Effect of vacuum thermal cyclic exposures on unidirectional carbon fiber/epoxy composites for low earth orbit space applications

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A B S T R A C T
The effects of the low earth orbit environment on three types of unidirectional high-modulus carbon fiber (M40 J, M55 J and M60 J)-reinforced composites were determined in detail. The synergistic environmental factors were the vacuum environment and thermal cycling. Cyclic thermal loading was performed in the temperature range between 120 °C and −175 °C for up to 2000 cycles under the high-vacuum state of 1.3 × 10⁻³ Pa. The material responses were characterized through an assessment of the physical, thermal and mechanical property changes. It follows from the experimental results presented that the synergistic actions of the vacuum and the thermal cycling on the composite property degradation can be attributed to the formation of microvoids and interfacial sliding at the fiber–matrix interface in the early stages of cycling. The implications of these degradation processes based on the dependence of composite properties on vacuum thermal cycling are also discussed.

1. Introduction

Advanced carbon fiber-reinforced composite laminates have been widely used in satellite structures, where the advantages of these materials—their high specific stiffness, near-zero coefficients of thermal expansion (CTE) and dimensional stabilities—make them uniquely suited for applications in a low-specific-weight environment [1–4]. However, since the beginning of composite structure applications, there has been a strong need to quantify the environmental effects on the composite materials based on the coupon-level laminate test data. Recent studies have shown that the environmental conditions that are the most representative of space and that tend to degrade the properties of composite laminates involve vacuum, thermal cycling [2,5–7], atomic oxygen (AO) [2,7–9], ultraviolet (UV) irradiation [2,5,7,8] and micrometeoroid particles [10]. In this respect, there is significant interest in the construction of an experimental database to capture the collective understanding of the degradation mechanisms of composite laminate in in-service environments [2,5–10]. It is necessary to be able to predict the long-term durability of composite laminates with engineering accuracy to use these materials with confidence in critical load-bearing structures.

One of environmental effects of space that is known to induce environmental degradation is thermal cycling because a satellite in low earth orbit (LEO, between 100 and 1500 km above the Earth's surface) passes in and out of the earth's shadow. The exterior surface is exposed to long-term periodic sharp temperature changes as a result [6,7,9]. This type of nonmechanical fatigue is of significant concern for the design of composite laminates because of the strong anisotropy of the thermoelastic characteristics of the long-fiber unidirectional plies. According to the anisotropic behavior of composite laminates, the imposed temperature variations induce thermal stresses at different levels [11,12]. For example, on the microscopic scale, the difference in the thermal dilatation between the fibers and the matrix increases the local stress state in the vicinity of the fiber–matrix interface, whereas, at the mesoscopic ply level, some ply stresses are built up because of the thermal expansion or contraction of the plies restrained by the adjacent plies of different laminate plies. This behavior is a particular concern for quasi-isotropic ([45/0]s/90°), [45/90°/−45°]s) and/or cross-ply asymmetric ([0/90°]) laminates. Considering the importance of the vacuum thermal cycling, several studies [6,7,12–15] have been conducted in this area. Gao et al. [6], Shin [7] and Funk and Sykes [15] in particular have focused on the fundamental understanding of cumulative damage development due to cyclic exposure. However, such research should be expanded. The limited number of thermal cycles (350 cycles or less) used in their studies should be expanded to draw a definite conclusion, and studying the effects of thermal cycling on more representative space-grade materials for LEO application combined in series with mechanical assessments is required to assess the long-term durability issues of these materials.
The goal of this study was to provide fundamental environmental degradation characteristics of unidirectional high-modulus carbon fiber/epoxy laminates that have been widely used in LEO satellite structures. This study significantly extends the scope of previous studies in indentifying degradation trends to investigate the cumulative damage effects of vacuum thermal cycling on the physical and thermal properties and to assess the degradation of the mechanical strength/elastic modulus in space-grade composites. To simulate a more realistic number of thermal loading events, analogous to what a LEO space-based system would experience while in service, vacuum thermal cycling experiments were expanded up to 2000 cycles a LEO space-based system would experience while in service, and to encompass an environmental severity over that expected during service life based on the previous studies [5, 6, 15, 20].

Common to each of the thermal cycling profiles, low temperature ($T_{low} = -175 \pm 5 \, ^\circ C$) and back to 120 \, ^\circ C so that the total duration of each cycle was approximately 43 min. A vacuum pressure of 1.3 \, Pa was employed throughout the thermal cycling test.

The extent of degradation was determined experimentally by exposing the test panels to vacuum thermal cycling. The experiments were repeated for 500 cycles (358 h), 1000 cycles (716 h), 1500 cycles (1074 h) and 2000 cycles (1432 h). Following the thermal cycling exposure, all partly- or fully-aged test panels were cut into the desired dimensions using a water-cooled diamond saw and subsequently dried in a vacuum oven at 60 \, ^\circ C for 24 h to remove moisture after the cutting operation. Series of physical and mechanical tests were then performed in the standard laboratory atmosphere of 23 \pm 2 \, ^\circ C and 50 \pm 5\% relative humidity on the baseline (unaged) and environmentally conditioned test samples.

2. Experimental procedures

2.1. Materials and specimen preparation

Three polyacrylonitrile (PAN)-based unidirectional Torayca® carbon fibers (Toray Industries, Inc., Japan), denoted as M40 J, M55 J and M60 J, were purchased in the form of prepreg tape and cured in an autoclave facility. These materials are attractive for space applications because of their low density, high modulus and dimensional stability. Three types of carbon fibers are high-modulus fibers, and have elastic moduli of 377 GPa, 540 GPa and 588 GPa for M40 J, M55 J and M60 J unidirectional fibers, respectively [16]. The corresponding linear longitudinal CTE values of carbon fibers are $-0.83 \times 10^{-6} \, ^\circ C^{-1}$, $-1.1 \times 10^{-6} \, ^\circ C^{-1}$ and $-1.1 \times 10^{-6} \, ^\circ C^{-1}$, respectively [16]. The fundamental characteristics of the three epoxy prepreg types used in this study are listed in Table 1.

The laminated composite panels were fabricated by stacking multiple layers of unidirectional prepregs. All panels for fabricating test specimens used in this study were unidirectional laminates with the lay-up sequences of [0\n]. The number of plies (n) to meet the required panel thicknesses depended on the test standards adopted for each test. Each panel manufactured for use as test samples should have a traceable reference edge [17]. All prepreg layers had a 0° orientation at nominal thicknesses of 1.0 mm, 1.5 mm and 3.0 mm. During a conventional lay-up process, the laminate stack was debulked at regular intervals (every 3 or 4 plies) to ensure that the prepreg conformed exactly to the curing tool and to remove most of the air bubbles from between the individual plies. After a conventional lay-up process, each laid-up stack was vacuum bagged and placed in an 2.4 m (diameter) \times 4.6 m (length) autoclave (Thermal Equipment Corporation, USA) for the curing process. Cure temperatures and cycle times were 176.7 \, ^\circ C for 180 min (M40 J/VISCOTEX6376), 121.2 \, ^\circ C for 100 min (M55 J/#2580-14) and 176.7 \, ^\circ C for 120 min (M60 J/#3800), respectively. A consolidation pressure was employed throughout the cure cycles with a full vacuum of 1 bar (107.9 kPa).

2.2. Environmental conditioning

The thermal cycling tests were conducted by using a thermal vacuum chamber (Hanchang Eng, South Korea), as shown in Fig. 1A. The experiment was performed in an environmental chamber with a proportional integral derivative (PID) programmable temperature controller. Test temperature range should be as large as practicable to meet environmental stress screening (ESS) purposes based on the guideline in MIL-STD-810F [18] and MIL-STD-1540C [19]. It is generally required to reveal potential flaws in material exposed to more extreme temperature change condition. In this study, extreme temperature conditions (120 \, ^\circ C to $-175 \, ^\circ C$) encompass an environmental severity over that expected during service life based on the previous studies [5, 6, 15, 20].

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Common to each of the thermal cycling profiles, low temperature ($T_{low} = -175 \pm 5 \, ^\circ C$) and back to 120 \, ^\circ C so that the total duration of each cycle was approximately 43 min. A vacuum pressure of 1.3 \, Pa was employed throughout the thermal cycling test.

2.3. Property evaluations

For qualitative evidence of the morphological changes in composites, planar and cross-sectional views of the specimens were obtained by using a S-2400 scanning electron microscope (SEM) instrument (Hitachi High-Technologies Co., Japan), as shown in Fig. 2. The laminates’ densities (g/cm³) were monitored before and after thermal cycling exposures according to the ASTM D 792 [21]. The volume fractions of fiber ($V_f$) and void ($V_v$) in each composite were measured by weighing a 25 cm² square of laminate sample and by dissolving the epoxy matrix in 70 wt.% nitric acid, based on the ASTM standards of D 3171 [22] and D 2734 [23], respectively. The glass transition temperature denoted as $T_g$ was measured by differential scanning calorimetry (DSC) using a DSC 50 instrument (Shimadzu Co., Japan) based on the ASTM E 1269 standard [24]. The $T_g$ is an important material property, defined as the specific temperature at which a material’s properties are drastically changed. For DSC experiments, the glass transition is defined as a change in the heat capacity as epoxy matrix is transformed from a glassy state to a rubbery state. Finally, a thermogravimetric analysis (TGA) experiment using a TGA 50 instrument (Shimadzu Co., Japan) was performed to compare the changes in the thermal stability of the epoxy matrix. The decomposition temperature ($T_d$) from the TGA experiments is defined by the

<table>
<thead>
<tr>
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</thead>
<tbody>
<tr>
<td>Manufacturer</td>
<td>Hexcel Composites, USA</td>
<td>Toray, Japan</td>
<td>Toray, Japan</td>
</tr>
<tr>
<td>Carbon fiber</td>
<td>Unidirectional M40JB-6 K</td>
<td>Unidirectional M55 J</td>
<td>Unidirectional M60 J</td>
</tr>
<tr>
<td>Epoxy matrix</td>
<td>6376 modified epoxy</td>
<td>#2580-14 modified epoxy</td>
<td>#3800 epoxy</td>
</tr>
<tr>
<td>Cure temperature</td>
<td>127 °C</td>
<td>127 °C</td>
<td>170 °C</td>
</tr>
<tr>
<td>Resin content</td>
<td>34 ± 2.5 wt.%</td>
<td>35 ± 3.0 wt.%</td>
<td>53 ± 5.0 wt.%</td>
</tr>
<tr>
<td>Volatile content</td>
<td>≤2%</td>
<td>0.34%</td>
<td>≤2%</td>
</tr>
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</table>

Table 1: Characteristics of three types of unidirectional carbon fiber/epoxy prepreg for space application: data is supplied by the prepreg manufacturers.
The changes in the mechanical properties of composite laminates exposed to vacuum thermal cycling environments were measured according to the appropriate ASTM standards: (1) interlaminar shear strength (ILSS, ASTM D 2344 [25]), (2) flexure strength/modulus (ASTM D 790 procedure A and B [26]), (3) longitudinal tensile strength/modulus (ASTM D 3039 [27]) and (4) longitudinal compressive strength/modulus (ASTM D 3410 [28]). All mechanical tests were conducted with a servo-hydraulic 100 kN MTS 810 testing machine (MTS Systems Co., USA) at a constant deformation rate up to ultimate failure. The sample dimensions and their referenced standards are shown in Fig. 3. A minimum eighteen samples was prepared at each environmental condition (i.e., thermal cycles) to facilitate statistically valid samplings. For each series of tests, the standard deviation $V(x)$, coefficient of variation $CV$ and $B$-value $B$ can be defined as follows [29]:

$$V(x) = \sqrt{\frac{\sum_{i=1}^{n} x_i^2 - nE(x)^2}{(n-1)}}$$

(1)

$$CV = 100 \times \frac{V(x)}{E(x)}$$

(2)

$$B = E(x) - kV(x)$$

(3)
where $x_i$ is the measured property, $n$ is the number of samples, $E(x)$ is the sample mean and $k$ is the $B$-value tolerance factor ($k = 1.97$) for the normal distribution that is based upon the number of samples. The fractographic images of the samples subjected to the mechanical tests were obtained through microscopic observations.

### 3. Results and discussion

#### 3.1. Baseline mechanical properties of carbon fiber/epoxy laminates

The mechanical properties of baseline composite laminates are summarized in Table 2, where average, CV and $B$-value are listed. In particular, the statistically based material properties with uncertainties were characterized as a $B$-value. A $B$-value is one in which 90% of the material property distribution is above the basis value with a 95% percent level of confidence based on the guideline in MIL-HDBK-17-1F [29].

#### 3.2. Changes in physical and thermal properties

The results for the physical and thermal properties for each material at each exposure condition are given in Table 3, where the most significant laminate properties, such as density, void content, $T_g$ and $T_d$, are listed. All physical and thermal properties were averaged from six replicates.

The exposure to cyclic vacuum thermal conditions apparently yields a decrease in the laminate densities, and the corresponding decrease in the rates after being submitted to 2000 cycles were 1.30% (M40 J), 1.22% (M55 J) and 2.55% (M60 J). One possible explanation for this reduction in laminate density is the coupling between the epoxy matrix-loss/shrinkage and formation of internal microvoids. Consequently, the results were verified by inspecting the electron micrographs of two perpendicular sides of the composite laminate, as shown in Fig. 4. These micrographs show typical internal damages on the free edge and on the as-molded surface: (1) matrix-crack formation at the as-molded surface (Fig. 4A); (2) plastic deformation in the epoxy matrix owing to ma-

### Table 2

<table>
<thead>
<tr>
<th>Test item (Test specification)</th>
<th>Material (Test specification)</th>
<th>Strength (MPa)</th>
<th>Modulus (GPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>M40 J [0/90]</td>
<td>80.9 ± 3.1</td>
<td>1368.6 ± 37.5</td>
</tr>
<tr>
<td></td>
<td>M55 J [0/90]</td>
<td>63.4 ± 1.2</td>
<td>1217.1 ± 27.9</td>
</tr>
<tr>
<td></td>
<td>M60 J [0/90]</td>
<td>50.8 ± 1.6</td>
<td>774.1 ± 9.4</td>
</tr>
<tr>
<td></td>
<td></td>
<td>E(x) ± V(x) (MPa)</td>
<td>CV (%)</td>
</tr>
<tr>
<td>ILSS (ASTM D 2344 [24])</td>
<td></td>
<td>E(x) ± V(x) (MPa)</td>
<td>CV (%)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>E(x) ± V(x) (MPa)</td>
<td>CV (%)</td>
</tr>
<tr>
<td>Flexural strength/modulus (ASTM D 790 [25])</td>
<td>M40 J [0/90]</td>
<td>1368.6 ± 37.5</td>
<td>2.7</td>
</tr>
<tr>
<td></td>
<td>M55 J [0/90]</td>
<td>1217.1 ± 27.9</td>
<td>2.3</td>
</tr>
<tr>
<td></td>
<td>M60 J [0/90]</td>
<td>774.1 ± 9.4</td>
<td>1.2</td>
</tr>
<tr>
<td>Longitudinal tensile strength/modulus (ASTM D 3039 [26])</td>
<td>M40 J [0/90]</td>
<td>1984.1 ± 67.3</td>
<td>3.4</td>
</tr>
<tr>
<td></td>
<td>M55 J [0/90]</td>
<td>1725.8 ± 94.0</td>
<td>5.4</td>
</tr>
<tr>
<td></td>
<td>M60 J [0/90]</td>
<td>759.1 ± 56.9</td>
<td>7.5</td>
</tr>
<tr>
<td>Longitudinal compressive strength/modulus (ASTM D 3410 [27])</td>
<td>M40 J [0/90]</td>
<td>1017.7 ± 8.9</td>
<td>0.9</td>
</tr>
<tr>
<td></td>
<td>M55 J [0/90]</td>
<td>694.6 ± 15.4</td>
<td>2.2</td>
</tr>
<tr>
<td></td>
<td>M60 J [0/90]</td>
<td>617.1 ± 15.0</td>
<td>2.4</td>
</tr>
</tbody>
</table>

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A M40 J, M55 J and M60 J denote M40 J/Vicotex6376, M55 J/#2580-14 and M60 J/#3800 unidirectional prepreg tapes, respectively.

B cured 1-ply thickness (60 vol.% fiber content); 0.100 mm for M40 J, 0.088 mm for M55 J and 0.038 mm for M60, respectively.
trix shrinkage (Fig. 4B); (3) microvoid formation; and (4) fiber–matrix interfacial debonding and sliding (Fig. 4C). All of these types of internal damages are known to result in property deterioration of composite laminates. The matrix shrinkage is particularly visualized as a dark and deep area, imprinted on the polished free-edge section, as shown in Fig. 4B. Typical microvoids formed at the fiber–matrix interface in the matrix-rich area are shown in Fig. 4C. It would appear that the constraint imposed on the expansion of the matrix is relieved by the interfacial debonding, followed by microvoid formation at the interface.

For quantitative comparison of the microvoids, the normalized volume fractions of microvoids versus the thermal cycles for each composite laminate are plotted in Fig. 5. In general, the microvoid expansion exponentially increased according to the number of cycles. As-fabricated M40 J and M55 J laminates achieve aerospace-grade void contents of less than 2.0 vol.%. The change in the microvoid volume fraction \( V_v \) of the M40 J laminate exposed to cyclic vacuum thermal conditions varies from 1.5 vol.% (baseline) to 2.3 vol.% (2000 cycles), as listed in Table 3. In contrast, the high level of microvoids present in the baseline M60 J laminate is probably due to fabrication and/or machining processes. Its rate of microvoid content formation was also rapid. It is well known that the microvoids act as microcrack initiation sites during thermal cycling, as exemplified by several studies [12,30,31].

It is well known that the vacuum thermal cycling also induce severe degradation in the epoxy matrix. This degradation is supposed to involve an irreversible modification of the molecular structures through the induction of chain scissioning, and it causes a change in the bulk material’s glass transition temperature as well. Such a degradation mechanism could be further supported by the permanent loss of thermal stability after aging. Both the \( T_g \) and the \( T_d \) of M40 J and M60 J laminates after 2000 cycles exposure experienced little or no change compared with those of the baseline material, as shown in Table 3: no signs of matrix degradation were observed. Despite the minimal differences found in the \( T_g \) of M55 J laminate, the adverse effects of the environments on the epoxy matrix’s cross-links were noticeable. Fig. 6A compares DSC thermograms of baseline and aged materials. Each of the diagrams shown in the figure represents the result from one particular sample: however, at least six replicates (using samples from different panels) were performed for each condition, and the results were quite repeatable. The results show that the reduction of the
was greater for cycled M55 J laminates, and the decrease in the rate was above 10% after being submitted to 2000 cycles. The TGA data supported this claim, as the \( T_d \) was found to decrease significantly (see Fig. 6B).

3.3. Changes in mechanical properties

The ultimate strength and elastic modulus of carbon fiber/epoxy laminates at standard laboratory atmosphere are compared in Fig. 7. The data represent the statistical mean values for at least eighteen samples, with the standard deviation shown. As a general rule, the strength and the modulus of the composite laminates were gradually reduced according to the number of cycles. The explanation for each property change will be discussed separately. The following sections compile the results for the different materials and contain comparisons of the composite laminates' properties.

3.3.1. Interlaminar shear strength and flexure strength/modulus

Both the ILSS and the flexure properties have been primarily determined for quality control and material selection, where comparative rather than absolute values are required. The ILSS values for samples exposed to cyclic vacuum thermal conditions were notably lower than those obtained from the baseline, and the corresponding decrease in the rates after 2000 cycles were 14.7% for M40 J and 12.2% for M60 J. In contrast, the change in the M55 J laminate was moderate at 5.8%. The most common failure mode observed was interlaminar yielding failure, although several samples corresponded closely to the local damage mode such as transverse ply cracking (i.e., flexure compression and flexure tension) [25].

For the flexure properties, severe degradation of the flexure strength was also observed because of the aging effect. Note that the flexure strength is the value of the stress at failure on the sample surface, and its failure mode is generally accompanied by the breaking of fibers rather than interlaminar shear [32]. Thus, it is less sensitive to the environment than the ILSS. In the case of M55 J laminates, it was observed that little additional strengthening was achieved in the early stage of cycling: the statistical strength ranges overlap. The studies by Gao et al. [6] and Papanicolau et al. [33] reported that further cross-linking of the epoxy matrix, which was induced by additional chemical reactions between unreacted groups, increased the flexure strength in the early stage of cycling. However, a direct comparison between the flexure and the DSC results may not be adequate because the \( T_g \) of the M55 J laminate was found to decrease significantly. Comparing different composite laminates revealed that the decrease in the flexure strength at 2000 cycles was 10.1% (M40 J), 13.3% (M55 J) and 0.8% (M60 J). It is probably due to the failure mode of flexure samples that most failures of M40 J and M55 J laminates presented interlaminar shear failure or ply-level buckling which was preceded by delamination of the outer ply. On the other hands, M60 J laminates exhibited that the specimen failure occurs on either one of its outer surface (i.e., fiber micro-buckling). The reductions in the flexure modulus occurred in the following order after 2000 aging cycles: 22.1% (M40 J) > 13.8% (M60 J) > 0.4% (M55 J).

3.3.2. Longitudinal tensile strength/modulus

The tensile properties of composite laminates are generally thought to be insensitive to the environment because of the carbon fibers’ inertness [34]. Adverse environments, however, may affect the epoxy matrix itself and the fiber/matrix interface, which directly contribute to the tensile strength and fracture toughness. In this study, the longitudinal tensile strength exhibited an intermediate decrease ranging from 6.7% to 11.1%, whereas the decrease of the tensile modulus occurred slowly because the normalized decrease fell between 1.8% and 6.9%. By comparing different composite laminates, the largest decrease in the strength and in the modulus were obtained for M60 J laminates. This is probably due to the high tensile modulus (588 GPa) of the M60 J carbon fiber itself, which is followed by an increase in the crystallinity and the orientation of the crystallographic basal planes parallel to the fiber axis, as illustrated in Fig. 8A [30,35]. A roughly linear relationship between the fiber modulus and the CTE exists, and as the longitudinal modulus of the carbon fibers increases, the longitudinal CTE decreases [36]. The incorporation of the highest modulus M60 J fiber with the lowest CTE (most negative) could

![Fig. 5. Normalized microvoid volume of unidirectional carbon fiber/epoxy laminates as a function of vacuum thermal cycling. Each value corresponds to the average of three samples: baseline \( V_f \) (88.38 ± 0.72 vol.%) and \( V_v \) (1.49 ± 0.25 vol.%) for M40 J; 53.02 ± 0.75 vol.% (\( V_f \)) and 2.02 ± 0.40 vol.% (\( V_v \)) for M55 J; and 38.19 ± 1.69 vol.% (\( V_f \)) and 3.71 ± 0.96 vol.% (\( V_v \)) for M60 J.]
result in more extreme behavior under transient thermal conditions because of the increased mismatch in the CTE between the composite constituents.

In addition, it is well reported that fiber–matrix interfacial adhesion controls the stress transfer between the fibers and the matrix, the stress relaxation and mechanisms of damage accumulation and propagation [31]. A schematic representation of the fiber–matrix interface is given in Fig. 8B. The fracture behavior of the fiber–matrix interfacial bond significantly influences the fracture strength and the toughness of composite laminates [37]. Typical tensile stress–strain diagrams of the M55 J laminates are plotted in Fig. 9A. It is clear in Fig. 9B that the toughness of the M55 J laminate decreased after being submitted to 2000 cycles: the ultimate strain was reduced by 44.5%. This observation can be explained by the presence of disjoining forces of molecular interactions between polymer molecules and adjacent phases [38,39]. The overall strength and modulus of such composite laminates were therefore dependent on the mechanical robustness in the interfacial zone because this occupies a rather large volume fraction within the composite microstructure.

3.3.3. Longitudinal compressive strength/modulus
In the case of changes in the longitudinal compressive strength, a decreasing trend similar to that of the longitudinal tensile strength could be observed for all of the laminates studied. The reason for the decrease in the compressive strength was attributed to the fragile fiber–matrix interface and the composite laminates had therefore become incapable of supporting the external compressive loading. In addition, the differences in fiber contents and internal irregularities (i.e., voids) played a role in the compressive behaviors of the laminates studied. The nominal Vf in the baseline was 68.6 vol.%, 53.0 vol.% and 38.2 vol.% for M40 J, M55 J and M60 J laminates, respectively. Sectioning of the as-fabricated laminates revealed that differences in the fiber content and their distribution were observed, as shown in Fig. 10. In particular, the uneven fiber distribution pattern led to the concentration of stress in the M60 J laminate subjected to loading, which degraded the longitudinal compressive strength (Fig. 10C).

3.3.4. Probability density function (PDF) analysis
At this point, it is also meaningful to compare the statistical properties of derived laminate properties with experimentally

Fig. 6. Typical DSC and TGA diagrams of the M55 J carbon fiber/epoxy laminate as a function of vacuum thermal cycling: (A) DSC thermograms; and (B) weight loss as a function of the TGA temperature, by showing (a) initial weight loss, (b) resin decomposition, (c) char decomposition and (d) fiber decomposition.
based values to account for uncertainties in engineering design. Any aerospace structures cannot be guaranteed to be absolutely safe because of the uncertainties in the in situ laminate properties. A probability analysis is thereafter required to show that the properties fall within a specified acceptable range for each specific application [40,41]. In this study, these differences were quantified both in terms of the mean values and the associated dispersion parameters. Examples of histograms for the four derived strength properties (ILSS, flexure, tensile and compressive) are shown in Fig. 11 in terms of probability densities with the distribution model superimposed. According to the guidelines of MIL-HDBK-17-1F [29], the normal, Weibull and lognormal laws can be defined by

\[
f(x; \mu, \sigma)_{\text{normal}} = \frac{1}{\sigma \sqrt{2\pi}} \exp \left( -\frac{(x - \mu)^2}{2\sigma^2} \right) \quad (\text{Normal distribution})
\]

\[
f(x; \mu, \sigma)_{\text{lognormal}} = \frac{1}{\sigma x \sqrt{2\pi}} \exp \left( -\frac{(\ln x - \mu)^2}{2\sigma^2} \right) \quad (\text{Lognormal distribution})
\]

\[
f(x; \beta, \theta)_{\text{Weibull}} = \frac{\beta}{\theta} \left( \frac{x}{\theta} \right)^{\beta-1} \exp \left( -\left( \frac{x}{\theta} \right)^\beta \right) \quad (\text{Weibull distribution})
\]
where \( \mu \) is the location parameter (i.e., mean value) and \( \sigma \) is the shape parameter (i.e., standard deviation). In the Weibull law, \( \theta \) and \( \beta \) are the scale and shape factors, respectively. The distribution parameters, along with the goodness-of-fit statistics according to the guidelines of MIL-HDBK-17-1F [29], are given in Table 4. Observed significance level (OSL) hypothesis tests were also performed. If the OSL for the Weibull distribution was greater than 0.05, this indicated an adequate fit of the data to the Weibull distribution. Based on these results and following the guidelines of MIL-HDBK-17-1F [29], it was appropriate to model the baseline properties with a Weibull law. In contrast, it is recommended to use a normal or lognormal law after the material has been subjected to cyclic vacuum thermal conditions. As shown in the figures, it is important to acknowledge this variability when designing with composite structures and to incorporate it into the design values of laminate properties [41]. The composite laminates exposed to cyclic vacuum thermal conditions have more intrinsic variability than the baseline because they exhibit wider discrepancies in each property. This behavior can be explained by instantaneous surface contact damage and subsequent diffusion through the internal laminate plies.

3.4. Fractography observation by microscopy

As mentioned above, thermal stresses are largely a consequence of very different CTEs between the fiber and the matrix in a unidirectional laminate. Therefore, the fiber–matrix interface plays a critical role in controlling the overall properties of the composite laminate, such as the off-axis strength, the fracture toughness and the environmental stability [37]. The representative fracture morphologies after being submitted to ILSS, flexure and longitudinal tests are shown in Fig. 12. Fractographic modification as a result of the cyclic vacuum thermal conditions was observed with fiber pull-outs and interfacial separations at the fiber–matrix interface. Therefore, it can be concluded that the predominant cause of thermal cycling damage is interfacial debonding and frictional sliding along the interface. The weak interfacial bond and the presence of some microvoids, and perhaps microcracking, should naturally facilitate the onset of frictional sliding during thermal cycling.

4. Conclusions

The ultimate goal of this study was to provide long-term durability data on three types of unidirectional high-modulus carbon fiber/epoxy laminates under vacuum thermal cycling for LEO satellite structure applications. From the study of the degradation mechanisms of the representative carbon epoxy composite laminates for space applications, the following conclusions can be drawn:

- The composite laminates examined in this study exhibited a gradual accumulation of damage as a result of the prolonged action of vacuum and thermal loading. Most of the property...
degradation occurred early on in the cycling process (up to 500 cycles) and then little additional degradation was observed. The intermediate changes (less than 10%) in the fiber-dominant properties, such as longitudinal tensile
strengths, could be observed at 2000 cycles. In contrast, the fiber–matrix interface-dominated properties, such as the ILSS and the compressive strength, were notably reduced up to a certain limit (up to 15%).

Although it is difficult to reach a definite conclusion because of the considerable scatter inherent in the data, the two material systems (M40 J/VISCOTEX6376 and M60 J/#3800) cured at 177 °C were more environmentally stable than the one cured at 121 °C (M55 J/#2580-14). The differences were most noticeable with the degradation of the thermal properties (T_g and T_d).

In addition, the microvoid formation and the subsequent reduction in the laminate density seem consistent with the decrease in the residual strength.

The vacuum thermal cycling resulted in the mechanical property degradation of the fiber–matrix interface, facilitated by the weak fiber–matrix bond. After the interfacial bond weakened, the thermal stress was relieved through interfacial sliding. Fractographic micrographs revealed that the loss of interfacial integrity between the carbon fibers and the epoxy matrix was the dominant mechanism in the permanent loss of the composite laminates’ properties: the loss of interfacial integrity induced the low-strength interfacial adhesion failure mode. Therefore, the formation of a more tenacious interfacial bond between the fiber and the matrix can improve the mechanical response of unidirectional carbon fiber/epoxy laminates exposed to cyclic vacuum thermal conditions.

**Table 4**

Distribution parameters of M40 J laminate according to vacuum thermal cycling: normal, lognormal and Weibull law.

<table>
<thead>
<tr>
<th>Materials</th>
<th>Properties</th>
<th>Normal law</th>
<th>Lognormal law</th>
<th>Weibull law</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>OSL, µ, σ</td>
<td>B (MPa)</td>
<td>OSL, µ_0, σ_0</td>
<td>B (MPa)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>ILSS (MPa)</td>
<td>Baseline</td>
<td>3.3E-01</td>
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<td>1000 cycles</td>
<td>1.3E-01</td>
<td>75.7, 4.86</td>
<td>66</td>
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<tr>
<td></td>
<td>2000 cycles</td>
<td>3.2E-04</td>
<td>69.1, 5.25</td>
<td>59</td>
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<tr>
<td>Flexure strength (MPa)</td>
<td>Baseline</td>
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<td>1369, 37.5</td>
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<td></td>
<td>1000 cycles</td>
<td>3.5E-02</td>
<td>1232, 14.6</td>
<td>1203</td>
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<tr>
<td></td>
<td>2000 cycles</td>
<td>5.7E-02</td>
<td>1231, 54.3</td>
<td>1124</td>
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<td>Longitudinal tensile strength (MPa)</td>
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<td>1984, 67.3</td>
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<tr>
<td></td>
<td>1000 cycles</td>
<td>1.9E-01</td>
<td>1905, 104</td>
<td>1699</td>
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<tr>
<td></td>
<td>2000 cycles</td>
<td>4.8E-02</td>
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<td>1751</td>
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<tr>
<td>Longitudinal compressive strength (MPa)</td>
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<td>1000</td>
</tr>
<tr>
<td></td>
<td>1000 cycles</td>
<td>3.4E-03</td>
<td>953, 29.9</td>
<td>894</td>
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<tr>
<td></td>
<td>2000 cycles</td>
<td>3.7E-03</td>
<td>897, 28.0</td>
<td>842</td>
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</tbody>
</table>

**Fig. 11.** Probability densities of the derived properties of the M40 J laminate according to vacuum thermal cycling: solid lines indicate the normal law; and dash lines indicate the Weibull law.
Fig. 12. Typical fracture morphologies after being submitted to ILSS, flexure and longitudinal tensile tests: the electron micrographs were obtained under 1000× magnification.

Remarks

Mention of any commercial or trade name in this article does not imply any endorsement of the products by the authors. The trade names have been used purely for identification purposes.

References


