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POLYMER INFILTRATION STUDIES

By

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POLYMER INFILTERATION STUDIES

Summary

Significant progress has been made on the preparation of carbon fiber composities using advanced polymer resins during the past three months. The results are set forth in recent reports and publications, and will be presented at forthcoming national and international meetings.

Current and ongoing research activities reported herein include:

- Prepregger hot sled operation
- Ribbonizing Powder-Impregenated Towpreg
- Textile Composites from Powder-Coated Towpreg: Role of Bulk Factor
- Powder Curtain Prepreg Process

During the coming months research will be directed toward further development of the new powder curtain pregregging method and on ways to customize dry powder towpreg for textile and robotic applications in aircraft part fabrication.

Studies of multi-tow powder prepregging and ribbon preparation will be conducted in conjunction with continued development of prepregging technology and the various aspects of composite part fabrication using customized towpreg. Also, during the period ahead work will continue on the analysis of the performance of the new solution prepregger.

Polymer Infilteration Studies

Polymer infilteration investigations are directed toward development of methods by which to produce advanced composite material for automated part fabrication utilizing textile and robotic technology in the manufacture of subsonic and supersonic aircraft. This object is to achieved through research investigations at NASA Langley Research Center and by stimulating technology transfer between contract researchers and the aircraft industry.

The powder curtain prepregging system, which was started up successfully last year has been used to produce over two hundred pounds of towpreg. It is currently undergoing modifications. The automated powder return system was contructed and is undergoing tests. Modification are being made to the powder curtain tube, for extended curtain width demonstration. These changes should provide better operating control over fugitive powder and improved towpreg quality control.

Issues in the use of powder coated towpreg for textile applications have been the subject of significant effort. Studies of ways to debulk powder preforms are being conducted, see attachments. An outline of planned textile composites research has been prepared to guide activities during the next two years, see attachments.

Consideration of the ways to customized towpreg for use in automated tow /fiber placement has resulted in several new approaches and will be the subject of a paper to be presented at the SAMPE. Noteworthy among the ideas that have been developed is the potential benefits from use of non-rectangular ribbon, and the thermal wave bonding model of tow placement with on-the-fly-cure. As described in the attached memoranda, several efforts to produce quality towpreg ribbon are underway. In addition to die forming methods, it is planned to investigate making unitape from powdered tow which can then be slit into the desired ribbon geometry.

The following attachments provide detailed information about several current and planned research projects.

Attachments

1. New NASA LaRC Multi-purpose Prepregging Unit.

2. Processing, Properties and Applications of Composites Using Powder-Coater Epoxy Towpreg Technology.

3. Ceramic Preconsolidation Tool for Thermoplastic Powder Towpreg.

4. Composite Part Fabrication.

5. Multi-tow Ribbon for ATP.

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A New NASA LaRC Multi-purpose Prepregging Unit

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ABSTRACT

A multi-purpose prepregging machine has been designed and built for NASA Langley Research Center. The machine has numerous advantages over existing units due to its various modular components. Each of these can be used individually or simultaneously depending on the required prepregging method.

A reverse roll coater provides the ability to prepare thin films from typical hot-melt thermoset formulations. Also, if necessary, the design allows direct fiber impregnation within the reverse roll coater gap. Included in the impregnation module is a solution dip tank allowing the fabrication of thermoplastic prepregs from solution. The proceeding modules within the unit consist of four nip stations, two hot-plates, a hot-sled option and a high temperature oven. This paper describes the advantages of such a modular construction and discusses the various processing combinations available to the prepregger.

A variety of high performance prepreg material systems were produced on IM7 (Hercules) carbon fiber. These included LaRC™ RP46, a PMR-type resin processed from methanol and two polyamide acids, LaRC™ IA and LaRC™ ITPI, prepregged from N-methyl pyrrolidinone (NMP). Parameters involved in the production of these prepreg materials are presented as are the mechanical properties of the resulting good quality laminates. A brief introduction into the existing prepregging science is presented. Topics relating to solution prepregging are identified with a focus on the current research effort and its future development.

Key Words: Prepregs, Equipment and Machinery

1.0 INTRODUCTION

As part of a polymer composite materials research program, a multi-purpose prepregging machine has been designed and built for NASA Langley Research Center (LaRC). The machine is capable of impregnating continuous fibers with high performance polymeric resins and has a number of advantages over existing units due to its various modular components. Each of the modules can be used individually or simultaneously depending on the required prepregging method.

This paper describes the advantages of such a modular construction and discusses the various processing combinations available to the prepregger. An analysis of the design and operating features of the prepreg tape machine was conducted to provide insight into the full utilization of the machine (1).

2.0 NASA LaRC Multipurpose Prepregger

The prepregger consists of various modules and is shown schematically in Figure 1. These modules are combined in different configurations depending on the method of impregnation.

2.1 Modules Spools of fiber tow are placed on the creel and each tow is threaded into the machine. The creel is split into two sides, each side consisting of five rows of ten fiber spools. Located in front of the fiber creels are a series of horizontal bars. When each tow from its corresponding row has been threaded over each bar on either side of the creel, the comb is threaded in a systematic manner (1) to prevent tow crossing and fiber damage.

The reverse roll coater module is traditionally used to prepare films onto release paper from hot-melt resin systems (Figure 2). The resulting films are brought into contact with the dry fiber web at the first nip roller station where the resin is released from the paper and impregnated into the web (Figure 3). The filming process is controlled by the gap dimensions at the coater (1). Resin is sheared onto the paper surfaces as the paper passes between the applicator and metering roll as illustrated in Figure 2. The rollers may be heated to control resin viscosity; also, if desired the dry fiber web can be passed through the roller gap and impregnation can be performed at this position.

Two dip tanks are available for running a solution impregnation operation. One tank is 38.1cm (15 inches) long and is used for prepreg tape 30.5cm (12 inches) wide. The tank has a maximum capacity of 3.3 liters and minimum capacity of 0.86 liters. The second is 12.7cm (5 inches) wide and was specifically designed for prepreg tape up to 7.62cm (3 inches) wide utilizing polymer solutions in research quantities. The maximum and minimum quantities for this second tank are 1.5 and 0.32 liters respectively. The dip tank assembly consists of three subassemblies. The first is the tank carriage and guides. The dip tank raises up to the fiber path which is pre-threaded for impregnation. The second subassembly consists of the dip pans and submerged impregnation bars hung from two steel rods over which the dry fiber web is passed. The impregnation bars are split in two and provide the prepregger with the option of passing the fiber web over two tiers, or kept as one unit and passed over the bottom tier. A heating unit is located underneath the dip tanks to provide viscosity control. By increasing the temperature, the viscosity may be lowered while maintaining a constant weight-percent loading of solids in the solution. The third subassembly is the metering rod assembly. The gap at the metering rods controls the prepreg resin content and is the most important part of the solution coating process. Resin content may be controlled by adjusting both the metering bar gap and/or the pressure. The gap is adjusted using wedge blocks that move inward to close the bars together. An adjustment screw is turned clockwise to adjust the metering gap and feeler gages are used for monitoring the gap dimensions.

The first nip station is used in conventional prepregging but it is not used in solution coating since the wet web is not amenable to nipping until initial drying has taken place in the first hot-plate. This first nip station is used for the resin film transfer process to impregnate fibers with hot-melt materials that have already been pre-filmed (Figure 3). The rollers have the option of being heated, if desired; this gives control over resin viscosity of hot-melt systems.

All nip rollers in the machine provide gap dimensions that remain constant along the roller length. A tuning bar is located in front of each roller and allows each wedge block, at either side of the roller, to be individually moved while maintaining the other stationary. Moving a particular wedge forwards or backwards provides the operator with the ability to raise and lower each side of the roller. Feeler gages are used to monitor the consistency of the gap dimensions. Two locks provide access to each wedge block movement and are situated on this bar and can be individually released.

Initial solvent devolatilization is achieved with hot-plates, Figure 1. The bottom paper remains on the underside of the prepreg to protect the plates from resin build-up. At the beginning of a run, ovens and hot-plates are raised to operating temperature prior to start-up. During operation, heat from the hot-plates is taken away by solvent evaporation and convection to air and by heating the web and paper as they pass over the plate. Since the prepreg web is very thin, its capacity to carry heat is small relative to the heat required for solvent evaporation.

To achieve a desired fiber aerial weight while maintaining fiber uniformity across the width (requiring tow-tow adhesion and no splits or gaps within the web), is an important aspect of product quality. Nip stations 2 and 3 are essential in the elimination of splits and gaps formed in the prepreg web during the devolatilization process. At this primary control point, the following parameters may be adjusted to eliminate splits:

- Gap width between rollers, creating resin squeeze out and lateral movement of the fibers
- Roller temperature (effects the flow properties of the prepreg)
- Back pressure to the nip rollers (effective when the rollers ride entirely on the web surface and are not forced apart by the wedge blocks)

Other parameters that are controlled/adjusted to contribute to the elimination of gaps and splits are:

- Web speed (this effects the pressure exerted on the web and the residence time in the devolatilization units)
- Web solution concentration (a function of the hot plate temperatures and original solution concentration since the polymer concentration in the web changes the web elasticity)

The above parameters help establish the range of operating conditions under which gap free prepreg can be produced. These conditions essentially define the operating protocol for this machine. As discussed later, this protocol will also include the use of the Prepreg Flow Number, (PFN), (2-4) that provides a temperature-pressure-velocity superposition relationship for the prepregging process.

A critical unit within the prepregger is the oven since it provides control over the solvent content in the final product. Operating variables which can be changed to remove solvent are oven temperature, air flow and web speed. Hence, residence time and temperature become important parameters in the equations for heat and mass transfer that describe the solvent vapor removal in the ovens (1).

The oven is heated by hot air. Air flows counter to the direction of travel of the prepreg web. This has the effect of both heating the solution within the web to vaporize the solvent and to sweep away solvent from the prepreg surface. Solvent evaporation has a cooling effect and should be a concern during the heat transfer analysis. The mass transfer process of devolatilization consists of solvent evaporation and diffusion through the prepreg thickness to the surface and convection into the air.

Nip station 4 provides the last opportunity to nip the prepreg web and to remove the remaining gaps and splits. The roller gap width and the roller pressure exerted on the web are the major parameters that can be changed to attain such goals. The solvent content within the prepreg must be monitored and controlled prior to this nip. Hence, resin content, distribution, and fiber aerial weight may be affected depending on the resin's prior history through the other modules.

The chill plate is located between nip station 4 and pull-roll station and cools the web prior to take-up and storage. It is capable of maintaining temperatures between 5-15°C. The chill plate is raised 0.95cm (3/8 inch) above the web line to ensure that the web remains in contact with the plate. The chill plate tends to "freeze" the resin and reduce flow, therefore, any effects on resin impregnation at the pull rollers are minimal. In turn, the lack of flow which freezing imparts on the prepreg web prevents resin loss through excessive squeeze-out and adhesion to the take-up paper. The chill plate may, therefore, have some effect on the final product quality in terms of reducing damage on wrapping as the release paper is removed.

The hot-sled module is designed to provide pressure needed to achieve impregnation during processing of high viscosity polymer solutions or melts (1). The hot-sled has several functions.

- It provides through-the-thickness penetration and wetting.
- It provides sufficient pressure to the fibers to create lateral movement and fiber nesting which helps eliminate splits and gaps.
- It increases contact pressure time. This improves fiber matrix adhesion of pre-impregnated material. Such material would include polymeric powders that coalesce together more efficiently under longer contact pressure times.
- It provides an alternate approach when nip stations 2 and 3 alone are inadequate to produce good quality product.

2.2 Safety Features Emergency stop buttons are located at several positions along the machine. The pneumatic circuitry is interfaced with the electrical circuitry and can be activated by pushing an emergency stop button when hazardous conditions such as loss of air flow in the ovens, excess solvent concentration, a high temperature alarm or interruption of the light screen occur. In the event of an emergency stop condition, the following steps automatically take place.

- All nip rolls open.
- The drive roll opens.
- Backing and metering rolls open.
- Heat to hot-plates and ovens is shut off and reset to a 260°C maximum.
- Electrical power to motors shuts off.
- Motor clutches disengage.
- Rewind clutches disengage.
- All sensors and instruments remain operable and continue to monitor the situation.

The emergency stop condition remains until the emergency condition has been cleared and the reset buttons on both the control cabinet and the control console have been pressed.

Since this machine can prepreg from solution, solvent vapor concentration must be detected to ensure a safe atmosphere and to prevent the possibility of explosion. Therefore, a real-time gas detection system has been designed into the system. Samples are drawn from the three hot-plate and oven assemblies using a Venturi vacuum pump. The sample gas is detected by a combustible sensor and a signal is sent to the system module where the concentration is displayed. If the concentration of any of the channels exceeds 25% of the lower explosion limit (LEL) of the solvent being tested (when calibrated correctly), the system will shut off all the heater and motor power to the machine.

The gas detection system is a back-up for the air flow control system in the oven or hot-plate hoods. An air flow sensor is located in the duct connecting all three hoods. If the blower is not on, or fails, all heating elements turn off until the problem is corrected. As a general rule, about 283,000 liters (10,000 cubic feet) per minute of air is mixed with every 3.785 liters (1 gallon) of evaporated solvent. If the air flow through the ovens is adequate, the system is safe without the gas detection system.

The required amount of air flow is based on line speed, percent solvent in solution and resin content and needs to be calculated (1) prior to every run. The air flow in each oven is adjusted accordingly with a damper. Make-up air in the hot-plates comes from the surrounding room; oven make-up air is supplied by an inlet blower.

All sprockets and chains are open in the back of the machine to facilitate maintenance of the bearings, chain, brakes and clutches. A light curtain which operates the entire length of the machine is used to prevent contact with moving parts. If the light screen is interrupted by an object, the motors stop and will not restart until the restart button is pushed. Indicator lights located on the body of the curtain are lit when the screens are operating correctly with no obstacles breaking the beam.

The chains are protected from the front operating side by chain guards which prevent the operator from inadvertently reaching the chain. All guards can be easily removed if required.

2.3 Operating Procedures The NASA machine is capable of fabricating pre-impregnated tape from a variety of material forms by the following methods:

- Solution prepregging with a dip tank
- Solution and hot-melt prepregging at the reverse roll coater
- Prepregging pre-cast films made on- or off-line at the reverse roll coater
- Collation and fusing of pre-impregnated tows.

These methods can be combined depending on the resin properties and solvent systems. Design equations describe the necessary procedures to estimate the required gap setting (1).

2.3.1 Reverse Roll Coating The off-line film coating process is most commonly employed to doctor onto release paper a film using hot-melt thermosets. The unit, illustrated in Figure 2, uses three rollers. Resin is applied to the preset gap between the backing and metering rolls, carried through the metering gap and pressed against the release paper surface which travels on a third roller in a counter direction. A thin resin film adheres to the paper surface after the resin profile has been sheared by the opposing motion of paper and resin. This shear splitting effect must be taken into account in calculating the metering gap dimensions. Prepreg resin content is primarily controlled by the film's resin aerial weight (1).

2.3.2 Direct Impregnation An alternative to the off-line paper coating process described above involves on-line coating which is similar to hot-melt prepregging. The fiber is impregnated on-line within the gap between the metering and applicator rolls, as illustrated in Figure 4. This process is typically performed with resin solutions that possess high viscosity's and can not be impregnated by the solution dip tank process. A standing puddle of resin is maintained at the gap with dams at either end of the rollers. Top and bottom release papers are brought into the metering bar gap along with the fiber web and resin system. Impregnation takes place at this junction.

• Use of trade names or manufacturers does not constitute an official endorsement, either expressed or implied, by the National Aeronautics and Space Administration.

2.3.3 Solution Dip-Tank The solution dip-tank and metering rods are shown in Figure 5. The gap between the metering rods is adjusted to control the amount of solution added to the fiber web. Impregnation pressure is a combination of mechanical and capillary pressures. A detailed description of how the impregnation bar geometry inside the dip tank effects these parameters is discussed elsewhere (1).

3.0 Results and Discussion

3.1 Resin Systems Processed The polymer matrix systems that have been prepregged with this machine are presented in Table I. Both hot-melt and solution techniques have been employed. For the latter, both N-methyl-pyrrolidinone (NMP) and methanol were the solvents of choice. Other high performance resins such as the LaRC™ RP46 system were studied. In this case methanol was the solvent.

3.2 Operating Conditions The prepregger processing parameters set forth in Table II need to be established for each individual prepregging run. The number of variables and conditions that need to be considered illustrate the complexity of the prepregging operation.

3.3 Properties A comparison of mechanical properties was made between laminates fabricated using prepreg made on the tape machine and prepreg prepared with a drum winder. A 30% solids solution of the LaRC™ IA amide acid in NMP was utilized in both prepregging operations. The results are given in Table III where 0° flexural strength and moduli show substantial improvements when a laminate was prepared using tape machine prepreg compared to laminates manufactured with drum-wound prepreg. The results are normalized for 60% fiber volume fraction.

3.4 Prepregging Science and Technology The most critical process during prepregging is the impregnation of the dry fiber web by the resin. Depending on how the impregnation takes place, i.e., through filming techniques or via the solution impregnation method, the lateral resin impregnating flow into the fibers must be addressed in terms of (a) the mechanical pressure being applied and (b) the capillary pressure found within the web pores. Capillary pressure is created by the fiber packing geometry and the solution-fiber wetting characteristics.

Flow modeling equations (2-5) are available for analysis of the prepregging process. Darcy's law is used to describe the impregnation rate, v, in the thickness, Y, direction. It is a function of the resin viscosity, μ, fiber bed permeability, k_f, and the applied pressure, P.

$$v = \frac{k_f}{\mu\ (T)}\left(\frac{dP}{dY}\right) \qquad 1.$$

This equation for resin flow through the web is the starting point for most prepregging analysis. Equations are needed to describe both impregnation through permeation and the resistance of this impregnation due to the resin's viscous flow. Seferis (2-4) has discussed fiber impregnation by introducing the term PFN (prepreg flow number) which describes the interrelationship between temperature, pressure and production rate. It is dependent on the operating conditions of the prepregger and the geometry of a fibrous preform.

$$PFN = \frac{KP_{eff}}{\mu V Y_f} \qquad 2.$$

K = Permeability
P_{eff} = Effective pressure
μ = Viscosity
V = Production rate
Y_f = Thickness of a collimated fiber tow

Seferis utilized Darcy's law with the mass and momentum conservation relationships to develop the PFN concept and discusses how it describes the interrelationship between temperature, production rate and impregnation pressure(2-4). The PFN number is the ratio of the impregnating forces to the resistance for impregnation. A PFN greater than one implies that resin is easily impregnated into the fiber web and a PFN less than one indicates that the resistance to viscous flow is high and the prepreg may be poorly wet-out. An excessively high PFN number could be detrimental to the hot-melt prepregging process. Too much flow may create resin starved prepregs due to excessive squeeze flow of the resin. The PFN approach was used to analyze the impregnation of fiber webs with hot-melt thermoset films. The PFN approach could be extended when impregnation occurs through a solution dip tank (1), although further parameters may need to be considered. Such parameters may include the change in the viscoelastic nature of the impregnated web as solvent is removed during the drying process and also the effect of entrained air which is brought into the dip tank by dry fiber tows.

In a solvent impregnated web, which is rich in solvent as well as polymer and fiber, the web becomes more elastic as solvent is removed during the drying process. The reduction in its viscous behavior and a concomitant increase in its elastic behavior provides a prepreg web that is more difficult to process. A major problem occurs when the increase in the web's elasticity causes web splitting and creates gaps during the drying oven process. This is an area for future analytical development.

3.5 Design Correlation's Operating experience has shown that the equations based on geometry and density used to calculate the metering bar gap dimensions are insufficient (1). Often, to achieve the desired prepreg resin content, the calculated metering bar gap setting had to be significantly increased.

Two experiments were performed to study this problem. Two 30% solids solutions of LaRC™ ITPI in NMP and Amoco's Udel Polysulfone in NMP were employed. These solutions were prepregged with Hercules IM7 carbon fiber. The metering bar gap was opened at 0.001 inch increments and prepreg samples taken. Each sample was B-staged at 250°C for 2 hours. Acid digestion studies were performed to determine the resin solids content. In Figure 6 the solids content on the prepreg is plotted versus the actual metering bar gap size. A curve representing the calculated theoretical prepreg solids content at various metering bar gap dimensions is also presented. A clear discrepancy can be observed between the calculated and actual percent solids on the prepreg. One possible explanation, which deserves further study, is that the prepreg web is not fully impregnated. The large amount of air bubbles observed within the dip tank suggests that entrapped air within the impregnation zone changes the solution density, thus the geometry and density equations may not be followed. A possible starting point for further study could utilize the calculation of the Prepreg Flow Number (PFN) (2-4) for each solution prepregged within the dip tank impregnation zone.

Devolatilization of solvent is a difficult process. The temperatures required to remove high boiling point solvents may chemically advance the resin beyond an acceptable level where processability is diminished. In certain instances where polyamide acids were being used, the excessive heating may cause the imidization reaction to proceed. This may reduce the processability of the resulting polymer.

The removal of splits is a vital process in the production of quality prepreg. It has been noted that the formation of gap-free prepreg at nip stations 2 and 3 is extremely important in producing good quality product. When gap-free prepreg enters the oven, the removal of more solvent may lead to the formation of more gaps. However, gap formation could be reduced by increasing fiber aerial weight at the dip tank. This ensures that the collimated fiber tows are kept together at the hot-plate and are less likely to split when the prepreg web is dried further in the high temperature oven.

In the two experiments discussed above, volatiles can be removed quite readily by increasing the oven temperature. For example, an increase in oven temperature from 188°C (370 °F) to 221°C (430 °F) decreased the percent volatiles by approximately 6%. However, the quality of the resulting prepreg was found to deteriorate as the temperature increased and the gap formation became more pronounced.

In an attempt to remove these gaps, the width of the dry web was decreased at the point of entry into the dip tank. It was thought that the desired width could still be obtained by controlling the gap size at the second nip station. In practice, this was found difficult to achieve. A decrease in the gap width tended to squeeze out excess resin rather than increase the width of the tow. That is, resin flows in the transverse direction more readily than carbon fibers move.

It was noted that the best quality prepreg was obtained when the prepreg web exiting from the #3 nip was completely gap-free. In these cases, further solvent removal tended to contract the web and few gaps were observed.

3.6 Future Work As discussed above, further development studies are required to establish design and operating information from which good quality prepreg can be produced. Theoretical and experimental studies relating to prepreg resin content, solvent devolatiliztion and the elimination of splits and gaps in the product are needed.

4.0 REFERENCES

1. S.P. Wilkinson, J.M. Marchello, and N.J. Johnston, NASA Technical Memorandum #TM L-17233 (1993).
2. J.C. Seferis and K.J. Ahn, SAMPE International Symposium, 34, 63 (1989).
3. K.J. Ahn, "Thermosetting Matrix Based Composite Impregnation Science and Engineering," Doctoral Dissertation, (J.C. Seferis Research Advisor) Department of Chemical Engineering, University of Washington, Seattle, WA (1990).
4. M.A. Hoisington, J.C. Seferis, and D. Thompson, International SAMPE Symposium, 37, 264, (1992).
5. S. Middleman, Fundamentals of Polymer Processing, McGraw Hill Inc., New York, NY (1977).

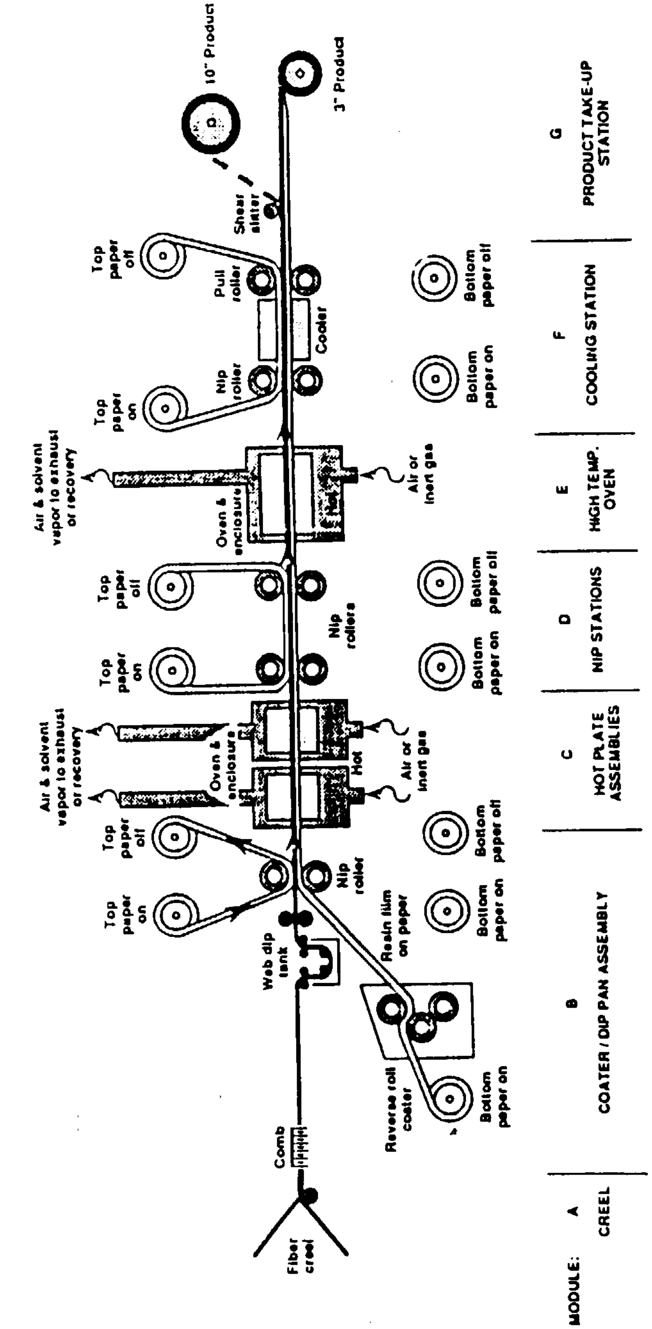

Figure 1. Schematic Diagram of the Tape Machine Modular Components.

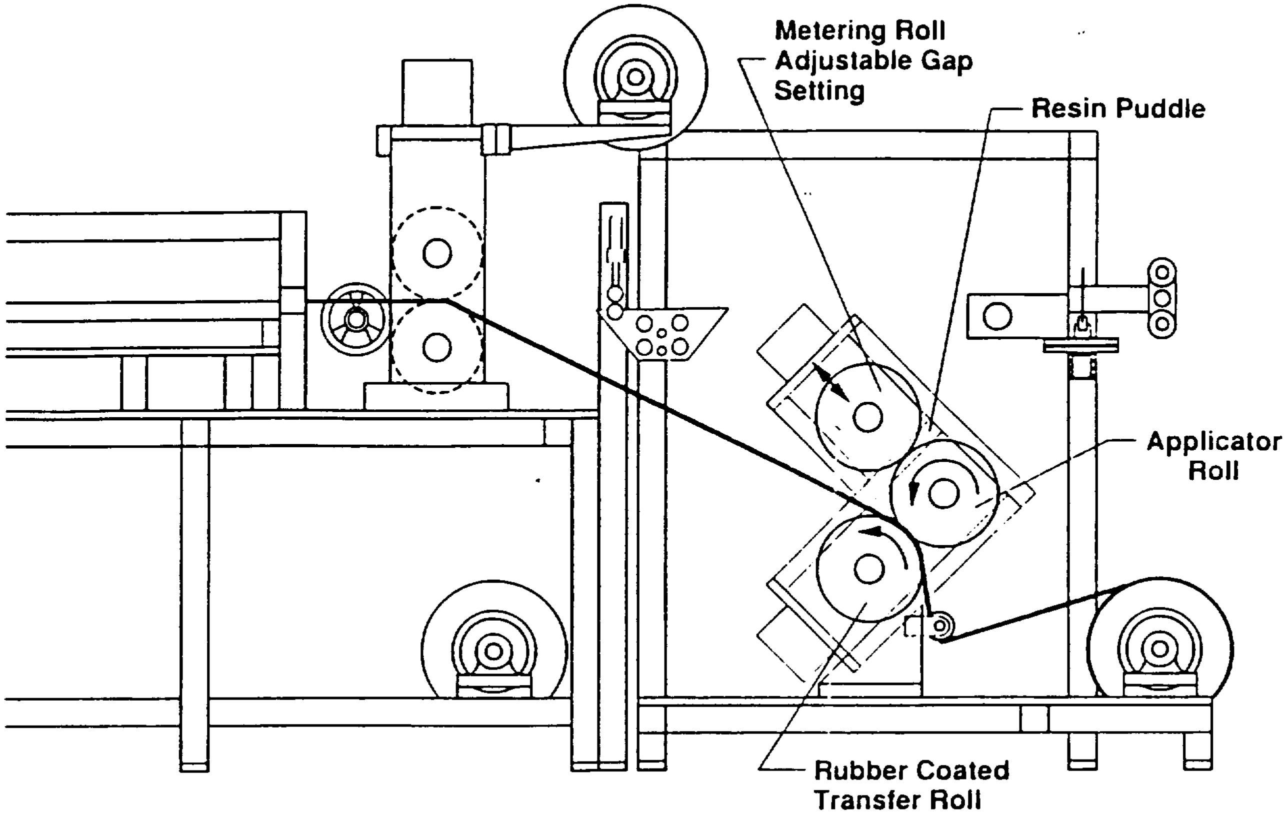

Figure 2. The Film Coating Process Using the Reverse Roll Coater.

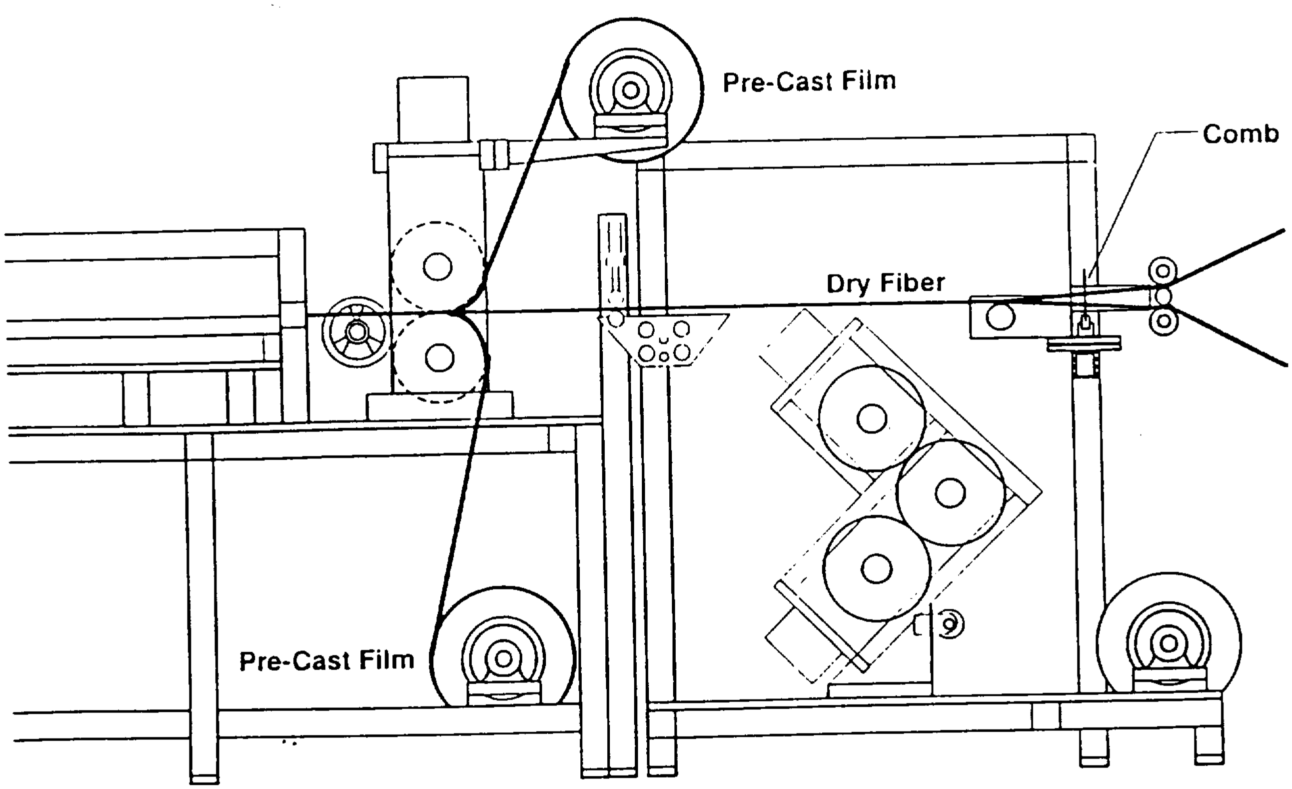

Figure 3. Common Fiber Impregnation Process Using Pre-Cast Films.

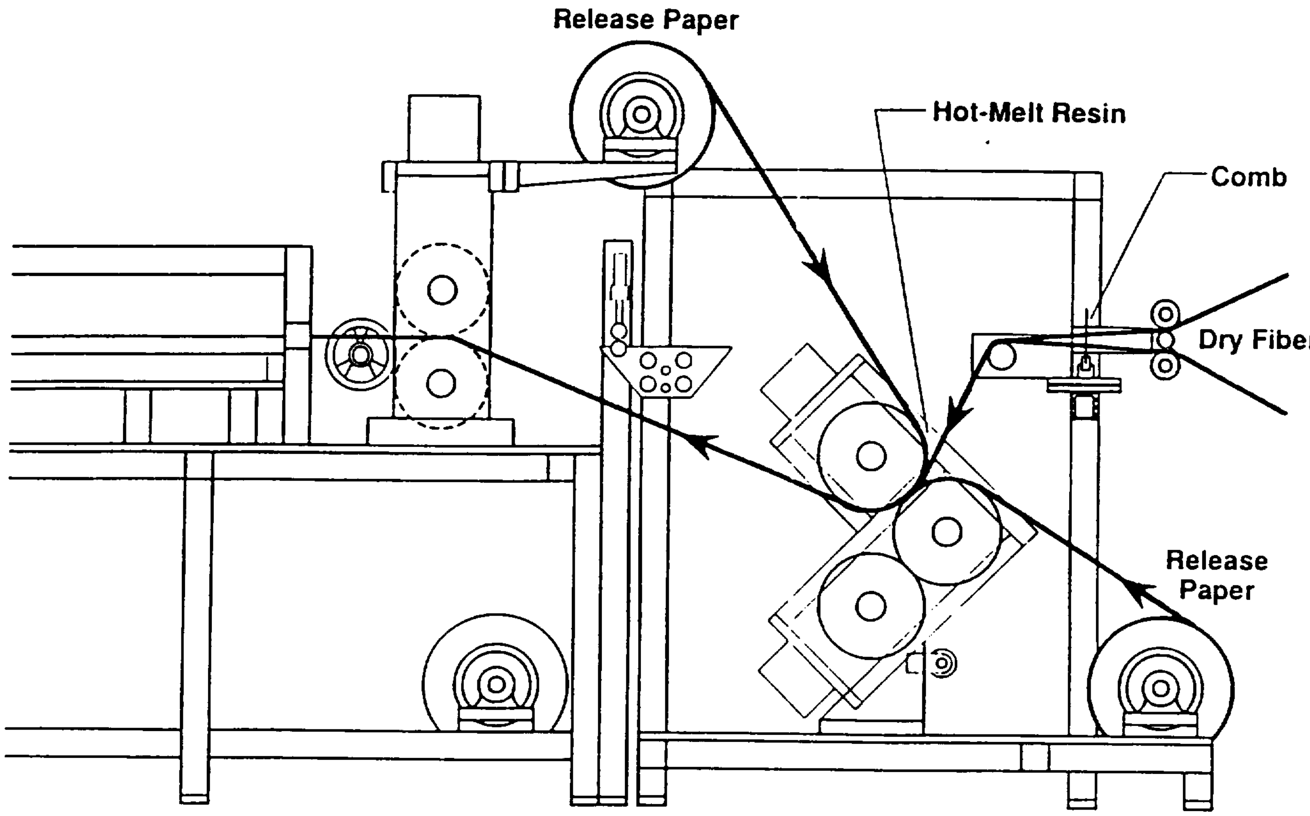

Figure 4. Direct Impregnation at the Reverse Roll Coater.

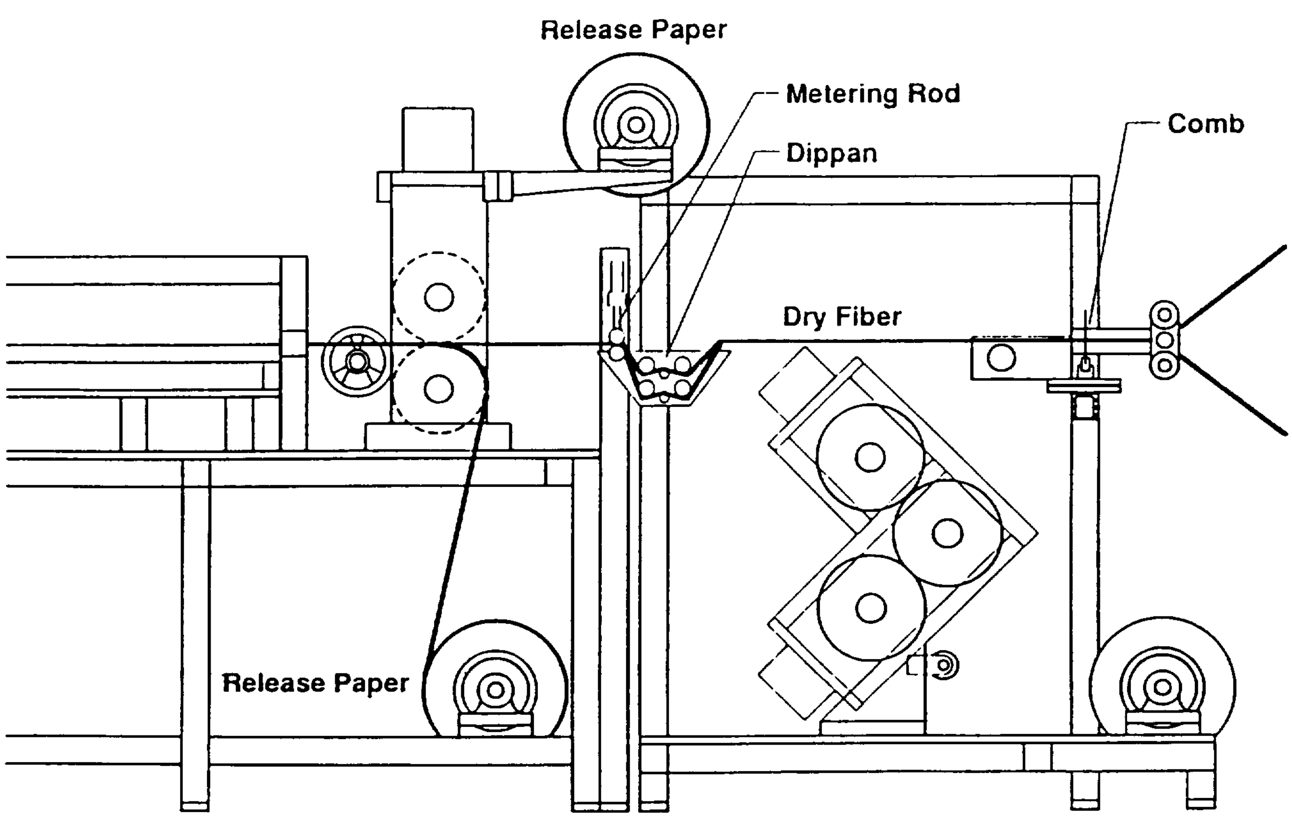

Figure 5. Solution Prepregging Using the Dip Tank Method.

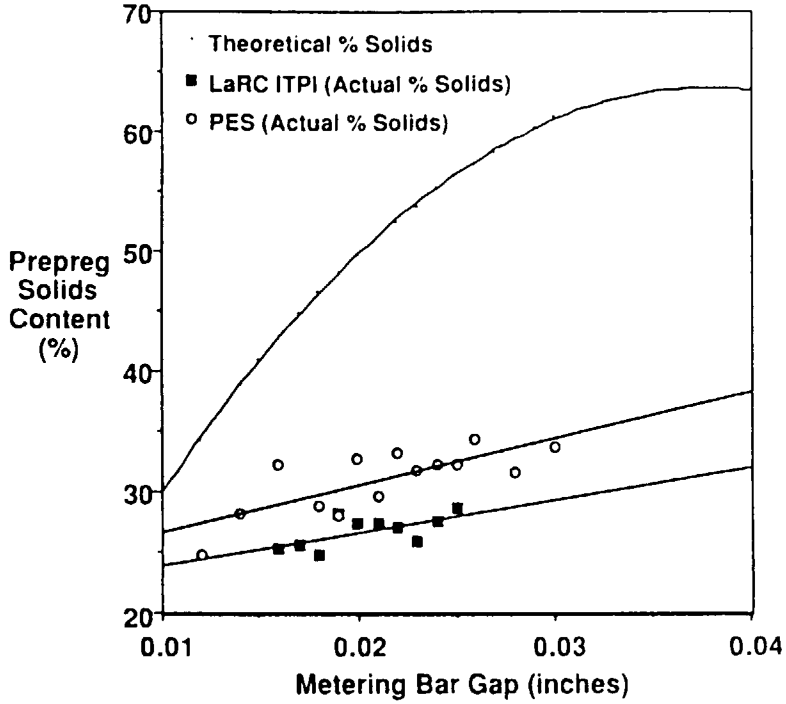

Figure 6 Actual Percent Solids of IM7/LaRC™ ITPI and IM7/Polysulfone Prepregs as a Function of Metering Bar Gap Dimensions

Table I
Material Systems Prepregged On The Tape Machine

Resin System	Solvent	% Solids in Solution
LaRC™ IA amid acid	NMP	30
LaRC™ ITPI amid acid	NMP	30
Polysulfone	NMP	30
High Mw Polyamide acid	NMP	30
LaRC™ ITPI isoimide/amide acid blend	NMP	23
LaRC™ RP46	Methanol	70

Table II
Machine Operating Parameters

Prepreg Batch Number ____
Coating Method ____
Fiber ____
Matrix ____
% Solids ____
FAW ____
Width (cm) ____
Fiber Yield (g/m^2) ____
Dry Resin Content ____
Number of Tows ____
Comb #/Angle ____
Prepreg Thickness (cm) ____
Line Speed (M/sec) ____
Dip Tank Metering Gap ____
Nip Roll Data (Temp/Pressure/Gap/Speed Ratio)
Hot Plate Temperatures (°C) ____
Oven Temperatures (°C) ____
Unwind Tension Settings ____
Rewind Pressures ____
Product Pressures ____

Table III
0° Flexural Properties for LaRC™ IA/IM7 Composites

	Tape Machine Prepreg	Drum Wound Prepreg
0° Flex Strength (MPa)		
25°C	1611±63	1474±8
177°C	1163±50	956±52
0°Flex Modulus (GPa)		
25°C	137±6	93±8
177°C	132±6	98±1

All results are normailzed for 60% fiber volume fraction.

Preliminary abstract for NASA"S Fourth Advanced Composites Technology Conference in Salt Lake City, Utah, June, 1993

Processing , Properties and Applications of Composites Using Powder-Coated Epoxy Towpreg Technology

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ABSTRACT

Composite manufacturing using the current prepregging technology of impregnating liquid resin into three-dimensionally reinforced textile preforms can be a costly and difficult operation. Alternatively, using polymer in the solid form, grinding it into a powder, and then depositing it onto a carbon fiber tow prior to making a textile preform is a viable method for the production of complex textile shapes. The powder-coated towpreg yarn is stable, needs no refrigeration, contains no solvents and is easy to process into various woven and braided preforms for later consolidation into composite structures.

NASA's Advanced Composites Technology (ACT) program has provided an avenue for developing the technology by which advanced resins and their powder-coated preforms may be used in aircraft structures. Two-dimensional braiding and weaving studies using powder-coated towpreg have been conducted to determine the effect of resin content, towpreg size and twist on textile composite properties. Studies have been made to customize the towpreg to reduce friction and bulk factor. Processing parameters have been determined for three epoxy resin systems on eight-harness satin fabric, and on more advanced 3-D preform architectures for the downselected resin system. Processing effects and the resultant mechanical properties of these textile composites will be presented and compared.

INTRODUCTION

For composite materials to be utilized as primary structures in subsonic and supersonic aircraft applications, the total production costs of the composite parts must be decreased from their present levels. Developments in the fabrication of composite parts point toward cost reduction through increased automation. In conjunction with the development of automated fabrication techniques, NASA Langley Research Center (LaRC) and BASF Structural Materials, Inc. have developed methods of prepregging carbon fiber with thermoplastic and thermosetting polymer powders.

Under NASA's Advanced Composites Technology (ACT) Contract, NAS1-18888, Lockheed Aeronautical Systems Company (LASC) is investigating the use of textile composites for primary aircraft structures. By coupling powder-coated towpreg with existing, highly automated textile processes, the resulting impregnated fabrics, broad goods and preforms can be easily molded into parts. These combined fabrication processes may be a low cost alternative to resin transfer molding (RTM) of dry preforms in cases where complex mold geometries and tightly fabricated preforms pose wet-out problems. Additionally, the physical size of aircraft primary structures makes tooling for RTM an expensive manufacturing option. The powder-coated process may offer the only viable method of part fabrication if high melt viscosity polymers are required to obtain improved composite properties, such as thermal stability and/or fracture toughness.

The following work has been conducted to generate fabrication technology for 3-D composite structures from powder coated 3-D textile preforms. Issues related to the weaving of towpreg yarns, debulking of 2-D and 3-D preforms, and processing and properties of eight-harness satin fabric flat panels have been addressed in detail. In the weaving study, the size, level of twist , and the rigidity of powder-coated tow were systematically varied to determine the effects of each process variable on resulting fabric quality. Debulking trails were performed to determine how effective various pressure/temperature combinations were on reducing bulk in as-fabricated 2-D and 3-D textile preforms. A layup and consolidation scheme was adopted which produced good quality flat laminates from 3M AMD 0036[1] epoxy powder coated preforms. Mechanical property tests were performed on these laminates to compare with identical laminates fabricated using the 3M PR-500 RTM epoxy resin system.

2-D WEAVING STUDIES

Towpreg Variables vs. Weavability

The primary objective of the 2-D weaving study was to learn how to convert powder-coated yarn into quality fabric. In earlier studies [1,2], the process of powder-coating tow and its weaving or braiding into preforms for part fabrication was found generally to be less expensive and inflicted less damage to the fibers when large (12,000 filament and more) tow bundles are used. Offsetting the advantage of using large tow bundles are factors such as potential difficulty in consolidation and possible reduction in composite properties. Yarn splitting and loose fibers on the yarn surface cause difficulties in weaving. To overcome this, yarn shaping, twisting, serving, wetting, and sizing are common practices used to reduce the separation of filaments, decrease tow-to-tow abrasion, and minimize fiber loss.

All towpreg used in the 2-D weaving portion of this study was fabricated using a dry powder process developed by NASA [3]. The dry powder prepregging process involves three steps: tow spreading, polymer deposition, and polymer fusion onto the fibers. The carbon fiber tow bundle was first pneumatically spread to approximately 3 inches in width, then impregnated with powder by means of a dry, recirculating, fluidized powder chamber. Radiant heating was used to sinter or fuse the polymer powder particles to the tow. The powder line was upgraded to speeds of 11-16 yards/min and over 21,000 yards of towpreg were produced for this study.

The effects of varying yarn bundle sizes and yarn twist on the weavability of dry polymer powder-coated fibers were studied in detail. The mechanical properties of composite made from the resultant woven cloth were determined. G30-500[1] (BASF) and AS-4[1] (Hercules) carbon fibers in tow bundles of 3k, 6k and 12k filaments were used. Each was impregnated with a thermoplastic polyimide, LARC™-TPI[1] 1500 medium flow powder (Mitsu Toatsu Chemicals). Weaving was performed on towpreg yarns that had twist levels of zero twist or 0.4 twists per inch (tpi). After establishing a weaving protocol, an experiment epoxy (3M AMD-0029[1]), was fabricated into towpreg and woven into eight-harness satin fabric.

Towpreg was woven into eight-harness satin fabric under NASA Contract NAS1-18358 by Textile Technologies, Incorporated[1] (TTI) in Hatboro, PA. The initial work was performed on yarns containing 6k filaments. During this phase, the set-up of the loom and the weaving of the towpreg were examined for ways to minimize damage imparted to the yarn. The towpreg yarn prepared at LaRC was rewound at TTI onto 40 separate spools in order to produce a balanced 4 inch wide fabric with 10 picks per inch (ppi). Two rewinding machines were used to determine how best to rewind towpreg. A rewinder that yielded a parallel winding pattern was found to cause less fiber damage than a rewinder that gave a cross winding pattern.

The spools of rewound towpreg were loaded into an Iwer 1200 rapier-type loom. Weaving powder-coated tow in a conventional rapier loom results in less fiber damage on the fill yarns, since the fill yarn is taken directly from the manufacturer's spool. Initial weaving efforts revealed problems with the surface of some of the warp towpreg yarns having loose filaments, which accumulated in the heddles and reed. To alleviate this problem, the towpreg was twisted to a carbon fiber manufacturer's standard of 0.4 tpi. Use of twisted towpreg greatly improved the weaving operation.

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[1]Use of trade names or manufacturers does not constitute an official endorsement, either expressed or implied, by the National Aeronautics and Space Administration.

An analysis is given in Table 1 of the powder-coated fabric produced for this study from both twisted and untwisted yarn. The weave counts, linear weights and fabric thicknesses are presented for 3k, 6k, and 12k towpreg made with LARC™-TPI. Noticeable fiber damage was observed in the woven material. While twisting improves weavability, the action of twisting was found to impart damage to the towpreg yarn. The method of tow twisting at LaRC was performed off-line after the prepregging had been completed, and required additional fiber handling. It is likely that improvements in the twisting equipment and on-line twisting can reduce fiber damage.

Table 1. Towpreg 8-harness Satin Fabric Parameters for 2-D Weaving Study.

Towpreg Specification	Weave Count (ppi)	Areal Weight (oz/yd^2)	Thickness (inches)
6k (G30-500) / LARC™-TPI, Untwisted	10.2 x 9.8	14.10	0.067
6k (G30-500) / LARC™-TPI, Twisted	10.2 x 9.8	14.27	0.071
6k (G30-500) / LARC™-TPI, Untwisted	10.1 x 10.0	13.22	0.077
6k (G30-500) / LARC™-TPI, Twisted	10.2 x 9.3	14.73	0.103
12k (AS-4) / LARC™-TPI, Twisted	8.2 x 8.2	23.91	0.126
3k (G30-500) / LARC™-TPI, Twisted	20.0 x 19.8	12.63	0.058

Towpreg flexural rigidly was also systematically varied since a previous study indicated that yarn flexural rigidity was an important parameter in successfully weaving towpreg [4]. Powder-coated yarn rigidity is a function of percent resin content, oven temperature, and yarn residence time in the oven. These parameters were appropriately altered to furnish the required rigidity variations. Samples were taken from each lot of towpreg yarn, and flexural rigidity was measured by ASTM method D1388-64. Towpreg flexural rigidity values utilized in this study are listed in Table II.

Towpreg Variables vs. Mechanical Properties

A mechanical testing program was developed to determine the effects of tow bundle size and twist on the mechanical properties of unidirectional and eight-harness satin fabric laminates. To investigate the effects of tow bundle size, powder-coated towpreg made from LARC™-TPI and 3k and 6k G30-500, and 12k AS-4 carbon filaments were frame-wrapped into unidirectional panels to obtain the flexural strength and modulus, and the transverse flexural strength [5].

A vacuum press was used to remove air from void spaces in the LARC™-TPI/carbon fiber specimens consolidated for the weaving study. At maximum temperature, pressure was applied at 1.5 psi/min to 60 psi in order to allow sufficient time for resin flow, adhesion, and fiber movement. The pressure ramp was followed by a hold period of one hour for final

Table 2. Flexural Rigidity Results for Powder Coated Tow and Fabrics Used in 2-D Weaving Study

Tow Description	Overhang (in)	Areal Weight (oz/yd^2)	Rigidity (oz-yd)
6k LARC™-TPI, Twisted Tow	4.0	2.44	1.54×10^{-2}
6k LARC™-TPI, Twisted Woven Cloth	3.25	13.28	4.45×10^{-2}
6k LARC™-TPI, Twisted Tow	9.0	1.66	1.17×10^{-1}
6k LARC™-TPI, Twisted Woven Cloth	7.0	12.79	4.24×10^{-1}
12k LARC™-TPI, Twisted Tow	5.5	6.15	9.87×10^{-2}
12k LARC™-TPI, Twisted Woven Cloth	4.0	25.85	1.60×10^{-1}
12k LARC™-TPI, Untwisted Tow (34.6 w/o resin)	6.75	5.94	1.76×10^{-1}
12k 3M epoxy, Untwisted Tow (32 w/o resin)	5.0	5.85	7.09×10^{-2}

consolidation and stress release at 662°F or 698°F for the unidirectional laminates and 698°F for the woven eight-harness satin prepreg cloth. The part was cooled below Tg at a rate sufficient to stop consolidation before final part thickness was reached. This avoided resin squeeze-out and resulting dry spots. The mechanical properties generated from these tests for untwisted tow are compared to the tow bundle size in Figures 1 and 2.

To determine the effects of twisted tow on mechanical properties, tests were conducted on unidirectional composites specifically, 12k carbon fiber (AS-4) towpreg of LARC™-TPI with a twist level of 0.4 tpi. Flexural strength and modulus (ASTM D790-84a) values were obtained and compared to untwisted towpreg (Table III). In addition, compression tests were performed tests were performed by the IITRI method (ASTM method D3410-87, procedure B). The values for compressive strength, modulus, and Poisson's ratio for twisted tow and untwisted tow are also listed in Table III.

The eight-harness satin fabric, woven from powder-coated towpreg, was consolidated into panels, and then cut into tension and short block compression specimens. The tension specimens were tested untabbed using hydraulic grips. A tensile load was applied only in the warp direction. Insufficient material was available for testing in the fill direction. The short block compression specimens were tested in both the warp and fill directions. Tension and compression moduli for eight-harness satin fabric composites are shown as a function of tow bundle size and twist in Figure 3.

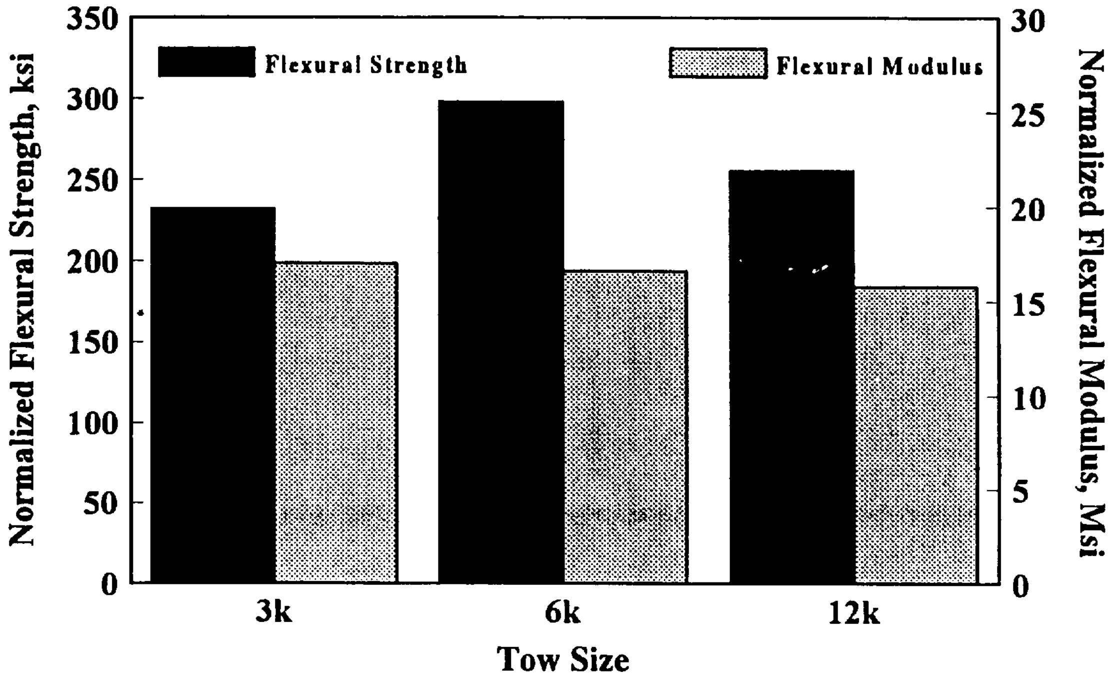


Figure 1. Flexural Properties vs. Tow Size in Unidirectional LARC™-TPI/AS-4 or G30-500 Composites (Normalized to 60 volume percent fiber)

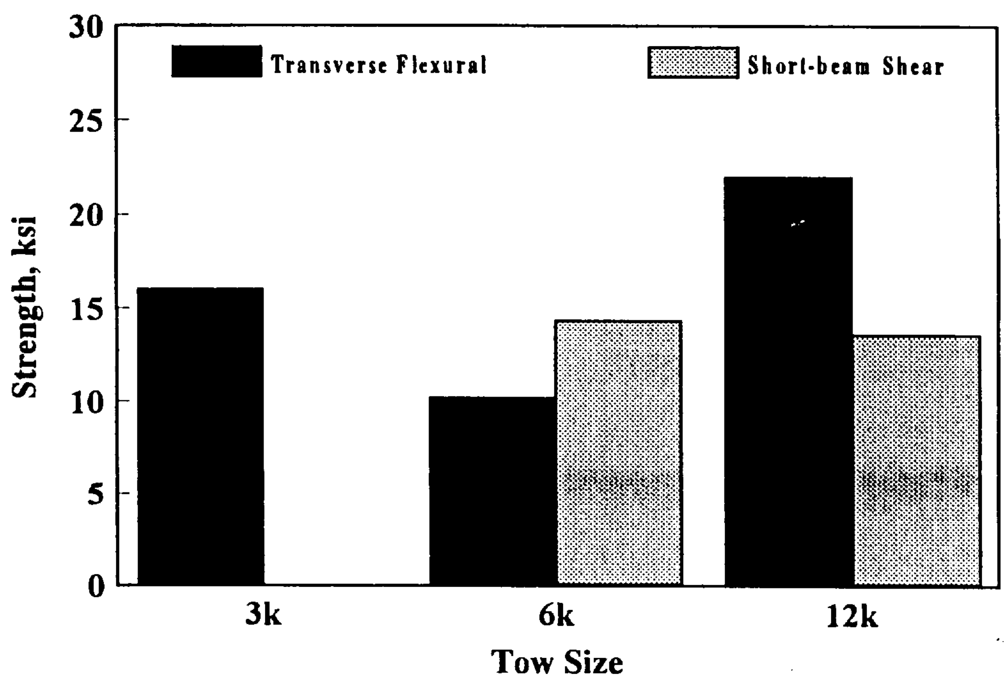


Figure 2. Mechnaical Properties vs. Tow Size in Unidirectional LARC™-TPI AS-4 or G30-500 Composites.

Table 3. Twisted and Untwisted Towpreg Mechanical Properties in Unidirectional 12k AS-4/LARC™- Thermoplastic Polyimide (TPI) Composites.

Mechanical Properties	UnTwisted Towpreg	Twisted Towpreg (0.4 tpi)
Flexural Strength[1], ksi	255.3 ± 14.1	248.4 ± 15.9
Flexural Modulus[1], Msi	15.6 ± 0.2	16.2 ± 0.4
Compression Strength, ksi	165.3 ± 1 2.2	140.4 ± 9.7
Compression Modulus, ksi	17.1 ± 0.8	15.7 ± 0.9
Poisson's Ratio[2]	0.35 ± 0.02	0.38 ± 0.03

[1] Values have been normalized for 60% fiber volume fraction.
[2] Based on IITRI compression data (by ASTM Method D3410-87, procedure B).

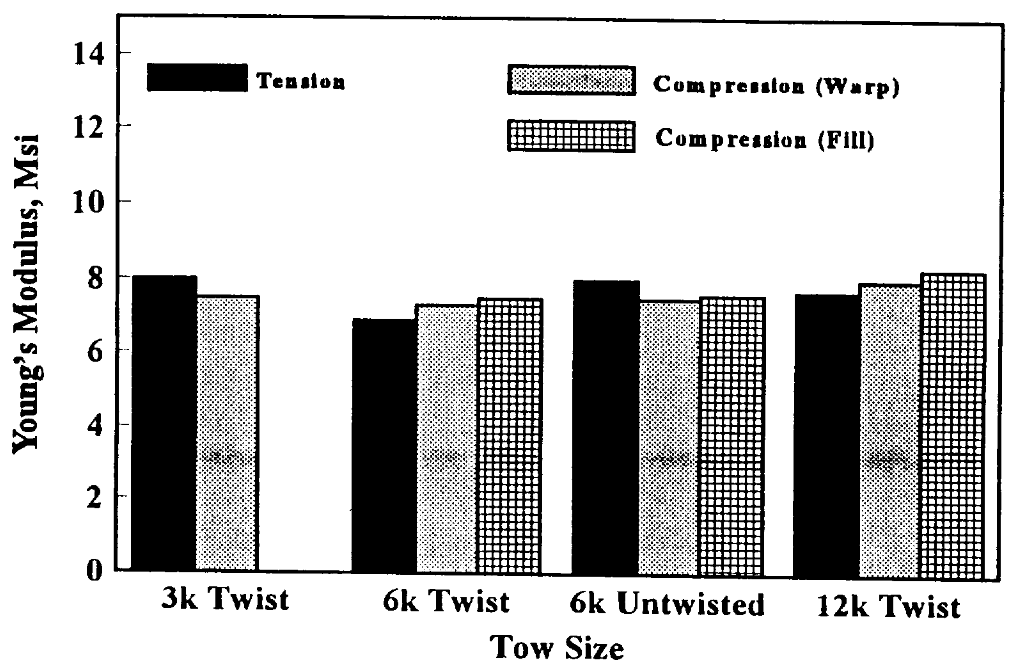


Figure 3. Young's Modulus (Short Block Compression) vs. Tow Size for 8-Harness Satin Woven LARC™-TPI/AS-4 or G30-500 Composites.

Discussion of Weaving Study Results

All weaving operations require that care be taken to minimize fiber damage. There should be as few as possible eyelets, bends and other tow touch points. Tensioning should be kept low. Rewinding and other handling activities should be minimized.

An important observation regarding weaving and tow size selection is the relation between fiber damage and tow size. During both powder prepregging and towpreg weaving, fiber damage is greater for the smaller tow bundles, because damage occurs primarily to the fibers that are at the bundle surface. For a given total amount of fiber, the use of small tows results in larger tow surface area and correspondingly higher fiber damage.

For well consolidated unidirectional laminates, the tow bundle size should have no effect on mechanical properties. No apparent pattern was found in the mechanical properties of the unidirectional laminates as a result of tow bundle size (Figures 1 and 2) with a possible exception in the transverse flexural strength values (Figure 2). Unidirectional composites made only with 6k material was consolidated at 698°F, whereas the 3k and 12k materials were consolidated at 662°F. The increase in temperature for the consolidation cycle may have resulted in an increase in consolidation due to a decrease in resin viscosity. As the processing cycles for LARC™-TPI fiber reinforced composites are improved, more mechanical property data will be generated.

In contrast to the unidirectional mechanical properties, the fabric composites were expected to exhibit increased mechanical property values with decreasing tow bundle size due to the contribution of crimp, which increases with increasing tow bundle size. The limited data obtained for composites made with eight-harness satin woven cloth (Figure 3) show no apparent effect of tow size or twist on tension and compression modulus. Because of a lack of material, each data point shown in Figure 3 presented an average value taken from testing three to five specimens. A large scatterband was observed for the strength data, consequently, more tests will be required to develop statistically significant strength values.

Mechanical properties of composites made of twisted towpreg exhibited lower properties, except flexural modulus, than specimens with untwisted tow (Table 3). At 0.4 tpi the non-alignment effect of fibers in a twisted yarn is negligible [6]. This is illustrated in the compression and flexural modulus values. Composites made with twisted towpreg had a 15 percent lower compression strength than those made with untwisted towpreg. Apparently, the additional fiber damage that resulted from the current method of twisting caused the reduction in strength. Twisting the towpreg improved weavability, since it caused the yarn to take on a cross-section that was round and compact. In order to create a suitable yarn for weaving, either the current method of twisting must be improved or the towpreg must be shaped with heated dies or rollers to achieve the same cross-section with less damage.

PROCESSING SCIENCE FOR POWDER-COATED PREFORMS

Special consideration in consolidating woven goods, as distinct from consolidating unidirectional tape, must be given to the elimination of intra- and inter-tow voids within fabric, as well as the elimination of inter-ply voids which are also of concern in conventional tape processing. The established steps [7] in the consolidation of woven materials are illustrated in Figure 4. They are: intimate contact of the polymer-polymer interface at numerous sites across the composite; followed by deformation and interdiffusion of polymer chains to cause the interface to disappear.

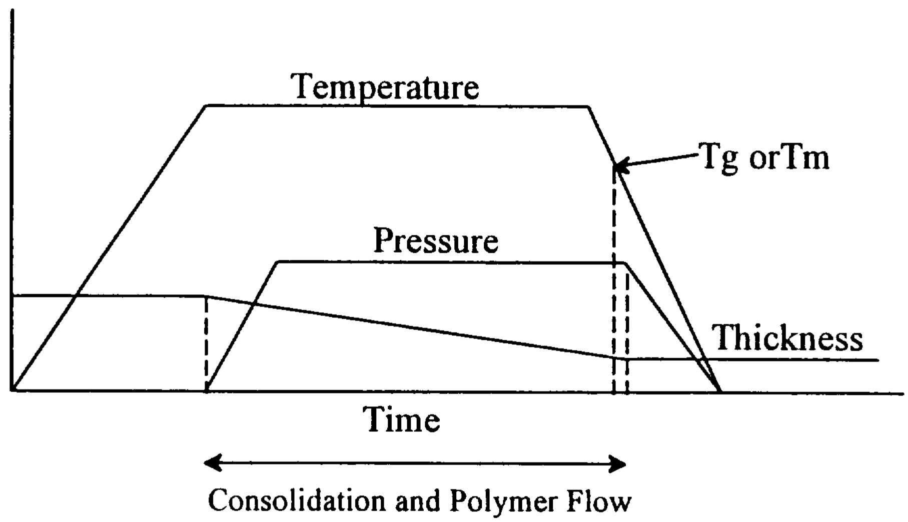


Figure 4. Towpreg Textile Preform Consolidation Cycle

Vacuum and autoclave pressure is applied to induce the resin flow, wetting of fibers, and fiber movement necessary to eliminate voids and fill the intra- and inter-tow spaces. The change in thickness during consolidation is about 4 to 1 for textile preforms. The debulking of preforms is part of the ongoing investigation. The ramping of the pressure allow the fibers time to move into a compact arrangemet,with minimum fiber crimping and breakage, and provides time for resin flow and adhesion.

The composite panel is held above Tg or Tm to anneal the part and releive internal stress which are generated duing the cooling down portion of the cure cycle. Once the material is cooled below Tg, pressure is released so that consolidation is stopped before resin can squeeze out of the layup, causing resin poor areas with lowered mechanical properties.

Flat test panels have been manufactured from woven 8-harness satin fabric from towpreg produced in both wet and dry powder impregnation processes. These methods have been previously described [8]. Early work was done using towpreg having 35 ± 2 weight percent resin. After analysis of the resulting parts, the resin content was raised to 38 ± 2 weight percent for subsequent material. It was evident that the textile operations were causing some powder losses during preform fabrication.

Bulk in Powder Coated Textile Preforms

A bulk factor is inherent in the powder towpreg manufacturing process. Figure 5 is a schematic of the acquisition of bulk during the impregnation of the tow with powder particles. This occurs regardless of the method of towpreg production, although towpreg processed by a dry (for example, electrostatic) technique generally has a slightly higher bulk than that produced using a wet (slurry) technique. This occurs when the tows are spread before the impregnation step. The process of debulking a composite prior to final consolidation is standard throughout the industry. Experience has shown that debulking increases the quality of a thermoset laminate. The debulk cycle removes air that becomes trapped between plies as they are laid down, with the result being both improved handling of the preform and ease of insertion into the tooling. High bulk factors are becoming a very challenging technical problem working with powder towpreg preforms. Typically, bulk factors run about 4:1 in powder towpreg preforms.

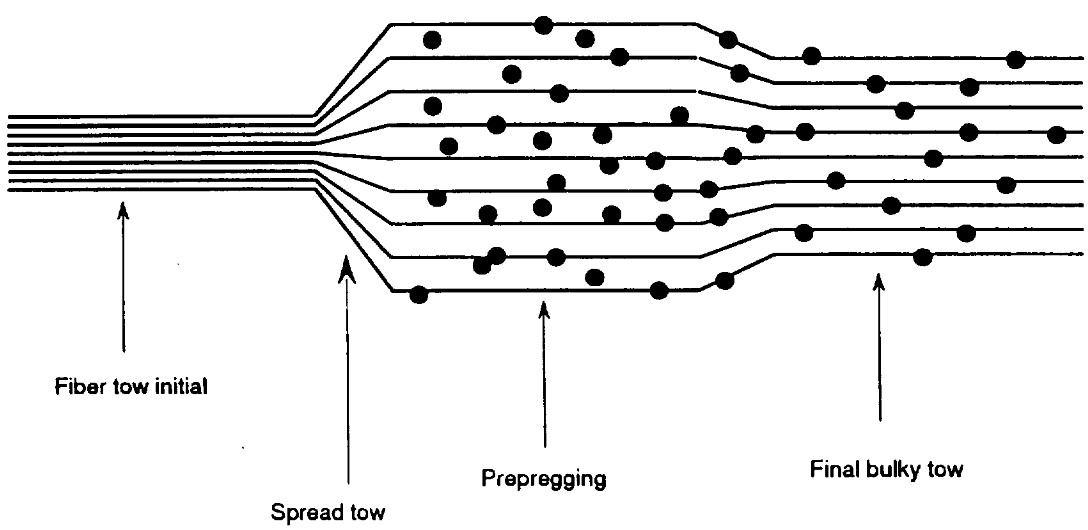


Figure 5. The Origination of Bulk in Powder Towpreg.

Bulk is not a great problem in flat laminates, however, the problem requires special attention whenever complex contours, angles, webs, or dropoffs are produced, if dimensional tolerances are to be maintained. When 3-D weaves or braids are used as preforms, a priority is to minimize fiber bulk in the z-direction. This is critical because z-directional tows restrict the ability of the preform to be compressed (and thus more difficult to consolidate), and then if the preform does compress, the excess fiber will buckle inside the part. Figure 6 shows the effect of bulk on the consolidation characteristics of a 3-D preform.

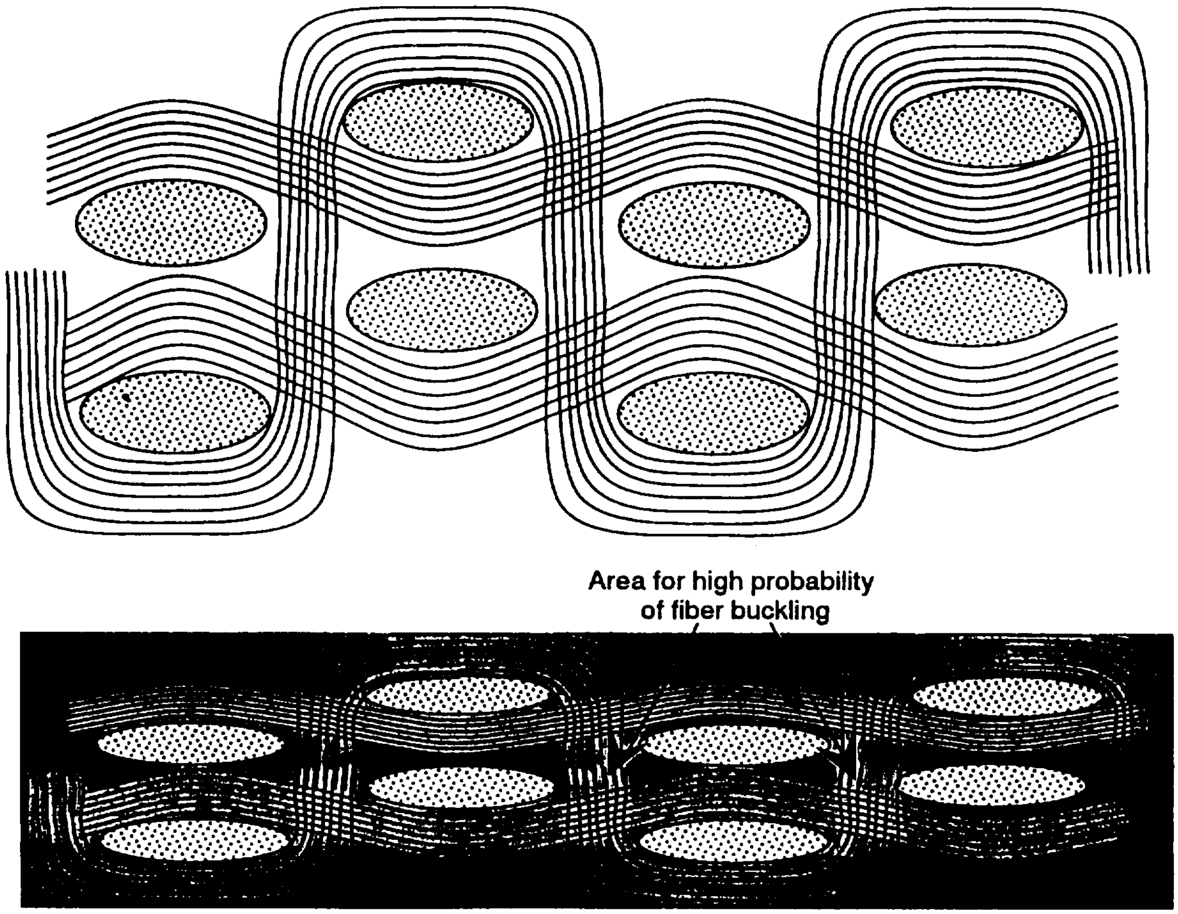


Figure 6. The Effect of Bulk on the Consolidation Characteristics of a 3-D Preform.

Debulking of 2-D and 3-D Textile Preforms

A debulking study has been initiated to address the issues related to consolidation of advanced textile preforms. The preform types which have been, or will be, under consideration are: 1) 2-D braid (supplied by Fiber Innovations[1]); 2) 3-D through the thickness weave (Techniweave[1]); 3) 3-D Multi-axial knit (Tech Textiles[1]); 4) 3-D weave (Fiberite[1], Fabric Development[1]). Initially, the study will establish the time, temperature, and pressure parameters for debulking flat samples of several advanced textile preform types, using towpreg produced at BASF (resin content: 38 ± 2 % by weight). Upon completion of the debulking study and establishment of process parameters, the verification of stiffened panel preform quality, tooling and manufacturing approaches shall be demonstrated. The manufacturing demonstrations will consist of debulking and processing representative sections of a side panel, such as an integrally stiffened window belt.

Prior to embarking on the debulking study, a short aging study was conducted on the AMD 0036 neat resin. Samples of the resin were held at 200°F for varying times, then analyzed rheometrically and by Differential Scanning Calorimeter (DSC). These analyses were done on samples which had been held at temperature for 15 minutes, 45 minutes, and 60 minutes at a heating rate of 4°F per minute. Viscosity data in Table 4 show that the minimum viscosity is virtually unchanged after aging at 200°F for 30 minutes. The DSC was run at a heating rate of approximately 18°F/minute. The results, also shown in Table 4, were compared with an as-received sample of the same batch, as well as with a sample from a second batch of powdered AMD 0036. The results show little difference between unaged and aged resin, for both the onset and peak exotherm temperature.

Table 4. Results of Differential Scanning Calorimetry and Rheometric Studies of Aged AMD 0036 Epoxy Resin.

Sample (Batch #IM2P)	1st Peak (J/oz)	2nd Peak (J/oz)	Total (J/oz)	Peak Exotherm (°F)	Minimum Viscosity (Poise)
#1--As Received	1700.2	430.4	2130.6	456.3	1300
#2--15 min. at 392°F	1632.4	498.6	2131.0	452.7	
#3--30 min. at 392°F					1360
#4--45 min. at 392°F	1666.7	404.9	2071.6	452.3	
#5--60 min. at 392°F	1486.8	315.6	1802.4	456.4	
#6--As Received Batch # 8126			2280.7	455	

2-D braided preforms manufactured by Fiber Innovations, Inc. were fabricated using BASF[1] produced AS-4/RSS-1952 towpreg, at a resin content of 39 ± 2% by weight. All towpreg was 6k, and samples 10.5" x 6" having 10 ends/inch were braided using 72 carriers, and a 3.25-inch mandrel diameter. Preforms containing 6 plies were braided; a dry lubricant was applied to the axial tows to reduce friction during the braiding of the towpreg. A silicone rubber vacuum table, with heat on the top only, was used, and the debulk cycle was as follows: 1) apply full vacuum; 2) heat from room temperature to 200°F; 3) hold for 15 minutes; and 4) turn the ply stack over, and repeat steps 1-3.

The typical initial thickness of the 6-ply braided preforms was 0.50". After the debulk cycle, the preform thickness was 0.22". After curing, the thickness of the sample was 0.124". This represents a debulk from 4:1 to 1.8:1. While the 2-D braided preforms react well to a pre-consolidation debulk cycle, issues relating to the incorporation of the dry lubricant must still be resolved before these materials will be considered for test panels. These issues relate to the reactivitiy of the dry lubricants under consideration and the effects of their presence on the mechanical properties of the resulting laminate.

Initial efforts at debulking 3-D preforms centered on the 3-D through the thickness woven preform manufactured by Techniweave, Inc. The nature of the z-direction fibers in these preforms is such that it is advantageous to try to debulk during preform fabrication.

On-line debulking studies will begin in the early summer at Techniweave. Currently, post-fabrication debulking of 3-D woven shaped preforms is being studied by Lockheed.

Two methods of debulking the Techniweave preforms have been undertaken. In the first method, hard tooling was made to debulk the T-stiffened preform . Three debulking cycles were conducted on the preforms. First, the preform was heated under vacuum pressure to 200°F, and held for 20 minutes. The bulk factor of the skin portion was reduced from 2.5 to 2.0, and the stiffener from 3.2 to 3.1. The second cycle was also done at 200°F, using autoclave pressure of 100 psi for 20 minutes. This time, the bulk factor of the skin was reduced from 2.0 to 1.2, and in the stiffener from 3.1 to 2.3. To reduce the bulk of the stiffening element further, a third cycle was conducted using moveable plates, rather than angle tools, to apply pressure to the perpendicular members of the preform individually. Under autoclave pressure of 100 psi for 20 minutes at 200°F, the bulk factor of the stiffener was reduced to 2.0. The step-wise debulking which occurred using this method is shown in Figure 7. The large surface area of the skin portion of the preform restricted the tool from sliding inward to the stiffener, preventing an even application of tool pressure, which caused an uneven debulk of the vertical stiffening member. After final consolidation of the part, it was evident that the cross-section of the skin/stiffener intersection was not symmetric about a neutral axis, however, the thickness of the respective horizontal and vertical members of the part were within specification.

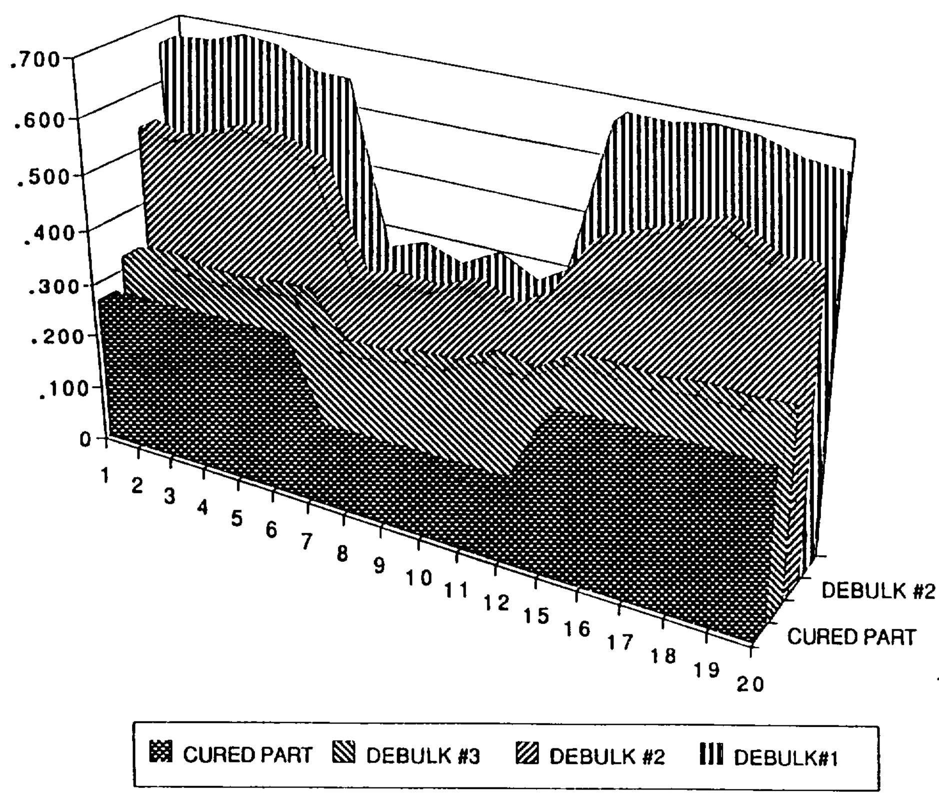


Figure 7. Results of Debulking Trial on 3-D Woven T-Stiffened Preform.

The results of the first debulking effort led to a redesign of the tooling for the T-stiffened Techniweave preform. In this approach, the part is vacuum bagged, placed in an autoclave and debulked using full vacuum pressure plus autoclave pressure of 100 psi. This is a two step approach where the stiffener will be debulked before debulking the skin. The preform will be hot debulked from both sides towards the center line of the blade using a combination of hard and rubber tooling material. The rubber is sized to provide approximately 150 psi on the stiffeners. Again, the part is heated to 200°F, and held for 20 minutes. Preliminary results show this approach to be successful in providing an even debulk of the preform. Both the vertical and horizontal members were debulked from approximately 4:1 to less than 2:1, however, there was a small amount of fiber bridging in the skin/stiffener intersection. The results of this trial are currently being further analyzed.

Consolidation of 2-D Woven Fabric

Epoxy powder coated eight-harness satin fabric was laid-up, bagged and autoclave processed into flat panels for mechanical property testing for three powdered epoxy resin systems: AMD 0036[1] (3M), RSS-1952[1] (Shell), and CET-3[1] (Dow). Powder towpreg manufactured by Custom Composites Materials, Inc.[1] (CCMI) of the 3M resin system was woven into 8-harness satin fabric by Textile Technologies, Inc.[1] (TTI). The resin content of this material was only about 27 weight percent; powder was added by hand to bring the panels up to 35 ± 2%. The other two resin systems were evaluated using towpreg produced by BASF[1], and woven into 8-harness satin fabric by Fabric Development, Inc[1]. Flat panels having fiber volume fraction of 58% to 60% were fabricated and evaluated by Lockheed [8].

Early attempts at consolidating the RSS-1952 and AMD 0036 coated powder fabrics were not successful due to the inherent hydroscopic tendencies of the powdered resins. Since the towpreg will be stored at room temperature, long term exposure to a high humidity environment may be experienced. Some moisture may also be used by the textile operators for lubrication during fabric production.

During the first few trials, water droplets formed between the part and the caul plate during the cure cycle. Some drying cycle tests were performed to determine the time required to dry a given number of plies in an oven/autoclave. The results resulted in a one hour drying cycle at 120°F under full vacuum pressure, during the autoclave cure cycle to insure that trapped moisture was eliminated before consolidation in the autoclave. High quality laminates were made using this approach, and a study to evaluate the necessary drying segment length for particular 3-D preforms will be initiated at Lockheed in the future.

Towpreg flat panel laminates were cured using a caul plate with 0.050" thickness after early trials showed that 0.080" thick cauls were too thick, given the low viscosity of the resin systems. The center regions of these panels were porous due to lack of pressure caused by the bowing of the caul, from edge bleeding of the resin. When resin extruded from the perimeter of the panel, the edge of the caul was unsupported and the caul subsequently bent. A more uniform pressure distribution having only local edge deformation was delivered using the thinner caul.

The low viscosity of the resin systems used in the program prompted excessive resin bleed during consolidation, therefore, an air dam around the perimeter of the parts was incorporated into the bagging scheme. The air dam was placed roughly 0.5" away from the apart to minimize bleed, yet maximize breather paths. The resulting layup bagging scheme is shown in Figure 8. A noticeable gap (0.5") is seen in Figure 8 between the edge of the caul and the edge of the part. This configuration is intended to allow for better breathing of the laminate during cure. Porosity tends to occur when adequate breathing channels are not available, even with the low viscosity of the resin. This is more than likely a result of high bulk factors which are inherent in the powder towpreg preforms. Future work is palnned to determine whiy this caul plate configuration must be used to fabricate quality laminates.

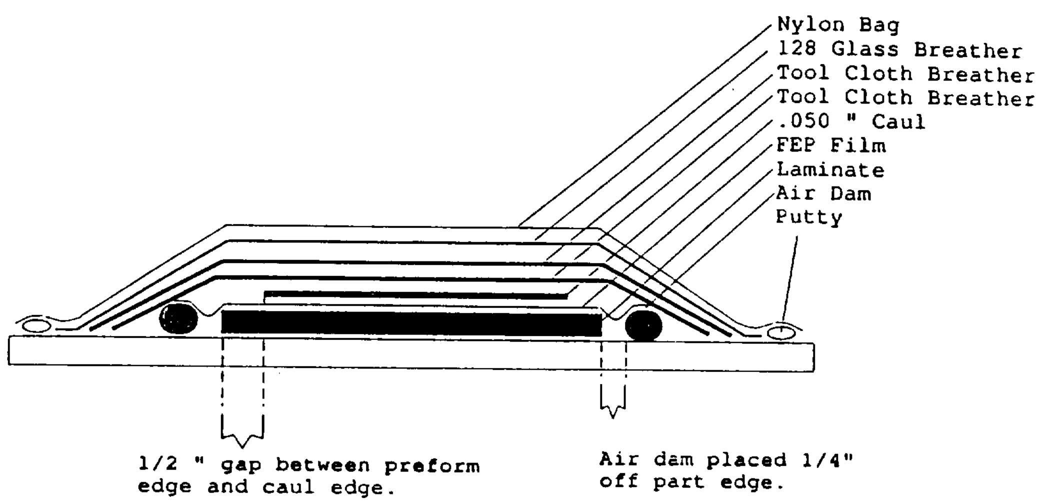


Figure 8. Layup Bagging Scheme for Powder Coated Flat Panel Laminates.

Some alterations to these generalized consolidation guidelines have been incorporated because of resin specifics. Initial cycles using the Lockheed standard heating rate of 5°F/min produced porous panels, however, well consolidated panels resulted when the heating rate was lowered to 3.5°F/min. The AMD 0036 epoxy resin requires a lower heating rate than standard epoxies in the cure cycle to minimize porosity. The CET-3 system required a thicker caul plate, since this system had a higher melt viscosity than the 3M resin. The lack of edge thinning due to bowing of the caul was reduced, and less bleeding was observed about the perimeter of the CET-3 parts.

Mechanical Properties of AS-4/AMD 0036 Flat Panels

The quality of the flat panel laminates met Lockheed ultrasonic specification. Self-calibration of 65% reflected through ultrasonic signal is considered acceptable. C-scans are performed using a 0.5-inch diameter transducer, at a frequency of 5 MHz, gain of 16 db, and a focal length of 4.0 inches.

Mechanical property testing of powder coated 8-harness satin fabric panels fabricated at Lockheed was conducted at Delsen Testing Laboratories, Inc. Test results normalized for fiber volume fraction for the PS-501 (3M) and RSS-1952 (Shell) systems are presented in Table 5, and compared to similar results for the PR-500 RTM resin [8]. PS-501 data represents the earlier lab scale version of the AMD 0036 epoxy system. Testing of the Dow CET-3 flat panels is currently underway.

Test results indicate that the PS-501 system is superior to the RSS-1952 material in all tests, with the exception of open hole tension and compression, both at room temperature (dry) and 180°F (wet). In the open hole tests, RSS-1952 outperforms the PS-510 resin system by 15% in compression; in tension the difference is only 2.7 ksi (6.4%). The PS-501 compression after impact (CAI) results are 40% higher than RSS-1952, for the NASA CAI test specimens. A photomicrograph from a CAI specimen is presented in Figure 9. The micrograph shows a classic compression failure, with a shear line across the failed region, when viewed from the panel profile. No global bending or buckling deformation was observed. Some splintering of the plies was observed on the back side of the panel, due to the impact.

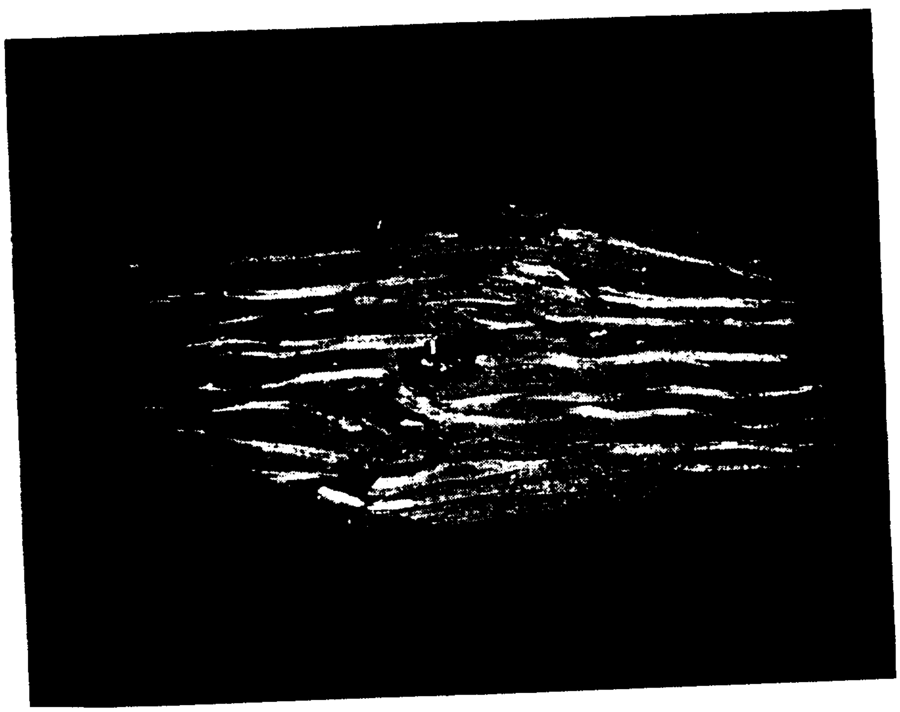


Figure 9. Photomicrograph from PS-501 CAI Specimen, Showing Compression Failure.

The PS-501 flat panel test results compare favorably with panels fabricated from the PR-500 RTM resin, except in the case of the hot-wet results, where the RTM panels were higher. The strength loss in these tests was from 21% to 28%. 3M Company has indicated

Table 5. Normalized Mechanical Test Results for Powder Coated 8-harness Satin Fabric Flat Panels

Test Type	PS-501 3M	RSS-1952 Shell	CET-3 Dow	PR-500 RTM Resin
0° Tension				
RT				
Ultimate Strength, ksi	99.2	91.8		94.7
Modulus, Msi	10.04	8.77		9.17
Poison's Ratio	0.042	0.042		0.079
Strain To Failure, %	1.17	0.00		1.10
-65°F				
Ultimate Strength, ksi	104.4	93.2		99.3
Modulus, Msi	9.92	8.96		8.92
Poison's Ratio	0.048	0.048		0.065
Strain to Failure, %	1.08	1.01		1.15
Fiber Volume, %	52.5	54.8		57.0
Orientation / Ply	0° / 4	0° / 4		0° / 3
0° Compression				
RT				
Ultimate Strength, ksi	103.1	81.1		100.9
Modulus, Msi	9.13	8.91		8.98
Poisson's Ratio	0.053	0.021		0.028
180°F, wet				
Ultimate Strength, ksi	70.8	50.3		87.8
Modulus, Msi	8.96	8.08		8.93
Poisson's Ratio	0.064	0.065		0.040
Fiber Volume, %	53.98	53.3		56.1
Orientation / Ply	0° / 8	0° / 8		0° / 8
45° Tension				
Ultimate Strength, ksi		10.6		16.1
Modulus, Msi		0.59		0.75
Fiber Volume, %		54.6		58.7
Orientation / Ply		45° / 6		45
Unnotched Tension				
Ultimate Strength,ksi	65.3	64.4		7.1
Modulus, Msi	6.84	5.84		6.47
Strain To Failure, %	1.21	1.07		1.09
Fiber Volume, %	61.5	54.4		56.2
Orientation / Ply	quasi / 8	quasi / 8		quasi / 8

Test Type	PS-501 3M	RSS-1952 Shell	CET-3 Dow	PR-500 RTM Resin
Open Hole Tension, ksi				
RT	41.9	44.6		47.8
-65°F	41.6	43.0		45.4
Fiber Volume, %	54.3	52.3		53.6
Orientation / Ply	quasi / 8	quasi / 8		quasi / 8
Unnotched Compression				
RT				
Ultimate Strength,ksi	70.3	59.3		72.4
Modulus, Msi	6.24	6.09		6.30
Strain to Failure, %	1.34	1.07		1.35
180°F, wet				
Ultimate Strength, ksi	50.3	37.5		64.3
Modulus, Msi	6.14	5.89		6.30
Strain To Failure, %	0.86	0.66		1.22
Fiber Volume, %	54.4	52.7		54.6
Orientation / Ply	quasi / 8	quasi / 8		quasi / 8
Open Hole Compression, ksi				
RT	38.1	44.3		41.4
180°F, wet	30.5	29.4		34.1
Fiber Volume,%	58.9	46.6		54.7
Orientation / Ply	quasi / 8	quasi / 8		quasi / 8
Compression After Impact (SACMA)				
Ultimate Strength, ksi		23.9	30.9	34.1
Modulus, Msi		6.36	5.82	6.64
Strain to Failure, %		0.39	0.50	0.51
Fiber Volume, %		55.6	60.4	59.8
Orientation / Ply		quasi / 12	quasi / 12	quasi / 12
Compression After Impact (NASA)				
Ultimate Strength, ksi	40.5	28.3		39.3
Modulus, Msi	6.40	6.52		6.54
Strain to Failure, %	0.67	0.44		0.63
Fiber Volume, %	55.6	55.5		57.5
Orientation / Ply	quasi / 18	quasi / 18		quasi / 18

that the hot-wet properties will improve in the scaled-up (AMD 0036) version of the powder. Some of the tests in Table 5 will be repeated using CCMI produced AMD 0036 towpreg, to evaluate any mechanical property differences between the electrostatic method and the slurry method of producing towpreg. All of the hot-wet tests will also be repeated.

SUMMARY

This study has dealt with textile applications of powder towpreg, focusing on weaving and consolidation. Some of the operating and design issues in these processes have been resolved while others have been highlighted for further attention. Making textile preforms from polymer powder-coated towpreg depends upon a number of material properties and equipment parameters. An optimal weaving protocol requires tow handling minimization, and tow bundle twisting of about 0.4 twists per inch. These textile techniques are important factors for automating the production of quality composite parts from powder-coated towpreg preforms.

The consolidation cycle for woven towpreg must account for the inter-bundle crimp of the weave. Since a higher towpreg resin content is needed to fill the interstitial spaces in woven materials than for unidirectional tape, in general, composites made from woven material will have a lower fiber volume fraction. Further studies are required to establish the optimum fiber volume fraction for powder-coated preforms. Because of the initial bulk associated with woven materials, vacuum pressure must be applied during the cure cycle to prevent the formation of air voids. During consolidation, the fibers in woven materials must move and realign, while resin must flow to fill the interstitial spaces. A gradual increase in pressure over time provides for greater ease of fiber movement and resin flow before the fibers align in a tight, compact arrangement. Attention to these factors, together with the general practice of holding for a period of time at maximum pressure and temperature followed by cooling at a rate which minimizes internal stresses, yielded composite specimens of woven material that were void-free, as determined by ultrasonic C-scans.

Mechanical properties were determined from composite specimens made with carbon fiber tow bundles of 3k, 6k and 12k that were coated with LARC™-TPI 1500 medium flow grade powder. Testing was performed on unidirectional and eight-harness satin fabric composite specimens. No apparent effect of tow size was found in the unidirectional or the eight-harness satin fabric composite specimens. The lower compression strength dispalyed by the unidirectional composites made with twisted towpreg was due to apparent fiber damage that occurred during the twisting of the tow bundle.

The matter of optimum tow bundle size when comparing the mechanical properties remains unresolved. Fiber damage has been observed to be less when larger tows are used. Weaving equipment capabilities are somewhat independent of tow size. It appears that the choice of tow bundle size is an open one in regard to properties, but that tows containing at least 12,000 or more fibers are favored, especially in regard to powder processing and weaving costs.

An aging study on AMD 0036 resin system showed that exposure to 200°F for up to one hour has little effect on the rheological characteristics of the material. Debulking studies on 2-D textile preforms showed that 200°F and vacuum pressure was sufficient to reduce the bulk factor from 4:1 to less than 2:1. Autoclave pressure is necessary to debulk 3-D woven preforms, due to the presence of the z-directional fibers.

The NASA ACT Program has selected 3M AMD 0036 as the powder system to be carried throughout the rest of the contract. The mechanical properties of flat panel laminates fabricated using towpreg coated with the lab scale formulation of AMD 0036 are superior to other powder-resin systems investigated, and comparable to the PR-500 RTM resin, except in hot-wet tests. It is anticipated that the hot-wet properties will be improved during the scale-up production, and tests to confirm this are planned.

ACKNOWLEDGEMENTS

The authors gratefully acknowledge the assistance of Mr. John Snoha for preparing the powder-coated towpreg and the consolidated woven cloth panels. Mr. Ruperto Razon for the unidirectional specimen preparation, and Mr. Benson Dexter for his assistance with the weaving studies and mechanical tests. Additional thanks is extended to Mr. Michael Cannon, Mr. Jay Shukla, Mr. Carl Hartup, and Dr. Sally Wu for their contributions to the development and understanding of the processing science of powder coated textiles.

REFERENCES

1. J. Marchello, and R. M. Baucom, "LARC™ Dry Powder Towpreg Process", *Science of Advanced Materials and Process Engineering Series,*vol. 36, April 1991, p. 68.

2. M. K. Hugh, J. M. Marchello, R. M. Baucom, and N. J. Johnston, "Composites from Powder-Coated Towpreg: Studies with Variable Tow Sized. *Science of Advanced Materials and Process Engineering Series,* vol. 37, March 1992, p. 1040.

3. R. M. Baucom, and J. M. Marchello, "LARC™ Powder Prepreg System", *SAMPE Quarterly,* vol. 21, no. 4, July 1990, p. 14.

4. D. E. Hirt, J. M. Marchello, and R. M. Baucom, "Study of Flexural Rigidity of Wearable Powder-Coated Towpreg", 22nd International SAMPE Technical Conference, November 1990, p. 360.

5. D. F. Adams, T. R. King, and D. M. Blackketter, "Evaluation of the Transverse Flexure Test Method for Composite Materials", *Composites Science and Technology,* vol. 39, 1990, p. 341.

6. C. Zweben, W. S. Smith, and M. W. Wardle, "Test Methods for Fiber Tensile Strength, Composite Flexural Modulus, and Properties of Fabric-Reinforced Laminates", Fifth Conference on Composite Material: Testing and Design, ASTM STP 674, 1979, p 228.

7. M. K. Hugh, J. M. Marchello, J. R. Maiden, and N. J. Johnston, "Weavability of Dry Polymer Powder Towpreg", 3rd NASA Advanced Composites Technology Conference, NASA CP-3178, Part I, June 1992, p. 175.

8. J. G. Shukla and T. D. Bayha, "Advanced Resin Systems and 3-D Textile Preforms for Low Cost Composite Structures", 3rd NASA Advanced Composites Technology Conference, NASA CP-3178, Part I, June 1992, p. 159.

Ceramic Preconsolidation Tool for Thermoplastic Powder Towpreg

The ceramic preconsolidation tool facilitates isolation of hot and cool contact zones which are required for the conversion of bulky powder coated carbon fibers into preconsolidated towpreg ribbons.

Langley Research Center, Hampton, Virginia

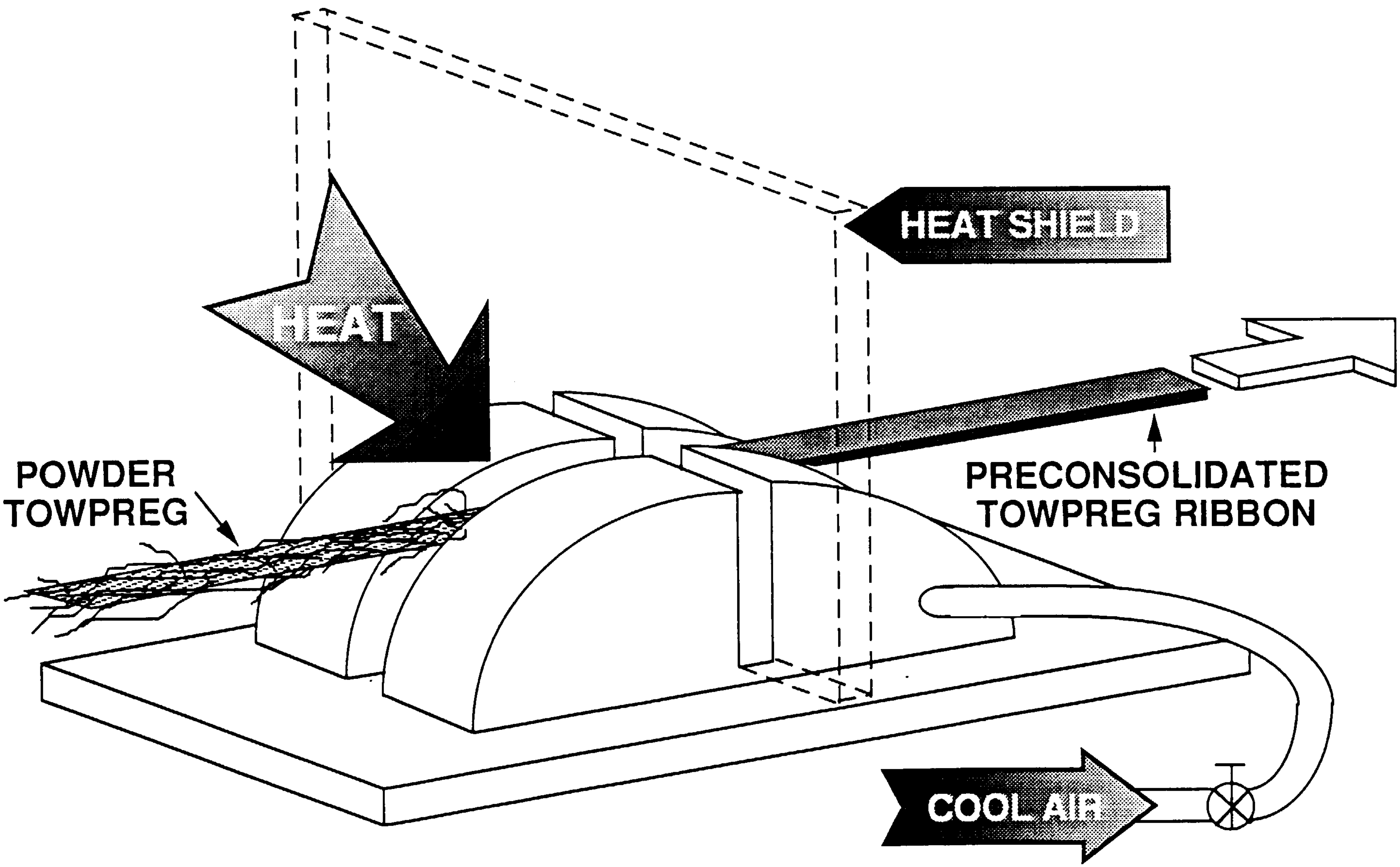
The potential use of various high-temperature performance thermoplastics as carbon fiber composite matrix systems has prompted research to identify novel methodology in the production of advanced composite prepreg systems Many of these polymers are not amenable to solvation in environmentally friendly solvents. When these polymers melt they typically exhibit high viscosity and are difficult to utilize in conventional prepreg operations due to reduced polymer flow during impregnation. Several techniques have been developed to plasticize these polymers but the plasticizers tend to reduce the mechanical properties of the bulk thermoplastic.

Current research at NASA LaRC has led to the development of novel tooling and prepregging techniques which convert powder-coated towpreg into preconsolidated ribbon without the use of any plasticizers or solvents of any kind. Relying on melt-flow and proper application of tension, friction and cooling, this processing method converts powder towpreg into ribbon with consistent polymer distribution and cross-section.

This innovative debulking technique draws on the advantages of pulling unidirectional filaments through a heated / cooled open-faced die. The ceramic tool was constructed at NASA LaRC, specifically for this purpose. After the bulky towpreg leaves the pay-out creel, it is warmed by a flow of heated gas. Initial contact between the melted towpreg and the tool occurs on a curved surface which is channeled into the tool. As the bundle is drawn over the tool. it spreads out to fill the channel. This accomplishes two things: first, it allows for melt squeeze flow to distribute the polymer more evenly throughout the fibers and secondly, it facilitates fiber nesting under tension which provides debulking. The heated towpreg now has the shape of a wide and flat ribbon. An air insulated region divides the two sections of the tool which provides sufficient thermal barrier necessary to separate the actively cooled zone of the tool from the heated zone. The ribbon next enters the cool zone of the tool and cools to a solid under controlled conditions. Shrinkage of the polymer and its reduced stickiness allow it to separate from the tool surface.

*This development is part of an ongoing research effort headed by Robert M Baucom of **NASA LaRC** Polymeric Materials Branch, and conducted by Donald A Sandusky of the College of William & Mary Applied Science Department, and Joseph M Marchello of Old Dominion University.*

CERAMIC PRECONSOLIDATION TOOL



April 15, 1993

Memorandum

To: N. Johnston, R. Baucom, M. Hugh, J. Hinkley, D. Sandusky, and S. Wilkinson

From: J.M. Marchello JMM

Subject: Composite Part Fabrication

Towpreg, polymer powder impregnated fiber tow, is now being produced at semi-commercial levels (over 100 pounds per month) by several companies (Custom Composites, BASF/Hexcel, EniChem and perhaps others). In the period ahead, these processes, and NASA-LARC's process, will continue to be developed while significant quantities of towpreg are made for test panels and part fabrication studies.

In conjunction with the increased availability of towpreg, our thoughts have shifted more toward composite processing and part fabrication. Some recent discussions have dealt with the issues involved in making different aircraft parts. The purpose of this memorandum is to summarize and share this information, and to obtain further comments.

The development of methods for using towpreg in the fabrication of composite parts for commercial aircraft is an important goal of the ACT and HSR (HSCT) programs. A knowledge about, and acceptance of, newly developed methods is vital to the Trade Studies and to the Design-Build team deliberations, which are concerned with increased use of composites in commercial aircraft.

Starting with towpreg yarn, there are several approaches to making composites. These are generally considered to be the use of textile technology, thermoforming, and automated tow/tape placement.

Textile technology may be used to weave or braid the towpreg into broadgoods and preforms for part fabrication. An important goal of the textile studies is to develop a powdered preform molding alternative to the RTM process for producing net shape parts. Having the polymer powder dispersed in the fiber bundle reduces melt-flow requirements and allows composites to be made from high melt viscosity polymers. Major developmental concerns with textile applications are tow friction (abrasion) during braiding and debulking of preforms during consolidation. These issues are being addressed in studies of towpreg surface treatment for braiding, debulking (cold) during weaving, and special tooling for debulking (hot) preforms during consolidation and part fabrication.

Thermoforming, primarily pultrusion and die forming, using towpreg offers a means of making parts directly and of making customized ribbon for ATP. Development of automated tow placement methods focuses on reduction of labor costs, beyond those of automated tape placement, through the development of special towpreg ribbons and robotic heads that can handle complex tool surfaces with sharp turns, ribbon add and drop, honeycomb cores, and in-situ consolidation. These issues are being addressed by studies of towpreg ribbonizing, autohesion bonding kinetics, and robot head modifications.

Direct part pultrusion is limited to making long fairly uniform parts, such as floor boards and support members. Reduction in the cross section of a part along the length may be achieved to some extent by using a contracting die and tow dropping methods. For example, a J bar of varying height might be pulled starting with the die fully extended. As pulling proceeds, the die J height is reduced and internal tows are dropped to accomodate the reduced die area along the part length.

With high temperature polymers, the intermediate composite products, from which parts are to be fabricated, are much stiffer, or boardier, and more difficult to work with than conventional prepreg or aluminium. This requires new approaches to fabrication and suggests possible utilization of carpentry and cabinet making techniques. There is a precedence for this. In 1940, when he could not get aluminum to build a protype of his planned 800 passenger flying boat, Howard Hughes used wood and made the Spruce Goose. For the HSCT aluminum is inadequate, so stiff, wood like, composites will need to be used. Instead of SST, the next plane might be called SSG for Son of Spruce Goose.

Current composite aircraft part fabrication studies deal with wing and fuselage sections and their supporting stringer members, window frames and window belt sections, air ducts, and floor boards. As a final thought topic, in the quest to make an all-composite airplane, consider the following possible way of making an aircraft landing gear column.

Landing gear columns contain hydraulic fluid and are made of high strength steel to handle high compression, impact, and flexural loads. The objective, then is to make a thick walled composite pipe that can handle the environment and loads of aircraft landing.

Composite are generally thought as being strong in tension and weak in compression. The reason for this is that the fibers are thin and they buckle readily under compression loading. If fiber buckling can be prevented, the composite might exhibit high compressive and flexural strength.

A composite landing gear column might be fabricated as follows. Filament wind several layers of towpreg on a metal pipe, having the diameter needed for the internal diameter of the cylindrical column, and consolidate. Cool and remove the pipe tool. (Alternately, the pipe tool might be left in and removed later.) This step produces a composite pipe with properties like an NOL ring. Attach on the outside of this composite pipe a large number of towpregs aligned in the axial direction. To ensure that they remain oriented in the axial direction these towpregs should be held with some tension. Consolidate so that the linear towpreg layer is debulked and resin bonded to the composite pipe. Tightly filament wind several towpreg layers around this tube and consolidate. Repeat the process by adding another layer of linear towpregs, consolidate, filament wind with towpreg and consolidate. Continue this buildup process to the external diameter of the part.

The result of the above process is a hollow composite cylinder consisting of a series of rings of linear (about 90%) and wrapping (about 10%) tows. The wrapping tows would serve to prevent buckling of the linear tows, which would provide the strength properties needed for a light weight landing gear column.

April 13, 1993

Memorandum

To: N. Johnston, R. Baucom, D. Sandusky, M. Hugh, J. Hinkley, and S. Wilkinson

From; J. Marchello JMM

Subject: Multi-tow Ribbon for ATP

There are several important aspects of customized towpreg ribbon that we have been discussing in recent weeks. The purpose of this memorandum is to attempt to summarized and share these thoughts, and to obtain additional comments.

Ribbon architecture concepts, that we have developed and published during the past year or so, deal with elimination of tow-tow gaps, improved bonding, rapid laydown, the ability to handle sharp placement turns and concave tool surfaces, and in-situ consolidation. In this regard we have proposed non-rectangular ribbon cross-sections (triangle, trapezoid, and parallelogram) that provide for ribbon nesting during placement, and the use of narrow ribbon for small radius laydown turns. These considerations have led to studies of towpreg ribbonizing, autohesion bonding kinetics, and robot head design modifications.

The standard of comparision for ATP ribbon is APC-2. It is 5.6 mills (142 microns) thick and 250 mills (6350 microns) wide. That corresponds to a cross-sectional area of 901,700 square microns. With 65% fiber volume, the fiber area is 586,105 sq. microns. With AS4 fibers (7 micron diameter or 38.5 sq. micron area) the fiber count of the ribbon is 15k. With IM7 (5 micron diameter or 19.6 sq. micron area) the fiber count is 30k.

Since the HSR/HSCT program will use IM7 fiber, to make a ribbon having the same area as APC-2, we would need to use either one 6k and two 12k tows, or five 6k tows. This is why we want to acquire a multi-tow creel for the ribbonizing studies.

An important feature of our customized ATP ribbon effort rests on the goal of developing automated (robotic) tow/fiber placement to the point where the robot head can do anything that can be done by human hands. This means handling complex tool surfaces that require placement during sharp turns, in confined highly curved areas. Add to this the desire for high (commerical) laydown rates and in-situ (on-the-fly) consolidation. Also, in many cases the part will consist of a honeycomb core with a composite skin. To avoid damage to the core/tool, ATP placement of such skins will require using less than 50 psi of pressure, and good adhesive bonding to the honeycomb.

To place tow ribbon without buckling during sharp turns, it is necessary to use narrow ribbons. For example, 1/8 inch (125 mill) wide ribbon can be placed at a turning radius of 5 inches. Sharper turns would require narrower ribbon.

To achieve tow gap reduction, or elimination, by nesting, the ribbon sides must be at angle other than 90^o (say 45^o) so that the ribbons will slide together while under the roller pressure during laydown. Consideration of the requirements for nesting and bonding of shaped ribbon indicate that the ribbon should be thicker than APC-2, perhaps about 20 mills, to provide adequate surface for lateral tow movement and bonding.

The above suggests development of customized ribbon having an average width of about 100 mills and a thickness of around 20 mills. Such a ribbon would have a cross sectional area of 1,290,320 sq. microns. With 65% fiber volume, the ribbon would have a fiber count of 43k. It could be made from seven 6k tow, or perhaps four 12k tows.

Using this thicker and narrower ribbon provides the potential to laydown at turning radii less than 5 inches and, since it is larger, to increase laydown rates. However, it may give rise to concerns about achieving adequate isotropic properties and about interlaminer microcracking.

With APC-2 ribbon placement, the ply thickness is 5.6 mills, subquent plys are laid at different angles (0,$\pm$45,90 degrees) which provides for isotropic composite properties. For a given composite thickness, using 20 mill ribbon would reduce the degree of isotropy since there would be fewer plies. However, braiding the powdered tows prior to ribbonizing would give some degree of isotropy, but only within the width of the ribbon. A braided and consolidated 20 mill thick ribbon, laid at different angles (0,$\pm$45,90), would result in improved isotropy in the composite over unbraided ribbon, but it would have less isotropy than that obtained using APC-2. This could result in somewhat reduced composite mechanical properties, which must be balanced against the potential benefits of automated sharp turn laydown, reduced gaps, and higher placement rates (less labor).

It has been observed that microcracking increases with tow bundle size. That is, when the ply consists of unidirectional fibers, increased ply (ribbon) thickness results in more microcracks. Thus, a 20 mill thick ribbon of unidirectional fibers would be expected to exhibit more microcracks than a 5.6 mill ribbon. If the 20 mill ribbon were made from braided tows, the possibility of microcracking would be reduced.

In view of the above considerations regarding microcracks and isotropy, it seems that we should direct some of our ribbonizing efforts toward shaping (customizing) braided towpreg. We might braid six 12k towpreg yarns and pull the braid through a die to make a 20 mill ribbon. For comparsion, we could pull six 12k towpreg yarns, unbraided, throught the die to make a 20 mill ribbon. These ribbons could be made into test panels to study mechanical properties and microcracks, and compared to thinner ribbons.