F. Carbon-Fiber Systems Integration

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Objectives

- Develop a subscale, modular test facility for demonstrating advanced carbon-fiber conversion technology.
- Integrate advanced-technology conversion modules into a functional and reliable system.
- Validate system process models and system inputs into cost analyses.
- Provide a capability for developing and testing conversion protocols for new polymer precursors.
- Provide the capability to produce small quantities of finished carbon fiber.
- Demonstrate advanced conversion technology to prospective commercialization partners.

Approach

- Procure and install a subscale, conventional carbon-fiber pilot-line.
- Construct a prototypical advanced-technology carbon-fiber line using equipment and process specifications developed in other carbon-fiber conversion projects.
- Locate the advanced technology and conventional lines adjacent to each other, highly instrument them for process characterization, and equip them with versatile fiber transport equipment.
- Develop a small tube-furnace system for developing and evaluating conversion processes for prospective precursor fibers available in quantities of a few grams or less.

Accomplishments

- Completed acceptance testing for conventional pilot-line.
- Measured conventional pilot-line emissions and determined necessary emission controls/containment for sustained operation.
- Completed partial upgrade of conventional pilot-line, constructed some equipment for future installation and designed carbon-proofing and automation systems.
- Completed installation and commenced experimental operation of microwave-assisted-plasma (MAP)-carbonization advanced-technology module.
- Commenced design and specification of a precursor evaluation system.

Future Direction

- Complete conventional pilot-line upgrades and validation, and update safety basis for sustained operation.
- Add capability to develop and evaluate conversion processes for precursor fibers available in gram quantities; evaluate alternative precursors and develop conversion protocols for selected precursors.
- Procure, install, and commission other advanced-technology modules when the processes and equipment designs are sufficiently mature.
- Perform precursor studies and parametric process studies.
- Investigate and demonstrate stable operating envelope, as well as system reliability, availability, and maintainability.
- Develop partnership(s) to commercialize the technology.

Introduction

The purpose of this project is to integrate advanced carbon-fiber technology developed in the ALM efforts. The project encompasses the acquisition and operation of conventional conversion technology for benchmarking and validation; integration of advanced-technology conversion modules into a functional system; and developing and/or evaluating conversion protocols for prospective precursors.

A small pilot facility will be designed specifically for processing tows of commercial-grade (large-tow) fiber, but will be sufficiently versatile that it can potentially process some higher-strength fibers. It will be designed to process a variety of polymer precursors; it may be adaptable to pitch precursors (this would require equipment and process modifications), but that is not a project requirement.

The heart of the pilot-scale conversion facility will be two adjacent, subscale, carbon-fiber conversion lines. One line will be based on conventional pre-treatment, conversion, and post-treatment processes and the other will embody advanced-technology processes. The facility will be highly instrumented for characterizing fiber properties and process parameters. It will also be highly modular and will be equipped with versatile tow-transport equipment so that tows can be routed through any combination of conventional- and advanced-technology conversion, pre-treatment, or post-treatment modules. The system-integration line will be capable of running a single tow or any number of tows up to its maximum capacity, which will be at least five 50k tows. The integration line may eventually be mated with upstream or downstream processes. The facility is shown schematically in Figure 1. Essential advanced-technology modules are stabilization, oxidation, and carbonization. Optional advanced-technology modules include graphitization, surface treatment, advanced instrumentation and control, and downstream processing. The integration line will use conventional pre-treatment, sizing, and fiber-transport equipment.

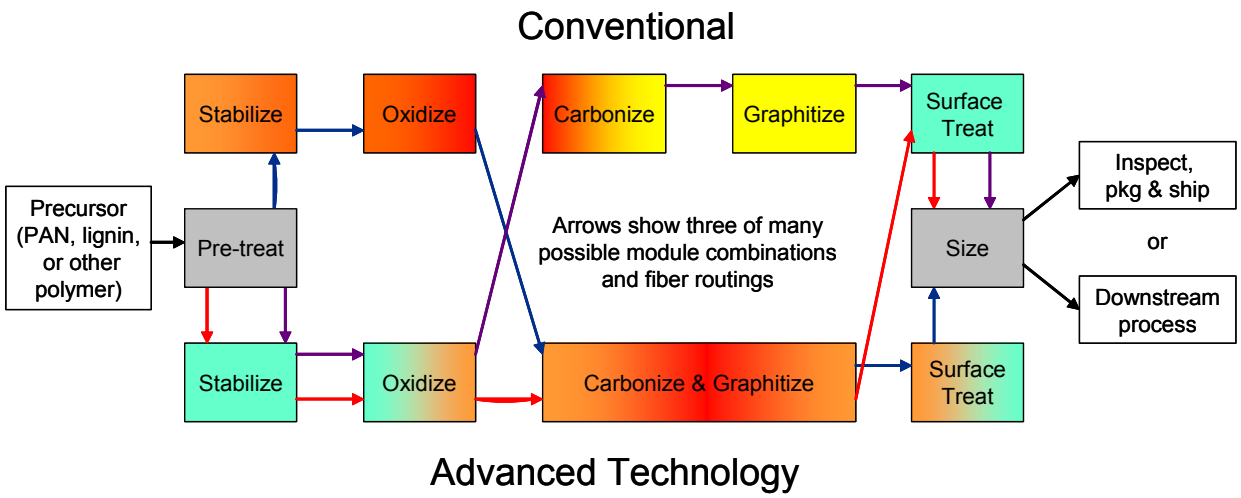

Figure 1. Schematic of carbon-fiber pilot-lines.

New precursor-development projects initially produce much smaller quantities of precursor fibers than are required by the aforementioned pilot- lines. Therefore, a tube-furnace system will be assembled for evaluating small quantities of prospective precursors and developing their conversion protocols.

Project Deliverables

At the end of this project, advanced-technology modules will have been assembled into a functional, prototypical, carbon-fiber conversion line with 5 – 10 large-tow capacity, and alternative precursor-conversion protocols will have been developed and/or evaluated. The following tasks will have been completed:

- confirm that advanced-technology conversion modules work together as a functional system;
- determine interfaces to optimize system
- cost and performance;
- determine and/or characterize system processing envelope, utility requirements, effluent streams, etc.;
- demonstrate system process control, stability, repeatability, scalability, safety, and reliability/availability/maintainability;
- investigate system fault tolerance and response;
- evaluate alternative precursor fibers and
- develop conversion protocols for selected precursors;
- scale alternative precursor tow sizes and validate their conversion at the pilot-scale; and
- validate that cost and technical targets were achieved.

Completion of these tasks should sufficiently demonstrate the advanced-technology's value, robustness, and scalability to convince an industrial partner to commercialize it.

Current Status

Conventional Pilot-Line

In fiscal year (FY) 2006, the conventional pilot-line was cleaned, serviced, and operationally tested. Acceptance test data are shown in Table 1. Acceptance data were slightly lower than the commercial-fiber baseline, but were considered quite good for a short first run; therefore, ORNL accepted ownership of the equipment. Figure 2 shows the conventional pilot-line installed at ORNL.

The previous owner of the pilot-line did not find it necessary to automate its operation. They run a 24/7 production facility, so it is quite simple for production operators to make manual adjustments to the pilot-line during its startup, operation, and shutdown. In contrast, ORNL research is a single-shift operation. Therefore, automation is necessary to enable safe and effective single-shift operation of the conventional pilot-line. Operational and environmental testing indicated the need for

Table 1. ORNL acceptance test data for conventional pilot-line.

Property	Parameter	Acceptance Run	Baseline*
Tensile modulus, Mpsi	Mean	30.4	28.7
	Standard deviation	0.90	0.71
	Design basis**	27.7	26.6
Ultimate strain, %	Mean	1.12	1.20
	Standard deviation	0.10	0.08
	Design basis	0.81	0.97
Ultimate strength, kpsi	Mean	349	364
	Standard deviation	34.3	22.0
	Design basis	246	298

* Baseline is commercial-grade fiber, tested at ORNL

** Design basis is mean less three standard deviations

Figure 2. Conventional pilot-line installation at ORNL.

modifications to the exhaust system, gas-purging system, and control system, in addition to "carbon proofing" to protect nearby electronics and high voltage electrical equipment. Modifications made in FY 2006 included:

- steam heat for tow pre-treatment (the pilot-line seller initially undersized the steam generator, so this required two iterations);
- chilled-water cooling for furnace seals at fiber inlets to and exits from the furnaces;
- addition of carbon-particulate filtration in the exhaust system; and
- improved purge-gas manifolding.

The project team designed and constructed or acquired, for future installation,

- sizing squeeze rolls,
- sizing dryer,
- ozone generator for ozone surface treatment, and
- ozone monitoring equipment.

The project team initiated carbon-proofing, as well as upgrades to the purge-gas delivery system, control system, and exhaust system; the necessary hardware for these modifications will be procured and installed in FY 2007. High-speed winder acquisition was planned to commence in FY 2006, however, budget reductions delayed acquisition by a year. Winder specifications were developed in FY 2006. The microwave-assisted plasma (MAP) carbonization module (see 3.E) was prepared for integration with other modules. Advanced oxidation parameters were found to depend on the advanced stabilization route (see 3.C). Therefore, the advanced stabilization-oxidation interface, and the respective process parameters were experimentally investigated (see 3.D).

Advanced-Technology Pilot-Line

The MAP carbonization module is the base module for the advanced-technology integration line. Other modules will be added to it later. It is now in place and operational. The MAP carbonization milestone of three-tow operation at 1 m/min was satisfied in FY2006; hence, the MAP unit is now ready for integration. The project team will continue scaling and maturing the MAP carbonization technology under integration and/or other future projects.

Several high-speed winders and tensioners are needed to support operation at the bandwidth (number of tows) and range of speeds targeted for the conventional and advanced-technology lines. Specifications have been drafted, but the capital procurement is scheduled for FY 2007 and beyond, per funding availability.

Metrics have been established for the systems-integration project. They are shown in "spider chart" form in Figure 3. Current status, as expected very early in the project, is well off the target for most metrics.

Precursor Evaluation System

Due to its size, the aforementioned pilot facility requires ≥ 1k tows, preferably many meters long. The tensioning system is not sufficiently precise to handle tows smaller than 1k without breaking them. Startup time for the conventional pilot-line graphitization furnace is about 20 hours, so restringing and restarting after tow breakage requires many hours. This is incompatible with the

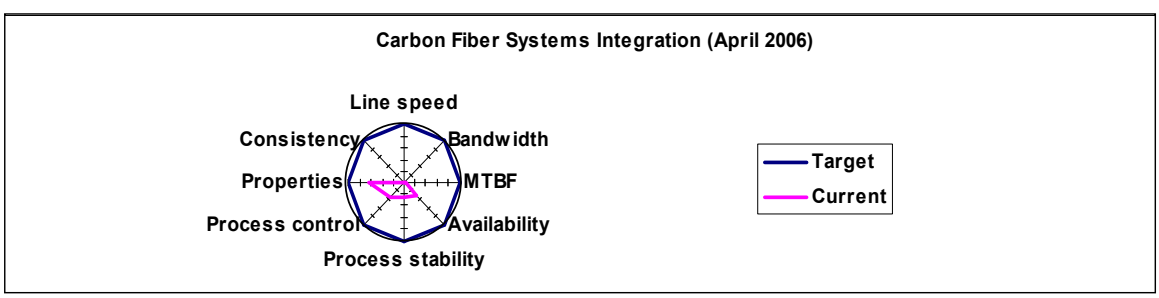

Figure 3. "Spider chart" showing carbon-fiber systems-integration metrics and estimated current status.

need for inexpensive, fast-turnaround, precursor evaluations. New precursor-development projects need many intermittent trials on very small tows, as they initially extrude one or a few filaments in short lengths for initial testing. The conventional pilot-line is not amenable to this kind of operation; therefore, a compact tube-furnace system with precision tensioners will be assembled for evaluating prospective precursors and developing their conversion protocols. The tube-furnace system will have rapid start-up/shutdown capabilities and will be capable of heat-treating short lengths of single filaments or any number of filaments up to approximately 3k tow size. Controlled tension can be applied to tows larger than approximately 10 - 20 filaments. Smaller tows can be "restrained" to provide some tension. Short fiber lengths can be readily transported through the system on trays or similar compact carriers. During FY 2006, the researchers commenced designing and specifying the tube-furnace system. It is expected to become operational in the second quarter of FY 2007.

Future Direction

During FY 2007, conventional pilot-line modifications and the advanced-technology pilot-line design and procurement will continue. The conventional and advanced-technology pilot-lines will be carbon-proofed (to better protect proximate electronic and high-voltage equipment), and conventional pilot-line operation will be automated. The researchers will investigate methods for resolving sealing and materials issues on the MAP carbonization advanced-technology module. Researchers plan to commence investigating the oxidation-carbonization interface, specifically addressing whether it is possible to carbonize fibers at a reduced oxidation level. Major procurements will include special fiber-handling equipment (a capital procurement) for the conventional and advanced-technology pilot-lines; furnaces, controllers, and transporters/tensioners for the precursor-evaluation system; and instruments that can be used for *in-situ*, real-time measurement of temperatures, gas compositions, etc. at various points in the advanced conversion processes. If lignin precursor fibers become available, a lignin-conversion process will be developed. If work is initiated on textile-based precursors, the researchers will optimize the textile-PAN conversion protocol; convert textile-based materials in the conventional pilot-line; and distribute finished textile-PAN tows to the three domestic automotive OEMs.

Conclusions

The development of a "carbon-fiber systems-integration" facility for testing and demonstrating new carbon-fiber manufacturing technology is underway. A conventional carbon-fiber pilot-line has been installed and tested. Modifications are underway to enable sustained, automated operation of the conventional pilot-line. An advanced-technology pilot-line will be constructed next to the conventional pilot-line, with capability to utilize any combination of conventional and advanced-conversion processes provided by the two adjacent lines. The MAP carbonization module is the first advanced-technology module and has been installed adjacent to the conventional pilot-line. MAP carbonization scaling is underway to increase line speed and bandwidth. Advanced stabilization and oxidation projects are developing modules that will be installed in the advanced-technology pilot-line in future years. A precursor evaluation system is being developed to evaluate conversion protocols for lignin and other alternative precursor fibers. Metrics have been established against which progress toward system integration and commercialization can be evaluated.

Presentations/Patents/Publications

None.