Nondestructive Evaluation of the Effects of Thermal-Mechanical Aging on IM7/IK3B Stiffness

MICHAEL D. SEALE AND ERIC I. MADARAS
NASA Langley Research Center
Mail Stop 231
Hampton, VA 23681

ABSTRACT: The introduction of new, advanced composite materials into aviation systems requires a thorough understanding of the long-term effects of combined thermal and mechanical loading on these types of materials. Ultrasonic Lamb waves offer a promising method of evaluating stiffness in composite materials. To help assess the effect of aging, a suitably designed Lamb wave measurement system is being used to obtain bending and out-of-plane stiffness coefficients of composite laminates undergoing thermal-mechanical loading. In this study, a series of 16-ply graphite-fiber-reinforced amorphous thermoplastic polyimide laminates, IM7/K3B, were aged in load frames equipped with special environmental chambers. The samples were subjected to both high and low strain profiles. The bending and out-of-plane shear stiffnesses as well as time-of-flight measurements for samples that have undergone up to 4,993 fatigue cycles of combined thermal and mechanical aging are reported. The Lamb wave generated elastic stiffness results show decreases of up to 20% with extended aging.

KEY WORDS: Lamb waves, thermal-mechanical aging, composites, nondestructive evaluation, ultrasonics.

INTRODUCTION

ADVANCED AEROSPACE COMPOSITE materials are required to respond well to combined thermal and mechanical loading. The materials used in the design of these structures need to perform well under these strenuous conditions for thousands of hours. Therefore, it is of interest to investigate the feasibility of nondestructively monitoring thermal-mechanical aging in composites. Among the various nondestructive techniques available, guided acoustic plate waves (Lamb waves) offer a convenient method of evaluating stiffness changes in polymer-matrix composite
materials. Studies have been conducted which show a reduction in Lamb wave velocity due to a loss of stiffness caused by matrix cracking [1-4]. Karim et al. [5] and Mal et al. [6] have used inversion techniques to determine the material parameters of composites from experimental Lamb wave data. Lamb wave techniques have also been used to study delaminations [7-10], porosity [10,11], and fiber misalignment [11].

Under general thermal-mechanical loading, both fatigue damage and thermal degradation of composites may occur. Street et al. [12] have used measurements of interlaminar fracture toughness, interlaminar shear strength, and hardness to investigate the response of AS4/3501-6 composites to elevated temperatures for exposure times up to 30 minutes. Deterioration of the matrix was found to be the primary degradation mechanism. Herakovich and Hyer [13] thermally cycled graphite/epoxy composites and measured the crack density as a function of cycles. Transverse matrix cracking was found to be the main cause of degradation. Stansfield and Pritchard [14] have subjected XAS/Fibredux 914 composites to rapid changes in temperature. These fluctuations were found to lead to both moisture absorption and thickness changes. Henaff-Gardin et al. [15] subjected T300/914 composites to cyclic thermal loading and used X-ray radiography to observe cracking. The crack density was found to increase with the number of thermal cycles. Tao et al. [16] used interlaminar shear strength and fracture toughness to monitor the effects of thermal as well as thermal-mechanical aging in IM7/K3B laminates. They found that the effects of aging were similar for both measurement methods.

Although these studies have examined thermal degradation [12-16] and thermal-mechanical aging [16] in polymer-matrix composites, few studies have been conducted which monitor either of these damage mechanisms using ultrasonic nondestructive evaluation techniques. However, ultrasonic Lamb waves have been shown by Seale et al. [1] to be an effective method for characterizing fatigue damage in AS4/3501-6 composites as well as thermal damage in AS4/977-3 composites. This study will explore the use of Lamb waves to monitor the effects of combined thermal-mechanical aging in thermoplastic composite specimens. The Lamb wave scanning system was used to measure time-of-flight and stiffness values on thermal-mechanically aged composites. The resulting stiffness reduction for samples which have been aged up to 4,993 fatigue cycles is discussed.

MATERIAL AND TEST PARAMETERS

The composite material studied was a graphite-fiber-reinforced amorphous thermoplastic polyimide, IM7/K3B. The 122.0 cm by 30.5 cm samples were manufactured with 16 plies and had a stacking sequence of [45/0/-45/90]_2S. The samples had a nominal thickness of 0.223 cm. The samples were subjected to thermal-mechanical aging in either 98-kN or 222-kN capacity load frames equipped with environmental chambers which have a usable temperature range of -54°C to +344°C. The grips for the load frames were outside of the chambers, and the chamber dimensions were 40 cm wide by 67 cm tall by 40 cm deep. Thus, only the middle 67-cm section along the length of the samples was subjected to thermal extremes. The upper and lower portions of the samples remained outside the chambers.

The thermal-mechanical fatigue profile used for this study is shown in Figure 1. In the figure, the units for strain and temperature have been omitted due to the fact that different ranges were used for the various samples. The load was applied in the 0° direction (along the length of the specimens), and both high and low strain profiles were used. The strain levels for the low-strain profiles ranged from 0 to 2,000 microstrain with a strain at or above 1,040 microstrain for
180 minutes. The strain levels for the high-strain profiles ranged from 0 to 3,000 microstrain with a strain at or above 1,560 microstrain for 180 minutes. The temperature extreme for all of the samples was chosen to be -18°C to +177°C with a sustained temperature of +177°C for 180 minutes.

**THEORY**

For a composite lamina with the $x$-axis defined as in the fiber direction, the $y$-axis transverse to the fibers, and the $z$-axis out of the plane of the plate, the stress-strain relationship for a lamina assumed to be under plane stress is given by

$$
\begin{bmatrix}
\sigma_1 \\
\sigma_2 \\
\tau_4 \\
\tau_5 \\
\tau_6
\end{bmatrix} =
\begin{bmatrix}
Q_{11} & Q_{12} & 0 & 0 & 0 \\
Q_{12} & Q_{22} & 0 & 0 & 0 \\
0 & 0 & Q_{44} & 0 & 0 \\
0 & 0 & 0 & Q_{55} & 0 \\
0 & 0 & 0 & 0 & Q_{66}
\end{bmatrix}
\begin{bmatrix}
\epsilon_1 \\
\epsilon_2 \\
\gamma_4 \\
\gamma_5 \\
\gamma_6
\end{bmatrix}
$$

where the subscripts 1,2,4,5, and 6 represent the $xx$, $yy$, $yz$, $xz$, and $xy$ components, respectively. The normal and shear stresses are given by $\sigma$ and $\tau$, respectively, and $\epsilon$ and $\gamma$ represent the normal and shear strains, respectively. The $Q_{ij}$ for $i,j = 1, 2, 6$ and for $i,j = 4, 5$ are the plane-stress stiffness components and shear stiffness components, respectively. Relationships between the $Q_{ij}$ and

![Figure 1. Typical thermal-mechanical fatigue profile.](image-url)
the elastic stiffness constants, $c_{ij}$, can be found in Jang [17], and equations relating the engineering constants $(E_1, E_2, G_{12}, v_{12},$ and $v_{21})$ to the $Q_{ij}$ can be found in Daniel and Ishai [18]. The bending stiffnesses, $D_{11}$ and $D_{22}$, and the out-of-plane stiffnesses, $A_{44}$ and $A_{55}$, are obtained by integrating the $Q_{ij}$ through the thickness of the plate. These stiffness values are defined as

$$D_{ii} = \int_{-h/2}^{h/2} (Q'_{ii}) k_j z^2 dz \quad i = 1, 2$$

(2)

and

$$A_{jj} = k_j^2 \int_{-h/2}^{h/2} (Q'_{jj}) k_j dz \quad j = 4, 5$$

(3)

where the subscript $k$ represents each layer in the laminate, $k_j$ is a shear correction factor, and $h$ is the total thickness of the plate [19]. The $Q'_{ij}$ are the transformed stiffness coefficients rotated according to the orientation of each ply with respect to the wave propagation direction. For a further description of plate theory and how the laminate stiffnesses relate to the Lamb wave velocity, the reader is referred to Tang et al. [19].

**LAMB WAVES**

Lamb waves arise from a coupling between the shear-vertical (SV) and compressional (P) waves reflected at the stress-free boundaries at the top and bottom of a thin plate. There are two classes of Lamb mode solutions, symmetric and antisymmetric, which are defined in terms of the displacement of the plate with respect to the mid-plane. As the frequency of the wave increases, higher order symmetric and antisymmetric modes begin to propagate. For reference, the Lamb wave dispersion curve for the thermoplastic samples used in this study is shown in Figure 2. At low frequencies (below 500 kHz), only the lowest order symmetric mode, $S_0$, and lowest order antisymmetric mode, $A_0$, propagate. In this region, the $S_0$ or extensional plate mode is almost nondispersive, and the $A_0$ or flexural plate mode is highly dispersive. The velocity of each Lamb mode is directly related to the properties of the material. Therefore, an effective tool exists to calculate the stiffness of a composite by measuring the velocity of these waves. This study will investigate solutions for the flexural mode.

For the propagation in the $0^\circ$ direction, the effects of altering $A_{55}$ and $D_{11}$ on the flexural dispersion curve are shown in Figure 3. The full stiffness values used in the calculation are given by Equations (2) and (3) and are assumed to be representative of an unaged sample. As can be seen in the figure, decreasing $D_{11}$ by 25% does not significantly change the curve. However, a larger shift in the curve is seen when $A_{55}$ is reduced by 25%. Also, the behavior of the dispersion curve is dominated by $D_{11}$ at low frequencies and by $A_{55}$ at higher frequencies. Figure 4 shows a plot of the percent reduction in velocity as a function of frequency for 25% reductions in each stiffness value. The figure clearly shows that changing $D_{11}$ has a greater effect on the velocity at lower frequencies, and changes in $A_{55}$ alter the dispersion curve at higher frequencies. The dispersion curve for propagation in the $90^\circ$ direction is the same as shown in Figure 3 except the constants controlling the behavior are $D_{22}$ and $A_{44}$. 
Figure 2. Lamb wave dispersion curve for thermoplastic composite samples.

Figure 3. Flexural dispersion curves using full (unaged) values of $D_{11}$ and $A_{55}$, with $D_{11}$ reduced by 25%, and with $A_{55}$ reduced by 25%.
MEASUREMENTS AND RESULTS

A new scanning system has been used that determines the velocity of the lowest order antisymmetric Lamb mode over a wide frequency range. In this measurement, one transducer is used to generate the Lamb mode and a second transducer is used as a receiver. For each frequency, the receiving sensor is moved by small increments to assure that the same peak in the waveform is followed over the total separation distance. The velocity at each frequency is calculated from the known transducer separation and the measured time-of-flight. The velocity measurements are accurate and repeatable to within 1%, resulting in reconstructed stiffness values repeatable to within 4% [20].

The elastic bending and out-of-plane shear stiffnesses of the material are computed from a reconstruction of the lowest order antisymmetric dispersion curve which best fits the data. For a laminated composite, the $D_{11}$, $D_{22}$, $A_{44}$, and $A_{55}$ stiffness matrix components can be determined. A mechanical scanner is used to move the sensors over the surface to map the time-of-flight, velocity, and stiffness of the entire specimen. Access to only one side of the material is required, and no immersion or couplants are required because the sensors are dry-coupled to the surface of the plate.

The Lamb wave scanning system was used to measure the stiffness in the 0° and 90° directions on unaged and aged specimens. The sensor separation was varied from 2.75 cm to 4.75 cm in increments of 0.4 cm. An 8-cycle Gaussian-enveloped sine wave was used to generate the signal, and the received signal was sampled at 25 MHz. The frequency was swept from 30 kHz to 200 kHz in 10-kHz steps, and the velocity at each frequency was obtained. The sample thickness was given above, and the density, estimated from common values for polymer matrix composites,
was taken to be 1560 kg/m$^3$. From the data for the 0° measurements, the dispersion curve was reconstructed, and values for the out-of-plane shear stiffness, $A_{55}$, and bending stiffness, $D_{11}$, were calculated. In a similar manner, the stiffnesses $A_{44}$ and $D_{22}$ were obtained from velocity measurements in the 90° direction.

Shown in Figure 5 are the experimental velocity measurements in the 0° direction for an unaged sample and one with 4,936 cycles of aging at a high strain level. Also shown are the reconstructed dispersion curves. As can be seen from the figure, the dispersion curve for the aged sample is clearly shifted from that of the unaged sample. The dispersion curves shown in Figure 5 were reconstructed using values for the out-of-plane stiffness, $A_{55}$, and the bending stiffness, $D_{11}$, which best fit the experimental data. For the curves shown, the value of $A_{55}$ decreased by 22% and the value of $D_{11}$ decreased by 6% for the aged sample as compared to the unaged sample.

It is expected, and previous strain gage measurements show [1], that matrix cracking due to fatigue damage in composites leads to a decrease in elastic moduli. Shown in Figure 6 is a photomicrograph of the edge of a sample aged 3,529 cycles, which shows matrix cracking occurring. The stiffness $A_{55}$ is controlled heavily by the matrix since the out-of-plane shear carrying capabilities of the composite are matrix dominated. Therefore, the large decrease in the stiffness $A_{55}$ was as expected. On the other hand, the stiffness $D_{11}$ is controlled by the fibers if the bending occurs in the fiber direction and by the matrix if the bending occurs perpendicular to the fibers. Due to the fact that a quasi-isotropic architecture was used for these samples, the bending stiffness will be affected by both the fiber and the matrix. Since fiber damage is probably not occurring in the samples, the value of $D_{11}$ will be influenced only by the matrix cracking. Therefore, a smaller decrease in $D_{11}$ would be expected.

![Figure 5](image.png)

**Figure 5.** Experimental dispersion curves for an unaged sample and a sample aged 4,936 cycles at a high strain level. Also shown are the reconstructed dispersion curves for the unaged (solid line) and aged (dashed line) samples.
Stiffness mappings for samples with a variety of aging profiles and times were made in situ as well as with the specimens removed from the environmental chambers. With the samples out of the chambers, a stiffness map of 75 different regions along the entire length of the panel could be obtained. For the in-situ measurements, only 20 stiffness measurements in the middle of the specimens could be made due to the constraints of the chambers. Measurements were made along the length of the samples to acquire mappings of $A_{55}$ and $D_{11}$ as well as across the width to obtain mappings of $A_{44}$ and $D_{22}$.

Sample results of stiffness mappings of $A_{55}$ are shown for three samples in Figure 7. Stiffness mappings were also obtained for values of $A_{44}$ from measurements in the $90^\circ$ direction. The results of these scans are shown in Figure 8. In the figures, the stiffnesses have been normalized to the average stiffness measured for the unaged sample in each set. The higher stiffness values in the regions at the bottom of the aged specimens were due to that portion of the sample remaining outside of the chambers and therefore not subjected to the same aging process as the rest of the sample. The average normalized stiffnesses and standard deviation for all of the samples measured are compiled in Table 1. Also shown in the table are the values of $D_{11}$ and $D_{22}$ measured for each sample.

The tabulated values indicate an increase in standard deviation for the aged samples as compared to the unaged samples. The larger standard deviation in the aged samples is most likely due to localized damaged regions developing in the specimens due to the aging process. The tabulated values also indicate that the standard deviations in the bending stiffnesses, $D_{11}$ and $D_{22}$, are 2 to 7 times greater than the standard deviations in the out-of-plane shear stiffnesses, $A_{55}$ and $A_{44}$. The large standard deviations in the values were most likely due to the insensitivity of the dispersion curve to changes in the bending stiffness over the measurement frequency range (30 kHz to 200 kHz). In this region, the parameter dominating the behavior of the curve will be the out-of-
Figure 7. Normalized stiffness mappings of $A_{55}$ for aged samples.

Figure 8. Normalized stiffness mappings of $A_{44}$ for aged samples.
plane shear stiffness (see Figure 4). Only a few data points exist at the very low frequencies (below 50 kHz), where the bending stiffness controls the behavior of the curve. Due to this lack of data, the constants $D_{11}$ and $D_{22}$ will not be as accurate as the constants $A_{44}$ and $A_{55}$. Therefore, the subsequent discussion will concentrate on the out-of-plane stiffness measurements.

Shown in Figure 9 are the normalized values of $A_{55}$ as a function of fatigue cycles for samples aged at both low and high strain levels. The results of $A_{44}$ from $90^\circ$ measurements for the same specimens are shown in Figure 10. A significant decrease was observed in both values of out-of-plane stiffness with extended aging. Additionally, it was observed that the stiffness for the high strain profile showed a greater decrease than the stiffness for the low strain profile at times approaching 5,000 fatigue cycles.

Plotted in Figure 11 as a function of fatigue cycles are the normalized stiffnesses $A_{44}$ and $A_{55}$ for the specimens aged at low strain levels. The same properties for the high strain profiles are shown in Figure 12. In both figures, the values of $A_{55}$ showed a greater decrease at each value of fatigue cycle than the values of $A_{44}$ for all but one data point. This behavior was expected, since the mechanical load was applied in the $0^\circ$ direction during the aging process. Loading in this direction tends to produce cracks in the $90^\circ$ plies as well as the $\pm45^\circ$ plies (see Figure 6). This type of damage would be indicated by a decrease in $A_{55}$. Since the loading occurred in the $0^\circ$ direction, matrix cracking in the $0^\circ$ plies probably did not occur or would not be as pronounced as in the $90^\circ$ plies. Therefore, the measurement of $A_{44}$ is sensitive only to the cracks in the $\pm45^\circ$ plies. This leads to a less significant decrease in stiffness for $A_{44}$, which is consistent with the observations.

In addition to the stiffness measurements, time-of-flight scans in the $0^\circ$ and $90^\circ$ directions were conducted on the samples which were removed from the environmental chambers. A 4-cycle Gaussian sine wave at a fixed frequency of 200 kHz was used to generate the antisymmetric

<table>
<thead>
<tr>
<th>Aging Cycles</th>
<th>Strain Profile</th>
<th>Normalized $D_{11}$</th>
<th>Normalized $A_{55}$</th>
<th>Normalized $D_{22}$</th>
<th>Normalized $A_{44}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>----</td>
<td>1.00 ± 0.11</td>
<td>1.00 ± 0.02</td>
<td>1.00 ± 0.10</td>
<td>1.00 ± 0.02</td>
</tr>
<tr>
<td>1,092</td>
<td>Low</td>
<td>1.01 ± 0.22</td>
<td>0.91 ± 0.06</td>
<td>0.97 ± 0.14</td>
<td>0.92 ± 0.08</td>
</tr>
<tr>
<td>2,339</td>
<td>Low</td>
<td>0.97 ± 0.06</td>
<td>0.90 ± 0.02</td>
<td>1.02 ± 0.08</td>
<td>0.95 ± 0.03</td>
</tr>
<tr>
<td>2,353</td>
<td>Low</td>
<td>0.90 ± 0.08</td>
<td>0.92 ± 0.02</td>
<td>0.86 ± 0.07</td>
<td>0.94 ± 0.02</td>
</tr>
<tr>
<td>4,420</td>
<td>Low</td>
<td>0.98 ± 0.11</td>
<td>0.88 ± 0.03</td>
<td>0.94 ± 0.11</td>
<td>0.90 ± 0.04</td>
</tr>
<tr>
<td>1,086</td>
<td>High</td>
<td>0.96 ± 0.12</td>
<td>0.91 ± 0.04</td>
<td>0.81 ± 0.06</td>
<td>0.87 ± 0.04</td>
</tr>
<tr>
<td>1,143</td>
<td>High</td>
<td>0.98 ± 0.09</td>
<td>0.90 ± 0.04</td>
<td>0.92 ± 0.15</td>
<td>0.88 ± 0.05</td>
</tr>
<tr>
<td>2,353</td>
<td>High</td>
<td>1.00 ± 0.15</td>
<td>0.91 ± 0.02</td>
<td>0.94 ± 0.11</td>
<td>0.92 ± 0.02</td>
</tr>
<tr>
<td>4,936</td>
<td>High</td>
<td>0.93 ± 0.16</td>
<td>0.80 ± 0.05</td>
<td>0.81 ± 0.06</td>
<td>0.85 ± 0.03</td>
</tr>
<tr>
<td>4,993</td>
<td>High</td>
<td>0.88 ± 0.08</td>
<td>0.83 ± 0.05</td>
<td>0.91 ± 0.11</td>
<td>0.87 ± 0.06</td>
</tr>
</tbody>
</table>
Figure 9. Normalized $A_{55}$ versus fatigue cycles for samples aged at low and high strain levels. (Note: the low-strain values have been displaced by +100 cycles in order to display the error bars clearly.)

Figure 10. Normalized $A_{44}$ versus fatigue cycles for samples aged at low and high strain levels. (Note: the low-strain values have been displaced by +100 cycles in order to display the error bars clearly.)
Figure 11. Normalized $A_{44}$ and $A_{55}$ versus fatigue cycles for samples aged at low strain levels. (Note: the values of $A_{44}$ have been displaced by +100 cycles in order to display the error bars clearly.)

Figure 12. Normalized $A_{44}$ and $A_{55}$ versus fatigue cycles for samples aged at high strain levels. (Note: the values of $A_{44}$ have been displaced by +100 cycles in order to display the error bars clearly.)
Lamb wave and the received signal was sampled at 25 MHz. The scan area was 73.0 cm by 24.0 cm for wave propagation in the $0^\circ$ direction and 73.0 cm by 22.0 cm for wave propagation in the $90^\circ$ direction. The step size for all scans was 1.0 cm and the transducers were held fixed at a separation distance of 2.75 cm. The results for the $0^\circ$ and $90^\circ$ scans are shown in Figures 13 and 14, respectively. In the figures, the values have been normalized to the average time-of-flight for the

**Figure 13.** Normalized time-of-flight scans for wave propagation in the $0^\circ$ direction for aged samples.

**Figure 14.** Normalized time-of-flight scans for wave propagation in the $90^\circ$ direction for aged samples.
unaged sample. As can be seen in the figures, and consistent with the previous stiffness measurements, an increase in time-of-flight was observed for the aged samples. The degraded stiffness in the aged specimens led to a slower Lamb wave velocity, which was reflected by the longer time-of-flight.

In addition to the longer overall time-of-flight, more features were observed in the samples than were seen in the stiffness scans. This was due to the smaller step size used in the time-of-flight scans. In the 0° scan for the unaged sample, a horizontal strip near the center of the sample was observed to have a lower time-of-flight. For this specimen, there was a visible wrinkle, which led to a slight thickening in that region. An increase in thickness translates to a faster Lamb wave velocity. This is in agreement with the indicated decrease in time-of-flight in that section of the panel. This wrinkle was not observed, however, in the 90° scan. This was probably due to the fact that the wave was travelling across the wrinkle in the 0° scan and along the wrinkle in the 90° scan. With the 1.0-cm step spacing, the sensors may have “jumped” over the wrinkle when the scanning direction was parallel to the wrinkle.

The sample aged at low strain levels did not show a significant change in the time-of-flight when compared to the unaged sample. However, some features were observed along the ±45° directions in the aged sample. This was most likely due to matrix cracking along those plies due to the loading. Additionally, the scan in the 0° direction for the sample aged at low strain levels appears more uniform than the 0° scan for the unaged sample. This could be due to a manufacturing anomaly in the sample that caused the wrinkle described above. For the sample aged at high strain levels, an overall increase in time-of-flight was observed for scans in both directions.

CONCLUSIONS

Lamb wave imaging is a unique tool for nondestructively measuring the elastic properties of a composite material. The method requires access to only one side of a specimen with no immersion or couplants. This study has shown the technique to be a very effective method for providing a quantitative measure of degradation due to thermal-mechanical aging of thermoplastic composite materials. The out-of-plane stiffness in both the 0° and 90° directions showed a significant decrease with extended aging times. The bending stiffnesses provided inconclusive results due to the large standard deviations in the values. Time-of-flight scans also showed an increase for aged samples as compared to an unaged sample. This corresponded to a slower velocity, which was in agreement with the reduced stiffness measured in the aged sample.

ACKNOWLEDGMENTS

This work was performed while Michael D. Seale held a National Research Council NASA-LARC Research Associateship. The authors would like to thank Steve Ziola, Wei Huang, and John Dorighi of Digital Wave Corporation for their technical support involving the scanning system. They would also like to thank Karen Whitley of NASA Langley Research Center and Steve Grossen of Lockheed Martin for the photomicrograph shown in this work.

REFERENCES


