Compaction of Thick Composites: Simulation and Experiment

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SUMMARY: A one-dimensional cure simulation model was developed for thick composite laminates including the effects of bleeder plies. To validate the model, 3-cm and 5-cm thick laminates were fabricated on a hot press using AS4/3501-6 graphite/epoxy prepreg. The fiber compressibility parameters were calibrated for the 3-cm thick laminate and then used to predict the compaction behavior of the 5-cm-thick laminate. The nonsymmetrical temperature distribution resulting from the presence of bleeder plies was well predicted by the model. Some of the discrepancies in the predicted compaction behavior could be attributed to those in the predicted temperature distribution. The temperature prediction was better for the thinner laminate.

KEYWORDS: thick composites, compaction, process simulation

INTRODUCTION

Thick-section composite structures are more difficult to fabricate because of their exothermic behavior. Finding an optimum cure cycle depends on the laminate thickness and a trial-anderror approach is not efficient. A solution to this problem is to develop a validated simulation model to predict the curing process.

A number of models for curing of composite laminates have appeared in the literature [1-4]. They are all based on a coupled formulation of energy balance and force balance. The energy balance equation yields temperature distribution and the force balance equation compaction behavior. The temperature distribution affects resin viscosity and hence the compaction behavior. These models, however, did not include the effects of bleeder plies, which are more frequently used with thick laminates in order to absorb excess resin. Also, they have not been fully validated experimentally.

The objective of the present study is to propose a one-dimensional cure simulation model for thick composites and provide experimental validation. The model recognizes the presence of bleeder plies, which have been neglected so far. The pertinent heat transfer and compaction equations are taken from [5]. The cure kinetic equation proposed by Lee et al. [6] is used together with the Dave's equation for fiber compressibility [3]. Outputs are temperature distribution, degree of cure, fiber volume fraction, and laminate thickness, all as functions of cure time. The governing equations are solved using a finite difference method.

MODEL FORMULATION

Heat Transfer

One-dimensional through-the-thickness heat transfer in a composite laminate is described by

$$\rho_c C_c \frac{\partial T}{\partial t} - K_c^T \frac{\partial^2 T}{\partial z^2} = V_m \rho_m H_{Rm} \frac{\partial \alpha}{\partial t}$$
(1)

where ρ is density, *C* is specific heat, *T* is temperature, *t* is time, K^T is thermal conductivity, *z* is the position coordinate from the bottom laminate surface, *V* is volume fraction, H_R is total heat of reaction, and α is degree of cure. Subscripts *c* and *m* represent composite and matrix, respectively. The composite properties are determined from its constituent properties as shown in the Appendix. All material properties are also provided in the Appendix.

The heat transfer in the bleeder plies is described by

$$\rho_b C_b \frac{\partial T}{\partial t} - K_b^T \frac{\partial^2 T}{\partial z^2} = \phi \rho_m H_{Rm} \frac{\partial \alpha}{\partial t}$$
(2)

where ϕ is porosity of the bleeder. Properties of the filled region of the bleeder are determined from the bleeder and resin using the rule of mixtures. Thus the right-hand side of equation 2 vanishes in the unfilled region. Furthermore, the convection term due to resin flow is neglected since the resin velocity and temperature gradient are assumed small.

The composite-bleeder assembly is subjected to a uniform heating on top of the bleeder and at the bottom of the laminate. Thus the boundary conditions are

$$T = T_b(t) \qquad \text{at } z = 0 \tag{3}$$
$$T = T_b(t) \qquad \text{at } z = h + h_b$$

where *h* and h_b are laminate and bleeder thickness, respectively. $T_b(t)$ is the temperature history prescribed on the boundaries. At the laminate/bleeder interface, temperature and heat flux are continuous. Initially, temperature and degree of cure are uniform throughout the laminate.

Compaction

The analysis of resin flow and fiber deformation in composite is based on the squeezed sponge model. The governing equations for 1-D compaction are given by [2, 4]

$$\frac{\partial p}{\partial t} = \frac{V_f}{S_f} \frac{\partial}{\partial z} \left(\frac{K_f^p}{\mu} \frac{\partial p}{\partial z} \right)$$
(4)

$$p + \sigma_{fe} = p_a \tag{5}$$

where p_a and p are applied pressure and resin pressure, respectively. K_f^p and S_f are permeability and tangent compliance, respectively, of the fiber preform and μ is resin viscosity. Note that p_a is maintained constant during curing. The tangent compliance gives a

change in fiber volume fraction resulting from a change in effective compressive fiber stress σ_{fe} :

$$S_f = \frac{\partial V_f}{\partial \sigma_{fe}} \tag{6}$$

A fiber compressibility equation relating V_f and σ_{fe} [2] is given in the Appendix.

The initial and boundary conditions for the composite are

$$p = p_{a} t = 0$$

$$\frac{\partial p}{\partial z} = 0 z = 0$$

$$p = p_{b/c} z = h$$
(7)

where $p_{b/c}$ refers to the pressure at the interface of the composite and the bleeder. The pressure at the top of the bleeder is assumed zero so that the excess resin can bleed out from the laminate without any resistance.

The resin flow in the bleeder is assumed to follow the Darcy's equation. The resin pressure distribution inside the bleeder is then given by

$$p_b = \frac{p_{b/c}}{h_f} \left(h + h_f - z \right) \tag{8}$$

where subscript *b* denotes the bleeder and h_f is the distance to the flow front from the top laminate surface. The superficial resin velocity v_b relative to the bleeder is related to the interface pressure by

$$v_b = \phi \frac{dh_f}{dt} = \frac{K_b}{\mu} \frac{p_{b/c}}{h_f}$$
(9)

The conservation of resin mass across the composite/bleeder interface requires that the superficial fiber velocity at the interface be equal to the resin velocity inside the bleeder:

$$-v_f\Big|_{z=h} = v_b \tag{10}$$

The conservation of mass inside the composite together with the Darcy's equation yields the superficial fiber velocity in term of the resin pressure gradient as

$$v_f = V_f \frac{K_f^p}{\mu} \frac{\partial p}{\partial z} \tag{11}$$

The flow continuity across the composite/bleeder interface thus manifests itself in the following equation:

$$-V_f K_f^p \left. \frac{\partial p}{\partial z} \right|_{z=h} = K_b \left. \frac{P_{b/c}}{h_f} \right. \tag{12}$$

The change in composite thickness is finally obtained as

$$\Delta h = h_o - h = \int \left| \frac{K_f^p}{\mu} \frac{\partial p}{\partial z} \right|_{z=h} dt$$
(13)

where subscript *o* is used to denote the initial state.

The equations introduced so far are sufficient to solve the coupled problem of laminate compaction and resin filling of the bleeder. They were solved using a finite difference method. It was found that 36 nodes and 5-sec time interval had yielded the desired convergence.

EXPERIMENTAL PROCEDURE

The model was validated by curing 3-cm and 5-cm thick laminates on a Tetrahedron hot press. The material was Hercules AS4/3501-6 prepreg tape. The manufacturer's recommended cure cycle was used for all experiments. Prior to curing, the prepreg stacks were de-balked and the entire bagging assembly was held under load so that the initial thickness could be stabilized before cure. The amount of compaction during cure was determined by measuring the change in the gap between the upper and lower platens using a LVDT. An external pressure of 0.689 MPa was applied and vacuum was maintained internally throughout the curing process.

The in-plane dimensions of the 3-cm thick laminate were 15.24cm x 15.24cm, and the ply orientations were $[0/90]_{114s}$. Fifteen bleeder plies were placed on top of the laminate to absorb excess resin for the 3-cm thick laminate while twenty-one bleeder plies were used for the 5-cm thick laminate. Aluminum molds were placed around the laminate to prevent fiber washout and resin flow from the sides. A silicon rubber was placed between the laminate and the aluminum mold so that an adiabatic condition could be maintained at the sides as much as possible. The bagging schedule and the placement of thermocouples are shown in Figure 1.

RESULTS/DISCUSSION

Because of the presence of the bleeder plies, which have a lower thermal diffusivity than the composite, the temperature distribution in the 3-cm thick laminate was not symmetric across the thickness, Figure 2. The excess resin pooled in the bleeder plies generates exothermic heat and thus contributes to the higher peak temperature at the top of the laminate. The overall agreement between the predictions and the experimental results is quite good.

The thickness change during cure is shown in Figure 3. The disturbances seen around 150 min. into cure were caused by an inadvertent, slight loss of pressure on the machine. This did not have any significant effect on the experimental results since the laminate had already reached full compaction when this incidence happened. The final thickness of the laminate was 27.45mm.



Fig. 1: Bagging schedule and thermocouple locations

The prediction shows a quicker compaction than actually observed. One possible reason might be that the viscosity predicted by the model was lower than actual. A lower viscosity would offer less resistance to the fiber deformation and hence result in a quicker compaction. It should be noted that the fiber compressibility model had been calibrated to match the initial and final thicknesses of the 3-cm laminate. As predicted, the top 8 bleeder plies were not saturated with the resin.

The temperature profile for the 5-cm thick laminate is shown in Figure 4. Again, the profile is not symmetric and the composite/bleeder interface temperature is higher than at the laminate bottom. The model-experiment correlation is not so good as for the 3-cm laminate: the predicted temperature rise is much slower than actual in the initial phase of cure. This is not surprising because any error in simulation would manifest itself more clearly when the thickness is larger.

The thickness change for the 5-cm thick laminate is shown in Figure 5. The final thickness of the laminate is 46.48mm. This value is only slightly higher than the one predicted by the fiber compressibility model using the same parameters as determined from the 3-cm laminate result. The discrepancy in compaction behavior between the prediction and experiment could be explained by a combination of the discrepancy in temperature and the discrepancy in viscosity. The 3-cm laminate data indicates that the predicted viscosity might be lower than actual. However, the large temperature lag exhibited by the simulation would push the viscosity higher and predict a slower compaction.

CONCLUSIONS

A 1-dimensional cure simulation model including the effects of bleeder plies has been developed for thick composite laminates. The model can predict the degree of cure, temperature distribution and laminate compaction. Laminates of 3-cm and 5-cm thickness have been fabricated to validate the model. The model-experiment correlation for temperature is good for the 3-cm laminate but not so good for the thicker 5-cm laminate. The discrepancies in the compaction behavior could be explained by the possible inaccuracy in the viscosity model used and the discrepancy in the predicted temperature.



Fig. 2: Model-experiment correlation for temperature distribution in 3-cm thick laminate at a) middle of laminate b) interface between laminate and bleeder c) middle of bleeder stack.



Fig. 3: Model-experiment correlation for compaction of 3-cm thick laminate.



Fig. 4: Model-experiment correlation for temperature distribution in 5-cm thick laminate at a) middle of laminate b) interface between laminate and bleeder c) middle of bleeder stack.



Fig. 5: Model-experiment correlation for compaction of 5-cm thick laminate.

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APPENDIX

<u>Matrix</u>

Density [7]:

$$\rho_{m} = 90\alpha + 1232 \qquad \alpha \le 0.45$$

$$\rho_{m} = 1272 \qquad \alpha > 0.45$$
Specific heat [7]:
Thermal conductivity [7]:

$$K_{m}^{T} = 0.161161 + (0.001465T - 0.017163)\alpha \quad (T \text{ in } ^{\circ}\text{C})$$
Curing rate:

$$\frac{d\alpha}{dt} = \begin{cases} (K_{1} + K_{2}\alpha)(1 - \alpha)(0.47 - \alpha) & \alpha \le 0.3\\ K_{3}(1 - \alpha) & \alpha \ge 0.3 \end{cases}$$

$$K_{i} = A_{i} \exp\left(-\frac{\Delta E_{i}}{RT}\right) \quad I = 1, 2, 3 \quad (T \text{ in } K)$$
Viscosity:

$$\mu = \mu_{\infty} \exp\left(\frac{U}{RT} + \kappa\alpha\right) \quad (T \text{ in } K)$$

Fiber preform

Compressibility equation [3]:
$$e_o = 1.476$$
, $a = 0.491$, $b = 3.745$
 $e = \frac{1 - V_f}{V_f} = -1.5519 \times 10^{-6} \sigma^{fe} + e_o$, $0 \le \sigma^{fe} \le 68707$ Pa
 $= -a \log_{10} (\sigma^{fe}) + b$, $\sigma^{fe} \ge 68707$ Pa

<u>Composite</u>

Density:

Specific heat:

$$\rho_c = \rho_m V_m + \rho_f V_f$$
$$\rho_c C_c = \rho_m V_m C_m + \rho_f V_f C_f$$

Transverse thermal conductivity [8]:

$$\frac{K_{c}^{T}}{K_{m}^{T}} = \left(1 - 2\sqrt{\frac{V_{f}}{\pi}}\right) + \frac{1}{D} \left[\pi - \frac{4}{\sqrt{1 - \left(D^{2}V_{f}/\pi\right)}} \tan^{-1}\frac{\sqrt{1 - \left(D^{2}V_{f}/\pi\right)}}{1 + D\sqrt{V_{f}/\pi}}\right], \quad D = 2\left(\frac{K_{m}^{T}}{K_{f}^{T}} - 1\right)$$

A_l , min ⁻¹	2.101 x 10 ⁹	ρ_f , kg/m ³	1.79×10^3
A_2, \min^{-1}	-2.014 x 10 ⁹	C_f , J/kg ^o C	7.12×10^2
A_3, \min^{-1}	1.960×10^5	K_f^T , W/m°C	26
ΔE_1 , J/mol	8.07×10^4	V_{fo} (3 cm)	0.4038
ΔE_2 , J/mol	7.78×10^4	V_{fo} (5 cm)	0.3907
ΔE_3 , J/mol	5.66 x 10 ⁴	V_b	0.34
H_R , J/g	473.6	$k_b^T / (\boldsymbol{\rho}_b C_b), \mathrm{mm}^2$	0.1
U, J/mol	9.08×10^4	h_b (3 cm), m	0.010516
μ_{∞} , Pa \cdot s	7.93 x 10 ⁻¹⁴	h_b (5 cm), m	0.012385
К	14.1		

Table: Material parameters

Note that the same units are used in the equations.

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