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Residual stresses created during curing of a polymer matrix composite using a viscoelastic model

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ABSTRACT

A modified viscoelastic constitutive model is proposed to predict cure-induced residual stresses in polymer matrix composites. The modifications rely on using the inverse of the Deborah number to describe regimes corresponding to a fully relaxed state, a viscoelastic state and an elastic state. The composite is only in a viscoelastic state for limited ranges of the Deborah number. By considering the evolution of the Deborah number during curing of the AS4/3501-6 composite, the composite is in a fully relaxed state when it is cured at high temperature and the degree of cure is lower than 0.73, and no further changes in the viscoelastic characteristics when the degree of cure is higher than 0.8. A 3D simulation of a composite laminate plate is used to predict the evolution of the residual stresses. The analysis reveals that the accurate simulation on the cure-induced residual stress should include the last two states of the entire cure process, and consider the stress relaxation, thermal deformation and chemical shrinkage.

1. Introduction

The creation of residual stress during curing of polymer matrix composites (PMCs) arises because of their inherent anisotropy and inhomogeneity. Their presence can lead to potentially lowering the performance of composite structures, and consequently cure-induced residual stresses should be considered in the design of composite structures. The earliest analyses of residual stresses in PMCs were based on the assumption that PMCs are in the stress-free state at the cure temperature. Therefore residual stresses only develop during the cooling process and can be predicted using elastic models [1,2]. These traditional elastic models omit the effects of chemical shrinkage, the development of residual stress before cool-down and stress relaxation during the cooling process. These factors are known to have significant effects on the final residual stresses in PMCs [3,4]. To capture the influence of these factors, a variety of revised cure-dependent elastic models have been proposed [4–7], such as a path dependent constitutive model [4,5] and the cure-hardening, instantaneous linear elastic (CHILE) model [7]. However, these models, do not consider stress relaxation during the cooling process.

Cure-dependent viscoelastic models have also been developed to predict the development of residual stresses in PMCs. These models recognize that the polymer matrix of PMCs changes from a liquid-like material in the early stages of cure to a viscoelastic solid at the end of curing [8,9]. Some of the earliest work on the residual stresses in composite materials using a viscoelastic approach was performed by Weitsman [10], although focusing only on the cooling process. White and Hahn [11,12] developed a process-dependent viscoelastic model to investigate the formation of residual stress in composite laminates during the entire cure cycle. They validated their model through the intermittent cure of non-symmetric cross-ply laminates and measuring process induced residual curvatures. White and Kim [3] introduced a thermo-viscoelastic constitutive equation that depended on the degree of cure and temperature. They then investigated the formation of residual stresses in Hercules AS4/3501-6 composite during an entire cure cycle using a 2D finite element model. Similarly, Lee and Kang [13] performed a thermo-viscoelastic analysis to determine residual stresses in a laminated shell during curing.

More recently, thermo-viscoelastic analyses have been used to explore the development of residual stresses in complex shaped

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composite structures [14,15]. However, thermo-viscoelastic properties for PMCs during curing are difficult to be accurately characterized and modeled. Furthermore, the utilization of anisotropic thermo-viscoelastic models leads to long calculation times and requires large amounts of memory to permit storage of the internal state variables [4]. It is therefore necessary to modify the constitutive models or to develop more efficient numerical methods to minimize the computational resources required. Clifford et al., [16] proposed hybrid finite element models containing both shell and solid elements to reduce the required computational resources.

The approach adopted in the present work was to consider whether a cure-dependent viscoelastic model could be modified and to explore methods to achieve this. The model is first introduced in section 2, and based on an analysis of the model, a parameter is proposed to represent the viscoelasticity of PMCs, and a modified viscoelastic model is proposed. In section 3 the development of viscoelasticity of the Hercules AS4/3501-6 composite during the typical cure cycle is analyzed using data given by White and Kim [3]. In section 4 the application of the modified model is performed and the analysis then applied in section 5 to curing of the Hercules AS4/3501-6 composite. The modified viscoelastic model is compared with predictions using the original viscoelastic model and a cure-dependent elastic model. The effects of stress relaxation, chemical shrinkage and thermal deformation on the development of residual stresses in a $[0/90]_s$ AS4/3501-6 laminate during the entire cure cycle are also analyzed.

2. Viscoelastic constitutive equations

The most general viscoelastic constitutive equation for relaxation of residual stresses generated in composites during curing can be expressed using a hereditary integral, where the stress $\sigma_i(t)$ and mechanical strain $\varepsilon_j(t)$ at any time are given by Ref. [13].

$$\sigma_i(t) = \int_{-\infty}^{t} C_{ij}(\alpha, T, t-\tau)\frac{\partial \varepsilon_j(\tau)}{\partial \tau}d\tau \tag{1}$$

$$\varepsilon_j(t) = \tilde{\varepsilon}_j(t) - \beta_j \Delta T - \eta_j \Delta\alpha \tag{2}$$

where α, T and t are the degree of cure, temperature and the current time respectively, C_{ij} is the stiffness matrix, which is dependent on the degree of cure, temperature and time, and the indices $i, j = 1$–6. $\tilde{\varepsilon}_j$ is the total strain, β_j and η_j are the thermal expansion coefficients (CTE) and chemical shrinkage coefficients (CCE), respectively, ΔT and $\Delta\alpha$ are the changes in temperature and degree of cure, and τ is a dummy variable.

If the material is treated as being thermo-rheologically simple at a constant degree of cure α, and there is no strain history prior to $t = 0$, according to the time–temperature superposition principle, Eq. (1) can be rewritten as [13].

$$\sigma_i(t) = \int_0^{\xi_t} C_{ij}(\alpha, T_r,\ \xi_t - \xi')\frac{\partial \varepsilon_j(\xi')}{\partial \xi'}d\xi' \tag{3}$$

where T_r is the reference temperature. ξ_t and ξ' are reduced times, given by:

$$\xi_t = \xi(t) = \int_0^t \frac{1}{a_T(\alpha, T)}ds \ \text{ and } \ \xi' = \xi(\tau) = \int_0^\tau \frac{1}{a_T(\alpha, T)}ds \tag{4}$$

where a_T is a shift factor. For the composite during curing, a_T is functionally dependent on both α and T [13].

In Eq. (3), $C_{ij}(\alpha, T_r, \xi_t - \xi')$ is the stiffness matrix at the reference temperature, and noted as $C_{ij}(\alpha, \xi)$. It can be approximated by the Prony series as [3]:

$$C_{ij}(\alpha, \xi) = C_{ij_\infty} + \sum_{m=1}^{M} C_{ij_m} \exp\left(\frac{-\xi}{\rho_m(\alpha)}\right) \tag{5}$$

where C_{ij_∞} are the fully relaxed stiffnesses, C_{ij_m} are the cure-dependent discrete stiffnesses of elements associated with the $\rho_m(\alpha)$ discrete relaxation times. Substituting Eq. (5) into Eq. (3), the viscoelastic constitutive equation becomes

$$\sigma_i(t) = C_{ij_\infty}\varepsilon_j(t) + \sum_{m=1}^{M} C_{ij_m}\int_0^{\xi_t} \exp\left(-\frac{\xi_t - \xi'}{\rho_m(\alpha)}\right)\frac{\partial \varepsilon_j(\xi')}{\partial \xi'}d\xi' \tag{6}$$

In order to solve the integral in Eq. (6), a recursive algorithm [17] is applied. The integral in Eq. (6) is referred to as $q_{jm}(t)$, and assuming a sufficiently small time increment Δt, the integral $q_{jm}(t)$ can be rewritten as

$$\begin{aligned} q_{jm}(t) = &\int_0^{\xi_{t-\Delta t}} \exp\left(-\frac{\xi_t - \xi'}{\rho_m(\alpha)}\right)\frac{\partial \varepsilon_j(\xi')}{\partial \xi'}d\xi' \\ &+ \int_{\xi_{t-\Delta t}}^{\xi_t} \exp\left(-\frac{\xi_t - \xi'}{\rho_m(\alpha)}\right)\frac{\partial \varepsilon_j(\xi')}{\partial \xi'}d\xi' \end{aligned} \tag{7}$$

where $\xi_t = \xi_{t-\Delta t} + \Delta\xi_t$ and $\Delta\xi_t = \int_{t-\Delta t}^{t}\frac{1}{a_T(\alpha,T)}ds$.

If it is assumed that the degree of cure and temperature are constant during the small time step Δt, and $\varepsilon_j(\xi')$ is a linear function of the reduced time ξ'. Then the two integrals in Eq. (7) can be solved, and a recursive formulation for $q_{jm}(t)$ is given by

$$q_{jm}(t) = A_{1m}(t)q_{jm}(t-\Delta t) + A_{2m}(t)\Delta\varepsilon_j^t \tag{8}$$

where $A_{1m}(t) = \exp\left(-\frac{\Delta\xi_t}{\rho_m(\alpha_t)}\right)$ and $A_{2m}(t) = \frac{1}{\Delta\xi_t/\rho_m(\alpha_t)}\left(1 - \exp\left(-\frac{\Delta\xi_t}{\rho_m(\alpha_t)}\right)\right)$, α_t is the degree of cure at the current time t, $\Delta\varepsilon_j^t$ is the strain increment in time step Δt.

Substituting Eq. (8) into Eq. (6), the current stress is finally expressed as

$$\sigma_i(t) = C_{ij_\infty}\varepsilon_j(t) + \sum_{m=1}^{M} C_{ij_m}q_{jm}(t) \tag{9}$$

In Eq. (9), the first part represent the contribution of the fully relaxed stiffnesses, and the second part represent the contribution of the discrete stiffnesses to the residual stress. The relaxation behavior of the residual stress contributed by the discrete stiffnesses is controlled by the parameters $A_{1m}(t)$ and $A_{2m}(t)$. It is noticeable that both $A_{1m}(t)$ and $A_{2m}(t)$ are determined by the parameter $\Delta\xi_t/\rho_m(\alpha_t)$. Here we introduce the notation

$$1/De_m = \Delta\xi_t/\rho_m(\alpha_t) \tag{10}$$

These non-dimensional parameters are referred to as the inverse of the Deborah numbers [18], widely used in rheology. If the Deborah number is large the material behaves as a solid, when it is small the material is effectively a fluid.

Both the relaxation parameters$A_{1m}(t)$ and $A_{2m}(t)$ are shown as functions of the inverse of the Deborah numbers in Fig. 1. It can be