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Angular dependence of hygroelasticity in unidirectional glass-epoxy composites

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Swelling and hygroelasticity of unidirectional glass fibre-reinforced epoxy composites in boiling water is studied with respect to the angle θ between the direction of orientation and the direction of measured dimensional change. The results are expressed in terms of the dependence of θ of a hygroelasticity coefficient. A very good agreement is observed between the experimental results and the theoretical equation, already verified in the case of the angular dependence of the coefficient of thermal expansion. This is taken as additional support for the analogy between thermoelastic and hygroelastic behaviour in composites. The experimental longitudinal and transverse hygroelasticity coefficients are found to be different from predictions based on Schapery's equation. However, it is shown that these equations can produce possible valid estimates of μ_L and μ_T provided the mechanical properties of the swollen constituent materials are used.

1. Introduction

The understanding of hygroelasticity defined as reversible swelling of polymers and of composites is very important, since most of the applications result in their exposure to liquids or vapour. By knowing the hygroelastic behaviour it is possible to predict the dimensional change and the lifetime of the material. The problem of the analogy between hygroelasticity and thermoelasticity in polymers and in composites has attracted our attention several times during the last two years. The hypothesis put to test was that hygroelasticity follows the same physical laws of thermoelasticity. So far, this hypothesis has been checked experimentally in two studies with positive results in both of them. The first study [1] showed that in oriented PMMA the coefficient of hygroelasticity is affected by the extent of orientation exactly as is the coefficient of thermal expansion. The axial coefficient of hygroelasticity exhibited a linear decrease, and the transverse coefficient exhibited an increase with increasing extent of orientation – in strict analogy with thermoelasticity. The second study [2] showed that the longitudinal and the transverse coefficients of hygroelasticity of a unidirectional glass fibre-reinforced epoxy composite were reasonably predictable from the coefficients of the constituent materials by Schapery's equations, proposed for the prediction of the coefficients of thermal expansion [3].

The present study was conducted in order to examine the angular dependence of hygroelacticity in unidirectional composites. Following a similar line of thought as above, the question asked was whether or not the Fahmy–Ragai approach to the angular variation of thermal expansion coefficient [4] could give a valid estimate of the angular dependence of hygroelasticity.

The coefficient of hygroelasticity was previously defined as follows [2]

$$\mu = \frac{\Delta L/L_0}{\Delta W/V_0} \tag{1}$$

where $\Delta L/L_0$ is the relative length change, V_0 is the initial volume of the material, and ΔW is the weight increase due to liquid absorption (see also [5]). Since thermoelastic and hygroelastic strains are second rank tensors, the coefficient in a direction θ is given by the following transformation (see for example [6]).

$$\mu_\theta = \mu_L \cos^2\theta + \mu_T \sin^2\theta \tag{2}$$

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where θ is the angle between the fibre direction and the direction of measured expansion, and μ_L ($\theta = 0°$) and μ_T ($\theta = 90°$) are the longitudinal and the transverse coefficients of hygroelasticity, respectively.

Equations based on energy principles were proposed by Schapery [3] for longitudinal and transverse thermal expansivity of composites consisting of a matrix with relatively stiff parallel fibres that are continuous and straight. Assuming the applicability of Schapery's equation, μ_L and μ_T are given as follows:

$$\mu_L = \frac{E_m \mu_m V_m + E_f \mu_f V_f}{E_m V_m + E_f V_f} \tag{3}$$

and

$$\mu_T = (1 + \nu_m)\mu_m V_m + (1 + \nu_f)\mu_f V_f - \mu_L(\nu_f V_f + \nu_m V_m) \tag{4}$$

where the subscripts m and f denote matrix and fibre properties, respectively, ν is Poisson ratio, V is the volume fraction, and E is Young's modulus.

2. Experimental

The materials employed were unidirectional E-glass fibre-reinforced epoxy composites in the form of thin plates (0.8 to 1.2 mm). The glass fibres were silane-treated EC 14-300-K937 (Vetrotex), suitable for epoxy resins. The specifications of the materials are given in Table I.

Four square (4 cm x 4 cm) test specimens were cut from each plate at different angles θ with fibre direction as follows: $\theta = 0°, 15°, 30°, 45°$. Dilatation measurements were made with each test specimen along its two major dimensions resulting in additional angles θ of 45°, 60°, 75°, 90°.

For hygroelasticity measurements specimens were immersed in boiling distilled water for 24 h, and taken out periodically for weighing and dilatation recordings. The recordings were carried out at 23° C, and each specimen was cooled down in distilled water before readings were taken. ΔL was measured by placing each specimen in a stainless steel fixture attached to a dial gauge capable of measuring to 0.5 μm.

TABLE I Specifications of composites

Sample	Resin	V_f	V_m	V_v*
A	Araldite F/HT972	0.28	0.55	0.17
B	Araldite F/HT972	0.40	0.52	0.08
C	ERL2256	0.51	0.48	0.01

* Volume fraction of voids.

The exposure of the test specimens to boiling water was expected to result in a gradual degradation of the fibre–matrix interfacial bond, and therefore to affect the hygroelastic behaviour. It was found [7], however, that the effect of the degradation is important during the first 8 h, and that thereafter it becomes negligible. Thus, as a precaution, the interface was conditioned by water-boil treating each specimen for 8 h and then drying it at 120° C to obtain a zero water content before applying the above hygroelasticity measurement procedure.

3. Results and discussion

Fig. 1 presents an example of plots of $\Delta L/L_0$ versus swelling period for the various angles θ. The plots represent the smoothed data reproduced from the experimental data; similar plots were made for the $\Delta W/V_0$ results. Since the $\Delta W/V_0$ data do not show any significant trend with θ, the results in Fig. 1 can already be taken to represent the hygroelastic behaviour as a function of θ.

The coefficients of hygroelasticity were worked out from plots of $\Delta L/L_0$ against $\Delta W/V_0$ generated from the smoothed data taken at 1 h intervals. Each of these plots was linear up to 10 h of water-boil treatment, producing the appropriate value μ by its slope. Following Fahmy and Ragai's approach the *measured* values of the hygroelasticity coefficients at 0° (μ_L), and at 90° (μ_T) were then used to plot

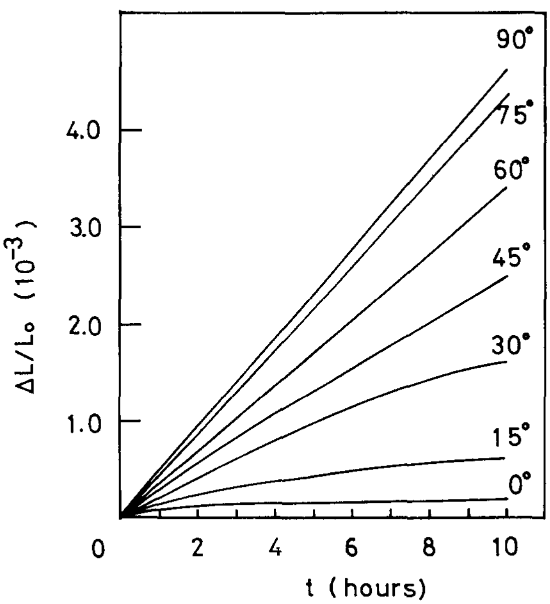
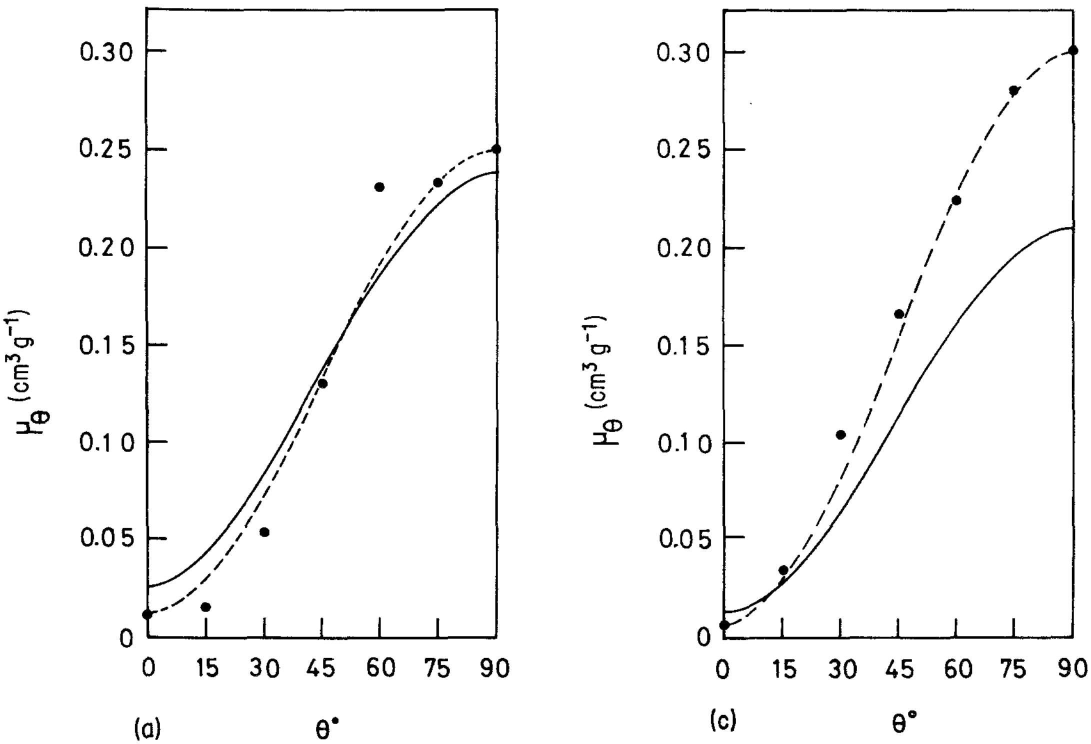


Figure 1 Relative dimensional change versus water-boil period as a function of the angle θ.



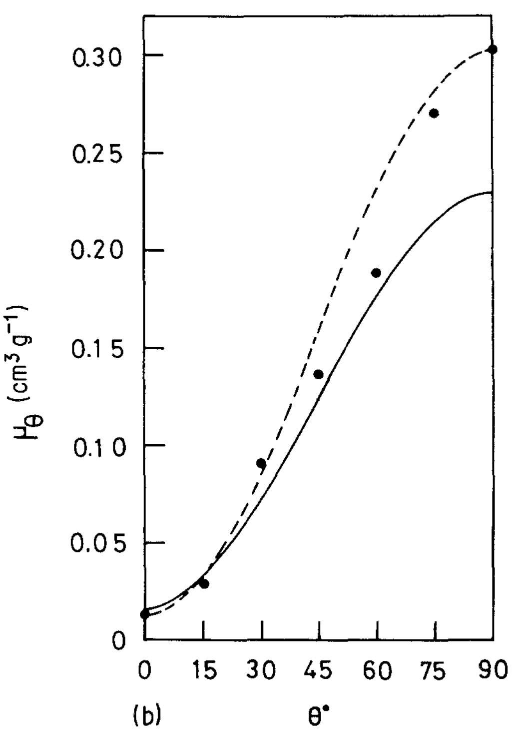


Figure 2 Coefficients of hygroelasticity as functions of θ, compared with the Equation 2, based on experimental μ_L and μ_T (– – –), and on μ_L and μ_T worked out by Schapery's equations taking the constituents' original properties (——). (a) Sample A, (b) sample B, (c) sample C.

Equation 2. The experimental values of μ_θ are presented in Fig. 2 for the three composites tested. It is seen that in each case, and in particular in sample C containing fewer voids, the experimental results are in close agreement with Equation 2 predictions. These results indicate that once μ_L and μ_T are known it is then possible to work out the coefficient of hygroelasticity at any direction.

Fig. 2 also includes plots of Equation 2 worked out by μ_L and μ_T as predicted by Equations 3 and 4. It is seen that the experimental μ_L and μ_T values are different by an average of − 35% and of + 25%, respectively from the theoretical predictions. These predictions are based, however, on the original properties of the constituent materials using the following data: $E_m = 3.0$ GPa, $\mu_m = 0.33$, $\nu_m = 0.35$, $E_f = 70.0$ GPa, $\mu_f = 0$, $\nu_f = 0.20$. The difference between the experimental and the calculated values of μ_L and μ_T obviously results in deviations of the experimental results from the theoretical μ_θ curve.

The inability of Equations 3 and 4 to produce valid predictions of μ_L and μ_T for the samples tested here, may be a result of two causes. The first is the weak interface as obtained by the pretreatment of the samples, and the second is the relatively high swelling extent. Regarding the second point, it should be noted that the present study employed much thinner specimens than the previous one [2], hence the present ratio of surface area to volume of test specimens was much higher, and so were the swelling extents (typical $\Delta W/V_0$ values of $16 \times 10^{-3}\,\mathrm{cm}^3\,\mathrm{g}^{-1}$ after 10h for sample C compared with $5 \times 10^{-3}\,\mathrm{cm}^3\,\mathrm{g}^{-1}$ for specimens of a similar fibre length in [2]). Such higher swelling is expected to produce a marked change in the original properties of the matrix, e.g., to decrease E_m and to increase ν_m. If, for example, the original E_m is reduced by swelling to 2.0 GPa and ν_m is increased to 0.40, the experimental μ_L and μ_T values will only differ by an average of -8% and of $+17\%$, respectively from the predictions. It is, therefore, thought that Schapery's equations can produce a valid estimate of μ_L and μ_T provided the mechanical properties of the swollen constituents are used.

Due to the strong effect of the mechanical properties of the swollen matrix on the values of μ_L and μ_T it is impossible to compare samples A, B and C, since either their void contents or the matrix materials are different. This is expected to produce a larger effect, and in fact to mask the effect of the varied fibre volume fraction.

4. Conclusions

From the above results we conclude as follows: (i) Equation 2 is very successful in predicting angular dependence of the coefficient of hygroelasticity in unidirectional glass-epoxy composites; and (ii) Schapery's equations can produce a better estimate of the longitudinal and transverse coefficients provided the mechanical properties of the *swollen* constituents are used in the equation.

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