EVALUATION OF THE COEFFICIENTS OF MOISTURE EXPANSION USING TRANSIENT SIMULATED LAMINATES METHODOLOGY (TSL)

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SUMMARY: In this study, Transient Simulated Laminates methodology(TSL) was demonstrated in order to evaluate hygrothermal environmental effects on the graphite/epoxy laminate. This method was based on the comparison between experimental results and numerical simulated results. In the theoretical part, the Rayleigh-Ritz method was extended to predict the shape of unsymmetric laminates $[0/0/90/90]_T$ in moist environments. In the experimental part, the curvature changes and the weight changes of the unsymmetric laminate were measured continuously during water absorption. This method was effective to understand the behavior of the laminate in the hygrothermal environments. It was found that there are discrepancies between experimental results and numerical results at the high moisture contents, specifically at the high water bath temperature during water absorption.

KEYWORDS: moisture, temperature, environmental effects, unsymmetric laminate, potential energy method, graphite/epoxy, water absorption, Thermal Analysis.

INTRODUCTION

After curing process, some kind of flat laminates show not only flat shapes, but also curved shapes, because of the existence of internal stresses. It is a well-known fact that some thin unsymmetric laminates at room temperature have cylindrical shapes rather than the saddle shapes predicted by classical lamination theory. A Rayleigh-Ritz method, in which the total of potential energy was minimized on the stable shape, was carried out by Hyer in order to predict the cylindrical shape of thin unsymmetric laminates[1]. It was reported that if the size of the laminate was larger than the critical size, the cylindrical shape provides a stable solution at room temperature.

There were other approaches focussed on the through-the-thickness deflections of laminates. One of these researches was the Process Simulated Laminate(PSL) theory, which was developed to evaluate the internal stresses in laminates, induced by thermal strain during the curing process[2]. The internal stresses were calculated by measuring the curvatures of the anti-symmetric laminates, which were made by machine milling or the separation by kapton films.

In this study, the moisture expansion of laminates during water absorption at various temperatures was evaluated using the Transient Simulated Laminates methodology (TSL).

This method was based on the comparison between experimental results and numerical simulated results. The numerical simulated results were carried out by the expanded Hyer's method to predict the shape of unsymmetric laminates in moist environments. The experimental results were carried out by measuring curvatures during water absorption, similar to the PSL method[3].

THEORETICAL FORMULATIONS

In general, it is assumed that a cured laminate is flat, stress-free and strain-free at the stress-free temperature under dry condition. The stress free temperature is generally 30°C below the curing temperature. As the laminate cools, the out-of-plane deflection develops from the differences in the thermal expansion properties of each ply. This assumption neglects the effects of any mechanical constraints during the autoclaving and bagging process. After the laminate cools to room temperature, the out-of-plane deflection changes as the temperature changes and as the moisture content of the laminate changes. The change of the out-of-plane deflection, induced by the change in moisture content, is brought about by the differences in the moisture expansion properties of each ply, just as the thermal expansion properties bring about the out-of-plane deflections as the temperature changes. As it is assumed that there are no external forces and momentum during cooling and water absorbing, the total of potential energy is the same as the total of strain energy in the laminate. The total strain energy W is represented as volume integral of strain energy density ω as follows[4]:

$$W = \int_V \omega \, dV \tag{1}$$

$$\omega = \frac{1}{2}\{\varepsilon\}^T D\{\varepsilon\} - \{\varepsilon\}^T D\{\alpha\}\Delta T - \{\varepsilon\}^T D\{\beta\}\Delta M \tag{2}$$

In Eqn 2, {ε} is the total strain vector, *D* is the stiffness matrix of the material, {α} is the coefficient of the thermal expansion vector, {β} is the coefficient of the moisture expansion vector, ΔT is the temperature difference, and ΔM is the moisture difference. In this equation, the total strain is consists of linear summation of thermal and moisture induced strains. The stiffness matrix, the coefficient of thermal expansion and the coefficient of moisture expansion are assumed constant in the range of temperature and moisture variation in this study.

In order to formulate the strains in the material, the in-plane and out-of-plane deflections of the mid-plane in global coordinates are assumed as follows:

$$\begin{aligned} u &= cx - \frac{a^2x^3}{6} - \frac{abxy^2}{4} \\ v &= dy - \frac{b^2y^3}{6} - \frac{abx^2y}{4} \\ w &= \frac{1}{2}\left(ax^2 + by^2\right) \end{aligned} \tag{3}$$

In this equation, *a*, *b*, *c*, and *d* are constants to be determined; *u*, *v*, and *w* are the deflections of the mid-plane in *x*, *y*, and *z* directions, respectively. The saddle shape and cylindrical shape of the unsymmetrical laminate can be predicted with this functional form of the out-of-plane deflection. When *a* equals -*b*, a saddle shape solution would be described. When *a* or *b* equals to zero, a cylindrical shape solution would be described; these two solutions show a snap-through phenomenon, which means that the one shape could be snapped into another shape. The reason why the deflections are assumed in this equation is that the in-plane strains

were similar to the predictions of classical lamination theory, in which they were independent of *x* and *y*, and there is no in-plane shearing strain. Using Eqn 3, the strains of mid-plane are given as follows:

$$\begin{aligned}
\varepsilon_x^0 &= \frac{\partial u}{\partial x} + \frac{1}{2}\left(\frac{\partial w}{\partial x}\right)^2 &&= c - \frac{aby^2}{4} \\
\varepsilon_y^0 &= \frac{\partial v}{\partial y} + \frac{1}{2}\left(\frac{\partial w}{\partial y}\right)^2 &&= d - \frac{abx^2}{4} \\
\varepsilon_{xy}^0 &= \frac{1}{2}\left(\frac{\partial u}{\partial y} + \frac{\partial v}{\partial x} + \left(\frac{\partial w}{\partial x}\right)\left(\frac{\partial w}{\partial y}\right)\right) &&= 0
\end{aligned} \tag{4}$$

The strains in any points in the laminates, which are based on von Karaman plate theory, are given as follows[5]:

$$\begin{aligned}
\varepsilon_x &= \varepsilon_x^0 - z\frac{\partial^2 w}{\partial x^2} &&= c - \frac{aby^2}{4} - az \\
\varepsilon_y &= \varepsilon_y^0 - z\frac{\partial^2 w}{\partial y^2} &&= d - \frac{abx^2}{4} - bz \\
\varepsilon_{xy} &= \varepsilon_{xy}^0 - z\frac{\partial^2 w}{\partial x \partial y} &&= 0
\end{aligned} \tag{5}$$

Eqn 2 is expanded and substituted by using Eqn 5 as follows:

$$\begin{aligned}
\omega = {} & \frac{1}{2}\overline{Q}_{11}\varepsilon_x{}^2 + \overline{Q}_{12}\varepsilon_x\varepsilon_y + \frac{1}{2}\overline{Q}_{22}\varepsilon_y{}^2 \\
& - \left(\overline{Q}_{11}\alpha_x + \overline{Q}_{12}\alpha_y\right)\varepsilon_x\Delta T - \left(\overline{Q}_{12}\alpha_x + \overline{Q}_{22}\alpha_y\right)\varepsilon_y\Delta T \\
& - \left(\overline{Q}_{11}\beta_x + \overline{Q}_{12}\beta_y\right)\varepsilon_x\Delta M - \left(\overline{Q}_{12}\beta_x + \overline{Q}_{22}\beta_y\right)\varepsilon_y\Delta M
\end{aligned} \tag{6}$$

In this equation, $\overline{Q}_{ij}$ is the reduce stiffness, α_x, α_y are the coefficient of the thermal expansion of x-direction and y-direction, and β_x, β_y are the coefficient of the moisture expansion of x-direction and y-direction. In this study, it is reasonable to assume α_{xy} and β_{xy} are both zero. According to the Rayleigh-Ritz method, when the first variation of the total strain energy equals zero, the total strain energy becomes minimized and it gives an upper-estimated solution. The first variation is given as follows:

$$\delta W = \left(\frac{\partial W}{\partial a}\right)\delta a + \left(\frac{\partial W}{\partial b}\right)\delta b + \left(\frac{\partial W}{\partial c}\right)\delta c + \left(\frac{\partial W}{\partial d}\right)\delta d \equiv 0 \tag{7}$$

This Eqn 7 leads to the next four equations:

$$\frac{\partial W}{\partial a} = 0, \quad \frac{\partial W}{\partial b} = 0, \quad \frac{\partial W}{\partial c} = 0, \quad \frac{\partial W}{\partial d} = 0 \tag{8}$$

By solving these four equations, the constants a, b, c, and d are fixed. The curvatures of the laminate κ_x, κ_y, κ_{xy} are given as follows:

$$\kappa_x = \frac{\partial^2 w}{\partial x^2} = a, \quad \kappa_y = \frac{\partial^2 w}{\partial y^2} = b, \quad \kappa_{xy} = \frac{\partial^2 w}{\partial x \partial y} = 0 \tag{9}$$

EXPERIMENTS

Three types of experiments were carried out in this study. First two experiments were investigated in order to determine the coefficients of thermal expansion and moisture expansion of the test specimens using of unidirectional test specimens. The unsymmetrical test specimens were used only in third experiment. All experiments were performed using Cytec-Fiberite HYE970/T300, a controlled-flow epoxy resin impregnated into a unidirectional graphite fiber tape. The resin content was 37% by weight. The laminates were made under the manufacturer recommended curing cycle, which included a curing temperature of 177°C (350°F) for two hours and a curing pressure of 552kPa (80 psi). The test specimens were cut from the laminates and all edges were trimmed and polished to give straight lines with right angles and smooth surfaces.

The coefficients of thermal expansion and the coefficients of moisture expansion of the material in the longitudinal and transverse directions were determined by experiments. The coefficients of thermal expansion were determined by measuring the dimensional changes of 26-ply, unidirectional test specimens using a thermomechanical analyzer (TA Instrument TMA 2940). The coefficients of moisture expansion were determined by measuring the dimensional changes and the weight change of 140×140mm quadrangle-shape, 4-ply, unidirectional test specimens, soaked in water baths at three constant temperatures: 28, 50, and 80°C. The measurements of dimension and weight were carried out at the room temperature under dry condition. It was assumed that the dimensional change in the through-the-thickness direction was negligible.

In order to compare between numerical simulated and experimental results, unsymmetric laminate test specimens were performed in moisture absorption studies. The stacking sequences of the test specimens were $[0/0/90/90]_T$ and the sizes were approximately 150×150×0.75mm. These test specimens were dried at 40°C in an oven with desiccant for 3-days to obtain initial moisture free condition, and they were subsequently immersed in water baths at three constant temperatures: 28, 50, and 80°C. All absorption experiments were conducted in de-ionized water.

The weights and curvatures of the test specimens were recorded periodically at room temperature throughout the experiments by removing the samples from the water baths, wiping water to get dry surfaces, and cooling them to room temperature. The weight changes due to the moisture absorption ΔM were defined as follows:

$$\Delta M = \frac{m - m_0}{m_0} \tag{10}$$

where, m is the transient weight of the test specimen, and m_0 is the initial weight.

The curvature of the test specimens along the x-direction and y-direction were calculated as follows:

$$\kappa_x = \frac{8h_x}{l_x^{\,2} + 4h_x^{\,2}}$$
$$\kappa_y = \frac{8h_y}{l_y^{\,2} + 4h_y^{\,2}} \tag{11}$$

In Eqn 11, κ is a curvature of the test specimen along each direction with suffix x and y, and *h* and *l* are the height and the length of the base of an arc as shown in Fig. 1.

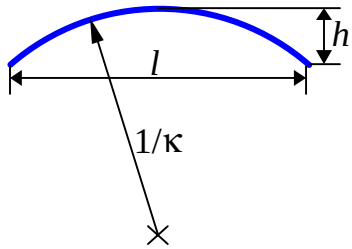

Fig. 1: Schematic figure of cylindrical shape and definitions of variables l, h, κ

Since all test specimens were described as the cylindrical shapes, therefore, either κ_x equaled zero or κ_y equaled zero, the indices will be dropped to simplify the notation and the curvature will be referred to as κ. The curvature change Δκ was defined as follows:

$$\Delta\kappa = \frac{\kappa - \kappa_0}{\kappa_0} \tag{12}$$

In this equation, κ is the transient curvature of the test specimen, and κ_0 is the initial curvature.

RESULTS AND DISCUSSIONS

Evaluation of Physical Properties

The coefficients of thermal expansion were found to be 3.57×10^{-6} °C^{-1} along the fiber direction and 4.96×10^{-5} °C^{-1} along the transverse direction, extrapolated at zero heating rate. The coefficients were determined over a temperature range of 25 to 100°C. As there were no dimensional changes along the transverse direction in a temperature range of 150 to 180°C, it was reasonable to assume the stress free temperature was 30°C below the curing temperature, i.e. 147°C.

The coefficients of moisture expansion were found to be zero along the fiber direction and 0.34 (kg/kg)$^{-1}$ along the transverse direction. These values were independent of the three water bath temperatures: 28, 50, and 80°C. The coefficients were determined up to a 1.6% weight change. There are non-linear dimensional changes at a weight change higher than 1.6%.

Numerical Simulated Results

In this numerical simulation, the curing temperature was 177°C, the stress free temperature was 30°C below the curing temperature, and the room temperature was 21°C. Therefore, the temperature difference ΔT was fixed at -126°C, which was calculated as (21°C - 147°C). The mechanical properties used in the calculations were as follows:

$$E_L = 124 \text{ GPa}, E_T = 8.6 \text{ GPa}, \nu_{LT} = 0.28, G_{LT} = 6.0 \text{ GPa}$$

The effects of the size of the square test specimens were carried out as a function of the weight change. Figure 2 shows the characteristics of the predicted shapes. The curvature at zero length is same as the curvature calculated by the classical lamination theory. On each weight-change percentage, if the length of the size of the test specimen is greater than a critical value, there are three possibilities of the shape: one is the saddle shape and the others are the cylindrical shapes. It was found that the cylindrical shape gave a stable solution, rather than the saddle shape gave an unstable solution if the length of the size was larger than the critical size. As the weight change increases, the curvature decreases to zero, finally the curvature goes to negative, and the critical sizes are shifted larger. In this study, the 150mm size of the test specimens was found larger enough to perform the cylindrical shape during water absorption.

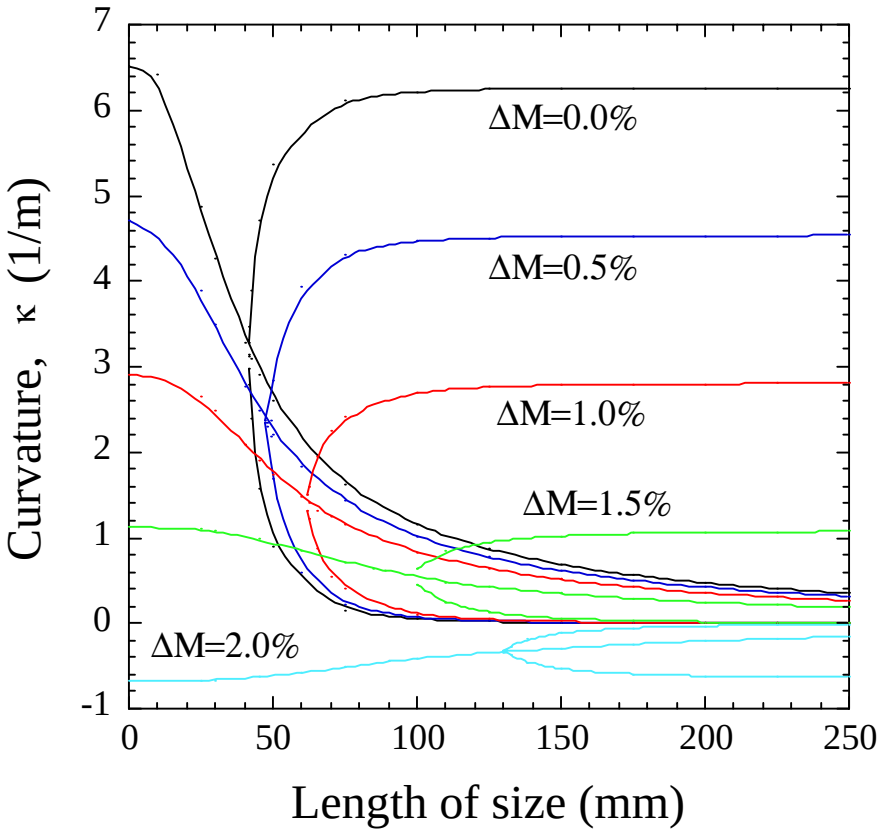

Fig. 2: The effects of the size of the square laminate on the curvature as a function of the weight change

Relationship between Curvature and Weight Change

Figure 3 shows the weight change and the curvature change of three test specimens as a function of normalized time, which was calculated as a square root of time divided by thickness, at the water bath temperature of 50°C. At the other water bath temperature, tendencies of the weight change and the curvature are similar to Fig. 3, except the equilibrium moisture contents and time-span related to the water diffusion rates. The weight changes of these test specimens were considered mainly from water absorption, dominated by the diffusion following to Fick's second law. As time past, the moisture contents increased and the curvatures decreased because of the differences in the moisture expansion properties of each ply.

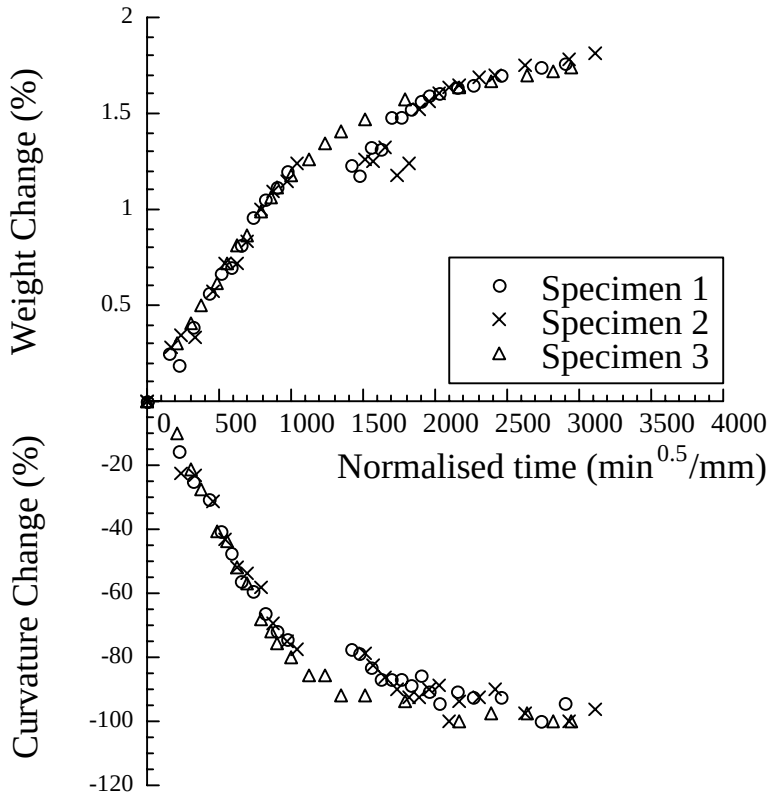

Fig. 3: Relationship of the weight change ΔM, the curvature change Δκ, and normalized time at the water bath temperature of 50°C

The relationship of the curvature change and the weight change of test specimens, comparing between numerical simulated results and experimental results, is shown in Fig. 4. The experimental results in the range of weight change from 0% to 1.0%, and the numerical simulated results seemed to be linear relationships between the curvature change and the weight change. The numerical values are almost corresponded to the experimental values, except the high weight-change region. In the high weight-change region, that is, high moisture contents region, the test specimen had a constant curvature, even if the weight was changed. The reason is considered the effect of physical structure changes, such as changes of polymer-network structure and rearrangements of molecular.

Additionally, the three slopes of the line, fitted to the experimental results of each water bath temperatures: 28, 50, and 80°C, were slightly different from each others. The number of test specimens to calculate the slope of the line is three specimens at each water bath temperature.

The differences in the slopes are considered out of range of experimental errors. As the water bath temperature increases, the slope of the fitted line to the results at that water bath temperature decreases. This means that the test specimen, soaking in the water bath at lower temperature, gives lower curvature at the same weight change percentage. In this temperature range of 28°C to 80°C, it is reasonable to assume that mechanical properties are constant, and it was found that coefficients of thermal and moisture expansion were constant by the experiments, which were described before. There is one possibility to explain this result, that the differences of the equilibrium moisture contents and Fick's diffusion rates of each temperatures affect the physical state of laminate specimen. In this numerical simulation, there is no consideration about hygrothermal cycling effects; therefore, it is not available to predict these discrepancies of the slopes.

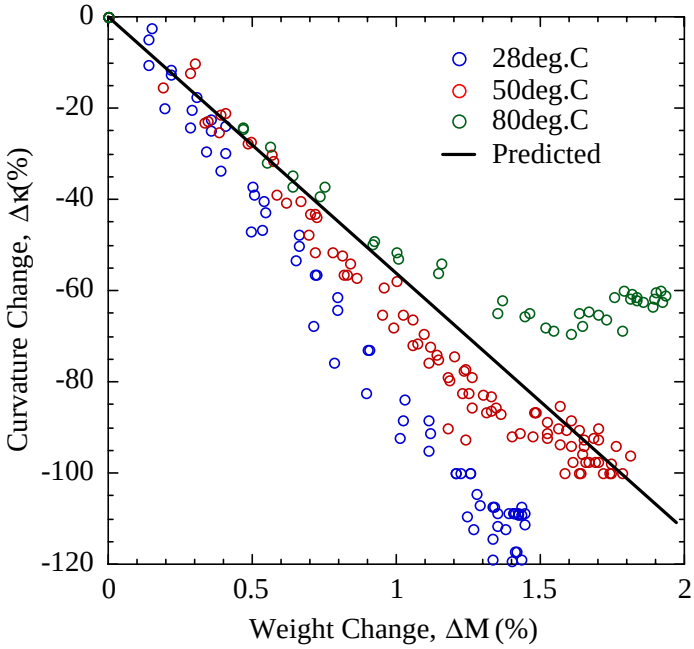

Fig. 4: Relationship between the weight change ΔM and the curvature change Δκ, at the three water bath temperatures: 28, 50, 80℃

CONCLUSIONS

In this study, Transient Simulated Laminates methodology(TSL) was demonstrated in order to evaluate hygrothermal environmental effects on the graphite/epoxy laminate. Although theoretical modeling of laminate was based on the simple linear summation of thermal and moisture induced strains, the measurements of curvature changes and weight changes of the unsymmetric laminate, comparing with the numerical simulated results, was effective to understand the behavior of the laminate in the hygrothermal environments. It was found that there are discrepancies between experimental results and numerical results at high moisture contents, specifically at the water bath temperature of 80°C. It was considered from the effects of physical structure change. In order to confirm these effects, further investigation will be required in hygrothermal cycling experiments, and theoretical modeling will be improved in the consideration of hygrothermal cycling.

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