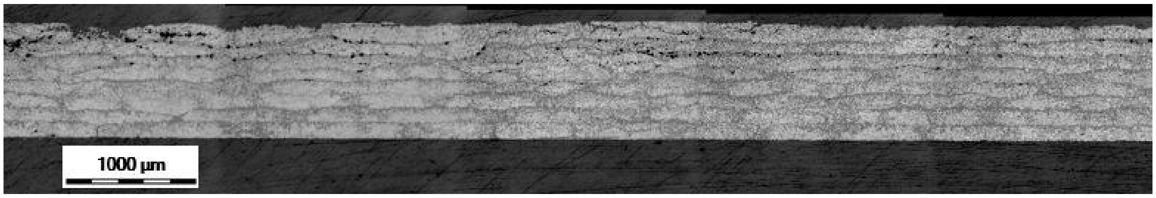


Figure 3.19 Microscopic image for [0]$_8$ laminate sample

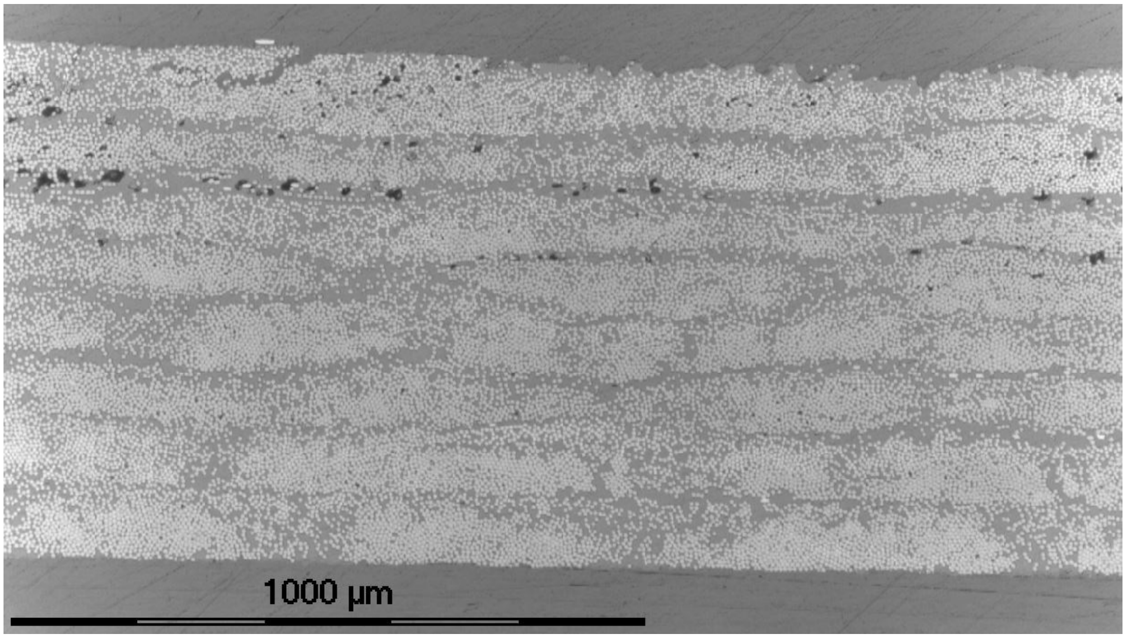


Figure 3.20 Microscopic image for [0]$_8$ laminate sample (Higher magnification)

Figures 3.19 and 3.20 show microscopic images for $[0]_8$ laminates made by AFP. From those pictures, it is noted that there are defects inside laminate such as voids and resin rich areas. Resin rich areas which are found inside layers are due to prepreg quality. Resin rich areas which are located at interlayer boundaries can be explained by high surface roughness of the material. Surface roughness creates empty channels along fiber direction. When AFP head laid down a tow on substrate, it squeezed resin to fill the spaces created by surface roughness. These areas can be reached only by resin but it is difficult for fibers to go in. Voids are observed to be at interface between two adjacent layers starting from 6th layers (Figure 3.19). The reason is by laying down the next ply on the substrate, roller did a re-compaction pass on the previously laid layers and enhanced the reduction of void. The bottom plies had more repass so had less void while for few plies on the top, they had less numbers of repass and had more void. Void fraction was measured by 2D microscopic analysis. For this laminate, that value is around 0.95%. Effect of repeat compaction will be described in section 3.4.

$[\mathbf{0}]_{\mathbf{12}}$ *laminates*

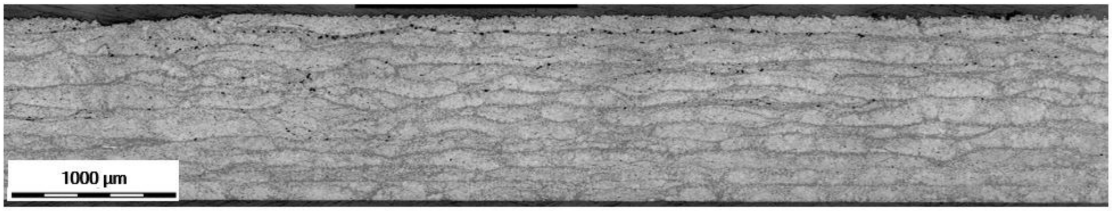


Figure 3.21 Microscopic image for $[0]_{12}$ laminate sample

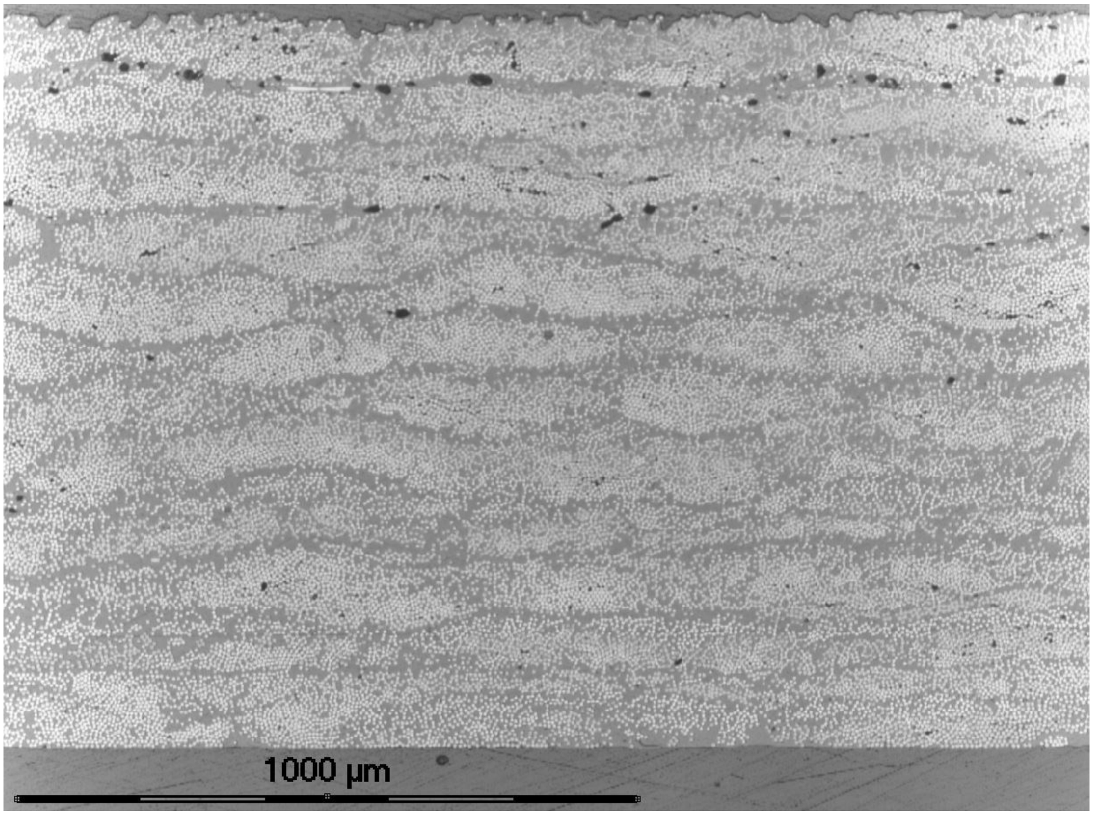


Figure 3.22 Microscopic image for $[0]_{12}$ laminate sample (Higher magnification)

Figures 3.21 and 3.22 show microscopic images for $[0]_{12}$ laminates made by AFP. From those pictures, it is noted that there are defects inside laminate such as voids and resin rich areas. Resin rich areas which are found inside layers are due to prepreg quality. Resin rich areas which are located at interlayer boundaries can be explained by high surface roughness of the material. Surface roughness creates empty channels along fiber direction. When AFP head laid down a tow on substrate, it squeezed resin to fill the space created by surface roughness. These areas can be reached only by resin but it is difficult for fibers to go in. Voids are observed to be at interface between two adjacent layers at top layers (Figure 3.21). 2D microscopic analysis was performed to determine void content in the laminate. Void fraction for this laminate is around 0.93%. Void appeared in $[0]_{12}$ laminate does not start from 6th ply but from 8th ply. It can be explained that by laying down the next ply on the substrate, roller applied a re-compaction pass on the previous laid layers and enhanced the reduction of void. The bottom plies had more repass so had less void while for few plies on the top, they had less numbers of repass and had more void.

$[45/-45]_{2S}$ *laminate*

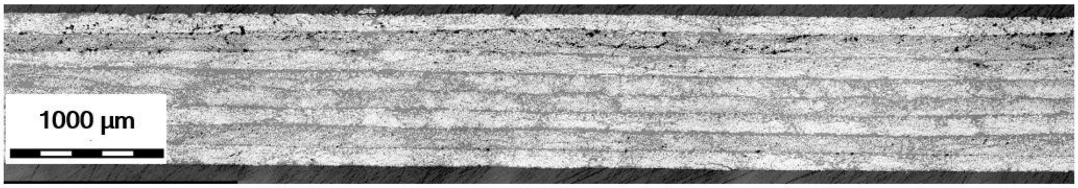


Figure 3.23 Microscopic image for [45/-45]$_{2S}$ laminate sample without caul plate

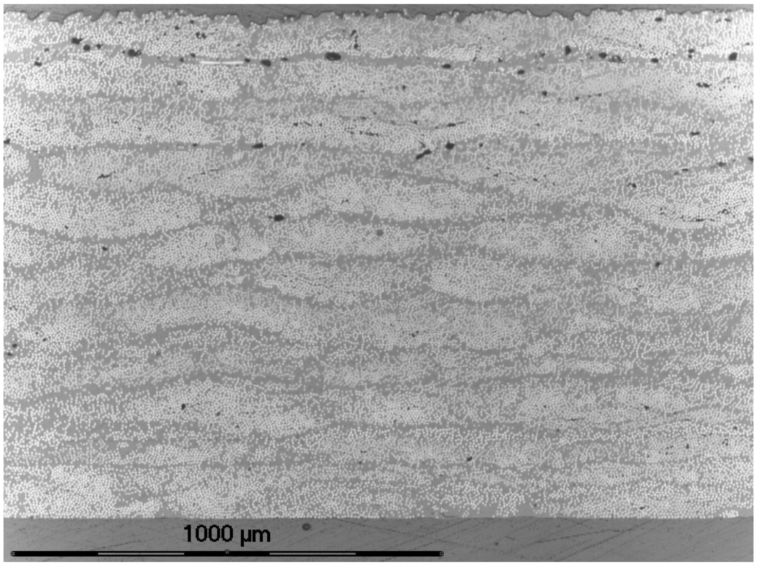


Figure 3.24 Microscopic image for [45/-45]$_{2S}$ laminate sample without caul plate (higher magnification)

Figures 3.23 and 3.24 show microscopic images for $[45/-45]_{2S}]$ laminates made by AFP without caul plate. From those pictures, it is noted that there are defects inside laminate such as void and resin rich area. Resin rich areas which are located at interlayer boundaries can be explained by high surface roughness of the material. Surface roughness creates empty channels along fiber direction. When AFP head laid down a tow on substrate, it squeezed resin to fill the space created by surface roughness. These areas can be reached only by resin but it is difficult for fibers to go in. Voids are observed to be at interface between two adjacent layers (Figure 3.23). Void fraction for this laminate is around 0.93%. In this case, interlayer voids appeared earlier starting from 2nd ply, not from 6th ply as in $[0]_8$ laminate and 8th ply as in $[0]_{12}$ laminate. The layup direction can be the reason of this difference. In $[0]_8$ and $[0]_{12}$ laminates, material was placed down in only one direction which was fiber direction. This direction is same as the direction of the channel created by surface roughness. In $[45/-45]_{2S}$ laminate, except for plies 4 and 5, all other two adjacent plies have different layup directions. This means the incoming tapes did not follow the empty channel direction but went perpendicular to it. This led to entrap more air between two adjacent layers and created voids.

3.3.2 Crystallinity

Crystallinity of laminates after manufacturing were checked by using DSC with the same procedure as with prepreg.

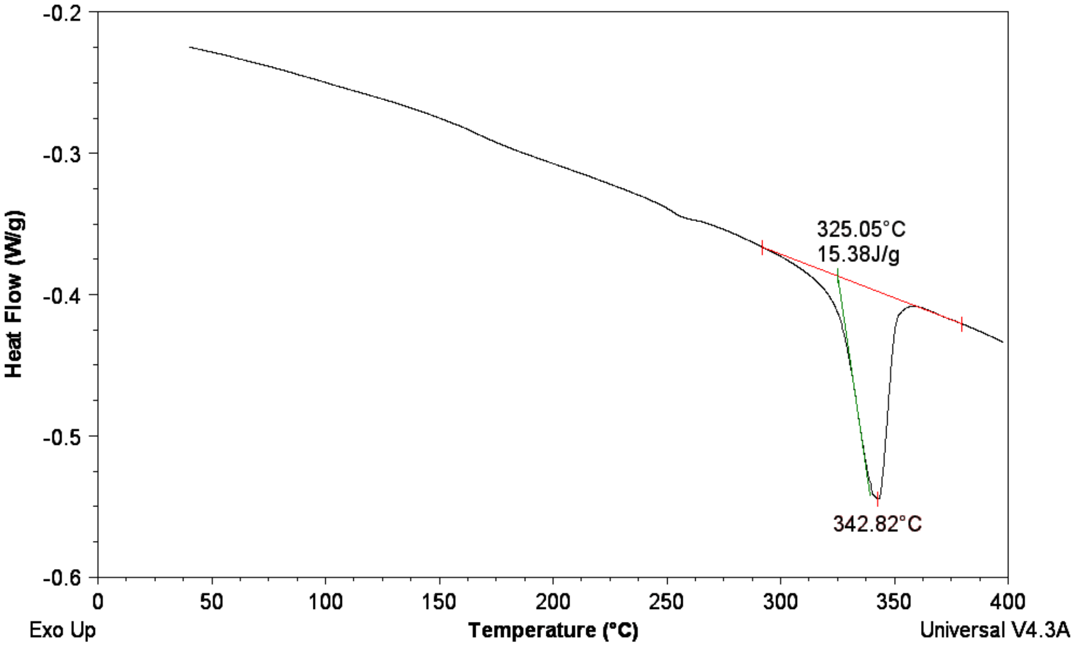


Figure 3.25 Heat flow of [0]$_8$ laminate sample

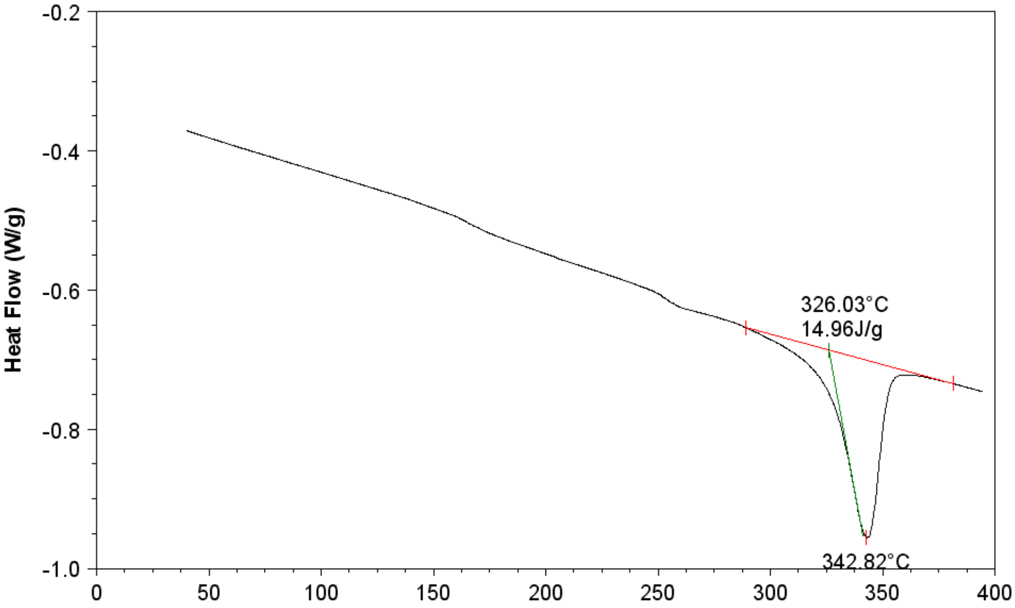


Figure 3.26 Heat flow of [0]$_{12}$ laminate sample

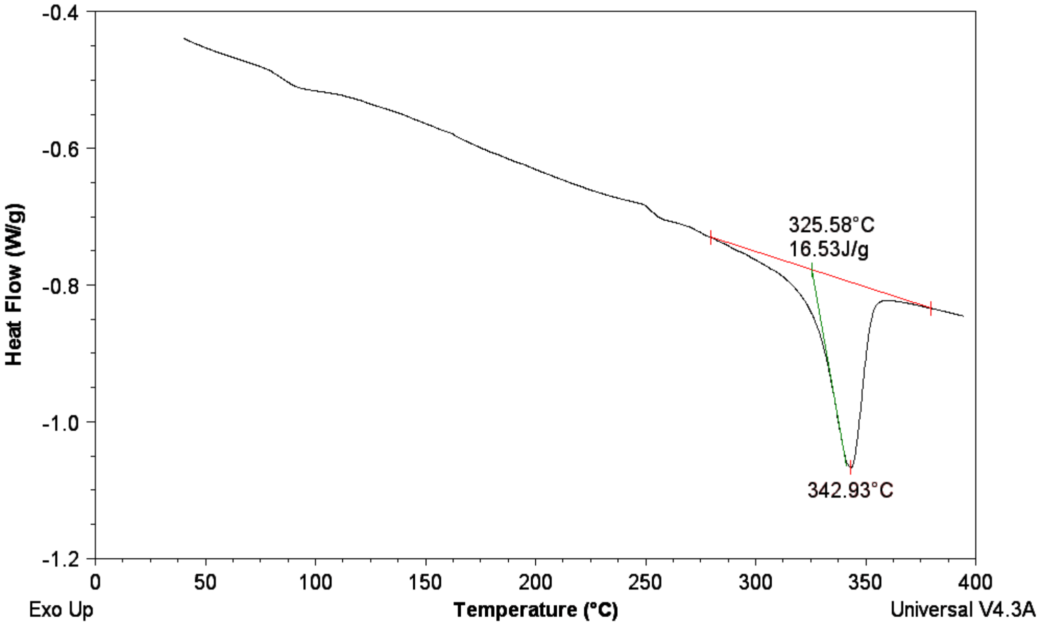


Figure 3.27 Heat flow of [45/-45]$_{2S}$ laminate sample

Figures 3.25, 3.26 and 3.27 show heat flow of $[0]_8$, $[0]_{12}$ and $[45/-45]_{2S}$ respectively. In all these three figures, it is noted that there is no peak representing crystallization of PEEK. In other words, the PEEK resin has reached it maximum level of crystallization.

3.4 Manufacturing thermoplastic laminates with repass

Interply voids which were observed in the laminates can affect their mechanical properties. During manufacturing, surface finish can become rougher, even some fibers can be pulled off due to the sticking of the tape to roller. This effect is more evident when the substrate becomes too hot. The bad surface finish of the previously laid layer is the reason for interply voids and resin rich area when the next ply is laid down. Judging from the quality of previous made laminates, the first few layers have good quality, while the top layers have more voids. Layers that were deposited early have several repasses of compaction over them while layers that are laid later have less number of repasses. In order to improve the quality of the laminate, reducing defects and eliminating voids, repass for the top layers were made. Repass after material deposition can improve surface quality, makes the surface smoother before laying the next ply.

$[0]_8$, $[0]_{12}$ and $[45/-45]_{2S}$ laminates were made with the same manufacturing parameters as previous experiments. The difference this time is repass was applied after material deposition starting from 4th ply for $[0]_8$, $[0]_{12}$ laminates, and starting from 3th ply for $[45/-45]_{2S}$ laminate. The moving speed of roller for repass is 2 in/s. Compaction forced is kept at 100 lbf, HGT is 825^{0}C, and Nitrogen flow rate is 60 SLPM.

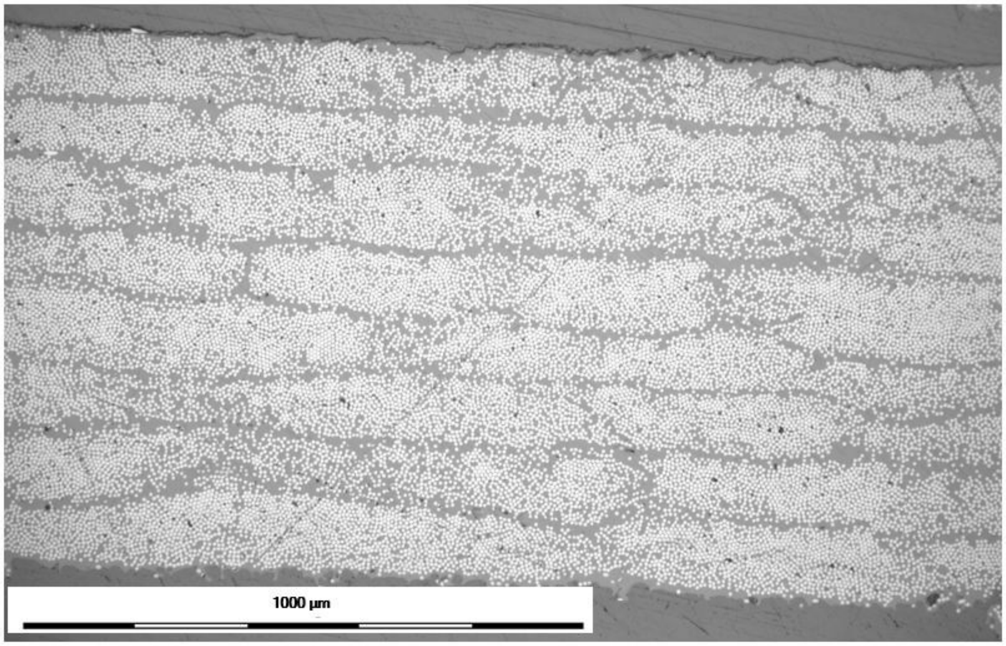


Figure 3.28 Microscopic image for $[0]_8$ laminate sample with 1 repass

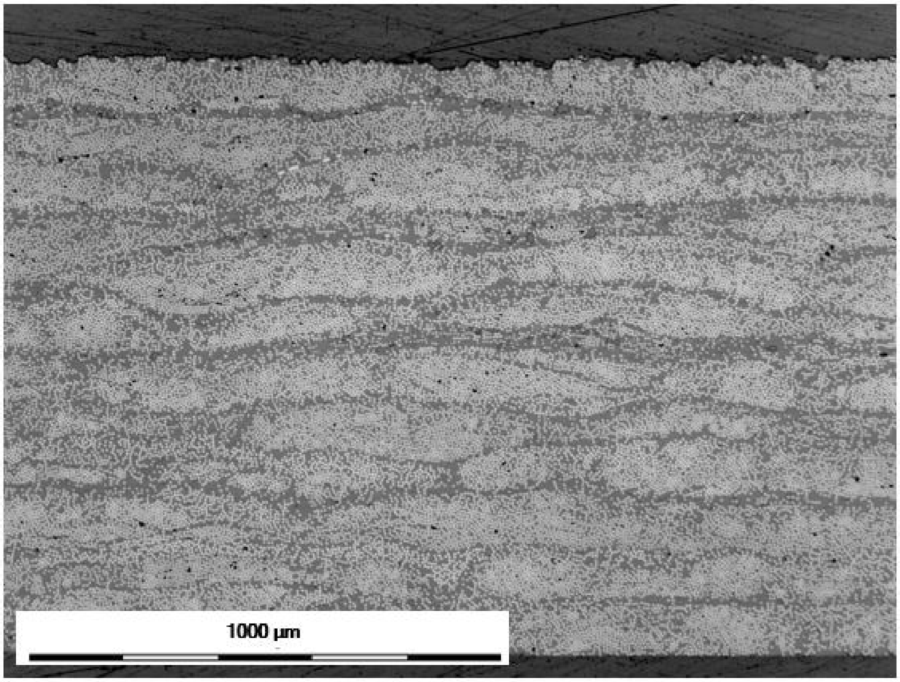


Figure 3.29 Microscopic image for $[0]_{12}$ laminate sample with 1 repass

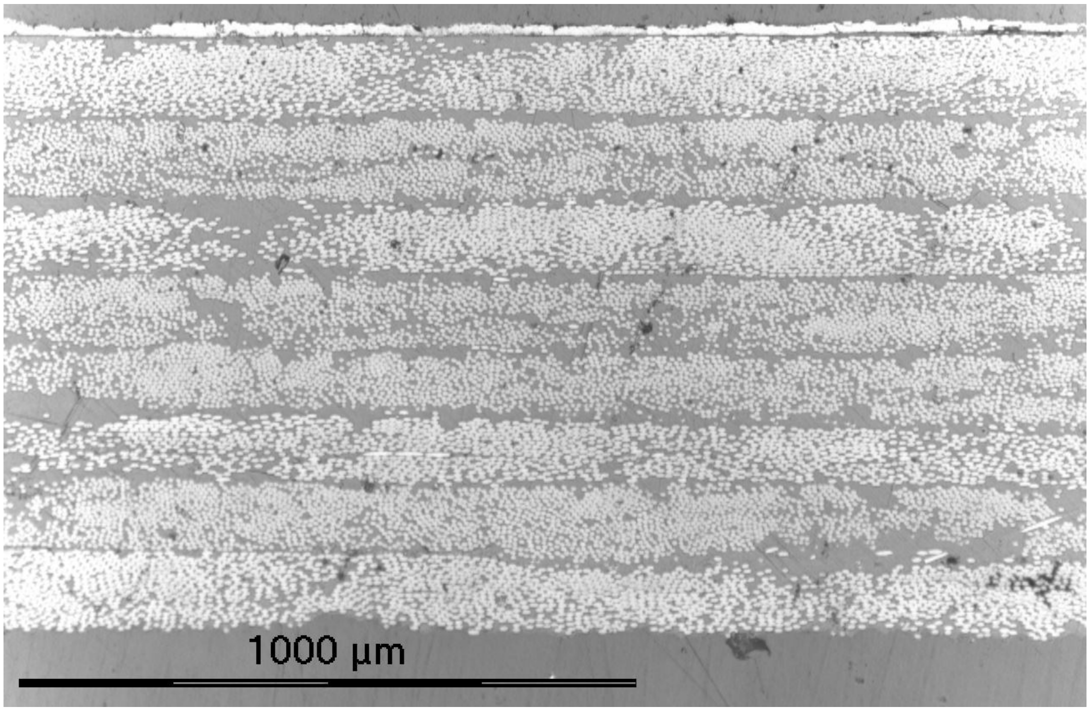


Figure 3.30 Microscopic image for $[45/\text{-}45]_{2S}$ laminate sample with 1 repass

Figures 3.28, 3.29 and 3.30 show the microscopic image for $[0]_8$, $[0]_{12}$ and $[45/-45]_{2S}$ consequently. Comparing with those without repass (Figures 3.19, 3.21, and 3.23), it is clearly seen that the interply voids fraction reduces significantly. However, resin rich areas do not reduce when applying repass. This is because the resin is entrapped inside channels along fiber direction and it cannot be removed with repass.

3. 5 Discussion

From different laminates made by AFP using heated mandrel, it can be concluded that using heated mandrel is necessary to get flat laminates. Manufacturing parameters, especially heated mandrel temperatures depend on layup sequence, thickness of the laminate, and relative direction of the incoming tape with the fiber direction of the previously laid ply. Thickness of the laminate, and layup sequence affect heat transfer from bottom to top layers. Heat conductivity coefficient along fiber direction (z) is much higher than in transversal to fiber direction (Table 3.6). Therefore, laminates will have different thermal conductivities in different directions depending on their layup sequences. This leads to quantity of heat retained inside laminates not the same. Temperature inside substrate should be in a range which is high enough to melt resin and bond tape to substrate but it should be low enough that substrate is not too soft. If substrate is too soft, roller digs into substrate and ploughs fibers of the substrate out. It creates damage on surface of the substrate so incoming tape cannot be bonded to substrate.

Repass after layup process for each layer reduces voids by improving surface finish after depositing material by the first pass.

DOC of final laminates were examined using DSC and it is noted that all thermoplastic resin has reached its maximum degree of crystallinity.

$k_{xx}(W/m \cdot {}^{o}C)$	6.8
$k_{yy}(W/m \cdot {}^{o}C)$	0.658
$k_{zz}(W/m \cdot {}^{o}C)$	0.658

Table 3.6 Thermal conductivity at room temperature of APC-AS4 unidirectional laminate[38]

CHAPTER 4 Characterization of laminates made by AFP with in-situ consolidation

The purpose of this chapter is to characterize the mechanical properties of flat thermoplastic composites laminates made by AFP and compare with samples made by HLU with autoclave. All tests were implemented on the MTS hydraulic machine with the maximum load of 100 kN.

4.1 Tests description and standard

4.1.1 Tensile Test

The tensile test is used to determine in-plane tensile properties such as strength and modulus of the composite. The tensile test in this study follows ASTM D3039-07 [39]. Laminates $[0]_8$ made by AFP were first trimmed to have 10 in long plates. Tabs were then bonded with high strength adhesive to the plate on both sides. The middle area without tab is 5.5 in long. Finally, this plate was cut to have 5 specimens of 10 in x 0.5 in. The strain transducers were implemented at mid-length, mid-width location of specimens. Force was applied to the specimens at the specified rate (2 mm/s) until failure; stress versus strain data were recorded. Finally, tensile modulus of elasticity, tensile strength were obtained from the curve.

Figure 4.1 Specimens for tensile test

4.1.2 Compression Test

The compression test is used to determine in-plane compressive strength and modulus of the composite. ASTM D3410-03 was used as standard. Laminates $[0]_{12}$ made by AFP were first trimmed to have 5.5 in long plates. Tabs were then bonded with high strength adhesive to the plate on both sides. The middle area without tab is 0.5 in long. Finally, this plate was cut to have 5 specimens of 5.5 in x 0.5 in. The strain transducers were implemented at mid-length, mid-width location of specimens. The gage length in case of compression test is 0.5 in. The IITRI Compression Test Fixture is used for this test. This fixture which is introduced by Illinois Institute of Technology Research Institute [40] and is started to be used in ASTM standard in 1987 [41], is to assure that the specimen will fail by compression and not by other modes. Force was applied to the specimens at specified rate (1.5 mm/s) until failure. Stress versus strain data were recorded. Afterward, compressive modulus and compressive strength were obtained from the curve.

Figure 4.2 Specimens for compression test

4.1.3 In-plane shear Test

The purpose of in-plane shear test is to characterize in-plane shear properties such as shear strength and modulus. In-plane shear test follows ASTM D3518 [42]. Laminates $[45/-45]_{2s}$ made by AFP were first trimmed to have 10 in long plates. Tabs were then bonded with high strength adhesive to the plate on both sides. The middle area without tab is 5.5 in long. Finally, this plate was cut to have 5 specimens of 10 in x 1 in. The strain transducers were implemented at mid-length, mid-width location of specimens. The gage length in case of compression test is 5.5 in. Afterward, force was applied to the specimens at the specified rate (2mm/s) until failure. Stress versus strain data were recorded. Finally, in-plane shear modulus, shear strength were obtained from the curve.

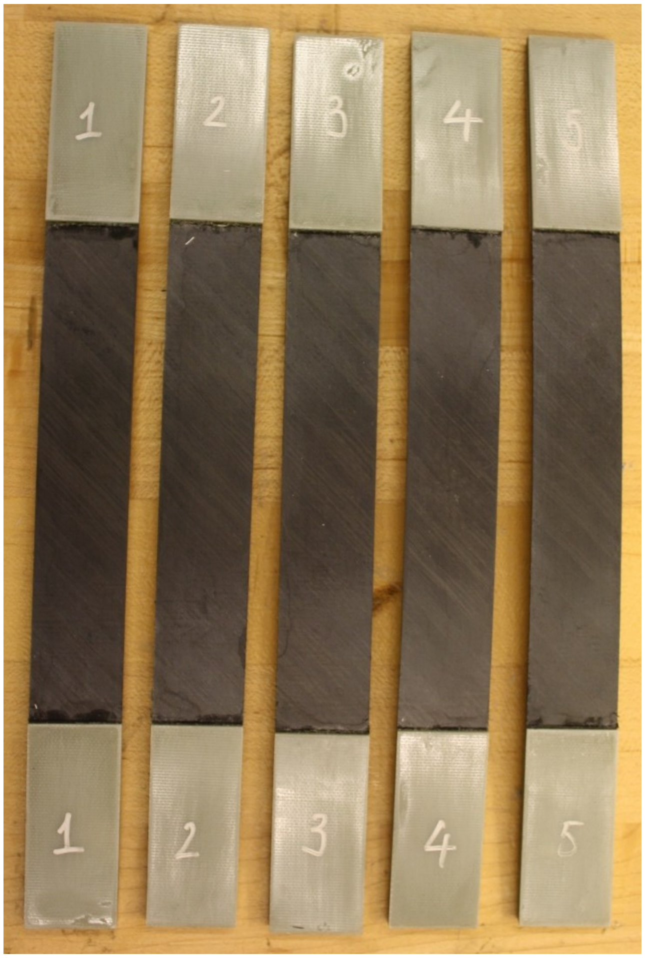


Figure 4.3 Specimens for in-plane shear test

4.2 Results and discussion

4.2.1 Tensile Test

Figure 4.4 shows the Stress-Strain curve of the tensile test. Five samples were tested and sample after failure is shown in Figure 4.5. During testing, it is noticed that delamination happened first, then followed by fiber fracture. The failure mode and area is DGM. The first character represents for failure mode, D means *Delamination*. The second character represents for failure area, G means at *Gage*. The third character represents for failure location, M means *Middle.*

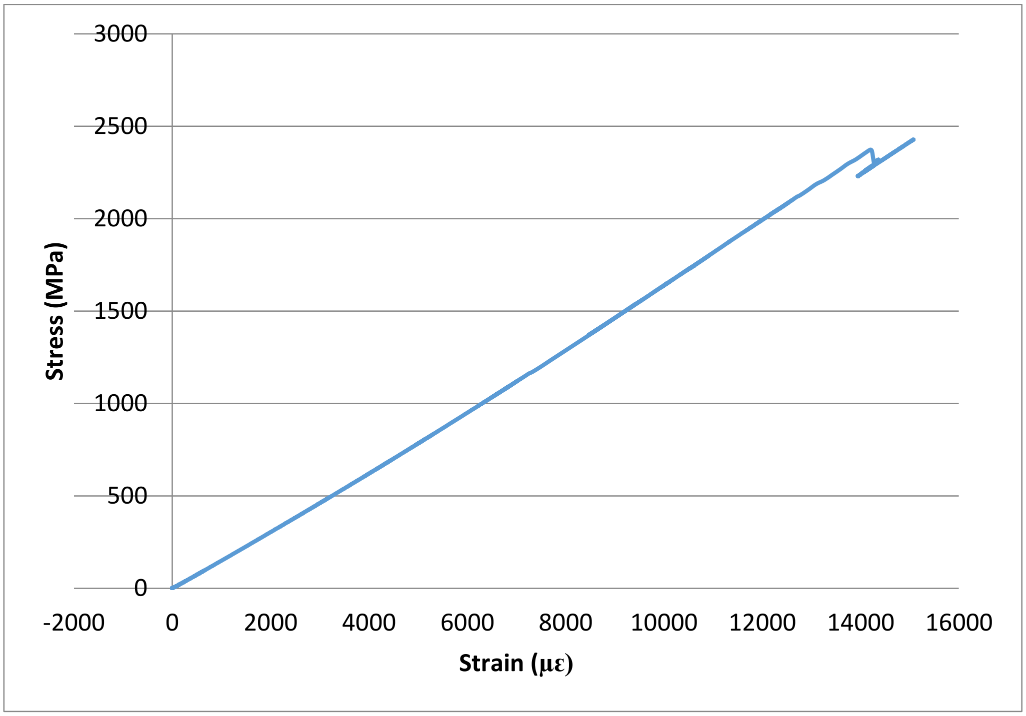


Figure 4.4 Stress vs strain curve for tensile test

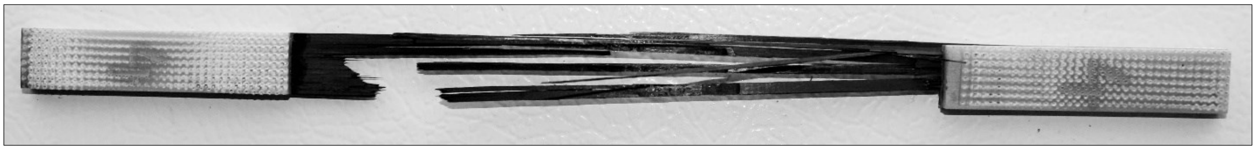


Figure 4.5 Sample after tensile test

	Tensile Modulus (GPa)			Tensile Strength (MPa)		
	Average Value	Standard Deviation	Coefficient of Variation (%)	Average Value	Standard Deviation	Coefficient of Variation (%)
AFP	165	0.23	0.14	2420	93.09	3.84
Autoclave [18]	136	3.61	2.66	2446	44.34	1.81

Table 4.1 Tensile modulus and strength of laminates made by AFP and by Autoclave.

Table 4.1 summarizes the results of tensile test coupons made by AFP. Results for samples made by compression in autoclave obtained by Cai et al.[18] are presented as reference point to compare. The average value of tensile modulus in case of laminate made by AFP is 165.15 GPa, coefficient of variation is 0.14 shows consistent and representative results. Figures 4.6 and 4.7 show the comparisons between tensile modulus and tensile strength between laminates made by AFP and by autoclave respectively. The tensile strength for laminate made by AFP is 2420 MPa, for laminate made by autoclave consolidation is 2446 MPa, the difference is 1.06% which is negligible. However, tensile modulus of laminates made by AFP increases by 22.2% from 136 GPa in case of autoclave to 165 GPa in case of AFP. The difference in modulus can be explained by the reduction of thickness by 19% of laminates made by AFP (0.033 inch) compared with those made by consolidation in autoclave (0.041 inch). The fiber volume fraction v_f of laminates made by AFP was measured using 2D microscopy analysis. Its value is 61%. Even this v_f is close to the value given by CYTEC [4], the measurement of v_f from prepreg tape using the same methods shows that v_f of the prepreg varies in a large range and has the average value of 50%. Maybe the variability in manufacturing method created this variation. Tensile strength was not increased same as tensile modulus. This is still a question but one proposed reason is because of de-bonding between fiber and matrix during cooling down process. The laminate was cooled down from melting temperature to tool temperature in a very short amount of time. The mismatch between CTE of fiber and matrix may break the bond of fiber-matrix.

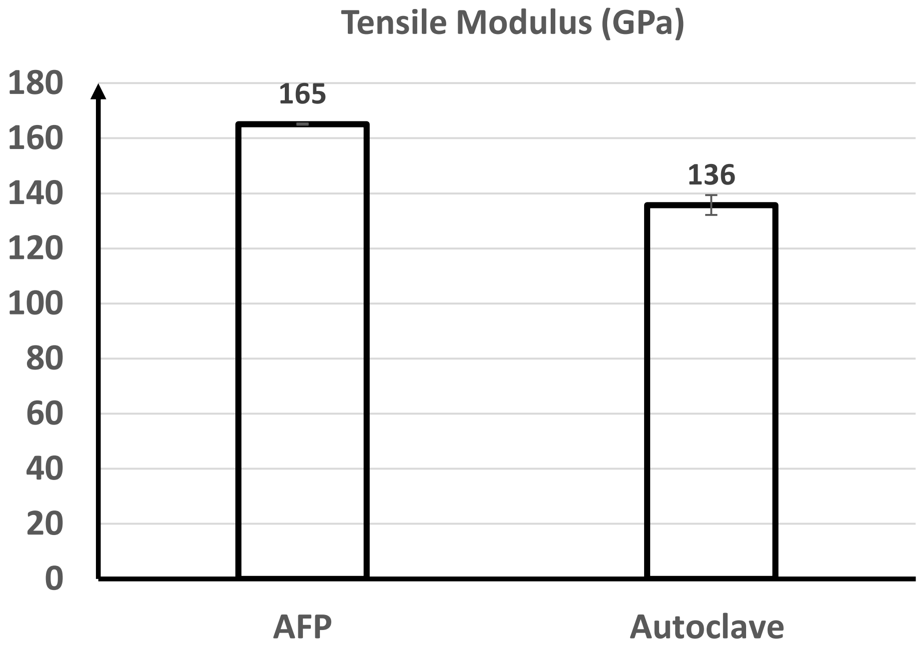


Figure 4.6 Tensile modulus comparison between AFP and Autoclave

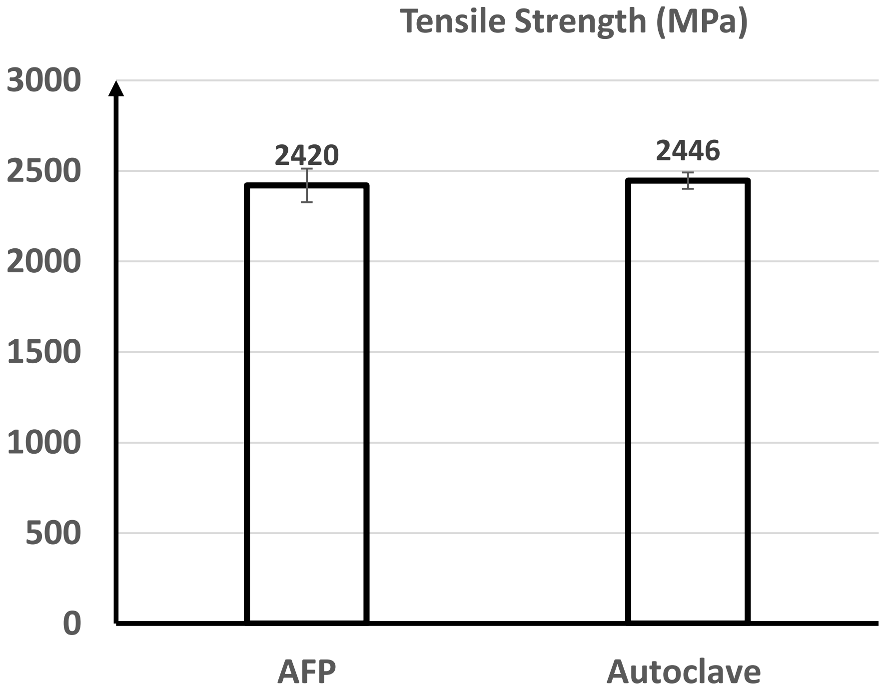


Figure 4.7 Tensile strength comparison between AFP and Autoclave

4.2.2 Compression Test

Figure 4.8 shows the Stress-Strain curve of the tensile test. Five samples were tested and sample after failure is shown in Figure 4.9. The failure mode and area is HAT. The first character represents for failure mode, H means *Through-thickness*. The second character represents for failure area, A means at *Grip/tab*. The third character represents for failure location, T means *Top.*

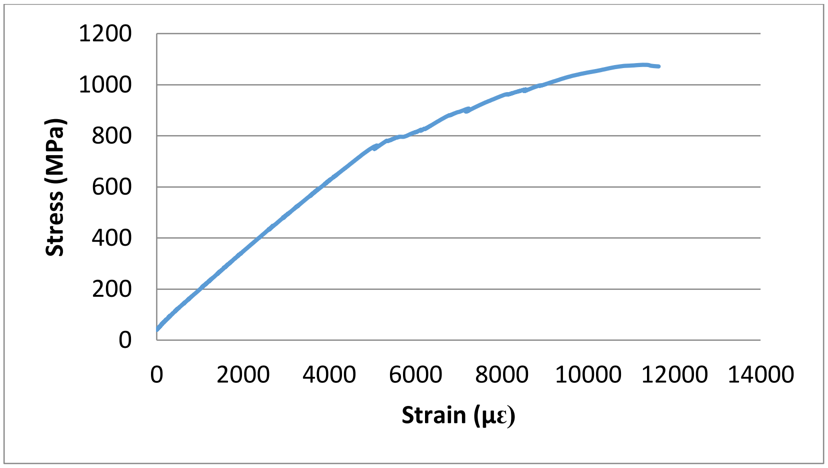


Figure 4.8 Stress vs strain curve for compression test

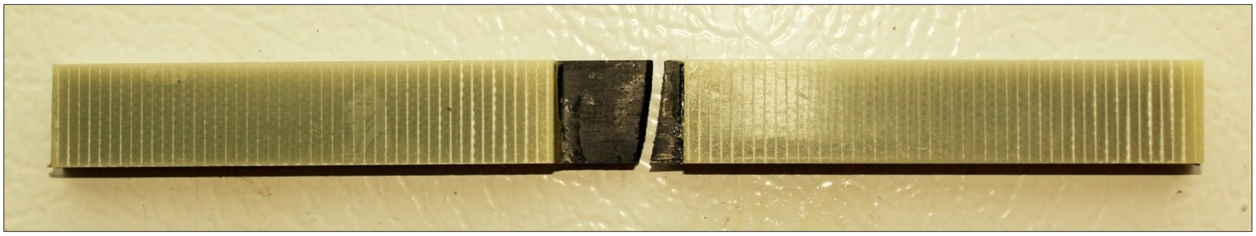


Figure 4.9 Samples after compression test

	Compressive Modulus (GPa)			Compressive Strength (MPa)		
	Average Value	Standard Deviation	Coefficient of Variation (%)	Average Value	Standard Deviation	Coefficient of Variation (%)
AFP	144	7.08	4.92	1055	37.70	3.57
Autoclave [18]	123	0.40	0.32	1407	9.44	0.67

Table 4.2 Compressive modulus and strength of laminates made by AFP and by Autoclave.

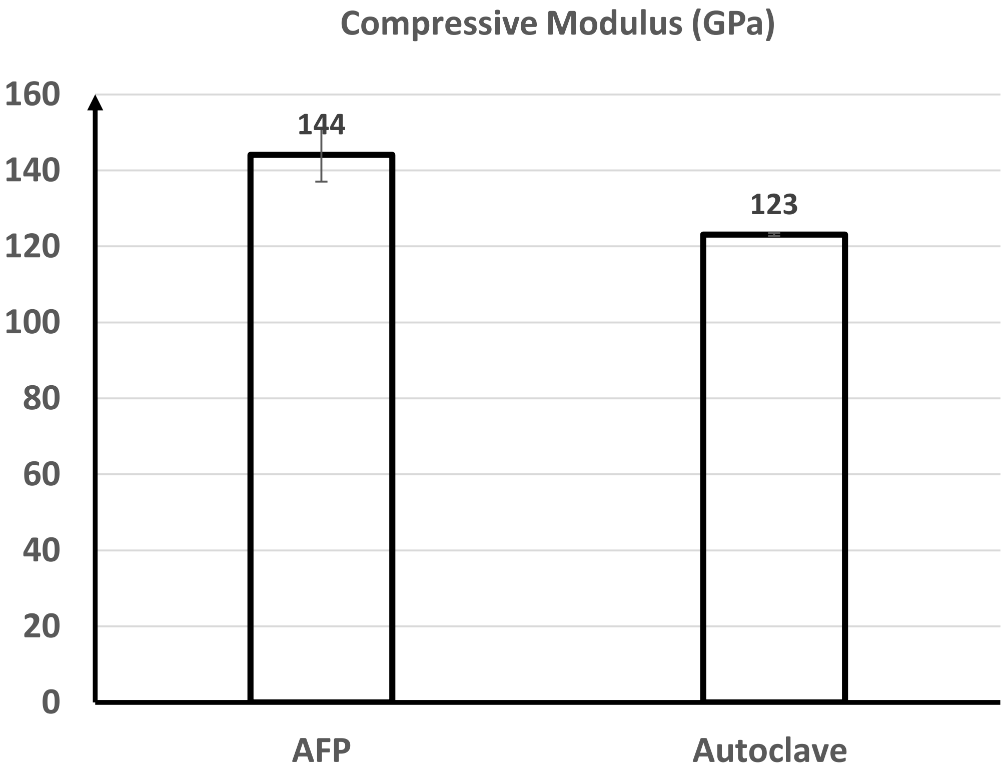


Figure 4.10 Compressive modulus comparison between AFP and Autoclave

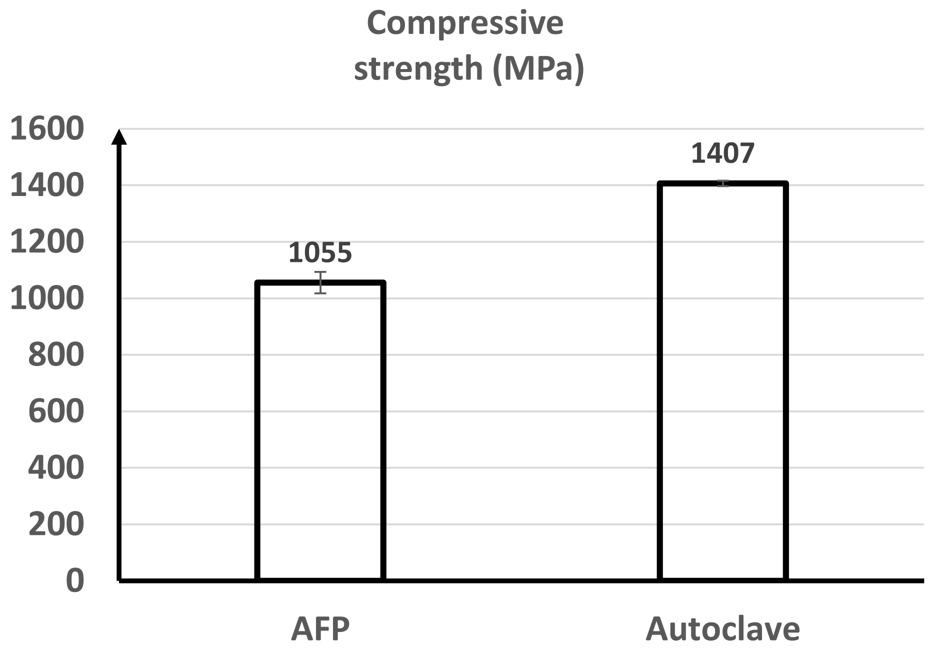


Figure 4.11 Compressive strength comparison between AFP and Autoclave

Table 4.2 summarizes results of compressive tests made by AFP. Results for autoclave obtained by Cai et al. [18] are presented as reference point to compare. The average value of compressive modulus in case of laminate made by AFP is 144 GPa. Coefficient of variation is 4.92 shows consistent and representative results. Figures 4.10 and 4.11 show the comparisons between compressive modulus and compressive strength between laminates made by AFP and by Autoclave respectively. The compressive strength for laminate made by AFP (1055 MPa) is lower by 25% than laminate made by autoclave (1407 MPa). However, compressive modulus of laminates made by AFP increases for 17% from 123 GPa in case of autoclave to 144 GPa in case of AFP. Increase in compression modulus can be explained by reduction of thickness and thus increasing in fiber volume fraction v_f. The fiber volume fraction v_f of laminates made by AFP was measure using 2D microscopy analysis. Its value is 61%. Even this v_f is close to the value given by CYTEC [4], the measurement of v_f from prepreg tape using the same methods shows that v_f of the prepreg varies in a large range and has the average value of 50%. Maybe the variability in manufacturing method created this variation. Compression strength was decreased by 25%. One reason can be the higher waviness of fiber in laminates made by AFP. Figures 4.12 and 4.13 shows the microscopic image for unidirectional laminate made by AFP and laminate made by autoclave consolidation. It is noticed that laminate made by AFP has more wavy fibers than that made by autoclave consolidation. This leads to the reduction in compressive strength.

Figure 4.12 Microscopic image along fiber direction of $[0]_{12}$ laminate made by AFP

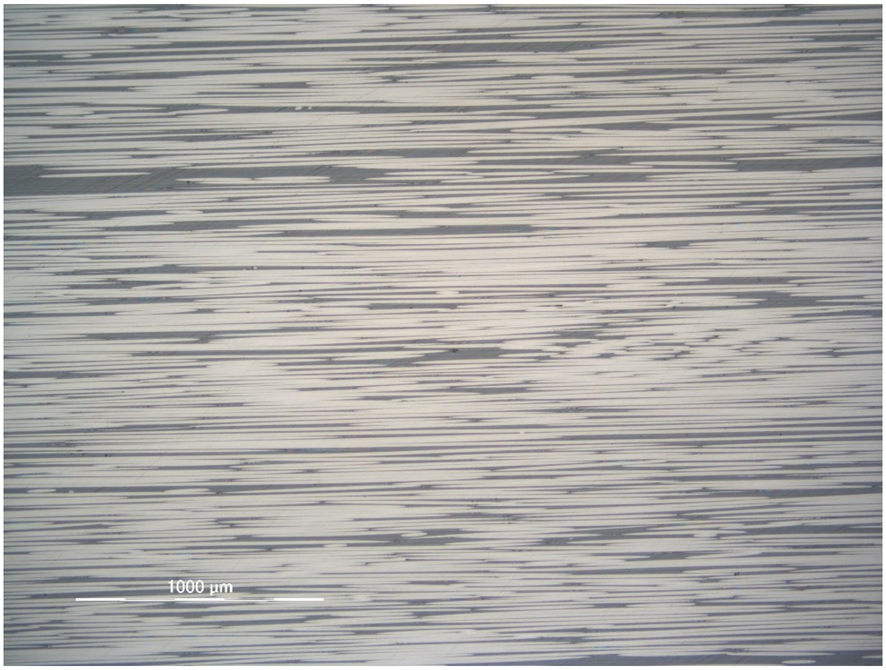


Figure 4.13 Microscopic image along fiber direction of laminate made by autoclave

4.2.3 In-plane shear Test

Figure 4.14 shows the Stress-Strain curve of the tensile test. Five samples were tested and sample after failure is shown in Figure 4.15.

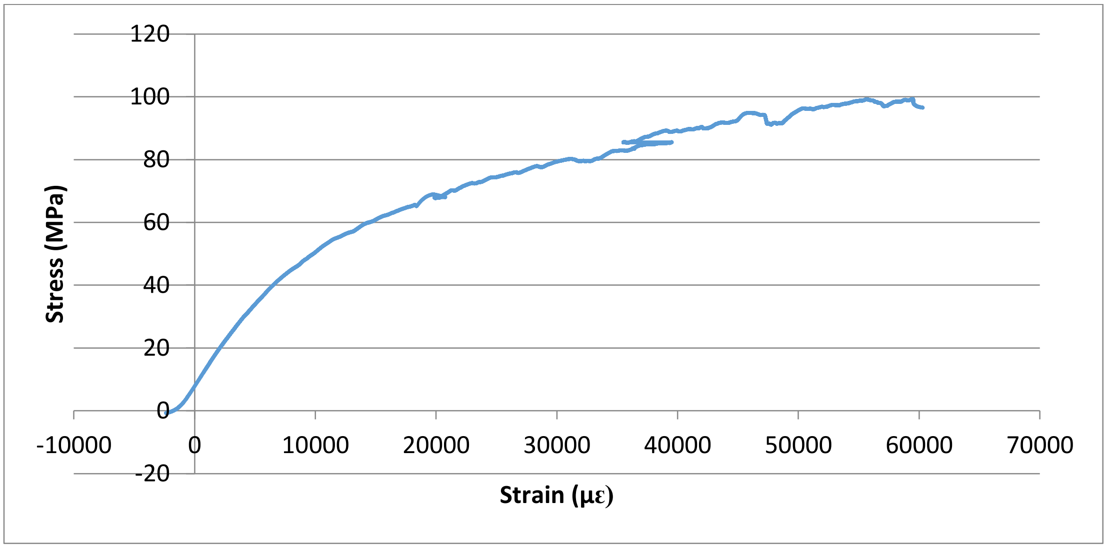


Figure 4.14 Stress vs strain curve for shear test

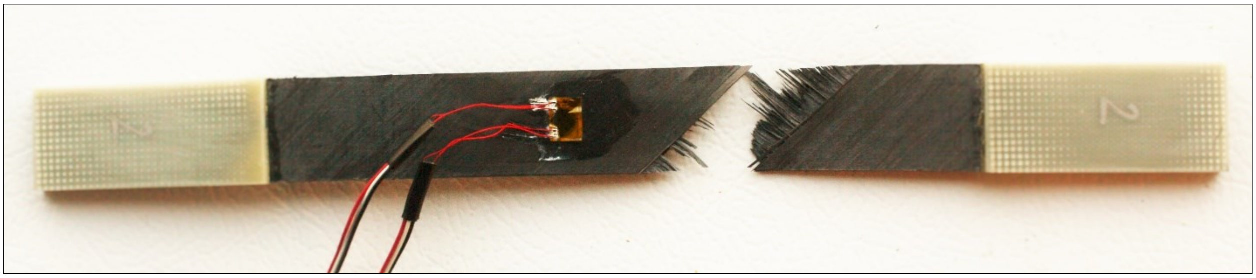


Figure 4.15 Samples after in-plane shear test

	Shear Modulus (GPa)			Shear Strength (MPa)		
	Average Value	Standard Deviation	Coefficient of Variation (%)	Average Value	Standard Deviation	Coefficient of Variation (%)
AFP	5.3	0.28	5.28	101	4.59	4.54
Autoclave [18]	5.5	0.20	3.64	80	1.27	1.59

Table 4.3 Shear modulus and Shear Strength for laminates made by AFP and by autoclave

Table 4.3 shows shear modulus and shear strength of laminate made by AFP. The average value of shear modulus in case of laminate made by AFP is 5.28 GPa. Coefficient of variation is 5.28 shows consistent and representative results. Figures 4.16 and 4.17 show the comparison between shear modulus and shear strength between laminates made by AFP and by Autoclave respectively. The shear modulus for laminates made by AFP (2420 MPa) is slightly lower than laminate made by autoclave (2446 MPa), the difference is 1.06%. Shear strength of laminates made by AFP is 101 MPa which is 20% higher than shear strength of laminates made by autoclave (80 MPa). However, shear strength of laminates made by autoclave was obtained with ASTM D5379[18] test while laminates made by AFP were subjected to ASTM D3518 test. The difference of test conditions may create the different results.

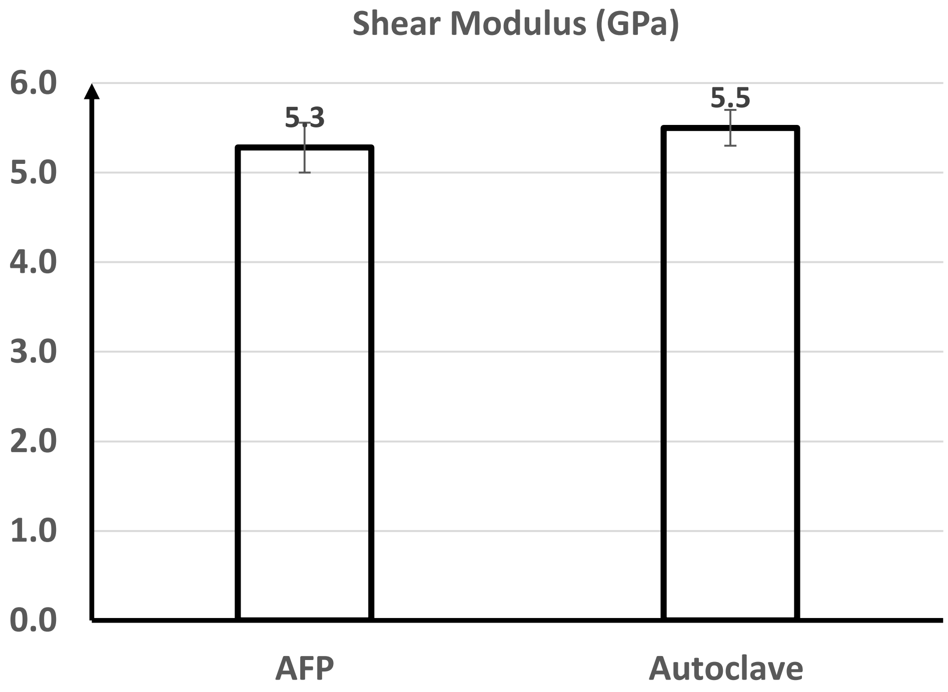


Figure 4.16 Shear modulus comparison between AFP and Autoclave

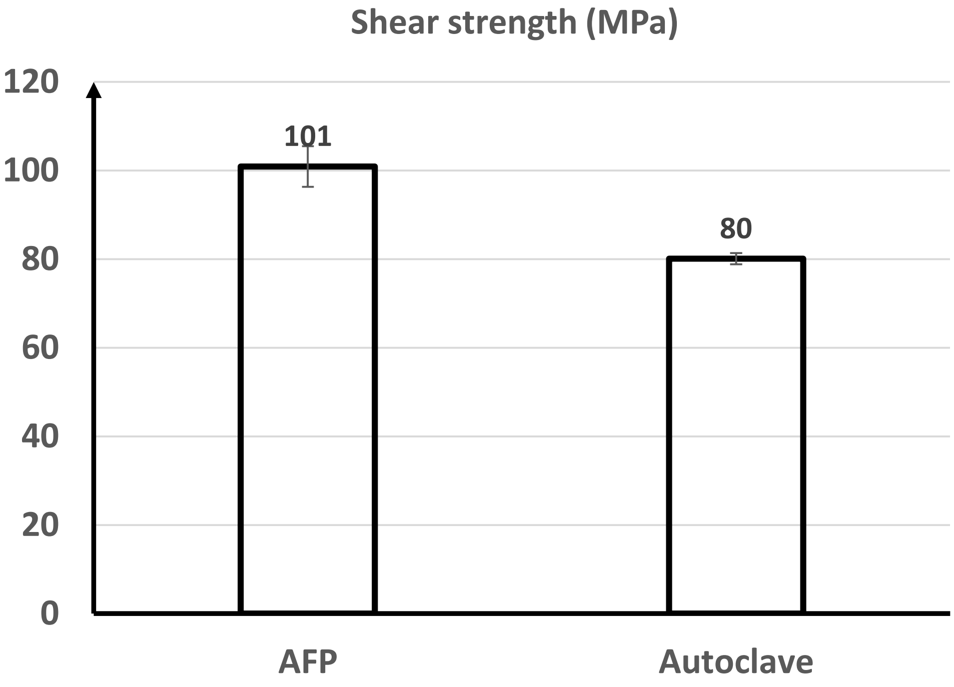


Figure 4.17 Shear strength comparison between AFP and Autoclave

4.3 Discussion

From experimental results of tensile, compression and in-plane shear test, it is noted that tensile and shear properties of laminates made by AFP are better than those of laminates made by autoclave. In more detail, tensile modulus in case of AFP is 22.2% higher than in case of autoclave while tensile strength is almost the same. Shear modulus shows 4.16% reduction and. Compressive modulus of laminate made by AFP is 17% higher than that by autoclave. However, compressive strength reduced for 25%. The improvement in tensile modulus and compression modulus can be explained by the reduction of thickness of laminates manufactured by AFP compared to those made by autoclave consolidation. Compaction force is 100 lbf while contact area between roller and substrate is very small so that applied pressure on to substrate is even higher than in autoclave.

Tables 4.4 and 4.5 show the work done by Lamontia et al. [28, 30], Comer et al. [29]. Lamontia (from Accudyne) used a complex the thermoplastic deposition head as described in Chapter 2. The heating system uses nitrogen gas. In OHC test, lay-down speed was not mentioned but the maximum capacity of lay-down speed for their machine is 3.28 in/s (5mpm) [28]. They obtained 84% of autoclave level for OHC strength and 100% for flexural strength. Comer et al. [29] used the same thermoplastic resin PEEK but different type of carbon fiber. The lay-down speed is fast (7.87 in/s), compared to 1 in/s in this study. Laser system was used as a heat source. They obtained 78% of autoclave level for OHC strength and 70% for flexural strength. Moreover, the obtained laminates shows high void fraction (3%). OHC modulus and flexural modulus were obtained from stress-strain curves, it is clearly seen that both OHC modulus and flexural modulus of laminates made by AFP are lower than those of laminates made by autoclave. The high speed of material deposition may be the reason of high void fraction so decreases these properties.

Material	Autoclave		ATP/AFP				Translation
	Modulus (GPa)	Strength (MPa)	Heat source	Lay-down speed (in/s)	Modulus (GPa)	Strength (MPa)	
AS4/PEEK[28]	-	327	Gas	-	44.8	276	84%
IM7/PEEK[29]	-	316	Laser	7.87	-	248	78%

Table 4.4 Open-hole-compression (OHC) properties of laminate made by autoclave and by AFP

Material	Autoclave	ATP/AFP			Translation
	Strength (MPa)	Heat source	Lay-down speed (in/s)	Strength (MPa)	
AS4/PEEK[30]	~1500	Gas	1.18	>1500	100%
IM7/PEEK [29]	1775	Laser	7.87	1207	70%

Table 4.5 Flexural properties of laminate made by autoclave and by AFP

CHAPTER 5 Conclusion

A successful procedure for making flat thermoplastic composite laminates with different layup sequences and different thicknesses by AFP heated by a hot gas torch and simple tape deposition head was developed.

A new approach to manufacture flat laminates was proposed. Heated mandrel was used to eliminate residual stress by keeping the whole laminate above T_g during manufacturing process. Details of the procedures are:

- For 8 layers and 12 layers unidirectional laminates, the same parameters can be used: HGT is 825^0C, compaction force is 100 lbf, steel mandrel temperature is 200^0C, and layup speed is 0.5 in /s for 1st ply and is 1 in /s for the rest. The difference is for 12 layers unidirectional laminates, starting from 9th ply, compressed air was used to cool down the substrate. The reason is if no cooling air applied to the substrate, its temperature will increase when laying down the following plies. High temperature makes the substrate become too soft and cannot sustain compaction and pushing forces from roller. After finishing lay-down process, laminates are left for natural cooling by turning off the heater.
- For 8 layers $[45^0/-45]_{2s}$ laminates, cooling the substrate surface during manufacturing is necessary. The effect of ploughing material when substrate temperature is too high is clearer in this case. It can be explained by the relative lay-down direction of the incoming tape with fiber orientation of previously laid adjacent ply. In unidirectional laminates, two adjacent plies have the same fiber orientation so when manufacturing ply n^{th}, roller moves in the same direction as fiber orientation of ply $n-1^{th}$. It creates a tension inside fibers. However, in case of $[45^0/-45]_{2s}$ laminate, except between plies 4th and 5th, others two adjacent plies have different fiber orientations. Fibers are being placed laterally by roller during deposition. Therefore, when laying down ply n^{th} $(n \neq 5)$, roller has to move perpendicular to fiber direction. It creates a force pushing fibers away from their previous position so damage the substrate. Manufacturing parameters for $[45^0/-45]_{2s}$ laminate are: HGT is 825^0C, compaction force is 100 lbf, steel mandrel temperature is 200^0C for 1st and 2nd plies, and then it is reduced to 155^0C for plies 3rd to

8th, and layup speed is 0.5 in /s for 1st ply and is 1 in /s for the ply two to eight. After finishing lay-down process, a caul plate was placed on top of the laminate and it was clamped to the bottom plate by C-clamps. The assembly of two plates with the laminate in the middle was then naturally cooled down by turning off the heater. The hot plate on top of the laminate helps to have a balance cooling on two sides of the laminate, and prevents thermal residual stresses during cooling.

Crystallinity of obtained laminates was examined by DSC and results show thermoplastic resin PEEK reached its maximum level of crystallization.

Repass is suggested to reduce void in the laminate. Without repass, interply void appeared starting from the 6th ply in case of unidirectional laminates, and starting from the 2nd ply in case of $[\pm 45^0]_{2s}$ laminate. After applying repass, there is almost no interply void in all cases. Furthermore, thickness of each layer is more uniform.

Secondly, thermoplastic laminates made by AFP with proposed parameters were characterized. Tensile, compression and in-plane shear tests were done and mechanical properties such as tensile strength and modulus, compression strength and modulus and shear strength and modulus were found. Results were then compared with those made by autoclave consolidation.

- Tensile modulus is higher by 22%, tensile strength is at the same level.
- Compression modulus is higher by 17%, compression strength reduces for 25%
- Shear modulus is at the same level. Shear strength increases by 20% but the two test are different so it is not a fair comparison.

The improvement in tensile modulus and compression modulus was explained by the reduction of thickness of laminates made by AFP compared to those made by autoclave consolidation. Furthermore, examining prepreg tape and laminates made by AFP shows an increment by11% of fiber volume fraction in laminates made by AFP.

Reduction of compressive strength needs further study. One proposed reason is due to more fiber waviness in laminate made by AFP. Fibers in laminate made by autoclave consolidation are straighter than those so it leads to higher compressive strength.

CHAPTER 6 Future work

This research proposed a manufacturing method to make flat thermoplastic laminates made by AFP with different laminate configurations. This can be done heated mandrel to keep the whole laminate above glass transition temperature during manufacturing. The future work should focus on optimizing process parameters. Secondly, obtained laminates still have a knock-off of 25% in compressive strength compared with those made by autoclave, it is required to find a solution to improve that property. Another study needs to be done is improvement of layup speed. In this study, maximum layup speed is 1 in/s. In order to facilitate the manufacturing thermoplastic laminate by AFP, it is better to layup at higher speed without decreasing quality of the laminate. Finally, some others tests should be performed to obtain others mechanical properties of laminates made by AFP.

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