

A THREE-DIMENSIONAL FINITE ELEMENT ANALYSIS OF PROCESS-INDUCED RESIDUAL STRESS IN RESIN TRANSFER MOLDING PROCESS

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Abstract

A three-dimensional finite element analysis of curing and process induced residual stress in Resin Transfer Molding (RTM) process is presented_o The finite element method (FEM) was employed to solve the coupled equations involved in the transient heat transfer and the cure kinetics of the resin and the distributions of internal temperature and cure degree of the composite at any instant time are obtained. The self-consistent field micro-mechanics model was used to predict the cure dependent composite mechanical properties. Thermal expansion and cure shrinkage were included in the analysis. The thermo-elastic mechanical governing equations were solved with the use of the incremental stress-strain relationship based FEM and the residual stress development was predicted. The present results were validated by the comparisons with the pertinent literature. The numerical example of a half cylinder was presented. The results show that it is necessary to carry out the three dimensional analysis due to the complex distributions of temperatures, cure degrees and process-induced stress for thick parts, which can be predicted at any point within composite structures in the present analysis.

1 Introduction

Determination of cure cycle and processinduced residual stress are two of the most critical issues in composite manufacturing process. The poor design of cure cycles may make the residual stress significantly develop during the process, which can have a great impact on the performance of composite structures. Process-induced residual stress can reduce both ultimate strength and fatigue life, and is more important to composite parts with strict tolerance requirements. The numerical simulation of process is a effective tool for reducing process costs and improving part quality.

The finite element method is widely used to simulate curing process for the sake of its adaptation to solve the complex shape problems ^[1-4]. These simulations of curing process can predict the distributions of temperatures and degree of cure within the composite materials. The residual stress analysis is necessary to carried out if one wants to evaluate how these gradients of temperature and degree of cure influence on the quality of parts. Chen ^[5] used a visco-elastic micro-mechanical model to study the effect of the cooling rate on the curing process induced residual stress. Andrew Johnston^[6] proposed a plane strain finite element model for simulation of the process-induced deformation during autoclave processing. Tarsha-Kurdi ^[7] investigated the influence of autoclave pressure on curing stresses in carbon/epoxy laminates and it showed the initial size of laminates and autoclave pressure can significantly affect the residual deformation.

The resin volume shrinkage is another important mechanism contributions to processinduced stress besides thermal expansion mismatch. The effective mechanical properties of resin can change drastically associated with the cross-link polymerization reaction. Therefore, thermal and chemical strains and the cure-dependent material properties should be included in the analysis of residual stress. Bogetti^[8,9] simulated the curing process and analyzed the curing process-induced stress for autoclave thick section parts. It pointed out that the complex spatially varying thermal and degree of cure gradients can lead to residual stress development greatly. White^[10,11] employed twodimensional simulation and visco-elastic model to study the curing process and residual stess. Teplinsky ^[12] combined the one-dimensional simulation with incremental laminate theory to predict the process-induced stress. ABQUS software was used to analyze the residual stress during autoclave process for thick section woven fabric composite parts by Huang^[13].

Although a quite amount of studies were made to model compression molding process, little is aimed to model process-induced residual stress in transfer molding (RTM) resin process. Golestanian^[14] stated that he first used the curedependent mechanical properties in predicting residual stresses in RTM process. However, his model is based on shell element, which neglect the variation of temperature in the direction of thickness. It may be successful in predicting residual stresses in thin laminates. The temperature, degree of cure and residual stress can change from point to point within composite materials for thick section parts. It is necessary to develop three-dimensional model to achieve the spatially varying distributions of temperature and degree of cure and evaluate the residual stresses at any point.

This paper is focused on developing threedimensional finite element program to simulate curing process and analyze process-induced stress within the complex shape thick parts. A threedimensional finite element formulation is proposed to avoid limitations of one- and two-dimensional analysis. The self-consistent field micro-mechanics model is used to predict cure dependent mechanical properties. The cure simulation and residual analysis are performed and results are compared with the relative literature.

2 Approach

2.1 Cure simulation

The thermal curing process of resin matrix composites is a themo-chemo coupled process. The resin is taken as not flowing in the stage of cuing and the convection thermal conduction is neglected. Thus the thermal conduction equation with chemical reaction as written in Eq.(1) can be used to describe a full three dimensional cure process^[4]:

$$\rho_c C_{pc} \frac{\partial T}{\partial t} = \nabla (k_c \nabla T) + (1 - f) q_r^{\bullet}$$
(1)

where $\rho_c \, \, \, \, C_{pc}$ are density, specific heat and k_c is thermal conductivity tensor, respectively. They can be determined according to mixture law.

$$\rho_{c} = f\rho_{f} + (1 - f)\rho_{r}$$

$$c_{pc} = \frac{f\rho_{f}c_{pf} + (1 - f)\rho_{r}c_{pr}}{\rho_{c}}$$
(2)

$$k_c = \frac{k_f k_r \rho_c}{f \rho_f k_f + (1 - f) \rho_r k_r}$$

where f stands for volume fraction of fiber, subscription r stands for resin, f stands for fiber, c stands for composite $_{\circ}$ The internal thermal resource •

 q_r is the heat reaction of resin, described as Eq.(3):

$$\mathbf{q}_r = \rho_r H_r \frac{d\alpha}{dt} \tag{3}$$

where ρ_r is density of resin, H_r is total reaction heat of unit mass resin when resin finishes the curing reaction. $d\alpha/dt$ is cure rate of resin, α is degree of cure

The cure kinetics of resin in this paper are selected from references $^{[2, 11]}$ and are listed as Eq.(4) :

$$\frac{da}{dt} = k_0 \exp(-\Delta E_c / RT) \alpha^m (1 - \alpha^n) \quad (4.1)$$
$$\frac{d\alpha}{dt} = (k_1 + k_2 \alpha^m) (1 - \alpha^n) \quad (4.2)$$

where : $k_1 = A_1 \exp(-\Delta E_1/RT)$

$$k_2 = A_2 \exp(-\Delta E_2/RT)$$

where R universal gas constant, T is temperature, $k_0,A1,A2, \Delta E_c$, ΔE_1 , ΔE_2 are experimental constants.

A variational principle is applied to Eq. (1) and after the virtual temperature (δ T) is introduced and volume integration is taken over the domain, the Eq.(1) is rewritten as Eq.(5):

$$\int_{V} (\rho_{c} C_{pc} \frac{\partial T}{\partial t}) \delta T dV = \int_{V} (\nabla(k_{c} \nabla T))$$
(5)

$$+(1-f)q_r)\delta TdV$$

The eight node three-dimensional elements is used to discretized Eq.(5). Temperature (T) at any point within an element is interpolated as in Eq.(6):

$$T = \sum_{i=1}^{8} N_i T_i \tag{6}$$

where Ni and Ti are shape functions and the nodal temperature at ith node, respectively.

The final discretized governing equation can be expressed as below:

$$CT + KT = F \tag{7}$$

where C is the thermal capacitance matrix, T is the nodal temperature vector, K is thermal conductivity matrix, F is thermal load vector. They are expressed as Eq.(8):

$$C = \sum_{e} \int_{V^{e}} \rho_{c} c_{cP} N_{i} N_{j} dV$$

$$K = \sum_{e} \int_{V^{e}} (\nabla N_{i} k_{c} \nabla N_{j}) dV \qquad (8)$$

$$F = \sum_{e} \int_{V^{e}} N_{i} (1 - f) \dot{q}_{r} dV$$

$$\dot{T} = \frac{\partial T}{\partial t}$$

The above Eq.(8) can be solved using the direct time integration method. The thermal load F from chemical reaction of resin is directly related to the degree of cure in the resin. The degree of cure at the time step (n+1), α_{n+1} can be approximated as Eq.(9):

$$\alpha_{n+1} = \alpha_n + \left(\frac{d\alpha}{dt}\right)_n \Delta t \tag{9}$$

where α_n and $(d\alpha/dt)_n$ are degree of cure and the rate of cure at the time step (n).

The Eq.(1) can be solved with the use of the formula above at each time step under the corresponding boundary conditions. The size of time increment must sufficiently small for accurate cure simulation, and is taken as 10 s in the present work, which gives a converged solution.

2.2 Material model

2.2.1 Cure dependent resin modulus

The modulus of resin is assumed to follow a simple rule of mixture as shown in Eq.(10) ^[9].

$$E_r = (1 - \alpha_{\text{mod}})E_r^0 + \alpha_{\text{mod}}E_r^\infty$$
(10)

$$\alpha_{\rm mod} = \frac{\alpha - \alpha_{gel}}{\alpha_{end} - \alpha_{gel}} \tag{11}$$

where E_r^0 and E_r^∞ are the assumed fully uncured and fully cured temperature dependent resin modulus.

The simple expression is used since it was shown to offer an good representation of modulus for the resin considered in this study^[11]. The present model assumes the poisson ratio and the fiber properties are constant during cure. The instantaneous resin shear modulus during cure is determined based on the isotropic material relation.

$$G_r = \frac{E_r}{2(1+\nu_r)} \tag{12}$$

2.2.2 Resin volumetric shrinkage model

Resin shrinkage occurs during cure and provide an important source of internal loading. Assuming a uniform strain contraction for all principal strain components, the incremental isotropic resin shrinkage strain of a unit volume element, $\Delta \varepsilon^{sh}$, and the incremental resin volumetric shrinkage have a relationship given by Eq.(13).

$$\Delta \varepsilon^{sh} = (\sqrt[3]{1 + \Delta V_r}) - 1 \tag{13}$$

A given incremental change in the degree of cure, $\Delta \alpha$, and the associated incremental change in specific volume of resin, ΔV_r , can be related to the total specific volume shrinkage of the completely cured resin, V_{sh}^T , as Eq.(14):

$$\Delta V_r = \Delta \alpha V_{sh}^T \tag{14}$$

The cure shrinkage strain in the resin during cure is the cumulative sum of all the incremental contributions. The fiber itself is assumed not to undergo any chemical contraction during cure.

2.2.3 Effective elastic modulus of composite unit cell

The composite mechanical properties strongly depend on the fiber and resin constituent properties, and fiber volume fraction. The self-consistent field micromechanical model is widely used to compute the instantaneous mechanical properties for fiber reinforced composites^[9]. unidirectional Huang^[14] used TEXCAD model to predict the effective unit cell modulus. We adopt the similar method in this paper. At first the composite unit cell is classified into N sorts of unidirectional reinforced composite (if necessary, the resin is also treated as one sort of such composite). Then the self-consistent model is employed to obtain effective mechanical properties of Nth sort composite. Finally, the effective of unit cell is taken to be the superposition of total N sorts of composite mechanical properties on the basis of theirselves spatial directions.

$$[C_{eff}] = \sum_{m=1}^{N} (f_m [T]_m^T [C_m] [T]_m)$$
(15)

where $[C_{eff}]$ is effective stiffness matrix of unit cell, f_m is fiber volume fraction of mth sort composite, $[T]_m$ is the coordinate transfer matrix of stress and strain between the local coordinate system of of mth sort composite and the global unit cell coordinate system.

2.2.4 Effective thermal expansion coefficients and shrinkage strain of composites

The thermal expansion and shrinkage stains are also dependent on the fiber and resin constituent properties, and fiber volume fraction. A simple model on the basis of a rule of mixture is used to predict the effective thermal expansion coefficients and shrinkage strain of composites^[9].

$$\varepsilon_{1} = \frac{\varepsilon_{1f} E_{1f} f + \varepsilon_{1r} E_{1r} (1 - f)}{E_{1f} f + E_{1r} (1 - f)}$$
(16)

$$\varepsilon_{2} = (\varepsilon_{2f} + v_{12f}\varepsilon_{1f})f + (\varepsilon_{2r} + v_{12r}\varepsilon_{1r})(1-f)$$
$$- \left[v_{12f}f + v_{12r}(1-f)\left[\frac{\varepsilon_{1f}E_{1f}f + \varepsilon_{1r}E_{1r}(1-f)}{E_{1f}f + E_{1r}(1-f)}\right] \quad (17)$$

where subscription 1stand for longitudinal direction, 2 stands for transverse direction. When the effective thermal expansion coefficient are evaluated, the formula $\alpha_1 = \varepsilon_1, \alpha_2 = \varepsilon_2$ is taken, and when the effective shrinkage strains are evaluated, the formula $\varepsilon_1^{sh} = \varepsilon_1, \varepsilon_2^{sh} = \varepsilon_2$ is taken,

The fiber is assumed to be zero shrinkage strain in this study.

2.3 Final Paper Format

2.3 Process induced residual stress

The cure simulation is carried out to yield the temperature and degree of cure distributions within the composite parts in a single time increment before the stress and deformation are calculated. The instantaneous effective material properties and the resin shrinkage load are computed according to these distributions as above discussed. These results are taken as the input parameters for residual stress calculations at the current time step.

The total incremental strain $\Delta \mathcal{E}$ is given by

$$\Delta \varepsilon = \Delta \varepsilon \,^{e} + \Delta \varepsilon \,^{th} + \Delta \varepsilon \,^{sh} \quad (18)$$

where \mathcal{E}^{e} is strain induced by mechanical load. $\Delta \mathcal{E}^{th}$

The thermal strain $\Delta \varepsilon^{th}$ can be expressed as

$$\Delta \varepsilon^{th} = \alpha \cdot \Delta T \tag{19}$$

The incremental form of stress and strain relationship is give by

$$\left[\Delta\sigma\right] = \left[C_{eff}\right] \left\{ \Delta\varepsilon^{e}\right\} + \left[\Delta\varepsilon^{th}\right] + \left[\Delta\varepsilon^{sh}\right\} \quad (20)$$

The Eq.(20) is solved with the use of the finite element method at each time step. The total strain can be obtained taking the cumulative sum of incremental strain of each time step. The details can be found elsewhere [9,,13].

3 Numerical examples

3.1 Example one

The first example is selected for Bogetti^[8,9] to validate the present program. Material system is about glass/polyester. Table 1 gives the composite thermal properties and Table 2 gives the cure kinetic parameters. The characteristic values of polyester resin during cure are seen in Table 3 and the mechanical properties of glass and polyester are listed as Table 6. The example consists of a unidirectional laminate with the dimensions of with $0.1524 \times 0.1524 \times 0.0254m$. The model of cure kinetic is as shown in Eq.(4.1).

Table 1 Thermal properties of Glass/Polyester for two-dimensional example

$\rho_c kg/m^3$	C _{pc} J/(W℃)	$\frac{K_{c22}=K_{c33}}{w/(m^{\circ}C)}$	K _{c11} /K _{c33}
1890	1260	0.2163	2

Table 2 Cure kinetic parameters of Glass/Polyester for two-dimensional example

Glass/1 ofyester for two-dimensional example				
m	n	k0(s-1)	$\Delta \operatorname{Ec}(J/\operatorname{mol})$	Hr(J/kg)
0.524	1.476	6.1667×1020	1.674×105	77500

The same boundary conditions are used in the example, namely the mold temperature is applied at the top and bottom surface of the part and the isolate heat condition is applied at the lateral surfaces. The temperature history at the centroid of the part is as shown in Fig.1. The results agree well with the Bogetti's. The present predicted temperature peak is 126.9 oC, whereas the Bogetti's is 126 oC or so. Sinc e the exact cure cycle curve cannot be obtained, the inflexion of the time in the cure cycle curve may be a little different from its original value of the Bogetti's. The error leads to that the time when the temperature peak occurs is 168 minute (Bogetti's is 164 minute) for the present simulations.



Fig.1 The history of temperature of example one



Fig.2 Comparisons between the present analysis with the Bogetti's results ^[15] for example one.

The in-plane transverse stress for three different volume shrinkage rates 6%, 3%, 1% and 0% are shown in Fig.2. A little difference between the present solutions and the Bogetti's is observed, especially near the mid-plane of the part. The maximum error between the present solutions and the Bogetti's is %8. The reason is that Bogetti used the laminated theory, while the three-dimensional finite elements are used in the present analysis. The significant self-equilibrating stresses remain after complete cure. The magnitude of the resin volumetric shrinkage strongly affects the stress development. These results are agreement with Bogetti's.

4.2 Example two

Material system is about glass/epoxy, and fiber volume fraction is 60%. The effective unit cell is shown as in Fig.3. The simple unit cell used here is for its very small thickness of 0.18mm. The third example is a half cylinder with a height 0.1m, an inner radius 0.0035 and an outer radius 0.005. A total volumetric shrinkage of resin is assumed to be



Fig.3 The effect unit cell of composite materials for half cylinder example

5%.Table 4 gives the composite thermal properties and Table 5 gives the cure kinetic parameters. The characteristic values of epoxy resin during cure are seen in Table 3 and the mechanical properties of glass and epoxy are listed as Table 6. The model of cure kinetic is as shown in Eq.(4.2).

Table 3 Resin characteristic elastic modulus during cure

-				0			
	Properties		Polye	Polyester		Epoxy	
-	$E_{m}^{0}(MPa)$		2.75	2.757		7	
	$E_{m}^{\infty}(MPa)$ 2.757×103 3.447		$3.447 \times$	103			
-	Table 4 Thermal properties of Glass/Epoxy for						
half cylinder example							
m	n	A_1	A_2	ΔE_1	ΔE_2	Hr	
		(s^{-1})	(s^{-1})	(J/mol)	(J/mol)	(J/kg)	
0.95	0.84	1.75×10^{5}	3.88×10^5	6.51×10^4	540×10^4	459224	

Table 5 Cure kinetic parameters of Glass /Epoxy for half cylinder example

	ρ	Cp	K
	kg/m ³	J/(W℃)	w/(m℃)
Glass	2560	712.35	0.137
Epoxy	1150	8210	8.673

Table 6 Fiber and resin constituent mechanical properties

Properties	Glass	Polyester	Epoxy
$E_1 (MPa)$	7.308×10^4	Eq.(6)	Eq.(6)
$E_2 (MPa)$	7.308×10^{4}	Eq.(6)	Eq.(6)
ν_{12}	0.22	0.40	0.35
V ₁₃	0.22	0.40	0.35
ν_{23}	0.22	0.40	0.35
G ₁₂ (MPa)	2.992×10^{4}	Eq.(6)	Eq.(6)
G ₁₃ (MPa)	2.992×10^4	Eq.(6)	Eq.(6)
$G_{23} (MPa)$	2.992×10^4	Eq.(6)	Eq.(6)
$\alpha_1(1/°c)$	5.04×10 ⁻⁶	7.20×10^{-5}	5.76×10 ⁻⁵
$\alpha_2(1/°c)$	5.04×10 ⁻⁶	7.20×10 ⁻⁵	5.76×10 ⁻⁵





Fig.5 The distributions of temperatures at time 30 min and 38 min.



Fig 6 The history of reidual stresses for half cylinder example

In residual stress analysis, perfect bonding between the laminate and the mould wall is assumed. Though the assumption still is a quite open issue, here we follow the previous convention to take it. The process-induced residual stresses are solved as discuss above at each time step. The total stress is the cumulative sum of each step solutions.

The developments of the temperature are shown in Figs.4 It can be seen that temperatures at central locations are higher than those at the positions close to the surface. It resulted in a temperature difference of about 24.2oC in the thickness direction. The significant temperature overshoots are observed from 36 min. The two different distributions of the temperature at the time of 30 and 38 min are shown respectively in Fig.5.It can be found that the regions with highest temperature tends to offset towards the center as the resin cures. It also indicates that the fraction of residual stress before the cooldown is about 50 percent of its maximum value. The stress σ xx is greatest among the three axial directions on the



Fig 7 The deformation of half cylinder

global coordinate system, as seen in Fig.6. The deformed shape is plotted as in Fig.7. The maximum displacements along three different directions, Ux,Uy,Uz are 1.11,0.545 and 0.59 mm respectively.

4 Conclusions

A three-dimensional finite element analysis of curing and process induced residual stress in Resin Transfer Molding (RTM) process is presented_o The finite element method (FEM) was employed to solve the coupled equations involved in the transient heat transfer and the cure kinetics of the resin and the distributions of internal temperature and cure degree within the composite at any instant time are obtained. A comparison with the Bogetti's results validated the present simulations. The self-consistent field micromechanics model was used to predict the cure dependent composite mechanical properties. Thermal expansion and cure shrinkage were included in the analysis. The thermo-elastic mechanical governing equations were solved with the use of the incremental

stress-strain relationship based FEM and the residual stress development was predicted. The numerical examples including a half cylinder were presented. The results show that it is necessary to carry out the three dimensional analysis due to the complex distributions of temperatures, cure degrees and process-induced stress for thick parts, which can be predicted at any point within composite structures in the present analysis.

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References:

- [1] Mi Ae Choe, Mi Hye Lee, Jaeeon Chang, Threedimensional simulation of the curing step in the resin transfer molding process. Polymer Composites, 1999;20(4):543-552.
- [2] Xiao Lin Liu, I.G.Crouch, Y.C.Lam, Simulation of heat transfer and cure in pultrusion with a general-purpose finite element package. Composites Science and Technology, 2000;60:857-864.
- [3] Hoon Cheol Park, Nam Seo Goo, Kyung Jae Min, Kwang Joon Yoon, Three-dimensional cure simulation of composite structures by the finite element method. Composite Structure, 2003;62:51-57.
- [4] Hoon Cheol Park, SUNG W.Lee, Cure simulation of thick composite structures using the finite element method. Journal of Composite Materials, 2001;35(3):188-201.
- [5] Yu Chen, Zihui Xia, Fernand Ellyin, Evolution of residual stresses induced during curing processing using a viscoelastic micromechanical model. Journal of Composite Materials, 2001;35(6):522-542
- [6] Andrew Johnston, Reza Vaziri, Anoush Poursartip, A plane strain model for processinduced deformation of laminated composite structures. Journal of Composite Materials, 2001,35(16):1435-1469.
- [7] Kheir Edddine Tarsha-Kurdi, Philippe Olivier, Thermoviscoelastic analysis of residual curing stresses and the influence of autoclave pressure on these stresses in carbon/epoxy laminates. Composites Science and Technology, 2002;62:559-565.
- [8] Travis A. Bogetti, Joha W.Gillespie,Jr. Twodimesional cure simulation of thick thermosetting composites. Journal of Composite Materials,1991;25(3):239-273

- [9] Travis A. Bogetti, Joha W. Gillespie, Jr. Processinduced stress and deformation in thick-section thermoset composite laminates. Journal of Composite Materials, 1992;26(5):626-660
- [10] S.R.White, H.T.Hahn, Process modeling of composite materials: residual stress development during cure.Part I. Model formulation, Journal of Composite Materials, 1992;26(16):2402-2422
- S.R.White, H.T.Hahn, Process modeling of composite materials: residual stress development during cure.Part II. Experimental Validation, Journal of Composite Materials, 1992;26(16):2423-2453
- [12] Sh.Teplinsky, E.M.Gutman, Computer simulation of process induced stress and strain development during cure of thick-section thermosetting composites, Computational Materials Science, 1996;6,:71-76
- [13] Xiaogang Huang, Joha W.Gillespie Jr, Travis Bogetti, Process induced stress for woven fabric thick section composite structures, Composite Structures, 2000;49:303-312
- [14] Hossein Golestania, A. Sherif El-Gizawy. Modeling of Process Induced Residual Stresses in Resin Transfer Modeled Composites with Woven Fiber Mats, Journal of Composite materials, 2001;35(1):1513-1528