PROPERTIES OF PMR POLYIMIDE COMPOSITES MADE WITH IMPROVED HIGH STRENGTH GRAPHITE FIBERS

Raymond D. Vannucci
Lewis Research Center
Cleveland, Ohio

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Raymond D. Vannucci
National Aeronautics and Space Administration
Lewis Research Center
Cleveland, Ohio 44135

ABSTRACT

Recent graphite fiber developments have resulted in high strength, intermediate modulus graphite fibers having improved thermo-oxidative resistance. These improved fibers, obtained from various commercial suppliers, were used to fabricate PMR-15 and PMR-II polyimide composites. Studies were performed to investigate the effects of the improved high strength graphite fibers on composite properties after exposure in air at 600°F. The use of the more oxidatively resistant fibers did not result in improved performance at 600°F. Two of the improved fibers were found to have an adverse effect on the long-term performance of PMR composites. The influence of various factors such as fiber physical properties, surface morphology and chemical composition are also discussed.

1. INTRODUCTION

PMR polyimides were developed at the NASA Lewis Research Center in response to the need for processable, high temperature resistant matrix resins for fiber reinforced composites. The advantages and versatility of PMR polyimides have been reviewed. At the present time two versions of PMR differing in chemical composition have been identified. The earlier version, PMR-15, is readily available in prepreg form from various commercial suppliers. Graphite fiber reinforced composites prepared with the more recently developed second generation material, PMR-II, exhibited improved performance at 600°F. Because of their excellent processing and elevated temperature performance characteristics, PMR materials are now gaining wide acceptance as engineering materials for high temperature applications. The fibers of primary interest for these applications have been the high strength, intermediate modulus graphite fibers. Recent studies have shown that the thermo-oxidative resistance of high strength graphite fiber reinforced polyimide composites is highly dependent on the oxidative resistance of the fiber. Primary factors contributing to poor fiber oxidative resistance are fiber sodium content and carbon content. The use of early technology high strength graphite fiber having low oxidative
resistance limited the useful life of PMR-15 composites in air at 600°F to about 600 hours. PMR composites prepared with Celion 6000 (a later technology, high strength, low sodium content) graphite fiber exhibited a useful life in air at 600°F of about 1200 hours. More recently, high strength, intermediate modulus, graphite fibers exhibiting significantly improved thermo-oxidative resistance have been introduced by Hercules (HTS-2), Union Carbide (Thornel B) and Great Lakes Fiber (Fortafil 3).

The purpose of this study was to investigate the effects of these improved high strength, intermediate modulus, graphite fibers on the mechanical properties and thermo-oxidative stability of PMR-15 and PMR-II polyimide composites. Composite weight loss and mechanical property retention characteristics were determined in air at 600°F. This paper also compares the effects of fiber physical properties, surface morphology and chemical composition on PMR composite properties.

2. EXPERIMENTAL PROCEDURES

2.1 MONOMERS AND PMR SOLUTIONS

The monomers used in this study are shown in Table I. The monomethyl ester of 5-norbornene-2,3-dicarboxylic acid (NE), 4,4'-methylenedianiline (MDA), para-phenylene-diamine (PPDA), and 3,3',4,4'-benzophenonetetracarboxylic dianhydride (BTDA) were obtained from commercial sources. The 4,4'-[hexafluoropropylidene]-bis(phthalic anhydride) (HFDA) was prepared according to Serafini, Vannucci and Alston. The dimethyl ester of 3,3',4,4'-benzophenonetetracarboxylic acid (BTDE) and the dimethyl ester of 4,4'-[hexafluoropropylidene]-bis(phthalic acid) (HFDE) were prepared as 50 weight percent solutions by refluxing a suspension of the corresponding anhydrides in anhydrous methanol until the solids dissolved and then for an additional two hours.

The PMR monomer stoichiometry for PMR-15 solutions was 2 NE/3.087 MDA/2.087 BTDE. The solutions were prepared at room temperature by dissolving the monomers in a calculated amount of anhydrous methanol to give 50 weight percent solutions. The PMR-II solutions were prepared in a similar manner, using a monomer stoichiometry of 2NE/2.67 PPDA/1.67 HFDE.

2.2 GRAPHITE FIBERS

The following graphite fibers were used for composite fabrication and fiber weight loss studies: HTS-2, Fortafil-3, Thornel B (both sized and unsized) and Celion 6000. Forced air convection ovens were used for long-term isothermal exposure of bare fibers and composites at 600°F. The air change rate was 100 cm³/min. Fiber elemental composition was determined using a combination of emission and X-ray fluorescence spectroscopy and by combustion. Transmission electron photomicrographs of fiber surface replicas were used to study fiber surface morphology.

2.3 COMPOSITE FABRICATION

Prepreg tapes were made by drum winding and impregnating graphite fiber with PMR solutions calculated to yield composites having 58 volume percent fiber. The prepreg tapes were dried on the rotating drum for 1 hour at room temperature and then for another hour at 122°F to reduce the solvent content to a level that gave flexible tapes without excessive tack. The tapes were removed from the drum, cut into 3 inch by 10 inch plies and stacked unidirectionally to produce 9-12 ply laminates. Each prepreg stack was placed into a preforming mold and staged for 1 hour at 400°F under a pressure of approximately 0.1 psi. After staging the stack was then placed into a matched metal die and a thermocouple attached to the die. Composites were then molded by placing the die into a press heated to 600°F and applying a pressure of 500 psi when the die temperature had reached 450°F. After reaching 600°F, pressure and temperature were maintained for 1
hour. The composites were then cooled under pressure to 400°F, removed from the mold, and post-cured in air at 600°F for 16 hours.

2.4 COMPOSITE TESTING

Prior to specimen preparation all laminates were inspected for acceptance using an ultrasonic C-scan technique. Flexural strength tests were performed in accordance with ASTM D-790 using a three-point loading fixture and a span of 2.0, 2.5 or 3.0 inches. The thicknesses of the laminates ranged from 0.080 inch to 0.110 inch. The span/depth ratio used ranged from 24 to 29.

The rate of center loading for flexural testing was 0.5 inch/min. Interlaminar shear strength tests were performed in accordance with ASTM D-2344 at a constant span/depth ratio of 5. Elevated temperature tests were conducted in an environmental heating chamber following a 15 minute soak at the test temperature. The mechanical property values reported are averages of three or more tests at each condition.

The fiber content of composites was determined by H₂SO₄/H₂O₂ digestion.

3. RESULTS AND DISCUSSION

3.1 FIBER THERMO-OXIDATIVE STABILITY

Prior to composite fabrication, the thermo-oxidative stability (TOS) of the various graphite fibers used in this study was determined. Table II lists the percent of fiber weight loss after 1500 hours of exposure in air at 600°F. Also listed are the vendor data for fiber physical and mechanical properties. The fiber weight loss data show that HTS-2, Fortafil 3 (F-3) and Thornel B (T-B) are significantly more thermo-oxidatively resistant than Celion 6000 (C-6). It should be pointed out that the Celion (lot HTA-78322) fiber investigated in this study exhibited a weight loss of 25 percent compared to 17 percent found for the Celion (lot HTS-7-6YII) fiber studied in reference 6. The table also shows considerable differences in tensile strength and tensile modulus for fibers having the same density. The tensile strength and tensile modulus of F-3 fiber are 22 and 29 percent lower than the tensile modulus of T-B fiber. Two lots of T-B fiber were investigated, one without any size and one with a polyimide compatible size.

3.2 PMR-15 COMPOSITES

Ultrasonic C-scan inspection of the PMR-15 composites indicated that all of the composites were of high quality. In addition the void contents of all the laminates were found to be 1.0 percent or lower.

Because of a wide scatter in the data obtained for composites made from the first HTS-2 fiber examined (lot 113-4), a second lot of HTS-2 (lot 121-3) was also investigated. More consistent data were obtained for composites made from the second lot of HTS-2 fiber.

Table III shows the initial room temperature and 600°F mechanical properties of unidirectional graphite fiber reinforced composites fabricated with PMR-15 and C-6, F-3, HTS-2 and T-B. Comparison of room temperature interlaminar shear strength (ILSS) data shows that all the composites, except those made from both lots of HTS-2 fiber, exhibited essentially the same room temperature ILSS and that the 600°F ILSS of the composites made with the improved fiber was 7 to 24 percent lower than the 600°F ILSS of the C-6 composite. The ILSS data indicate that adequate resin/fiber bonding was obtained with all the fiber with (perhaps) the exception of the HTS-2 fibers. The room temperature flexural strengths of all the composites, except for composites made with HTS-2 (lot 113-4) and T-B (sized and unsized), are in good agreement with predicted values based on the simple rule of
mixtures. The room temperature flexural moduli of all the composites are lower than would be predicted using the simple rule of mixtures. The lower flexural properties at 600°F reflect the increased contribution of the matrix properties to composite properties at elevated temperatures.

In summary, the initial composite mechanical properties presented in the table indicate that all of the fibers (with the possible exception of HTS-2 as noted earlier) provided composites which could be used for long term isothermal exposure studies.

Figures 1-8 compare the weight loss characteristics and mechanical property retention of PMR-15 composites after isothermal exposure in air at 600°F. To facilitate the comparison of data obtained from the composites made with the improved fibers to the data obtained from composites made with C-6 fiber, the F-3, T-B and C-6 composite data are compared in Figures 1-4 and the HTS-2 and C-6 composite data are compared in Figures 5-8.

Figure 1 shows the composite weight loss behavior of PMR-15 composites made with C-6, T-B (sized), T-B (unsized) and F-3 fibers. Even though the Celion fibers used in this study exhibited lower TOS than the Celion fibers used in the earlier study, after 1500 hours of exposure in air at 600°F PMR-15 composites made with either lot of Celion exhibited identical levels of weight loss. The data presented in the figure clearly show that weight loss behavior during isothermal exposure of the composites was similar and the total composite weight losses exhibited by each of the composites after 1500 hours of exposure were not significantly different. Therefore, based on these results it may be concluded that the use of T-B (sized), T-B (unsized) and F-3 fibers did not provide composites having greater TOS than C-6 composites.

Figure 2 compares the flexural strength retention of C-6, F-3 and T-B/PMR-15 composites after exposure in air at 600°F. The figure shows that the T-B (sized and unsized) composites exhibited significantly lower flexural strength retention throughout the exposure time compared with the C-6 and F-3 composites. The C-6 and F-3 composites retained approximately 93 percent of their initial strength after 1500 hours of exposure.

Figure 3 compares flexural modulus retention of the C-6, F-3, and T-B/PMR-15 composites after isothermal exposure in air at 600°F. It can be seen that the flexural modulus retention of C-6, F-3 and sized T-B composites compare favorably out to 1500 hours of exposure while the modulus of the unsized T-B composite dropped considerably after 1200 hours of exposure.

The effect of isothermal exposure in air at 600°F on the ILSS on the PMR-15 composites is shown in figure 4. The figure shows that after 1500 hours of exposure the C-6 and F-3 composites retained 97 percent and 78 percent of their ILSS, respectively. The sized T-B and unsized T-B composites retained 87 percent and 65 percent of initial 600°F ILSS, respectively. While the use of sized T-B fiber resulted in only marginal improvement in T-B composite flexural strength over that of the unsized T-B composite, the sizing had a significant effect on the retention of T-B/PMR-15 ILSS at 600°F.

The results shown in figures 2-4 indicate that the use of the more thermo-oxidatively stable F-3 and T-B fibers did not result in PMR-15 composites with improved 600°F performance.

The properties of HTS-2/PMR-15 composites as a function of 600°F exposure time in air are shown in Figures 5-8. As mentioned earlier, due to the wide variation in HTS-2 composite properties after exposure at 600°F, two lots of HTS-2 fiber were investigated. Two of three PMR-15 composites made with lot 113-4 fiber exhibited rapid degradation and excessive loose surface
fiber after approximately 700 hours of exposure in air at 600°F making it impossible to machine test specimens. The third composite withstood 1500 hours of exposure at 600°F. A second lot of HTS-2 fiber, lot 121-3, provided more consistent composite data. These composites did not exhibit loose surface fiber until at least 1200 hours of exposure at 600°F.

The weight loss characteristics as a function of time in air at 600°F for HTS-2 and C-6 composites are shown in figure 5. It can be seen that both lots of HTS-2 fiber provided composites which exhibited the same weight loss behavior and total weight loss as did the C-6 composite. However, the data in the figure also clearly show the wide variation in weight loss properties both within and between the fiber lots. These data make it difficult to arrive at any conclusions concerning the propensity of HTS-2 fibers in providing PMR-15 composites with improved thermo-oxidative stability.

The variation of flexural strength and flexural modulus with exposure time in air at 600°F for HTS-2 and C-6 composites is presented in figures 6 and 7, respectively. As expected, there was considerable variation in the flexural strength retention characteristics of composites made from lot 113-4 fibers. Although the flexural strength retention of composites made from lot 121-3 fibers was found to be more consistent, it can be seen that their level of strength retention was considerably lower than that of the C-6 composite, especially after 400 hours of exposure.

The flexural moduli of the HTS-2 composites (figure 7) exhibited the same general behavior as was observed for composite weight loss and flexural strength, i.e., data scatter within and between fiber lots and more rapid degradation than C-6 composites. The higher levels of flexural moduli at room temperature (see Table III) and at 600°F, before and after extended exposure at 600°F, for some of the HTS-2 composites simply reflect the higher moduli of the HTS-2 fibers (see Table II).

The ILSS retention of HTS-2 and C-6 composites after long-term exposure to air at 600°F is shown in Figure 8. The figure shows that the ILSS of all of the HTS-2 composites are considerably lower than that of the C-6 composite throughout the exposure time. In view of the weight loss and flexural properties data for HTS-2 composites the ILSS results were not unexpected.

3.3 PMR-II COMPOSITES

Table IV lists the initial room temperature and 600°F mechanical properties of unidirectional reinforced C-6, HTS-2 (lot 121-3), F-3 and T-B composites made with PMR-II matrix resin. The data presented in Table IV follow the same general trends that were established with the PMR-15 matrix (see Table III). The agreement in fiber dominated (flexural) properties is not surprising and the similarity of the ILSS results indicate that the same level of interfacial bonding (chemical and/or mechanical) was attained with both matrices.

Figures 9-12 compare the weight loss characteristics and mechanical properties retention of the PMR-II composites after isothermal exposure in air at 600°F. The weight loss characteristics during extended exposure at 600°F are shown in Figure 9. In the earlier study (6) with Celion and PMR-II the composite weight loss, after 1800 hours in air at 600°F, was found to be 9 percent instead of the 16 percent found in this study. It can be seen in the figure that PMR-II composites made with HTS-2 and F-3 fibers resulted in significantly improved TOS during long term 600°F exposure compared to the C-6 composite. The HTS-2 and F-3 composites exhibited about a 6 percent weight loss after 1800 hours compared to 16 percent for the C-6 composite. The T-B composite exhibited a weight loss of 13 percent after 1800 hours of exposure. Although the 3 percent difference in composite weight loss between the
T-B and C-6 composites is not considered to be very significant, the 10 percent difference between HTS-2 and C-6 and between F-3 and C-6 is quite significant. These results are difficult to explain, particularly in view of the findings with PMR-15 which indicated that none of the more TOS fibers (with perhaps the exception of F-3) resulted in composites with improved TOS. It should be pointed out that, even after 1800 hours of exposure at 600°F, none of the PMR-II composites exhibited significant formation of loose surface fiber. In contrast, some of the PMR-15 composites exhibited significant formation of loose surface fiber after only 700 hours of 600°F exposure. To gain additional insight into other factors that might be responsible for the observed differences in composite weight loss behavior, the surface morphology and chemical composition of the fibers were investigated. The results of these studies will be discussed in a later section of the report.

Figure 10 compares the flexural strength retention of PMR-II composites after isothermal exposure in air at 600°F. It can be seen that the improved TOS of the HTS-2, F-3 and T-B composites did not result in improved flexural strength retention after long-term 600°F exposure. After 2000 hours of exposure the C-6 composite retained 80 percent of its strength compared to only a 31 percent retention for the T-B composite. After 1800 hours of exposure the F-3 and HTS-2 composites retained 73 and 60 percent of their initial strengths, respectively. It is interesting to note that, except for the C-6 composite, the relative order of TOS as assessed by composite weight loss and retention of composite flexural strength was the same, i.e., F-3 > HTS-2 > T-B.

Figure 11 shows the retention of flexural modulus for the PMR-II composites after exposure in air at 600°F. The figure shows that the C-6, F-3 and HTS-2 composites exhibited excellent retention of flexural modulus out to their exposure limits of 1800 to 2000 hours, while the T-B composite lost approximately 50 percent of its initial modulus after 2000 hours of exposure.

Figure 12 shows the ILSS retention of PMR-II composites after isothermal exposure in air at 600°F. The figure shows that the C-6, F-3 and HTS-2 composites retained 87, 85 and 69 percent of their initial 600°F ILSS, respectively, after 1800 hours of exposure. The T-B composite exhibited significantly lower ILSS retention, approximately 31 percent retention after 1800 hours of exposure. The relative order of TOS as assessed by ILSS retention is the same as noted earlier for composite weight loss and flexural strength retention.

In summary, the results of the data shown in figures 1-12 clearly indicate that PMR-15 and PMR-II composites made with the more TOS fibers did not exhibit improved long-term 600°F performance as compared with C-6/PMR-15 and PMR-II composites. Also, significant differences were observed in the composite weight loss of PMR-15 and PMR-II composites made from the more TOS fibers while the weight loss behavior of C-6/PMR-15 and PMR-II composites was comparable. Furthermore, although the improved fiber PMR-II composites exhibited lower weight loss compared to the C-6/PMR-II composites, the lower weight loss did not result in improved retention of composite mechanical properties during long-term exposure at 600°F. These findings suggest that, in addition to fiber TOS, fiber chemical composition and surface morphology may be other factors contributing to long-term elevated temperature performance of PMR composites.

3.4 FIBER CHEMICAL COMPOSITION AND SURFACE MORPHOLOGY

The major and minor elements present in the fiber are listed in Table V. As expected, the C-6 fiber is both lower in carbon and higher in sodium content than the more oxidatively stable fibers. By comparing the amounts of the other trace elements present in each of
the fibers, it can be seen that neither the HTS-2 or T-B fibers contained any trace element significantly higher in amount than that present in the C-6 fiber. The F-3 fiber contained significantly higher levels of sulfur, chlorine and potassium than the C-6 fiber. However, of the three more oxidatively stable fibers, the F-3 fiber provided PMR composites with the best overall elevated temperature performance. Taken as a whole, the fiber elemental analysis does not bring out any major factors which may be responsible for the high temperature performance of the improved fiber PMR composites.

Transmission electron photomicrographs (TEM) of surface replicas of the five fibers are shown in figures 13-17. Five fiber samples from each fiber lot were studied. The surface replicas shown are typical for each fiber. It can be seen that the surfaces of the HTS-2 (figs. 13 and 14) and the T-B (fig. 15) fibers are smoother and more regular (fewer and less pronounced surface striations) than those of the F-3 (fig. 16) and C-6 (fig. 17) fibers. The surface irregularities of the F-3 and C-6 fibers should result in enhanced mechanical bonding at the resin/fiber interface. The improved interfacial bonding should be reflected in higher initial mechanical properties and improved retention of mechanical properties at 600°F. Higher initial properties were found for the C-6 composites but not for the F-3 composites. The C-6 and F-3 composites, with the exception of ILSS retention for the F-3/PMR-15 composite, exhibited the best retention of elevated temperature mechanical properties. Thus, it appears that the enhanced resin/fiber interfacial bonding provided by fiber surface irregularities can provide better property retention characteristics.

4. SUMMARY OF RESULTS
1. HTS-2, Fortafil 3 and Thornel B graphite fibers were found to exhibit significantly improved thermo-oxidative stability at 600°F compared to Celion 6000 graphite fiber.
2. PMR-15 and PMR-II composites made with HTS-2, Thornel B and Fortafil 3 fibers did not exhibit improved 600°F performance compared with Celion 6000/PMR-15 and PMR-II composite performance.
3. The use of HTS-2 and Thornel B graphite fibers was found to adversely affect the long-term 600°F performance of PMR-15 and PMR-II composites.
4. The use of the more oxidatively stable fibers significantly lowered the 600°F weight loss characteristics of PMR-II composites. However, this was not reflected in improved retention of mechanical properties at 600°F.
5. Significant differences were found in the surface characteristics of Celion 6000, Fortafil 3, HTS-2 and Thornel B fibers which may have contributed to the 600°F performance of graphite fiber reinforced PMR-15 and PMR-II composites.

5. REFERENCES

TABLE I. - MONOMERS USED FOR POLYIMIDE SYNTHESIS

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<th>STRUCTURE</th>
<th>NAME</th>
<th>ABBREVIATION</th>
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<td>MONOMETHYLSTER OF 5-NORBORNEOL, 3-BICARBOXYLIC ACID</td>
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<tr>
<td><img src="image" alt="Structure" /></td>
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<td>PPDA</td>
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TABLE II. - MECHANICAL PROPERTIES OF FIBERS STUDIED(a)

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<th>FIBER</th>
<th>LOT</th>
<th>DENSITY LB/IN³</th>
<th>FILAMENTS PER TOW</th>
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<th>TENSILE MODULUS, ksi</th>
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(a) VENDOR DATA
(b) NASA DATA
### Table III. Mechanical Properties of Graphite Fiber P/M-15 Composites

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<tr>
<th>Fiber</th>
<th>Reinforcement</th>
<th>Interlaminar Shear Strength, KSI</th>
<th>Flexural Strength, KSI(a)</th>
<th>Flexural Modulus of Elasticity, KSI(b)</th>
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(a) Normalized to 60% Fiber
(b) Average value obtained from 3 laminates
(c) * * *

### Table IV. Mechanical Properties of Graphite Fiber P/M-II Composites

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<th>Fiber</th>
<th>Reinforcement</th>
<th>Interlaminar Shear Strength, KSI</th>
<th>Flexural Strength, KSI(a)</th>
<th>Flexural Modulus of Elasticity, KSI(b)</th>
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(a) Normalized to 60% Fiber

### Table V. Elemental Analysis of Carbon Fibers

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<th>Fiber</th>
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<th>P (ppm)</th>
<th>S (ppm)</th>
<th>Cl (ppm)</th>
<th>Na (ppm)</th>
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<th>Si (ppm)</th>
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<td>-100</td>
<td>280</td>
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<td>25</td>
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<td>410</td>
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<td>3</td>
<td>&lt;10</td>
</tr>
<tr>
<td>HTS-2 (Lot 121-4)</td>
<td>97.71</td>
<td>300</td>
<td>300</td>
<td>&lt;100</td>
<td>70</td>
<td>210</td>
<td>30</td>
<td>140</td>
<td>35</td>
<td>31</td>
<td>42</td>
<td>&lt;50</td>
</tr>
<tr>
<td>Fortafil J</td>
<td>98.28</td>
<td>100</td>
<td>1100</td>
<td>1030</td>
<td>35</td>
<td>91</td>
<td>20</td>
<td>41</td>
<td>380</td>
<td>170</td>
<td>10</td>
<td>&lt;50</td>
</tr>
<tr>
<td>Thorsen B</td>
<td>99.56</td>
<td>200</td>
<td>200</td>
<td>&lt;100</td>
<td>15</td>
<td>410</td>
<td>14</td>
<td>45</td>
<td>10</td>
<td>61</td>
<td>21</td>
<td>&lt;50</td>
</tr>
</tbody>
</table>
FIGURE 1. -WEIGHT LOSS OF PMR-15/GRAPHITE FIBER COMPOSITES EXPOSED IN AIR AT 600°F

FIGURE 2. -FLEXURAL STRENGTH OF PMR-15/GRAPHITE FIBER COMPOSITES EXPOSED AND TESTED IN AIR AT 600°F

FIGURE 3. -FLEXURAL MODULUS OF PMR-15/GRAPHITE FIBER COMPOSITES EXPOSED AND TESTED IN AIR AT 600°F

FIGURE 4. -INTERLAMINAR SHEAR STRENGTH OF PMR-15/GRAPHITE FIBER COMPOSITES EXPOSED AND TESTED IN AIR AT 600°F
FIGURE 13. - SURFACE OF HTS-2 (LOT 113-4) GRAPHITE FIBER TEM REPLICA

FIGURE 14. - SURFACE OF HTS-2 (LOT121-3) GRAPHITE FIBER TEM REPLICA
FIGURE 15. SURFACE OF THORNEL B GRAPHITE FIBER TEM REPLICA

FIGURE 16. SURFACE OF PORTAFIL 3 GRAPHITE FIBER TEM REPLICA
FIGURE 17. SURFACE OF CELION 6000 GRAPHITE FIBER TEM REPLICA
**Title and Subtitle**
PROPERTIES OF PMR POLYIMIDE COMPOSITES MADE WITH IMPROVED HIGH STRENGTH GRAPHITE FIBERS

**Author(s)**
Raymond D. Vannucci

**Performing Organization Name and Address**
National Aeronautics and Space Administration
Lewis Research Center
Cleveland, Ohio 44135

**Supplementary Notes**

**Abstract**
Recent graphite fiber developments have resulted in high strength, intermediate modulus graphite fibers having improved thermo-oxidative resistance. These improved fibers, obtained from various commercial suppliers, were used to fabricate PMR-15 and PMR-II polyimide composites. Studies were performed to investigate the effects of the improved high strength graphite fibers on composite properties after exposure in air at 600° F. The use of the more oxidatively resistant fibers did not result in improved performance at 600° F. Two of the improved fibers were found to have an adverse effect on the long-term performance of PMR composites. The influence of various factors such as fiber physical properties, surface morphology and chemical composition are also discussed.