

CHARACTERIZATION OF THERMAL EXPANSION OF THERMO-SET COMPOSITES

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ABSTRACT

Use of composite materials in engineering applications has been increased significantly during the last decades due to their good mechanical properties despite lighter weight. A significant advantage of such materials is the choice of selection among several polymers and different types of reinforcement (fibre/fabric) types to achieve required properties. But this heterogeneous nature also induces several problems and makes thermo-mechanical behaviour complex. Thermal expansion of thermoset composite material is thus a parameter to know accurately. In laminated composites, this parameter depends largely on the orientation of plies, fibre fraction, type of resin and fibres, etc. In this article, some results on the determination of coefficients of thermal expansion (CTE) of composite plates (with stacking sequence 0, 0/90, 0/45, +/-45) at different angles in XY, XZ, and XYZ plane using finite element analysis and homogenized properties are presented. Experimental values of these coefficients for 0/90 stacking are found in agreement with the simulations. Finally, a mathematical model, based on strain matrix is proposed for modelling these coefficients.

Keywords : thermoset composites, liquid infusion moulding, thermal expansion, dilatometer

1. INTRODUCTION

Use of composite materials in engineering applications has been increased significantly during the last decades due to their good mechanical properties despite lighter weight [1]. Depending of type of end use different types of reinforcement e.g. unidirectional fibres, woven fabric, knitted, or braded fabrics, are used to achieve required properties in composite part [2]. The polymers used in composites are classified as: thermoset and thermoplastic ones. Thermosetting polymers e.g. epoxy, unsaturated polyester, and vinylester are commonly used in aerospace and naval applications due to their low viscosity, which makes easy the impregnation of reinforcement and hence process of fabrication.

Knowledge of thermal expansion and /or shrinkage is essential to characterize the behaviour of thermoset composite materials [3]. In case of laminated composites, the differences between the coefficients of thermal expansion of the constituents, i.e. fibres, matrix, and

mould lead to the formation of residual stresses and deformations in the composite part [4-7]. Characterization of these coefficients is thus essential for modelling. Generally, it is considered that out of plane coefficient of thermal expansion (CTE) is equal to the transversal (in-plane) thermal coefficients [8].

In the present article, thermal expansion coefficient of a unidirectional glass/epoxy laminated composite plate was determined in different directions in the plane, and out of plane of the piece. It was found that the out of plane coefficients are not equal to the in-plane transversal coefficients, which is against the previously believed concept. Moreover, a significant effect of Tg was also observed on the thermal coefficients of the composite. These coefficients were also modelled using strain matrix and found in agreement with the experimental ones.

2. MATERIAL AND METHODS

In this study, a laminated composite plate (300 mm× 300

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mm× 40 mm) was fabricated using vacuum assisted resin transfer moulding (VARM) such as the fibre volume fraction is equal to 50%. All the plies were stacked at zero degree. The polymerization was carried out at room temperature.

Parallelepiped samples (10mm× 10mm× 30-40mm) were cut into the composite plate, in directions defined by a direction coefficient 0 or 1, according to the axis X, Y and Z: (1,0,0), (0,1,0), (0,0,1), (1,1,0), (0,1,1), (1,0,1) and (1,1,1). To determine the CTE, a classical dilatometer was used. Samples are given names (Table 1) for easy understating.

Table: Samples names and corresponding orientations

Name	S1	S2	S3	S4	S5	S6	S7
Orientation	100	010	001	110	101	011	111

For determination the Tg of composite, a differential scanning calorimeter (DSC Q200-TA instruments) was used.

2.1. DIFFERENTIAL SCANNING CALORIMETER

This technique has been used to identify heat of cure, specific heat capacity, glass transition temperature (Tg) and degree of cure (α) of polymer [9]. Differential Scanning Calorimeter (DSC) consists of two cells called reference and sample cell. During the heating, it measures the difference of heat that is consumed by sample as compared to the reference, for reaching a given temperature. The instrument used in our study was a DSC Q200 from TA instruments.

2.2. DESCRIPTION OF DILATOMETER

DI.24 ADAMEL LHOMARGY® is the dilatometer, which was used for the thermal expansion measurements of composite samples. It consists of an oven, an aluminium support, thermocouples, and LVDT sensors.

1. The oven consists of a heater based on the silicon carbide resistor, which can heat the sample from room temperature to 1600°C. The oven can be moved on a rail to facilitate the sample placement.
2. Silicon support, for placing the samples of dimensions 10 to 50 mm long in the oven. The cross-section of sample can be up to 10mm x10mm.

3. Two thermocouples are employed, one in the silicon support for the measurement of sample temperature, and second in the centre of the oven to control the heating temperature.

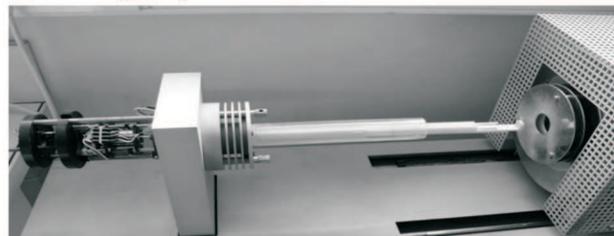


Figure 1: DI.24 (ADAMEL LHOMARGY® dilatometer

The head consists of a LVDT (Linear variable differential transformer) type displacement sensor, and two screws for adjusting the zero and initial load on the sample. The sample placed on aluminium support is hold with a pressing bar. This bar can be positioned with two screws to adjust zero strain and initial pressure on the sample (1mm in our case). This bar is linked to the LVDT sensor. The length variations during the heating/cooling ramps are then recorded. The precision of used instrument is +/- one micrometer.

3. RESULTS AND DISCUSSION

3.1. DETERMINATION OF TG

Glass transition temperature (Tg) is the temperature at which polymer converts from glassy state to rubbery state or vice versa. This temperature has a significant effect on thermo-mechanical properties of polymers and polymer reinforced composites. Therefore, it is an important factor to find out. In the present paper, differential scanning calorimeter was used to characterize Tg of glass/epoxy composite samples. 10-15 gram of composite sample were sealed in aluminium hermetic pan and scanned for the temperature range of 0°C -100°C at the heating rate of 4°C/min. The average value of Tg was found equal to 56 ± 2 °C.

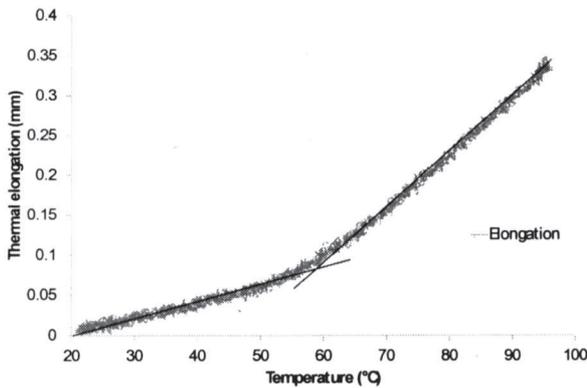
3.2 DETERMINATION OF THERMAL COEFFICIENTS

In order to determine the thermal coefficient, samples were submitted to the following thermal cycle:

1. Heating from room temperature to 100°C at the rate of 2°C/min
2. Cooling to 20°C at rate of 2°C/min

3. Maintaining at 20°C for 80 min

This cycle was repeated 2-3 times for a sample. For each orientation, two samples were tested, and then average of both results was taken as the final results.



Tg

Figure 2: Thermal elongation versus temperature response of samples 011

In Figure 2, Elongation response of composite sample 011(diagonal in xz plane) during the heating step is plotted versus temperature. Slope of a graph is directly proportional to the coefficient of thermal expansion, which can be found using the simple expression:

$CTE = \frac{1}{e_0} \left(\frac{\partial e}{\partial T} \right)$, where e_0 is the initial length of the sample, T is the temperature, and e is length at this temperature.

The elongation versus temperature curve can be divided into two linear zones in figure 2. The first one starts from room temperature to the temperature of 56-58°C. In this zone composite was in the glassy state. Therefore, slope of the graph of this portion when divided by sample length will result into CTE_{glassy}. Whereas, the slope of second linear portion after divided by sample length will result into CTE of composite in rubbery state. This change of slope occurs in the zone of glass transition of composite sample (56 ± 2 °C found by DSC). Therefore, it can be concluded that the change in slope is due to glass transition from glassy to rubbery state.

Figure 3 shows the comparison of CTEs below Tg of all the tested samples. It can be noted that CTE of the sample S₁ (along the fibres direction) is minimale (6.1×10⁻⁶ K⁻¹). The value of CTE in transversal direction (S₂) is higher

than the value of CTE through the thickness (S₃). This result was surprising but several repetitions on samples from different composite parts gave the same results which lead us to conclude that this result is correct.

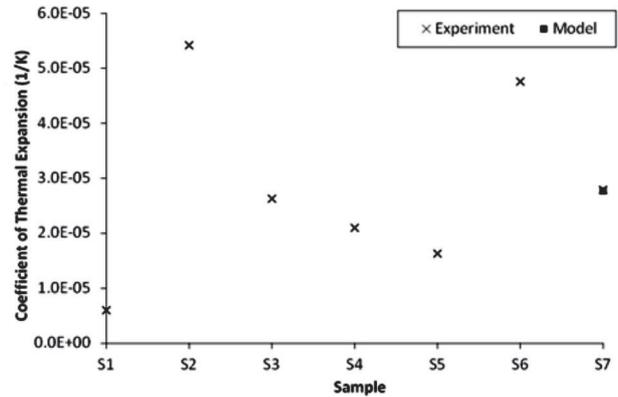


Figure 3: Linear coefficients of thermal expansion (1/K)

CTE of sample S₅ (diagonal in xz plane) has lesser value as compared to other diagonal samples (S₄, and S₆), which is normal because no component of CTE along y-axis was included in this coefficient. CTE of samples S₇ was found approximately equal to CTE along z-axis.

The next step was to propose a model for CTE, depending on angle of orientation of fibres. At first, an attempt was made to use strain matrix to calculate the CTE₁₁₁ (of a sample in space making 45° with its projection in xy plane) from the experimental values of other six CTEs, which was then compared with the experimental value. The strain ϵ_{xyz} (denoted by ϵ in equation below) is given by the matrix:

$$\epsilon = \begin{bmatrix} \frac{\epsilon_x}{\sqrt{3}} + \frac{\epsilon_y}{\sqrt{3}} + \frac{\epsilon_z}{\sqrt{3}} \\ \frac{\epsilon_x}{\sqrt{3}} - \frac{\epsilon_y}{\sqrt{3}} + \frac{\epsilon_z}{\sqrt{3}} \\ \frac{\epsilon_x}{\sqrt{3}} + \frac{\epsilon_y}{\sqrt{3}} - \frac{\epsilon_z}{\sqrt{3}} \end{bmatrix} \begin{bmatrix} 1 \\ 1 \\ 1 \\ \sqrt{3} \end{bmatrix}$$

For one degree rise in temperature (2.1) can be written as

$$C_{E_{111}} = \begin{bmatrix} \frac{C_{E_{10}} + \epsilon_{xy} + \epsilon_{yz}}{\sqrt{3}} & \frac{1}{\sqrt{3}} \\ \frac{\epsilon_{xy} + C_{E_{00}} + \epsilon_{yz}}{\sqrt{3}} & \frac{1}{\sqrt{3}} \\ \frac{\epsilon_{yz} + \epsilon_{xy} + C_{E_{00}}}{\sqrt{3}} & \frac{1}{\sqrt{3}} \end{bmatrix} \cdot$$

The values ϵ_{xy} , ϵ_{yz} , ϵ_{xz} can be found by solving the strain matrices in their respective planes. By solving (2.2), the value of CTE₁₁₁ was found equal to $2.79 \times 10^{-5} K^{-1}$. The experimental value ranges from 2.77×10^{-5} . The results show that a mathematical model based on above approach may be suitable for such estimations. Model can be improved by considering the other thermophysical phenomena such as Tg, etc.

CONCLUSION

In the present study, effect of fibre orientation on thermal expansion of glass/epoxy unidirectional laminated composite is studied. It was observed that Tg has a significant effect on CTE of composite. It is found experimentally, that CTE is minimum along the fibres in XY plane. Value of CTE, when taken out of plane, increases with angle and reaches maximum value along the Z-direction (out of plane). As a whole, value of thermal coefficient along Y-axis was the greatest among other directions in the composite part. More experiments at different orientations are required to propose a comprehensive mathematical model.

REFERENCES

[1] Reddy JN. Mechanics of laminated composite plates and shells: theory and analysis. 2nd ed: CRC Press, USA; 1945.

[2] Nawab Y, Legrand X, Koncar V. Study of changes in 3D-woven multilayer interlock fabric preforms while forming. Journal of The Textile Institute. 2012;103(12):1273-9.

[3] Parlevliet PP, Bersee HEN, Beukers A. Residual stresses in thermoplastic composites - a study of the literature. Part III: Effects of thermal residual stresses.

Composites Part A: Applied Science and Manufacturing. 2007;38(6):1581-96.

[4] Yoon KJ, Kim J-S. Effect of Thermal Deformation and Chemical Shrinkage on the Process Induced Distortion of Carbon/Epoxy Curved Laminates. Journal of Composite Materials. 2001;35(3):253-63.

[5] Yates B, McCalla BA, Sargent JP, Rogers KF, Phillips LN, Kingston-Lee DM. The thermal expansion of carbon fibre reinforced plastics. J Mater Sci. 1978;13(10):2217-25.

[6] Peeters LJB, Powell PC, Warnet L. Thermally-Induced Shapes of Unsymmetric Laminates. Journal of Composite Materials. 1996;30(5):603-26.

[7] Nawab Y, Jacquemin F, Casari P, Boyard N, Sobotka V. Shape evolution of carbon epoxy laminated composite during curing," Key Engineering Materials. Key Engineering Materials. 2012;504-506:1145-50.

[8] Gay D, Hoa SV, Tsai SW. Composite materials : Design and Applications: CRC Press; 2003.

[9] Schick C. Differential scanning calorimetry (DSC) of semicrystalline polymers. Analytical and Bioanalytical Chemistry. 2009;395(6):1589-611.