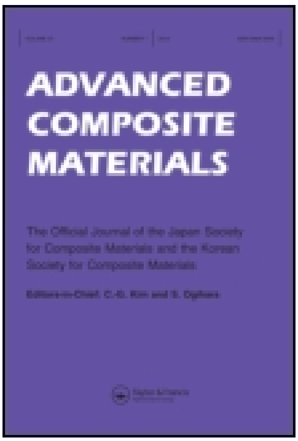

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Use of glass fibers in tailoring laminated composites with directionally negative and near-zero coefficients of thermal expansion

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Abstract—An analysis, together with experiments, has been made toward an intention indicated by the title of the paper. A simple closed-form expression is derived for the in-plane coefficients of thermal (thermoelastic) expansion (CTEs) in laminates of the type $[(\pm\theta)_n]_s$ from a consideration of the so-called 'normal-shear coupling' in anisotropic elasticity. By incorporating micromechanical expressions to estimate the lamina elastic propeties and CTEs, it is found that negative CTEs can occur if a polymer of very low modulus and very large CTE is used as the matrix material, and polypropylene is a candidate. This prediction is confirmed experimentally. Some stiffness-enhanced designs using 0° plies are also suggested.

Keywords: Glass fiber; polypropylene; laminated composites; thermal expansion.

1. INTRODUCTION

Laminated composites of fiber-reinforced plastics (FRPs) can exhibit 'anomalous' thermoelastic behavior, which is characterized by *negative* coefficients of thermal expansion (CTE) in one particular in-plane direction of the laminates. Experimentally, this behavior was reported first by Fahmy and Ragai [1] in carbon/epoxy composites and later by Strife and Prewo [2] in Kevlar/epoxy composites, both with simple $\pm\theta$ angle-ply lay-ups of unidirectionally fiber-reinforced plies. To our knowledge, no composite systems other than these have been identified as materials having such directionally negative CTEs. Does this imply that the anomaly in laminate CTEs never occurs in any FRPs that utilize glass fibers? If the answer is not affirmative, conventional 'low-cost' glass fibers may find new 'engineered' applications. With this possibility in mind, the work presented here was undertaken.

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In the following, we first deal with a simple, but most fundamental, bidirectionally fiber-reinforced laminate of the type $[(\pm\theta)_n]_s$ to show that a closed-form expression for the laminate CTE in its in-plane principal direction can be derived from a consideration of *normal-shear coupling* in the so-called *generally orthotropic* off-axis laminae. Using this macromechanical expression along with a set of micromechanical expressions for predicting the lamina elastic constants and CTEs, we then make a calculation to seek a candidate for the matrix material (along with its volume fraction) to be combined with glass fibers. Experimental results that follow will verify our prediction. Finally, with attention focused on laminated composite beams with *near-zero* CTEs, an ongoing effort to enhance the beam stiffness in bending by incorporation of 0° plies is described briefly.

2. SIMPLE $\pm\theta$ ANGLE-PLY LAMINATES

2.1. Closed-form expression for the laminate CTE

As is well known, thermoelastic properties of fiber composite laminates in general can be predicted by classical laminated plate theory (CLPT). However, this route of analysis requires rather complicated, but just algebraic, computations and does not necessarily illuminate the principles involved in our problem. Giving special attention to a simple $\pm\theta$ angle-ply laminate, we may take an alternative way of analysis. By making reference to Fig. 1, which illustrates a significant effect of normal-shear coupling characteristic of generally orthotropic (just like monoclinic) off-axis unidirectional (UD) laminae in the $\pm\theta$ laminate, it is easy to derive a closed-form expression for the laminate CTE under consideration:

$$\alpha_X(\theta) = \alpha_L \cos^2\theta + \alpha_T \sin^2\theta + \frac{\overline{S}_{16}(\theta)}{\overline{S}_{66}(\theta)}(\alpha_T - \alpha_L)\sin 2\theta. \tag{1}$$

Here, α_L and α_T are the longitudinal and transverse CTEs of UD lamina; $\overline{S}_{66}(\theta)$ and $\overline{S}_{16}(\theta)$ are the in-plane shear and normal-shear coupling compliances of $+\theta$ off-axis UD lamina in the X–Y coordinates fixed to the laminate, and these are given in terms of four effective elastic constants of UD lamina (i.e. the longitudinal Young's modulus E_L, the transverse Young's modulus E_T, the longitudinal shear modulus G_L, and the longitudinal Poisson's ratio ν_L) as

$$\overline{S}_{66}(\theta) = \left(\frac{1+2\nu_L}{E_L} + \frac{1}{E_T}\right)\sin^2 2\theta + \frac{1}{G_L}\cos^2 2\theta, \tag{2}$$

$$\overline{S}_{16}(\theta) = \left\{\frac{1}{E_L}\cos^2\theta - \frac{1}{E_T}\sin^2\theta + \left(\frac{\nu_L}{E_L} - \frac{1}{2G_L}\right)\cos 2\theta\right\}\sin 2\theta. \tag{3}$$

The expression above is quite informative in its form; the first two terms represent the free expansion of off-axis UD lamina and the last term an interactive synergistic

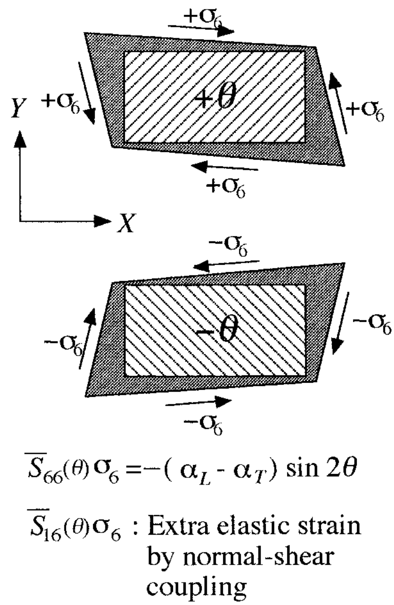

Figure 1. Schematic depiction illustrating the importance of normal-shear coupling for in-plane thermal expansion behavior of a symmetric, balanced, $\pm\theta$ angle-ply laminate. A rectangular piece of off-axis UD lamina does not remain rectangular after free expansion because of its anisotropic CTEs, i.e. $\alpha_L < \alpha_T$. When the $+\theta$ and $-\theta$ laminae are bonded together to form the laminate, equal-in-magnitude, but opposite-in-sense, in-plane shear stresses $+\sigma_6$ and $-\sigma_6$ are induced in the respective laminae. The elastic strains associated with these stresses must cancel out the mismatch in the in-plane shear component of strains due to anisotropic thermal expansion of the laminae; hence $\overline{S}_{66}\sigma_6 = -(\alpha_L - \alpha_T)\sin 2\theta$. Since the off-axis laminae each behave just like a monoclinic material, the shear stresses above cause elastic length changes in the X direction, i.e. extra normal strains, $S_{16}(\theta)\sigma_6$ and $-S_{16}(-\theta)\sigma_6$, are equally induced in the respective laminae, and these can be negative in a certain range of θ.

effect due to the lamination. When $E_L \gg E_T$, one gets an approximate equation:

$$\frac{\alpha_X(\theta) - \alpha_L}{\alpha_T - \alpha_L} \approx \sin^2\theta - \frac{\sin^2\theta + \frac{1}{2}\left(\dfrac{E_T}{G_L}\right)\cos 2\theta}{\sin^2 2\theta + \left(\dfrac{E_T}{G_L}\right)\cos^2 2\theta}\sin^2 2\theta. \tag{4}$$

Consider a special case of $E_T/G_L = 1$; then,

$$(\alpha_X(\theta) - \alpha_L)/(\alpha_T - \alpha_L) = -\sin^2\theta\cos 2\theta, \tag{5}$$

which gives a minimum value of $-1/8$ at $\theta = 30^\circ$, i.e. $\alpha_X = (9\alpha_L - \alpha_T)/8$ at this ply-angle. In fiber-reinforced plastics, $\alpha_T \gg \alpha_L$ because α_L is as small as the axial CTE of the fiber whereas α_T is dominated by the matrix CTE. Hence, it is evident that the condition $\alpha_L < 0$, which is usually met in carbon/epoxy and Kevlar/epoxy systems, is not essential for the laminate CTE $\alpha_X(\theta)$ to be negative.

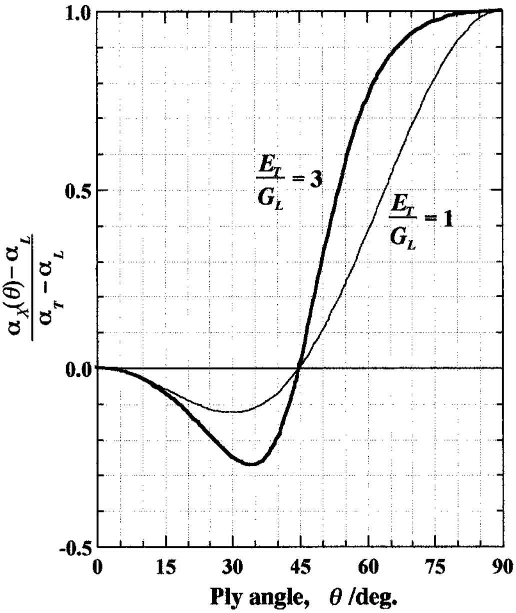

Figure 2. The laminate CTE, $\alpha_X(\theta)$, plotted against the ply angle θ, calculated from equation (4) with $E_T/G_L = 3$ (and with $E_T/G_L = 1$ for comparison). Note that the condition $E_L \gg E_T$ is assumed in equation (4).

Actually, $E_T/G_L \approx 3$ in most cases of practical interest (see footnote[1]) and then, the ratio $(\alpha_X(\theta) - \alpha_L)/(\alpha_T - \alpha_L)$ becomes more negative in a range of θ smaller than about 45° and takes a minimum value of -0.27 at $\theta \cong 33.5°$, as shown in Fig. 2. Thus, the approximation above suggests that $\alpha_X(\theta)$ can take negative values if $\alpha_T/\alpha_L \geqslant (1.27/0.27) \approx 4.7$.

2.2. Requirements for the matrix properties

The analysis shown above has been made entirely on a 'macromechanical' basis. For further examination to elucidate the possibility of glass fiber systems that we are particularly interested in, we need 'micromechanical' information about the lamina elastic properties and CTEs ($E_L, E_T, G_L, \nu_L, \alpha_L$ and α_T). To this end, we have used a set of expressions given in Table 1. The following values will be appropriate for the present examination: $E_\mathrm{f} = 75$ GPa, $\nu_\mathrm{f} = 0.2$ and $\alpha_\mathrm{f} = 5 \times 10^{-6}$ K^{-1} for the glass fiber, and $V_\mathrm{f} = 0.5$ and $\theta = 30°$ for the laminate. Asuuming $\nu_\mathrm{m} = 0.3$ as a typical value of Poisson's ratio for most polymeric matrices, we have calculated the laminate CTE $\alpha_X(\theta)$ as a function of both Young's modulus E_m and

[1] Using approximate expressions for E_T and G_L given in Table 1, one may derive $E_T/G_L \cong 4(\mathcal{P} + V_\mathrm{f})(1 + \mathcal{Q}V_\mathrm{f})/(1 + V_\mathrm{f})[(\mathcal{P} + V_\mathrm{f}) + (1 + \mathcal{Q}V_\mathrm{f})]$ with $\mathcal{P} \equiv (11 - 16\nu_\mathrm{m})/3(1 - 2\nu_\mathrm{m})$ and $\mathcal{Q} \equiv (11 - 16\nu_\mathrm{m})/(17 - 28\nu_\mathrm{m})$, where V_f is the fiber volume fraction and ν_m the matrix Poisson's ratio. Putting $V_\mathrm{f} = 0.5$ and $\nu_\mathrm{m} = 1/3$ gives $E_T/G_L \approx 3$.

Table 1.
Equations used to calculate the effective elastic properties and CTEs of unidirectional lamina

Longitudinal Young's modulus

$$E_L = (1 - V_f)E_m + V_f E_f + \frac{4\nu_f(1 - V_f)(\nu_f - \nu_m)^2}{(1 - V_f)/k_f + V_f/k_m + 1/G_m} \cong (1 - V_f)E_m + V_f E_f. \tag{a}$$

Longitudinal Poisson's ratio

$$\nu_L = (1 - V_f)\nu_m + V_f\nu_f + \frac{V_f(1 - V_f)(\nu_f - \nu_m)(1/k_m - 1/k_f)}{(1 - V_f)/k_f + V_f/k_m + 1/G_m} \cong (1 - V_f)\nu_m + V_f\nu_f. \tag{b}$$

Plane-strain bulk modulus

$$k_T = k_m + \frac{V_f}{1/(k_f - k_m) + (1 - V_f)/(k_m + G_m)} \cong \frac{k_m + V_f G_m}{1 - V_f}. \tag{c}$$

Longitudinal shear modulus

$$G_L = G_m + \frac{V_f}{1/(G_f - G_m) + \frac{1}{2}(1 - V_f)/G_m} \cong \frac{1 + V_f}{1 - V_f}G_m. \tag{d}$$

Transverse shear modulus

$$G_T = G_m + \frac{V_f}{1/(G_f - G_m) + (1 - V_f)(k_m + 2G_m)/2G_m(k_m + G_m)} \cong \frac{(1 + V_f)k_m + 2G_m}{(1 - V_f)(k_m + 2G_m)}G_m. \tag{e}$$

Transverse Young's modulus

$$E_T = \frac{4}{1/G_T + 1/k_T + 4\nu_L^2/E_L} \cong \frac{4}{1/G_T + 1/k_T}. \tag{f}$$

Longitudinal CTE

$$\alpha_L = \alpha_m + \frac{\alpha_f - \alpha_m}{1/K_f - 1/K_m}\left(\frac{3(1 - 2\nu_L)}{E_L} - \frac{1}{K_m}\right). \tag{g}$$

Transverse CTE

$$\alpha_T = \alpha_m + \frac{\alpha_f - \alpha_m}{1/K_f - 1/K_m}\left(\frac{3}{2k_T} - \frac{3(1 - 2\nu_L)\nu_L}{E_L} - \frac{1}{K_m}\right), \tag{h}$$

where V_f = the volume fraction of fiber (of circular cross-section); K = the bulk modulus; k = the plane-strain or transverse bulk modulus for transversely isotropic materials undergoing lateral dilatation without longitudinal extension ($k = K + G/3$ for isotropic materials); subscripts f and m refer to fiber and matrix, respectively. It is assumed that both constituents of the composite are isotropic.

CTE α_m of the matrix. Results of this calculation is summarized as a 2D contour map representation in Fig. 3. Here, diagrams (a) and (b) show the longitudinal and transverse CTEs of UD lamina, respectively. As is evident from diagram (c),

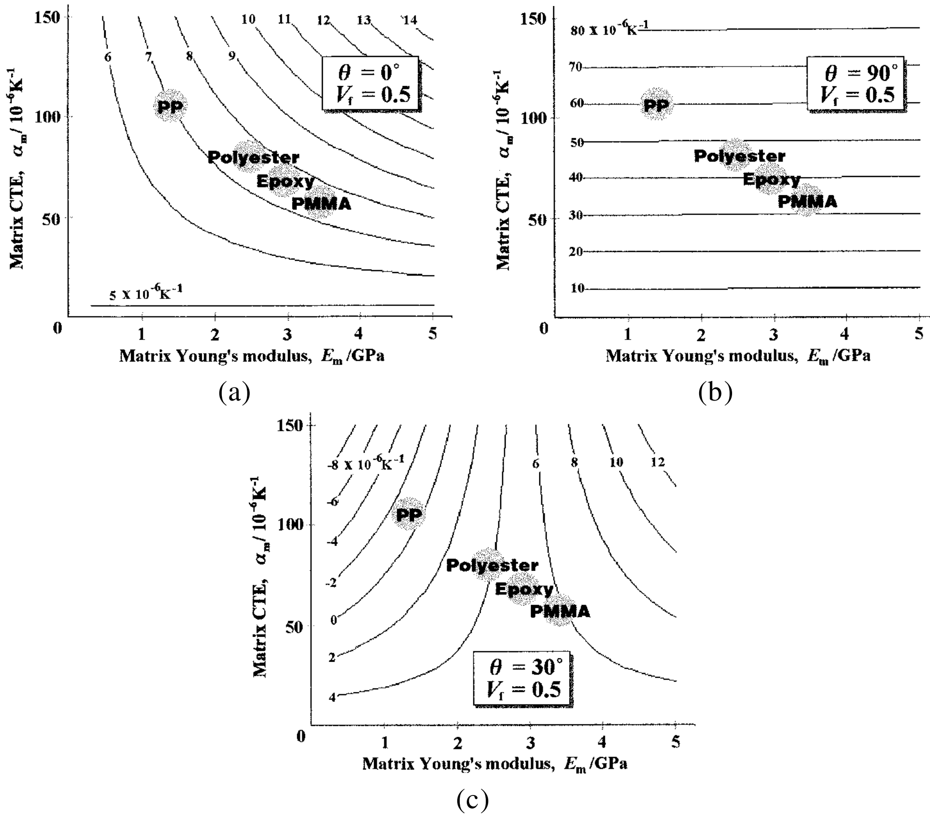

Figure 3. Contour maps showing the laminate CTEs as a function of the matrix Young's modulus, E_{m}, and the matix CTE, α_{m}, for (a) $\theta = 0°$, (b) $\theta = 90°$ and (c) $\theta = 30°$.

negative CTEs will occur if matrices of *very low modulus* and *very large CTE* are used, a candidate of which is polypropylene (PP). Note that the anomaly in laminate CTEs is never expected with such polymers of conventional GFRPs as epoxy and polyester.

2.3. *Experimental verification*

To verify our prediction for the glass/PP composite system experimentally, 8-ply laminates of three different lay-ups, $[0^{\circ}_{8}]$, $[(\pm 30^{\circ})_2]_{\mathrm{s}}$ and $[(\pm 45^{\circ})_2]_{\mathrm{s}}$, were fabricated using 45 vol% UD-glass/PP prepreg tapes of thickness 0.2 mm, and in-plane thermal expansions in two principal directions of these laminated orthotropic materials were measured in the range from room temperature (RT) to 120°C by means of scanning laser extensometry. Results are shown in Fig. 4, which displays typical strain–temperature curves on heating. It is evident that the negative CTE response occurs in the laminate with $\theta = 30°$. Close inspection of the thermal expansion data reveals that the slope of each curve changes at temperatures around 40°C and 90°C. Thus the average CTE values were determined separately in three

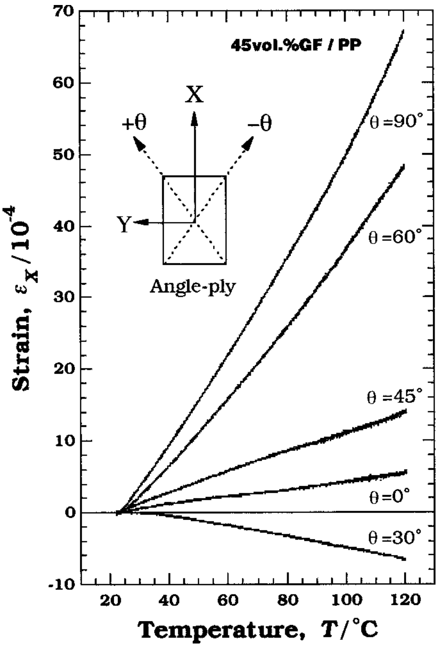

Figure 4. Typical thermal expansion curves on heating for $\pm\theta$ angle-ply laminates of 45 vol% glass fiber/polypropylene composite, measured in the X direction at a heating rate of 0.5°C/min.

different ranges: (I) RT to 35°C, (II) 45–85°C and (III) 100–120°C. In Fig. 5, results are plotted and compared with predictions by equations (1) to (3). Here, the observed CTE values at $\theta = 0°$ and 90° are used as the input data for α_L and α_T in equation (1), together with the calculated lamina elastic constants listed in Table 2. A good agreement can be seen for all other data points with the predicted curves.

3. LAMINATES WITH NEAR-ZERO CTEs

3.1. A primitive example

As is evident from the diagram shown in Fig. 5, the simple lamination with $\pm\theta$ plies of the 45 vol% glass/PP composite gives near-zero CTEs at certain values of θ while the determination of these θ values depends on the temperature ranges indicated. If attention is focused on range II (45–85°C) for example, the θ value of either 18° or 39° is suggested. Of these, the former angle is of interest for two reasons. First, a greater axial stiffness is expected in that laminate. Second, fiber misorientations that may be encountered in making experimental 'small-size' laminates will have a comparatively small effect. Thus, a $[(\pm 18°)_2]_s$ laminate was prepared, and its thermal expansion measured. The result confirmed our prediction; its average CTE in the range 45–85°C was about 0.6×10^{-6}/K, which can be well compared with the value 0.17×10^{-6}/K predicted.

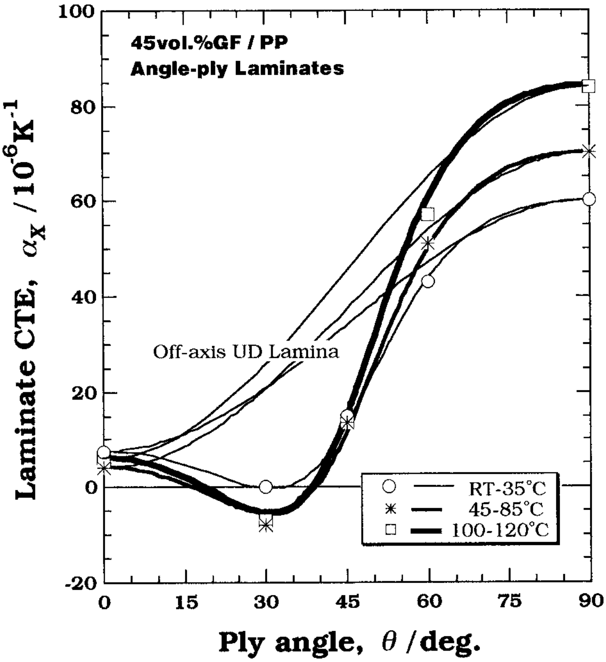

Figure 5. Comparison between measured and calculated in-plane CTEs of $\pm\theta$ angle-ply laminates. The calculation, based on equation (1), uses the measured CTE values at $\theta = 0°$ and $90°$ as input data for α_L and α_T. Calculated angular dependence of CTE for the off-axis UD lamina is also shown by dotted lines to illustrate the effect of the lamination.

Table 2.
Elastic properties of 45 vol% glass fiber/polypropylene unidirectional lamina, calculated from constituent elastic constants[a]

Longitudinal Young's modulus, E_L (GPa)	34.5
Transverse Young's modulus, E_T (GPa)	3.1
Longitudinal shear modulus, G_L (GPa)	1.3
Longitudinal Poisson's ratio, ν_L	0.25

[a] Young's modulus (E) and Poisson's ratio (ν) of glass fiber (GF) and polypropylene (PP) are: $E_{GF} = 75$ GPa, $\nu_{GF} = 0.2$, $E_{PP} = 1.3$ GPa and $\nu_{PP} = 0.3$.

3.2. Stiffness-enhanced designs

The directionally near-zero CTE behavior will be of practical interest, especially if the laminate is formed into slender configurations with various cross-sectional shapes. In making such axially very low-expansive solid or hollow beams as elements for structural application, their stiffnesses in axial bending (and also in stretching possibly) will be an important consideration. Thus, the incorporation of axially reinforcing 0° plies as outer skin layers is of further interest. While this is now under examination, some preliminary data show quite encouraging results. For instance, a 10-ply laminate of the type $[0°/(\pm\theta)_2]_s$, the simplest case

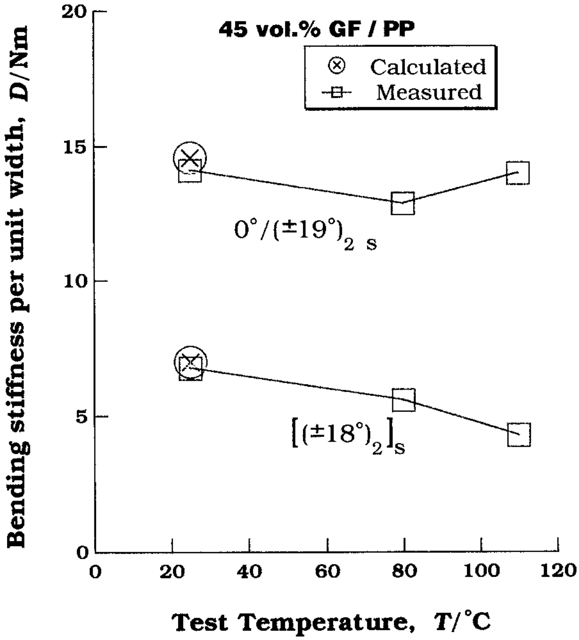

Figure 6. Axial bending stiffnesses of two different laminated beams, $[(\pm 18^\circ)_2]_s$ and $[0^\circ/(\pm 19^\circ)_2]_s$, measured at 25, 80 and 110°C by four-point flexural testing using specimens of length 180 mm, width 15 mm and of different thicknesses 1.6 and 1.9 mm for the respective laminates, with outer span 120 mm and inner span 40 mm. Constant load was applied in a stepwise manner and the resulting surface strain was meared with a foil strain gage. The strain response in the duration of 2 h was nearly constant at 25°C and 80°C but slightly changed at 110°C. Linearity between moment and curvature was good especially at 25°C and 80°C.

Table 3.
Calculated and measured CTEs of the $[0^\circ/(\pm 19^\circ)_2]_s$ laminate

CTE	Temperature range		
α (10^{-6} K^{-1})	25–35°C	45–85°C	100–120°C
Calculated	3.5	0.15	0.41
Measured	3.0	−0.14	−2.9

for stiffness enhancement, is expected to have an average CTE of 0.15×10^{-6}/K in the range 45–85°C when $\theta = 19^\circ$. Also, preliminary estimates by CLPT show that the axial bending stiffness of this particular laminate is about twice greater that of the $[(\pm 18^\circ)_2]_s$ laminate (although this naturally involves a thickness effect). Experimental results generally confirmed the above predictions, as summarized in Table 3 and Fig. 6. Of further interest are such laminates of sandwich constuction as $[0^\circ/(\pm\theta)_2/(\mathrm{PP})_n]_s$, which will be discussed elsewhere.

To summarize, the present investigation based on combined macro- and micromechanics analyses for simple $\pm\theta$ angle-ply laminates of fiber-reinforced composite has predicted and demonstrated a new type of fiberglass laminate that may find various engineered applications.

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