Two-Dimensional Heat Transfer Through Complex-Shaped Composite Materials – Part I: Literature Review and Model Development

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The energy equation describing two-dimensional transient heat transfer in a composite medium is solved numerically in order to predict the temperature profiles inside the curing composite material as a function of time. The model includes a heat generation term due to exothermic chemical reactions, several types of boundary conditions, thermal properties dependent on both temperature and direction, and dynamic boundary movement due to compaction. This paper addresses the physical and chemical phenomena associated with composite materials processing. A literature review of the published mathematical models is presented. The proposed model is then developed, and the discretization and solution techniques are outlined.

Nomenclature

$A_1, A_2, \ldots, A_5$	= coefficients of interior node transformed equation
a^{ij}	= components of covariant metric vector
C	= heat capacity, J/kg-K
f	= analytical function
$(g)^{1/2}$	= Jacobian
H_R	= heat of reaction, J/kg
k_{11}	= 11-component of thermal conductivity tensor, W/m-K
k_{22}	= 22-component of thermal conductivity tensor, W/m-K
P	= control function for ξ coordinate
Q	= control function for η coordinate
Q'	= heat generated per unit volume, W/m^3
T	= temperature, K
TOLD	= temperature at previous time step, K
t	= time, s
W	= relaxation parameter
x,y	= Cartesian coordinates
α	= degree of cure of extent of reaction
δ_{1j}, δ_{2j}	= decay factor for ξ and η point attraction
η	= coordinate of computational field
κ_{1j}, κ_{2j}	= line attraction amplitude for ξ and η, respectively
ξ	= computational field coordinate
ρ	= density, kg/m^3
σ_{1i}, σ_{2i}	= decay factor for ξ and η line attraction
ϕ_{1i}, ϕ_{2i}	= line attraction amplitude for ξ and η, respectively
$(\partial T/\partial t)_c$	= temperature rate in computational domain, K/s
$(\partial T/\partial t)_p$	= temperature rate in physical domain, K/s
11, 12	= subscripts designating components of thermal conductivity tensor
ξ,η,x,y	= subscripts designating partial derivatives with respect to ξ, η, x, and y, respectively

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Introduction

ADVANCED composites consist of reinforcing fibers such as glass, carbon, graphite, or metals in a polymeric, metallic, or ceramic matrix. The type and composition of the constituents and the processing procedure is varied in order to tailor the composite characteristics. These composite characteristics or properties are superior to those of the constituents and may include a high strength-to-density ratio, a high stiffness-to-density ratio, high or low conductivity, and resistance to corrosion, fatigue, and stress rupture.

The processing methods vary depending on the type of reinforcement and matrix. For thermoplastic materials, the techniques include injection molding, compression molding, and cold stamping. Thermoset processing may employ contact molding, matched die molding, injection molding, pultrusion, filament winding, or vacuum bag autoclave molding. Although this paper focuses on the autoclave curing of thermosetting composites, the strategy is applicable to many manufacturing processes for composite materials.

Autoclave Curing of Composite Materials

Autoclave curing typically consists of laying up resin impregnated fiber sheets or layers on a (forming) tool and covering it with a vacuum bag; a typical layup is shown in Fig. 1. The entire assembly is inserted into an autoclave (a pressure cooker). The autoclave temperature and pressure (cure cycle) are controlled in order to compact the composite, manage voids, and initiate the chemical curing reaction.

Composite cure cycles are usually developed by trial and error.[1] A cure cycle is chosen based on the constituent material properties, dimensions, geometry, desired composite properties, and processing experience; the part is then fabricated, and the composite properties are determined. The cure cycle is then modified, and the process is repeated experimentally until the desired composite properties are achieved. This procedure is time consuming and costly and rarely results in optimum operating parameters.

Process modeling can reduce the dependence on the experimental trial and error procedure, provide direction to process optimization, allow for the study of the effect of process variables on product properties, and become an integral part of the closed-loop control of the process. The model is limited by the numerical and physical approximations and the availability of property data.

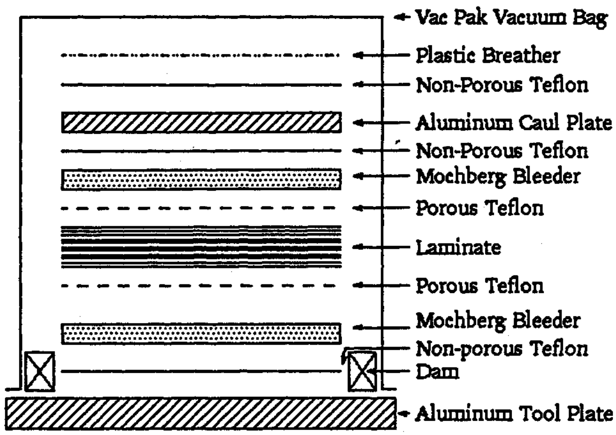

Fig. 1 Typical autoclave layup.

Problem Statement

The objective of this paper is to address the modeling of the curing of thermosetting composite materials. The existing heat transfer models are described. A model is then developed in order to predict temperature and degree of cure distributions during the processing of composite materials. Although this work deals mainly with heat transfer, the various phenomena are described here to present an overall picture of the process. The strategy presented for heat transfer can be extended to model other phenomena.[2]

The physical and chemical changes that occur during the curing process include heat transfer, resin flow, chemical reactions, physical thickness changes, and void formation. The uncured composite prepreg material as well as the bagging materials offer a resistance to the conduction of heat supplied to cure the resin. A heat balance is then used to describe the temperature distribution within the material. Accurate internal temperature knowledge is required to determine the autoclave curing conditions.

Under the influence of pressure gradients, the resin flows out of the composite into an absorbent material called a bleeder. The resin viscosity and reaction kinetics are dependent on the prevailing temperature and degree of cure. The cure reactions for most thermoset resins are exothermic and act as an internal heat source contributing to the energy balance. Endothermic reactions can be included as heat sinks.

The applied pressure and chemical reactions contribute to changes in the physical shape of the part being cured. During compaction, the boundaries of the composite are not stationary and their movement may not be uniform. Within the prepreg being cured, vapors are generated either as a product of the chemical reactions or by vaporization from saturated liquids. Air and contaminants entrapped in the plies during layup is also a source of vapor. Small bubbles are nucleated and then coalesce to form voids. Diffusion, pressure gradients, and thermal expansion govern void size, distribution, and movement.

The various phenomena described above need to be considered in order to predict certain cure parameters essential to controlling the quality of the composite. The temperature is critical because all properties and phenomena are dependent on it. The kinetics are needed to predict the degree of cure, an indication of the extent of polymerization. The resin flow/deformation must be considered to predict the compaction rate (affects heat transfer), pressure distribution and developed stresses. The void model is used to predict void formation and transport, the starting point for void management.

Review of the Literature

A thorough review of the application of numerical techniques to the modeling of transport processing is available.[3] The literature addressing heat transfer in composites will be discussed. Literature dealing with heat transfer through complex geometry, moving boundaries, or variable properties is summarized.

Loos and Springer[4] developed a one-dimensional thermochemical model to predict the temperature and degree of cure profiles during the autoclave processing of composite materials. This model does not include convection heat transfer at the boundaries. The rule of mixtures that calculates a volume-average property of the composite from constituents properties is used to determine the density and heat capacity. The thermal conductivity is approximated by the correlation of Springer and Tsai.[5] The resulting heat balance is solved using the finite difference technique. This model agrees with one-dimensional experimental results. The bleeder is the only bagging material considered; other bagging materials, such as the breather and nylon bag, could offer considerable resistance to heat transfer.

Mallow et al.[6] developed heat transfer and kinetic models to predict temperature and degree of cure distributions during composite materials curing. The resistance from various tool and bagging materials is considered. An empirical relationship for the convective heat transfer coefficient in the autoclave is used. The solution technique is the finite element method with a lumped mass approach for the constant thermal properties in each element. This code can accommodate three-dimensional heat transfer but cannot accommodate complex shapes. Model predictions agree with experimental data in shapes consistent with the assumptions.

Servais et al.[7] developed a code to predict the temperature profile through a laminate stack during the press cure cycle. The one-dimensional, transient energy equation with a temperature-dependent heat source and constant thermal properties is solved using an implicit finite difference technique. A weight averaging technique is used to combine the various thin materials and to obtain weight-averaged properties. The results show agreement with experimentally determined temperature profiles for large area, thin materials cured in presses.

Soni and Pagano[8] modeled the processing of carbon-carbon composites in order to predict temperature and degree of graphitization (equivalent to degree of cure in thermoset composite materials) in three-dimensional systems. This model uses the finite element method, is restricted to carbon-carbon composites, and is not sufficiently general to apply to other materials.

Ma et al.[9] developed a model to simulate steady-state two-dimensional heat transfer and the curing reactions of unsaturated polyesters in a rectangular die of a pultruder. Bulk thermophysical properties, which are weight-averaged and dependent on the fraction of resin cured, are used. The boundary conditions are in the form of specified temperatures or adiabatic walls. The energy equation is solved using the finite difference approach in rectangular coordinates. Although results of the code are reasonable, experimental verification is not available.

Smith and Guceri[10] presented a model of the heat transfer during laser processing of thermoplastic composites. The solution technique is the body-fitted coordinate (BFC) system generation coupled with the finite difference method. No experimental verification was presented.

Uchikawa and Takeda[11] used the boundary-fitted coordinate transformation to predict temperature distributions during the solidification of steel castings of arbitrary shapes. Constant density and thermal conductivity were used, and the latent heat of freezing was accounted for by a change in the apparent specific heat. No boundary movement was considered in the analysis.

McWorter and Sadd[12] used the BFC method to study steady-state heat conduction in anisotropic material of arbitrary shapes. Constant but directional thermal conductivities were used. The central finite difference method was utilized to discretize the energy equation for a steady-state stationary boundary system.

Goldman and Kao[13] studied the transient two-dimensional heat conduction using the rectangular and cylindrical BFC method for systems with constant properties and stationary boundaries.

Hsu et al.[14] presented a method to treat heat transfer problems in systems with constant thermal properties and a moving boundary using coordinate transformation and a control-volume approach.

Lin[15] and Meric[16] studied the transient one-dimensional heat conduction in a composite slab with a temperature-dependent thermal conductivity. Both models use the finite element method.

The models described in the literature typically are limited to one-dimensional conditions or simple geometries; others are constrained by constant constituent properties or fixed boundaries. The objective of this work is to extend the existing models to accommodate two-dimensional transient heat transfer through complex-shaped heterogeneous systems of variable properties, moving boundaries, and various boundary conditions. Historically, a moving complex-shaped boundary problem required a (complex) finite element solution method; however, the body-fitted coordinate system scheme is consistent with the (simpler) finite difference solution method.

Moving Boundaries and Grids

The finite difference methods are the most widely used techniques for the solution of fluid flow and heat transfer problems. This is mainly due to the ease of understanding, development, and programming of the technique. Recently, however, as problems with complex geometries are being addressed, the finite difference methods are used less frequently because of their inability to accommodate complex boundaries. Large errors arise if the boundaries do not pass through the grid points requiring interpolation in most sensitive regions. The errors introduced at the boundary can propagate into the field and result in inaccurate solutions. The finite element methods circumvent the problem mentioned above at the expense of simplicity and ease of programming. A method for generating a grid that conforms to the physical boundaries was introduced by Chu[17] and extended by Thompson et al.[18] This body-fitted coordinate system generation technique has eliminated the problem introduced by complex boundaries and allowed the return to the finite difference techniques.

The grid generation technique consists of specifying the boundary points that are then used to generate the interior points by one of two methods: the first consists of numerically solving a set of partial differential equations,[19] and the second makes use of algebraic interpolation.[20] The differential equations used in the grid generation include parabolic, hyperbolic, and elliptic equations. Algebraic methods are used to generate the grid points in the interior of the field by interpolating the values at the boundaries.

An adaptive grid is one that is moving with time; the grid points are allowed to adjust in order to provide a more accurate solution. Adaptive gridding allows the movement of these points as the solution progresses to accommodate improved resolution in regions of large gradients. Adaption can also be used in systems where physical changes, such as moving boundaries, occur. A constant coordinate line is coincident with the moving boundary at all times, and the remainder of the grid adapts to that change. Even though the grid points are dynamic in the physical space, all calculations are made on the the fixed computational grid without interpolation. A review of the adaptive grid generation methods has been documented by Thompson.[21]

Approach

The development of the model consists of discretizing the complex domain under study using the BFC system generation method. This grid generation method results in a rectangular computational domain with unity spacing. There is a one-to-one correspondence between every point in the computational domain and its image in the physical domain (knowing the solution in the computational field results in knowing it in the physical field). Then, the transient energy equation describing heat transfer in nonhomogeneous composite materials with variable properties and moving boundaries and accounting for heat generation by chemical reactions is derived. The energy equation and boundary conditions are then transformed to the computational domain and discretized using implicit finite differences. The difference equations are assembled and solved in the computational domain to obtain the temperature and degree of cure distributions as a function of time. The development of each of the procedures outlined follows.

Grid Generation

The grid generation technique used is based on the method of Thompson et al.[22] The method consists of specifying the boundary points and generating the interior points of the grid by solving the following system of Poisson equations in two dimensions

$$\begin{cases}\xi_{xx} + \xi_{yy} = P(\xi,\eta) \\ \eta_{xx} + \eta_{yy} = Q(\xi,\eta)\end{cases} \tag{1}$$

Transforming the above equation such that x and y become the dependent variables

$$(x_\eta^2 + y_\eta^2)x_{\xi\xi} - 2(x_\xi x_\eta + y_\xi y_\eta)x_{\xi\eta} + (x_\xi^2 + y_\xi^2)x_{\eta\eta} + g(Px_\xi + Qx_\eta) = 0 \tag{2a}$$

$$(x_\eta^2 + y_\eta^2)y_{\xi\xi} - 2(x_\xi x_\eta + y_\xi y_\eta)y_{\xi\eta} + (x_\xi^2 + y_\xi^2)y_{\eta\eta} + g(Py_\xi + Qy_\eta) = 0 \tag{2b}$$

Equation (2) shows the nonlinearity of the differential equation system. An iterative solution method is then necessary. The functions P and Q provide control of point location and coordinate line spacing for ξ and η, respectively. If P and Q are set to zero, the above equation reduces to the Laplace system that would result in the smoothest grid. Thompson et al.[22] have used the following functions for P and Q:

$$P(\xi,\eta) = -\sum_{i=1}^{n} \phi_{1i}\, \text{sign}(\xi - \xi_i) \exp(-\sigma_{1i}|\xi - \xi_i|) - \sum_{j=1}^{m} \kappa_{1j}\, \text{sign}(\xi - \xi_j) \exp[-\delta_{1j}\sqrt{(\xi-\xi_j)^2 + (\eta - \eta_j)^2}] \tag{3a}$$

$$Q(\xi,\eta) = -\sum_{i=1}^{n} \phi_{2i}\, \text{sign}(\eta - \eta_i) \exp(-\sigma_{2i}|\eta - \eta_i|) - \sum_{j=1}^{m} \kappa_{2j}\, \text{sign}(\eta - \eta_j) \exp[-\delta_{2j}\sqrt{(\xi-\xi_j)^2 + (\eta - \eta_j)^2}] \tag{3b}$$

where "sign" is the function that returns the sign of its operand. The first term in each of Eqs. (3) causes the constant ξ and constant η lines to be attracted to ξ_i and η_i constant coordinate lines, respectively. The second term results in point attraction (or repulsion) of the constant coordinate lines to the point represented by (ξ_j,η_j). The sign function controls the attraction or repulsion on both sides of the line or point. Without it, attraction on one side and repulsion on the other side occur. Thompson et al.[22] state that there were no difficulties encountered because of the discontinuity of this function. The amplitudes and decay factors in the above equations can either be specified as input by the user or generated internally based on some criteria of error distribution. With the emergence of adaptive grids, Eq. (3) will become less useful. However, until multidimensional adaption becomes well understood and developed, the above method seems to be adequate. The system of equations represented by Eq. (2) can be discretized using the finite difference approach. The difference equations can subsequently be solved using the successive-over-relaxation (SOR) method to obtain the (x,y) coordinates of the interior points of the grid as a function of (ξ,η) and thus establishing the correspondence between every point in the physical domain and its image in the computational domain.

Energy Balance

The two-dimensional transient heat transfer is described by[23]

$$\frac{\partial}{\partial x}\left(k_{11}\frac{\partial T}{\partial x}\right)+\frac{\partial}{\partial y}\left(k_{22}\frac{\partial T}{\partial y}\right)+Q'=\rho C\frac{\partial T}{\partial t} \tag{4}$$

Note that the properties are a function of temperature (and therefore of position as well). In the formulation of Eq. (4), the following assumptions were made:

1) The energy generation term Q' is due to an exothermic chemical curing reaction and is described by the kinetic model of Loos and Springer[4] as $\rho(\partial\alpha/\partial t)H_R$.

2) The fiber direction is either parallel or perpendicular to the x axis.

3) The heat generated by viscous dissipation and heat transfer by convection within the laminate are negligible compared with heat transfer by conduction because the resin flow is of creeping nature. Viscous dissipation and convective heat transfer cannot be neglected in some processes such as extrusion.

Interior Node Difference Equation

Using the following transformations

$$f_x=\frac{1}{\sqrt{g}}(y_\eta f_\xi - y_\xi f_\eta)=a^{11}f_\xi + a^{21}f_\eta \tag{5}$$

$$f_y=\frac{1}{\sqrt{g}}(-x_\eta f_\xi + x_\xi f_\eta)=a^{12}f_\xi + a^{22}f_\eta \tag{6}$$

$$\left(\frac{\partial T}{\partial t}\right)_p=\left(\frac{\partial T}{\partial t}\right)_c-\frac{\partial x}{\partial t}(a^{11}T_\xi + a^{21}T_\eta)-\frac{\partial y}{\partial t}(a^{12}T_\xi + a^{22}T_\eta) \tag{7}$$

the spatial and time derivatives of Eq. (4) are transformed to the computational domain, and the energy equation becomes

$$A_1T_{\xi\xi}+A_2T_{\eta\eta}+A_3T_{\eta\xi}+A_4T_\xi+A_5T_\eta+Q'=\rho C\frac{\partial T}{\partial t} \tag{8a}$$

where

$$A_1=[(a^{11})^2k_{11}+(a^{12})^2k_{22}] \tag{8b}$$

$$A_2=[(a^{21})^2k_{11}+(a^{22})^2k_{22}] \tag{8c}$$

$$A_3=[2a^{11}a^{21}k_{11}+2a^{12}a^{22}k_{22}] \tag{8d}$$

$$\begin{aligned}A_4&=(a^{11})^2k_{11\xi}+(a^{12})^2k_{22\xi}+a^{21}a^{11}k_{11\eta}+a^{22}a^{12}k_{22\eta}\\&+a^{11}a^{11}_\xi k_{11}+a^{12}a^{12}_\xi k_{22}+a^{21}a^{11}_\eta k_{11}+a^{22}a^{12}_\eta k_{22}\\&+\rho C\frac{\partial x}{\partial t}a^{11}+\rho C\frac{\partial y}{\partial t}a^{12}\end{aligned} \tag{8e}$$

$$\begin{aligned}A_5&=a^{11}a^{21}k_{11\xi}+a^{12}a^{22}k_{22\xi}+(a^{21})^2k_{11\eta}+(a^{22})^2k_{22\eta}\\&+a^{11}a^{21}_\xi k_{11}+a^{12}a^{22}_\xi k_{22}+a^{21}a^{21}_\eta k_{11}+a^{22}a^{22}_\eta k_{22}\\&+\rho C\frac{\partial x}{\partial t}a^{21}+\rho C\frac{\partial y}{\partial t}a^{22}\end{aligned} \tag{8f}$$

Equations (8) then represent the differential equation describing heat transfer and generation for an interior node in the computational field. The $(\partial x/\partial t)$ and $(\partial y/\partial t)$ represent the speed with which the node is moving due to compaction. Using an implicit finite difference method with second-order accurate difference for spatial derivatives and first-order accurate difference for the time derivatives, the above equation is discretized to yield

$$\begin{aligned}&\left(-2A_1-2A_2-\frac{\rho C}{\Delta t}\right)T_{i,j}+\left(A_1-\frac{1}{2}A_4\right)T_{i-1,j}\\&+\left(A_1+\frac{1}{2}A_4\right)T_{i+1,j}+\left(A_2+\frac{1}{2}A_5\right)T_{i,j+1}\\&+\left(A_2-\frac{1}{2}A_5\right)T_{i,j-1}+\left(\frac{1}{4}A_3\right)T_{i+1,j+1}\\&+\left(\frac{1}{4}A_3\right)T_{i-1,j-1}-\left(\frac{1}{4}A_3\right)T_{i+1,j-1}\\&-\left(\frac{1}{4}A_3\right)T_{i-1,j+1}+Q'_{i,j}=-\frac{\rho C}{\Delta t}\mathrm{TOLD}_{i,j}\end{aligned} \tag{9}$$

Equation (9) represents the implicit difference equation for an interior point in the grid. Note that the temperatures of nine different nodes are included in the equation and, thus, define a nine-point computational molecule.

Convective Boundary Condition

Several boundary conditions can be imposed on the problem depending on the type of heat transfer process being described. In order to accommodate the widest range of processes, three types of boundary conditions are treated: specified temperature at the boundary, specified heat flux, and a convective boundary.

In developing the heat transfer equations for convective boundary nodes, a heat balance is written. Several types of equations result depending on the position of the node under study. Using Fourier's law for the conductive heat flux and Newton's law of cooling for the convective heat flux, the energy balance equation for a boundary node is written. The energy balance for a node of a complex boundary is obtained in the same fashion as that of a rectangular boundary node. The only difference is in representing distances and areas available for heat transfer. The energy balance equation (for a node at a boundary of any shape) is then transformed and discretized using the same procedure outlined above for an interior node. The transformed equations include terms representing the speed with which the boundary is moving due to compaction.

Temperature Specified Boundary Condition

In processes such as a press where the temperature at the boundary is known, a different approach is used. The interior node equations remain the same. However, when a boundary node temperature is encountered, it is replaced by the value of the specified temperature at that particular boundary node. For a complex boundary, assign the temperatures of the boundary nodes of the physical domain to their respective images in the computational domain. A nonuniform boundary temperature profile can be accommodated.

Specified Heat Flux Boundary Condition

This boundary condition requires a new set of difference equations for the boundary nodes. The method is simple and can be summarized as follows: the same procedure used in the convective boundary can be used if one replaces Newton's law of cooling term by the specified flux (zero for insulated boundaries).

Properties

Historically, composite material properties have been evaluated as constant bulk properties. These approaches do not include the property's dependence on direction since average constant values throughout the material are assumed. Several empirical methods to determine composite properties have been tested using the computer code and compared. These methods include directional transport properties,[24] resistance method,[25] the expression developed by Ma et al.[9] and Springer and Tsai.[5] The expression of Saliba et al.,[24] which uses an effective thermal conductivity in the fiber and transverse direction, is also used. When studying heat transfer in composite materials, the properties at the interfaces between two different materials have to be determined. Simple arithmetic averages have been previously used. In this work, the method of Servais et al.[7] is utilized. This method consists of weight averaging the values between the two materials forming the interface.

Solution

The difference equations derived above are nonlinear (coefficient matrix contains temperature dependent properties) requiring an iterative solution method. The assembled difference equations were solved using the SOR method. Although this method is usually used with steady-state problems, it proved to be effective for solving for the temperature distribution at each time step. If the assembled equations can be represented with the following matrix equation $[C(T)][T]=[P]$, the solution approach is as follows:

1) Using initial conditions, calculate $[C]$ and $[P]$.

2) Using the difference equation for node (I,J) calculate an intermediate value of the temperature (TEMP) at node (I,J).

3) Calculate temperature at node (I,J): $T_{i,j} = W\text{TEMP} + (1-W)\ \text{TOLD}_{i,j}$.

4) Repeat steps 2 and 3 for all nodes.

Using the relaxed temperatures instead of initial conditions, the coefficient matrix $[C]$ is calculated, and the above procedure is repeated. The newly calculated values are compared with the previously calculated ones. This iteration is repeated until the maximum difference between two consecutively calculated values at the same node is within the allowed tolerance. Then, the time is updated, and the entire process is repeated at the new time step. Note that when marching through the grid, the most updated values of the previous points are used in the calculation so that maximum computational efficiency is obtained.

A Gauss-Jordan (G-J) direct method for solving the assembled equations at each iteration within a time step was also used. The two solution methods (SOR and G-J) were not compared. Although implicit finite differences (unconditionally stable) were used, parametric studies were conducted to find an envelope of suitable spatial and time step sizes.[23] This analysis is beyond the objectives of this paper and will not be presented here.

Summary

The modeling of thermoset composite materials processing was addressed. The various physical and chemical phenomena taking place during cure were summarized, and the effect of the chemical reactions and compaction on the heat transfer process was identified. A review of mathematical models which describe heat transfer through curing composite materials was presented. The models described in the literature are limited to one-dimensional conditions, simple geometry, constant properties, or fixed boundaries. The model described herein extended the existing models to accommodate heat transfer through complex-shaped, two-dimensional heterogeneous composite materials of variable properties, moving boundaries, and various boundary conditions. The model also includes heat generation due to exothermic chemical reactions. The solution method of the energy equation is the body-fitted coordinate system generation technique coupled with an implicit finite difference method. The resulting difference equations were solved iteratively to predict temperature and degree-of-cure profiles as a function of time.

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