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Shape evolution of carbon epoxy laminated composite during curing

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Abstract. The laminated composites undergo chemical deformation on cross-linking and thermal deformation while cooling, during the fabrication process. In thin laminates, due to large displacements and complex shape evolution, these deformations can only be explained by using a nonlinear strain-displacement relationship. In the present article, the thermal and chemical deformations occurring in carbon/epoxy laminates are calculated together for the first time by considering a non-linear geometrical approach, in order to understand the evolution of shape and hence residual stresses induced during the fabrication process.

Introduction

Thermoset laminated composite undergoes residual strains and stresses during the curing cycle due to mismatched coefficients of thermal expansion and chemical shrinkage of the constituents. These residual deformations and stresses depend mostly on the shrinkage behaviour of the polymer matrix as fibres have no chemical and very small thermal shrinkage. Matrix chemical shrinkage is a direct consequence of cross-linking of molecules while curing. Resulting residual stresses can lead to dimensional instability, warpage, ply cracking, delamination and fibre buckling. Hyer [1] was the pioneer among many authors [1-4] to quantify and calculate the thermal deformations in the laminated composites. Not many of the authors estimated or measured the residual stresses/deformation in the laminated composite caused by chemical shrinkage as they assumed that part is in stress free state until the complete cure. Bogetti and Gillespie [5] showed that during a curing cycle, the composite undergoes chemical residual stress in addition to thermal stress. Modelling of shape of the part, and total residual stress σ as a function of temperature T and degree of cure α can be done by taking into account the chemical shrinkage, as well as thermal expansion/contraction by using an advanced form of Hooke's law (1).

$$\sigma = C(T, \alpha) \left(\varepsilon - CTE(T, \alpha) \Delta T - CS(T, \alpha) \Delta \alpha \right) \quad (1)$$

Where ε is the residual strain which is calculated from the displacements. The rigidity matrix C , coefficient of thermal expansion (CTE), and coefficient of chemical shrinkage (CS) depend linearly or not on the degree of cure (α) and temperature (T). The coefficient of chemical shrinkage CS can be written mathematically as $CS = \frac{\Delta e_{\text{shrinkage}}}{e_i \Delta \alpha}$, where e_i is the initial thickness. Δe is the change in

thickness for $\Delta \alpha$ increment of the degree of cure. By using the above formulation, Abou Msallem [6] simulated the residual stresses and strains by assuming linear displacement-strain relationship.

Since in thin laminates, the displacements are very large with respect to thickness and evolution of the shape is complex, the linear strain-displacement relationship is not able to predict correctly the strain and hence the final shape of the laminated piece [7]. Therefore, a nonlinear relationship is required for this purpose.

In the present article, chemical deformations induced in the laminates are calculated for the first time by using nonlinear strain-displacement relationship. This calculation is coupled with a self-consistent model, which provides the thermo-physical properties of laminate, evolving with the cure of resin. Thermal deformation is calculated as well, and then the possible evolution of the shape of a composite part during the curing cycle is presented.

Calculation of Strains

Two approaches, i.e. linear approach or classical laminate theory, and non-linear one are found in literature for the calculation of strains in the laminated plate. Linear approach based on a linear strain-displacement, is not able to predict correctly the deformation [7] in a laminated piece. For example, room temperature shape of [0/90] non-symmetric laminates predicted by linear theory is a 'saddle' like, but in reality, it is cylindrical. This difference was explained by assuming a non-linear strains-displacement relationship [8].

The equations of strains are simplified by many authors [2, 4, 7]. Gigliotti [7] solved the displacement equations by using polynomial like Timoshenko's polynomial. It is one of the simplest ways that involves only five unknowns, but this method gives only the good results for [0/90] laminates. Dano and Hyer [4] used richer equations having 28 unknown coefficients, to solve the equations of strains for angle ply as well as cross- ply laminates.

To calculate the deformation curvatures and stability of the deformed shape, a minimization of energy approach is used. The total potential energy (E_{tot}) of laminated plate is given as follows:

$$E_{tot} = \frac{1}{2} \sigma_{ij} \epsilon_{ij} \quad (2)$$

Here σ_{ij} and ϵ_{ij} are the stress and strain matrices respectively. By Lagrange- Dirichlet theorem, if total potential energy (E_{tot}) is differentiated and value of first derivative is zero, then object will be in a state of equilibrium. In addition, this equilibrium will be stable if second derivative of total energy is positive. Evaluation of total energy at any instant by using this minimization of energy approach gives as by-product the curvatures of laminated plate subjected to chemical shrinkage and thermal expansion/contraction. The bending curvatures are denoted by 'a' and 'b' whereas 'c' denotes to twist curvature.

Material and Methods

Carbon/epoxy laminated composite piece [0₂/90₂] having dimensions 190×190×1 (mm³) and 57% fibre volume fraction is considered for this study. Following classical thermal cycle is used for curing of the part:

1. Heating from room temperature (20°C) to curing temperature 180°C at rate of 3°C/min
2. Maintaining at 180°C for 120 min.
3. Cooling to room temperature at 3°C/min.

Deformations during curing cycle. Fig.1. shows the graph of temperature and the corresponding degree of cure, during the curing cycle. The degree of cure is calculated by using a kinetics law [9]. The gel point α_{gel} of the studied epoxy resin was found equal to 0.52+/- 0.01[6].

In the first step of the thermal cycle, the resin undergoes chemical shrinkage and thermal expansion, but as the gel point is not reached yet so material is not so stiff to undergo chemical deformation. During the second step, the resin undergoes cross-linking resulting into the chemical shrinkage of matrix and development of mechanical properties. Since provided temperature is

constant and due to small thickness of the part, exothermy of cross-linking does not impact the temperature, the temperature remains constant and the chemical shrinkage is the only prominent phenomenon in this step, which causes the chemical deformation once the material passes the gel point. The chemical deformation zone is highlighted in the fig. 1. In the third step, the cured part is cooled from curing temperature to room temperature resulting into a thermal deformation. The thermal deformation zone can also be observed in the Fig.1.

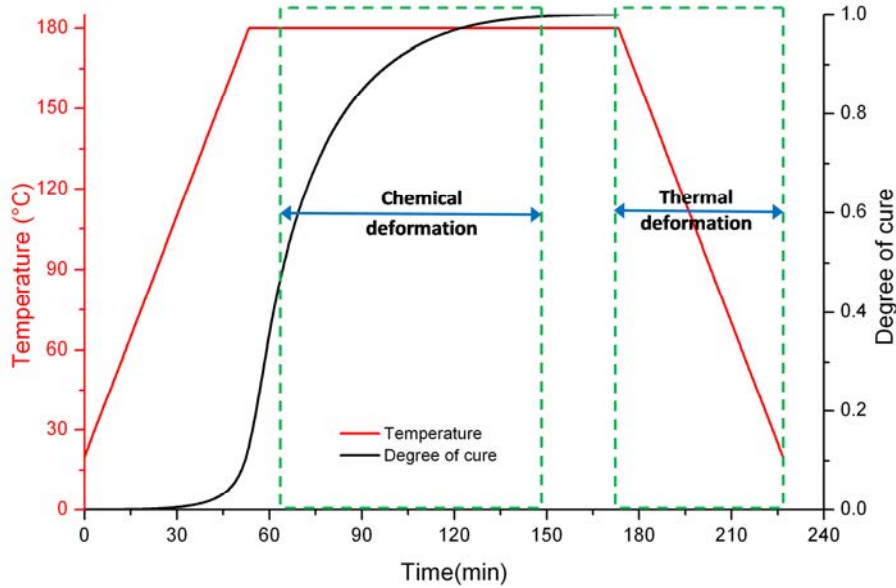


Fig. 1. Degree of cure and temperature variation during the thermal cycle

Self-consistent model for estimation of effective properties. The deformation induced in a composite piece depends on the mechanical properties of constituents, and the mismatched coefficients of thermal expansion and chemical shrinkage. Since the properties of composite are developing during curing, the calculation of chemical deformations is a little tricky.

Self-consistent models based on the mathematical formalism proposed by Kroner [10] and Eshelby [11] constitute a reliable method to predict the micromechanical behaviour of heterogeneous materials, i.e. composites. In the present study, an advance form of this model [6] is used to calculate the effective properties of composite i.e. coefficients of thermal expansion (CTE_1, CTE_2), coefficients of chemical shrinkage (CS_1, CS_2), Young's modulus (E_1, E_2) and shear modulus (G_{12}) during and after curing by using the properties of fibres (Table 1) and matrix.

Table 1. Mechanical and thermo-mechanical properties of fibres

Property	Value	Property	Value
E_1 [GPa]	230	G_{12} [GPa]	27.6
E_2, E_3 [GPa]	20.7	CTE_1 [K^{-1}]	-0.38×10^{-5}
ν_{12}, ν_{13}	0.2	CTE_2 [K^{-1}]	20×10^{-6}
G_{23} [GPa]	6.89	CS_1, CS_2	0

For the cured resin : Young modulus $E = 1.37$ GPa, Poisson's ratio $\nu^m = 0.35$, coefficient of thermal expansion $CTE^m = 44.9 \times 10^{-6} K^{-1}$, and coefficient of chemical shrinkage $CS^m = 0.019$ [9]. The effective estimated properties of cured composite are presented in Table 2.

Table 2. Effective properties of cured carbon/epoxy ply (57 vol% fibre).

CTE_1 [K^{-1}]	CTE_2 [K^{-1}]	CS_1 (%)	CS_2 (%)	G_{12} [GPa]	E_1 [GPa]	E_2 [GPa]	ν_{12}
0.21×10^{-6}	29.8×10^{-6}	-1.17×10^{-2}	-0.67	6.40	131.2	5.63	0.25

Determination of Thermal Deformation

Thermal curvatures induced in a [0/90] carbon/epoxy laminated plate, on cooling from curing temperature 180°C to room temperature, are calculated by a minimization of energy approach. Since the piece is thinner ($e = 1\text{mm}$) so it was assumed that there are no temperature gradients. The composite was supposed to be fully cured before cooling, so $\Delta\alpha$ was equal to zero in this step.

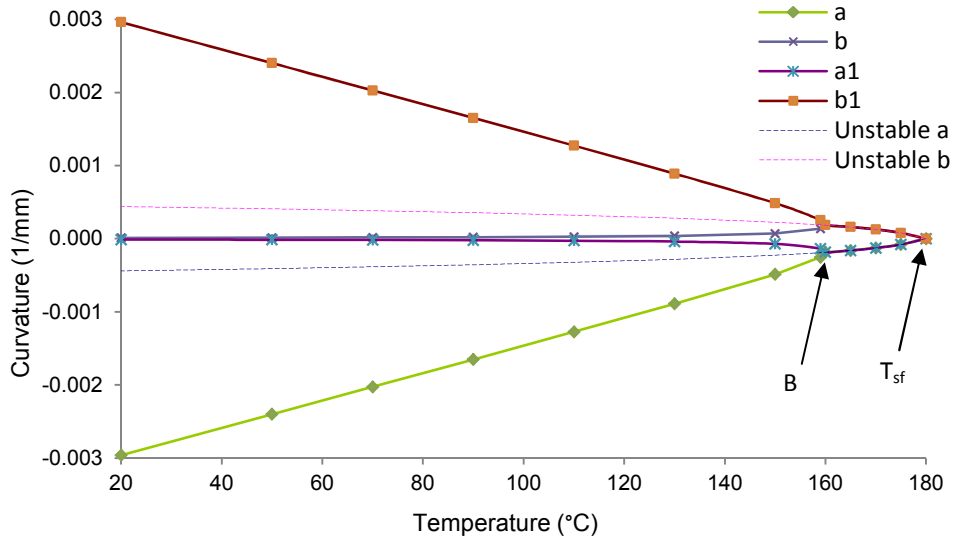


Fig. 2. Thermal curvatures of [0/90] carbon/epoxy laminate

If it is assumed that the chemical deformation is zero before the start of the thermal deformation zone, then the curing temperature 180°C will be the thermal stress free temperature (T_{sf}), and the thermal deformation at this point will be zero. Gigliotti's equations, being simple equations, are used to calculate the strain fields during cooling. Fig. 2 shows the calculated thermal bending curvatures (a and b). On solving the energy equation (2), a saddle shape ($a = -b$) is observed in the beginning. This pair of curves splits into three possible pairs of branches from the bifurcation point (B): a saddle shape (dashed curves), and two cylinders ($a > 0, b = 0$ or vice versa) with axis along x or y -axis. On verifying the three solutions by calculating the energy derivatives, it founds that saddle shape solution is not stable energetically after the bifurcation point. So experimentally, only the cylindrical solutions are possible. Gigliotti [7] presented the similar results on a carbon/epoxy laminated part. The twist curvature ' c ' in [0/90] laminates is zero, but in angle-ply laminates, it has a certain value in addition to bending curvatures. Curvatures of the angle-ply laminates were calculated by using relatively richer Hyer's strain equations [4]. These results are not presented here due to shortage of space.

Determination of Chemical Deformations

As discussed earlier, the chemical shrinkage is an important factor, which has not been taken into account yet, for the calculation of residual strains and stresses. In this section, calculation of the evolution of composite properties as a function of degree of cure, and for a given fibre volume fraction is estimated by using self-consistent model, and then chemical curvatures are calculated by using these properties.

The properties of fibres, which were used for the estimation of composite properties, are given in the Table 1. Since there is no chemical reaction in fibres during curing cycle, their properties are independent of cross-linking, but properties of the resin are dependent on cure kinetics. Young's modulus of the resin is not significant upto $\alpha = 0.8$, but from this point it increases sharply to reach a value of 1.37 GPa in the fully cured state [6]. The Poisson's ratio ν^m of the resin is 0.35 [6].

The cure shrinkage of the resin was determined experimentally and was found varying linearly with the degree of cure. The slope of cure shrinkage versus the degree of cure graph gave the coefficient of chemical shrinkage (CS^m) of the resin, which was found equal to -0.019 [9]. By using the properties of fibres and resin, the effective properties of composite during cross-linking were calculated for 57% volume fraction of the fibres.

Once the chemical and mechanical properties of composite part during the curing were estimated, the energy equation was solved to get the curvatures due to chemical shrinkage. The chemical curing region of the thermal cycle can be divided into two steps:

1. Heating step: temperature increases from room temperature to 180°C but no thermal or chemical deformations are expected as properties of composite plies, i.e. Young's modulus, coefficients of chemical shrinkage (CS_1 and CS_2) are negligible.
2. Isothermal step: the temperature is constant, so there is no thermal deformation, but due to the chemical shrinkage of resin, a chemical deformation is expected to appear.

Fig. 3 shows the estimated chemical curvatures while curing of [0/90] laminate with 57% fibre volume fraction. As the properties of resin are negligible until the gel point ($\alpha_{gel}=0.52$), no chemical curvatures are observed until this point.

When the curvatures start to appear, their magnitudes are not significant. Starting from α_{gel} to $\alpha=0.8$, two bending curvatures (a, b) with equal but opposite magnitude are observed. This means that piece will deform in the saddle like shape, but at $\alpha=0.875$ these curves separated into three pairs. So $\alpha=0.875$ is the bifurcation point for this specific composite. From this point, there are three possible solutions. On verifying the stability of solutions by a minimization of energy approach, the central pair (dashed curves) is found unstable. Therefore, in reality, a cylinder along x or y-axis can be observed. Curvatures of these cylindrical shapes increase with increasing the degree of cure until the fully cured state. As explained earlier, the mechanical properties of resin are negligible until $\alpha=0.8$ and it is not able to cause shrinkage in the ply. However, from this point, the properties evolve sharply and resin can induce shrinkage in ply. That is why, most of the deformation is observed after this point. The twist curvature 'c' is found zero.

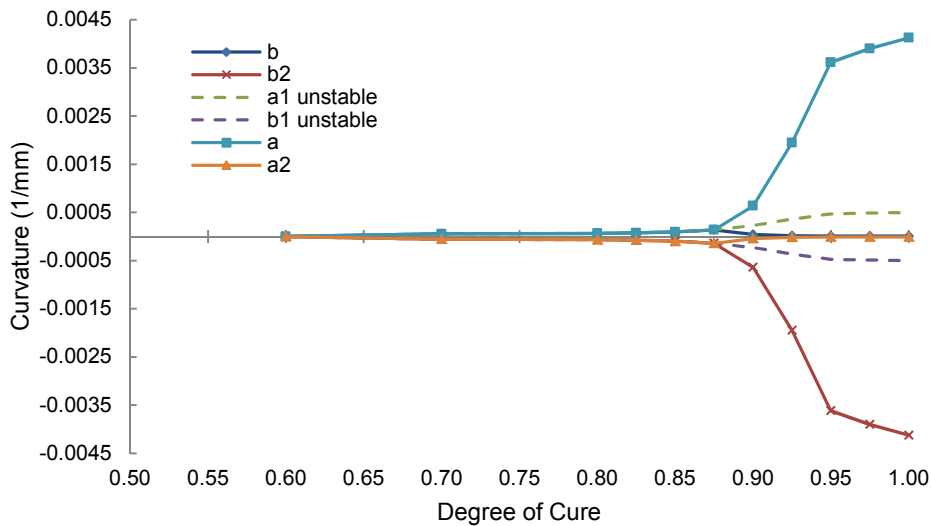


Fig. 3. Chemical curvatures of composite [0/90] as function of degree of cure

Conclusion

In this study, thermal and chemical deformations in a [0/90] carbon/epoxy laminated composite are predicted during the fabrication cycle, considering a non-linear displacement-strain relationship. Effective properties of composite, required for these calculations, were estimated from the properties of matrix and fibres by using a self-consistent model. It is observed that chemical

deformations begin to appear from the gel point. These deformations are so significant that they cannot be neglected. This study is an effort to understand the curing process. A work is in progress to establish an experimental setup for the experimental quantification of these deformations.

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