HIGH-TOUGHNESS, CONTROLLED FLOW BMI PREPREG FOR AEROSPACE APPLICATION

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ABSTRACT

Existing bismaleimide (BMI) prepreg systems offer high use temperatures for aerospace applications, however they are also prone to low toughness and excessive resin flow during cure. To overcome these difficulties, a novel BMI prepreg system – T1100G/4000 – has been developed, featuring toughening and flow control technologies. It exhibits high compression after impact (CAI) equivalent to a toughened epoxy system, and consistent cured ply thickness (CPT) due to excellent flow control. Additionally, T1100G/4000 showed excellent open hole tensile strength (OHT) and no microcracking after 1200 thermal cycles. The development of this new system as well as the system's performance will be explored in this paper.

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1. INTRODUCTION

Bismaleimide (BMI) resins are known for their high use temperatures, but also for their brittle behavior and difficult processing. Over the last few decades, work has been done throughout the composites industry to improve their performance and make them more viable in fiber reinforced plastic (FRP) composites. This includes the addition of thermoplastics and other toughening agents to improve the use of BMI in structural applications [1]. Toray has developed a BMI toughened prepreg that further advances this field of study, including a 40% improvement in compression after impact (CAI) over current commercially available BMI prepregs. Furthermore, in tensile properties, a 35% improvement in room temperature open hole tensile strength (OHT) was observed. Cured samples were subject to thermal cycling, and no microcracks are present up to 1200 thermal cycles.

The mechanical and processing improvements discussed here are accomplished by combining Toray's specialties in fiber development and resin formulation. Toray's BMI 4000 resin is formulated with the intent to make processing similar or equivalent to an aerospace grade epoxy, and is paired with Toray's high-strength, high-modulus fiber TORAYCA T1100G for improved mechanical performance.

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2. BACKGROUND ON BMI

BMI composites have been in use since the 1980s, and have grown in their number of applications as processing and toughening technologies have broadened the appeal to industry. An iconic example of BMI in industry is the F-22 Raptor, where BMI was implemented throughout the design due to its high strength and heat resistance [2].

The general structure of BMI is provided in Figure 1. The R-group often includes a benzene ring structure, though it sometimes can be an aliphatic chain. The maleimide groups are capable of bonding with comonomers or other maleimide groups.

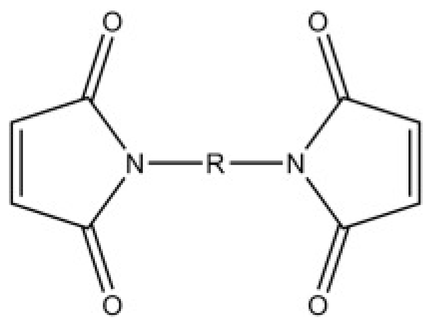


Figure 1. Generic chemical structure of BMI [1].

3. EXPERIMENTATION

The BMI prepreg discussed in this paper was produced at Toray Composite Materials America, Inc. with an optimal resin content of 35% (%Rc) and a fiber area weight (FAW) of 145 gsm. Actual resin content is discussed in the results section.

Unless otherwise described in this paper, composite laminates were cured according to the following parameters:

- Pressure: 0.6 MPa
- Curing conditions: 143 °C for 2 hours + 190 °C for 2 hours (ramp rate: 1.7 °C/min, cool down rate: 2 °C/min)
- Post cure conditions: 210 °C for 4 hours or 227 °C for 4 hours (ramp rate: 1.7 °C/min, cool down ramp rate: 2 °C/min)

All tests referred to in this paper are described below:

- Viscosity: parallel plate fixture on ARES apparatus (Patel Scientific), 40 mm diameter plate with 0.6 mm gap, frequency of 10 rad/s, ramp rate of 2 °C/min
- Resin flow index: an index value equal to the integral of the inverse viscosity curve from 50 °C to beginning of gelation
- Acid digest: ASTM D3171
- Tack: ASTM D8336-21 (for more information, please refer to [3]), 55% relative humidity

- Glass transition temperature (Tg): measured as the onset of the G' curve using a DMA850 (TA Instruments) with 3-point bending clamp fixture and ramp rate of 5 °C/min
- CAI: ASTM D7137, $[+45°/0°/-45°/90°]_{3S}$
- OHT: ASTM D5766, $[+45°/0°/-45°/90°]_{S}$
- Open hole compressive strength (OHC): ASTM D6484, $[+45°/0°/-45°/90°]_{2S}$
- Inter-laminar shear strength (ILSS): ASTM D2344, $[0°]_{20}$
- Thermal cycling condition in oven: 177 °C for 15 minutes then -55 °C for 15 minutes, ramp and cool down rate of 5 °C/min

4. RESULTS

4.1 Flow control and handling

BMI is known for extremely low viscosity at elevated temperatures, sometimes even below 1 P, necessitating extra bagging procedures such as edge damming around the laminate so that excessive resin bleed out doesn't occur. Resin flow can be managed through the addition of thermoplastics, fillers, and accelerators. However, while it is desirable to increase viscosity at elevated temperatures, many of these techniques also severely reduce tack of the prepreg, rendering the prepreg incompatible with certain layup techniques. It is important, therefore, to balance the tack and the viscosity at elevated temperatures for the intended application.

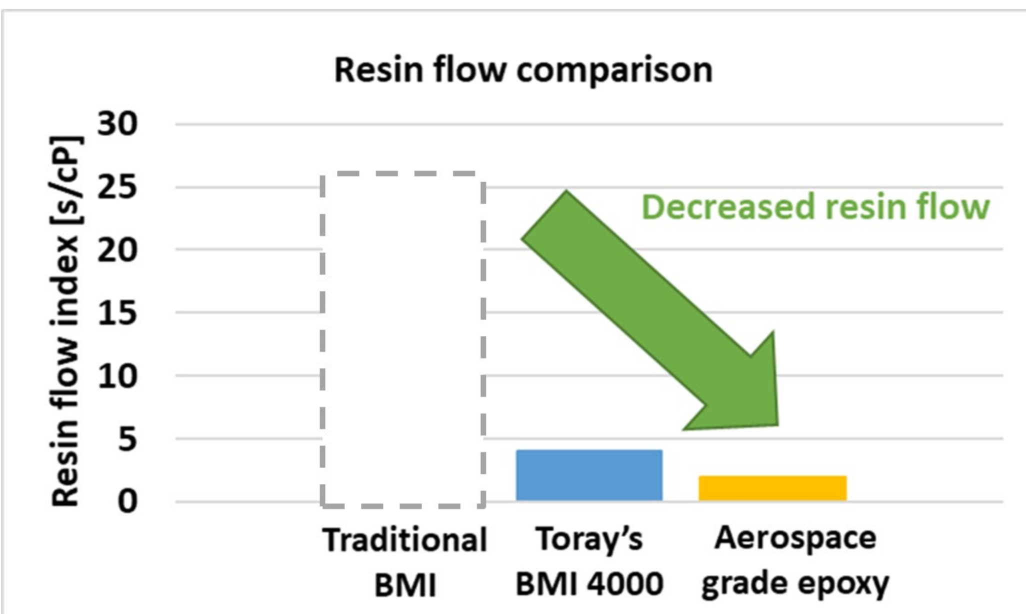


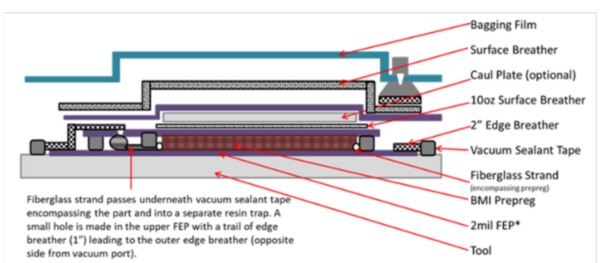
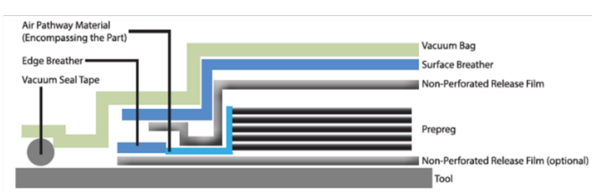
Figure 2.Resin flow index comparison between traditional BMI, Toray's BMI 4000, and an aerospace grade epoxy. A higher resin flow index indicates a resin with greater tendency to flow during cure due to

low viscosity. The gray dotted outline representing traditional BMI shows the variety of ranges of flow index possible in commercially available BMIs.

Figure 2 shows the comparison between a traditional BMI, Toray's BMI 4000, and a typical aerospace grade epoxy. The resin flow is reported as an index value equal to the integral of the inverse viscosity curve from 50 °C to beginning of gelation. A higher resin flow index indicates a resin with greater tendency to flow during cure due to low viscosity. As described earlier in this section, traditional BMI can have a very high flow index, up to a value of roughly 25 s/cP, compared to aerospace grade epoxies which usually exhibit a flow index between 1 and 10 s/cP. Toray's BMI 4000 has a flow index that falls within the same range as aerospace grade epoxy, a significant improvement over some traditional BMIs.

This improvement allows for decreased time and material costs in bagging procedures that have previously been necessary in order to contain resin within the laminate during cure. T1100G/4000 was cured in an autoclave using both an edge damming method for traditional BMIs and an epoxy bagging method. The cured ply thickness (CPT) was equivalent for both laminates regardless of bagging method (see Table 1 and Figure 3). Additionally, the cured laminates maintained their resin content and exhibited less than 1% void content as determined by acid digest.

Table 1. Comparison of T1100G/4000 laminates cured in autoclave with different bagging methods. The CPT and resin content are the same or similar for both methods.

<table>
<tr><th rowspan="2">Bagging method</th><th rowspan="2">Schematic</th><th rowspan="2">Average panel thickness [mm]</th><th colspan="2">CPT</th><th colspan="2">Resin content</th></tr>
<tr><th>Average [mm/ply]</th><th>Standard deviation [mm/ply]</th><th>Prepreg [wt%]</th><th>Cured panel [wt%]</th></tr>
<tr><td rowspan="2">Traditional BMI bagging</td><td rowspan="2">
</td><td rowspan="2">1.82</td><td rowspan="2">0.15</td><td rowspan="2">0.003</td><td>35.80%</td><td>35.87%</td></tr>
<tr><td colspan="2">Δ = + 0.07%</td></tr>
<tr><td rowspan="2">Epoxy bagging</td><td rowspan="2">
</td><td rowspan="2">1.82</td><td rowspan="2">0.15</td><td rowspan="2">0.002</td><td>35.80%</td><td>35.61%</td></tr>
<tr><td colspan="2">Δ = - 0.19%</td></tr>
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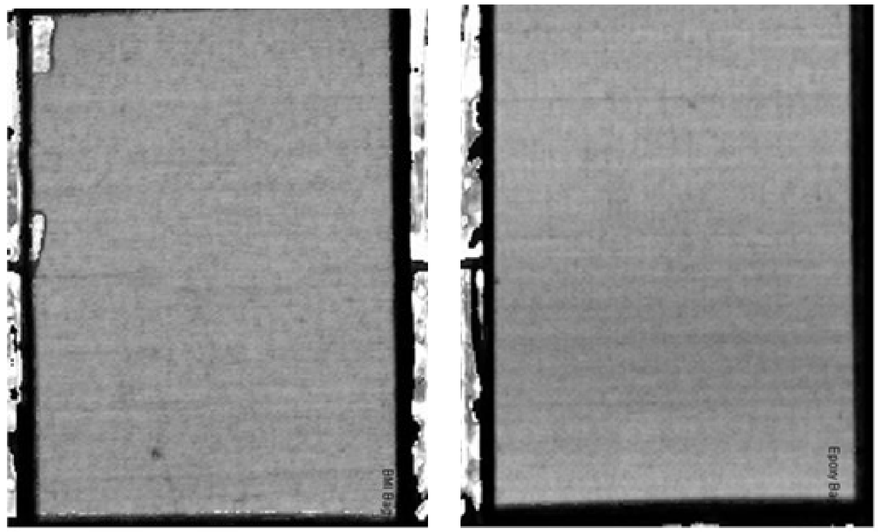
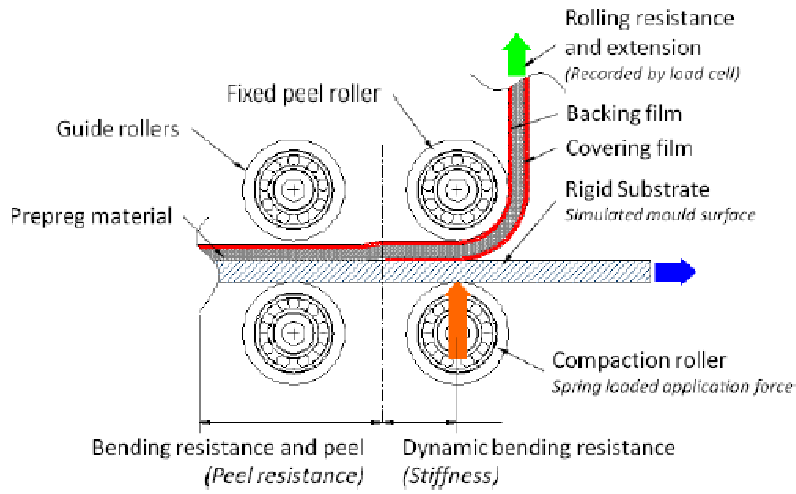


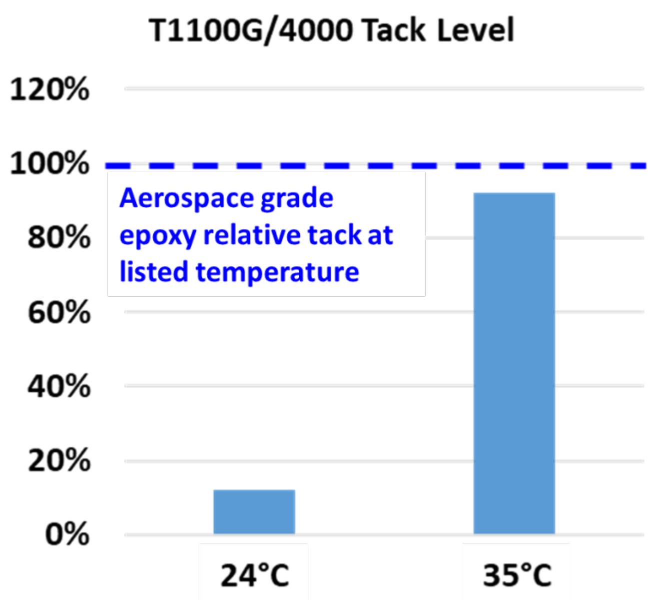
a) b)

Figure 3. Non-destructive inspection (NDI) scans of T1100G/4000 using (a) traditional BMI bagging method and (b) epoxy bagging method. The images show low void content and good laminate quality regardless of bagging method.

The prepreg is able to be laid up by hand under ambient conditions, and will conform to an aluminum tool at around 30 °C. It was found that T1100G/4000 tack is readily adjustable with temperature (see Figure 4), and that room temperature tack is suitable for automated fiber placement (AFP), automated tape laying (ATL), and other such automated processes.



a)



b)

Figure 4. Tack testing according to (a) ASTM D8336-21 [4] with data shared in (b).

4.2 Mechanical properties

Toray's T1100G/4000 system has pushed the performance envelope for BMIs, as shown in Figure 5. CAI was increased 40% due to toughening technology utilized in the 4000 resin (1500 in-lb/in loading). OHT at room temperature ambient (RTA) conditions was increased by 35% over traditional BMI prepreg due to Toray's high-strength, high-modulus fiber, TORAYCA T1100G. OHT also showed improvement for -54 °C cold temperature ambient (CTA) condition. OHC is maintained, even at 177 °C. The ETW condition ("elevated temperature wet") describes a soak condition of two weeks in water at 71 °C, then testing at an elevated temperature, in this case 177 °C.

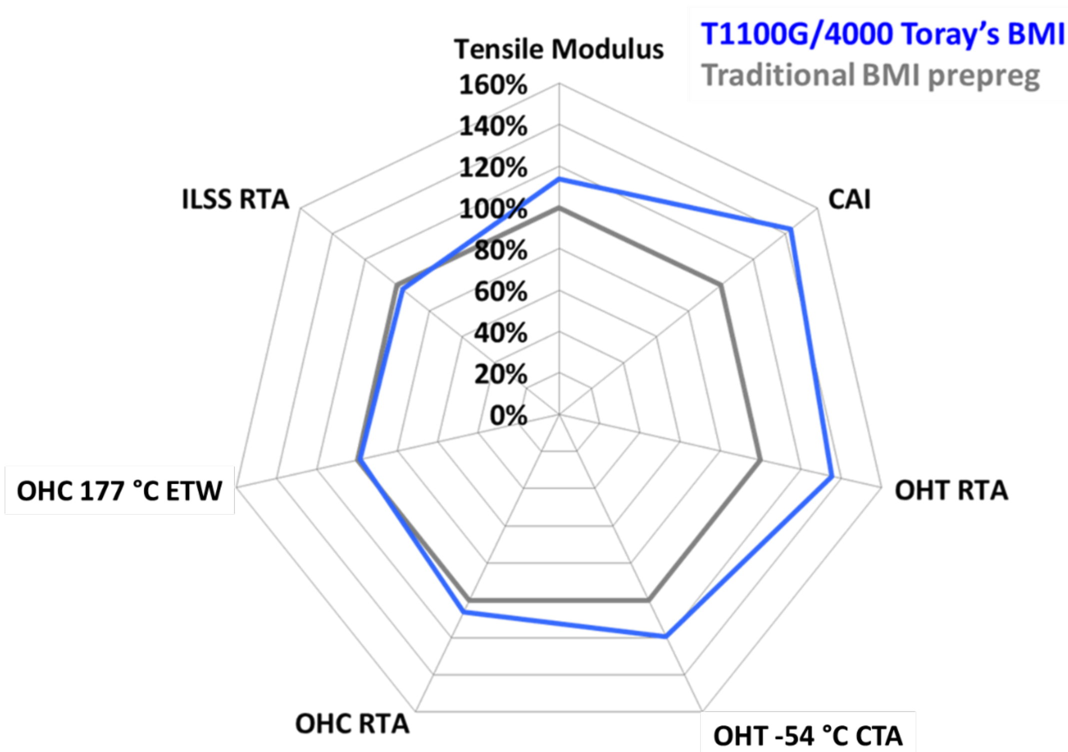


Figure 5. Radar chart comparing key mechanical properties of traditional BMI prepreg and Toray's T1100G/4000. Note that CAI data shared in this chart was impacted at a loading of 1500 in-lb/in.

Figure 6 shows the property trade-off between the Tg and CAI for T1100G/4000. Increasing Tg via increasing crosslinking density through extended cure can risk a decrease in toughness, yet T1100G/4000 does not show a significant drop in CAI as post cure temperature increases. The highest Tg shown in Figure 6 is roughly 250 °C, and with even higher post cure temperatures the dry Tg can be up to 290 °C (wet Tg, 205 °C). CAI, however, has not yet been tested at these higher post cure temperatures.

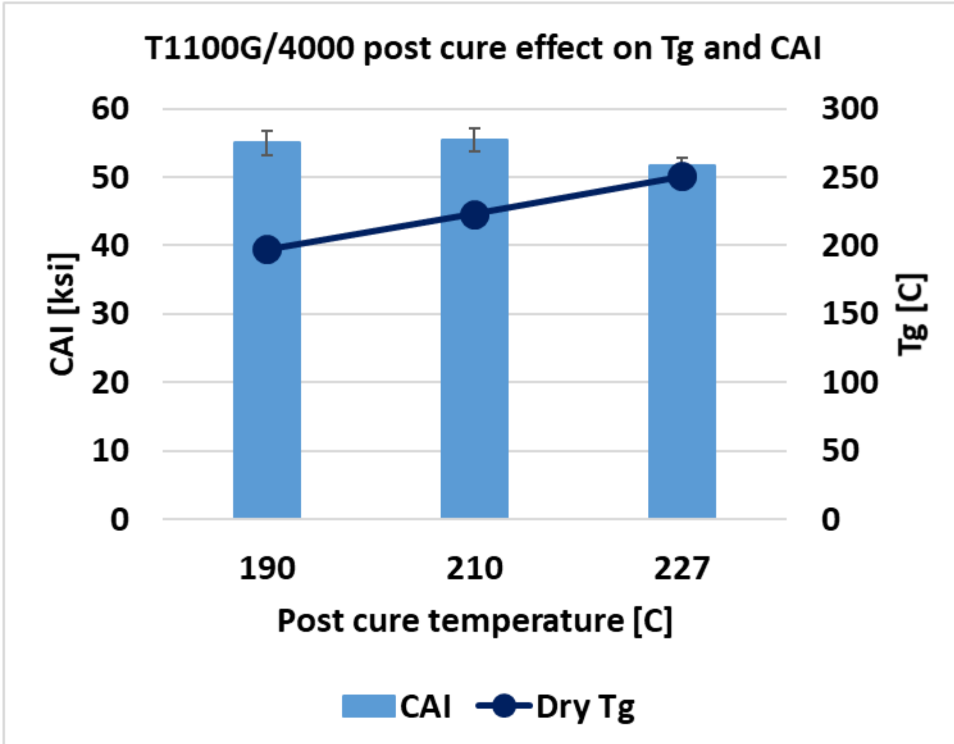
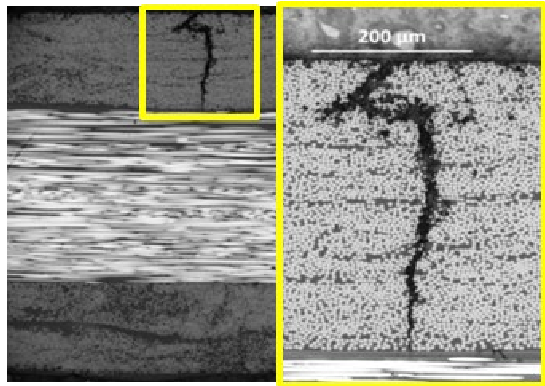


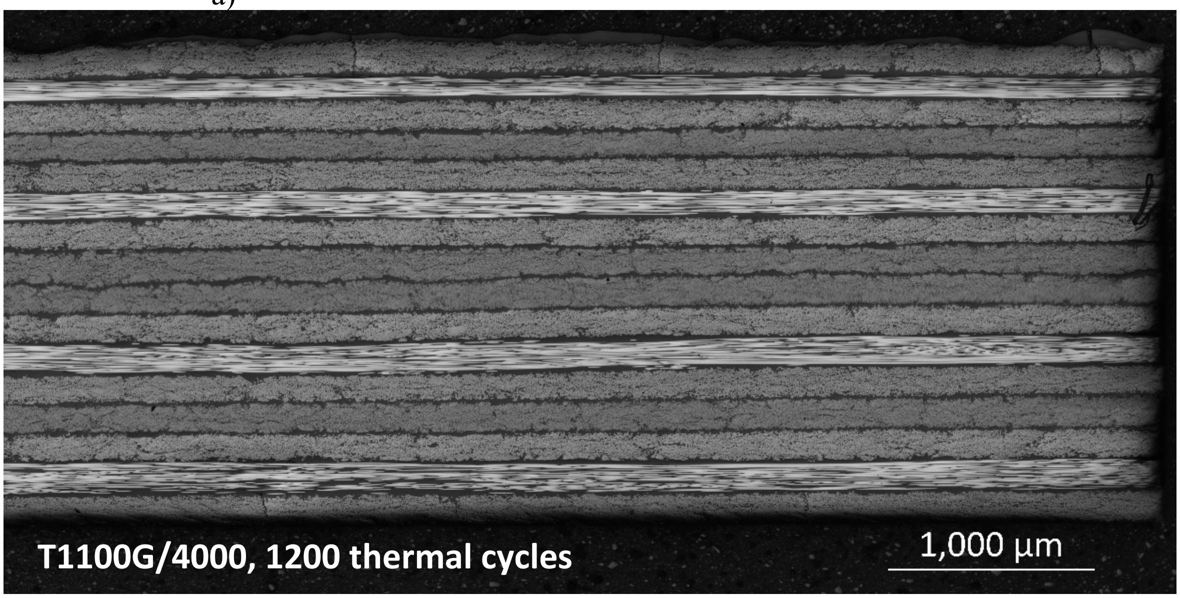
Figure 6. Post cure study comparison between traditional BMI prepreg and T1100G/4000. Note that the loading for CAI in this study was 1000 in-lb/in. All post cures were conducted for 4 hours.

4.3 Thermal cycling and microcrack studies

Brittle materials can exhibit small defects known as microcracks during cure, post cure, or service at fluctuating temperatures. Increasing toughness of the laminate is one method to reduce microcracking. For microcrack studies, a quasi-isotropic laminate ($[+45/0/-45/90]_{2S}$) was laid up, cured, and cut to a sample with dimensions of 7.62 cm x 10.16 cm (3 in x 4 in). Samples were placed in an oven following a temperature program of 177 °C for 15 minutes then -55 °C for 15 minutes, with a ramp rate of 5 °C/min. Small sections of these samples were then prepared for microscopy. Under these conditions, T1100G/4000 demonstrates microcrack resistance up to 1200 thermal cycles.



a)



b)

Figure 7. An example of microcracking in epoxy is provided in (a) for comparison, while an actual image of T1100G/4000 after 1200 thermal cycles is shown in (b). No microcracks are present within the bulk of the material.

5. CONCLUSIONS

This paper demonstrated the improvement of Toray's T1100G/4000 prepreg over traditional BMI in the areas of flow control, handling, tensile properties, and toughness. Toray's BMI 4000 has a resin flow index similar to that of an aerospace grade epoxy, allowing for easier manufacturing, handling, and curing. It was shown that T1100G/4000 has an increased OHT of 35%, an increased CAI of 40%, and maintains OHC performance. Future modifications of T1100G/4000 seek to improve compressive strength in order to push the BMI performance envelope even further.

6. REFERENCES

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[2] Boyd. J. "Bismaleimide composites come of age: BMI science and applications". *SAMPE Journal*. Vol. 35, No. 6, November/December 1999, pp 13-22.

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