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Simulations of an Infrared Composite Curing Process**

By Sawsane Nakouzi*, Johann Pancrace, Fabrice Schmidt, Yannick Le Maoult and Florentin Berthet

Using IR energy is an efficient method of curing composites. In this paper, we study IR interactions with the composite, which is placed in an IR oven. The liquid resin infusion technique is used for the impregnation process of fibers with resin. Numerical simulations of the curing process for a carbon fiber-reinforced epoxy (RTM6) system are presented. In-lab software called Rayheat based on ray tracing algorithms and developed in Matlab is used to compute the radiative heat flux that impacts the composite. A three-dimensional numerical model is developed in the finite element software Comsol Multiphysics, where the heat-balance equation is coupled with the cure kinetic model of the resin. The computed radiative heat flux is exported to Comsol Multiphysics and imposed as a boundary condition on the top surface of the composite. This numerical model allows calculation of the temperature distribution in the composite during curing, which is a key parameter that affects its mechanical properties. We can predict also the evolution of the degree of cure as function of time.

Epoxy resins have several applications in the aerospace and automobile industry. Because of their good adhesive properties, superior mechanical, chemical and thermal properties, and resistance to fatigue and microcracking, they produce high performance composites. Since it is necessary to optimize the manufacturing time and costs and to determine the performance of these composites, some researchers^[1-4] have studied infrared (IR) heating for the polymerization process. Others^[5,6] have used IR energy for the preheating process.

In the technology presented here, the composite is cured in an IR oven (see Figure 1) which includes halogen lamps. The liquid resin infusion (LRI) process is used to manufacture the composite, whereby liquid resin is infused through a fiber reinforcement previously laid up in a one-sided mold.

These epoxy resins release an exothermic heat flux during the curing process, which can possibly cause an excessive temperature in the thickness. Consequently, for the production of high performance composites, it is necessary to know the thermal behavior of the composite during curing. The most detailed models for the curing process of composites using IR heaters have been presented by Chern *et al.*^[1-4] They measured the radiative properties of graphite/epoxy and

glass/epoxy systems and modeled the radiative heat transfer in the glass/epoxy system as a volumetric radiation transport. Therefore, IR interactions with the graphite/epoxy system were modeled as a surface radiation transport.

Cosson *et al.*^[7] developed numerical algorithms, based on a ray-tracing method. In-lab software, called Rayheat, predicts the volumetric distribution of the radiation intensity in a semi transparent medium. This software also models IR interactions with highly absorptive mediums, where the radiation transport is a surface radiation. In this paper, we present simulations of the IR curing process of a carbon/epoxy system. Numerical simulations were performed in order to determine the temperature distribution in the composite thickness during the polymerization process. The heat balance equation is coupled with the exothermic heat and the radiative heat flux using the commercial software Comsol MultiphysicsTM. In order to introduce these simulations to an industrial composite part, we validate the simulations of a simple sheet of composite.

Liquid Resin Infusion (LRI)

LRI is used for the impregnation process of fibers with resin. In this process, presented schematically in Figure 2, liquid resin with low viscosity is infused through fiber reinforcement, which is placed in a single-sided mold sealed with a vacuum bag. Before being impregnated by resin, the fiber reinforcement must be preheated by the IR lamps, in order to maintain the low viscosity of the resin and to facilitate its path through the reinforcement. This process combines the advantage of infusion at ambient pressure, short cycle time and low cost equipment with the ability to produce parts of

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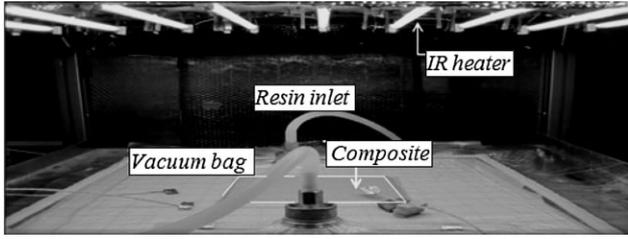


Fig. 1. IR oven. This figure presents the IR oven used in our experiments. The composite is placed at the center of the oven, facing the IR heaters. A liquid resin infusion process is used to impregnate the carbon fiber reinforcement.

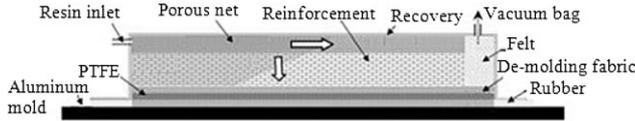


Fig. 2. Schematic presentation of the liquid resin infusion process. Reinforcement is placed in a one-sided mold. A vacuum pump assures the infusion of the resin through the reinforcement.

complex shape. After the reinforcement has been impregnated, IR heaters are turned on in order to start the curing process of the composite.

Materials

The physical geometry of the composite is assumed to be a sheet of dimension $160 \times 140 \times 6 \text{ mm}^3$. It is composed of a sequence of carbon plies embedded in an epoxy matrix. The carbon fiber lay-up shows plies rotated by 45° (nomenclature: [0/0/+ 45/90/-45/0] s). Commercial epoxy resin RTM6 provided by the Hexel Corporation was used in our study. This resin was chosen because it is currently used in the aerospace and automobile industries.

Short wave halogen lamps (in the range $0.6\text{--}6 \mu\text{m}$ and wavelength $\lambda_{\text{max}} = 1.2 \mu\text{m}$ at the lamp's maximum relative intensity) made by Toshiba Lighting Company were employed for the curing process. The lamps have variable power levels and a nominal power of 1000 W. A tungsten filament is the IR emitter; it is surrounded by quartz glass and a kind of halogen gas-filled lamp.

IR Interaction

IR radiation processing of the polymeric materials is fast, volumetric (in semi-transparent media), direct and controllable. Several researchers have studied^[8–11] the radiative heating processes of polymers and semi transparent mediums. Cosson *et al.*^[7] developed software called Rayheat in order to predict the distribution of the radiative intensity in PET preforms. Chern *et al.*^[11–4] successfully studied the radiative curing process of hoop-wound cylinders of graphite/epoxy and glass/epoxy. Previously, we have studied the curing process of a composite, the physical geometry of which corresponds to a carbon/epoxy block, manufactured by the LRI process. The recovery surface is semitransparent to IR radiation: 92% of the IR radiation is transmitted to the composite (this value was measured in an earlier work in the laboratory). Carbon fibers embedded in the epoxy resin

are very strong IR absorbers over broad spectral ranges at all wavelengths, while the composite has strong absorption bands in some portions of the spectrum.^[11] The mean value of the spectral reflectance of the carbon/epoxy system is 15%, as taken from the literature.^[2]

Ray Tracing

In ray tracing, we model radiation as rays of light. The propagation of the rays is only based on the laws of geometrical optics. The general idea is to simulate the interactions between the ray of light emitted by a source (in our case halogen lamps) and every object present in the scene (here, the IR oven), including multiple lamps, reflectors and the composite. In the ray tracing software, coiled tungsten is the IR emitter. It is assumed to be a Lambertian grey body^[7] and is modeled as a cylinder with equivalent diameter. That is, we define a number N of rays emitted from the lamp; this number is chosen in a way to have a good agreement between the calculations costs and the needed precision of the results (see Figure 3). Then we start from a random point P on the IR emitter and trace the ray emitted in a random direction. This direction d is defined by two parameters $\Theta \in [0, \pi/2]$ and $\varphi \in [0, 2\pi]$. These parameters are defined by stochastic variables:^[12]

$$\Theta = \arcsin \sqrt{X_1} \quad (1)$$

$$\varphi = 2\pi X_2 \quad (2)$$

where X_1 and X_2 are independent uniform stochastic variables in the range $[0,1]$.

Each ray emitted by the source is defined by its origin, direction, and spectral intensity. Then the ray is tested against all objects in the scene to determine what it hits and if it intersects the composite (Figure 3). The different interactions between the ray and the objects present in the scene change the ray properties. In our ray tracing software, assumptions found in the literature are made for the different optical properties of lamps, reflectors,^[10] recovery surface and the carbon/epoxy system.^[1,2] They are referenced in Table 1 below. The ray tracing method has been validated in a previous work in the laboratory^[7] with an analytical solution given by view factor.^[13]

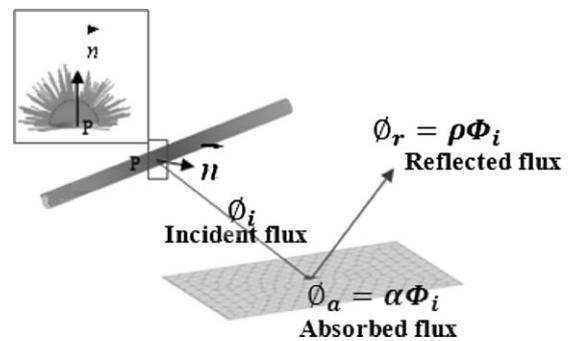


Fig. 3. Schematic presentation of the ray-tracing method. In this method we compute the origin and direction of each ray emitted by the IR heater and we study its intersection with the composite. Ray tracing method is used in our numerical simulations in order to compute the IR heat flux that impacts the top surface of the composite.

Table 1. Summary of the thermophysical properties used in the numerical simulations.

Property	Material	Expression	Reference
Volume fraction	Carbon fiber	$v_f = \left[\frac{\rho_f}{\rho_r} \left(\frac{m}{m_f} - 1 \right) + 1 \right]^{-1}$	[20]
	RTM6	$v_m = 1 - v_f$	[20]
Weight fraction	RTM6	$w_m = (v_m / \rho_f) / (v_m / \rho_f + (1 - v_m) / \rho_m)$	[18]
	Carbon fiber	$w_f = 1 - w_m$	[18]
Density $\rho(\text{kg}/\text{m}^3)$	Composite	$\rho_c = \frac{\rho_m \rho_f}{\rho_m w_f + \rho_f w_m}$	[18]
Heat Capacity (J/kgK)	RTM6 ($\alpha = 0$)	$C_{p,m}(0, T) = 1208.15 + 15.1969T - 0.049976T^2$	[20]
	RTM6 ($\alpha = 1$)	$C_{p,m}(1, T) = 816.29 + 13.35109T - 0.036553T^2$	[20]
	RTM6 (α)	$C_{p,m}(\alpha, T) = Cp(0, T) * (1 - \alpha) + Cp(1, T) * \alpha$	[20]
	Carbon fiber	$C_{p,f}(T) = 577.4 + 6.85165T - 0.018078T^2$	[20]
	Composite	$C_{p,c} = C_{p,m}w_m + C_{p,f}w_f$	[20]
Thermal conductivity $k(\text{W}/\text{mK})$	RTM6	$k_m(\alpha) = 0.1155 * \alpha^4 + 0.2222 * \alpha^3 - 0.9228 * \alpha^2 + 0.785$	[20]
	Carbon fiber	0.265	Transverse thermal conductivity ($v_f = 0.62$)
		3.1	Longitudinal thermal conductivity ($v_f = 0.62$)
	Composite	$k_{c,t} = \frac{k_m k_f}{(1 - v_m)k_m + v_m k_f}$	Transverse thermal conductivity.
		$k_{c,l} = v_m k_m + (1 - v_m)k_f$	Longitudinal thermal conductivity.

IR Oven and Boundary Conditions

Composite is exposed to radiation in the IR oven (see Figure 4). This one is composed of nine halogen lamps provided by Toshiba Lighting Company. The IR radiation is absorbed by the carbon ply present on the top surface of the composite. In addition this surface is a convective boundary surrounded by the air present in the oven, at temperature $T_{\text{air}}(t)$. The frame of the composite is so thin in comparison with the surface, the edge effect becomes negligibly small; that is, we can ignore the heat exchanges all around its contour. The bottom of the composite is in contact with a surface that is supposed to be a thermal insulation, but there is no real adiabatic surface. Bottom temperature was introduced as a boundary conditions in the finite element software Comsol. During curing, incident radiation is absorbed by the top surface of the carbon/epoxy composite. Then the composite is thermally heated in the thickness via conduction from its top surface. Figure 5 presents the IR density distribution as well as the boundary conditions.

Natural Heat Convection Coefficient

The natural heat transfer coefficient is obtained from a correlation for the horizontal plate with the heated surface

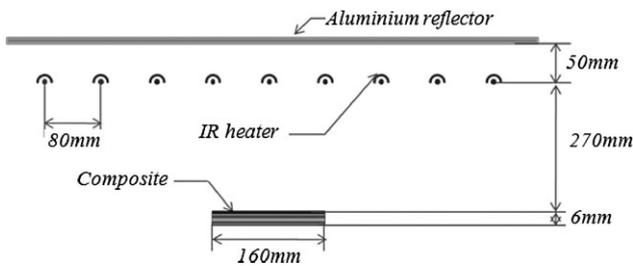


Fig. 4. Schematic presentation of the IR oven. The IR oven is composed of nine halogen lamps. This oven geometry configuration is used in our simulations in order to predict the heat flux on the top surface of the composite.

facing upward:^[14]

$$h = \frac{k_{\text{air}} Nu}{L} \quad (3)$$

Where Nu is the Nusselt number, k_{air} is the air thermal conductivity, and L is the characteristic length of the plate. With $Nu = c(Gr.Pr)^n$ where Pr is the Prandtl number, $c = 0.13$ and $n = 1/3$. The physical properties of atmospheric air in the previous equations are evaluated at a mean temperature, defined as: $T_m = T_s - 0.25(T_s - T_\infty)$, where T_s is the composite top surface temperature. The Grashof number is defined as $Gr_L \equiv (g\beta(T_s - T_\infty)L^3)/\nu^2$ where β is the thermal expansion coefficient, and is evaluated at $(T_s + T_\infty)/2$.

Kinetic Modeling

Kamal and Sourour^[15] have shown that the following model describes the cure kinetics of an epoxy resin:

$$\frac{d\alpha}{dt} = (k_1 + k_2 \alpha^m)(1 - \alpha)^n \quad (4)$$

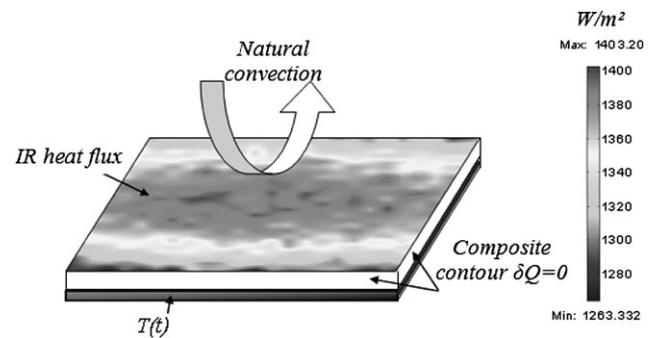


Fig. 5. Boundary conditions. IR heat flux that impact the top surface of the composite is taken as a boundary condition in the finite element software Comsol Multiphysics. The contour of the composite is so thin in comparison with its thickness that we can ignore heat exchanges all around its contour. The bottom surface of the composite is taken as a boundary condition.

$$k_i = A_i \exp\left(-\frac{E_i}{RT}\right) \quad i = 1, 2, 3 \quad (5)$$

where k_i are the rate constants with an Arrhenius type of dependence with temperature and m and n are catalytic constants.

Although the model of Kamal and Sourour contains several essential features, it ignores the influence of the glass transition temperature T_g on the diffusion of the macromolecules. This model has been extended in order to reflect the effect of vitrification, which induces a decrease of the resin reaction rate since mobility of macromolecules becomes very limited. This model was also used by other authors.^[16,17]

$$\frac{d\alpha}{dt} = \left(\frac{k_1 k_d}{k_1 + k_d} + \frac{k_2 k_d}{k_2 + k_d} \alpha^m \right) (1-\alpha)^n \quad (6)$$

$$k_d = k_3 \exp\left(-\frac{b}{f}\right) \quad (7)$$

$$f = f_0 + \delta(T - T_g) \quad (8)$$

where k_d is a diffusion factor, k_3 the diffusion rate constant with an Arrhenius type of dependence, b a constant of order one, T_g the glass transition temperature, f a free volume fraction of the polymer, f_0 a free volume fraction for a temperature equal to T_g , and δ the thermal expansion coefficient. For an amorphous polymer, which is the case of our composite, $f_0 = 0.025$ and $\delta = 4.8 \times 10^{-4} \text{ K}^{-1}$.

The glass transition temperature is modeled using the DiBenedetto equation:^[17,18]

$$T_g = T_{g0} + ((T_{g\infty} - T_{g0})\lambda\alpha) / (1 - (1-\lambda)\alpha) \quad (9)$$

Where T_{g0} and $T_{g\infty}$ are the glass transition temperature of unreacted and fully reacted resin, respectively, and λ is the material constant. These values were deduced from the literature.^[19]

The kinetic parameters of resins can be calculated using linear regression algorithms by fitting model equations with differential scanning calorimetry (DSC) experimental data. In this paper, the kinetic parameters of the RTM6 resin were taken from published estimates.^[19]

Energy Equation

The composite transient temperature can be evaluated by the heat balance equation assuming an instantaneous equilibrium temperature between the resin and the fibers at each time:^[18]

$$\rho_c C_{p,c} \frac{\partial T}{\partial t} = \nabla k_c \nabla T + v_m \rho_m H_u \frac{\partial \alpha}{\partial t} \quad (10)$$

Where H_u is the ultimate heat of reaction of the resin, and α the degree of curing. The material property parameters required in this analysis are listed in Table 1. The thermal conductivity of the composite k_c (W/mK) was measured by Leco for carbon-reinforced RTM6 resin.^[20]

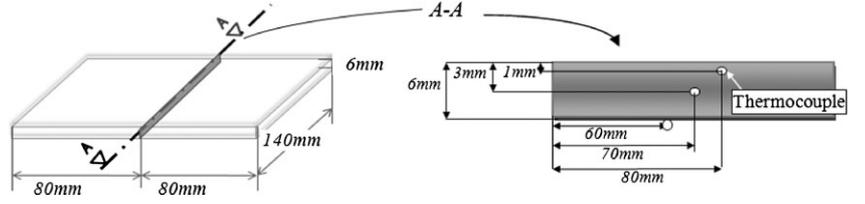


Fig. 6. Experimental set-up. Temperature acquisition is assured by three thermocouples type K, placed in the thickness of the composite and having an $80 \mu\text{m}$ diameter.

Experimental

Experimental measurements were carried out in order to validate numerical simulations of the IR curing process of carbon/epoxy composite. Temperature acquisition in the thickness of the composite is achieved using three thermocouples of type K, having an $80 \mu\text{m}$ diameter. These thermocouples were fixed in the fiber reinforcement through the thickness. Then, air was aspirated with the vacuum bag and the fibers were compacted. A schematic presentation of the thermocouple positions is presented in Figure 6.

Validation

Experiments show a good agreement with the numerical results. In the experimental process, carbon fibers placed in the mold were preheated by the IR lamps present in the oven. Then the IR heaters were turned off and the RTM6 resin at 80°C infused through the reinforcement using the LRI process. In order to refer to the same operating conditions between experiments and simulations (we did not address in our simulations the filling step, during which resin flows through the fiber reinforcement), IR heaters were turned on after infusion took place and stabilization of the resin in the mold. The curing process was achieved with 22% of the nominal power of the IR heaters. Because the degradation temperature of the matrix is 220°C , we turned off the lamps at 200°C . The lamps were then turned on again when the temperature decreased to 175°C (see Figure 7). We introduced the experimental operating conditions as input data in our simulations: oven and composite geometry, air temperature,

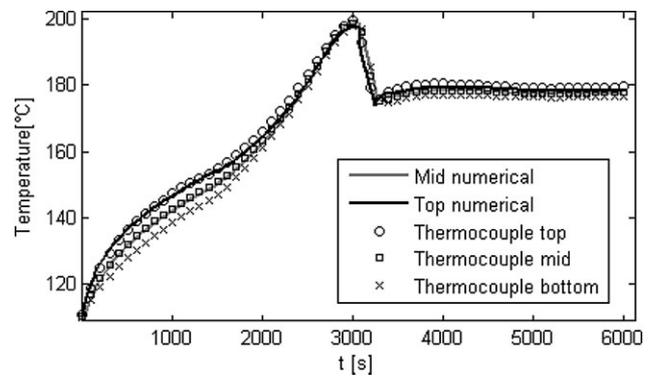


Fig. 7. Curing temperature validation: thermocouple temperature acquisitions through the thickness of the composite and numerical results. This figure shows the good agreement between the experimental data and the numerical results.

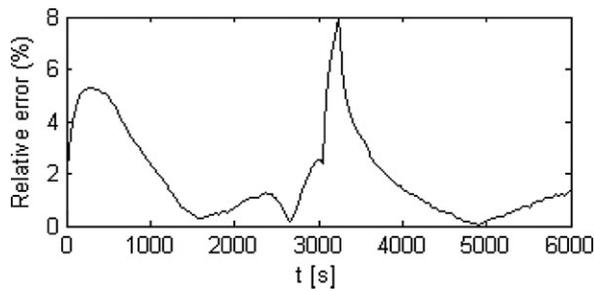


Fig. 8. The relative error between the thermocouple temperature and the numerical results.

initial temperature of the composite, bottom temperature of the composite.

The relative error between thermocouple's actual and numerically predicted temperature is calculated using Equation 11 and presented in Figure 8:

$$E = \sqrt{\frac{\sum_{i=1}^2 (T_{exp} - T_{num})^2}{\sum_{i=1}^2 T_{exp}^2}} \times 100 \quad (11)$$

Conclusions

A novel process for curing composites is the use of IR heaters. The composite is placed in an IR oven, facing the IR heaters. Numerical simulations were carried out in order to predict the thermal behavior of the composite during the curing process. Radiative heat flux is computed in Matlab, using an algorithm developed in-lab. This heat flux is exported to the finite element software Comsol Multiphysics, where it is taken as a boundary condition on the top surface of the composite. The exothermic heat flux released by the epoxy matrix during the curing process is coupled with the heat equation. Then, we can predict temperature distribution in the composite during the polymerization and the degree of cure. We validate our numerical results with experimental measurements. So far we have discussed IR curing of carbon fiber-reinforcement epoxy matrix. Curing glass/epoxy systems is our interest for future works, because of the volumetric distribution of the radiative intensity in the semi transparent mediums.

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