Fabrication Of Carbon-Boron Reinforced Dry Polymer Matrix Composite Tape[1]

Harry L. Belvin and Roberto J. Cano

NASA Langley Research Center
6A West Taylor Street
Hampton, VA 23681-0001

Monte Treasure and Thomas W. Shahood

Textron Specialty Materials
Two Industrial Avenue
Lowell, MA. 01851-5199

ABSTRACT

Future generation aerospace vehicles will require specialized hybrid material forms for component structure fabrication. For this reason, high temperature composite prepregs in both dry and wet forms are being developed at NASA Langley Research Center (LaRC). In an attempt to improve compressive properties of carbon fiber reinforced composites, a hybrid carbon-boron tape was developed and used to fabricate composite laminates which were subsequently cut into flexural and compression specimens and tested.

The hybrid material, given the designation HYCARB, was fabricated by modifying a previously developed process for the manufacture of dry polymer matrix composite (PMC) tape at LaRC. In this work, boron fibers were processed with IM7/ LaRC™ IAX poly(amide acid) solution-coated prepreg to form a dry hybrid tape for Automated Tow Placement (ATP). Boron fibers were encapsulated between two (2) layers of reduced volatile, low fiber areal weight poly(amide acid) solution-coated prepreg. The hybrid prepreg was then fully imidizied and consolidated into a dry tape suitable for ATP.

The fabrication of a hybrid boron material form for tow placement aids in the reduction of the overall manufacturing cost of boron reinforced composites, while realizing the improved compression strengths. Composite specimens were press-molded from the hybrid material and exhibited excellent mechanical properties.

KEY WORDS: Boron, Composite, Reinforcement

---

[1] This paper is declared a work of the U.S. Government and is not subject to copyright protection in the United States.

1.0 INTRODUCTION

As part of an effort to develop high performance composite technology for the aircraft industry and launch vehicle fabricators, NASA Langley Research Center (LaRC) has investigated ways to make intermediate composite material forms and develop the processes required for the manufacture of component structure from those materials. In the past, these composite research and development activities have included powder impregnation of carbon fiber tow bundles, dry (containing no solvent) PMC ribbon/tape fabrication from powder-coated towpreg, and ATP of ribbon/tape to form specimens for testing. Recently, a previously developed process for making solvent-free thermoplastic tape was modified to fabricate dry-tape from an alternative form of precursor (solution-coated prepreg). The NASA Langley 3-inch-wide-dry tape line was modified and processing conditions were established for producing high quality well-consolidated tapes from a modified solution-coated prepreg for ATP [1, 2].

During the past decade several methods have been developed for the fabrication of precursor materials for the use in dry tape fabrication. One method developed at NASA Langley is the powder curtain process (Fig. 1) [3]. In precursor fabrication methods such as the powder curtain process, the powder content along the length of the tow may vary. During tape fabrication, the variance may result in resin rich or resin poor areas, facilitating splits or gaps in the tape. The use of the solution coating process creates a more evenly distributed resin content along the length of the material. The process developed at NASA Langley Research Center using a modified prepreg as the precursor has the designation "wet to dry process" [4].

The object of this study was to develop fabrication procedures for the manufacture of a 7.6 cm (3 in.) wide dry carbon-boron reinforced composite tape. The wet to dry process developed at LaRC for making dry 7.6 cm (3 in.) wide tape for ATP provided the basis for this new material form. The current state of the art technology presses boron fibers into the face of standard wet prepregs to manufacture the hybrid material. The use of standard prepreg requires hand lay-up techniques and autoclave processing, which increases the overall component cost. During the course of the work, the NASA Langley 3-inch-wide-dry tape line (Fig 2) was used and new process parameters for the wet to dry process to fabricate the hybrid material were established. New parameters were needed because processing conditions of the boron reinforced precursor were considerably different from the original modified prepreg used.

2.0 MATERIALS[2]

IM7 12K, unsized carbon fiber from Hexcel, Salt Lake City, UT and a 30% solids poly(amide acid) solution of LaRC™IAX in N-methyl-2-pyrrolidinone (NMP) from Imitec Inc. of Schenectady, N.Y. were used to manufacture the precursor for this project. The boron was

[2] The use of trademarks or names of manufacturers in this report is for accurate reporting and does not constitute an official endorsement, either expressed or implied, of such products or manufacturers by NASA

supplied and inserted into the precursor tape by Textron Specialty Materials of Lowell, MA. The boron was inserted to a density of 200 ends per inch (EPI), using a proprietary process.

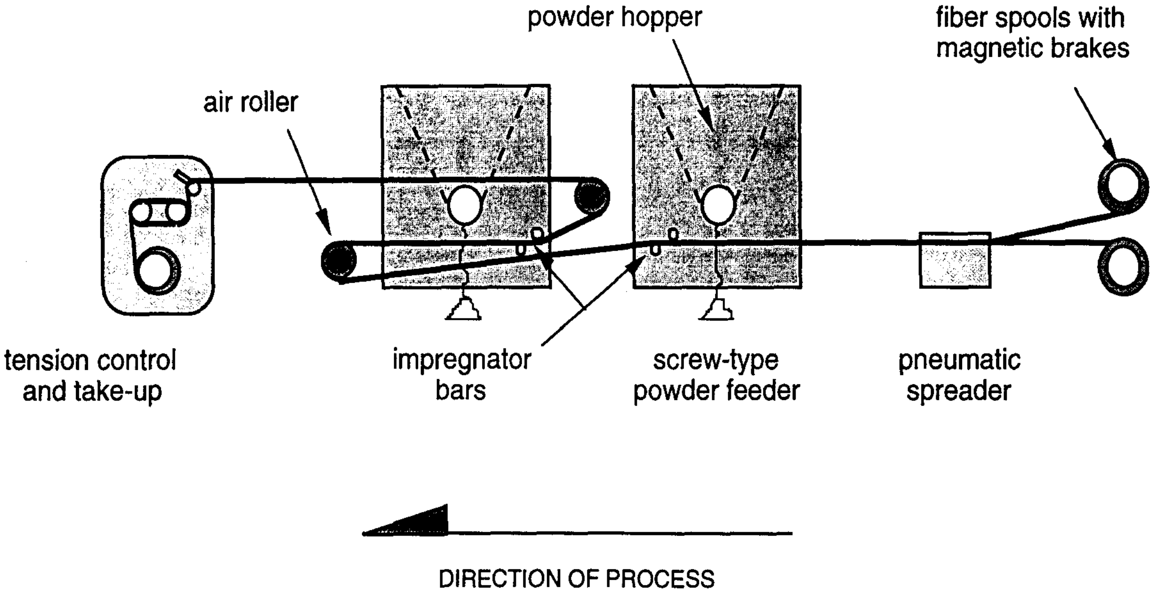

Figure 1. The NASA LaRC powder curtain process for powder impregnating carbon fibers

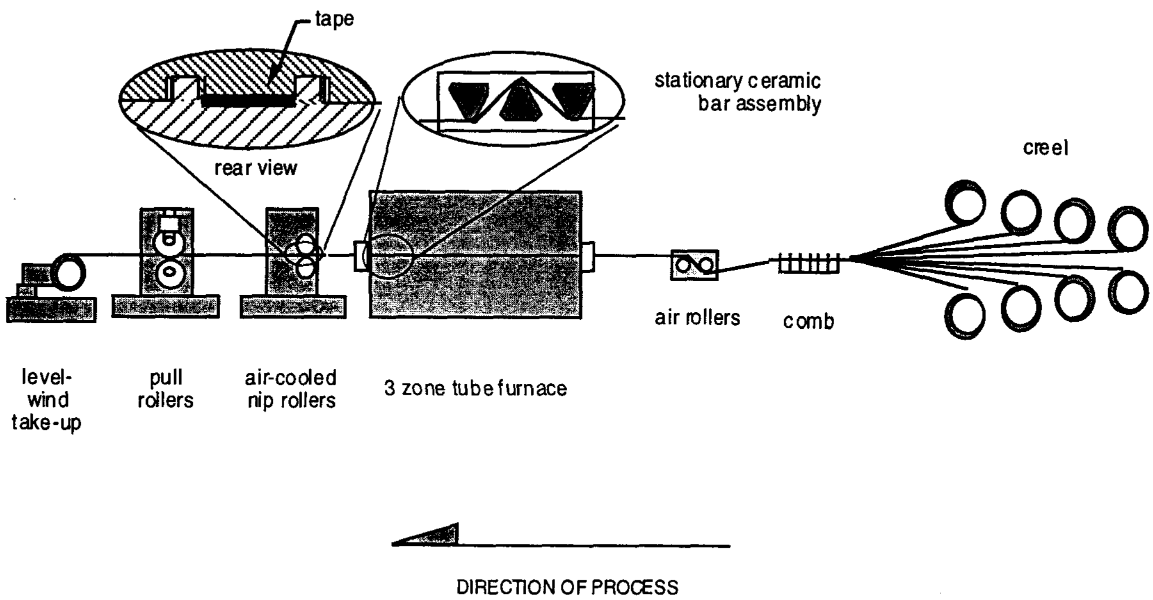

Figure 2. The NASA LaRC tape process for converting powder-coated towpreg into fully consolidated 7.6cm (3 in)-wide tape

3.0 EXPERIMENTAL

3.1 Boron/Carbon Reinforced Precursor Fabrication A 30% solids poly(amide acid) solution of LaRC™ IAX in NMP was used in this study. The solution was processed into a 15.24 cm (6.00 in.) wide prepreg on the NASA LaRC Multipurpose Prepregger (Fig. 3)[5]. The precursor tape contained a volatile content of approximately 18% and dry solids content of approximately 38% by weight [4]. The fiber areal weight was approximately 77 grams/cm^2. Typical poly(amide acid) prepreg has a volatile content of 20% to 24%, a dry solids content of approximately 35% with a fiber areal weight of 145 grams/cm^2. A slightly lower volatile content prepreg was fabricated to facilitate solvent removal and imidization process of the material during the final phase of precursor processing. The reduced fiber areal weight was needed to allow for the insertion of the boron fibers between the prepreg layers and maintain an approximate carbon fiber FAW of 150 grams/cm^2. Two rolls of the modified prepreg material were fabricated and sent to Textron Specialty Materials for the insertion of the boron fibers

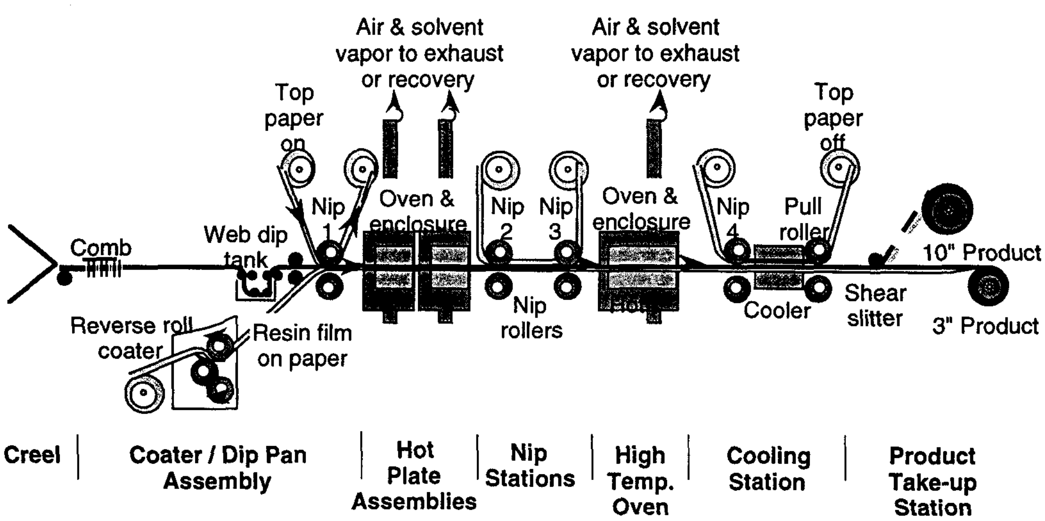

Figure 3. NASA LaRC Multipurpose Prepregger

between the two plies, using a proprietary process. The final hybrid boron/carbon reinforced material was shipped back to LaRC for slitting and final processing using the NASA LaRC 3-inch Wide Dry Tape line (Fig 2).

3.2 Boron/Carbon Reinforced Dry Tape Fabrication. The NASA LaRC 3-inch Wide Dry Tape line (Fig. 2) was used for the final processing phase of the boron reinforced precursor. Typical process parameters for LaRC™ IAX were modified due to the increased material present during processing. The typical wet to dry process uses a prepreg tape approximately 0.02 cm (0.008) thick and 8.26 cm (3.25 in.) in width with a fiber areal weight of approximately 150 grams/cm^2. The operational parameters typically used for processing

LaRC™ IAX, include a bar temperature range of 360°C(680°F) to 375°C(707°F), line speed of 3.05 m/min (10 ft./min) and spool tension ranges from 75-125 grams. The original parameters were modified to allow for the extra resin and volatile content in the precursor tape. Original IAX work without boron utilized a precursor tape with approximately 12% volatile content by weight. The bar temperature had a range of 390°C (734°F) to 410°C (770°F), while maintaining a line speed of 3.05 m/min (10 ft./min.) and a tension of 100 grams on the spool. A spool of LaRC™ IAX without boron reinforcement was also fabricated using the original parameters. This dry tape was used to manufacture the control specimens for comparison to the boron reinforced specimens as well as in the hybrid specimens where plies were needed without boron. Test specimens reinforced with boron were fabricated with one, two and three plies of boron contained within.

3.3 Dry Tape Evaluation A Shimadzu DSC-50 Differential scanning calorimeter (DSC) was used to verify that the processed tape glass transition temperature (Tg) was below the final cured Tg of the LaRC™ IAX thermoplastic polyimide (Fig. 4 & 5). A Jeol JSM-5600 scanning electron microscope was used to determine tape quality of both the control and the boron reinforced tapes (Fig. 6 & 7). Also, residual volatile contents were determined to aid in the cure cycle development for press molding the tape into test specimens. Volatile contents were generated by weighting the material, heating it to 250° C (482° F) and holding for an hour, then determining a final weight. The tape contained approximately 0.4 to 0.3 percent volatile content by weight, which did not exceed some fully imidized powder-coated tow preg after processing. The press molded specimens are the baseline to which the ATP specimens will be compared.

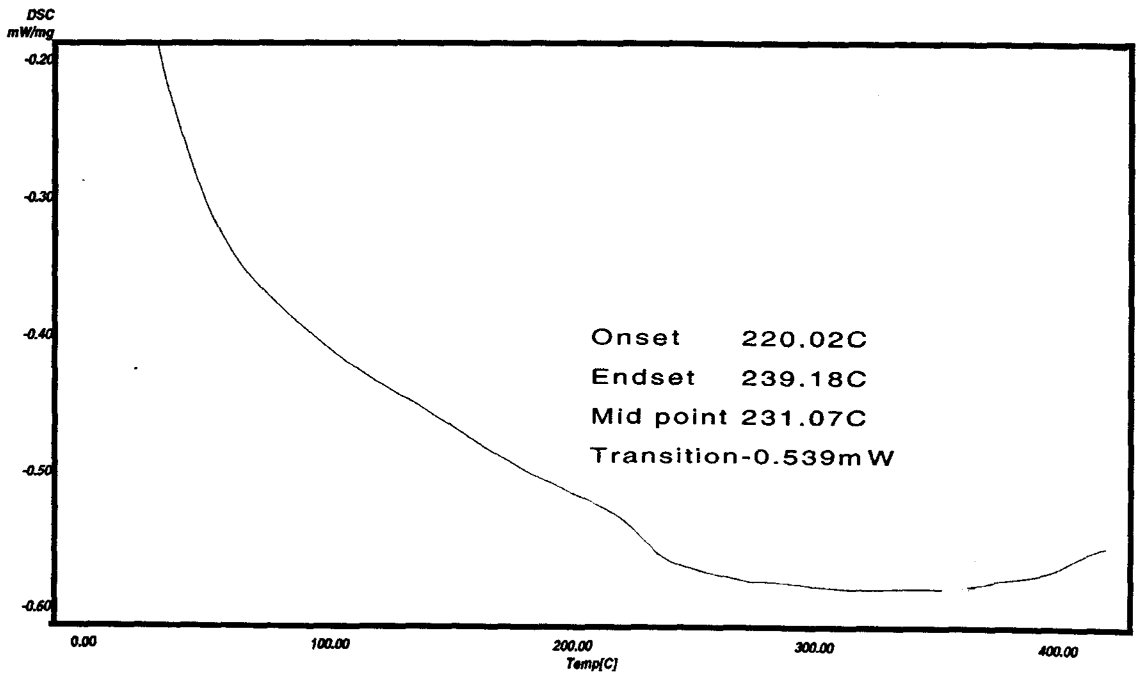

Figure 4. DSC of LaRC™IAX/Boron/IM7 thermoplastic polyimide tape, initial scan

3.4 Specimen fabrication A quantity of material was fabricated for the manufacture of specimens for 0° flexure, transverse flexure, and notched compression tests. In each test group, specimens were fabricated with one, two, and three plies of boron and a control (no boron reinforcement) for comparison. In each case, identical lay-ups for the hybrid and control specimens were fabricated. For the flexure tests (ASTM D-790), ten ply unidirectional panels were fabricated and for the notched compression test a modified version of the Northrop Corporation open hole compression test (OHC) was used. Lay-ups for each test are shown in

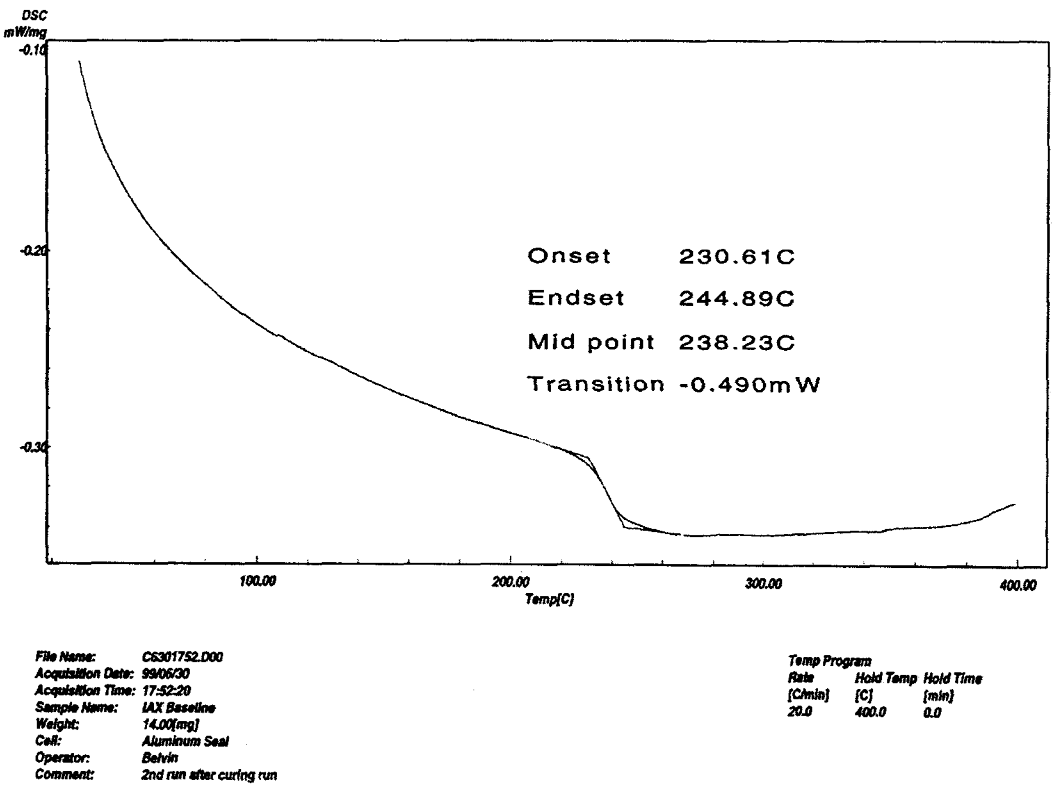

Figure 5. DSC of LaRC™IAX/Boron/IM7 thermoplastic polyimide tape after cure

Table 1. The panels were pressed molded using a modified cure cycle for the LaRC™ IAX (Fig. 8). A typical cure cycle for the LaRC™ IAX contains a drying hold at 225°C (437°F) for one hour [6]. Since the dry tape has an average volatile content of less than 1% by weight, the drying hold was reduced to 15 minutes. Ultrasonic evaluation using a Sonix FlexScan C-Scan was used to determine panel quality before the cutting of the test specimens. A combination of two techniques were used for the ultrasonic NDE, pulse echo and through transmission. All the panels were determined to be of acceptable quality for testing.

4.0 RESULTS and DISCUSSION

4.1 Tape Characteristics/Morphology. Approximately 8.167 kg (18 lbs.) of 7.62 cm (3.00 in) wide LaRC™ IAX tape was produced during this study. In general, the tape seemed well consolidated and broke cleanly. The fracture surface of the fibers appears to be well wet-out (Fig. 6). Approximately 1.81 kg (4 lbs.) of 7.62 cm (3.00 in) wide LaRC™IAX Hycarb was produced. The fracture surface illustrates that the boron is not totally encapsulated after processing (Fig. 7). The carbon filaments are not filling the gaps between the boron fibers.

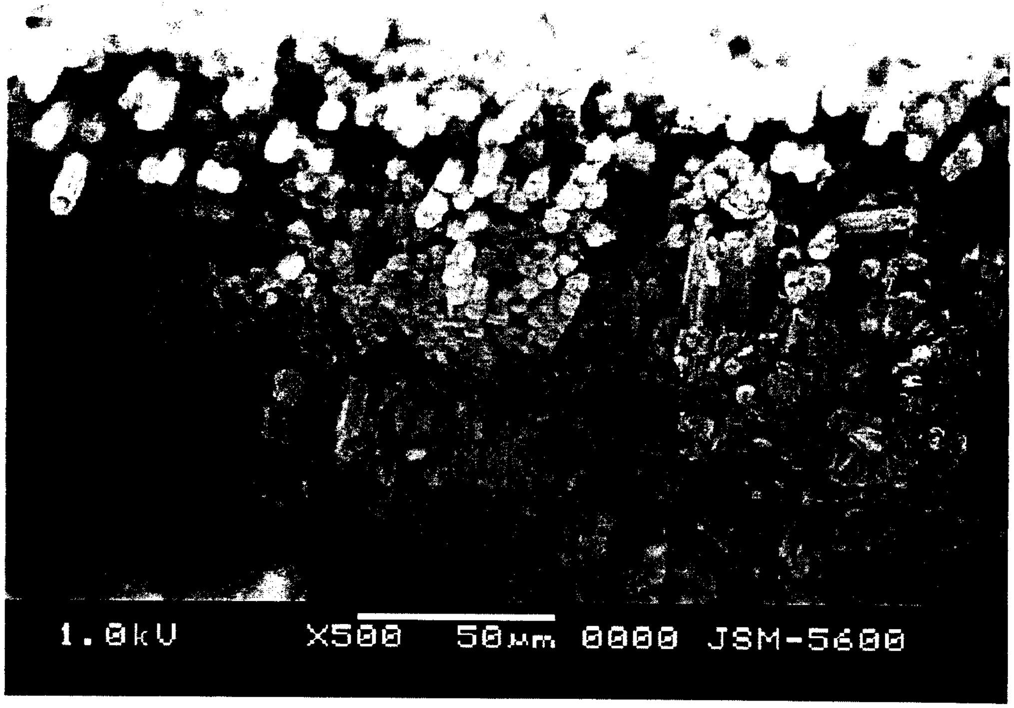

Figure 6. SEM of LaRC™ IAX/IM7 thermoplastic polyimide tape, fracture surface

Figure 7. Sem of LaRC™ IAX/Boron/IM7 thermoplastic polyimide tape

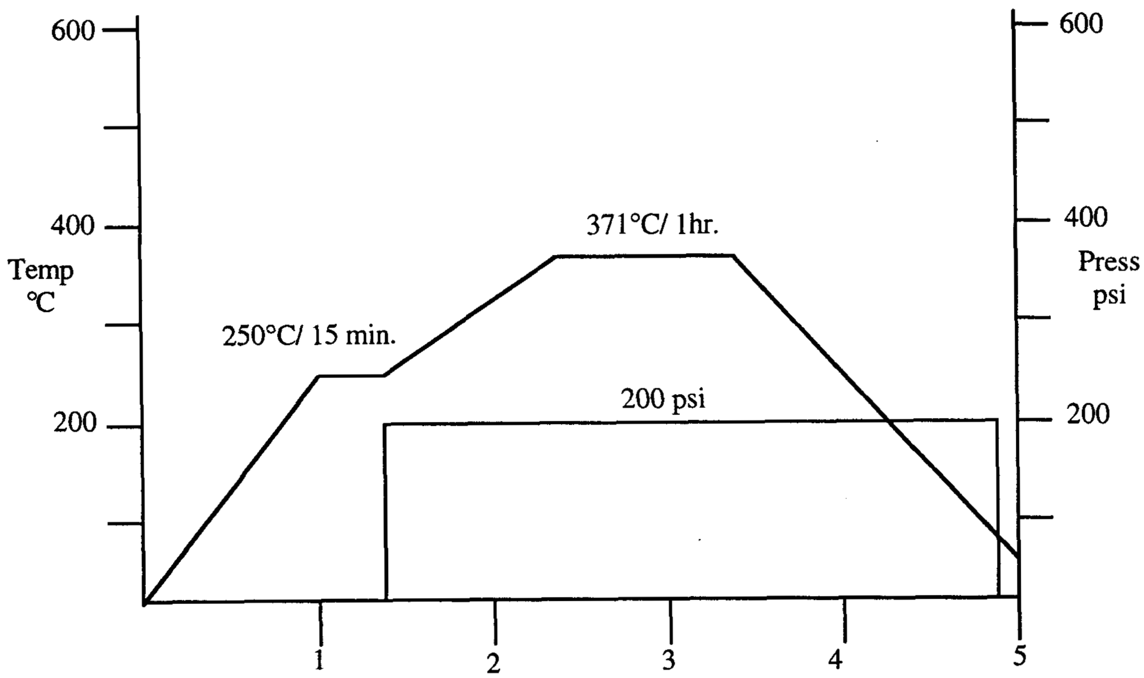

Figure 8. Modified cure cycle of LaRC™IAX

	0°Flex	**90°Flex**	**OHC**
Baseline	$[0]_{10}$	$[0]_{10}$	$[\pm 45/90/0_2/\pm 45/0_2/\pm 45/0]_s$
1 Ply Boron	$[0_5/B/0_5]$	$[0_5/B/0_5]$	$[\pm 45/90/0_2/\pm 45/0_2/\pm 45/0/B^*]_s$
2 Plies Boron	$[0_4/B/0]_s$	$[0_4/B/0]_s$	$[\pm 45/90/0_2/\pm 45/0/B/0/\pm 45/0]_s$
3 Plies Boron	$[0_4/B/0/B/0/B/0_4]$	$[0_4/B/0/B/0/B/0_4]$	$[\pm 45/90/0_2/\pm 45/0/B/0/\pm 45/0/B^*]_s$

Table 1. Test panel lay-ups for the test specimen fabrication.

* Designates only one ply of Boron at that position in the symmetric lay-up

4.2 Panel Morphology Photomicrographs of the cross section of a boron reinforced panel with one array of boron fibers and the baseline panel (Fig 9 & 10) exhibit a void fraction of less than 1%. The same was shown with the panels that contained two and three arrays of boron fibers. The boron fibers become totally encapsulated during the molding cycle. The ability of the resin to fully encapsulate the boron fibers during the molding cycle demonstrates the dry tapes sufficient residual processability after tape fabrication. This combined with a Tg lower than the fully-cured material suggests that the ATP process has a good chance of being successful with this material form.

4.3 Mechanical Properties Panels were manufactured to determine 0° flexure and transverse flexure properties at room temperature and 177°C and notched compression properties (OHC) at room temperature. The mechanical properties determined for the LaRC™ IAX/carbon/boron tape are shown in Figures 11,12, and 13. The 90° flexure test results

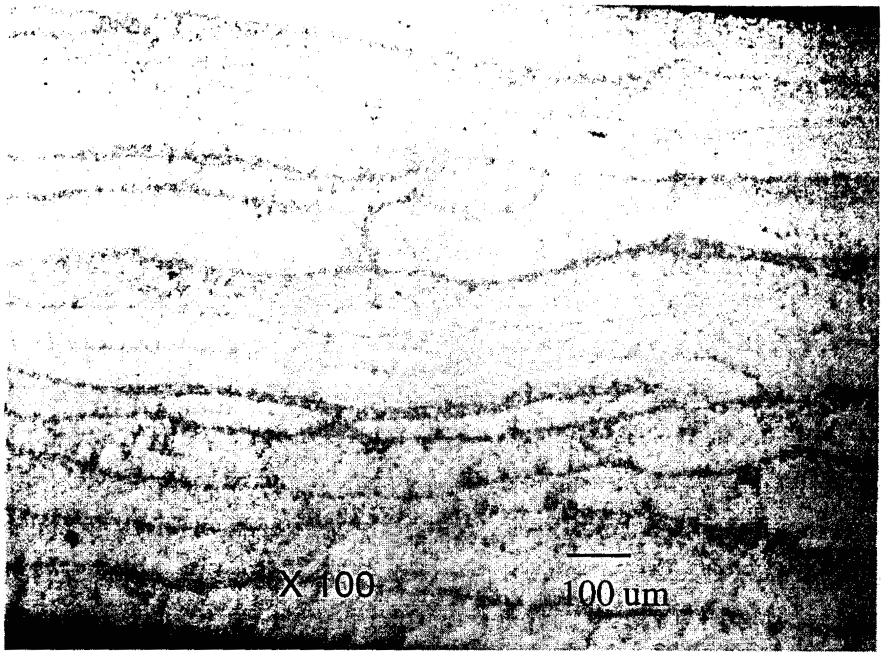

Figure 9. Photomicrograph of cross section of LaRC™ IAX/IM7 baseline panel .

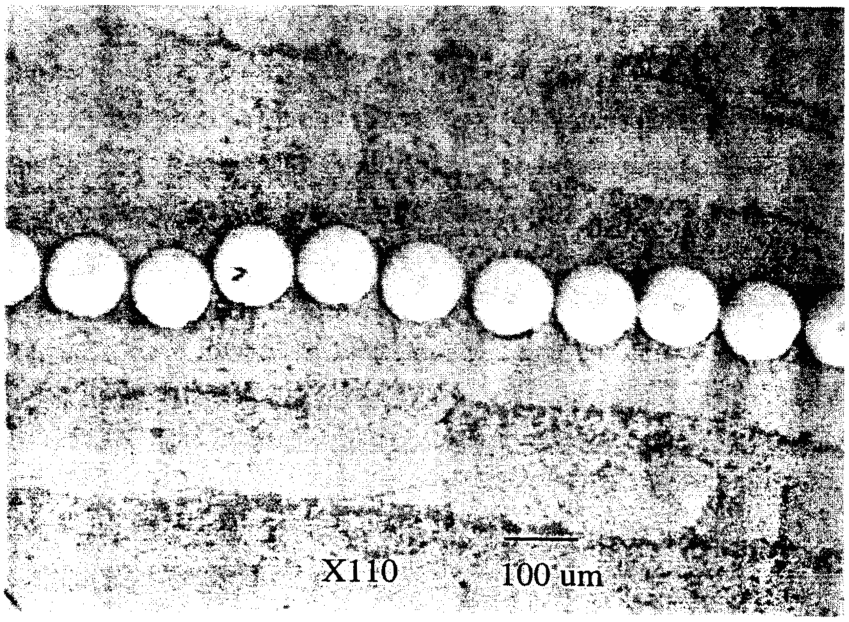

Figure 10. Photomicrograph of cross section of LaRC™ IAX/Boron/IM7 panel with one array of boron fibers

demonstrate that the boron did not adversely affect the resin dominated mechanical properties and provided an adequate interface between the boron fibers and the matrix. All the data obtained for the 90° flex test are comparable to the baseline data within the scatter of the data (Fig 12). The 0° flexure and the open hole compression data illustrate a marked improvement

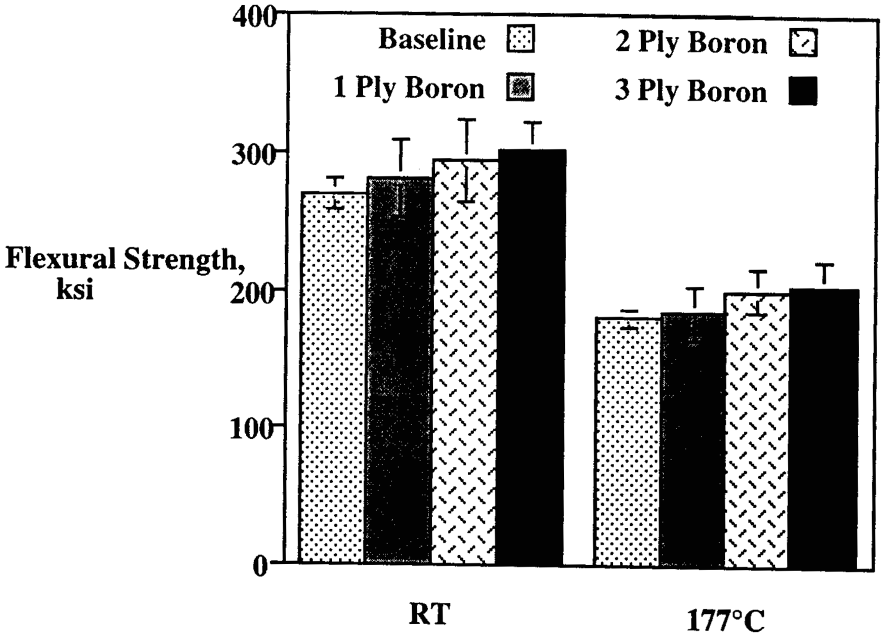

Figure 11. 0° Flex mechanical properties of LaRC™ IAX/Boron /IM7 and LaRC™ IAX/IM7 at room temperature and 177°C

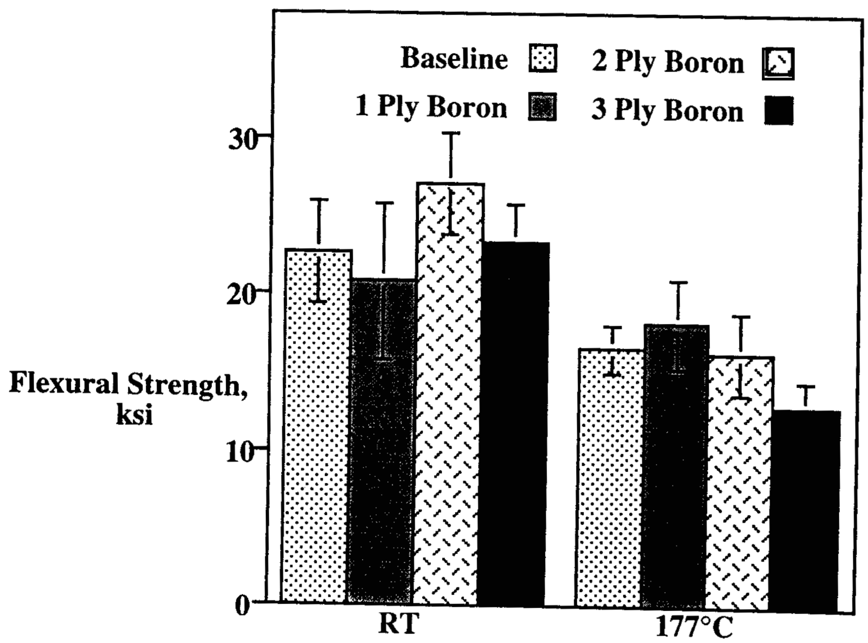

Figure 12. 90° Flex mechanical properties of LaRC™ IAX/Boron /IM7 and LaRC™ IAX/IM7 at room temperature and 177°C