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POWDER TOWPREG PROCESS DEVELOPMENT

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SUMMARY

The process for dry powder impregnation of carbon fiber tows being developed at LaRC overcomes many of the difficulties associated with melt, solution, and slurry prepregging. In the process, fluidized powder is deposited on spread tow bundles and fused to the fibers by radiant heating. Impregnated tows have been produced for preform, weaving and composite materials applications.

Design and operating data correlations have been developed for scale up of the process to commercial operation. Bench scale single tow experiments at tow speeds up to 50 cm/sec, have demonstrated that the process can be controlled to produce weavable towpreg. Samples have been woven and molded into preform material of good quality.

INTRODUCTION

Because of the high melt viscosities of thermoplastics and toxicity and handling problems with solvents, several years ago NASA Langley Research Center (LaRC) began investigating ways to powder coat carbon fiber using fluidized bed, slurry, and electrostatic technology (1). It was felt that powder coated prepreg would have good drape and perhaps some tack. Powder coated tow was thought to have the potential to be woven into cloths and preforms and to be filament wound and pultruded. Powder based composites offered the potential of being cost-effective to produce and fabricate.

Experiments at LaRC and elsewhere have confirmed many of the expectations for powder prepregging (2). Some of the important features of the powder coating process are these:

- Versatile: Thermoplastics and thermosets
- Operates at room temperature
- Involves no solvents
- Has manageable exposure to toxic materials
- Requires no significant refrigeration: reduces waste/spoilage
- Can be woven, pultruded and thermoformed
- Offers a viable alternative to RTM processing of textile preform composites

The dry powder process under development at LaRC overcomes many of the difficulties associated with melt, solution and slurry prepregging. In the process, fluidized powder is deposited on spread tow bundles and fused to the fibers by radiant heating. This paper reports the results of bench scale experiments that have provided test materials and design and scale up information needed for process development.

EXPERIMENTAL

The experimental system was composed in sequence of the tow feed spool with tow tension brake; the fluidization chamber with powder feeder; the electric oven; the quality control monitor; the take-up spool with tow speed control, Figures 1 and 2. Unsized Hercules AS-4 carbon fibers in 3K and 12K tows were used with six matrix polymer powders, Table I.

DESIGN RELATIONS

The experimental equipment and operating procedures have been described in detail in previous reports (2,3). The bench scale dry towpreg system was operated over a wide range of conditions to confirm design theory and operating correlations for each component and thereby provide the basis for scale up to produce commercial quantities of towpreg.

Tow Spreader

The tow bundle enters the spreader at the throat of a flat expansion section, air enters at the tow outlet and is drawn through holes in the sidewalls of the expansion section into a vacuum manifold. The angle of fiber spread in the pneumatic tow spreader is the result of the force balance between the tow tension applied by the brake and the air drag on the tow fibers due to flow toward the spreader walls. Tow spread is controlled and maintained by adjusting the tension and vacuum pressure.

Powder Deposition

In the powder deposition chamber, the expanded fiber tow behaves like a fibrous filter. Particle collection is by momentum impaction, inception owing to van der Waals forces, Brownian diffusion and in some cases electrostatic force. Theoretical analysis (2) provides the following design relationship for the level of powder deposition on the tow as it passes through the chamber where the tow residence time is

$$(P/[1-P]) = \{\eta_i U[D_p + D_f]\,(N/W_t)\}\,(Ln/U_t) \qquad (3.1)$$

$\theta_c = L/U_t$, P is the weight percent resin in the towpreg, W_t, the clean tow weight per unit length, U_t, the linear tow rate, D_p the particle diameter, U, the gas velocity, ρ_p, the particle density, D_f, the fiber diameter length, N the number of fibers in the tow, L the chamber, n the particle cloud density, and the average fiber collection efficiency $\eta_i = 0.00465$ over a wide range of chamber flow recirculation conditions. The appropriateness of this design relationship for the powder deposition rate was demonstrated for the powders over a wide range of operating conditions, Figure 3, during experiments at LaRC (2,3).

Powder Recirculation

The fluidized bed unit is comprised of two different particle fluidization systems - upflow and downflow. In the external return tube gas flows up through the fluidized powder and conveys it to the top of the fluidized deposition chamber. The powder and gas flow into the chamber, pass through and around the spread tow with some powder being deposited on it, and flow down to the fan inlet at the bottom. The fan accelerates the particles and gas into the external tube to complete the flow cycle.

Beginning at the fan outlet the pressure losses for each section of the powder recirculation system are as follows (4).

<u>Acceleration</u>:

$$\Delta P_1 = (\eta_t + \rho_g)U^2/2g_c \qquad (3.2)$$

where ΔP_1 is the pressure drop on acceleration to velocity U and n_t is the dispersed particle density in the external tube.

The suspension flows from the fan outlet into the tube with a reduction of the flow cross-sectional area. The pressure drop for flow contraction is

$$\Delta P_2 = K(n_t + \rho_g)U^2/2gc \qquad (3.3)$$

where $K = 0.4(1.25 - S_2/S_1)$, S_2 is the tube area and S_1 is the fan outlet area.

Vertical flow:

For solids-to-gas weight rate ratios over 50 the sum of the pressure drops due to friction can be estimated using the correlation for vertical flow.

$$\Delta P_3 = 2.5 L_e n_t U_s^{0.45} (D_p/D_t)^{0.25} \qquad (3.4)$$

The correlation is not dimensionless (4): L_e is the equivalent length of pipe, ft; n_t is the dispersed solids density, lb/ft^3; D_p is the particle diameter, ft; and, D_t is the tube diameter, ft. The solids flow slower than the gas. Solids flow slippage correlations for high particle loadings level off at the limit of one half the gas velocity,

$$U_s = U/2 \qquad (3.5)$$

Flow resistance due to smooth 90° bends may be expressed as $26D_t$ of equivalent pipe length (4) so that for the two bends

$$L_e = L + (2)(26)D_t = L + 52D_t \qquad (3.6)$$

combining equations gives

$$\Delta P_3 = 2.5(L + 52D_t)(n_t + \rho_g)(U/2)^{0.45}(D_p/D_t)^{0.25} \qquad (3.7)$$

Column pressure:

The vertical column pressure difference between that of the tube and the chamber is

$$\Delta P4 = (n_t - n)h \qquad (3.8)$$

where n_t is the particle cloud density in the vertical tube and n the particle cloud density in the deposition chamber.

Chamber flow:

The suspension flows from the tube into the chamber with an expansion of the flow cross-sectional area. The pressure drop for flow area expansion is

$$\Delta P_5 = (n_t + \rho_g)(U^2/2g_c)(1 - S_1/S_2) \qquad (3.9)$$

Friction resistance for flow downward in the deposition chamber is assumed to be negligible compared to the other flow resistances.

Total pressure loss:

The sum of the above pressure losses represents the system resistance to flow and is provided by the fan recirculation. Consideration of the magnitudes of the various terms indicates that flow friction in the pipe and support of the suspended column of solid particles make up over 90% of the pressure drop or work required of the fan.

Cloud Density

The fan horsepower, system total pressure losses, and powder material balance may be used to calculate the cloud density achievable for a specific design at a given set of operating conditions (2). As indicated in equation 3.1, the cloud density is an important factor in establishing the rate of powder deposition on the tow.

The stalling condition of the recirculation system is reached when the air flow in the tube, U, equals the maximum carrying velocity, $U_{g,m}$, at which point powder accumulates in the vertical tube stopping the fan. Operation just below the stalling point provides the maximum particle cloud density.

For upflow of gases and solids in a vertical pipe, the maximum carrying velocity can be estimated using the correlation (4)

$$U_{g,m} = 910\,(\rho_p/[\rho_p + 62.3])D_p^{0.60} \qquad (3.10)$$

The correlation is not dimensionless (4): $U_{g,m}$ is in ft/sec, ρ_p is the density of the solid particles in lb/ft^3, and D_p is the particle diameter in ft.

Powder-Tow Fusion

Towpreg flexibility and powder-fiber fusion are important for weaving and molding applications. These properties of the towpreg depend upon the temperature of the oven and the time that the powder laden tow takes to travel through the oven. Flexural rigidity data were obtained for towpreg having a range of resin content and fused at several different oven temperatures and residences times. The standardized cantilever test, ASTM D 1388-64, for fabrics, was used to determine the flexural rigidity of towpreg samples (5).

Quality Control

An instrument for on-line continuous detection of the towpreg resin content was developed in conjunction with Analytical Services and Materials, Inc. of Hampton, VA. Towpreg is composed of electrically conducting carbon fibers and dielectric polymer resin. The resin level monitor measures the electric capacitance of the towpreg, which is a function of its resin content.

Once the system design has been selected, the operating variables are used to achieve towpreg quality control during production. The process outputs to be maintained, within certain setpoints, by the control system are towpreg resin content, powder fusion, and flexibility. During continuous operation, regulatory control over output variances is accomplished by adjusting the tow speed, powder feed rate, and oven temperature.

COST ESTIMATION

A preliminary estimate of the cost to make towpreg by the dry powder process was made based on the cost of the bench scale equipment, Table II. There are no multi-tow dry powder units upon which to base cost projections for commercial operation. Assuming that the process can be scaled up to the 25 tow level in 5-tow units, projections of the cost of such a system were made and are presented in Tables II and III.

The impact of tow and resin costs are reflected in Table III and Figure 4. The powder impregnation process alone costs approximately $9.30 per pound of towpreg. Grinding solid resin to powder costs about $3.00 per pound. The cost of towpreg is the cost of the resin and tow plus the cost of milling the resin into powder and impregnating the fiber tow.

Towpreg may be stitched into uniweave tape/sheet or woven and braided into broadgoods and preforms. The cost to weave towpreg yarn in an 8 harness satin, 40 inches wide, is approximately $10.00 per pound. These estimates suggest that the costs incurred in producing powder coated towpreg should be comparable to, and perhaps even less than, those incurred for producing conventional hot melt prepreg.

RESULTS AND DISCUSSION

Extensive testing of the bench scale dry towpreg system over a range of conditions has confirmed design theory and operating correlations for each component and provides the basis for scale up to produce commercial quantities of towpreg. The unit operated reliably with the various powders, using both 3K and 12K tows, for periods as long as eight hours, at tow speeds as high as 50 cm/sec (2).

Test specimens of both unidirectional fiber towpreg and woven towpreg were prepared and molded for testing, Table IV. Weaving experiments indicate that the towpreg used as fill material must have a flexural rigidity below 10,000 mg-cm so that it bends and follows the shuttle action without breaking. Towpreg used as beam material must have a flexural rigidity below 100,000 mg-cm so that it does not break during heddle and comb action, and it must bind together the fibers so that they do not come loose, resulting in material thinning and comb clogging. This last condition requires using resin fusion to bind the unidirectional tow fibers in the beam material.

CONCLUDING REMARKS

Significant progress has been made on the development of the LaRC dry powder towpreg system. Polymer powders can be deposited on a moving carbon fiber tow in a recirculating fluidized bed and then fused to the tow using radiant heating. The flexibility of the resulting towpreg may be controlled by adjusting the temperature and time of the oven fusion process such that weavable towpreg can be obtained. Unidirectional and woven test samples, using both thermoplastics and thermosets, produced by the process have been of good quality.

Design information and operating data correlations have been developed for scale up to commercial operation. Cost estimates suggest that processing cost are comparable to those of conventional hot melt prepreg. In the future, from a part fabrication point of view, powder coated prepreg tape, woven broad goods and woven and braided preforms may be considered as options to similar materials made by other methods.

ACKNOWLEDGMENT

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TABLE I.- POWDER PREPREG PRECURSOR DATA*

Polymer Description	Particle Size, μ	Supply Source
LARC-TPI 2000	7.0	Mitsui Toatsu Chemical, Inc.
LARC-TPI 1500	19.0	Mitsui Toatsu Chemical, Inc.
PEEK 150	17.0	ICI Fiberite
PMR-15 PI	1.5	Dexter-Hysol Aerospace
PR500 Epoxy	19.0	3M Company
Fluorene Epoxy	3.0 (12.0)**	3M Company

*All prepreg utilized unsized Hercules AS-4 graphite fibers in 3K and 12K tows

**The fluorene epoxy was supplied in two different particle sizes

TABLE II.- TOWPREG COST ESTIMATION

	Dollars
Laboratory Single Tow System Equipment Cost	
Feed Spool Holder and Tow Tension Brake	1,500
Pneumatic Spreader and Vacuum System	2,000
Powder Screw Feeder	6,000
Powder Deposition Chamber and Recirculation Fan	2,000
Electric Oven	1,500
Resin Level Sensor and Controller	5,000
Traversing Takeup Spool and Tow Speed Control	6,000
Subtotal	24,000
Installation @ 25% of equipment cost	6,000
Total Cost	30,000
25 Tow System Equipment Cost Estimate*	
Feed Spool Creel and Tension Brakes	7,500
Pneumatic Spreaders and Vacuum Systems	10,000
Powder Screw Feeders	30,000
Powder Deposition Chambers and Recirculation Fans	10,000
Electric Ovens	7,500
Resin Level Sensors and Controllers	25,000
Takeup Spools and Speed Controls**	60,000
Subtotal	150,000
Installation @ 25% of equipment cost	37,500
Total Cost	187,500

*No multi-tow information is available. The above estimate assumes that the 25 tow system is made up of 5 deposition chambers, each handling 5 tows, complete with a pneumatic tow spreader and feeder. There are 5 resin level sensors, one for each chamber. The creel, ovens, and takeup system may be single units or multiple units.

**Estimate is for 25 spools with traversing mechanisms and speed control.

TABLE III.- PROJECTED ANNUAL COSTS*

	Dollars/Year	
	Epoxy	LARC-TPI
Direct Costs		
Equipment (3 year life)	62,500	62,500
Space (1,000 sq ft @ 10$/yr sq ft)	10,000	10,000
Utilities (20,000 KWH/yr @ $.15/KWH)	3,000	3,000
Personnel (1 FTE + benefits)	40,000	40,000
Tow (13,300 lb @ 20$/lb)	266,000	266,000
Resin (7,200 lb; Epoxy @ 20$/lb;	144,000	
LARC-TPI @ 150 $/lb		1,080,000
Subtotal	525,500	1,461,500
Indirect Costs @ 50%	262,750	750,750
Total Annual Cost	788,250	2,192,250

Estimated Towpreg Production Costs

The annual production rate is 8,500 kg or 18,600 pounds. With the above annual costs the cost to produce towpreg is estimated to be:

Epoxy/carbon fiber	42.38 $/lb
LARC-TPI/carbon fiber	117.86 $/lb

*Assumptions

The 25 tow system operates at 25 cm/sec or (50 ft/min) tow speed Standard 1.0 kg (2.2 lb) spools of 3K tow are used. The spool tow is 15,600 ft long giving a running time of 5.2 hours.

Allowing for loading, unloading, and maintenance it was felt that this level of operation would require one full-time technician.

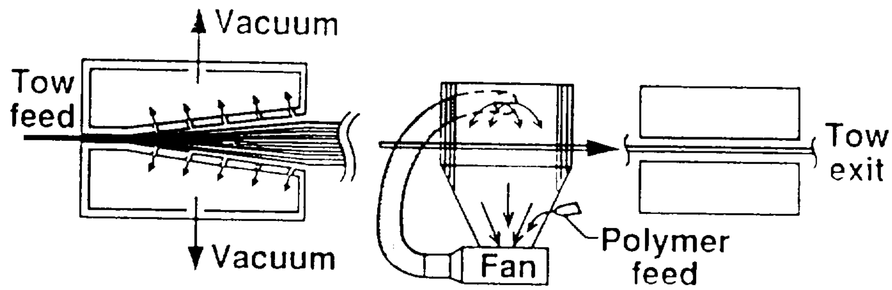
At this production rate 26 million meters (85 million feet) of 3K towpreg would be produced per year. For 35 wt % resin this amounts to 8,500 kg per year of towpreg (18,600 lb). Materials consumption, assuming 10% waste, would be 6,000 kg of tow per year (13,300 lb/yr) and 3,300 kg of resin per year (7,200 lb/yr).

Equipment life is taken to be 3 years using linear depreciation and no scrap value. With storage and handling access the system would occupy 1,000 sq ft. The major utility required is electricity for the motors and ovens estimated at 20,000 KWH/yr.

TABLE IV.- MECHANICAL PROPERTIES OF DRY POWDER COATED COMPOSITES

Composite- AS-4 Carbon Fiber-	Short Beam Shear strength ksi (MPa)	Flexure Strength ksi (MPa)	Flexure Modulus msi (GPa)
12K Unidirectional LARC-TPI 2000*	12.3 ± 1.3 (848 ± 8.9)	323 ± 14.1 (2227 ± 97)	19.4 (134)
3K 0/90 4-Harness woven, LARC-TPI 1500**, Specimen:			
0° Beam		134 ± 3.5 (924 ± 24)	8.1 (56)
0° Fill		83 ± 4.0 (572 ± 28)	5.7 (39)
3K Unidirectional PR 500***	14.5 ± 0.4 (100 ± 2.8)	206 ± 11.1 (1419 ± 76)	16.4 (113)
3K Unidirectional fluorene-based epoxy****	11.8 ± 0.6 (81.3 ± 4.1)	240 ± 15.8 (1654 ± 109)	15.4 (106)

	molding conditions	resin content
*	1 hr/660°F/800 psi	33.5 wt %
**	1 hr/700°F/300 psi	29 wt %
***	4 hr/350°F/85 psi	32.5 wt %
****	4 hr/350°F/85 psi	35 wt %



Prepreg Processing Modules

Tow spreader chamber

- Adjustable tow spread
- Minimum fiber damage
- Fast line speed

Powder coater

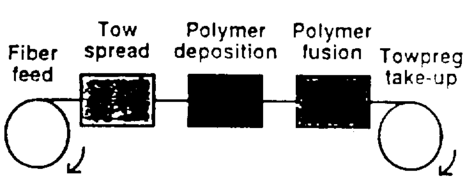
- Recirculating chamber
- Uniform coating
- Precise resin control

Powder fusion

- Convection furnace
- Individual fiber coating

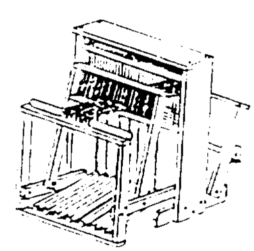
FIGURE 1. DRY POWDER PREPREGGING

Dry powder prepreg



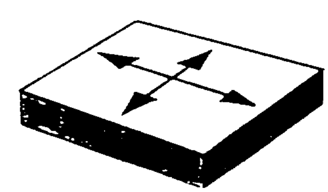
- Stable, adjustable tow spread
- Uniform polymer application/ distribution
- Continuous polymer mass fraction monitor/control
- Rapid towpreg production speeds

Powder prepreg weaving



- Towpreg weavibility established
- Towpreg drape limits determined
- 1,3 twill fabric preform woven

Composite evaluation



- 8-ply woven cloth laminated fabricated
- Excellent C-scan results
- Excellent flexural strength

FIGURE 2. INNOVATIVE PREPREG RESEARCH

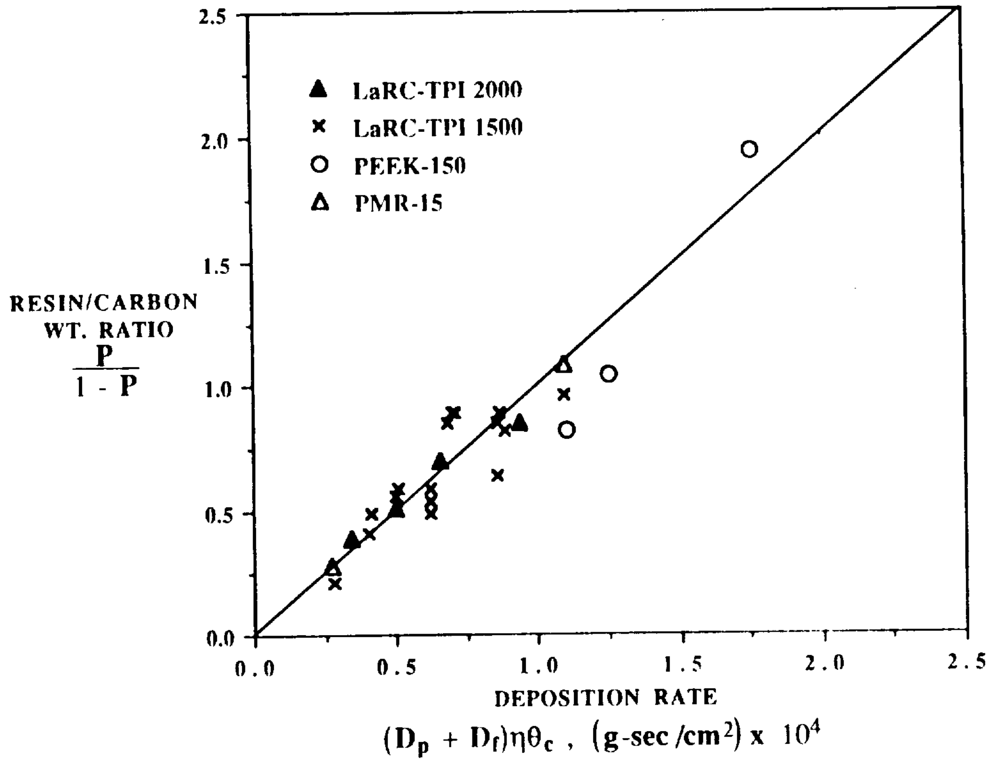


FIGURE 3. POWDER DEPOSITION DATA CORRELATION

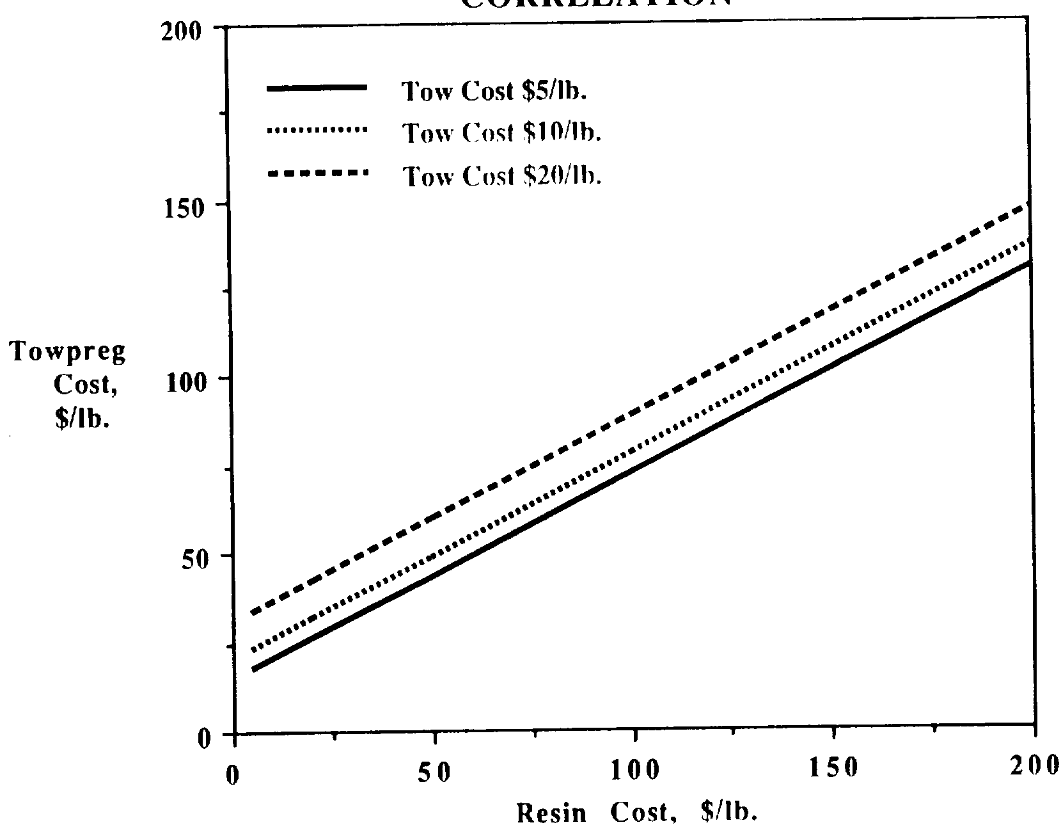


FIGURE 4. TOWPREG COST ESTIMATION