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CURING SIMULATION OF COMPOSITES COUPLED WITH INFRARED HEATING

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ABSTRACT : Because of higher specific strength and stiffness, low weight, and good resistance to corrosion, the use of composite materials in aerospace structures has increased. Aircraft industry has recently begun to investigate Liquid Composites Molding techniques (LCM) through research programs because of its ability to produce large parts at a low cost. In this paper, we have not addressed the filling step during which the resin flows through fibrous media, but we investigate the numerical simulation of curing reinforced RTM-6 by infrared heating. A ray tracing in-lab software called RAY-HEAT is used to compute the radiation energy due to the interaction between reinforced RTM-6 and lamps. Finite element based program COMSOL Multiphysics™ has been used to simulate the curing process. Thermochemical model has been implemented in order to compute reaction rate as a function of reaction temperature and degree of conversion using a cure kinetic model .

KEYWORDS: polymer composite, infrared heating, degree of cure, epoxy resin, graphite/epoxy, modeling.

1 INTRODUCTION

Using thermal radiation to cure the composite is more efficient, effective, and clean than standard thermal energy sources. Thermal radiation was successfully used to produce a high quality polymer matrix in less time [1]. The infrared radiation was studied and coupled with the energy equation and the exothermic heat released during the curing process in order to predict the composite temperature as a function of time and degree of cure.

2 INFRARED INTERACTION

Because the graphite fibers embedded in the epoxy resin are very strong infrared absorbers over broad spectral ranges at all wavelengths, the composite has strong absorption bands in some portions of the spectrum; we have neglected epoxy absorption in this paper [1, 2, 3, 4].

2.1 BOUNDARY CONDITIONS

The composite is assumed to be a block with a thickness range of 1-25mm. It is composed of the superposition of a sequence of carbon plies embedded in an epoxy matrix [5].

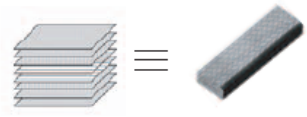


Figure 1: *.Schematic presentation of the composite [6].*

The infrared radiation is absorbed by the carbon ply present in the top surface of the composite. The boundary conditions taken in COMSOL Multiphysics™ are a surface incident heat flux on the top surface. There is also natural heat convection between this surface and the air present in the oven. The other boundaries are assumed to be adiabatic: the bottom surface is in contact with a mould and there is no heat exchange between the mould and the composite. The other sides of the composite are so thin in comparison with the top that they can be considered adiabatic.

3 ENERGY EQUATION

The composite transient temperature can be evaluated by the Fourier's modal equation assuming an instantaneous equilibrium temperature between the resin and the fibers at each time: [7,8]

$$\rho_c c_{p,c} \frac{\partial T}{\partial t} = \nabla(k_c \nabla T) + v_m \rho_m H_u \frac{\partial \alpha}{\partial t} \qquad (1)$$

Where H_u= ultimate heat of reaction, α = degree of cure, and the $\rho_c = \text{density}, c_{p,c} = \text{heat capacity}, \quad k_c =$ thermal conductivity of the composite are expressed using the rule of mixture:[8, 9, 10]

$$\rho_c = \frac{\rho_m \rho_f}{\rho_m w_f + \rho_f w_m} \qquad (2)$$

$$c_{p,c} = c_{p,m} w_m + c_{p,f} w_f \qquad (3)$$

$$k_c = \frac{k_m k_{f,t}}{(1-v_m) k_m + v_m k_{f,t}} \qquad (4)$$

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$$w_m = \frac{v_m/\rho_f}{v_m/\rho_f + (1-v_m)/\rho_m} \quad (5) \qquad w_f = 1 - w_m \quad (6)$$

Where $k_{f,t}$= transverse thermal conductivity of the fibers, w = weight fraction, v = volume fraction, the subscript f stands for the fibers, and m for the matrix.
The system of equations described above can be solved at each time step in order to calculate the distributions of temperature and degree of cure and their evolution with time.

4 KINETIC MODELING

4.1 GLASS TRANSITION TEMPERATURE

The glass transition temperature is modelled using the DiBenedetto equation:[9]

$$T_g = T_{g0} + \frac{(T_{g\infty} - T_{g0})\gamma\alpha}{1-(1-\gamma)\alpha}$$

Where T_{g0} and $T_{g\infty}$ = glass transition temperature of un-reacted and full-reacted resin, γ = material constant.
These values were deduced from the literature [8].

4.2 KINETIC MODEL

Kamal and Sourour [7] have shown that the following model adequately describes the cure kinetics of an epoxy resin:

$$\frac{d\alpha}{dt} = (k_1 + k_2\alpha^m)(1-\alpha)^n$$

$$k_i = A_i \exp\left(-\frac{E_i}{RT}\right) \qquad i = 1,2$$

Where k_1 and k_2 = rate constants with an Arrhenius type of dependence with temperature and m and n = catalytic constants.
Although the model of Kamal and Sourour [6] contains several essential features, it ignores the influence of the glass transition temperature T_g on the diffusion of the macromolecules.
This model [7] has been extended in order to reflect the effect of vitrification in the kinetic model of resin transfer molding. This model was also used by [8]

$$\frac{d\alpha}{dt} = \left(\frac{k_1 k_D}{k_1 + k_D} + \frac{k_2 k_D}{k_2 + k_D}\alpha^m\right)(1-\alpha)^n$$

$$k_D = k_3 \exp\left(-\frac{b}{f}\right)$$

$$f = f_0 + \delta(T - T_g) \quad [11]$$

Where k_D = diffusion factor, k_3 = diffusion rate constant, b = constant having the order of one, T_g = glass transition temperature, f = the free volume fraction of the polymer[11], f_0 = the free volume fraction for a temperature equal to T_g, δ = the thermal expansion coefficient for a temperature above T_g.
For an amorphous polymer [10] which is the case of our composite, $f_0 = 0{,}025$ and $\delta = 0{,}00048$.

4.3 KINETIC PARAMETERS

Kinetic parameters of RTM6 resin are calculated using linear regression algorithms by fitting model equations $\frac{d\alpha}{dt} = f(\alpha, T)$ with the Differential Scanning Calorimeter experimental data. These experimental DSC data were scanned isothermally for different temperatures: 140, 160, 170 and 180°C.
In the DSC, the rate of heat generation dQ/dt is recorded with time. As H_u is constant for a given curing process, [7, 8, 12] the rate of the degree of cure is calculated by:

$$\frac{d\alpha}{dt} = \frac{1}{H_u}\frac{dQ}{dt}$$

The degree of cure is obtained by integrating the DSC data and normalizing with H_u.

4.4 FITTED KINETIC PARAMETERS

The figure 2 below shows the temperature dependence of factors $K1$ and $K2$ which exhibits an Arrhenius type of temperature dependence.

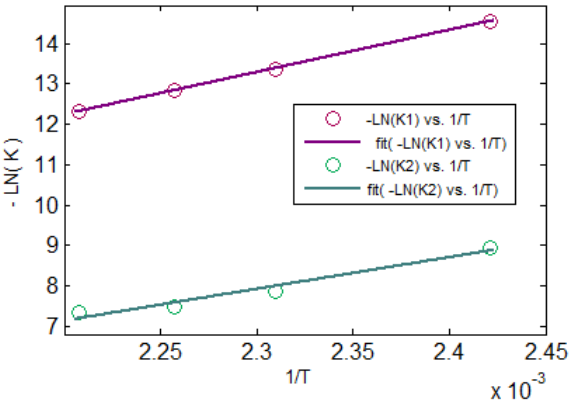


Figure 2: *Reaction rate constants k1 and k2.*

The estimated kinetics parameters from the fitting curves used in our simulations are listed in table 1 below.

Table 1: *Estimated parameters*

parameter	
A_1 $[min^{-1}]$	4.5E6
E_1 $[J/mol]$	74.69E4
A_2 $[min^{-1}]$	1.3E6
E_2 $[J/mol]$	58.37E4
A_3 $[min^{-1}]$	85.2E28
E_3 $[J/mol]$	2.16E5
m [-]	1.23
n [-]	1.36
b [-]	0.227
H_u [J/kg]	4.30E5
R [J/(mol-K)]	8.314

5 NUMERICAL EXAMPLE

The carbon fibers embedded in the epoxy are very strong absorbers in that they absorb infrared energy and convert this energy to heat the epoxy matrix surrounding the carbon fibers. Thermal radiative boundary conditions are imposed on the top surface of the composite and the curing temperature is controlled through the infrared emitters in order to optimize a uniform surface heat flux. The emitters used in our project are halogen lamps given from TOSCHIBA LIGNTNING.

Until now, the composite properties were taken from the literature [1, 9]; others are assumed to be constants in order to run some initial simulations of a curing process of RTM6 as a matrix combined with a fibrous reinforcement in the form of carbon plies.

In this numerical example, a high heat flux of $1600\ W/m^2$ was provided for the first 6 minutes in order to initiate and accelerate resin curing, and for the rest of time, the surface heat flux taken was $1200\ W/m^2$. These heat flux values was imposed in a way to cure the composite in a less time and do not exceed 220°C in temperature (the degradation temperature of the matrix).

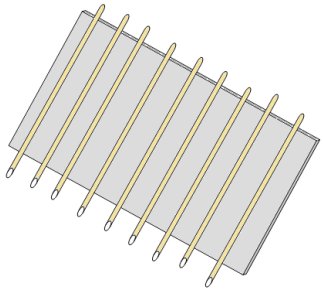


Figure 3: *Schematic presentation of the composite face to the infrared emitters.*

5.1 NUMERICAL RESULTS

Using COMSOL Multiphysics, the energy equation was coupled with the exothermic reaction in order to predict the temperature distribution in the composite and the degree of cure.

The figures 4 and 5 below show the temperature and the degree of curing respectively evolution with time at the center of the composite.

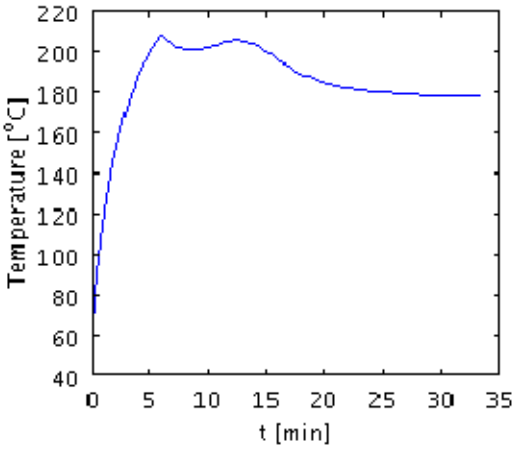


Figure 4: *Temperature evolution with time at the center of the composite*

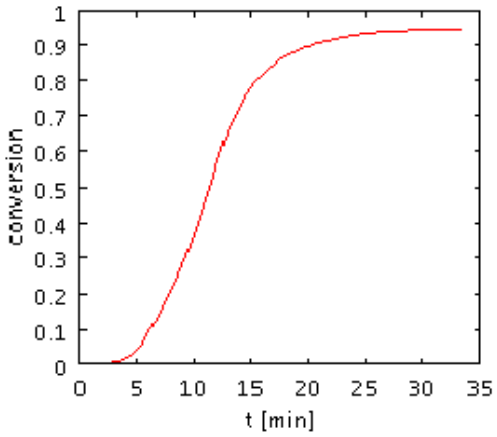


Figure 5: *Degree of cure evolution with time at the center of the composite*

The figures 6 and 7 below show the temperature and degree of cure respectively evolution with time and thickness of the composite.

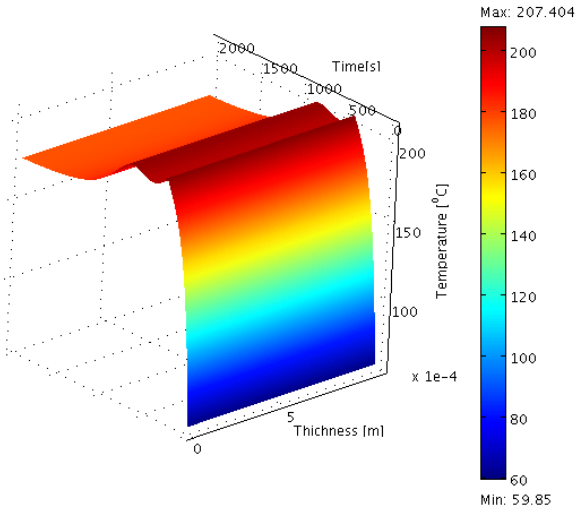


Figure 6: *Temperature evolution with thickness and with time*

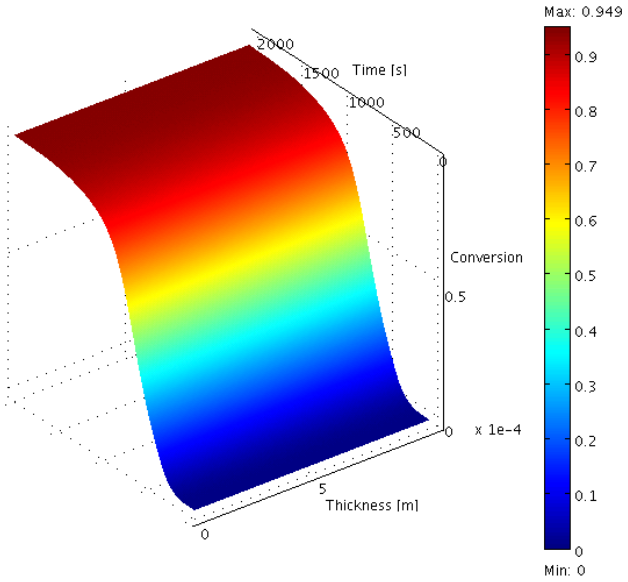


Figure 7: *Degree of cure evolution with thickness and with time*

These numerical simulations correspond for a 1mm thickness composite; there is no important variation of the temperature or degree of cure with the thickness. From these simulations, we can deduce that the cure of the composite is accomplished in a less time than the others modes of heating.
In the future work, we need to validate our numerical simulations with experimental infrared curing.

6 CONCLUSIONS

The cure model of resin is investigated in this study. A modification of the Kamal-Sourour cure model is given to account for the diffusion effect. Kinetic parameters of the cure model based on DSC Experiments were estimated using optimization algorithms. A thermal-kinetic model has been developed in COMSOL Multiphysics to simulate the cure process of pure resin with assumed material properties. In-lab software developed to predict the infrared heat flux on the top surface of the composite will be coupled with COMSOL in order to simulate the whole infrared curing process of the composite.

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