

Curing Induced Residual Stresses in Laminated Cylindrical Shells

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A viscoelastic finite element analysis is presented to investigate residual stresses occurred in a laminated cylindrical shell during cure. An incremental viscoelastic constitutive equation that can describe stress relaxation during the cure is derived as a recursive formula which can be used conveniently for a numerical analysis. The finite element analysis program is developed on the basis of a 3-D degenerated shell element and the first order shear deformation theory, and is verified by comparing with an one dimensional exact solution. Viscoelastic effect on the residual stresses in the laminated shell during the cure is investigated by performing both the viscoelastic and linear elastic analyses considering thermal deformation and chemical shrinkage simultaneously. The results show that there is big difference between viscoelastic stresses and linear elastic stresses. The effect of cooling rates and cooling paths on the residual stresses is also examined.

Key Words : Viscoelastic Analysis, Residual Stress, Laminated Cylindrical Shell, Cure, Degree of Cure, Finite Element Analysis

1. Introduction

A residual stress occurred in fiber-reinforced thermosetting composite materials during cure is one of severe factors that deteriorate the performance of a composite structure. The residual stress can bring about matrix cracking in composites even before external loads are applied and thus can reduce the stiffness of the composites. In addition, since the residual stress means a pre-loading, it can also cause the degradation of strength. Therefore, the residual stresses induced during cure should be considered in the design of composite structures because they generally have a bad effect on the stiffness and strength of the composite structure.

During the past decade, a number of viscoelastic analyses have been developed based on

Schapery's viscoelasticity model (1967) to predict viscoelastic behavior of composite structures. Tuttle and Brinson (1986) developed a numerical procedure for predicting nonlinear viscoelastic response of laminated composites based on classical lamination theory. Henriksen (1984) developed a two dimensional finite element analysis for nonlinear viscoelastic behavior of an isotropic material. Roy and Reddy (1988) presented a similar analysis which includes large displacement and moisture diffusion. Lin and Hwang (1989) developed a two dimensional finite element analysis for the linear viscoelastic response of anisotropic materials. Lin and Yi (1991) presented a similar analysis for generalized plane strain conditions. Kennedy and Wang (1994) presented a three dimensional finite element analysis which treats the nonlinear viscoelastic response of laminated composites.

The first analysis on residual stresses in a thermosetting matrix composite during the cure was performed by Hahn and Pagano (1975). They calculated linear elastic residual stresses under the assumption that a laminate was in a stress-free state at cure temperature immediately before cool-down. Bogetti and Gillespie (1992) analyzed the

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cure-induced residual stresses for thick laminates using an incremental elastic laminated plate theory and investigated the effect of chemical shrinkage on the development of residual stresses. Hodges et al. (1989) presented an experimental approach to find optimal curing conditions for reducing the residual stresses of carbon fiber/epoxy composite. White and Hahn (1991) developed a process model for the investigation of viscoelastic residual stress development in laminates during cure processing and validated the model by the intermittent cure of unsymmetric cross-ply laminates in which curing induced residual curvatures were measured. Kim (1996) introduced a viscoelastic constitutive equation depending on degree of cure and temperature by performing stress relaxation tests and investigated the residual stresses of Hercules AS4/3501-6 composite during the cure by analyzing two dimensional and axisymmetric problems.

The objective of this research is to investigate the residual stresses occurring in a laminated cylindrical shell during the cure by deriving an incremental viscoelastic constitutive equation and by performing the finite element analysis. The viscoelastic data of Hercules AS4/3501-6 provided by Kim (1996) is used in calculations. The viscoelastic finite element program will be developed on the basis of a 3-D degenerated shell element and the first order shear deformation theory, and is validated by comparing with an one dimensional exact solution. The effect of temperature, degree of cure and chemical shrinkage on the viscoelastic residual stresses induced in the laminated shell during the cure is studied. The effect of cooling rates and cooling paths on the residual stresses is also examined.

2. Viscoelastic Constitutive Equation

A linear viscoelastic constitutive equation to predict the relaxation of residual stresses induced in composite shells during cure can be expressed by the following hereditary integral (Flaggs and Crossman, 1981 ; Lin and Hwang, 1989) :

$$\sigma_i = \int_0^t C_{ij}(\alpha, T, t-\tau) \frac{\partial \bar{\epsilon}_j}{\partial \tau} d\tau, \quad (1)$$

(i, j=1,2, ..., 6)

$$\bar{\epsilon}_j = \epsilon_j - \beta_j \Delta T - \eta_j \Delta \alpha \quad (2)$$

where σ_i and ϵ_j represent stresses and strains, and C_{ij} , β_j , and η_j represent relaxation moduli, coefficients of thermal expansion, and coefficients of chemical shrinkage, respectively. ΔT and $\Delta \alpha$ indicate the changes in temperature and degree of cure. t denotes time and τ is a dummy variable for integration. Since thermoset composite materials, which are softened at the beginning of cure, become to be hardened as cure process goes on, mechanical properties of the thermoset composite materials are generally changed during manufacturing process. Lee and Springer (1990), and White and Hahn (1992) showed that they depended on the degree of cure. Hence, if the degree of cure changes, the composite material will show thermo-rheologically complex behavior which eventually makes it very difficult to solve a problem. But, if the degree of cure, denoted by α_0 , is maintained to be constant, the material can be assumed to be thermo-rheologically simple. By using a temperature-dependent reduced time, this postulate makes it possible for Eq. (1) to be expressed in the following form :

$$\sigma_i = \int_0^t C_{ij}(\alpha_0, T_0, \xi^t - \xi^\tau) \frac{\partial \bar{\epsilon}_j^\tau}{\partial \tau} d\tau \quad (3)$$

where T_0 indicates the reference temperature, and ξ represent the reduced time defined as

$$\xi^t = \int_0^t \frac{ds}{a_\tau(\alpha_0, T(s))}, \quad \xi^\tau = \int_0^\tau \frac{ds}{a_\tau(\alpha_0, T(s))} \quad (4)$$

where a_τ represents a shift factor expressed as a function of temperature at the constant degree of cure α_0 . Because generalized Maxwell models consisting of negative exponential functions are known as good approximations for the response of viscoelastic materials, the time-dependent relaxation moduli C_{ij} in Eq. (3) can be expressed in a finite exponential series of the form :

$$C_{ij}(\xi) = C_{ij}^\infty + \bar{C}_{ij} \sum_{m=1}^N W_m \exp\left(-\frac{\xi}{\tau_m}\right) \quad (5)$$

where C_{ij}^∞ represent fully relaxed moduli and $\bar{C}_{ij} = C_{ij}^0 - C_{ij}^\infty$ where C_{ij}^0 represent unrelaxed moduli. τ_m and W_m are relaxation times and weighting factors at the given degree of cure. All parameters

described above are determined from a viscoelastic experiment. Substituting Eq. (5) into Eq. (3), the viscoelastic constitutive equation is written as

$$\sigma_i^t = \int_0^t \left[C_{ij}^\infty + \bar{C}_{ij} \sum_{m=1}^N W_m \exp\left(-\frac{\xi^t - \xi^\tau}{\tau_m}\right) \right] \frac{\partial \bar{\epsilon}_j^\tau}{\partial \tau} d\tau \quad (6)$$

We will now proceed to simplify the hereditary integral appearing in the constitutive equation in the manner proposed by Henriksen(1984) and used by Kennedy and Wang(1994) and derive a recursive formula in which the solution at current time $t + \Delta t$ can be obtained using the solution known at the previous time t . The constitutive equation at current time $t + \Delta t$ can be written as follows.

$$\begin{aligned} \sigma_i^{t+\Delta t} = & \int_0^{t+\Delta t} C_{ij}^\infty \frac{\partial \bar{\epsilon}_j^\tau}{\partial \tau} d\tau \quad (7) \\ & + \sum_{m=1}^N \int_0^{t+\Delta t} \bar{C}_{ij} W_m \exp\left(-\frac{\xi^{t+\Delta t} - \xi^\tau}{\tau_m}\right) \frac{\partial \bar{\epsilon}_j^\tau}{\partial \tau} d\tau \end{aligned}$$

Now let us define an equation as follows.

$$\sigma_{im}^t = \int_0^t \bar{C}_{ij} W_m \exp\left(-\frac{\xi^t - \xi^\tau}{\tau_m}\right) \frac{\partial \bar{\epsilon}_j^\tau}{\partial \tau} d\tau \quad (8)$$

and then the second integral in Eq. (7) at time $t + \Delta t$ can be defined as

$$\begin{aligned} \sigma_{im}^{t+\Delta t} = & \int_0^{t+\Delta t} \bar{C}_{ij} W_m \exp\left(-\frac{\xi^{t+\Delta t} - \xi^\tau}{\tau_m}\right) \\ & \frac{\partial \bar{\epsilon}_j^\tau}{\partial \tau} d\tau \quad (9) \end{aligned}$$

This can be written in two parts.

$$\begin{aligned} \sigma_{im}^{t+\Delta t} = & \int_0^t \bar{C}_{ij} W_m \exp\left(-\frac{\xi^{t+\Delta t} - \xi^\tau}{\tau_m}\right) \frac{\partial \bar{\epsilon}_j^\tau}{\partial \tau} d\tau \\ & + \int_t^{t+\Delta t} \bar{C}_{ij} W_m \exp\left(-\frac{\xi^{t+\Delta t} - \xi^\tau}{\tau_m}\right) \frac{\partial \bar{\epsilon}_j^\tau}{\partial \tau} d\tau \quad (10) \end{aligned}$$

For a sufficiently small time increment Δt , the reduced time can be expressed as

$$\xi^{t+\Delta t} = \xi^t + \Delta \xi^{t+\Delta t} \quad (11)$$

and the increment in the reduced time $\Delta \xi$ is defined as

$$\Delta \xi^{t+\Delta t} = \int_t^{t+\Delta t} \frac{ds}{a_\tau} \quad (12)$$

By assuming that the degree of cure and the temperature are constant during the small time

interval Δt , Eq. (12) can be approximated as

$$\Delta \xi^{t+\Delta t} = \frac{\Delta t}{a_\tau} \quad (13)$$

Using Eqs. (8) and (11), the first integral in Eq. (10) is expressed as

$$\begin{aligned} I_1 = & \int_0^t \bar{C}_{ij} W_m \exp\left(-\frac{\xi^{t+\Delta t} - \xi^\tau}{\tau_m}\right) \frac{\partial \bar{\epsilon}_j^\tau}{\partial \tau} d\tau \\ = & \exp\left(-\frac{\Delta \xi^{t+\Delta t}}{\tau_m}\right) \int_0^t \bar{C}_{ij} W_m \exp\left(-\frac{\xi^t - \xi^\tau}{\tau_m}\right) \frac{\partial \bar{\epsilon}_j^\tau}{\partial \tau} d\tau \quad (14) \\ = & \exp\left(-\frac{\xi^{t+\Delta t}}{\tau_m}\right) \sigma_{im}^t \end{aligned}$$

In order to solve the second integral in Eq. (9), let us assume that the change in the strain is constant in an interval of $t < \tau < t + \Delta t$. Namely,

$$\frac{\partial \bar{\epsilon}_j^\tau}{\partial \tau} \cong \frac{\bar{\epsilon}_j^{t+\Delta t} - \bar{\epsilon}_j^t}{\Delta t} = \text{constant} \quad (15)$$

Using Eqs. (13) and (15), and assuming that moduli and weighting factors are constant for Δt , the second integral in Eq. (10) can be derived as follows.

$$\begin{aligned} I_2 = & \int_t^{t+\Delta t} \bar{C}_{ij} W_m \exp\left(-\frac{\xi^{t+\Delta t} - \xi^\tau}{\tau_m}\right) \frac{\partial \bar{\epsilon}_j^\tau}{\partial \tau} d\tau \\ \cong & \frac{\bar{\epsilon}_j^{t+\Delta t} - \bar{\epsilon}_j^t}{\Delta t} \bar{C}_{ij}^{t+\Delta t} W_m \exp\left(-\frac{\xi^{t+\Delta t}}{\tau_m}\right) \\ & \int_t^{t+\Delta t} \exp\left(\frac{\xi^\tau}{\tau_m}\right) d\tau \\ = & \frac{\bar{\epsilon}_j^{t+\Delta t} - \bar{\epsilon}_j^t}{\Delta t} \bar{C}_{ij}^{t+\Delta t} W_m \exp\left(-\frac{\xi^{t+\Delta t}}{\tau_m}\right) \\ & \int_{\xi^t}^{\xi^{t+\Delta t}} \exp\left(\frac{\xi^\tau}{\tau_m}\right) a_\tau d\xi \quad (16) \end{aligned}$$

Integrating this equation gives

$$\begin{aligned} I_2 = & \bar{C}_{ij}^{t+\Delta t} W_m \frac{\tau_m}{\Delta \xi^{t+\Delta t}} \left[1 - \exp\left(-\frac{\xi^{t+\Delta t}}{\tau_m}\right) \right] \\ & (\bar{\epsilon}_j^{t+\Delta t} - \bar{\epsilon}_j^t) \\ = & \bar{C}_{ij}^{t+\Delta t} \Gamma_m^{t+\Delta t} \Delta \bar{\epsilon}_j^{t+\Delta t} \quad (17) \end{aligned}$$

where $\Gamma_m^{t+\Delta t}$ and $\Delta \bar{\epsilon}_j^{t+\Delta t}$ are defined as

$$\Gamma_m^{t+\Delta t} = W_m \frac{\tau_m}{\Delta \xi^{t+\Delta t}} \left[1 - \exp\left(-\frac{\Delta \xi^{t+\Delta t}}{\tau_m}\right) \right] \quad (18)$$

$$\Delta \bar{\epsilon}_j^{t+\Delta t} = \bar{\epsilon}_j^{t+\Delta t} - \bar{\epsilon}_j^t \quad (19)$$

Therefore, substituting Eqs. (14) and (17) into Eq. (10) gives

$$\sigma_{im}^{t+\Delta t} = \exp\left(-\frac{\Delta \xi^{t+\Delta t}}{\tau_m}\right) \sigma_{im}^t + \Gamma_m^{t+\Delta t} \bar{C}_{ij}^{t+\Delta t} \Delta \bar{\epsilon}_j^{t+\Delta t} \quad (20)$$

where the initial values of Γ_m^0 and σ_m^0 at time $t=0$ are W_m and 0, respectively. From a computational point of view, Eq. (20) is much easier to deal with than Eq. (9) because Eq. (20) requires a knowledge of quantities at the previous time t , while Eq. (9) requires a knowledge of quantities over the complete history of the response of the material. Substitution of Eq. (20) into Eq. (7) leads to

$$\begin{aligned} \sigma_i^{t+\Delta t} &= \int_0^{t+\Delta t} C_{ij}^\infty \frac{\partial \bar{\epsilon}_j^\tau}{\partial \tau} d\tau + \sum_{m=1}^N \sigma_{im}^{t+\Delta t} \\ &= \int_0^{t+\Delta t} C_{ij}^\infty \frac{\partial \bar{\epsilon}_j^\tau}{\partial \tau} d\tau + \sum_{m=1}^N \exp\left(-\frac{\Delta \xi_m^{t+\Delta t}}{\tau_m}\right) \sigma_m^t \\ &\quad + \sum_{m=1}^N \Gamma_m^{t+\Delta t} \bar{C}_{ij}^{t+\Delta t} \Delta \bar{\epsilon}_j^{t+\Delta t} \end{aligned} \quad (21)$$

Similarly, the stresses at time t can be expressed as

$$\sigma_i^t = \int_0^t C_{ij}^\infty \frac{\partial \bar{\epsilon}_j^\tau}{\partial \tau} d\tau + \sum_{m=1}^N \sigma_{im}^t \quad (22)$$

By subtracting Eq. (22) from Eq. (21) and using Eq. (15), incremental stresses during the time interval Δt can be obtained as follows.

$$\begin{aligned} \Delta \sigma_i^{t+\Delta t} &= \sigma_i^{t+\Delta t} - \sigma_i^t \\ &= \left[C_{ij}^{\infty t+\Delta t} + \sum_{m=1}^N \Gamma_m^{t+\Delta t} \bar{C}_{ij}^{t+\Delta t} \right] \Delta \bar{\epsilon}_j^{t+\Delta t} \\ &\quad + \sum_{m=1}^N \left[\exp\left(-\frac{\Delta \xi_m^{t+\Delta t}}{\tau_m}\right) - 1 \right] \sigma_{im}^t \end{aligned} \quad (23)$$

This is a viscoelastic constitutive equation that represents the relationship of incremental stresses and strains during the small time step Δt . The stresses σ_{im}^t can be recursively calculated by Eq. (20). As mentioned above, the material is thermorheologically simple when the degree of cure is constant. Hence, Eq. (20) is valid only for the constant degree of cure. If the relaxation times τ_m depend on the degree of cure (Kim, 1996), Eq. (23) must be modified. If the degree of cure is assumed to be constant during the small time step Δt although it changes during entire curing process, Eq. (23) can be approximately used with modifications such that τ_m^t and $\tau_m^{t+\Delta t}$ are inserted, respectively, instead of τ_m in Eqs. (14) and (16), because Eq. (23) is the relationship of incremental stresses and strains during the small time step Δt . In this case, Eqs. (18), (20) and

(23) can be rewritten as

$$\begin{aligned} \Delta \sigma_i^{t+\Delta t} &= \sigma_i^{t+\Delta t} - \sigma_i^t \\ &= \left[C_{ij}^{\infty t+\Delta t} + \sum_{m=1}^N \Gamma_m^{t+\Delta t} \bar{C}_{ij}^{t+\Delta t} \right] \Delta \bar{\epsilon}_j^{t+\Delta t} \\ &\quad + \sum_{m=1}^N \left[\exp\left(-\frac{\Delta \xi_m^{t+\Delta t}}{\tau_m^t}\right) - 1 \right] \sigma_{im}^t \end{aligned} \quad (24)$$

where

$$\begin{aligned} \sigma_{im}^{t+\Delta t} &= \exp\left(-\frac{\Delta \xi_m^{t+\Delta t}}{\tau_m^t}\right) \sigma_{im}^t \\ &\quad + \Gamma_m^{t+\Delta t} \bar{C}_{ij}^{t+\Delta t} \Delta \bar{\epsilon}_j^{t+\Delta t} \\ \Gamma_m^{t+\Delta t} &= W_m^{t+\Delta t} \frac{\tau_m^{t+\Delta t}}{\Delta \xi_m^{t+\Delta t}} \left[1 - \exp\left(-\frac{\Delta \xi_m^{t+\Delta t}}{\tau_m^{t+\Delta t}}\right) \right] \end{aligned} \quad (25)$$

For the sake of simplicity, Eq. (24) can be written in the symbolic form.

$$\Delta \sigma^{t+\Delta t} = \mathbf{C}^{t+\Delta t} \Delta \bar{\epsilon}^{t+\Delta t} + \mathbf{H}^{t+\Delta t} \quad (27)$$

where \mathbf{C} and \mathbf{H} are called a time-dependent stiffness matrix and a hereditary stress vector, respectively, whose components are defined as

$$C_{ij}^{t+\Delta t} = C_{ij}^{\infty t+\Delta t} + \sum_{m=1}^N \Gamma_m^{t+\Delta t} \bar{C}_{ij}^{t+\Delta t} \quad (28)$$

$$H_i^{t+\Delta t} = \sum_{m=1}^N \left[\exp\left(-\frac{\Delta \xi_m^{t+\Delta t}}{\tau_m^t}\right) - 1 \right] \sigma_{im}^t \quad (29)$$

Note that the viscoelastic formulation given by Eq. (27) can be applied for the case of varying viscoelastic parameters (C_{ij}^∞ , \bar{C}_{ij} , τ_m) as well as constant parameters and will be very useful in applying for geometrically nonlinear analyses since it is defined as the incremental stress-strain relationship.

3. Viscoelastic Finite Element Formulation

An equilibrium equation at time $t+\Delta t$ using the principle of virtual work is expressed as

$$\int_V \delta(\epsilon^{t+\Delta t})^T \sigma^{t+\Delta t} dV = \delta W^{t+\Delta t} \quad (30)$$

where the superscript \mathbf{T} means transpose, \mathbf{V} and \mathbf{A} represent volume and area, and δW is the external virtual work defined as

$$\begin{aligned} \delta W^{t+\Delta t} &= \int_A \delta(u^{t+\Delta t})^T \bar{f}_s^{t+\Delta t} dA \\ &\quad + \int_V \delta(u^{t+\Delta t})^T \bar{f}_b^{t+\Delta t} dV \end{aligned}$$

$$= \int_A \delta(\Delta \mathbf{u}^{t+\Delta t})^T \mathbf{f}_s^{t+\Delta t} dA + \int_V \delta(\Delta \mathbf{u}^{t+\Delta t})^T \mathbf{f}_b^{t+\Delta t} dV \quad (31)$$

where \mathbf{u} , \mathbf{f}_s , and \mathbf{f}_b represent displacement, surface traction, and body force vectors, respectively. Note that $\delta \mathbf{u}^t$ is given as zero because the variation is taken about the configuration at time t . To use incremental stresses and strains, Eq. (30) can be rewritten as

$$\int_V \delta(\Delta \boldsymbol{\varepsilon}^{t+\Delta t})^T (\boldsymbol{\sigma}^t + \Delta \boldsymbol{\sigma}^{t+\Delta t}) dV = \delta W^{t+\Delta t} \quad (32)$$

where $\delta \boldsymbol{\varepsilon}^t$ is also zero because the variation is taken about the configuration at time t . By transforming Eq. (27) from material coordinates (on-axis) of a shell element to global coordinates and then substituting it into Eq. (32), the following equation can be obtained.

$$\int_V \delta(\Delta \boldsymbol{\varepsilon})^T \mathbf{D} \Delta \boldsymbol{\varepsilon} dV = \delta W - \int_V \delta(\Delta \boldsymbol{\varepsilon})^T \boldsymbol{\sigma} dV + \int_V \delta(\Delta \boldsymbol{\varepsilon})^T \mathbf{D} \mathbf{F}_{TC} dV - \int_V \delta(\Delta \boldsymbol{\varepsilon})^T \mathbf{F}_H dV \quad (33)$$

where the superscripts indicating the time are dropped hereafter for the sake of convenience, the stress and strain now refer to the global coordinates and \mathbf{D} , \mathbf{F}_{TC} and \mathbf{F}_H are, respectively, defined as

$$\mathbf{D} = \mathbf{Q} \mathbf{C} \mathbf{Q}^T \quad (34)$$

$$\mathbf{F}_{TC} = \mathbf{Q} \Delta \boldsymbol{\varepsilon}_{TC} \quad (35)$$

$$\mathbf{F}_H = \mathbf{Q} \mathbf{H} \quad (36)$$

where \mathbf{Q} represents a matrix that transforms the stress-strain law from the material coordinates to the local coordinates of the element and then from the local coordinates to the global coordinates (Bathe, 1982). The modulus \mathbf{C} in Eq. (34) is recalculated under the assumption that the stress normal to the shell surface is zero, and \mathbf{H} is given by Eq. (29). $\Delta \boldsymbol{\varepsilon}_{TC}$ represents a vector including both the thermal and chemical shrinkage strains defined by the second and third terms in the right hand side of Eq. (2).

On the basis of a 3-D degenerated shell element and the first order shear deformation theory (Chao and Reddy, 1984 and Panda and Natara-

jan, 1981), the incremental displacement vector $\Delta \mathbf{u}$ in an isoparametric element with p nodal points is defined as (Bathe, 1982)

$$\Delta \mathbf{u} = \sum_{k=1}^p \mathbf{N}^k(\xi, \eta) \Delta \bar{\mathbf{u}}^k + \frac{1}{2} \sum_{k=1}^p \mathbf{N}^k(\xi, \eta) t^k \zeta \Delta \mathbf{V}^k \quad (37)$$

where ξ , η , ζ denote the local coordinates of the element and $\Delta \bar{\mathbf{u}}^k$, \mathbf{N}^k , t^k represent the displacement vector, shape function and thickness at nodal point k of the element, respectively.

$\Delta \mathbf{V}^k$ represents, at nodal point k , the difference between unit vectors normal to the shell mid-surface at time $t + \Delta t$ and at time t , that is, $\mathbf{V}_{t+\Delta t}^k - \mathbf{V}_t^k$. The strain-displacement relation is written in the tensor form :

$$\boldsymbol{\varepsilon} = \frac{1}{2} ((\nabla \mathbf{u})^T + \nabla \mathbf{u}) \quad (38)$$

Expressing a virtual displacement vector and a virtual strain vector by a function of a virtual nodal displacement vector using Eqs. (37) and (38) gives

$$\delta(\Delta \mathbf{u}) = \mathbf{N} \delta(\Delta \bar{\mathbf{u}}), \quad \delta(\Delta \boldsymbol{\varepsilon}) = \mathbf{B} \delta(\Delta \bar{\mathbf{u}}) \quad (39)$$

where \mathbf{N} and \mathbf{B} represent a shape function matrix and a displacement-strain matrix, respectively (Bathe, 1982). By substituting Eq. (39) into Eq. (33), the viscoelastic finite element formulation is summarized as

$$\mathbf{K}^{t+\Delta t} \Delta \bar{\mathbf{u}}^{t+\Delta t} = \mathbf{F}^{t+\Delta t} \quad (40)$$

where the time-dependent stiffness matrix \mathbf{K} and the force vector \mathbf{F} are given by

$$\mathbf{K}^{t+\Delta t} = \int_V \mathbf{B}^T \mathbf{D}^{t+\Delta t} \mathbf{B} dV \quad (41)$$

$$\mathbf{F}^{t+\Delta t} = \mathbf{R}^{t+\Delta t} - \int_V \mathbf{B}^T \boldsymbol{\sigma}^t dV + \int_V \mathbf{B}^T \mathbf{D}^{t+\Delta t} \mathbf{F}_{TC}^{t+\Delta t} dV - \int_V \mathbf{B}^T \mathbf{F}_H^{t+\Delta t} dV \quad (42)$$

$$\mathbf{R}^{t+\Delta t} = \int_A \mathbf{N}^T \mathbf{f}_s^{t+\Delta t} dA + \int_V \mathbf{N}^T \mathbf{f}_b^{t+\Delta t} dV \quad (43)$$

After solving Eq. (40) with boundary conditions, displacements, strains and stresses at time $t + \Delta t$ are updated as follows.

$$\begin{aligned} \mathbf{u}^{t+\Delta t} &= \mathbf{u}^t + \Delta \mathbf{u}^{t+\Delta t} \\ \boldsymbol{\varepsilon}^{t+\Delta t} &= \boldsymbol{\varepsilon}^t + \Delta \boldsymbol{\varepsilon}^{t+\Delta t} \\ \boldsymbol{\sigma}^{t+\Delta t} &= \boldsymbol{\sigma}^t + \Delta \boldsymbol{\sigma}^{t+\Delta t} \end{aligned} \quad (44)$$

Table 1 Cure kinetics constants of Hercules 3501-6 resin

| | |
|----------------------------|----------------------|
| R (J/mol °K) | 8.314×10^3 |
| A_1 (min ⁻¹) | 2.101×10^9 |
| A_2 (min ⁻¹) | -2.014×10^9 |
| A_3 (min ⁻¹) | 1.960×10^5 |
| ΔE_1 (J/mol) | 8.07×10^4 |
| ΔE_2 (J/mol) | 7.78×10^4 |
| ΔE_3 (J/mol) | 5.66×10^4 |

4. Degree of Cure, Shift Factor, and Relaxation Time

The degree of cure α is defined as the ratio of the heat of reaction released up to time t and the total heat of reaction (Lee et al., 1982 and Dusi et al., 1987). If the degree of cure is zero, it means that composites are uncured. If the degree of cure is one, it means that the composites are fully cured. The degree of cure at time t is calculated by

$$\alpha(t) = \int_0^t \frac{d\alpha}{dt} dt \quad (45)$$

Since Hercules AS4/3501-6 graphite/epoxy composite is used in this paper, the rate of the degree of cure for Hercules 3501-6 resin is determined experimentally as below (Lee et al., 1982).

$$\frac{d\alpha}{dt} = (k_1 + k_2\alpha)(1 - \alpha)(0.47 - \alpha) \quad (\alpha \leq 0.3) \quad (46)$$

$$\frac{d\alpha}{dt} = k_3(1 - \alpha) \quad (\alpha > 0.3)$$

Constants k_1 , k_2 , k_3 are defined as

$$\begin{aligned} k_1 &= A_1 \exp(-\Delta E_1/RT) \\ k_2 &= A_2 \exp(-\Delta E_2/RT) \\ k_3 &= A_3 \exp(-\Delta E_3/RT) \end{aligned} \quad (47)$$

where R is the universal gas constant, and A_1 , A_2 , A_3 and ΔE_1 , ΔE_2 , ΔE_3 represent the pre-exponential factors and activation energies of which values are presented in Table 1. The shift factor a_T of AS4/3501-6 composite is functions of the degree of cure and the temperature, and

Table 2 Relaxation times and weighting factors at reference degree of cure ($\alpha_r = 0.98$)

| m | τ_m (min) | W_m |
|---|-----------------|-----------|
| 1 | $2.922137e+1$ | 0.0591334 |
| 2 | $2.921437e+3$ | 0.0661225 |
| 3 | $1.82448e+5$ | 0.0826896 |
| 4 | $1.1031059e+7$ | 0.112314 |
| 5 | $2.8305395e+8$ | 0.154121 |
| 6 | $7.9432822e+9$ | 0.2618288 |
| 7 | $1.953424e+11$ | 0.1835594 |
| 8 | $3.3150756e+12$ | 0.0486939 |
| 9 | $4.9174856e+14$ | 0.0252258 |

defined as (Kim, 1996)

$$a_T = 10 \left\{ -a_1 \exp\left(\frac{1}{\alpha - T_r}\right) - a_2 \right\} (T - T_r) \quad (48)$$

where the values of constants a_1 and a_2 are 1.4 and 0.0712, respectively, and T_r denotes the reference temperature determined as 25°C. The relaxation time τ_m is a function of the degree of cure and experimentally determined as (Kim, 1996)

$$\begin{aligned} \tau_m(\alpha) &= 10^{[\text{Log}(\tau_m(\alpha_r)) + \{f(\alpha) - (\alpha - \alpha_r) \text{Log}(\lambda_m)\}]} \\ f(\alpha) &= 0.0536 + 0.0615\alpha + 0.9227\alpha^2 \quad (49) \\ \lambda_m &= \frac{10^{9.9}}{\tau_m(\alpha_r)} \end{aligned}$$

where the reference degree of cure α_r is 0.98. The relaxation times $\tau_m(\alpha_r)$ corresponding to α_r and the weighting factors and the weighting factors W_m are presented in Table 2.

5. Numerical Results and Discussion

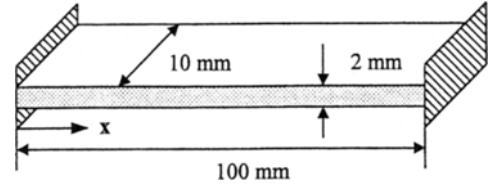
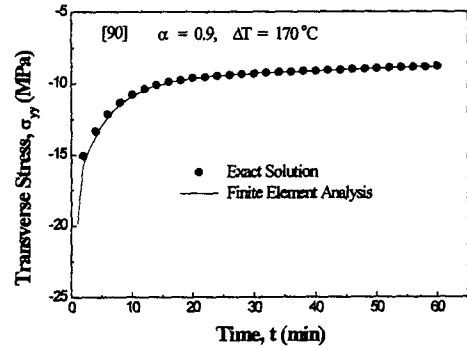
Material properties of Hercules AS4/3501-6 composite used in the analysis are presented in Table 3. These values are used to calculate the moduli C_{ij} given by Eq. (5). Since stress relaxation in the direction of a fiber is negligible, the components C_{11} , C_{12} , and C_{13} related to the fiber direction are assumed to be linearly elastic. The fully relaxed modulus C_{ij}^∞ is experimentally determined by 1/7 of the unrelaxed moduli C_{ij}^0 (Kim, 1996). The element used for the finite element

Table 3 Mechanical properties of AS4/3501-6 used in calculations

| | |
|-------------------------------------|------------------------------|
| Young's modulus, E_{11} | 125.38 GPa |
| Young's modulus, E_{22} | 8.1 GPa |
| Young's modulus, E_{33} | 8.1 GPa |
| Shear modulus, G_{12} | 4.1 GPa |
| Shear modulus, G_{13} | 4.1 GPa |
| Shear modulus, G_{23} | 2.75 GPa |
| Poisson's ratio, ν_{12} | 0.25 |
| Poisson's ratio, ν_{13} | 0.25 |
| Poisson's ratio, ν_{23} | 0.47 |
| Thermal expansion coeff., β_1 | $0.5e-6$ $1/^\circ\text{C}$ |
| Thermal expansion coeff., β_2 | $35.3e-6$ $1/^\circ\text{C}$ |
| Thermal expansion coeff., β_3 | $35.3e-6$ $1/^\circ\text{C}$ |
| Chemical shrinkage coeff., η_1 | $-1.67e-4$ |
| Chemical shrinkage coeff., η_2 | $-8.81e-3$ |
| Chemical shrinkage coeff., η_3 | $-8.81e-3$ |

analyses is a 8-node degenerated shell element with five degrees of freedom per node and $2 \times 2 \times 2$ Gauss integration points are used at each layer of the element to prevent shear locking. Because the accuracy of numerical results depends on the time increment Δt , the finite element analyses are executed for three time increments $\Delta t = 10$ sec, 30 sec, and 1 min for the purpose of examining the convergence of numerical solutions and it is found that there is no big difference between them. Therefore, to save computational time, the time increment $\Delta t = 1$ min is used for the numerical analyses presented below.

In order to verify the finite element program developed in this study, the following procedure is carried out. Since there does not exist an exact solution available for comparing with results calculated in this study, a laminated plate as shown in Fig. 1 is presented, of which both ends are clamped. The length, width, and thickness of the plate are 100 mm, 10 mm, and 2 mm, respectively, and fiber orientation is 90° normal to the x -axis. If the degree of cure is constant and the temperature is given by $\Delta T h(t)$ where $h(t)$ is a

**Fig. 1** Geometry of the clamped laminated plate**Fig. 2** Comparison of the finite element analysis with the exact solution for the transverse stress in the laminated plate

unit step function, the exact solution for a one dimensional problem can be obtained from Eq. (6). The total strain ϵ_2 in Eq. (2) is given as zero because the ends of the plate are clamped. Neglecting the deformation due to the chemical shrinkage and substituting Eqs. (2) and (4) into Eq. (6) and integrating it, the one dimensional exact solution is given as

$$\sigma_2^i = \left[E_{22}^\infty + \bar{E}_{22} \sum_{m=1}^N W_m \exp\left(-\frac{t}{\tau_m a_T}\right) \right] (-\beta_2 \Delta T) \quad (50)$$

where the subscripts 2 represent the direction normal to the fiber orientation and E represents Young's modulus. For the finite element analysis, the plate shown in Fig. 1 is divided by 10 meshes in length and 2 meshes in width. When the degree of cure α and the temperature change ΔT are 0.9 and 170°C , respectively, the results calculated by Eq. (50) and the finite element analysis are compared in Fig. 2. The result of the finite element analysis is calculated at the center of the plate. Because two results agree well each other as shown in Fig. 2, validation of the finite element program developed in this paper can be accomplished.

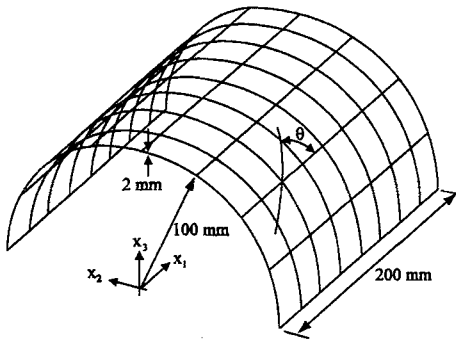


Fig. 3 Geometry of the laminated semi-circular shell

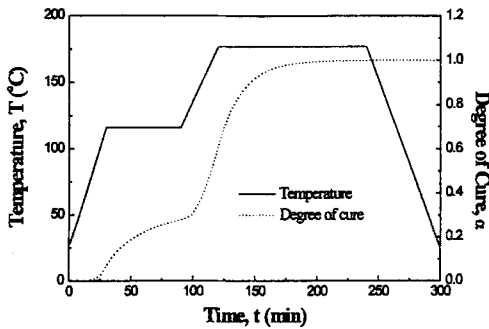


Fig. 4 The temperature and the degree of cure during cure

To predict residual stresses occurred in a laminated cylindrical shell during cure, the laminated semi-circular shell as shown in Fig. 3 was selected. The radius, length and thickness of the shell are 100 mm, 200 mm and 2 mm respectively. The laminated shell consists of 4 plies. All four edges of the shell are given as free and temperature distribution is assumed to be uniform. For the finite element analysis, the laminated shell is uniformly divided by 8×8 elements. Total numbers of the elements and nodes are 64 and 225, respectively. A cure cycle used in the calculations is presented in Fig. 4. Total cure time is 300 minutes, and temperatures at the first dwell known as a consolidation stage and at the second dwell known as a cure stage are defined as 116°C and 177°C, respectively. The degree of cure calculated for this cure cycle is presented in Fig. 4 as dot lines and increased rapidly near the time about 100 minutes.

To investigate the viscoelastic effect on the

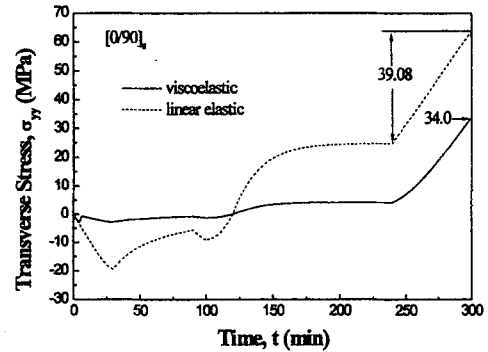


Fig. 5 Comparison of the viscoelastic transverse stress with the linear elastic transverse stress during cure

residual stresses in the laminated shell during the cure, the analysis is performed for $[0/90]_s$ laminate where a positive fiber orientation θ is shown in Fig. 3. Numerical results calculated for both viscoelastic and linear elastic analyses considering thermal deformation and cure-induced chemical shrinkage simultaneously are presented in Fig. 5. The transverse stresses normal to the fiber orientation are calculated at the first ply 0° and near the center of the shell, and the results show that they are quite different as shown in Fig. 5. Especially, it is found that the viscoelastic stress is considerably relaxed with the advance in cure, and that the final residual stress for the viscoelastic analysis is much lower than that for the linear elastic analysis. Generally, calculating the residual stress using the linear elastic analysis, the stress is assumed to be free at the second dwell called the cure stage and calculated only at the cooling stage. Under this assumption, the calculated linear elastic stress is 39.08 MPa during cooling as shown in Fig. 5 and the final viscoelastic stress is 34.0 MPa at the end of cure. This indicates 15% difference with respect to the viscoelastic stress and, in addition, the linear elastic analysis has disadvantage that the residual stress can not be calculated in the entire curing process. But, the viscoelastic analysis makes it possible to predict the residual stresses in the entire curing process. Especially, from the viscoelastic result in Fig. 5, the residual stress at the second dwell is approximately 4.1 MPa. Although this value is not big, it may cause matrix cracking in the

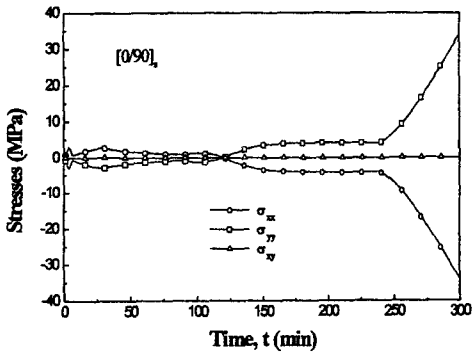


Fig. 6 Residual stress development at the first ply (0 degree) during cure

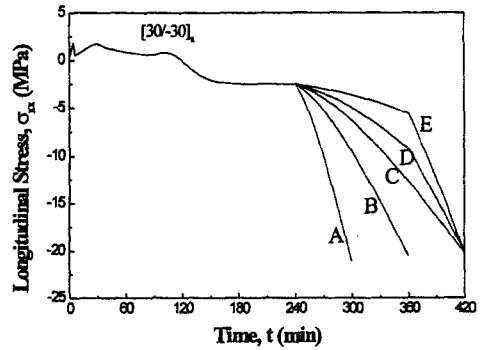


Fig. 9 Longitudinal stress development at the first ply (30 degree) for various cure cycles

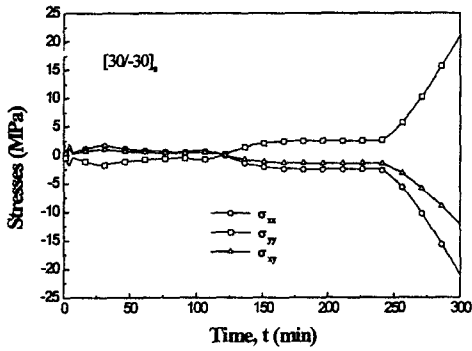


Fig. 7 Residual stress development at the first ply (30 degree) during cure

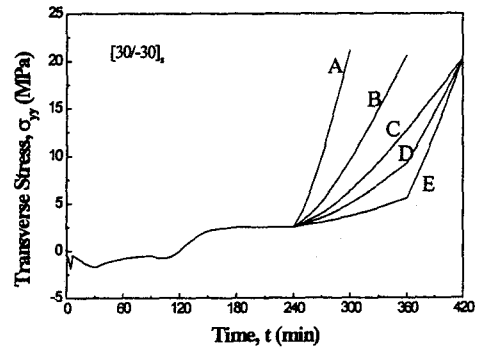


Fig. 10 Transverse stress development at the first ply (30 degree) for various cure cycles

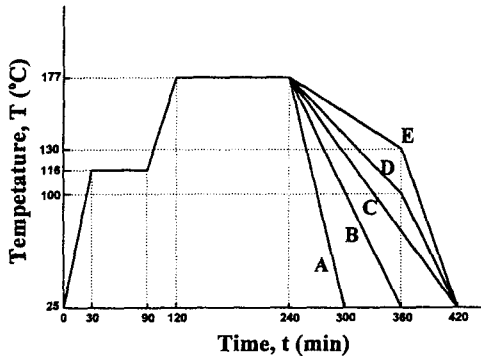


Fig. 8 Cure cycles with different cooling paths

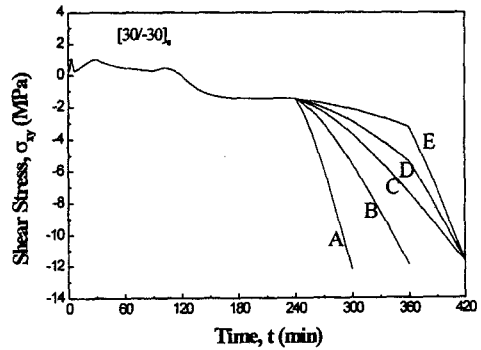


Fig. 11 Shear stress development at the first ply (30 degree) for various cure cycles

laminated shell during cure. The reason for this is that the transverse strength of composites becomes to be rapidly decreased as temperature increases, and that mechanical properties of the composites change very complicatedly when the degree of cure changes rapidly as shown in Fig. 4 and the transverse strength is expected to be quite small, specially, near the cure time 100 minutes.

Next, the residual stresses are calculated during

the cure for laminated shells consisting of $[0/90]_s$ and $[30/-30]_s$ stacking sequences. The results are presented in Figs. 6 and 7 where σ_{xx} , σ_{yy} and σ_{xy} represents longitudinal, transverse and in-plane shear stresses, respectively. The stresses are calculated at the first plies (0° and 30°) and near the center of the shell. As shown in the figures, the longitudinal and transverse stresses for the cross-

Table 4 Comparison of final residual stress for cure cycles with different cooling paths

| Cure cycle | Longitudinal stress, σ_{xx} (MPa) | Transverse stress, σ_{yy} (MPa) | Shear stress, σ_{xy} (MPa) |
|------------|--|--|-----------------------------------|
| A | -21.289 | 21.168 | -12.208 |
| B | -20.737 | 20.619 | -11.892 |
| C | -20.416 | 20.300 | -11.708 |
| D | -20.330 | 20.215 | -11.658 |
| E | -20.369 | 20.253 | -11.681 |

ply laminated shell are greater than those for the angle-ply laminated shell, whereas the in-plane shear stress is greater in the angle-ply laminated shell.

Finally, in order to investigate the effect of cooling rate on the residual stresses, five cure cycles are selected as shown in Fig. 8 and they are given to be equal from the beginning of the cure cycle to the end of cure stage. The results of stresses analyzed for $[30/-30]_s$ laminated shell with different cooling paths are presented in Figs. 9, 10 and 11 and the stresses are also calculated at the first ply and near the center of the shell. The final residual stresses for these cure cycles are compared in Table 4. Comparing the results of the cure cycles A, B and C, the stresses are slightly reduced as the cooling time is increased. The cooling times of the cure cycles C, D and E are fixed as 180 minutes and only their cooling paths are different. In this case, as shown in Table 4, the stresses for the cooling path D are smaller than those for the paths C and E. Therefore, an optimal cooling path showing the minimum residual stresses is expected to exist near the path D if the cooling time is fixed as 180 minutes.

5. Conclusions

In this study, the viscoelastic finite element analysis has been performed to investigate the residual stresses occurred in laminated shells during the cure. The incremental viscoelastic constitutive equation that can describe the stress relaxation during the cure is derived as a recur-

sive formula which can be used conveniently for a numerical analysis. The stress relaxation is defined as functions of the degree of cure and the temperature. The finite element analysis program is developed on the basis of the 3-D degenerated shell element and the first order shear deformation theory, and is verified by comparing with the one dimensional exact solution.

To investigate the viscoelastic effect of on the residual stresses in the laminated shell during the cure, both the viscoelastic and linear elastic analyses are performed for $[0/90]_s$ laminated semi-circular shell considering thermal deformation and chemical shrinkage simultaneously. The results show that there is big difference between the viscoelastic stresses and the linear elastic stresses. The longitudinal, transverse and in-plane shear stresses are calculated and compared during the entire curing process for $[0/90]_s$ cross-ply and $[30/-30]_s$ angle-ply stacking sequences.

Finally, the effect of both the cooling rate and the cooling path on the residual stresses is investigated for five cure cycles with different cooling paths. The residual stresses calculated for the cooling times 60, 120 and 180 minutes are shown to be slightly reduced as the cooling time is increased. The results show that the final residual stresses also depend on the cooling path when the cooling time is fixed.

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