Assessment of the Thermoelastic Properties of an Injection molded short-fiber Composite: Experimental and Modelling

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ABSTRACT: The accurate prediction of both the elastic properties and the thermal expansion coefficients is very important for the precise simulation of such processes as injection molding of short-fiber polymer-matrix composites. In this work, a two-step homogenization procedure is applied and compared with experimental values obtained on a polyarylamide/glass fiber composite for a broad range of temperatures. It is observed that the stiffness averaging version of the model surpasses the compliance averaging variant, especially when it is combined with a precise evaluation of the fourth-order orientation tensor. It is also demonstrated that the orthotropic closure approximations are significantly better than previous ones (linear, quadratic, and hybrid) and than a very recent one. Among the orthotropic closure approximations, the fitted ones lead to acceptable results, which are very close to those obtained with the experimentally measured fourth-order orientation tensor

Key words: Short-fiber composites, Thermoelastic properties, Orientation tensor, Injection molding, Micromechanical modeling

1 INTRODUCTION

Short-fiber reinforced polymer-matrix composites constitute an important class of technical materials, because of their technological and economical interests. In particular, such composites with non-aligned reinforcements are of considerable importance because of their good thermoelastic properties. Even if both the matrix and the fibers are isotropic, the composite is usually anisotropic because of the non random orientations of the fibers: it is stiffer and stronger along the direction of dominant orientation, for instance. In the present paper, both the elastic and thermal properties of a polyarylamide/glass fiber composite, obtained by injection molding, are estimated and measured.

2 THEORY

Several theories ([1, 2] for instance) proceed in two steps to predict the overall thermoelastic properties of such materials: first, the properties of a unidirectional composite are estimated, and then an orientation averaging procedure is applied over all directions. For the first step, it has been shown by Tucker and Liang [3] that, among a set of available theories, the Mori and Tanaka model [4] gives satisfactory results when compared with finite element simulations. This is consistent with the previous comparisons with experimental results performed by Peyroux [5], for instance. In this paper, emphasis is put on the orientation averaging stage. Materials with isotropic distributions of fiber orientations [6,7] and/or planar isotropic distributions [8] have been considered much less often than unidirectional composites, and even fewer studies have focused on general orientation distributions [9].

The auxiliary unidirectional composite is generally transversely isotropic. Consequently, the orientation averaging (4) leads to:

$$C_{ijkl} = C_1 a_{ijkl} + C_2 (a_{ij}\delta_{kl} + a_{kl}\delta_{ij}) + C_3 (a_{ik}\delta_{jl} + a_{il}\delta_{jk} + a_{jl}\delta_{ik} + a_{jk}\delta_{il}) + C_4 \delta_{ij}\delta_{kl} + C_5 (\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) \quad (1)$$

where the coefficients *Ci* are linear combination of the unidirectional rigidity tensor coefficients. Alternatively, an averaging of the compliance tensor of the auxiliary unidirectional composite leads to:

$$S_{ijkl} = S_1 a_{ijkl} + S_2(a_{ij}\delta_{kl} + a_{kl}\delta_{ij}) + S_3(a_{ik}\delta_{jl} + a_{il}\delta_{jk} + a_{jl}\delta_{ik} + a_{jk}\delta_{il}) + S_4\delta_{ij}\delta_{kl} + S_5(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) \quad (2)$$

but, $\mathbf{S}^{UD}$ in spite of being the inverse of $\mathbf{C}^{UD}$, this effective compliance is not the inverse of the effective stiffness obtained above. This is a limitation of the present two-step approach. Many authors [10,11] prefer stiffness averaging to compliance averaging, because of a better agreement with experimental elastic constants, but without discussing the effect on thermal properties. In the present work, both the elastic and thermal properties predicted for the composite are considered, and they are compared with experimental results.

The second and the fourth-order orientation tensors are needed to apply most micromechanical models, but the fourth-order one is unavailable usually and, therefore, several closure approximations have been tested: the classical ones (linear, quadratic, hybrid), orthotropic ones [12,13] and a more recent one [14].

The thermal properties of a composite are closely related to its elasticity. This is especially true for a short-fiber composite containing two phases only, where the relation obtained by Levin [15] applies:

$$\begin{aligned} \boldsymbol{\alpha} &= (\mathbf{S^f} - \mathbf{S}):(\mathbf{S^f} - \mathbf{S^m})^{-1}:\boldsymbol{\alpha}^\mathbf{m} + (\mathbf{S^m} - \mathbf{S}):(\mathbf{S^m} - \mathbf{S^f})^{-1}:\boldsymbol{\alpha}^\mathbf{f} \\ \boldsymbol{\kappa} &= (\mathbf{C^f} - \mathbf{C}):(\mathbf{C^f} - \mathbf{C^m})^{-1}:\boldsymbol{\kappa}^\mathbf{m} + (\mathbf{C^m} - \mathbf{C}):(\mathbf{C^m} - \mathbf{C^f})^{-1}:\boldsymbol{\kappa}^\mathbf{f} \end{aligned} \quad (3)$$

with $\boldsymbol{\kappa} = \mathbf{C}:\boldsymbol{\alpha}$. It means that the thermal expansion can be evaluated directly from an estimation of the effective compliance or rigidity tensor.

3 EXPERIMENTAL

A short-fiber composite (IXEF 1002 supplied by Solvay) has been analyzed in this study. It was made of a polyarylamide (semi-crystalline aromatic polyamide) matrix containing 16.5 % (volume fraction) of glass fibers with an aspect ratio equal to 25. Both constituents were isotropic, with an influence of temperature on the Young modulus, Poisson's ratio and thermal expansion coefficient of the matrix as shown in Fig. 1, whereas these parameters were assumed to be temperature-independent in the fibers, and equal to 74 GPa, 0.25 and 5 10-6 C-1, respectively. The matrix Young modulus was measured by tensile and dynamic torsion tests; the expansion coefficient by dilatometry tests [16] and the Poisson's ratio was deduced from the compressibility modulus K measurement as $\nu = (3 - E/K)/6$.

60x60x1mm3 plates of this polyarylamide/glass fiber composite have been mold injected through a 0.8mm-thick end-fan gate to provide a parallel flow front. Orientation distributions and moduli have been measured in the center of the plates. Since the thickness of the plate is small, the fiber distribution does not change significantly through the thickness (for instance, a11 in the flow direction does not vary more than 15%) and no skin-core distribution appears. The Young moduli (at room temperature and at 120 C) have been measured with tensile tests, using a uniaxial extensometer; and the thermal expansion coefficients (for a whole range of temperatures) with dilatometry tests. Because the specimens were very thin, the Young moduli were measured along the injection flow (axis 1) and the transverse direction (axis 2) only, whereas the thermal expansion could be obtained also through the plate thickness (axis 3).

The fiber orientation tensors were measured accurately by analyzing a series of images obtained with a scanning electron microscope, following a procedure that reduced the possible artifacts by using inclined polished cuts (this will be reported in a separate paper). The components of the measured tensor deduced from the observation of a large number of fibers are:

$$a_{ij} = \begin{pmatrix} 0.793 & 0.016 & 0.053 \\ 0.016 & 0.179 & 0.006 \\ 0.053 & 0.006 & 0.028 \end{pmatrix} \quad (4)$$

where it can be observed that the distribution is dominantly in the injection plane (the a_{11} and a_{22} terms are large) but is not strictly planar, and that many fibers are parallel to the injection flow (a_{11} is the largest component). These a_{ij} values will be used in all the numerical applications that follow. The fourth-order orientation tensor also was deduced from the direct observation of the fibers. This experimental fourth-order orientation tensor allows computing the elastic and thermal properties without using any closure hypothesis, and this will be applied in the discussion below.

4 RESULTS

Firstly, the stiffness averaging version of the model surpasses the compliance averaging variant. Moreover, this procedure allows more flexibility through the role of the fourth-order orientation tensor [16]. The use of the various closure approximations for predicting the elastic properties was analyzed, but all the closure approximations give acceptable results except the one developed by Doghri and Tinel [14] which overestimates E_{11}.

The stiffness averaging version of the model also surpasses the compliance averaging variant for predicting the expansion coefficients, especially if the fourth-order experimental orientation tensor is used. The linear, quadratic, and hybrid closure approximations lead to unacceptable results by predicting negative expansion coefficients when the contrast of mechanical properties between the two phases is high, that is at high temperature. For this reason, Doghri and Tinel [14] have developed a new closure approximation. It leads to acceptable prediction for α_{11}, but gives very close values for α_{22} and α_{33} , which is not suitable (Fig. 2). Then α_{22} is too high at high temperature.

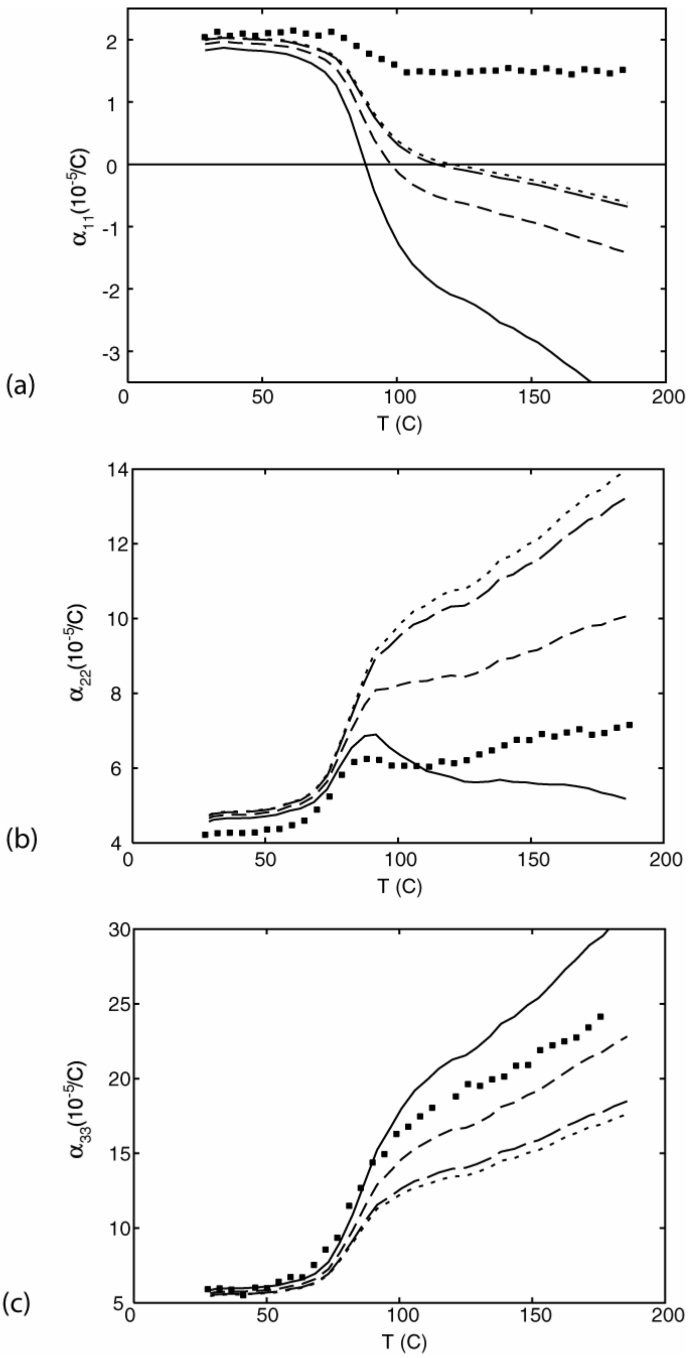

Fig. 1. Thermal expansion coefficients along the three axes of the composite: experimental values (symbols) and predictions given by the stiffness averaging procedure using the linear (unbroken line), quadratic (dotted line), and hybrid (type A: long dashes, type B: short dashes) closure approximations.

The orthotropic closure approximations lead to acceptable results. Their combination with a simple stiffness averaging procedure leads to good predictions for elastic moduli and thermal expansion (Fig. 2).

Among the studied closure approximations, the fitted Cintra and Tucker closure [12] is the best, since it performs as well as the experimental fourth-order orientation tensor. Nevertheless, some discrepancies can be observed for α_{22} and α_{33}.

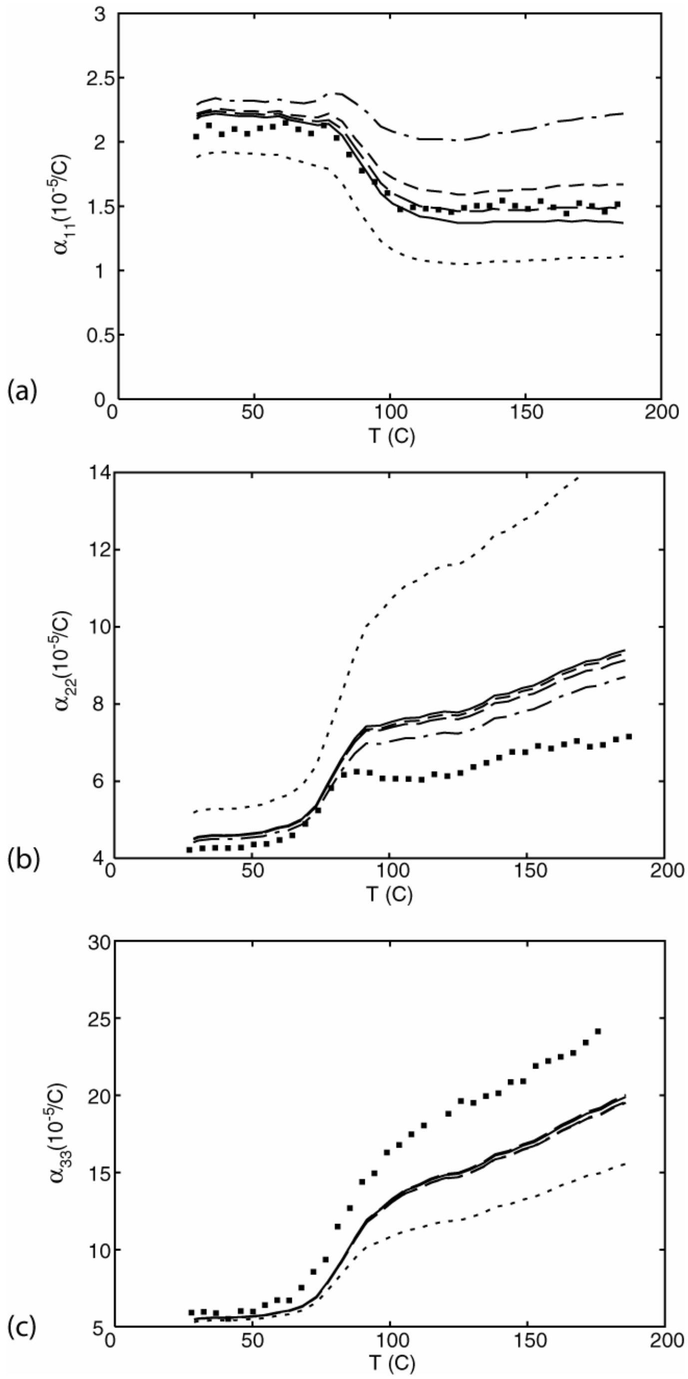

Fig. 2. Same as Figure 1, but the experimental values (symbols) are now compared to the predictions given by the stiffness averaging procedure using either the experimental fourth-order orientation tensor (unbroken line), or the closure approximations proposed by Cintra and Tucker (smooth closure: mixed dashes, fitted closure: long dashes), Chung and Kwon (short dashes), Doghri and Tinel (dotted line).

This may be due to the complex flow-induced crystalline microstructure of the polymer matrix, which cannot be considered as fully isotropic. The dilatometric behavior of the pure injection molded matrix is transversely isotropic, with a larger expansion coefficient along the thickness direction due to some added small lamellar particles to enhance nucleation. We can note that the use of the stiffness averaging procedure and the experimental fourth-order tensor gives very close results to orthotropic closure approximations ones.

5 CONCLUSIONS

The comparison with experimental values of both the elastic and thermal properties of a short-fiber composite provides a selective procedure to test predictive models. The two-step homogenization procedure that applies orientation averaging to an auxiliary unidirectional composite with the same fiber content as the misoriented composite is able to predict the elastic and thermal properties of a short-fiber composite accurately, but a good closure approximation should be used. Orthotropic closure approximations give the best results.

ACKNOWLEDGEMENTS

The authors acknowledge LEGRAND, MOLDFLOW and SOLVAY companies for their financial support. They wish to thank H. Alglave, J. Correa, D. Delaunay, R. Fulchiron, S. Karpp-Pfordt, P. Kennedy, M. Laplanche, R. Le-Goff, V. Leo, J.M. Rossignol, R. Zheng for the enriching discussions during all the FISH (FIber filled polymers: injection and SHrinkage) project.

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