NON ISOTHERMAL SIMULATION OF THE RESIN TRANSFER MOLDING PROCESS

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SUMMARY

In this work the non isothermal nature of the process has been analyzed, together with the influence of the curing reaction on the temperature profiles. The thermal analysis has been considered for the resin-fibers system, the dry fibers and the mold, to evaluate for the whole system the temperature field. The numerical simulations have been performed by using an epoxy resin and a fiber glass mat. The temperature and degree of cure fields are presented and compared with the literature data.

KEYWORDS: Resin Transfer Molding, non isothermal filling simulation.

INTRODUCTION

Resin Transfer Molding is an efficient and attractive process for the manufacturing of high performance polymer composites. In this process, a thermoset polymer resin is injected into a mold, where a fiber preform is placed beforehand. The resin impregnates the preform and fills the mold. Near the end of the filling, the resin reacts to form a cross-linked polymer network (curing reaction).

The process involves different physical and chemical phenomena. The resin flow is coupled to heat transport, chemical reaction and rheological changes. The mold filling, in fact, doesn't take place under isothermal conditions. Since the resin is injected at room temperature and the mold is pre-heated, heat transport from the mold walls and the fiber preform to the resin occurs. Further, the curing reaction is highly exothermic. The flow pattern is also influenced by the resin viscosity that depends on the temperature and on the conversion. At the starting, due to the heating from the mold, the resin viscosity may drop by orders of magnitude; when the curing reaction starts, the viscosity increase as the resin solidifies.

Due to the complex nature of the problem, process modeling and numerical simulation can represent an useful tool to understand, control and manage the different effects affecting the product quality.

The process simulation would be useful in addressing several critical issues. It will allow to the designer to anticipate some problems such as dry spots, premature resin cure and formation voids. Before the mold is built, the designer could optimize the filling and the curing stage by controlling the processing parameters (the injection pressure, the placement of the gate and the mold temperature) and reducing the tooling costs. A model for the simulation of the non-isothermal mold filling stage of the RTM manufacturing process has been developed. The non isothermal nature of the process has been analyzed, together with the influence of the curing reaction on the filling. The thermal analysis has been considered for the resin-fibers system, the dry fibers and the mold, to evaluate for the whole system the temperature field as function of the filling time. In the fig.1 the analyzed system at a generic time is reported.



Fig.1: Geometric schema of the analyzed system.

PROCESS MODELING

One-dimensional simplified resin flow model has been used, while the temperature field has been considered two-dimensional in the plane x,z (x is the flow resin direction and z the thickness direction). A chemo-rheological model has been introduced to analyze the influence of the temperature and the cure degree on the viscosity.

Our model is based on the following hypothesis:

the resin is assumed incompressible and newtonian;

the flow in the fiber preform is assumed to be governed by the Darcy law;

the tensor permeability is symmetric and constant;

thermal equilibrium between the liquid and solid phases, i.e. the resin and the fiber preform at each point at the left of the resin flow front have the same temperature.

For Darcy flow the average velocity \underline{v} of the resin is given by the expression:

$$v = -\frac{K}{\mu} \sum_{-}^{K} p \tag{1}$$

where $K_{=}$ is the permeability of the porous medium, ∇p the pressure gradient and μ the viscosity.

Applying our assumptions and the continuity equation to the equation 1), the governing equation for the pressure distribution becomes:

$$\frac{d}{dx} \cdot \left(\frac{K_x}{\mu} \frac{dp}{dx}\right) = 0 \tag{2}$$

 K_x is one of the principal components of the permeability tensor. Since the permeability tensor is symmetric, it's possible to diagonalyze it everywhere in the mold, i.e. to find a principal coordinate system. The boundary conditions on equation 2) are zero pressure at the flow front and constant injection pressure.

By applying the boundary conditions, we have the following solutions for the pressure distribution and the resin velocity:

$$p(x) = \frac{p_0}{\int_{0}^{x_{fr}} \mu(x) dx} \left\{ \left[\int \mu(x) dx \right]_{x = x_{fr}} - \int \mu(x) dx \right\}$$
(3)

$$u = \frac{dx}{dt}\Big|_{fronte} = K_x \frac{p_0}{\int_0^{x_{fr}} \mu(x) dx}$$
(4)

where x_{fr} denotes the location of the resin flow front at a fixed time.

The heat balance equations have been considered for the walls of the mold, for the dry fibers and for the resin-fibers system, accounting, in all cases, for the conduction heat transfer along the x and z directions and the time variation term. For the resin-fibers system the energy equation includes also the convection heat transport and the curing reaction heat.

The energy equations have the following forms:

$$\left[\rho_{r}c_{pr}\left(1-V_{f}\right)+\rho_{f}c_{pf}V_{f}\right]\frac{\partial T_{c}}{\partial t}+\rho_{r}c_{pr}\left(1-V_{f}\right)\mu\frac{\partial T_{c}}{\partial x}=K_{xc}\frac{\partial^{2}T_{c}}{\partial x^{2}}+K_{zc}\frac{\partial^{2}T_{c}}{\partial z^{2}}+\rho_{r}H_{r}\left(1-V_{f}\right)\dot{\alpha}$$
(5)

$$\rho_f c_{pf} \frac{\partial T_f}{\partial t} = K_{xf} \frac{\partial^2 T_f}{\partial x^2} + K_{zf} \frac{\partial^2 T_f}{\partial z^2}$$
(6)

$$\rho_m c_{pm} \frac{\partial T_m}{\partial t} = K_{xm} \frac{\partial^2 T_m}{\partial x^2} + K_{zm} \frac{\partial^2 T_m}{\partial z^2}$$
(7)

where the pedix r and f relate to the properties of the resin and the fibers respectively, $\dot{\alpha}$ is the rate of the reaction, H_r is the heat generated by the curing reaction, V_f is the fiber volume fraction, ρ , c_p and k are the density, the heat capacity and the thermal conductivity. To complete the model, the resin conservation equation has to be specified:

$$\frac{\partial \alpha}{\partial t} + u \frac{\partial \alpha}{\partial x} = \dot{\alpha} \tag{8}$$

 α is the degree of cure. The second term accounts for the convection of the degree of cure due to the resin flow inside the mold.

INITIAL AND BOUNDARY CONDITIONS

The initial conditions for the equation are the following:

t=0	x=0	z ### [0, a[;]δ-a, δ]	$T=T_{0p}$	(9)
t_0	<i>т шин</i> 10 11		$\mathbf{T}_{-}\mathbf{T}$	

t=0 x ###
$$]0, 1]$$
 z ### $[a, 0-a]$ $I = I_{0f}$ (10)
t=0 x=0 z ### $[a, \delta-a]$ $T=T_{0c}$ (11)

t=0 x=0 z ### [a,
$$\delta$$
-a] α =0 (12)

 T_{0c} is the resin-fiber system temperature, T_{0p} the mold temperature and T_{0f} the fibers temperature. The thermal equilibrium approach has been used also for the injection zone, where both the temperature and the conductive flux present a discontinuity. Along the injection zone the temperature T_{0c} of the resin-fiber system respects the following heat balance equation:

t≥0 x=0 z ### [a, δ-a]

$$\rho_r c_{pr} (1 - V_f) T_{0r} + \rho_f c_{pf} V_f T_f = \left[\rho_r c_{pr} (1 - V_f) + \rho_f c_{pf} V_f \right] T_{0c}$$
(13)

The heat conductive transport has been neglected. At the starting the fibers temperature T_f is T_{0f} , while during the filling T_f has been considered equal to the temperature T_c , evaluated with the energy equation 5). T_{0r} is the injection resin temperature.

The boundary conditions along the external section are:

$$\begin{array}{cccc} x=0 & z \# \# [0, a[;]\delta - a, \delta] & T=T_p & (14) \\ x \# \# [0, 1] & z=0, \delta & T=T_p & (15) \end{array}$$

$$x = l$$
 $z ### [0, \delta]$ $T = T_p$ (16)

x=0 z ### [a,
$$\delta$$
-a] T=T_{0c} (17)

x=0 z ### [a,
$$\delta$$
-a] α =0 (18)

The continuity of the temperature and the conductive flux have been imposed along the rigid interface section:

x ###]0, x_{fr}[z=a,
$$\delta$$
-a $T_c=T_m$ $K_{zm}\frac{\partial T_m}{\partial z} = K_{zc}\frac{\partial T_c}{\partial z}$ (19)

x ###]
$$\mathbf{x}_{fr}$$
, l-a[z=a, δ -a $T_f = T_m$ $K_{zm} \frac{\partial T_m}{\partial z} = K_{zf} V_f \frac{\partial T_f}{\partial z}$ (20)

x=1-a z ### [a,
$$\delta$$
-a] $T_f = T_m$ $K_{xm} \frac{\partial T_m}{\partial x} = K_{xf} V_f \frac{\partial T_f}{\partial x}$ (21)

The last boundary condition is the specification of the resin temperature along the flow front surface. At this stage we investigate the influence of the heat transport along the resin flow front surface on the thermal profiles. The temperature front respects a heat balance equation on the moving surface, where theoretically the temperature is continues and the conductive flux presents a discontinuity. The boundary condition is: $x=x_{c}$, z, ### la δ -a[

$$uT_{fr}\left[\rho_{f}c_{pf}V_{f} + \rho_{r}c_{pr}\left(1 - V_{f}\right) - \rho_{f}c_{pf}V_{f}\right] = -K_{xc}\frac{\partial T}{\partial x}\Big|_{x_{fr}^{-}} + K_{xf}V_{f}\frac{\partial T}{\partial x}\Big|_{x_{fr}^{+}}$$

In the fig.2 the qualitative thermal profiles along the moving surface are reported. \underline{v} is both the resin velocity and the speed propagation of the conductive flux discontinuity.



Fig.2: Thermal profiles along the moving surface.

(22)

Generally the thermal front condition is simpler and consists of imposing a constant temperature front equal to the fibers mat temperature [1] or formulating a balance equation that neglects the fibers conductive flux [2-5]. This is surely true near the end of the filling but not at the starting when the temperature gradients between the resin and the fibers mat are high.

CURE AND CHEMO-RHEOLOGICAL MODEL

The only terms which still need to be defined are the rate of reaction $\dot{\alpha}$ and the dependence of the viscosity on the degree of cure and on the temperature. The numerical simulations have been performed with an epoxy resin.

For this resin, the following models represent the curing and chemo-heological behavior:

$$\dot{\alpha} = 2f[\mathbf{I}]_{t=0}K_p(1-\alpha) \cdot \left[1 - \exp(-K_d t)\right]$$
(23)

$$\mu = A_{\mu} \exp\left(\frac{E_{\mu}}{RT}\right) \left(\frac{\alpha_g}{\alpha_g - 1}\right)^r$$
(24)

where

$$K_d = A_d \exp\left(-\frac{E_d}{RT}\right) \tag{26}$$

$$K_{p} = A_{p0} \exp\left(-\frac{E_{p0}}{RT} \left(1 - \frac{\alpha}{\alpha_{f}}\right)^{m}\right)$$
(27)

$$F = a_1 + a_2 \left[\alpha - \frac{c_1 \left(T - T_{ref} \right)}{c_2 + \left(T - T_{ref} \right)} \right] + a_3 \left[\alpha - \frac{c_1 \left(T - T_{ref} \right)}{c_2 + \left(T - T_{ref} \right)} \right]^2$$
(28)

In the following tables the values of the above parameters have been reported:

Table 1 : Kinetic parameters.				
f	0.1			
m	4			
$\alpha_{ m f}$	0.68			
A _d	$5.55*10^{16}$			
A _{p0}	$0.38*10^{10}$			
E _d (J/mole)	$1.41*10^5$			
E _{p0} (J/mole)	4.27*104			
[I] _{t=0}	0.005208			
[R] _{t=0}	0			
H _r	2.44*10 ⁵			

A_{μ} (N s/m ²)	2.16*10-4
E_{μ} (J/mole)	26214
a ₁	-0.9621
a ₂	6.482
a ₃	-3.205
c ₁	2.131
c ₂	374.6
ag	1
$T_{ref}(K)$	328

Table 2 : Rheological parameters.

NUMERICAL IMPLEMENTATION

The domain has been discretized by using a constant step dz in the thickness direction and a variable step dx in the flow direction, determined from the advancement of the resin flow front. At each time the resin advancement has been determined with the equation 4), that has been discretized in the following form:

$$dx_{i} = K_{x} \frac{p_{0}dt}{\sum_{i=0}^{N} \frac{1}{2} (\mu_{i} + \mu_{i-1}) dx_{i}}$$
(29)

The integral of the viscosity distribution has been evaluated with the trapezoidal rule.

The energy equations and the resin conservation equation have been discretized with the finite difference method using the Crank-Nicolson method. The spatial derivative have been evaluated with central or backward formulas relating to the conductive or convection term respectively.

The resolution has been done with an iterative algorithm until the convergence of the temperatures.

RESULTS

The numerical simulations have been performed with parameters similar to Wang et al.[7].We have used an aluminum mold (thickness 158mm). The dimension of the mold cavity was 254*9.48 mm. In the table 3 the thermal properties of the resin and the fiber glass have been reported.

Property	Resin	Fiber
Density	1100	2560
(Kg/m3)		
Specific heat	1680	670
(J/kg K)		
Thermal	0.168	0.417
conductivity		
(W/m K)		

Table 3 : Thermal properties of the resin-fiber system.

A random fiber mat with a porosity of 0.8 has been used. The permeability value of this mat has been extrapolated from the data of [8]. The initial temperatures of the mold walls and the

fiber mat have been set at 62 °C. The injection pressure and temperature of the resin were respectively $4.48 \times 10^5 \text{ N/m}^2$ and 29 °C.

Under these conditions, the filling time is resulted 124.5s. The figure 3 shows the temperature field in the whole system at the end of the filling. In the first part of the mold the temperature is lower than in the last zone, where the curing reaction starts. In this zone the temperature attains the highest value 62.3 °C at the center of the mold.

During the filling, before the resin reaches each location inside the mold, the temperature is around the initial temperature (62° C). Then, due to the heat convective transport of the resin, a cooling can be observed until the end of the filling. This behavior can be noticed in the figures 4 and 5, where the temperature of six locations (fixed in figure 1) as function of the time have been reported.

The figures 4 and 5 refer respectively to the locations x_1 and x_2 , 8.5 cm and 17 cm far from the injection section (see fig.1). Along the interface with the mold walls (locations 1,4), due to the high conductivity of the aluminum mold, the temperature is constant.



Fig.3: Temperature distribution at the end of the filling (124.5 s).



Fig.4: Temperature profiles at the location x_1 *.*



*Fig.5: Temperature profiles at the location x*₂*.*

Along the thickness a temperature gradient can be observed both the filling and the curing stage. Since the curing reaction starts in the last part of the mold and propagate in the other parts, exothermic temperature peaks are present at different times. The figures 6 and 7 show the temperature and degree of cure fields after 300 s. The maximum value of the temperature is 93 °C.



Fig.6: Temperature distribution after 300 s.



Fig.7: Degree of cure distribution after 300 s.

The curing reaction starts in the last part of the mold for the heating of the walls mold, then, due to the reaction heat, propagates in the other zones. Along the thickness, a gradient of conversion can be noticed as shown in figure 8, where the conversion distribution after 1000 s has been reported.



Fig.7: Degree of cure distribution after 1000 s.

Moreover, in the first zone of the mold along the flow front surface our numerical simulations predict the conductive flux discontinuity. Near the flow front surface the dry fibers temperature is influenced by the resin flow, i.e. a cooling of the dry fibers can be observed, as shown in figure 8. At the starting of the filling, this effect is important and can have influence on the thermal profiles and on the filling time.



Fig.8: Thermal profiles along the flow front surface at the location 3(18 s).

CONCLUSIONS

A model for the simulation of the RTM process has been developed. In our model a particular boundary condition has been specified along the flow front surface, where the temperature front respects a jump equation. At the starting of the filling, along this section the conductive flux presents a discontinuity and the dry fibers temperature decrease sharply. Since this effect can have influence the thermal profiles and the filling time, further investigations have to be made. The numerical simulations have been performed by using an epoxy resin and a glass fibers system and parameters similar to Wang et al. Results well predict qualitatively the experimental literature data. As the experimental data, a cooling can be observed at each location after the arrival of the resin until the end of the filling. Further exothermic peaks are present at the center of the mold.

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