Determination of the Local Thermal Heat Exchange during Resin Flow in a Fibrous Medium

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Abstract

Liquid composite molding processes generally involve injection of a polymeric resin in a fibrous preform previously placed in a closed mold. Resin kinetics largely depend on the temperature cycle applied and, as far as thick composites parts are concerned, they can greatly impact on the temperature profile, especially in the core of the part where high temperature range can be reached, affecting the part mechanical properties. Thermal analysis of the system is usually done at the macro-scale level. However, at micro level and because of resin flow across the fibrous preform, local thermal effects have to be considered. A heat dispersion coefficient for instance will account for the hydrodynamic effects so as to improve significantly the accuracy of the temperature profile prediction at steady state. To improve prediction of transient temperature profiles, local heat transfer between resin and fibers needs to be considered. The characterization of this coefficient is conducted following an inverse method, numerical solutions parametered by this coefficient being derived from a non-local thermal equilibrium (or two-equation model) and compared with experimental temperature profiles drawn for several injection velocity cases. Significant improvement in the prediction of transient temperature profile is then obtained. Correlation between the injection velocity and the local heat exchange coefficient is also shown.

Introduction

Liquid Composite Molding injection processes are widely used in the aerospace and automotive industries. They consist in injecting resin into a fiber preform placed in a one or two-sided closed mold. Resin Transfer Molding is one of these processes where a two sided mold for fast manufacturing cycles can be obtained. The fiber reinforcements considered are glass, carbon or aramid fibers, through which a reactive thermoset resin is forced to flow. Thermoset resins involve a curing stage that can last more than half the cycle time. In order to reduce this step, the mold is heated as the curing reaction can be catalyzed by heat, and cold resin (at room temperature) is injected. Prediction of the curing time is important so as to assure a complete filling of the mold before the resin starts to gel. As the polymerization process strongly depends on the temperature cycle, accurate prediction of the temperature profile during the filling stage is required. Beside heat conduction through the mold walls and convection of heat as the resin flow front advances, local heat transfer phenomena due to the flow of resin though the porous medium have to be taken into account. The two principal ones, heat dispersion and local heat transfer, are presented and discussed here. In particular, a methodology to characterize local heat transfer coefficient between resin and fibers is proposed.

Nomenclature

a_{fm} : specific area
Cp: specific heat at constant pressure
d_p : composite length scale
h: reference thickness
h_{fm} : heat exchange coefficient
H: heat exchange parameter
k: thermal conductivity
k_e: effective thermal conductivity
k_D: dispersion coefficient
T: temperature
t: time
$<v_f>$: average flow front velocity

Subscripts and superscripts
f: fabric

m: matrix

e: composite

ρ: density

ϕ: porosity

Pe: Peclet Number

K: permeability tensor

η: viscosity

α: thermal diffusivity

Background : Heat transfer models in LCM processes

Heat transfer in composite manufacturing involves conduction of heat towards the mold walls, convection by the resin as it flows through the preform and heat generation as the resin cures. For LCM processes, two heat transfer models are reported in the literature. The Non Local Thermal Equilibrium (NLTE) model or the two-equation model is derived by considering the energy balance in the two components (resin and fibers) separately [1-5]:

$$\phi(\rho C_p)_m \frac{\partial\langle T_m\rangle}{\partial t} + (\rho C_p)_m \nabla(\langle v_m\rangle\langle T_m\rangle) = \nabla(K_m \bullet \nabla\langle T_m\rangle) + H(T_m - T_f) + \phi\dot{s} \quad (1.)$$

$$(1-\phi)(\rho C_p)_f \frac{\partial\langle T_f\rangle}{\partial t} = \nabla(K_f \bullet \nabla\langle T_f\rangle - H(T_m - T_f)) \quad (2.)$$

where ρ, *K* and *Cp* are the density, the heat conductivity tensor and the heat capacity of the materials, ϕ is the porosity of the porous media, $\langle v_f \rangle$ is the injection average velocity, $\dot{s}$ is the heat generated by the curing reaction, and the subscripts *f* and *m* refer to the fiber (fabric) and to the resin (matrix) properties. *H* is the heat exchange parameter and is defined by [6] :

$$H = a_{fm} h_{fm} \quad (3.)$$

where h_{fm} is the heat exchange coefficient between resin and fibers, and a_{fm} is the specific area representing the laminate architecture at tow scale, expressed for a woven fabric by [7]:

$$a_{fm} = \frac{4}{d_p} \quad (4.)$$

d_p being the length scale of the composite, set to be a fiber tow diameter as far as woven fabric are considered.

This model is precise but difficult to use as the heat exchange coefficient needs to be determined [8-12]. The alternative is to use the Local Thermal Equilibrium (LTE) model or the one-equation model based on the assumption that local resin and fibers temperatures are equal :

$$\langle T_m \rangle = \langle T_f \rangle = \langle T \rangle \qquad (5.)$$

Applying the LTE assumption to equations 1 and 2 and using the volume averaging method [5], the one-equation model is obtained, assuming that no curing reaction is involved:

$$\left(\phi\left(\rho C_p\right)_m + (1-\phi)\left(\rho C_p\right)_f\right)\frac{\partial\langle T\rangle}{\partial t} + \left(\rho C_p\right)_m \langle v_m\rangle \cdot \nabla\langle T\rangle = \nabla \cdot \left(K_e \cdot \nabla\langle T\rangle\right) \qquad (6.)$$

The effective conductivity tensor K_e is also called the stagnant conductivity and may be estimated as the average of the materials thermal conductivities, as described below :

$$K_e = \phi K_m + (1-\phi) K_f \qquad (7.)$$

This assumption is valid under certain conditions. For instance, the temperature gradient between the two phases must be negligible, thus that the ratio of the thermal conductivities should be approximately unity. Numerical computations show that almost no temperature difference exists when non-metallic materials are considered, thus that the local thermal equilibrium assumption can be valid for LCM processes [13]. However, using one model or the other depends on the injection parameters. For instance, it was shown that the one-equation model breaks down when high injection velocities are used or when a significant heat generation occurs in one of the two phases [13-16].

Lin et. al developed a numerical model to compare the LTE with NLTE [10]. Lagrangian coordinate system was used in the flow front region whereas an Euler coordinate system was used in the main flow region for better accuracy. At low flow rates, both methods gave similar results. At high flow rates, the LTE model deviated from NLTE.

Quintard and Whitaker [12] also obtained similar conclusions. Jiang, et. al [13] also showed that the local equilibrium model and the two-equation model give similar results as they observed that small temperature gradients develop between the fluid and solid phases when glass materials are used as solid phase. They also pointed out that better results are obtained with NLTE when constant heat flow is assumed at the boundary compared to constant temperature boundary conditions.

Heat dispersion during liquid flow in porous media

Issues in heat transfer in porous media are directly linked with the fact that resin can not follow a unidirectional path as it fills the preform. Although at the macro-scale the resin velocity profile can be considered as uniform, at the micro-scale and because of the preform structure, the resin flow is disturbed, creating local convective effects. Local variations of velocity [5] are associated with the thermal conductivity tensor K by adding a thermal dispersion coefficient K_D to the effective thermal conductivity K_e in the one-equation model.

$$K = K_e + K_D \quad (8.)$$

This coefficient is expected to depend on the velocity of the flow front, on the porosity and structure of the medium and on the thermal characteristics of the materials [1-5].

Very few values of the dispersion coefficient calculated from experimental work are found in the literature, especially because experimental work is difficult to conduct. Dessenberger and Tucker [5] experimentally investigated the dispersion coefficient as a function of the Peclet number. They used a radial injection scheme in a two-layers random glass fiber preform and tracked the temperature at three locations along the radius. The dispersion coefficient was characterized by an optimization routine using the Local Thermal Equilibrium model. They found that the dispersion coefficient would increase linearly with the Peclet number being between 0.1 and 1.2, then it would linearly decrease with $\mathrm{Pe}^{-5.775}$, and for Peclet number higher than 2.5, the dispersion would remain constant. Metzger et al. also confirmed the linearity of the transverse dispersion coefficient with the Peclet Number [17].

Hsiao et. al [18] obtained good result when comparing experimental temperature in the mid-plane with a numerical solution. Their results showed a linear relation of the dispersion coefficient with the Peclet number for different material structures created from different fiber preform architectures.

Characterization of the heat dispersion coefficient was performed for a given resin /fiber system [19] from an inverse method based on an analytical solution derived from the one-equation model at steady state [20-21] under the following conditions and assumptions:

- the inlet injection flow rate remains constant throughout the injection,
- no curing reaction is initiated,
- the fibrous materials used are isotropic in the in-plane directions,
- no resin viscosity change with temperature, which is a valid assumption as far as thermoset resins are concerned provided that no curing reaction is initiated.
- the dispersion along the in-plane directions can be neglected, as long as the thickness is at least 10 times smaller than the plane dimensions.
- as the conduction mainly takes place in the through-thickness direction, the in-plane conduction term is not considered.

The data needed for the determination of the dispersion coefficient are the fiber volume fraction, the thermal properties of the materials, and the mold walls temperature. The heat dispersion coefficient is defined by an optimization program that helps to determine the steady state numerical temperature profile along the flow front direction closest to the experimental one. Steady state temperature profiles obtained numerically and experimentally are shown on Fig. 1.

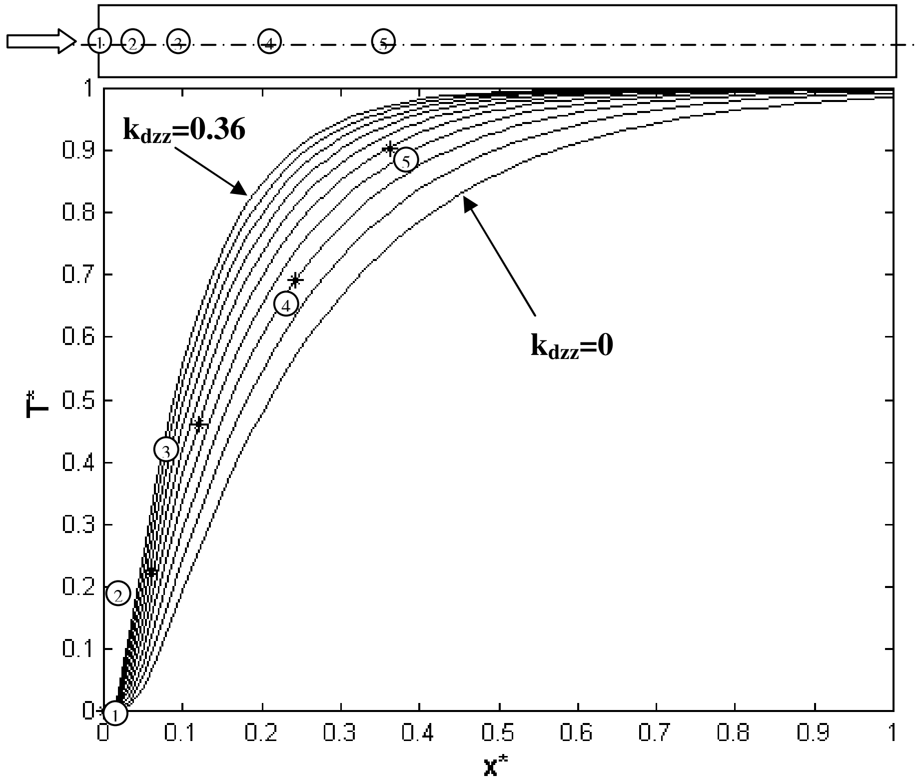

Figure 1 Numerical steady state temperate profiles (solid lines) obtained for different heat dispersion coefficient vs. experimental temperature profile (stars)

Good correlation at steady state is thus obtained between the experimental and the analytical temperature profile along the mold when heat dispersion is considered. However, at the transient stage, large discrepancies are observed, as shown in Fig. 2. It was thus shown that the one-equation model was accurate in predicting the temperature profile at steady state during a LCM injection. When fast injection schemes or highly reactive resin systems are involved, steady state is not necessarily reached. For these conditions, the two-equation model is required, thus accurate characterization of the heat exchange coefficient h_{fm} would be needed. Next section is dedicated to the description of the h_{fm} coefficient characterization methodology and on the results obtained.

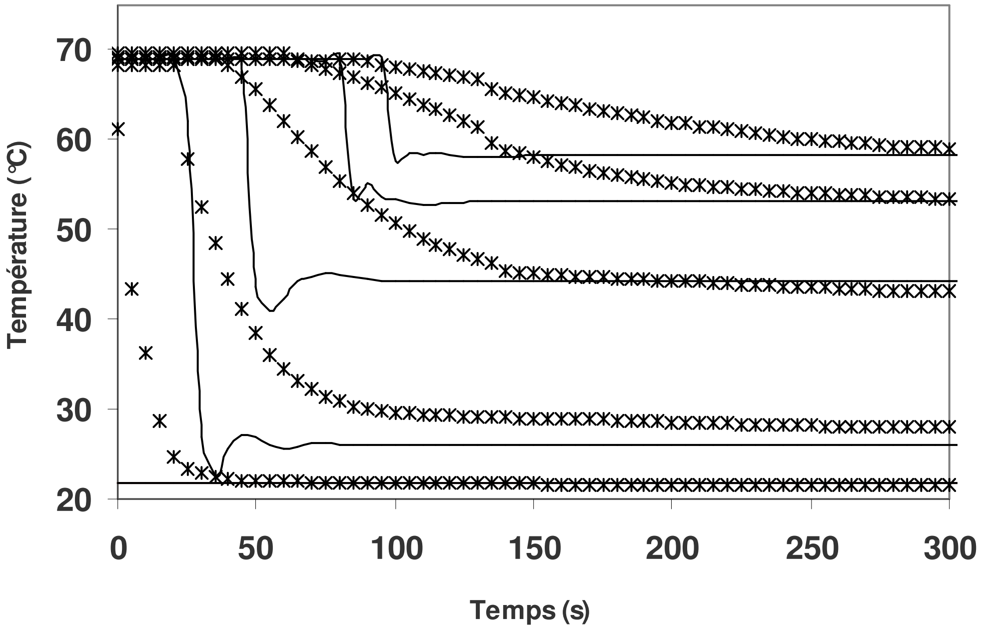

Figure 2 Numerical (solid lines) and experimental (dotted lines) transient temperature profiles at different location along the flow front

The dependence of the dispersion coefficient with the velocity was studied in the linear and in the radial [19]injection case in order to establish a relation between the thermal dispersion and the Peclet number define as:

$$Pe = \frac{\langle v_f \rangle d_p}{\alpha} \quad (9.)$$

where d is the diameter of the fiber tows, $<v_f>$ is the resin average velocity and α is the thermal diffusion of the fluid phase.

A linear correlation was found between the heat dispersion coefficient and the Peclet number, either for a glass or a carbon fiber preform.

Heat exchange coefficient determination

Attempts to numerically characterize the heat exchange coefficient at the pore size level showed little accuracy. Generally, the preform is modeled at the pore size level where fibers or fiber tows are modeled as cylinders or square rods. The velocity profile is determined and then the local heat transfer between the fabric and the fibers is calculated, using the non-local thermal equilibrium model. Nakayama and Kuwahara [22] modeled a closed cell as shown on Fig. 3 to represent the flow around a cylindrical rod.

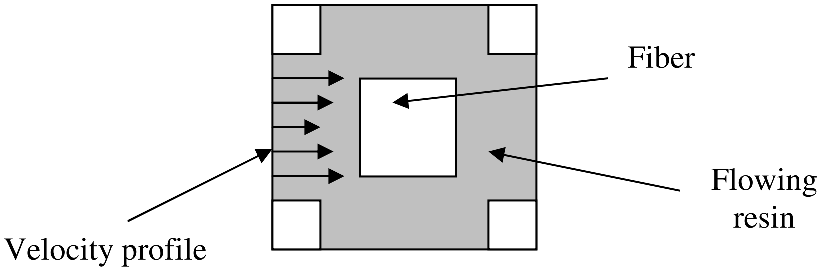

Figure 3 Schematics of a unit cell approach

Other examples of heat exchange coefficient experimental characterization can be found in the literature. Ramond et. al [23] used infrared thermogravimetry on thin plates where a two equation model was needed as the composite medium considered was a polymeric matrix in a metallic network. The experiment is based on the thermal response of the plate under a flash excitation.

The method presented here for the heat exchange coefficient is based on an inverse method, the numerical solution being derived from the two-equation model under the same conditions and assumptions as for the derivation of the analytical solution for the heat dispersion characterization. Under those assumptions, the two-equation model becomes :

$$\phi\left(\rho C_p\right)_m \frac{\partial T_m}{\partial t} + \left(\rho C_p\right)_m \langle v_m \rangle \frac{\partial T_m}{\partial x} = \left(k_m + k_d\right)\frac{\partial T_m}{\partial z^2} + H\left(T_f - T_m\right) \quad (10.)$$

$$(1-\phi)\left(\rho C_p\right)_f \frac{\partial T_f}{\partial t} = k_f \frac{\partial T_f}{\partial z^2} - H\left(T_f - T_m\right) \quad (11.)$$

The equations are simultaneously solved using a multiphysics FE-based calculation tool (COMSOL Multiphysics 3.2). The program is inserted in an optimization program written with Matlab 6.5. The numerical temperature-histories for different locations along the flow direction are compared to experimental temperature profiles. The closest numerical solution to the experimental ones will determine the heat exchange coefficient corresponding to the injection considered.

Experimental set-up

The fiber reinforcement used is a plainweave 5x4 woven fabric (Veterotex America) with an area weight of 816g/m^2. To obtain a preform with 49% volume fraction, 16 layers are necessary for a mold gap of ½ inch (1.24 cm). Figure 4 presents the fiber preform placed in the mold. The resin system used is a vinylester resin (Derakane 411C50, Dow Chemichals) with a viscosity of 0.2 Pa.s in average, initiated with 1.5% of trigonox. No catalyst was added, as the curing reaction can be initiated by heat. Materials thermal characteristics are reported in Table 1. The resin exothermic peak is expected to occur three hours after being held at the mold wall temperature, which is a long time after the resin injection stage has been completed so that no heat from the curing reaction is released. The thermocouples were placed in the mid-thickness of the mold, taped to the fiber layer as shown in Fig. 4. The first thermocouple is placed at the inlet to record the inlet temperature. Then, two thermocouples are placed one inch apart from each other, five other thermocouples are taped every two inches. The recording of the temperature at the different locations is continued until the end of the experiment. When the resin reaches the heated fibers, the temperature drops, as the average temperature of resin and fiber will be lower as the injected resin is cold.

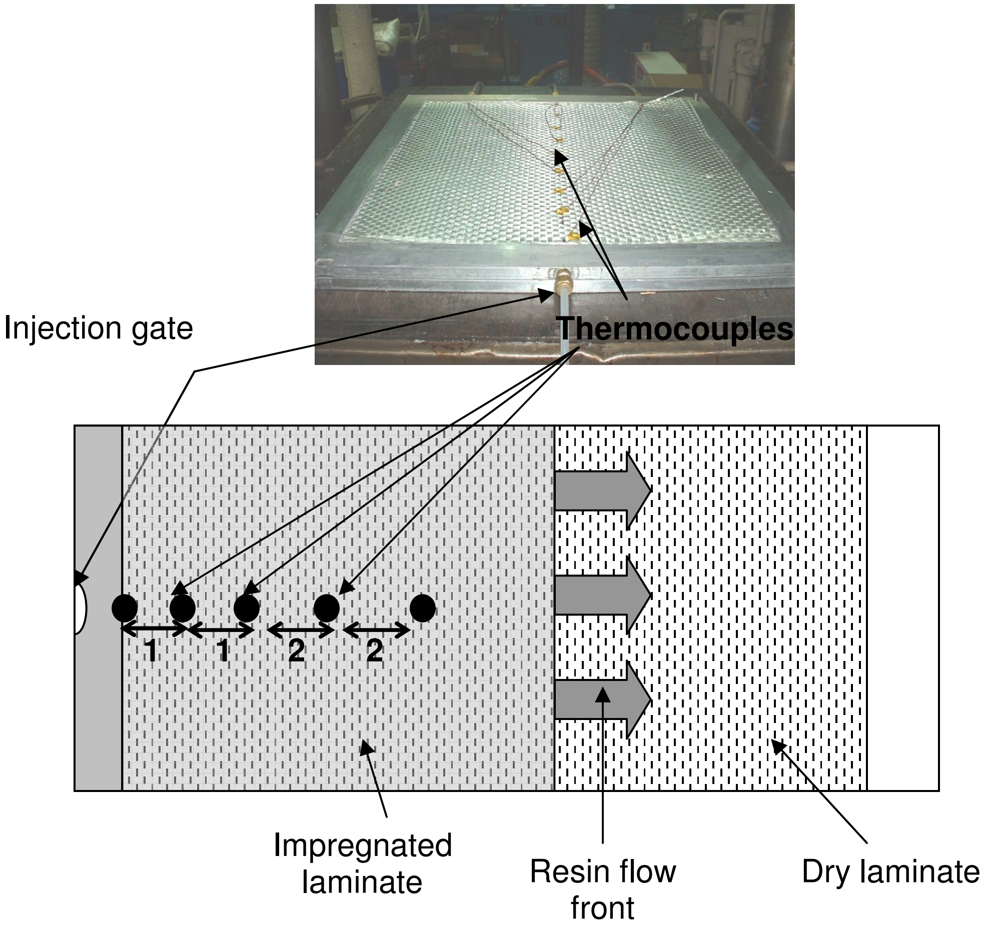

Figure 4 Experimental injection set-up and thermocouples location (unit : inch)

Three experiments were conducted at different injection velocities. First step is to define the heat dispersion coefficient by comparing steady state temperature profiles from analytical solutions with the experimental one, as described in a previous section. Then numerical temperature history can be compared to the experimental temperature profiles so as to define the heat transfer coefficient between fibers and resin.

Table 1 Material properties

Characteristics	Symbol	Unit	Vynilester resin	Glass fibers
Viscosity	μ	Pa.s	0.2	N/A
Area weight	A	g/m^2	N/A	816
density	ρ	Kg/m^3	1040	2560
Thermal conductivity	K	W/m.K	0.110	0.417
Specific heat capacity	C_p	J/KgK	1350	670

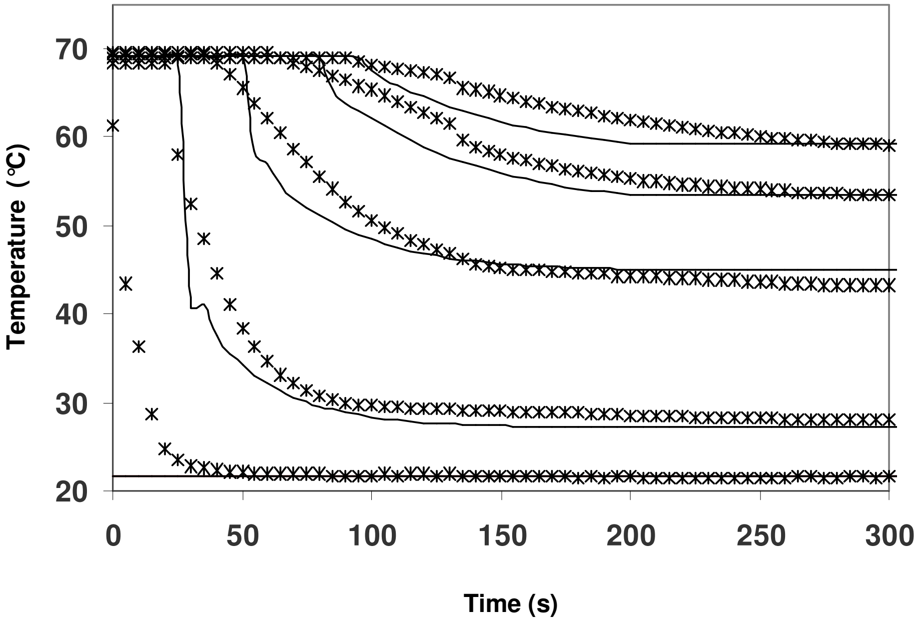

Figure 5 Numerical (solid lines) and experimental (*) temperature profiles

Data analysis

The heat transfer coefficient is determined by an optimization program that searches for the numerical solution parametered by the heat transfer parameter *H* closest to the experimental temperature profile.

Numerical curves are more similar to experimental curves when a two-equation model that includes the heat exchange effects between the resin and the fibers is used (Figure 5). Fig. 6 represents the experimental temperature profiles along with the numerical solution derived from the one- and the two-equation model. For the first thermocouple located at the mold inlet, the two numerical solutions are superposed in the two first cases. Local thermal discrepancies of 15°C can be reached between the two numerical models.

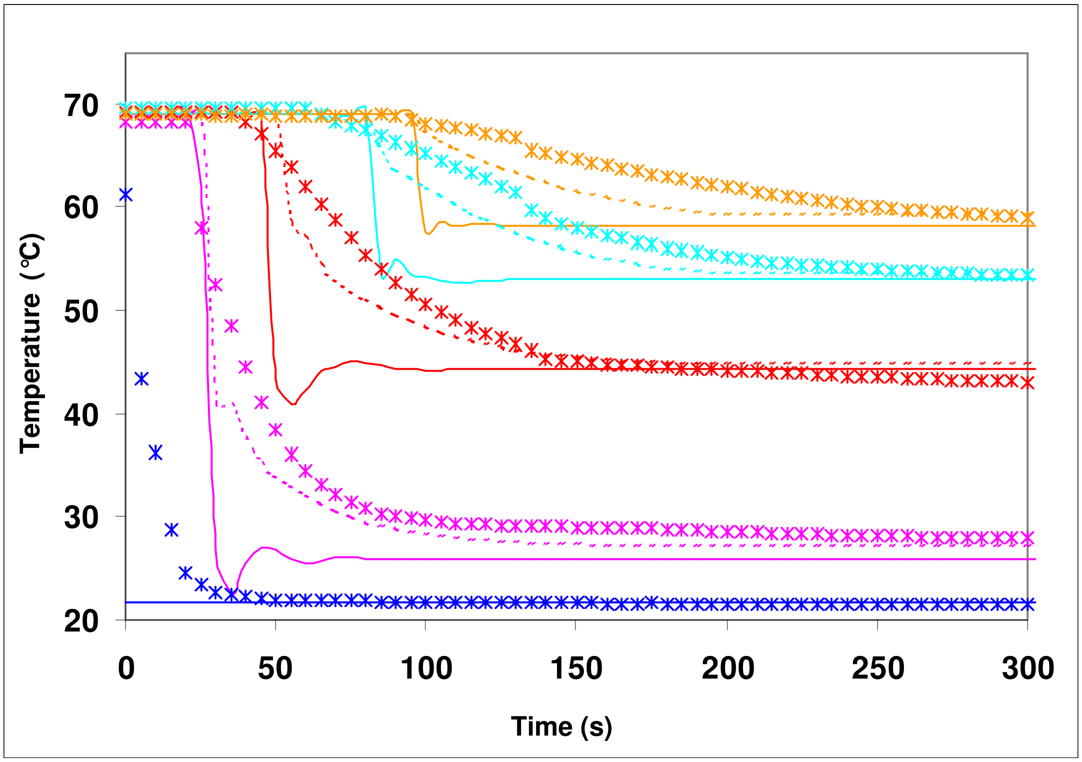

Figure 6 Comparison between one-equation model (solid lines), two-equation model (dotted lines), and experimental profiles (*) at different locations along the flow front direction

The heat exchange coefficient obtained for the three different injection velocities are reported on Fig. 7 along with the injection velocity.

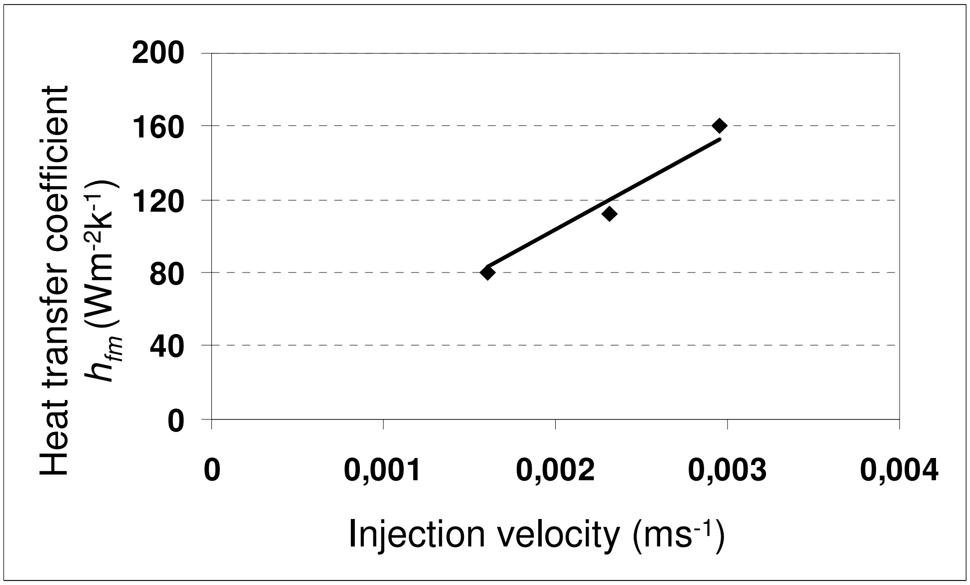

Figure 7 Heat transfer coefficient vs. injection velocity

According to Fig. 7, the heat exchange coefficient is a linear function of the injection velocity over the range studied with values that agree in order of magnitude with the literature [24-25]. The order of magnitude observed for the coefficient is similar to the one usually observed for organic composite materials. When the injection velocity increases, forced convection effects also increase, thus higher heat exchange coefficient is measured.

Discussion

Injection velocity is one of the main parameters that helps defining if the two-equation model is necessary for accurate temperature prediction during injection. For fast injection schemes, heat transfer between resin and fibers will have to be considered as the flow front would reach the end of the mold while local heat transfer will still be in a transient stage. On another hand, for low injection schemes, the temperature will homogenize between the resin and the fibers as fast as the resin flow front advances. The other parameter to take into account is the preform thickness. For thin parts (few millimeters), heat conduction will be faster than heat transfer form the rein to the fibers. So the one-equation model is sufficient in that particular case. If a thick sectioned part is now considered, heat conduction in the core of the material will have less importance compared to heat exchange between resin and fibers. The field of application of each equation model thus depends on the injection parameters as well as on the part geometry. The LTE model would be used for dispersion coefficient characterization or when steady state analysis can be sufficient, like in slow processes, or when poorly reactive resin systems are considered. NLTE model would be required for the other cases.

Conclusion

For accurate prediction of transient temperature profiles during the filling of a fibrous preform, it is necessary to consider the heat transfer coefficient between resin and fibers at the local scale. Numerical temperature profiles were obtained by implementing the two-equation model in a finite element code. The one-equation model without local heat exchange effects was also studied. It was shown that large temperature difference between the prediction of the two models can be obtained. The one-equation model would only give accurate temperature prediction at steady state. This model should only be used in slow processes, with slow temperature changes, whereas the two-equation model is needed for fast injection processes in order to correctly predict the curing kinetics of the resin system.

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