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Temperature field of thick thermoset composite laminates during cure process

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Abstract

The development of temperature field of thick thermoset matrix laminates manufactured by autoclave vacuum bag process were measured and compared with the numerically calculated results. The finite element formulation of the transient heat transfer problem was carried out for polymeric matrix composite materials from the heat transfer differential equations including internal heat generation produced by exothermic chemical reactions. The finite element analysis software, which was based on the general finite element software package, was developed for numerical simulation of the entire composite process. From the experimental and numerical results, it was found that the measured temperatures profiles were in good agreement with the numerical ones, and conventional cure cycles recommended by prepreg manufacturers for thin laminates should be modified to reduce out-of-plane temperature gradient.

Keywords: C. Finite element analysis; Temperature field

1. Introduction

Thick composite laminates have found several applications in aerospace, military, marine and civil structures. Future increase of the usage of thick composites relies heavily on the successful fabrication of parts with dependable quality and at low cost. Unfortunately, process conditions for thick composites are not well defined. The manufacturer's recommended cycle (MRC) is often inadequate for thick composites because of adverse effects involving large out-of-plane temperature gradient and mid-plane heat generations are neglected.

There have been many studies on the curing of thick thermoset matrix composites. Oh and Lee [1] studied the cure cycle for glass/epoxy composite laminate by 3-dimensional finite element analysis. An optimized cure cycle with the cooling and reheating steps was developed by minimizing the objective function to reduce the temperature overshoot in the composite. Bogetti and Gillespie [2] developed a two-dimensional cure simulation analysis of thick thermoset composites and predicted the temperature and degree of cure distributions within an arbitrary cross-sectional geometry. Ciriscioli et al. [3] measured the temperature, ionic conductivity, and compaction in 16–200 ply thick graphite/epoxy laminates. They also compared the data to the results calculated by the Loos-Springer CURE model. Twardowski et al. [4] compared the experimental temperature profiles of a thick part to the results predicted by a one-dimensional computer simulation, from which the effect of initial degree of cure and consolidation were investigated. Hojjati and Hoa [5] constructed model laws based on dimensionless parameters for cure of thermoset composites and

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predicted the temperature and degree of cure distributions of a thick composite based on the model. Michaud et al. [6,7] predicted the cure behavior of a thick vinyl ester matrix composite manufactured by resin transfer molding and investigated the effect of cure inhibitors and fibers on the cure kinetics. White and Kim [8] developed the stage cure technique for fabricating thick composites and investigated the effect of stage cure on the mode I interlaminar fracture toughness and shear strength. Yi et al. [9] conducted the transient heat transfer finite element analysis by assuming thermal properties such as density, heat capacity, and thermal conductivity as a function of temperature and degree of cure. Kim and Lee [10] developed an autoclave cure cycle with cooling and reheating steps using finite difference analysis and experiment. They showed that the developed cure cycle was effective for the reduction of temperature overshoot. Blest et al. [11] studied the modeling and simulation of resin flow, heat transfer, and the cure of multiplayer thermoset composite laminates during an autoclave processing. They showed the approximate validity of the model by comparing the numerical results with the known experimental data. Joshi et al. [12] presented a procedure to use a general-purpose finite element package for cure modeling and demonstrated the modeling of cure of a thick laminate, a honeycomb sandwich panel, and an I-beam. Marinez [13] used a one-dimensional finite difference analysis and an error function defined by the temperature difference between the target temperature and the laminate temperature to calculate the time-optimal autoclave cure schedule with minimum error. Park and Lee [14] developed a two-dimensional cure simulation by finite element method. They calculated through-the-thickness temperature distributions of composite structures including mandrels. Melnik [15] studied the thermoset composite applied to biochemical engineering. He studied the thermal degredation by heat transfer and cure kinetics. Loos and Springer [16] developed a one-dimensional model to simulation the cure-process of a flat-plate by solving the governing equation using finite difference method. Rai and Pitchumani [17] have used mixed PDE/ODE methods to solve for coupled temperature and cure fields.

In most of the previous studies, the temperature and degree of cure distributions were predicted using one- or two-dimensional finite difference analysis. Only a few literatures studied the temperature distribution by finite element analysis. Although several researches have developed special-purpose numerical software to study the cure process [1–4,6–14], general-purpose numerical software with ability to perform a transient heat transfer analysis can also be used for cure model if appropriate user programs are added to the software [1,12]. Because general-purpose FE package have well developed pre- and post-processors, it is beneficial if they can be used for the cure model. The objective of this study is to gain a fundamental understanding of the cure process unique to thick composite laminates. One-dimensional transient heat transfer finite element analysis during autoclave cure cycle for a 2 cm thick carbon fiber/epoxy laminate is analyzed by commercial finite element software, ANSYS. The simulation of the cure process accounts for thermal and chemical interactions associated with cure. The finite element model composed of the tooling and vacuum bag assembly as well as the laminate is used to investigate the effect of the geometry of the tooling and bagging materials on the temperature profiles.

2. Thermo-chemical model

It is assumed that the convective heat transfer effect by the resin flow is negligible and the resin and fiber are at the same temperature at any specific time. With these assumptions, the one-dimensional model of heat transfer including the governing equation can be expressed as follows [5,9,16,18]:

$$\rho C_p \frac{\partial T}{\partial t} = \frac{\partial}{\partial z}\left(k\frac{\partial T}{\partial z}\right) + \rho H_u \frac{d\alpha}{dt}, \tag{1}$$

$$\frac{d\alpha}{dt} = g(\alpha, T) = K(T)\alpha^m(1-\alpha)^n, \tag{2}$$

$$K(T) = A\exp\left(-\frac{E}{RT}\right), \tag{3}$$

$$m = C_1\exp(-C_2T), \tag{4}$$

$$n = C_3\exp(-C_4T), \tag{5}$$

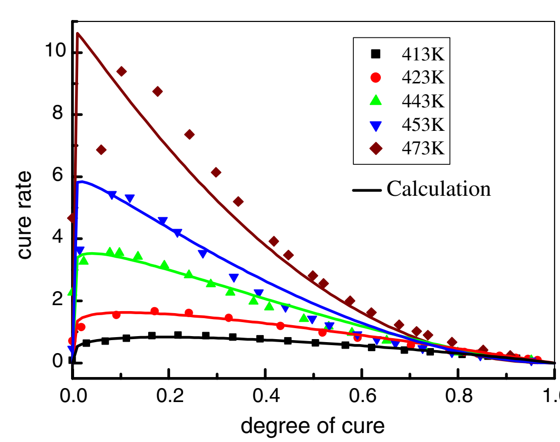

Fig. 1. Rate of heat generation of T300/HD03 prepreg during the isothermal scanning.

Table 1
Cure kinetic parameters of the carbon/epoxy composite

Cure kinetic parameters					
Constant for m	C_1	1.879e − 10	Pre-exponential factor	A (min^{-1})	2.263e7
	C_2	6.06e − 2	Activation energy	E (J/mol)	5.682e4
Constant for n	C_3	1.94e − 3	Heat of reaction	H_u (J/g)	313.84
	C_4	−1.49e − 2			

where T is temperature; ρ, C_p and k are density, specific heat, and thermal conductivity of composite, respectively; $d\alpha/dt$ is cure rate; H_u is heat of reaction generated during dynamic scanning; A is pre-exponential factor; E is activation energy; R is universal gas constant; C_1, C_2, C_3 and C_4 are constant, respectively.

The cure kinetic model of the carbon/epoxy prepreg (T300/HD03) was obtained using the PE DSC-7. The isothermal scanning tests were conducted with constant temperatures of 413, 423, 433, 443, 453, 463, and 473 K. The dynamic scanning tests were also performed at a constant rate of 10 K/min. Fig. 1 shows comparison between the rate of heat generation measured during the isothermal scanning and those calculated by the cure kinetic model of Eq. (2). The developed cure kinetic model agreed well with the experimental results. The cure kinetic parameters of the carbon/epoxy composites are presented in Table 1.

The internal heat generation rate was calculated using Eqs. (2)–(5). The simple rule of mixture was assumed to be valid for the physical and thermal properties of the composite such as density, specific heat, and longitudinal conductivity. The transverse conductivity was calculated using the following Tsai–Halpin model [19]:

$$\frac{k}{k_m} = \frac{k_f + k_m + (k_f - k_m)V_f}{k_f + k_m - (k_f - k_m)V_f}, \tag{6}$$

where k_m is the conductivity of resin; k_f is the fiber conductivity in the transverse direction of fiber; V_f is the fiber volume fraction of the composite.

3. One-dimensional finite element formulation

The temperature profiles in the laminate during the cure process can be obtained through a transient heat transfer analysis including the internal heat generation. The combined thermal and cure problem for a given temperature cycle, $T_\infty(t)$, is referred to as the primal thermo-chemical analysis. We will derive the finite element formulations for the thermal problem and cure problem separately, then combine these two to get the complete finite element equations for the thermo-chemical model. We will solve the temperatures and degree of cure simultaneously and accurately.

The finite element formulation of Eq. (1) is well documented and the reader is referred to the text by the Reddy [20] in this regard. We choose the same finite element shape functions $N(z)$ for nodal solution vectors $\mathbf{T}$ (temperature) and α (degree of cure), and approximate the nodal solutions as

$$T(z,t) \approx N\{z\}\mathbf{T}(\mathbf{t}), \quad \alpha(z,t) \approx N\{z\}\alpha(\mathbf{t}). \tag{7}$$

So the finite element equation for an element are obtained as:

$$[\mathbf{K}_T^e + \mathbf{K}_h^e]\{T^e\} + [\mathbf{M}_T^e]\left\{\frac{\partial \mathbf{T}^e}{\partial t}\right\} = \{\mathbf{F}^e\} + \{\mathbf{Q}^e\}, \tag{8}$$

$$\mathbf{M}_\alpha^e\left\{\frac{d\boldsymbol{\alpha}}{dt}\right\} = \{\mathbf{F}_\alpha^e\} \tag{9}$$

with

$$\mathbf{K}_T^e = \int_z \left(\frac{\partial \mathbf{N}^T}{\partial x} k_x \frac{\partial \mathbf{N}}{\partial x}\right) dz, \quad \mathbf{K}_h^e = \mathbf{N}^T h \mathbf{N},$$
$$\mathbf{M}_T^e = \int_z \mathbf{N}^T \rho C_p \mathbf{N}\, dz, \quad \mathbf{M}_\alpha^e = \int_z \mathbf{N}^T\mathbf{N}\, dz,$$
$$\mathbf{F}^e = \int_z \mathbf{N}^T \rho H_u \frac{d\alpha}{dt}\, dz, \quad \mathbf{F}_\alpha^e = \int_z \mathbf{N}^T \frac{d\alpha}{dt}\, dz, \quad \mathbf{Q}^e = hT_\infty,$$

where $\mathbf{K}_T^e$ and $\mathbf{K}_h^e$ are element conductance and element convection matrices due to conduction and convection, respectively; $\mathbf{M}_T^e$ and $\mathbf{M}_\alpha^e$ are the element capacitance matrices, respectively; $\mathbf{F}^e$, $\mathbf{F}_\alpha^e$ and $\mathbf{Q}^e$ are heat load vectors arising from internal heat generation, cure reaction and surface convection. Since both $\mathbf{F}^e$ and $\mathbf{F}_\alpha^e$ depend on $\mathbf{T}$ and α, Eqs. (8) and (9) comprise a transient coupled problem.

Assembly of the elemental contribution and combined with Eq. (9) results in the global system equations:

$$[\mathbf{K}]U + [\mathbf{M}]\dot{U} = \mathbf{F} \tag{10}$$

with

$$\mathbf{M} = \begin{bmatrix} \mathbf{M_T} & -\rho \mathbf{H_u} \mathbf{M_\alpha} \\ \mathbf{0} & \mathbf{M_\alpha} \end{bmatrix}, \quad \mathbf{K} = \begin{bmatrix} \mathbf{K_T} + \mathbf{K_h} & \mathbf{0} \\ \mathbf{0} & \mathbf{0} \end{bmatrix},$$
$$U = \begin{Bmatrix} \mathbf{T} \\ \boldsymbol{\alpha} \end{Bmatrix}, \quad F = \begin{Bmatrix} \mathbf{Q} \\ \mathbf{F_\alpha} \end{Bmatrix}.$$

To solve Eq. (10), the time domain is discretized using the θ method. The solution at the next time step using:

$$U^n = U^{n-1} + \left[(1-\theta)\dot{U}^{n-1} + \theta\dot{U}^n\right]\Delta t, \tag{11}$$

where $\boldsymbol{U}^n$ and $\boldsymbol{U}^{n-1}$ are the solution at the previous and current time steps respectively; Δt is the time step. The parameter θ is an adjustable parameter varying between 0 and 1 and the algorithm depends on the chosen value of θ . If θ between 0.5 and 1, the integration procedures are known as implicit methods. Eq. (10) can be discretized as

$$\left[\frac{\mathbf{M}}{\Delta t}+\theta\mathbf{K}^n\right]\boldsymbol{U}^n=\left[\frac{\mathbf{M}}{\Delta t}+(1-\theta)\mathbf{K}^{n-1}\right]\boldsymbol{U}^{n-1}+\theta\mathbf{F}^n+(1-\theta)\mathbf{F}^{\mathbf{n-1}}. \quad (12)$$

Since non-linearities occur from thermal properties and internal heat generation which are dependent on temperature and degree of cure, an iterative procedure is necessary to solve the system of equations. The Newton–Raphson algorithm, due to its quadratic convergence characteristics, is employed and the tangent stiffness matrix is updated at each iteration.

4. Experiment

Experiments were conducted to verify the validation of simulation as well as to investigate the effectiveness of the convectional cure cycle recommended by the prepreg manufacturer. The unidirectional carbon/epoxy prepreg used in this work was T300/HD03. The laminate was stacked to the dimension of 300 mm × 300 mm × 20 mm. After the stacked prepreg was paced on the tool plate (thickness = 15 mm) covered by the release film, the caul plate (thickness = 5 mm) and dams (thickness = 25 mm) covered by the release film were placed on the top and four edges of the laminate. Then, the assembly was bagged with a standard nylon bagging film and cured in an autoclave using the convectional cure cycle. The typical lay-up of autoclave-cured composites scheme and experimental apparatus are shown in Figs. 2 and 3, respectively. The individual material properties are shown in Table 2.

The convectional cure cycle is composed of two stages: first stage for consolidation and second stage for full cure. In the first stage, the autoclave temperature is increased to 130 °C with the rate of 2 °C/min and kept at this temperature for 60 min. During the first stage, the excess resin is squeezed out of the laminate. In the second stage, the temperature is increased to 170 °C, which is the cure temperature of the resin, with the same heat rate and maintained at 170 °C for 180 min to complete the cure. The thermal contact resistance was assumed to be negligible, and the convective heat transfer coefficient of 70 W/m^2. K was used between the autoclave air and vacuum bag [1].

In order to investigate the variation of temperature inside the laminate during the cure process, three thermocouples were placed at the 40-ply above the bottom, center of laminate, and 40-ply below the top of the laminate. The location of the thermocouples is shown in Fig. 2.

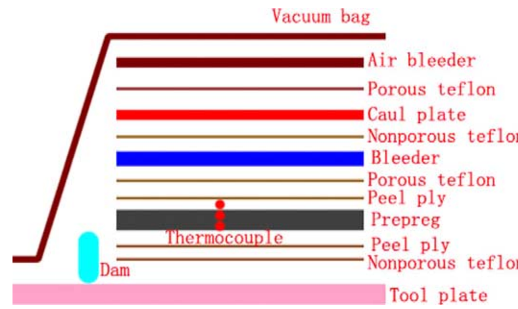

Fig. 2. The typical lay-up of autoclave-cured composites.

Fig. 3. The experimental apparatus.

Table 2
Physical and thermal properties of the used materials [21]

Properties	Value	Properties	Value
Density of resin (g/cm^3)	1.25	Conductivity of Teflon film (W/m K)	0.40
Density of fiber (g/cm^3)	1.80	Conductivity of bag (W/m K)	0.24
Density of aluminum (g/cm^3)	2.72	Conductivity of bleeder (W/m K)	0.07
Density of teflon film (g/cm^3)	2.2	Specific heat of resin (J/g K)	1.260
Density of bag (g/cm^3)	1.14	Specific heat of fiber (J/g K)	0.712
Density of bleeder (g/cm^3)	0.26	Specific heat of aluminum (J/g K)	0.903
Conductivity of resin (W/m K)	0.24	Specific heat of Teflon film (J/g K)	1.05
Conductivity of fiber (W/m K)	2.51	Specific heat of bag (J/g K)	1.67
Conductivity of aluminum (W/m K)	220	Specific heat of bleeder (J/g K)	1.35

5. Results and discussions

Fig. 4 shows that temperature profiles through the thickness are not symmetric along the center of the laminate. This is due to the fact that the thickness of the tool plate is different from that of the caul plate. Furthermore, thermal conductivities of bleeder materials are smaller than those of composites. Increasing the thickness of bleeder is expected to significantly alter the temperature distribution in laminates. This also found by Oh and Lee [1].

The temperature profiles at the three points in alminates were obtained through the experiment and compared with the numerical simulation results. Fig. 5 shows the comparison of the temperature profiles obtained by the simulation and experiment, in which the predicted temperature profiles were in good agreement with the experimental results. A temperature overshoot at the center of laminate was observed from the experimentals results. The maximum temperatures at the midpoint of center section was 191.7 °C. From Fig. 5, it was found that the resin degradation would occur because the maximum temperature of the laminate exceeded 182 °C which was the glass transition temperature of the fully cured resin [22]. At beginning, the maximum temperature occurs on the outside and the minimum temperature is at the center of the plate. When exothermic heat is generated from chemical reactions inside the composite, temperatures at the center increase and become higher than the autoclave temperature. Higher temperatures also result in a faster cure.

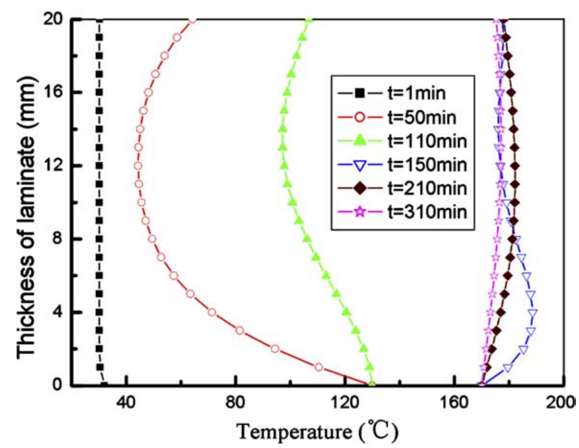

Fig. 4. Temperature distribution.

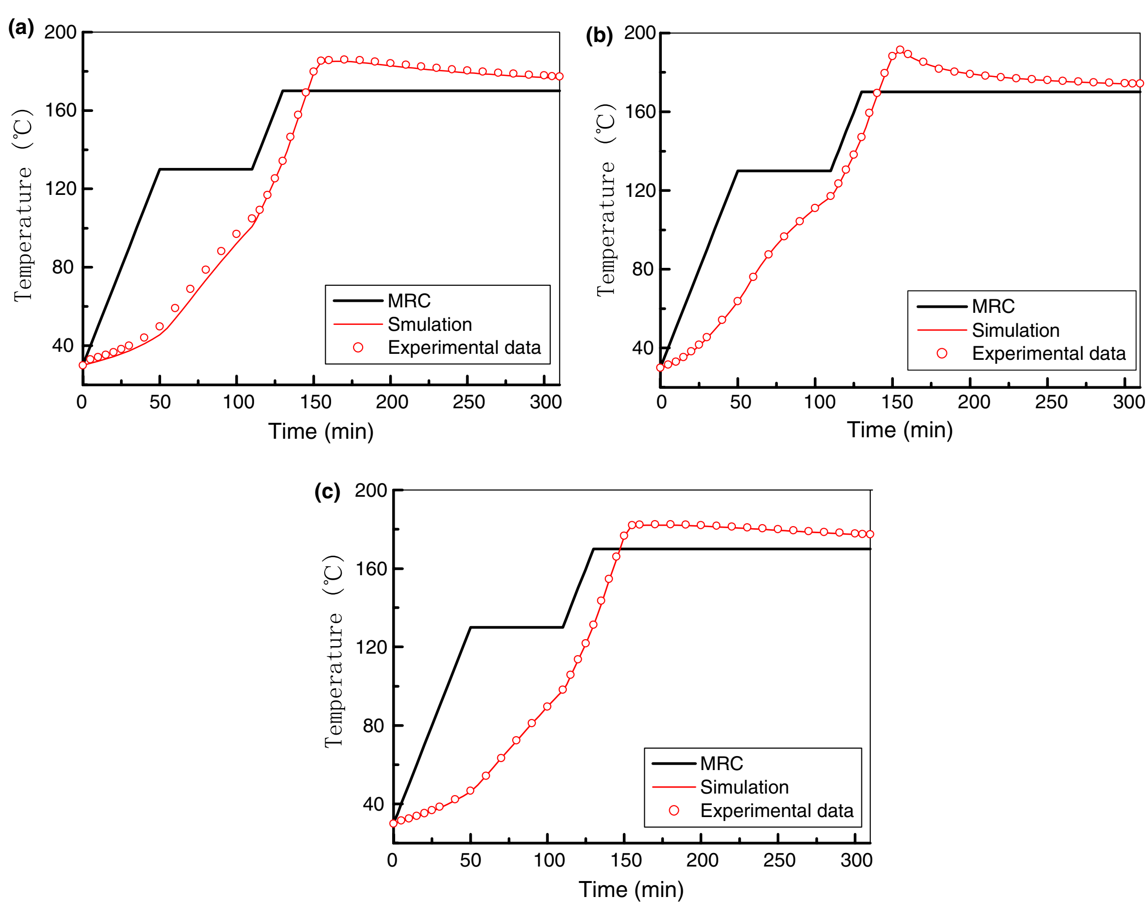

Fig. 5. Temperature profiles of the carbon/epoxy laminate cure by the convectional cure cycle: (a) bottom temperature; (b) middle temperature; (c) top temperature.

The profiles of degree of cure at the center section are presented in Fig. 6. At the initial stage of curing, a lower degree of cure was observed at the midpoint of the center section due to the low thermal conductivity of laminate; however, the degree of cure increased abruptly due to the internal exothermic reaction as the autoclave temperature increased.

The temperature profiles of the laminate are influenced by the convective heat transfer coefficients between the vacuum bag and the autoclave air, the thickness of bleeder, and the geometry of mold assembly such as thickness of the tooling and caul plate. In order to investigate the effect of the convective heat transfer coefficient on the temperature profile, the one-dimensional FEM analysis was performed varying the coefficients. The temperature profiles at the center of laminate are presented in Fig. 7. As the coefficient decreased, the temperature overshoot decreased while the time to reach the maximum temperature and the temperature difference from the autoclave air increased. The low convective heat transfer makes the internal exothermic reaction occur at a slow rate, which reduces the temperature overshoot. On the contrary, the high convective heat transfer coefficient promotes the fast exothermic reaction, which increases the temperature overshoot.

Fig. 8 shows the maximum temperature at the center of the laminate and corresponding times for various thickness of the mold assembly such as the tool plate and caul plate. As the thickness of mold assembly increased, the maximum temperature decreased a little, while the corresponding time increased a little because the thickness increase of the mold assembly increased the thermal resistance between the laminate and autoclave air, which caused the slow heating of the laminate. From Figs. 7 and 8, it was found that the convective heat transfer coefficient between the vacuum bag and autoclave air had a great influence on the temperature profiles in the laminate, while the thickness of mold assembly seldom affected them.

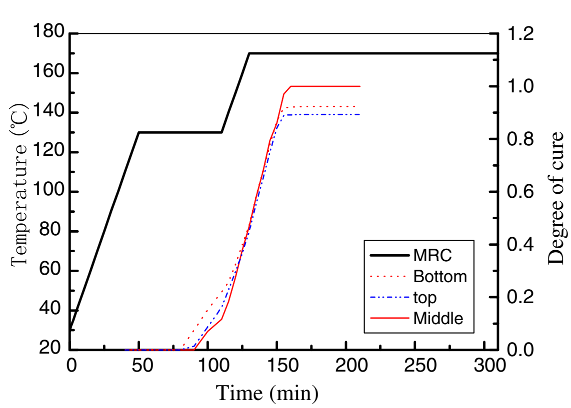

Fig. 6. Degree of cure profiles of the carbon/epoxy laminate cured by the conventional cure cycle.

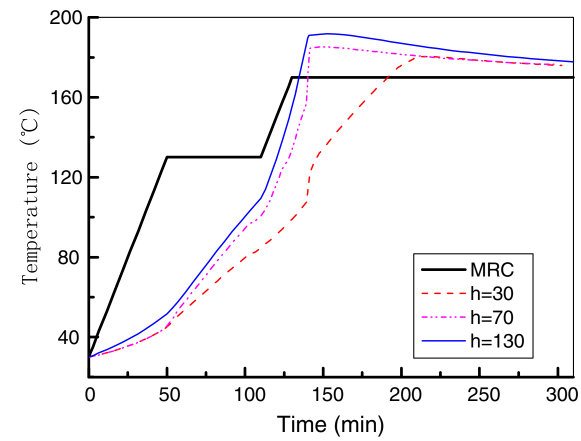

Fig. 7. Effect of convective heat transfer coefficients (unit: W/m^2 K) on the midpoint temperature.

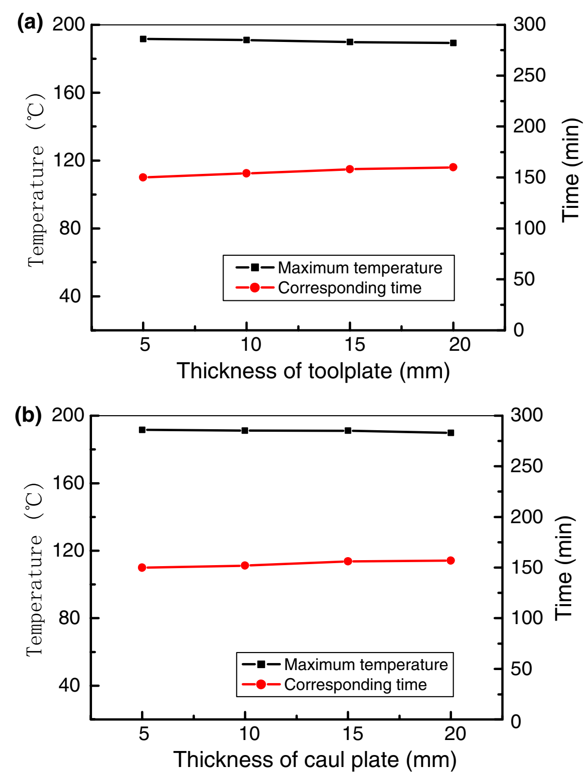

Fig. 8. Maximum temperatures and corresponding times for various (a) thickness of tool plate; (b) thickness of caul plate.

6. Conclusions

Non-linear transient heat transfer finite element procedures and codes are developed to simulate cure processes of polymer matrix composites. The temperature and degree of cure distributions in thick carbon fiber/epoxy laminates during process were calculated using the one-dimensional finite element analysis, and it was found that the predicted temperature profiles agreed well with the experimental ones. The manufacturer's recommended cycle produced a large temperature overshoot at the midpoint of the laminate because of exothermic reaction and low thermal conductivity of matrix. This can lead to matrix degradation, nonuniform cure and consolidation, and residual stress. The temperature profiles in the laminate were much affected by the bleeder materials and the convective heat transfer coefficient between the vacuum bag and autoclave air, while they were little influenced by the thickness of mold assembly. To perform more accurate simulations, temperature and cure dependent thermal properties, and heat transport due to resin flow should include in the finite element model.

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