Mingfa Ren, Qi Wang*, Jie Cong and Xin Chang

Study of one-dimensional cure simulation applicable conditions for thick laminates and its comparison with three-dimensional simulation

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Abstract: The comparison of one- and three-dimensional cure simulation of thick thermoset matrix laminates was conducted in this study. The applicable conditions of one-dimensional cure simulation were investigated. The transient heat conduction equation coupled to the cure kinetics was solved numerically using one- and three-dimensional finite element analysis. The evolution of temperature and degree of cure of the laminates during the curing process obtained by the simulation agreed well with the published experimental results. The results indicate that a wider one-dimensional analysis applicable region around the center point will be obtained in the laminate with a higher span-to-thickness ratio and in a less anisotropic material system. In the applicable region, the accuracy of the one-dimensional cure simulation can satisfy the engineering request and save the computational cost. While beyond the region, there is a steep increase in deviation of the one- and three-dimensional simulation results.

Keywords: cure simulation; degree of cure; finite element analysis; one- and three-dimensional analysis; thermoset matrix laminates.

1 Introduction

Advanced composite materials combined the advantages of lightweight, high stiffness, strength and durability, which made them an attractive option to be used in aerospace and other industries [1, 2].

To improve the load-carrying capacity of composite structures, the section thickness of laminates is increased, and thick-section laminated composite structures are being utilized increasingly in design as primary load-carrying components [3, 4]. For example, at the wing root of large aircrafts are several hundreds of plies, whose thickness can be 30–50 mm. The thickness of the central wing jointing area of the Airbus A380 can even be 160 mm. However, the reduction of manufacturing cost and the improvement of quality and reliability of composite structures are two main problems that are needed to be overcome [5], especially for the thick-section composite structures.

During the curing process of the composite structure, the cure cycle applied on the prepregs in an autoclave is the key to initiate an exothermic chemical reaction, which will influence the temperature distribution of the prepregs, and the temperature distribution will determine the service performance of the composite structure [6–10]. High temperature can shorten the curing time, but may result in residual stress and obvious material degradation, while low temperature can lead to unacceptable long curing time [11]. Therefore, to get high-quality and low-cost products, the curing process must be optimized [12].

To improve the process level, the conventional method used is the trial and error test combined with empirical rules of thumb [13]. This method requires not only an extremely long period but also high cost, and it is especially not applicable for thick-section composite structures because the results from one experiment may not be applicable to laminates with different thicknesses. In contrast, the numerical simulation method can predict heat transfer, resin flow, fiber compaction and residual stress development during the curing process in a shorter time and the cost can be greatly reduced [14].

In the past decades, a number of attempts have been made in the numerical simulation of the thermosetting composite curing process by various models with different dimensions. Loos and Springer [15] developed a one-dimensional (1-D) model relating the cure cycle to the thermal, chemical and physical processes to simulate the curing process of a flat plate using the finite difference method. Twardowski et al. [16] compared

*Corresponding author: Qi Wang, Department of Engineering Mechanics, Dalian University of Technology, Dalian 116024, China, Phone: +86 15640947652, e-mail: 499455885@mail.dlut.edu.cn

Mingfa Ren, Jie Cong and Xin Chang: State Key Laboratory of Structural Analysis for Industrial Equipment, Department of Engineering Mechanics, Dalian University of Technology, Dalian 116024, China

the temperature profiles of a thick part predicted by a 1-D computer simulation to the experimental results and investigated the effect of the initial degree of cure and consolidation. Kim and Lee [17] developed an autoclave cure cycle with cooling and reheating steps using finite difference analysis and experiment. Guo et al. [18] conducted a 1-D transient heat transfer analysis during autoclave cure cycle using the finite element model composed of tooling, vacuum bag assembly and laminate. Teplinsky and Gutman [19] conducted a 1-D cure simulation analysis of a plate to investigate process-induced stress and strain development in thick-section polymer matrix composites. The 1-D analysis is based on the assumption that heat transfer in other directions is negligible. Therefore, 1-D analysis has certain limitations, for instance, it cannot apply to the curing process for the complex cross-section structures where a multidimensional heat conduction may happen.

Bogetti and Gillespie [20] developed a 2-D cure simulation and predicted the temperature and degree of cure distributions within an arbitrary cross-sectional geometry using the alternating direction explicit (ADE) finite difference scheme. Telikicherla et al. [21] also considered a 2-D time-dependent heat conduction equation by an ADE finite difference procedure, addressing the effects of heating rate, laminate thickness, bleeder material and convective heating. Blest and Duffy [22] performed a 2-D simulation of resin flow, heat transfer and curing of laminates with varying composite thicknesses using a finite difference scheme on a moving grid. Park and Lee [23] developed a 2-D cure simulation of thick composite structures by the finite element method. They pointed out that simplifying the 3-D problem as a 2-D one may have caused additional error, and a full 3-D cure simulation is needed for more accurate cure simulation.

Oh and Lee [24] analyzed the temperature, degree of cure and resin pressure distributions in thick laminates during the curing process using the 3-D finite element model. Cheung et al. [25] conducted a 1-D heat transfer analysis for a flat plate and a 3-D one for a curve-shaped part using the Galerkin finite element method. Zhu et al. [26] developed a 3-D coupled thermo-chemo-viscoelastic finite element model to simulate heat transfer, curing and residual stress development of an L-shaped composite part. Park et al. [27] developed a 3-D finite element program which can simulate the curing process of composite structures with arbitrary geometry under non-uniform autoclave temperature distribution. Shojaei et al. [28] performed a 3-D process cycle simulation of resin transfer molding (RTM), which consists of filling and curing stages. The computer code provides various information

for process cycles to design the optimum processing conditions.

In most of the previous studies, the curing process simulations were conducted in 1- or 2-D analysis. Especially for the flat plate, most academics considered it as a 1-D heat transfer question. These analyses were under the assumptions of uniform temperature distribution and symmetry in geometry [27]. Therefore, 1-D analysis has the advantages of simplicity and rapidity. However, lack of research mentioned the applicable conditions of 1-D simulations. In fact, the 1-D analysis may not get the accurate temperature and degree of cure distributions when the thickness of the plate is large or the region of interest is close to the plate edge. Three-dimensional analysis can get the results at any positions of the structure with an arbitrary section at the expense of a long simulation time. For some large structures, the 3-D simulation time can be tens of hours, which may make the numerical simulation method lose its superiority.

In this study, 1- and 3-D finite element analyses were developed to simulate the curing process of thick thermosetting composite laminates. The comparison of 1- and 3-D curing simulation results of thick thermosetting laminates with different span-to-thickness ratios and thermal properties was conducted. To improve the computational efficiency of cure simulation and realize the further work of cure process optimization, the applicable conditions of 1-D cure simulation were studied. The relationship of 1-D analysis applicable region and properties of thick thermosetting laminates was investigated.

2 Equations and model

2.1 Thermo-chemical model

The 3-D, transient orthotropic heat transfer equation with an interior heat source can be expressed as:

$$\rho c_p \frac{\partial T}{\partial t} = \frac{\partial}{\partial x_i} \left(k_{ij} \frac{\partial T}{\partial x_j} \right) + \rho V_R H_R \frac{\partial c}{\partial t} \text{ in } V \quad (i, j = 1, 2, 3) \quad (1)$$

where ρ denotes the composite density, c_p the specific heat, T the temperature, t the time, x_i the coordinates, k_{ij} the components of the thermal conductivity tensor and c the degree of cure, which is defined as the ratio of the heat released by the reaction to the ultimate heat of reaction H_R . The last term in Equation (1) corresponds to the internal heat generation associated with the exothermic cure reaction and is described by a cure kinetics model [29]:

$$\frac{\partial c}{\partial t} = g(T, c) \text{ in } V \quad (2)$$

where $g(T, c)$ represents some function of T and c , and takes on specific forms depending on the material system under consideration [30]. In the present study, we use the following cure kinetics model for a graphite/epoxy material [15, 31]:

$$\begin{aligned} \frac{dc}{dt} &= (k_1 + k_2 c)(1 - c)(0.47 - c) & \text{for } (c \leq 0.3) \\ \frac{dc}{dt} &= k_3(1 - c) & \text{for } (c > 0.3) \end{aligned} \quad (3)$$

k_1 , k_2 and k_3 are defined by the Arrhenius rate expressions:

$$\begin{aligned} k_1 &= A_1 \exp(-\Delta E_1 / RT) \\ k_2 &= A_2 \exp(-\Delta E_2 / RT) \\ k_3 &= A_3 \exp(-\Delta E_3 / RT) \end{aligned} \quad (4)$$

where R and T are the universal gas constant and absolute temperature, respectively. A_1 , A_2 and A_3 are the pre-exponential coefficients, and ΔE_1 , ΔE_2 and ΔE_3 are the activation energies.

The temperature and degree of cure fields described by Equations (1) and (2) are subjected to the initial conditions $T = T_0$, $c = 0$ at $t = 0$, as well as the boundary conditions:

$T = T_{\text{applied}}(t)$ on the outer surface and $-K \frac{\partial T}{\partial n} = 0$ on the symmetry surface.

2.2 Finite element model

Before solving the transient coupled thermo-chemical problem, the finite element formulation is required to be derived. Some researchers have solved the coupled governing Equation (1) and Equation (2) iteratively, i.e. the thermal equation first, then the cure kinetics equation, or vice versa. However, the temperature and degree of cure obtained by this method do not exactly correspond to each other, even when the time step is small. Our approach treats both equations with a single method, allowing us to solve for the temperature and degree of cure simultaneously. In the thermo-chemical model, each node has two degrees of freedom: temperature and degree of cure. We choose the same finite element shape functions N for nodal solution vectors T (temperature) and c (degree of cure), and approximate the solutions as:

$$T(t) \approx \mathbf{N}\mathbf{T}(t), \quad c(t) \approx \mathbf{N}\mathbf{c}(t) \quad (5)$$

The finite element formulations can be derived for the heat transfer and thermal cure kinetics equations [32]:

$$\mathbf{C}_T \dot{\mathbf{T}} - \rho H_R \mathbf{C}_c \dot{\mathbf{c}} + (\mathbf{K}_T + \mathbf{K}_h) \mathbf{T} = \mathbf{P}_h \quad (6)$$

$$\mathbf{C}_c \dot{\mathbf{c}} = \mathbf{P}_c \quad (7)$$

where

$$\begin{aligned} \mathbf{C}_T &= \int_{-1}^1 \int_{-1}^1 \int_{-1}^1 \mathbf{N}^T \rho c_p N J d\xi d\eta d\zeta, \quad \mathbf{C}_c = \int_{-1}^1 \int_{-1}^1 \int_{-1}^1 \mathbf{N}^T N J d\xi d\eta d\zeta, \\ \mathbf{K}_T &= \int_{-1}^1 \int_{-1}^1 \int_{-1}^1 \mathbf{B}_{Tc}^T \mathbf{K} \mathbf{B}_{Tc} J d\xi d\eta d\zeta \\ \mathbf{K}_h &= \int_{-1}^1 \int_{-1}^1 \int_{-1}^1 \mathbf{N}^T h N J d\xi d\eta d\zeta, \quad \mathbf{P}_h = \int_{-1}^1 \int_{-1}^1 \int_{-1}^1 \mathbf{N}^T h T_\infty J d\xi d\eta d\zeta, \\ \mathbf{P}_c &= \int_{-1}^1 \int_{-1}^1 \int_{-1}^1 \mathbf{N}^T g(c, T) J d\xi d\eta d\zeta \end{aligned} \quad (8)$$

In the above equations, $\mathbf{K}$ is the 3×3 thermal conductivity matrix and $\mathbf{B}_{Tc}$ is the gradient interpolation matrix.

Because the thermo-chemical problem is a coupled one, Equations (6) and (7) can be combined into a matrix form as:

$$\begin{bmatrix} \mathbf{C}_T & -\rho H_R \mathbf{C}_c \\ 0 & \mathbf{C}_c \end{bmatrix} \begin{Bmatrix} \dot{\mathbf{T}} \\ \dot{\mathbf{c}} \end{Bmatrix} + \begin{bmatrix} \mathbf{K}_T + \mathbf{K}_h & 0 \\ 0 & 0 \end{bmatrix} \begin{Bmatrix} \mathbf{T} \\ \mathbf{c} \end{Bmatrix} = \begin{Bmatrix} \mathbf{P}_h \\ \mathbf{P}_c \end{Bmatrix} \quad (9)$$

For simplicity, Equation (9) can be rewritten in shorthand as:

$$\mathbf{C}\dot{\mathbf{U}} + \mathbf{K}\mathbf{U} = \mathbf{P} \quad (10)$$

in which the coefficient matrices and vectors are:

$$\mathbf{C} = \begin{bmatrix} \mathbf{C}_T & -\rho H_R \mathbf{C}_c \\ 0 & \mathbf{C}_c \end{bmatrix}, \quad \mathbf{K} = \begin{bmatrix} \mathbf{K}_T + \mathbf{K}_h & 0 \\ 0 & 0 \end{bmatrix}, \quad \mathbf{U} = \begin{Bmatrix} \mathbf{T} \\ \mathbf{c} \end{Bmatrix}, \quad \mathbf{P} = \begin{Bmatrix} \mathbf{P}_h \\ \mathbf{P}_c \end{Bmatrix} \quad (11)$$

To solve Equation (10), the time domain is discretized using the Newmark- β method, and $\mathbf{U}(t)$ is approximated as:

$$\mathbf{U}(t) \approx \mathbf{U}(t - \Delta\tau) + \Delta\tau[(1 - \beta)\dot{\mathbf{U}}(t - \Delta\tau) + \beta\dot{\mathbf{U}}(t)] \quad (12)$$

where $\Delta\tau$ is the step time and β is an adjustable parameter chosen between 0 and 1. In this study, β is set to be 0.5 for its unconditional stability and second-order accuracy (Δt^2). Assuming that the specific heat and density of the material are constant during cure, Equation (10) can be discretized as:

$$\left(\frac{\mathbf{C}}{\Delta t} + \beta \mathbf{K}^n \right) \mathbf{U}^n = \left(\frac{\mathbf{C}}{\Delta t} + (1 - \beta) \mathbf{K}^{n-1} \right) \mathbf{U}^{n-1} + \beta \mathbf{P}^n + (1 - \beta) \mathbf{P}^{n-1} \quad (13)$$

where superscripts n and $n - 1$ represent the value at the current and previous time steps, respectively. The distribution of temperature and cure degree in the laminate can be solved iteratively at each time step with the Newton-Raphson scheme.

3 Module verification

Based on the aforementioned equations, user subroutines for 1- and 3-D cure simulation of thermosetting composite structures were developed in the finite element software ANSYS. To verify the accuracy of user subroutines, the curing process of a unidirectional graphite/epoxy composite laminate with dimensions of $15.24 \text{ cm} \times 15.24 \text{ cm}$ and a uniform thickness of 2.54 cm is considered. Because of symmetry, a 3-D finite element model for one eighth of the laminate and a 2-D finite element model (1-D heat transfer) for one fourth were developed. The element types used for the 1- and 3-D finite element models were the 4-node element and the 20-node element, respectively. The part is subjected to a typical cure cycle for which experimental results were available. The cycle involves a 60-min dwell at 116°C and a 120-min dwell at 177°C . The temperature boundary conditions defined by the cure cycle were imposed on the top and bottom plate surfaces, while the other surfaces are adiabatic due to the symmetry. The thermal properties and cure kinetic parameters of the graphite/epoxy are listed in Table 1.

At first, to verify that the finite element model is independent of the mesh size, a mesh convergence test was conducted. The result is given in Figure 1. We see that when mesh size changes from 4 mm to 3 mm , the variation of the result is only 0.2% , so the solution is convergent when the mesh size is 3 mm .

The temperature and degree of cure evolution at the center of the plate are presented in Figures 2 and 3. These results agree well with the experiment in [33]. The

Figure 1: Mesh convergence test.

Figure 2: Temperature profile in the center of the graphite/epoxy laminate.

Figure 3: Degree of cure profile in the center of the graphite/epoxy laminate.

Table 1: Thermal properties and cure kinetic parameters for graphite/epoxy.

Thermal properties		Cure kinetic parameters	
$\rho \text{ (m}^3\text{)}$	1578	$A_1 \text{ (min}^{-1}\text{)}$	2.102×10^9
$C_p \text{ (J/kg} \cdot \text{K)}$	862	$A_2 \text{ (min}^{-1}\text{)}$	-2.104×10^9
$K_{22} = K_{33} \text{ (W/m} \cdot \text{K)}$	0.4135	$A_3 \text{ (min}^{-1}\text{)}$	1.960×10^5
K_{11}/K_{33}	1, 5, 10	$E_1 \text{ (J/mol)}$	8.07×10^4
$H_r \text{ (J/kg)}$	1.986×10^3	$E_2 \text{ (J/mol)}$	7.78×10^4
		$E_3 \text{ (J/mol)}$	5.66×10^4

temperature results emphasize the importance of the exothermic aspect of the chemical reaction. Because of the exothermic cross-linking reaction, two overshoots are observed while the cure degree is aggressively increasing at where the cure temperature is held. The two thermal spikes during the curing of the center point are shown in the Figure 2. There is a 9°C difference compared to the boundary for the first spike and a 30°C difference for the second. There is a transition for the degree of cure profile at the value of 0.3 because of the change in cure kinetic models at the time of 103 min. It is found that a longer time step would result in a less required time in solving and a more coarse result, or vice versa. To ensure the efficiency and accuracy, the time step size was chosen to be 10 s, yielding a total of 1860 steps.

4 Results and discussion

4.1 Example 1

To investigate the difference of the 1- and 3-D cure simulations, an orthogonal graphite/epoxy laminate with dimensions of $200\text{ mm} \times 200\text{ mm} \times 40\text{ mm}$ is established, as shown in Figure 4, in which the span-to-thickness ratio is 5 ($L/D=5$).

Temperature and degree of cure profiles at the center of the laminate obtained by the 1- and 3-D simulations are shown in Figures 5 and 6. In the same time step size and

Figure 4: Model of the orthogonal graphite/epoxy laminate.

Figure 5: Temperature profile of the center point of the laminate.

Figure 6: Degree of cure profile of the center point of the laminate.

mesh size, the terminal times of the 1- and 3-D simulations are 26 and 425 min, respectively.

From the results, obvious discrepancies between the two simulations can be observed. Before the temperature overshoot, the temperature predicted by the 3-D cure simulation is higher than that predicted by the 1-D cure simulation, while the result is converse after the temperature overshoot. This is because in the 3-D analysis, heat conduction is allowed from three directions, while in the 1-D analysis, heat transfer is allowed only from the top and bottom surfaces. The difference of the temperature evolution resulted in the difference of degree of cure, as shown in Figure 6. Therefore, 1-D cure simulation is not accurate enough. However, because of the simplicity and rapidity of the 1-D analysis, the applicable conditions of the 1-D cure simulation need to be studied.

Figure 7 presents the temperature of points on the diagonals of the midfaces of the thick laminates with span-to-thickness ratios ranging from 5 to 10 at the maximum exotherm of the center point which are obtained by both 1- and 3-D cure simulations. It can be concluded that the temperature of the laminate gradually decreased from inside to outside according to the results of the 3-D simulation. The temperature contour of one eighth of the laminate with the span-to-thickness ratio of 10 is shown in Figure 8, which was obtained by the 3-D simulation at the time of maximum exotherm. There is a region around the center point in which the temperature is consistent with the center point from the contour, whereas outside this region, complex temperature gradients appear. The thermal gradient along the thickness at the center section is more severe than that at the side section.

Because only the heat conduction along the thickness is considered in the 1-D simulation, a uniform temperature distribution was obtained by 1-D simulation in the

Figure 7: Temperature of laminates with different span-to-thickness ratios.

Figure 8: Temperature contour of one eighth of the laminate.

Figure 9: Percentage of 1-D analysis applicable region.

same plane. Therefore, the 1-D analysis can meet the accuracy requirement in the regions, and the regions become smaller with the span-to-thickness ratio of the laminates decreasing (increasing of thickness). While beyond the regions, there is a steep increase in the deviation of 1- and 3-D simulation results. Considering the acceptable error range is $\pm 1^\circ\text{C}$, which is the temperature-controlling precision of the composite autoclave, the percentage of the 1-D analysis applicable region is shown in the Figure 9. When the span-to-thickness ratio is lower than 6, the error generated by the 1-D simulation is out of the acceptable error range even at the center of the laminates.

4.2 Example 2

The anisotropy of the composite structure has a significant influence on the difference of the 1-D and 3-D cure simulations. The effect of the anisotropic heat conduction is examined in a $200\text{ mm} \times 200\text{ mm} \times 40\text{ mm}$ orthogonal graphite/epoxy laminate. The typical graphite/epoxy composite K_{11}/K_{33} ratios of 1, 5 and 10 were investigated.

The temperature of the points on the diagonals of the midfaces at the maximum exotherm of the center point (about 124.5 min) is shown in the Figure 10. When the value of K_{33} is invariant, the results obtained by 1-D cure simulation of laminates with K_{11}/K_{33} ratios of 1, 5 and 10 are all the same.

The 3-D cure simulation results demonstrate that high K_{11}/K_{33} ratios accentuate the uneven distribution of temperature fields. In the case of $K_{11}/K_{33} = 1$, the temperature of the points are consistent with the center point within 97.2 mm from the center point, when $K_{11}/K_{33} = 5, 10$, the temperature of the points are consistent with the center

Figure 10: Temperature of laminates with different K_{11}/K_{33} ratios.

point within 70.7 and 44.2 mm, respectively. Similarly, a steep increase in the deviation of the 1- and 3-D simulation results disappeared when the position of the points is beyond the preceding regions. Thus, it can be seen that the anisotropy of the composite increases the difference of the 1-D and 3-D cure simulations. In the laminates with same span-to-thickness ratios, the laminates in the less anisotropic material system will have a wider region in which the 1-D simulation is applicable.

4.3 Example 3

In this numerical example, we performed a cure simulation for a typical composite part, advanced grid-stiffened (AGS) composite structure (as shown in Figure 11). The dimensions of the AGS composite structure is listed in Table 2. The maximum span-to-thickness ratio of the AGS composite structure is 19.4, and the minimum is 8.3.

The material system used for the AGS composite structure is graphite/epoxy, with properties shown in

Figure 11: AGS composite structure.

Table 2: Dimensions for the AGS composite plate.

Dimension parameters	Value/mm
t_1	3
t_2	3
l_1	3
l_2	50

Table 1. Because the AGS composite structure has orthogonal grids, the 3-D finite element model can be established as a periodic unit cell, as shown in Figure 12. Meanwhile, to compare with the 3-D analysis, a 1-D model with the minimum thickness ratio of the AGS composite structure is established. The temperature and degree of cure evolutions of the center point on the interface between the ribs and the skin are shown in Figure 13. As the results show,

Figure 12: Three-dimensional AGS composite structure finite element model.

Figure 13: The temperature and degree of cure evolutions of the AGS composite structure.

the temperature and degree of cure evolutions obtained from the 1- and 3-D simulations were well consistent, which can support the above-mentioned conclusions. However, the total cost simulation time for the 1- and 3-D analyses is 0.8 and 32.3 h, respectively. Consequently, the computational cost can be saved 97% by 1-D cure simulation for the AGS composite structure. Therefore, the optimization design for cure cycle and other process parameters can be conducted by 1-D simulation to get the high-quality products.

5 Conclusions

A user subroutine for simulating the temperature and degree of cure evolution of a thermosetting composite was developed. One- and three-dimensional coupled thermo-chemical cure simulations of composite plates were conducted. The results of the simulations have a good agreement with the published results; thus, the accuracy of the user subroutine is verified.

A comparison of the 1- and 3-D cure simulations of thick orthogonal graphite/epoxy laminates with different span-to-thickness ratios was conducted. The results indicate that there is a region around the center in which the 1-D analysis can meet the accuracy requirement for the laminate with a high span-to-thickness ratio. However, the discrepancy between the 1- and 3-D cure simulations will sharply increase beyond the regions. When the span-to-thickness ratio is lower than 6, the 1-D analysis will produce a considerable error even at the center of the laminate. Additionally, it was found that the anisotropy of the composite increases the difference of the 1-D and 3-D cure simulations, and a wider 1-D analysis applicable region will be obtained in the composite structure with a less anisotropic material system.

The 1-D analysis applicable conditions proposed in this study provide a distinguishable region in which the 1-D analysis has a high precision. Therefore, the computational efficiency of cure simulation can be significantly improved. Thus, this work lays the foundation for the further work of manufacturing process optimization at a low cost and in a short period, finally achieving the goal of getting high-quality, long-lasting and reliable performance products.

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