AN IMPROVED COMPRESSION MOLDING TECHNOLOGY FOR CONTINUOUS FIBER REINFORCED COMPOSITE LAMINATE - Part I: AS-4/LaRC-TPI 1500 (HFG) Prepreg System

Tan-Hung Hou and Paul W. Kidder
Lockheed Engineering & Sciences Company
Hampton, Virginia 23666

Rakasi M. Reddy
Department of Mechanical Engineering & Mechanics
Old Dominion University
Norfolk, Virginia 23508

Contract NAS1-19000
Grant NAG1-569
May 1991
AN IMPROVED COMPRESSION MOLDING TECHNOLOGY FOR CONTINUOUS FIBER REINFORCED COMPOSITE LAMINATE - Part I: AS-4/LaRC-TPI 1500 Prepreg System

Table of Contents

FOREWORD ................................................................. iii
ABSTRACT ................................................................. 1
INTRODUCTION ........................................................... 2
MOLDING EXPERIMENTS
   Run MW101 ............................................................... 3
   Run MW102 ............................................................... 4
   Run MW103 ............................................................... 5
   Run MW104 ............................................................... 6
   Run MW104B ............................................................. 7
CONSOLIDATION MECHANISM ............................................. 9
COMPOSITE MECHANICAL PROPERTIES
   Short Beam Shear (SBS) Strength .................................. 11
   Flexural Strength .................................................... 13
CONCLUSIONS .......................................................... 14
REFERENCES ............................................................. 16
This work was sponsored by and performed at the Polymeric Materials Branch (PMB), NASA Langley Research Center (LaRC). Mr. Robert M. Baucom (PMB) was the Technical Monitor for this investigation.

The authors benefited fruitfully from technical discussion with Mr. Timothy W. Towell (AMB) and Dr. Norman J. Johnston (PMB) during the course of this investigation. Financial support of LaRC through a contract NAS1-19000 to Lockheed Engineering & Sciences Company, Hampton, Virginia and a grant NAG1-569 to the Old Dominion University, Norfolk, Virginia are also gratefully acknowledged.
AN IMPROVED COMPRESSION MOLDING TECHNOLOGY FOR CONTINUOUS FIBER REINFORCED COMPOSITE LAMINATE - Part I: AS-4/LaRC-TPI 1500 (HFG) Prepreg System

Tan-Hung Hou and Paul W. Kidder
Lockheed Engineering & Sciences Company
Hampton, Virginia 23666

Rakasi M. Reddy
Department of Mechanical Engineering & Mechanics
Old Dominion University
Norfolk, Virginia 23508

ABSTRACT

Poor processability of fiber reinforced high performance polyimide thermoplastic resin composites is a well recognized issue which, in many cases, prohibits the fabrication of composite parts with satisfactorily consolidated quality. Without modifying the resin matrix chemistry, improved compression molding procedures have been proposed and investigated with the AS-4/LaRC-TPI 1500 High Flow Grade (HFG) prepreg system in this study. Composite panels with excellent C-scans can be consistently molded by this method under 700°F and a consolidation pressure as low as 100 psi. A mechanism for the consolidation of the composite under this improved molding technique is discussed. This mechanism reveals that a certain degree of matrix shear and tow filament slippage and nesting between plies occur during consolidation, which leads to a reduction of the consolidating pressure necessary to offset the otherwise intimate inter fiber-fiber contact and consequently achieves a better consolidation quality. Outstanding short beam shear strength and flexural strength were obtained from the molded panels. A prolonged consolidation step under low pressure, i.e., 100 psi at 700°F for 75 minutes, was found to significantly enhance the composite mechanical properties.
INTRODUCTION

Compression molding of flat panels of fiber reinforced resin matrix composite laminates is the simplest form of a molding process which employs matched metal dies. In this process, individual prepreg plies were cut into the desired dimensions from a flat sheet of prepreg material. The number of prepreg plies are dictated by the desired final part thickness. The prepreg plies were oriented during the stacking sequence to yield orthotropic or quasi-isotropic laminates. The plies were stacked inside the cavity of the female mold and subjected to consolidation under heat and compression forces.

Despite the simplicity in tooling design, the compression molding of a composite laminate is by no means a trivial process [1-4]. Among others, processing parameters such as heat transfer phenomena [5-7], resin flow behavior [8,9], fiber wetting, adhesion and elastic behavior [10,11], volatile escape mechanism [12a,b], and bulk consolidation [4,13], etc. are issues imperative to be understood and controlled. For thermosetting materials, extra attention is needed for the resin chemorheology (viscosity-cure time profile) and reaction heats [14-16]. For the crystallizable resins, such as Poly-Ether-Ether-Ketone (PEEK) polymer, the effects of molding temperature, cooling scheme, post cure annealing and related residual stresses on the formation of crystallinity within the molded specimen are critical for the fabrication of composite parts with controlled properties [17-19].

For thermoplastic resins, such as the class of aromatic polyimide materials, resin flow behavior is one of the major issues involved in the compression molding process. The stiff backbones of the molecules with aromatic rings give rise to the high glass transition temperature of the polymers. Although some unique properties such as thermal and thermo-oxidative stability, superior chemical resistance and excellent mechanical properties are achieved, the high viscosity associated with such a molecular structure often leads to poor processability. Using conventional molding technology, void-free and well consolidated composite laminates with good mechanical properties are often difficult to achieve.

An improved molding technique that did not involve modifying the resin matrix properties is employed in this investigation. The technique was demonstrated with the AS-4/LaRC-TPI 1500 prepreg system. It was shown that well consolidated composite panels with exceptional C-scans and mechanical properties can be consistently molded by this method with a far less demanding pressure level than that used by conventional molding techniques.
MOLDING EXPERIMENTS

The LaRC-TPI 1500 prepreg system selected for this study was fabricated in-house at Langley Research Center. The LaRC-TPI 1500 high flow grade (HFG) polymer was manufactured by Mitsui Toatsu Chemical, Inc. of Japan. Thermal and rheological properties of this class of polymers have been reported previously [20]. It was impregnated into fiber tows by means of a proprietary neutral buoyancy slurry technique [21,22] followed by pultrusion through a hot die at 650°F. The prepreg sheet appeared reasonable smooth. However, the resin content is not uniformly distributed. The acid digestion was done in concentrated sulfuric acid mixed with an equal weight of 30% hydrogen peroxide. Preliminary acid digestion of the prepreg sampled from three locations arbitrarily selected indicate resin contents of 42.8, 33.1 and 24.3% by weight.

SEM photomicrographs of the prepreg are shown in Figure 1. Although dry fibers are visible in a few areas near the tow center, the prepreg appears to possess a reasonably acceptable level of resin impregnation with no apparent separation among fiber tows.

Run MW101

A 10 ply 3.000" x 2.750" composite laminate was processed by a vacuum press according to the cure cycle shown in Table 1. The cure cycle consists of two cure steps with various combinations of temperature and pressure. A full vacuum of 29" Hg was used at each step in the cycle. The cycle was interrupted at the completion of cure step 1 (650°F/0psi/0.5hrs) so that information regarding any geometrical changes could be measured. The results are also included in Table 1. In the first step of the cure, the molding process included the following: two stops measuring 3.000" long by 0.125" high and 0.125" wide were inserted at the sides of the mold. Prepreg plies were cut to the size of 3.000" x 2.750" and stacked between the stops. At room temperature, the height of the stacked laminate is always higher than the stops. After insertion in the press the mold was closed to the stops with the application of 100 psi pressure and remained in contact with the prepreg transmitting the platen temperature to the laminate after the male mold contacted the stops. This loosely compacted structure in the unconsolidated laminate provides the necessary escape paths for the volatiles generated during the B-stage. It is noted that at the end of (the first) 650°F cure step, the composite panel ended up with a 0.5% weight loss. This is expected because of the hot melt process used during the prepping stage when the prepreg was pulled through the pultrusion die. The thickness of the laminate was 0.117 inch. This thickness is shorter than the height of the molding stops, indicating a process of resin diffusion after melting among
individual prepreg plies. The C-scan of this laminate was poor because of the loosely compacted structure existing in the unconsolidated laminate.

The stops were removed from the mold after the first (650°F/0psi/.5hr.) cure step was completed. A 10 minute hold at 750°F and 500 psi throughout the molding cycle completed the second cure step. A large change in panel dimensions resulted. The thickness was decreased from 0.117 to 0.056 inches, while the lateral in-plane dimension increased from 2.750" to a full 3.000", completely filling the mold cavity. Excess resin was squeezed out in both in-plane directions indicating excellent flow properties. Fiber plus resin wash-out in the lateral in-plane direction were also observed. The C-scan shown in Figure 2 indicates a panel with excellent consolidation quality.

Table 1. Geometrical changes of the 10 uni-ply composite laminate in Run MW101

<table>
<thead>
<tr>
<th>Press Cycle (°F/psi/hr)</th>
<th>Wt. (g)</th>
<th>Wt. Loss (g)</th>
<th>Wt. Loss (%)</th>
<th>Thickness (in)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ambient</td>
<td>13.096</td>
<td>---</td>
<td>---</td>
<td>---</td>
</tr>
<tr>
<td>1 650/0/.5</td>
<td>13.022</td>
<td>0.074</td>
<td>0.565</td>
<td>0.117±.003</td>
</tr>
<tr>
<td>2 750/500/.2</td>
<td>12.268</td>
<td>0.828</td>
<td>6.321</td>
<td>0.056±.002</td>
</tr>
</tbody>
</table>

Run MW102

Another 10 ply 3.000" x 2.750" composite laminate was processed in a vacuum press. In this run the B-stage step with internal molding stops was eliminated. The laminate was molded directly by a single step: 750°F for 10 minutes with 200 psi applied throughout the molding cycle. Geometrical changes of the composite panel are tabulated in Table 2. It is noted that the final thickness of the consolidated panel is comparable to the that of run MW101, and so is the weight reduction (6.107 vs. 6.321%). The lateral in-plane dimension increased from 2.750" to a full 3.000", completely filling the mold cavity. Excess resin was squeezed out in both in-plane directions indicating excellent flow properties. Fiber plus resin wash-out in the lateral in-plane direction were also observed. The C-scan shown in Figure 3 indicates a panel possessing a
consolidation quality of better than 90%, despite the fact that a small deterioration in quality is evident when compared with panel MW101 of Figure 2.

Table 2. Geometrical changes of the 10 uni-ply composite laminate in Run MW102

<table>
<thead>
<tr>
<th>Press Cycle (°F/psi/hr)</th>
<th>Wt. (g)</th>
<th>Wt. Loss (g)</th>
<th>Wt. Loss (%)</th>
<th>Thickness (in)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ambient</td>
<td>13.104</td>
<td>---</td>
<td>---</td>
<td>---</td>
</tr>
<tr>
<td>1</td>
<td>750/200/2</td>
<td>12.302</td>
<td>0.802</td>
<td>6.107</td>
</tr>
</tbody>
</table>

Run MW103

Another 10 ply 3.000" x 2.750" composite laminate was processed by a vacuum press according to the cure cycle shown in Table 3. The cure cycle is identical to that used in run MW101 (Table 1), except that 100 psi pressure was employed in the final consolidating step. In the first step of the cure cycle (650°F) using internal molding stops, the composite laminate realized a similar level of weight loss (0.47% ) as that of panel MW101. The thickness (0.112") of the unconsolidated panel is lower than the height of the stops, indicating a process of resin matrix diffusion after melting among individual prepreg plies. The C-scan of this laminate was poor because of the loosely compacted structure existing in the unconsolidated laminate.

The stops were removed from the mold after the first (650°F/0psi/.5hr.) cure step was completed. A 15 minute hold at 750 °F and 100 psi throughout the molding cycle completed the second cure step. A large change in panel dimensions resulted. The thickness was decreased from 0.116 to 0.058 inches, while the lateral in-plane dimension increased from 2.750" to a full 3.000", completely filling the mold cavity. Excess resin was squeezed out in both in-plane directions. The similar flow behavior observed to that of run MW102 under higher consolidation pressure (200 psi) indicates the excellent flow properties possessed by this high flow grade of LaRC-TPI 1500 polyimide. Fiber plus resin wash-out in the lateral in-plane direction was also observed. The weight loss at the completion of the final consolidation step is 3.62%, which is about 60% of the higher consolidation pressure steps (runs MW101 and MW102).
The C-scan shown in Figure 4 indicates a well consolidated composite panel with over 90% void free quality. The overall consolidation quality is poorer than the other panels reported above (Figures 2 and 3). This is attributed to the lower consolidation pressure used in this experiment.

Table 3. Geometrical changes of the 10 uni-ply composite laminate in Run MW103

<table>
<thead>
<tr>
<th>Press Cycle (°F/psi/hr)</th>
<th>Wt. (g)</th>
<th>Wt. Loss (g)</th>
<th>Wt. Loss (%)</th>
<th>Thickness (in)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ambient</td>
<td>12.912</td>
<td>---</td>
<td>---</td>
<td>---</td>
</tr>
<tr>
<td>1 650/0/.5</td>
<td>12.851</td>
<td>0.061</td>
<td>0.472</td>
<td>0.116±.002</td>
</tr>
<tr>
<td>2 750/100/.25</td>
<td>12.442</td>
<td>0.470</td>
<td>3.640</td>
<td>0.058±.002</td>
</tr>
</tbody>
</table>

Run MW104

In order to compare the composite consolidation qualities between molding processes either with or without internal molding stops, a 10 ply composite laminate was processed by the conventional molding technology in a vacuum press. Prepreg plies were cut to the size of 3.000" x 3.000" which fit the cavity of the mold exactly. The molding cycle is shown in Table 4. In this run the B-stage step with internal molding stops was eliminated. Because of the minimal weight loss observed in the earlier experiments (runs MW101 and 103), elimination of the B-stage step is believed to have insignificant effects on the final consolidation quality of the composite panel.

The laminate was molded directly in a single step: 750°F for 15 minutes at 100 psi throughout the molding cycle. Geometrical changes of the composite panel are also tabulated in Table 4. It is noted that the weight reduction (3.07% vs. 3.62%) is comparable to that of run MW103. Excess resin was squeezed out in both in-plane directions indicating similar flow properties to that of run MW103. Fibers plus resin wash-out in the lateral in-plane direction was also observed.
It is noted that, despite the use of the same consolidating pressure (100 psi), the final thickness of the consolidated panel is 12% thicker (0.065" vs. 0.058") than that of run MW103 which employed the internal molding stops. The appearance of the panel surfaces are not as smooth as panel MW103. Panel MW104 molded without stops could not deform laterally, consequently the interply nesting and compaction in the previous panels (MW101-103) did not occur resulting in a poor quality panel. When compared to panel of MW103, it is conceivable that a gain of 12% in the final thickness will translate to a 12% loss of compaction in the total panel volume, as a result of the elimination of the use of molding stops. Such a reduction in the level of volume compaction will inevitably jeopardize the consolidation quality of the composite panel.

Precisely because of these reasons, it is shown in Figure 5 that the panel of run MW104 exhibits poorer consolidation quality when compared to that of MW103.

Table 4. Geometrical changes of the 10 uni-ply composite laminate in Run MW104

<table>
<thead>
<tr>
<th>Press Cycle (°F/psi/hr)</th>
<th>Wt. (g)</th>
<th>Wt. Loss (g)</th>
<th>Wt. Loss (%)</th>
<th>Thickness (in)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ambient</td>
<td>13.680</td>
<td>---</td>
<td>---</td>
<td>---</td>
</tr>
<tr>
<td>1 750/100/.25</td>
<td>12.256</td>
<td>0.424</td>
<td>3.099</td>
<td>0.065±0.002</td>
</tr>
</tbody>
</table>

Run MW104B

The panel MW104, which failed to be completely consolidated by compression molding as discussed above, was trimmed .125" on each side in the in-plane lateral direction. The resulting 3.000" x 2.750" panel was again molded in a 3.000" x 3.000" mold following the identical cure cycle (750°F for 15 minutes with 100 psi throughout the cycle).

At the completion of the molding cycle, the composite panel had expanded to its full dimension of 3.000" x 3.000". A small weight loss (0.345%) was recorded for this panel (see Table 5). The C-scan shown in Figure 6 indicates a well consolidated composite panel. This run is essentially a repeat of run MW103 which produced a panel with an identical C-scan. It demonstrates that a LaRC-TPI 1500 (HFG) composite panel with outstanding C-scan quality can
be molded with a consolidation pressure as low as 100 psi by means of the improved molding technology.

When compared to the panel from run MW104, it is clear that the failure of complete consolidation for that panel was not due to the high viscosity of the matrix resin. The success of the molding experiment MW104B, which salvaged and upgraded the poorly consolidated panel of run MW104 into a composite laminate with exceptional C-scan quality, illustrates that there is a significant level of external load absorbed by the intimate interply fiber-fiber contact when the conventional molding technique is employed. In the improved molding technique used in run MW104B, the amount of load absorption was greatly reduced by the mechanism of lateral flow and interply fiber-fiber nesting. Consequently, better flow behavior was realized due to a higher effective pressure experienced by the matrix resin within the composite laminate.

Table 5. Geometrical changes of the 10 uni-ply composite laminate in Run MW104B

<table>
<thead>
<tr>
<th>Press Cycle (°F/psi/hr)</th>
<th>Wt. (g)</th>
<th>Wt. Loss (g)</th>
<th>Wt. Loss (%)</th>
<th>Thickness (in)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ambient</td>
<td>12.170</td>
<td>---</td>
<td>---</td>
<td>0.065±.002</td>
</tr>
<tr>
<td>1</td>
<td>750/100/.25</td>
<td>12.128</td>
<td>0.042</td>
<td>0.056±.002</td>
</tr>
</tbody>
</table>
CONSOLIDATION MECHANISM

As noted above, a 12% difference in the degree of volume compaction is observed between panel MW103, which employed internal molding stops, and panel MW104, which did not. Despite the use of an identical molding cycle, i.e., 15 minute hold at 750°F and 100 psi, a dramatic difference in the consolidation qualities between these two composite panels was noted from the C-scans shown in Figures 3 and 5. Such a difference is attributable to the observed 12% difference in the volume compaction experienced by these two panels. The following discussion will address the origins which give rise to the observed variation of the volume compaction.

In the case of run MW103, two molding stops measured .250" in total width were used during the initial B-stage at the 650°F cure step. Prepreg plies were cut to 3.000" in the fiber direction and 2.750" in the lateral direction. The whole assembly (prepreg plies and molding stops) were fitted into the space of a 3.000" x 3.000" mold cavity. After the B-stage cure step the stops were removed. A 15 minute hold at 750 °F and 100 psi was then applied to the composite panel to complete the final consolidation step. At the completion of the molding cycle, the composite panel had spread to fill the mold dimensions of 3.000" x 3.000" and was fully consolidated. Obviously, the extra space provided by the removal of the molding stops during the final consolidation step contributes to the higher volume compaction level noted for this panel (MW103). Given the final panel (MW103) thickness of 0.058" (see Table 3), it is calculated that

\[
\frac{\text{Effective volume of stops}}{\text{Volume of panel}} = \frac{3.000" \times 0.250" \times 0.058"}{3.000" \times 3.000" \times 0.058"} = 0.0833
\]

where the effective volume of stops denotes the extra space (volume), provided by the removal of the stops, is available for the panel to spread laterally during the consolidation step. It shows that this effective space only accounts for an 8.33% difference in volume compaction discussed above between the fully consolidated panels MW103 and MW104. The remaining 3.67% (= 12% - 8.33%) difference in the degree of volume compaction must be accounted for by other reasons.

It is conceivable that, without stops (which act as side restraints), the unidirectional fiber/resin matrix deforms or spreads laterally from the center outward upon the application of pressure. The fiber/resin matrix moves as a unit in parallel with all the other fiber bundles. The space created by the removal of the stops is filled as the fiber/resin matrix comes in contact with the sidewalls of the mold. Such a spreading mechanism will effectively prevent a certain degree of external load from being absorbed by the otherwise intimate interply fiber-fiber contact, and

9
consequently allow a certain degree of interply fiber-fiber nesting to occur. The spreading and compaction mechanism allows further consolidation which leads to a dense void free laminate. The remaining 3.67% difference noted above is attributable to interply fiber nesting.

The observed higher degree of volume compaction with panel MW103 can therefore largely be attributed to these two reasons discussed above.

Unlike the prepreg systems made using the technique of solution prepregging, which contain resin matrices with limited imidization reaction, the LaRC-TPI prepreg system investigated here has been pultruded through a hot die after being impregnated by the slurry technology [2]. The resin matrix is essentially thermoplastic. This is evident by noting that there was only 0.5% weight loss occuring after B-staging at 650°F for 0.5 hours. Consequently, the molding step of B-staging with internal stops used for panel MW103 can be eliminated. It is only necessary to cut the prepreg plies 2.750" wide in the in-plane lateral dimension to facilitate the required interply fiber nesting mechanism to occur within the laminate during the consolidation stage.
COMPOSITE MECHANICAL PROPERTIES

The temperature and pressure cycles developed in this investigation have been shown to consistently produce composite panels from the AS-4/LaRC-TPI 1500 prepreg material with exceptional C-scan quality. However, C-scan is known only as a useful first line quality control tool. A composite laminate with good C-scan quality does not guarantee a comparable level of good mechanical properties. Because of the much lower consolidation pressure used in this work when compared to conventional molding processes, it is therefore necessary to evaluate the mechanical properties of these consolidated composite panels.

Short Beam Shear (SBS) Strength

A 21 ply 3x3 composite laminate was molded (Run MW105) by a vacuum press according to the cure cycle shown in Table 6. Prepreg was cut into 3.000" by 2.750" and then stacked in a 3.000" by 3.000" female mold cavity. B-staging of the laminate was performed at 600°F for 30 mins in the mold with two stops fitted to the two sides of the mold lateral to the fiber direction. Due to the use of molding stops, the laminate experienced null pressure. An unconsolidated panel with loosely compacted structure resulted at the completion of the B-stage.

B-stage of the panel was followed by a consolidation step. During this step, a pressure of 200 psi was applied at 750°F for 15 minutes. The resultant weight loss percentage and final panel thickness are comparable to those found in other experiments (Run MW103, for example). On a per ply basis, the final thickness of MW105, i.e., 0.113/21=.0054", is noted to be at the same level of magnitude as .0057" found in MW102, .0058" found in MW103 and .0056" found in MW104B, indicating a similar degree of interply fiber-fiber nesting being achieved. A C-scan of the panel is shown in Figure 7.

Table 6. Geometrical changes of the 21 uni-ply composite laminate in Run MW105

<table>
<thead>
<tr>
<th>Press Cycle (°F/psi/hr)</th>
<th>Wt. (g)</th>
<th>Wt. Loss (g)</th>
<th>Wt. Loss (%)</th>
<th>Thickness (in)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ambient</td>
<td>25.950</td>
<td>---</td>
<td>---</td>
<td>---</td>
</tr>
<tr>
<td>1</td>
<td>600/0/.5</td>
<td>25.779</td>
<td>0.171</td>
<td>0.659</td>
</tr>
<tr>
<td>2</td>
<td>750/200/.25</td>
<td>25.178</td>
<td>0.772</td>
<td>2.975</td>
</tr>
</tbody>
</table>
Twenty seven (27) SBS specimens were machined from panel MW105 and tested at 4 temperatures. The test temperatures were room temperature, 93°C (200°F), 150°C (300°F) and 177°C (350°F). Results are shown in Figure 8. Also shown in Figure 8 are data reported by Ohta et al. [22] on the same composite system. The SBS strengths from panel MW105 are considered reasonably good. However, they are consistently lower than those obtained by Ohta et al. under all test temperatures. For example, a 20% lower SBS strength is observed at room temperature. The difference in properties could be attributed to different processing conditions as tabulated below:

<table>
<thead>
<tr>
<th>MW105</th>
<th>Ohta et al. [22]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pressure (psi)</td>
<td>200</td>
</tr>
<tr>
<td>Temperature (°F)</td>
<td>750</td>
</tr>
<tr>
<td>Time (mins)</td>
<td>15</td>
</tr>
<tr>
<td>Resin Content (% wt.)</td>
<td>47.7</td>
</tr>
<tr>
<td>Prepreg ply</td>
<td>21</td>
</tr>
</tbody>
</table>

It is readily noticeable from these two sets of molding conditions that significant differences exist in both magnitude and duration of the molding pressures. In order to investigate the effect of the molding pressure duration on the SBS strength of the composite laminate, a new panel (Run MW107) was molded according to the molding cycle tabulated in Table 7.

The usual B-stage step at 600°F was performed with the aid of molding stops. With the molding stops removed, the composite panel was then subjected to the consolidation step with 200 psi applied at 700°F for 75 minutes. The percentage weight loss and the per ply final panel thickness are comparable to those found in panel MW105. A C-scan of the panel is shown in Figure 9.

Table 7. Geometrical changes of the composite laminate (24 uni-ply, 3x3) in Run MW107

<table>
<thead>
<tr>
<th>Press Cycle (°F/psi/hr)</th>
<th>Wt. (g)</th>
<th>Wt. Loss (g)</th>
<th>Wt. Loss (%)</th>
<th>Thickness (in)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ambient</td>
<td>27.690</td>
<td>---</td>
<td>---</td>
<td>---</td>
</tr>
<tr>
<td>1 600/0/.5</td>
<td>27.548</td>
<td>0.142</td>
<td>0.513</td>
<td>0.224±.002</td>
</tr>
<tr>
<td>2 700/200/1.25</td>
<td>27.027</td>
<td>0.663</td>
<td>2.394</td>
<td>0.120±.002</td>
</tr>
</tbody>
</table>
Twenty two (22) SBS specimens were machined from panel MW107 and tested at 4 temperatures as in the case of MW105 discussed above. Results are shown in Figure 10. Also shown in Figure 10 are data reported by Ohta et al. on the same composite system. It is noted that an increase in the duration of consolidation pressure resulted in a 15% increase (16.5 ksi in MW107 vs. 14.3 Ksi in MW105) in the room temperature SBS strength. The SBS strength at room temperature reaches 92% of the value reported by Ohta et al.. Considering that only one fifth of the consolidation pressure (200 psi in MW107 vs. 1,000 psi in Ohta et al.) was actually employed, this result is noteworthy.

**Flexural Strength**

Flexural strength specimens were prepared from panel MW103 and MW104B, and measured under room and elevated (177°C) temperatures. Five specimens were used at each temperature. The results are shown in Figure 11. Also included in Figure 11 are those data reported by Ohta et al. [22] on the same composite system. As noted from the figure, the flexural strength from the panels molded in this study is inferior at both test temperatures. The room temperature flexural strength, for example, reaches only 61% of that obtained by Ohta et al..

Again, the observed difference can be attributed to the different processing conditions as tabulated below:

<table>
<thead>
<tr>
<th></th>
<th>MW103</th>
<th>MW104B</th>
<th>Ohta et al. [22]</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Pressure (psi)</strong></td>
<td>100</td>
<td>100</td>
<td>1,000</td>
</tr>
<tr>
<td><strong>Temperature (°F)</strong></td>
<td>750</td>
<td>750</td>
<td>700</td>
</tr>
<tr>
<td><strong>Time (mins)</strong></td>
<td>15</td>
<td>15</td>
<td>60</td>
</tr>
<tr>
<td><strong>Resin Content (% wt.)</strong></td>
<td>48.1</td>
<td>46.2</td>
<td>42.0</td>
</tr>
<tr>
<td><strong>Prepreg ply</strong></td>
<td>10</td>
<td>10</td>
<td>8</td>
</tr>
</tbody>
</table>

Significant differences exist in both magnitude and duration of the molding pressures. From the study of the effect of the molding pressure duration on the SBS strength discussed above, it is believed that, with the lower consolidation pressure employed, the enhanced consolidation quality resulting from the prolonged pressure application will also lead to an enhanced flexural strength of the composite laminate.
CONCLUSIONS

The following conclusions can be drawn from this investigation:

1) Excellent 3x3 composite panels were produced as evidenced by the C-scans, in spite of wide fluctuations in the resin content of random specimens taken from the prepreg. The specimens varied in resin content from 24.3 to 42.8% by weight.

2) Despite the extreme variation in resin content, the level of the thru-thickness impregnation of the tow bundles and the uniformity of fiber tow distribution in the prepreg system as shown by the SEM photomicrographs in Figure 1 are adequate for the compression molding process. The melt viscosity for the resin matrix (less than 1,000 poises at 750°F [20]), is also proven to be adequately low for the required flow properties.

3) It has been demonstrated that the composite consolidation quality can be significantly enhanced by means of internal molding stops. With the aid of molding stops for the prepreg system currently investigated, a composite panel with good C-scan quality can be molded with a consolidation pressure as low as 100 psi. The benefits gained through the aid of internal molding stops are applicable for prepreg systems which undergo imidization reactions during the molding cycle [23]. For the thermoplastic matrix studied, the benefits to the composite consolidation quality using molding stops are realized from the following mechanism:

Before the final consolidation stage of the cure cycle, the molding stops are removed. The removal of the molding stops creates an excess volume within the cavity of the mold, which allows in-plane lateral movement of the fiber/resin matrix in response to the applied consolidating pressure. Such movement shears the resin matrix and allows slippage and nesting of tow filaments to occur between plies. A smaller degree of applied pressure is absorbed by the otherwise intimate inter fiber-fiber contact which exists in the prepreg pattern cut to the full dimensions of the mold in the conventional compression molding process. Consequently, better consolidation quality is achieved, minimizing residual void content within the laminate.

Simple compression molding of a full (3.000"x3.000") cut pattern will bring the tow filaments in contact and without lateral flow will not allow slippage and nesting to occur. A
higher degree of external load is offset by intimate inter fiber-fiber contact, consequently producing thicker composite panels with voids and poor C-scans.
REFERENCES


AS-4/LaRC-TPI 1500 Composite Prepreg

SEM Magnification: 80x

Figure 1. Scanning Electron Micrographs of the AS-4/LaRC-TPI 1500 (HFG) prepreg system.

SEM Magnification: 200x
**Cure Cycle** - 650°F/0psi/.5hrs, 750°F/500psi/.2hrs.

<table>
<thead>
<tr>
<th>File: MW101</th>
<th>Pulse-Echo Amplitude Plot</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td></td>
</tr>
<tr>
<td>90</td>
<td></td>
</tr>
<tr>
<td>80</td>
<td></td>
</tr>
<tr>
<td>70</td>
<td></td>
</tr>
<tr>
<td>60</td>
<td></td>
</tr>
<tr>
<td>50</td>
<td></td>
</tr>
<tr>
<td>40</td>
<td></td>
</tr>
<tr>
<td>30</td>
<td></td>
</tr>
<tr>
<td>20</td>
<td></td>
</tr>
<tr>
<td>10</td>
<td></td>
</tr>
<tr>
<td>0</td>
<td></td>
</tr>
</tbody>
</table>

**C-Scan Gain: 60%**

<table>
<thead>
<tr>
<th>File: MW101</th>
<th>Pulse-Echo Amplitude Plot</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td></td>
</tr>
<tr>
<td>90</td>
<td></td>
</tr>
<tr>
<td>80</td>
<td></td>
</tr>
<tr>
<td>70</td>
<td></td>
</tr>
<tr>
<td>60</td>
<td></td>
</tr>
<tr>
<td>50</td>
<td></td>
</tr>
<tr>
<td>40</td>
<td></td>
</tr>
<tr>
<td>30</td>
<td></td>
</tr>
<tr>
<td>20</td>
<td></td>
</tr>
<tr>
<td>10</td>
<td></td>
</tr>
<tr>
<td>0</td>
<td></td>
</tr>
</tbody>
</table>

**C-Scan Gain: 90%**

Figure 2. C-scans of composite panel MW101.
Figure 3. C-scans of composite panel MW102.
AS-4/LaRC-TPI 1500 Composite Panel MW103


C-Scan Gain: 60%

C-Scan Gain: 90%

Figure 4. C-scans of composite panel MW103.
AS-4/LaRC-TPI 1500 Composite Panel MW104

Cure Cycle - 750°F/100psi/.25hrs.

C-Scan Gain: 60%

C-Scan Gain: 90%

Figure 5. C-scans of composite panel MW104.
Cure Cycle - 750°F/100psi/.25hrs.

File: MW104B
Pulse-Echo
Amplitude Plot

C-Scan Gain: 60%

File: MW104B
Pulse-Echo
Amplitude Plot

C-Scan Gain: 90%

Figure 6. C-scans of composite panel MW104B.
AS-4/LaRC-TPI 1500 Composite Panel MW105


C-Scan Gain: 60%

C-Scan Gain: 90%

Figure 7. C-scans of composite panel MW105.
AS-4/LaRC-TPI 1500 (HFG) COMPOSITE

![Graph showing Short Beam Shear (SBS) strength of composite panel MW105, compared to those reported by Ohta et al. [22].](image)

Figure 8. Short Beam Shear (SBS) strength of composite panel MW105, compared to those reported by Ohta et al. [22].
AS-4/LaRC-TPI 1500 Composite Panel MW107

Cure Cycle - 600°F/0psi/.5hrs., 750°F/200psi/1.25hrs.

C-Scan Gain: 60%

Figure 9. C-scans of composite panel MW107.
Figure 10. Short Beam Shear (SBS) strength of composite panel MW107, compared to those reported by Ohta et al. [22].
Figure 11. Flexural strength of the composite panels MW103 and MW104B, compared to those reported by Ohta et al. [22].
Poor processability of fiber reinforced high performance polyimide thermoplastic resin composites is a well recognized issue which, in many cases, prohibits the fabrication of composite parts with satisfactorily consolidated quality. Without modifying the resin matrix chemistry, improved compression molding procedures have been proposed and investigated with the AS-4/LaRC-TPI 1500 High Flow Grade (HFG) prepreg system in this study. Composite panels with excellent C-scans can be consistently molded by this method under 700°F and a consolidation pressure as low as 100 psi. A mechanism for the consolidation of the composite under this improved molding technique is discussed. This mechanism reveals that a certain degree of matrix shear and tow filament slippage and nesting between plies occur during consolidation, which leads to a reduction of the consolidating pressure necessary to offset the otherwise intimate inter fiber-fiber contact and consequently achieves a better consolidation quality. Outstanding short beam shear strength and flexural strength were obtained from the molded panels. A prolonged consolidation step under low pressure, i.e., 100 psi at 700°F for 75 minutes, was found to significantly enhance the composite mechanical properties.