Study on Cure Deformation of Metal/CFRP Laminated Composites

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Abstract. During the curing process of composite materials, the differences of the heat transfer ability and mechanical properties of material in various directions result in residual stresses and curing deformation in the composite after curing. Curing deformation has adverse effect on the mechanical performance of composites. The development of internal stress during curing may cause matrix cracking, delamination, warpage and rebound deformation of composites. In this paper, numerical simulation is used to predict the curing deformation of components, thereby reducing production costs and improving production efficiency as well. According to the curing kinetic equation, the HETVAL subroutine was firstly developed to calculate the temperature field and the curing degree field of the curing process. UMAT and UEXPAN subroutines were then applied to solve the mechanical constants and thermal strain and curing shrinkage strain during the curing process. After the proposed model was verified, the deformation of the metal composite co-cured components was studied. It was found that when the thickness ratio of metal to CFRP is one to one the composite gains the greatest geometrical deviation after curing.

1. Introduction
Composite materials have been widely used in many fields due to their high specific strength, large specific modulus, fatigue resistance and corrosion resistance. However, due to the thermal expansion and contraction effect of the material itself during the curing process of the composite material, the chemical shrinkage effect of the matrix and the thermal expansion coefficient of the composite material and the mould do not match, which inevitably causes deformation after moulding, especially large size and complicated structure member. Curing deformation can cause assembly stress, reduce structural strength, shorten fatigue life, and even lead to component scrapping. In order to ensure the quality of component manufacturing while minimizing cost, it is important to use numerical simulation to predict curing deformation. In the early research, Cowley [1] approximated the curing deformation of the orthogonally laid strip laminates to the cylindrical shape, and applied the classical laminate theory to estimate the curvature deformation of the cylinder. Yi [2] et al. used nonlinear finite element method to calculate the residual stress during the cooling process of composites. Kappel [3] reported a preliminary experimental study on the mechanical interaction and spring deformation of unidirectional, crossed and quasi-isotropic laminates with C-type and L-type specimens. Zhang [4] proposed a modified viscoelastic constitutive model for predicting curing residual stress of polymer matrix composites. Ersoy [5] predicted the spring of AS4/8552 composite C-section parts. Hörberg [6]
introduced the experimental results and analysis of the influence of laminate thickness on the elastic deformation and shape deformation of thermosetting composite L-shaped steel. Brauner [7] presented a simulation method for analysis of induced deformation of composite multi-segment flaps.

This paper aims to study the curing process and the deformation of composite materials by simulation. Using the material customization function in the finite element software ABAQUS, a subroutine for calculating the curing exotherm and curing degree as well as the mechanical constant and strain was developed. Then, the correctness of the model is verified by comparison with the examples given in the literature. Finally, the model was used to study the geometrical deformation of metal/CFRP composites.

2. Theoretical basis of numerical simulation

2.1. Thermal-chemical theory
The curing process of the thermosetting resin matrix composites is a simultaneous physical and chemical process of heat conduction and curing reaction, the essence is a transient heat conduction problem with a nonlinear internal heat source. Wherein, the internal heat source is derived from the chemical reaction exotherm of the resin matrix. In this paper, a three-dimensional heat conduction model is used to calculate the temperature distribution, and the curing reaction kinetic equation is introduced to calculate the internal heat source.

\[
\frac{\partial T}{\partial t} = \rho_c \frac{\partial}{\partial x} \left( k_{xx} \frac{\partial T}{\partial x} \right) + \frac{\partial}{\partial y} \left( k_{yy} \frac{\partial T}{\partial y} \right) + \frac{\partial}{\partial z} \left( k_{zz} \frac{\partial T}{\partial z} \right) + q
\]

where \(\rho_p\) is the density of composite material; \(c_p\) is the specific heat capacity of the composite; \(k_{xx}, k_{yy}, k_{zz}\) are the thermal conductivity of the composite in the x, y and z directions respectively; \(t\) is the curing time; \(T\) is the transient temperature at the current moment; \(q\) represents the heat generation rate of the curing reaction, which can be expressed as:

\[
q = \rho_r (1 - V_f) H_r \frac{d\theta}{dt}
\]

where \(\theta\) is the degree of cure; \(\rho_r\) is the density of the resin; \(V_f\) is the volume fraction of the fiber; \(H_r\) is the total heat released by the resin per unit mass in the curing process; \(\frac{d\theta}{dt}\) represents the rate of curing reaction and the specific form depends on the type of resin. In this paper, using the improvement equation of the curing mechanics of polyester resin proposed by Johnston [8] is as follows:

\[
\frac{d\theta}{dt} = \frac{K \theta^m (1 - \theta)^n}{1 + e^{C \theta_{CO} + \theta_{CT} T}}
\]

where \(A\) is the frequency factor; \(\Delta E\) is the activation energy; \(C, \theta_{CO}, \theta_{CT}\) are the diffusion constant; \(R\) is the gas constant; \(m\) and \(n\) are the reaction series.

2.2. Determination of mechanical properties of resin
During the curing process, the fiber hardly changed in volume during the whole process, and the resin undergoes a liquid-gel state-glass state, and the mechanical properties vary greatly. This paper uses Johnstonl's improved model to describe the change in mechanical properties of the resin during curing.
\[
E_r = E^0_r, (T^* < T^*_{c1})
\]
\[
E_r = E^0_r + \frac{T^* - T^*_{c1}}{T^*_{c2} - T^*_{c1}} (E^\infty_r - E^0_r), (T^* < T^*_{c2})
\]
\[
E_r = E^\infty_r, (T^* > T^*_{c2})
\]

where \( E_r \) and \( \nu_r \) are the instantaneous modulus of elasticity and poisson’s ratio of resin respectively; \( E^0_r \) is the elastic modulus of resin before curing; \( E^\infty_r \) is the elastic modulus of fully cured resin; \( \nu^\infty_r \) is the poisson’s ratio of fully cured resin; \( T^*_{c1}, T^*_{c2} \) are constants, indicating the beginning and end of glass transition; \( T^* \) is the difference between resin temperature and instantaneous glass transition temperature; \( T^0_g \) is the glass transition temperature before curing; \( a_{fg} \) is a constant, determined by experiment; \( \bar{\theta} \) is the degree of cure.

2.3. Curing shrinkage strain

During the curing process, the volume shrinkage of the resin is also one of the reason for the composite deformation to cause residual stress, which is related to the polymerization reaction of the resin under the temperature load. Assuming that the dimensions of the cube micro-elements in the three directions are \( l_1, l_2, l_3 \) and the variation are \( \Delta l_1, \Delta l_2, \Delta l_3 \), the volume change of the micro-element is:

\[
\Delta V = l_1 \Delta l_2 \Delta l_3 + l_2 \Delta l_3 \Delta l_1 + l_3 \Delta l_1 \Delta l_2 + l_1 \Delta l_2 \Delta l_3 + l_2 \Delta l_1 \Delta l_3 + l_3 \Delta l_1 \Delta l_2 + \Delta l_1 \Delta l_2 \Delta l_3
\]

Volume change rate is:

\[
\Delta \nu = \frac{\Delta V}{V} = \epsilon_1 + \epsilon_2 + \epsilon_3 + \epsilon_r \epsilon_2 + \epsilon_r \epsilon_3 + \epsilon_r \epsilon_2 \epsilon_3
\]

Since the resin is isotropic, the shrinkage strain of the resin is:

\[
\epsilon^m_{sh} = \sqrt{1 + \Delta \nu} - 1
\]

The chemical shrinkage strain of the three main directions of the composite material can be calculated by the following formula:

\[
\begin{align*}
\epsilon^c_1 &= 0 \\
\epsilon^c_2 &= \epsilon^c_3 = \left(\sqrt{1 + \Delta \nu} - 1\right) \left(1 - V_f\right) \left(1 + \nu_r\right)
\end{align*}
\]

where \( V_f \) is the fiber volume content; \( \nu_r \) is the resin Poisson's ratio. Since the 1 direction is constrained by the fiber and the strain is small, it is considered that the contraction strain in the 1 direction is 0.

2.4. Thermal strain

During the cooling phase of the curing process, thermal deformation occurs inside the laminate due to the inconsistent thermal expansion coefficients of the composite material in all directions. The formula for calculating the thermal strain of a composite material is:

\[
\begin{align*}
\epsilon_1^T &= \beta_1 \Delta T \\
\epsilon_2^T &= \epsilon_3^T = \beta_2 \Delta T = \beta_3 \Delta T
\end{align*}
\]
where $\beta_1$, $\beta_2$, $\beta_3$ are coefficients of thermal expansion in the direction of the composite material 1, 2 and 3; $\Delta T$ is the temperature difference.

### 3. Numerical simulation

In order to verify the algorithm, a laminated plate of AS4/8552 material is used to simulate the curing process. The laminate dimensions are shown in figure 1, where $L=300\text{mm}$, $W=30\text{mm}$, $B=1\text{mm}$. Single layer thickness is $0.125\text{mm}$ and layup sequence is $[0_4/90_4]$. The thermochemical parameters of AS4/8552 materials and the mechanical properties are shown in reference [9].

![Figure 1. Dimension diagram of laminate](image)

#### 3.1. Temperature and degree of cure

In this paper, the ABAQUS three-dimensional finite element simulation analysis software is used to simulate the temperature field and curing degree field of the laminate. At the same time, the HETVAL subroutine is used to calculate the curing exotherm of the resin matrix, and the temperature data is transmitted to ABAQUS for update. Finally, the curve of temperature and curing degree changing with time in the curing process of the center point of the laminates is obtained, and compared with the results in the literature, as shown in figure 2.

![Figure 2. Comparison curve of temperature and curing degree at the center point of the laminate](image)

(a)Comparison curve of temperature with time (b)Comparison curve of curing degree with time

From the figure 2, the settlement result of this paper is basically consistent with the temperature and curing degree change curve calculated in literature [11], in which the maximum relative error of temperature is 0.9% and the maximum relative error of curing degree is 1.2%, within an acceptable range. Therefore, the program and calculation result in this paper are correct.

#### 3.2. Curing deformation

Based on the calculation results of temperature and curing degree, curing deformation of laminates is simulated. According to the experimental results in literature [10] and the calculation results in literature [11] show that according to the $[0_4/90_4]$ laid the curing deformation of unsymmetrical laminated is near to the cylindrical shape, with the curvature of the cylindrical to represent the...
deformation degree of laminated plates. The curvature calculation method as shown in figure 3, where L is half of the chord length of the deformed cylinder, and U is the maximum deviation displacement in the Z direction.

![Figure 3. Diagram of curvature calculation](image)

The formula for calculating the cylinder curvature is:

$$t = \frac{1}{R} = \frac{2U}{L^2 + U^2}$$  \hspace{1cm} (13)

Figure 4 is a deformation diagram of the asymmetrically laminate obtained after curing, and the curvature of the cylinder is calculated and compared with the curvatures in [10] and [11], as shown in table 1.

![Table 1. Comparison of the calculation results in this paper with the literature](image)

|                  | t / m$^{-1}$ | error/% |
|------------------|--------------|---------|
| Experimental results in reference [10] | 4.07         | 0       |
| Calculation results in reference [11]  | 4.37         | +7.37   |
| The calculation results in this paper  | 3.98         | -2.2    |

The error between the calculation result and the experiment is 2.2%, which verifies the correctness of the simulation method and the program. Figure 5 shows the displacement change in the Z direction during the curing of the laminate. It can be seen from figure 5 that the laminate has two obvious deformations during the curing process. The first deformation is caused by the curing shrinkage of the resin, and the second deformation is caused by the thermal expansion and contraction caused by the temperature drop of the laminate. At about 9000s, the displacement has a negative value, that is, the laminate has a downward bend. This is because the gel point of the selected material is 0.651. When
the degree of cure reaches 0.651, the temperature is still rising. At this time, the phenomenon of curing shrinkage and thermal expansion occurs at the same time. The deformation caused by thermal expansion is greater than the deformation of the curing shrinkage, resulting in deformation.

![Displacement vs Time Graph](image)

**Figure 5.** The displacement of the center point of the laminate in the Z direction

4. **Deformation analysis of co-curing of metal and composite laminates.**

The fiber metal laminate is co-cured based on the epoxy resin and the metal. During the curing process, different materials are bonded to each other, and no subsequent bonding process is needed. This simplifies the process and ensures that pure composite materials will not lose very important strength and stiffness when their properties are damaged. AS4/8552 composite materials and aluminum are co-cured to study the deformation results of dimension and layup sequence.

4.1 **Finite element model**

The dimensions of the metal aluminum and composite materials are all based on 300 mm × 30 mm × 1 mm, and the finite element model is shown in figure 6. The ABAQUS is used to establish a three-dimensional finite element model. The thermal conduction unit DC3D8 is used to simulate the temperature field and the curing degree field. The calculation results are imported into the stress analysis model, and the three-dimensional solid element C3D8R is used to simulate the residual stress and curing deformation. On this basis, the deformation laws of metal and composite materials with thickness ratios of 3 mm, 2 mm and 1 mm, that is (metal thickness: composite thickness) of 3:1,2:1,1:1,1:2,1:3 and composite lamination angles of 0°, 45° and 90° are studied respectively.

![Co-curing Model of Metals and Composites](image)

**Figure 6.** Co-curing Model of Metals and Composites

4.2 **Result analysis**

The simulation of the curing process of the metal-composite with different thickness ratios is carried out, and the displacement of the Z direction in the three cases of the ply angles of 0°, 45° and 90° is obtained, as shown in figure 7.
Figure 7. Comparison of displacement in the Z direction with different thickness ratios

It can be seen from the figure that the deformation amounts corresponding to different thickness ratios are different. Within the three ply-angles, the amount of curing deformation shows a certain regularity. When the thickness ratio of the metal to the composite material is 1:1, that is, the thickness of the metal and the composite material is the same, the deformation amount of the laminate in the Z direction is the largest. Keeping the thickness of the composite material constant and increasing the thickness of the metal, the corresponding deformation amount is gradually reduced. Under the same conditions, the thickness of the metal is kept constant, and when the thickness of the composite is increased, the corresponding deformation amount also tends to decrease. At the same time, it can be seen that increasing the thickness of the metal is less than increasing the thickness of the composite during the curing process. This is because the metal has only thermal expansion and contraction during the curing process, and the coefficient of thermal expansion is larger than that of the composite. The chemical reaction does not occur in the whole process, and the recovery ability after deformation is strong. Therefore, in the manufacture of the laminate, appropriately increasing the content of the metal can effectively reduce the deformation of the member after the completion of the curing.

5. Conclusion
(1) Based on the time-varying characteristics of the mechanical parameters of composite materials, a three-dimensional finite element calculation model of composite materials is established by using mathematical models including thermochemical model, resin curing shrinkage model and residual stress model, which can be used to simulate the curing process of composite materials.
(2) In comparison with the results of the references, it is proved that the algorithm developed for calculating the curing process of the composite material is reasonable and effective.
(3) Based on the algorithm proposed in this paper, the metal composite co-cured components are simulated and proved that when the thickness of metal and composite materials is the same, the deformation after curing is the largest. Appropriate increase of metal thickness can reduce deformation to a certain extent.

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