4. POLYMER COMPOSITES R&D

A. Development of Manufacturing Methods for Fiber Preforms

Principal Investigator: Jeff Dahl
Ford Motor Company, Research and Innovation Center
P.O. Box 2053, MD 3135 RIC, Dearborn, MI 48121-2053
(313) 845-1039; fax: (313) 390-0514; e-mail: jdahl@ford.com

Project Manager: C. David Warren
Oak Ridge National Laboratory
P.O. Box 2009, Oak Ridge, TN 37831-8050
(865) 574-5069; fax: (865) 574-0740; e-mail: warrencd@ornl.gov

Technology Area Development Manager: Joseph A. Carpenter
(202) 586-1022; fax: (202) 586-1600; e-mail: joseph.carpenter@ee.doe.gov
Field Technical Manager: Philip S. Sklad
(865) 574-5069; fax: (865) 576-4963; e-mail: skladps@ornl.gov

Contractor: U.S. Automotive Materials Partnership
Contract No.: FC26-02OR22910

Objective

- Develop and demonstrate new fiber preforming processes to decrease cost, increase manufacturing rates and improve reproducibility of large preforms for composite molding.
- Provide process development support to the Automotive Composites Consortium's (ACC) Focal Project-3.

Approach

- Identify carbon-fiber properties required to permit rapid processing and achieve desired performance levels.
- Investigate materials, process equipment, and tooling technology to further reduce the cost and enhance the quality of chopped-fiber performs.
- Characterize chopped-fiber preforms utilizing experimental carbon-fiber rovings to determine important characteristics (permeability, light transmission, areal density distribution, etc.).
- Explore the extension of automated preforming technology to make preforms with a thermoplastic matrix.

Accomplishments

- Toho Tenax carbon-fiber roving development program progressed with successful direct splitting of carbon-fiber rovings.
- Preforming process development conducted on the Focal Project 3 B-pillar test section.
- Initiated preliminary thermoplastic P4 (TP-P4) processing studies

Future Direction

- Experimental preforming and molding evaluation of mechanically-split, low-filament-count Toho Tenax carbon-fiber rovings.
- Further characterization and investigation of low-cost, carbon-fiber rovings with reduced individual bundle size.
- Investigate the use of thermoplastic-matrix materials in the P4 process and develop the TP-P4 process concept.

Introduction

This project has focused on the development of the P4 process, a fully-automated robotic preforming process. A prototype, two-station manufacturing cell was designed, fabricated and installed at the National Composite Center (NCC) in Kettering, Ohio. This equipment is currently being utilized to support preforming and material development efforts within the Automotive Composites Consortium (ACC).

In order to obtain higher mass savings with composites relative to steel (50-70%), carbon fiber must be utilized as the reinforcing fiber. The extension of this preforming technology to manufacture carbon-fiber preforms is now in progress to support the development of ultra-light weight vehicle structures. A major focus of this program is the development of a carbon-fiber material suitable for chopped-fiber processes. Current production carbon-fiber materials are not amenable. Advances in carbon-fiber roving and preforming technology will be demonstrated in the structural automotive parts designed and prototyped as part of the ACC's Focal Project 3 (see report 4.D).

Preliminary investigation to determine the feasibility of using a modified P4 process (i.e., TP-P4) to generate thermoplastic/glass fiber preforms and blanks for compression molding was initiated.

Carbon-Fiber Roving Development

To date, chopped carbon-fiber preforming research and development has been limited due to material format issues. In order to address fundamental material format issues with off-the-shelf carbon fibers, development programs have been initiated with two carbon-fiber manufacturers (Hexcel and Toho) to expedite material research. The focus of these programs is to investigate the technology required to achieve a reduction in bundle size, to determine the effects of bundle size in the preforming/molding processes and to ascertain the effect on composite material performance.

Hexcel Carbon Fibers

A carbon-fiber roving development program to investigate the effect of individual bundle size on preforming, molding and composite material performance using Hexcel carbon fibers is nearly completed. The development effort focused on assessing the effect of individual bundle size on P4 preforming, structural reaction injection molding (SRIM), and the resultant composite material performance. It was theorized that a reduction in bundle size would improve material distribution in the preforming process and, therefore, positively impact the mechanical performance of molded structures. Based upon the data gathered to date, composite material mechanical performance was positively impacted by a reduction in bundle size.

Evaluation of the carbon-fiber rovings is nearly complete with preform permeability evaluation remaining. Remaining preform fabrication includes permeability samples, both in-plane and through the thickness. Upon completion of preform permeability test equipment at ORNL, in-plane and through-thickness permeability measurements will be conducted using preforms fabricated with each of the seven carbon-fiber rovings previously manufactured.

Toho Tenax Carbon Fibers

A research program to develop carbon-fiber rovings more amenable to the P4 preforming process was originally developed and initiated with Fortafil Fibers. Toho Carbon Fibers acquired Fortafil on September 1, 2004; therefore, the program is now officially with Toho Carbon Fibers. This program focuses on developing cost-effective methods to reduce the individual carbon-fiber bundle sizes within a roving by using relatively low-cost, 80k carbon fiber as the input material. A carbon-fiber roving specification was developed jointly between the ACC and Fortafil as the target material for this program. The overall program is comprised of five phases.

Toho Tenax CF Phase 1: The first phase of the work was to investigate whether or not splitting the precursor could give a final carbonized tow with fiber bundles of the desired dimensions.

The major effort at Toho was focused on evaluating precursor samples produced by spinning small-filament-count strands through spinnerets selectively blanked to give the desired number of filaments. In total, four samples were evaluated at Toho. The first three contained 42 strands of 2,000 filaments each; the last sample contained only 28 strands but was spun with greater separation between the spinneret hole sections. All samples were spun as 1.5 denier/filament (dpf) and grouped into bundles containing 167k total filaments. The progression to larger total filament count and strand number in the course of this work was necessitated by the requirement to spin more dope through the spinneret to stabilize the spinning process. All samples were crimped and packaged in the normal way.

Two of the samples were oxidized on Toho's pilot line and then carbonized and sized on their production line. The precursor visually appeared to be more segmented than standard precursor; however, once the material was tensioned in oxidation, the strands were not easily distinguishable. While the final carbon-fiber product does have a separated appearance, cross filaments are plentiful and some of the strands appear to have consolidated to form larger strands.

The last of three precursor samples produced was also converted into carbon fiber at Toho. This particular sample contained 28 individual strands of 2,000 filaments each. The tow was carbonized and sized with the same polyurethane size used in the ongoing carbon slitting effort (see Phase 2). Because the tow stiffness was enhanced by the size, separating the strands in the tow was even more difficult than for the two similar tows converted and sized with epoxy.

The precursor-splitting trials generally demonstrated that the use of divider pins or grooved bars is feasible to keep strands separate in the precursor state during the spinning process attributable to the toughness of the polymer and the larger filament diameter. However, the use of the same techniques in the carbon conversion process will likely cause excessive filament damage once the polyacrylonitrile (PAN) fiber is converted to a brittle material of much smaller diameter. The implication of the results to date was that the strands must either be maintained as separate entities until sizing application and drying or created at the point of size application and maintained separate through drying, as is done in the carbon slitting process.

The results of these trials were conveyed to precursor manufacturing personnel for final consideration. The decision to terminate this development approach was made after discussing the technical problems with personnel at the precursor manufacturing facility. In the final analysis, the precursor manufacturing team was unable to introduce entanglement or twists in the individual strands of a tow, and carbon-fiber manufacturing personnel were negative on the feasibility of maintaining separation of the strands in the carbon conversion process without causing excessive filament damage. Development activities to produce a stranded roving by modifying the form of the precursor were subsequently discontinued.

Toho Tenax CF Phase 2: The second phase of the project involves direct mechanical slitting of carbon fiber, focusing, in particular, on what happens at the point of subdivision of the tow into strands.

Various types and configurations of slitter rolls were evaluated. The most effective slitting configuration achieved to date uses a single roll, eccentric in shape, with flat bottom grooves, and rotated counter to the direction of the moving tow faster than the absolute speed of the fiber (Figure 1). The eccentric design of the roll allows cross filaments to move between grooves in a gradual way to avoid a build-up of filaments that would otherwise cause stoppage of the operation. The use of multiple rolls and various groove geometries was also evaluated. Additional rolls do not enhance the slitting effectiveness, however.

Critical to the consolidation of the strands after their creation at the surface of the slitter roll is the

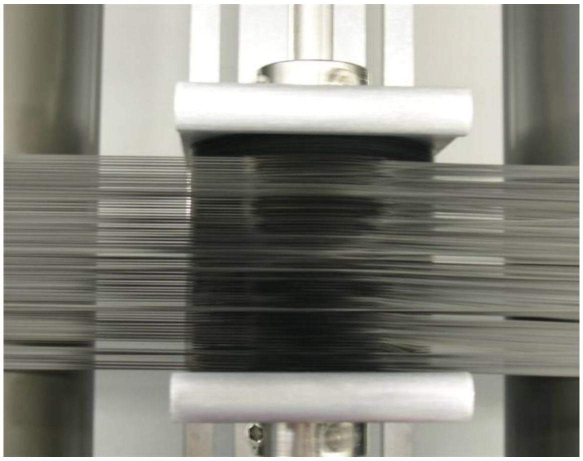

Figure 1. Prototype tow-splitting device.

application of a water-based size. Initial slitting trials used a hand sprayer to apply water directly onto the fiber and slitting roll, but techniques have since been developed to apply the size prior to the slitting roll by drip or kiss-roll application.

Upon completion of the above tasks, lab-scale production of three roving samples was conducted for material evaluations in the P4 process. Carbon-fiber rovings with nominal strand counts of 25, 35, and 50, were produced on the lab-scale slitting line (Figure 2). All of the rovings were produced from 80k carbon tows having a TEX value of approximately 4380. The rovings were sized with the same commercially-available, water-based polyurethane used for all of the carbon slitting trials to date. Some minor fuzzing was encountered in the production of the strands resulting in the formation of small fuzz balls, most of which were manually removed. It is envisioned that in a commercial production operation, such fuzz agglomerations would be removed by patented production technology already owned by Toho Carbon Fibers.

Figure 2. Prototype tow-splitting line.

Following receipt of the materials, a processing evaluation of the three rovings (25, 35 and 50 split) was conducted on the ACC's P4 preforming machine. These panels were then molded via SRIM and characterized to determine the mechanical properties of the composite. Five kilograms of fiber with each of three split configurations (25, 35, and 50 splits, yielding about 1.5 k, 2.5k, and 3k strands) were evaluated by the ACC. Preforming of the fibers was acceptable for an initial evaluation, although further sizing work is needed to deal with fiber handling issues. The results of mechanical testing from this preforming and molding trial are shown in Table 1.

Table 1. Mechanical Properties of Toho Tenax split-tow carbon-fiber composite.

	Tensile		Compression
	Strength MPa	Modulus GPa	Strength MPa
Control 12x3k	207	33.4	247
Toho 50x1.5k	194	29.7	250
Toho 35x2.5k	190	32.2	298
Toho 25x3k	190	32.5	297

Mechanical properties are similar to the control, but do not show an improvement with fiber bundle size. Since other work in the ACC has shown the advantage of smaller fiber bundles, this indicates a need for further sizing development to enhance the properties of the random chopped composite.

Toho Tenax CF Phase 3: This phase calls for the development of appropriate fiber sizings for improved handling and fiber/matrix interface with a polyurethane resin system. This has been initiated in conjunction with the processing studies on the fiber-slitting line, and will be continued by Toho research labs in Japan.

Toho Tenax CF Phases 4& 5: The fourth phase is acquisition of a pilot-line for fiber production. The actual extent of the pilot-line is still under discussion with Toho management, and the fifth phase, the optimization of the pilot-line, production of fibers for ACC evaluation, and assessment of manufacturing costs, will be determined after the results of that discussion are clear.

The program has an anticipated completion date of September 30, 2006. The date has been delayed from the initial schedule by a combination of factors, including the lack of success of the precursor-slitting trials and the acquisition by Toho Tenax. Continuation of this project is currently under review by Toho Tenax management.

Thermoplastic P4 (TP-P4)

Preliminary Processing Studies: To prove out the process concept, an initial series of experimental studies was conducted to demonstrate deposition of polymer matrix materials and reinforcement fibers. In total, 80 blanks were manufactured under a variety of conditions to test deposition reliability and throughput capability. The key technical hurdles, related to fiber and matrix deposition, are considered to be:

- Verification of throughput capability for 45s production cycle times.
- Demonstration of control over fiber distribution.
- Demonstration of process robustness for continuous, uninterrupted operation.

During the concept feasibility trials, attention was focused on material deposition throughput. At present, the TP-P4 processing equipment has shown capability for fiber and polymer deposition, but only at reduced output. This limitation is a function of the equipment design, which was originally commissioned for a previous project. Hence, increased material output will require a re-design of equipment. An example of the material blank that is produced using the current development cell is shown in Figure 3.

As the photograph shows, random deposition of the fibers and matrix is achieved creating a blank similar to conventional glass mat thermoplastic (GMT). Overall, the process concept feasibility studies were considered a success, although the remaining technical hurdles in the list above need to be addressed, and are the subject of further trials scheduled in 2006. The key issue, however, remains throughput capability. Figure 4 shows material throughput requirements if the process were applied

Figure 3. TP-P4 chopped-fiber blank.

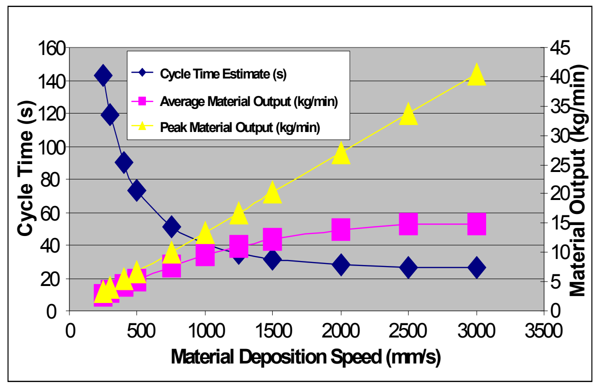

Figure 4. Material deposition vs. production cycle time.

to the production of a vehicle liftgate inner panel. In this example, the deposition speed in the TP-P4 cell is chosen in order to meet the specified annual production volume. For instance, at a high production volume of 100k units per year or more, a cycle time of 40s is desirable to prevent the need for a second manufacturing cell. Based upon these constraints, an average deposition rate of 10kg/min would be required, with a peak deposition capability of 15kg/min. The current TP-P4 is operating at about 10% of this capability. Hence, a parallel effort is focused on the design of new deposition equipment, capable of meeting the target throughput requirements. Fabrication and testing of the new equipment will commence if justified by the results of an economic evaluation of the TP-P4 process technology.

Assessment of Pre-Consolidation Methods: Following material deposition and stabilization, the requirement for a further processing step is anticipated in order to promote wetting of the fiber reinforcement by the polymer matrix. Hence, a variety of techniques are under investigation to determine the most cost-effective processing route. These include double-belt lamination, hot-air or infrared heating, and batch thermal cycling of matched tooling.

The function of the pre-consolidation method within the TP-P4 process is placed into context using Figure 5. In this chart, the material porosity is shown at different stages of processing. Since porosity has a direct impact on final part properties, it is important to track this parameter and determine the effect of processing on the final outcome. When determining the right choice of pre-consolidation method, two factors need to be considered. Firstly, a high level of porosity may be tolerated if a near-zero value can be achieved during the compression molding process. Secondly, the type of porosity, whether within or between fiber bundles (Figure 6), will have an effect on what is manageable during processing. Hence, it may not be required to drive down the bulk porosity to 2% or 3% during consolidation. Instead, the overall process needs to be considered, and the consolidation route selected, based upon process economics. Regarding the latter, results from consolidation studies, which are ongoing, will be used to specify equipment requirements for full-scale manufacture. Equipment pricing will subsequently be used as input into future cost analysis. The results of this analysis will be presented in a future report.

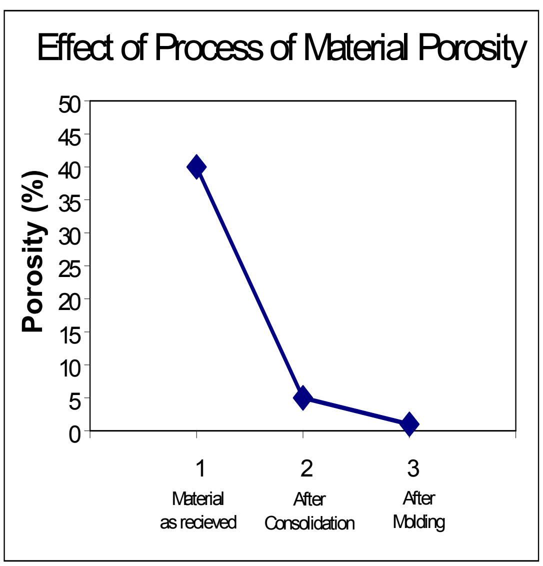

Figure 5. The effect of processing on material porosity.

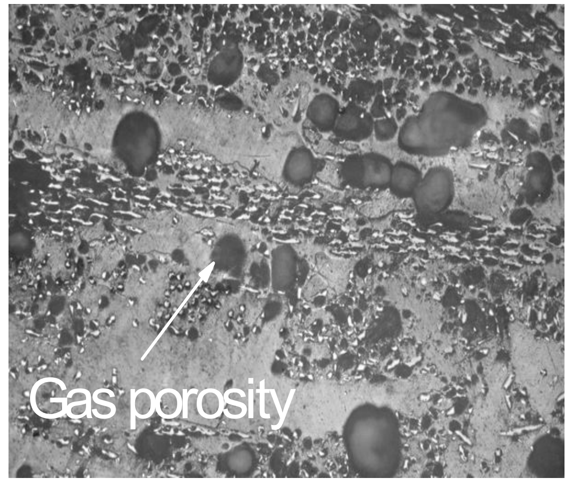

Figure 6. Porosity distribution within TP-P4 materials following pre-consolidation.

Economic Analysis: In parallel to process development studies, an analysis of the TP-P4 process has commenced to understand the process economics. The work is divided into three distinct phases.

Phase 1: Preliminary cost analysis based upon assumptions of TP-P4 capability
Phase 2: Experimental studies to corroborate process/equipment assumptions from Phase 1.
Phase 3: Refine cost model and update results based upon studies conducted in Phase 2.

During the first phase of the project, a baseline cost model was created for process technologies that are direct competitors to the TP-P4 concept. These models are now established and will provide a realistic assessment of the cost savings forecast using the new technology. In order to execute a comparative analysis, two target components have been selected as the focus of case studies (a SUV liftgate inner panel and a rear seat-back structure). To date, materials and process data have been gathered for input into the cost model. Outstanding data required to enable the first analysis are being sought with a first series of cost analysis results due by Nov 30th 2005.

TP-P4 Summary: A new, high-volume process technology for the manufacture of thermoplastic composite components is currently under development. The project, which received ACC Board of Directors approval in Q2 2005, commenced by using existing and modified process equipment to prove out initial concept feasibility. Preliminary process studies have demonstrated manufacturing capability and further process investigations are ongoing to improve production robustness and throughput capability. Further scale up of R&D activities will ultimately be contingent upon a positive economic assessment. Preliminary results of the process cost analysis are due by Nov 30th 2005.

Focal Project 3 B-Pillar Preforming

In support of Focal Project 3 (FP3), preforming process development has been conducted to facilitate manufacture of B-Pillar Inner and Outer preforms. Preforming development efforts were performed using the revised B-pillar preforming tooling. In the revised tooling, the 'B' surface is the preform deposition surface and the 'A' surface is the consolidation surface. Although the preform and molding tool compatibility issue has been addressed with the revised preform tooling, the inverse orientation of the deposition surface has created additional issues during the material deposition process.

In an attempt to resolve these issues, it was determined that baffles would be constructed and utilized on the underside of the preform screen. Baffling was constructed by trimming previously-molded B-pillars so that only the flanges and a section of the walls adjacent to the flanges remained. The baffling was installed on the underside of the tooling by fastening it to the existing screen every 250 mm (Figure 7). The baffling was initially installed flush against the screen so that airflow was essentially blocked in the 1.5 mm flanges and lower section of the vertical regions. However, some air flow did occur in these regions, but at much lower values.

Air flow rates were significantly reduced with the baffles installed, primarily in the 1.5 mm flange regions by as much as ten times based upon average anemometer readings (Figure 8). Air flow measurements were recorded as averages taken over ten seconds, with the anemometer held at approximately the same angle and spacing each time.

Figure 7. Molded Baffling Installed on Underside of Preforming Tool

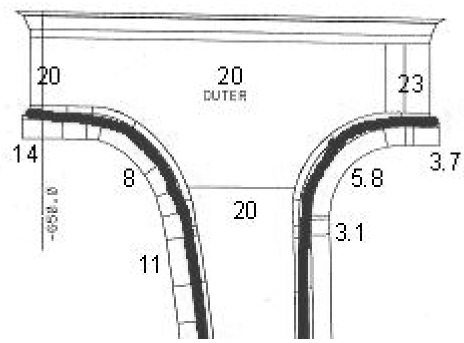

Figure 8. Air flow rates on outer B-pillar with baffles installed (the 2 dark areas).

High air flow rates on the unbaffled tooling (Figure 9) combined with the orientation of flanges relative to thicker regions, encouraged fibers to congregate in the flanges while being chopped. This resulted in excess thickness in the flanges and made it difficult to fill the outside radii with proper fiber amounts. Areal density sampling data indicated regions exceeding 100% by volume for the target fiber volume fraction of 40% at a 1.5 mm thickness. Large areal density variability within the preform has led to subsequent molding issues including fiber-wash and dry regions in the parts. Excessive material density creates "dry spots" in the molded components. Also, fiber crush existed primarily in

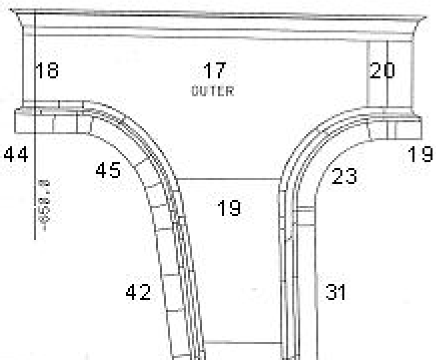

Figure 9. Air-flow rates on outer B-pillar without baffles.

flanges adjacent to thicker regions on the bottom 6 and 8 mm regions of outer and inner B-pillars, respectively.

After inspecting molded B-pillars, it was evident that the air baffles had positively affected preforming optimization efforts with respect to fiber density and dispersion. However, issues still remained such as high fiber density in the 1.5 mm flanges. Adjustments were made to the spacing of the baffles by placing flat washers (~1.5 mm thickness) between the baffles and preform screen. Several spacing ranges were evaluated during preforming. Optimal spacing, and, hence, optimum air flow values were achieved via a 3.0 mm spacing.

Several preforming process variables were also altered in an attempt to optimize the B-pillar preforming along with the air flow being optimized via baffling. Different fiber lengths (12.5, 15, 25, 30, 50, and 100 mm) were utilized throughout different locations. Tool Center Points (TCP) were modified to place the chopper gun closer to the screen in certain regions. Additionally, fiber output, robot speed, air ejector pressure, screen air flow, and robotic positions were modified to varying degrees in an attempt to optimize B-pillar preforms.

Minimal, positive results were realized through modifications to the above listed preforming process variables. Problems still exist with the quantity of fibers that can be placed in the flanges sections (1.5 mm) and radii relative to the amount that can be placed in the thicker regions. When fibers are placed in the flanges at or near the target fiber volume fraction (40%), very low fiber volume fractions will exist in the adjacent radii and thicker regions. However, if fibers are placed in the thicker regions and radii at the target fiber volume fraction, excess fibers will exist in the flanges that are adjacent to these regions.

The combination of near-target fiber volume fraction (40%) in the flanges and slightly reduced fiber volume fraction (30-35%) in the adjacent thicker regions seems to be the best compromise. However, this presents problems in the molding stage of manufacturing, such as fiber-wash, race-tracking, areas of very low fiber density, and areas of excess fiber density leading to fiber crush. Overall, there appears to be more compromise with the revised tooling than with the original tooling, despite baffling being installed on the revised tooling. This is due to the location of the flanges relative to the thicker regions combined with the tooling surface proximity to the flanges, which increases the air flow rates in the region and causes fibers to congregate into the flanges. Also, the flanges are located below the thicker regions, allowing gravity to have a greater effect on the congregation of fibers in the flanges. Optimization efforts are ongoing in an attempt to resolve these issues.