

# Non-isothermal simulation of the structural reaction injection moulding process.

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## Abstract

We present a non-isothermal simulation model for the structural reaction injection moulding of thin parts. This model takes the effect of mechanical dispersion into account in the energy and species balance, by assuming perfect mixing in the gap and negligible mixing along the mid-surface. Several examples are analysed, which highlight the effect of the heated walls on the general heat transfer, especially when curing is considered.

## INTRODUCTION

Among the various kinds of moulding processes, structural reaction injection moulding (SRIM) occupies a very specific place for the manufacturing of large fibre reinforced parts. In this process, a reactive mixture (activated either by heat or by mixing) is injected into a mould where a continuous fibre preform has been placed beforehand. The resin fills the spaces between the fibres, and the final part is demoulded after sufficient curing. During injection, fibre orientation remains mostly unchanged, which allows for a good control of the mechanical properties of the part. Even if several attempts to use a thermoplastic resin instead of a thermoset have been reported in literature, the relatively low viscosity of the reactive mixture remains a serious advantage of thermosets, regarding injection pressure and wetting of the reinforcement.

Due to the complex resin flow through the fibre preform, and the coupled influence of chemical reactions and temperature (both on each other and on the viscosity), numerical simulation represents a very useful tool in order to better understand the different effects affecting the final part quality. This paper presents a model for the non-isothermal simulation of the structural reaction injection moulding process, together with the influence of the reaction on the filling. We investigate the thermal behaviour of the resin and the reinforcement, with the assumption of perfect mechanical dispersion in the gapwise direction and negligible dispersion in the mid-surface directions. All the unknown fields are therefore two-dimensional.

## GOVERNING EQUATIONS

The equations governing the filling stage of SRIM can be derived from a multiphase flow analysis [1,2]. In what follows, subscripts  $f$  and  $r$  relate to the (solid) fibre mat and the (liquid) resin, respectively. The flow of the resin through the porous mat is governed by the

Darcy equation :

$$\alpha_r \underline{v} = -\frac{1}{\eta} \underline{K} \cdot \underline{\nabla} p \quad , \quad (1)$$

where  $\underline{v}$  and  $p$  are the average velocity and pressure in the liquid phase,  $\alpha_r$  is the resin volume fraction (also referred to as the porosity),  $\eta$  is the viscosity (depending on temperature and reaction degree), and  $\underline{K}$  is the permeability tensor of the fibre mat. Both  $\alpha_r$  and  $\underline{K}$  may vary throughout the mould.

Considering the filling of thin parts of thickness  $2h$ , with an incompressible Newtonian mixture and through a non consolidating and stationary fibre preform (both with constant thermal properties), the pressure field can be shown to be two-dimensional. The continuity equation is written as

$$\underline{\nabla}_m \cdot (\underline{S} \cdot \underline{\nabla}_m p) = 0 \quad , \quad (2)$$

where  $\underline{\nabla}_m$  denotes the constrained gradient in the mid-surface directions. The fluidity tensor  $\underline{S}$  is obtained from the viscosity and the permeability tensor by a gap-averaging procedure :

$$\underline{S} = \int_{-h}^h \frac{1}{\eta} \underline{K} dz \quad . \quad (3)$$

Due to the usually higher out-of-plane permeability, the velocity profile is essentially parallel to the mid-surface of the cavity.

The energy balance is obtained using the thermal equilibrium hypothesis, which assumes that a unique temperature is observed at any location in the cavity for both resin and mat. Letting  $\rho_r$ ,  $\rho_f$ ,  $c_{pr}$  and  $c_{pf}$  stand for the specific masses and heats of the resin and the fibres, and  $\Delta H$  and  $T$  denote the reaction heat and the common resin and mat local temperature, the energy equation is written as [1] :

$$\begin{aligned} & \left( \alpha_r \rho_r c_{pr} + \alpha_f \rho_f c_{pf} \right) \frac{\partial T}{\partial t} + \alpha_r \rho_r c_{pr} \underline{v} \cdot \underline{\nabla} T \\ & = \underline{\nabla} \cdot \left( \left( \underline{k} + \alpha_r \rho_r c_{pr} \|\underline{v}\| \underline{L} \right) \cdot \underline{\nabla} T \right) \\ & + \alpha_r \rho_r \Delta H f(C, T) + \frac{1}{\eta} \underline{\nabla} p \cdot \underline{K} \cdot \underline{\nabla} p \quad , \end{aligned} \quad (4)$$

where the classical terms, accounting for time variation and advection of temperature, conduction, reaction and viscous heating, can easily be identified. In the thermal conduction term, besides the conductivity tensor  $\underline{k}$ , an additional dispersion effect is taken into account, which is associated with the geometrical tortuosity of the preform. This effect is characterised by a mechanical dispersion tensor  $\underline{L}$ .

The species balance is

$$\alpha_r \frac{\partial C}{\partial t} + \alpha_r \underline{v} \cdot \underline{\nabla} C = \underline{\nabla} \cdot \left( \alpha_r \|\underline{v}\| \underline{L} \cdot \underline{\nabla} C \right) + \alpha_r f(C, T) \quad , \quad (5)$$

where  $C$  denotes the reaction degree. Mechanical dispersion is again taken into account in this equation. Both temperature and reaction degree are three-dimensional fields, and equations (4) and (5) are to be solved through the gap. Further simplifications will be considered in the next section.

Two constitutive equations are needed for the viscosity  $\eta$  and the reaction evolution law  $f$ . In the following, we consider a polyester-styrene reactive mixture, together with initiators and inhibitors. The viscosity is a function of temperature and reaction degree, according to the model of Gonzalez and Macosko [3]. We have used the reaction kinetics described by Stevenson [4], whose interest resides in its capability to accurately describe the separate effects of the components on the reactions. The reaction degree  $C$  therefore consists of four fields, namely the initiator, inhibitor and free radical concentrations, and the curing degree.

## RESULTS AND DISCUSSION

### *Isothermal simulation*

In order to illustrate the effect of the reinforcement on the flow front evolution, we first consider the isothermal filling of an automotive bonnet (Fig. 1). Only eqs (2) and (3) are solved, using a method developed in the frame of thermoplastic injection moulding (TIM) [5]. This approach combines finite elements with a sophisticated front tracking and remeshing technique along the mid-surface [6].

Because of the symmetry of the whole part, only one half of the bonnet has been covered by a finite element mesh (Fig. 1.a). The principal axes of the fibre preform are oriented along the x- and y-directions. During filling, several temporary meshes are automatically generated, such as shown in Fig. 1.b. We first consider an isotropic fibre mat, in which case the flow fronts are identical to those that would be observed in TIM (Fig. 1.c). In a second step, when the permeability tensor is non-isotropic (Fig. 1.d), the flow fronts are observed to be tremendously influenced by the reinforcement. Note that the position of the last front is an important information for mould design, in order to avoid air entrapment.

### *Non-isothermal simulation, perfect gapwise dispersion*

During filling, the walls are heated in order to activate the curing reaction before demoulding. Non-isothermal effects must therefore be incorporated in the model. Due to the complex form of eqs (4) and (5), several attempts have been reported in literature to simulate the non-isothermal behaviour of the two-phase medium while neglecting dispersion effects [7]. In this paper, on the contrary, complete mixing is assumed in the gapwise direction, while negligible mixing is assumed in the mid-surface directions [8]. The temperature and reaction degree are two-dimensional, allowing for a gap averaging of eqs (4) and (5). The resulting equations are :

$$\begin{aligned} & \left( \bar{\alpha}_r \rho_r c_{pr} + \bar{\alpha}_f \rho_f c_{pf} \right) \frac{\partial T}{\partial t} + \bar{\alpha}_r \rho_r c_{pr} \bar{v} \cdot \nabla T \\ & = -\frac{q_{wall}}{2h} + \bar{\alpha}_r \rho_r \Delta H f(C, T) + \frac{1}{\eta} \nabla p \cdot \bar{K} \cdot \nabla p \quad , \end{aligned} \quad (6)$$

$$\frac{\partial C}{\partial t} + \bar{v} \cdot \nabla C = f(C, T) \quad , \quad (7)$$

where  $q_{wall}$  stands for the outgoing flux on the walls, and  $\bar{\alpha}_r, \bar{\alpha}_f, \bar{v}$  and  $\bar{K}$  stand for the gap-averaged values of  $\alpha_r, \alpha_f, v$  and  $K$ .

From equation (6), it is clear that the presence of the fibre mat reduces the average heat advection velocity to a lower value  $\bar{v}_{heat}$  than the material velocity  $\bar{v}$  :

$$\bar{v}_{heat} = \frac{\bar{\alpha}_r \rho_r c_{pr}}{\bar{\alpha}_r \rho_r c_{pr} + \bar{\alpha}_f \rho_f c_{pf}} \bar{v} \quad (8)$$

This effect is illustrated in Fig. 2, where the filling of a long rectangular plate is considered in a situation where the influence of the heated walls is not taken into account ( $q_{wall} = 0$ ). The injection temperature is 333 K and the initial mat temperature is 303 K. Due to the hyperbolic nature of eq. (6), a boundary condition is needed at the flow front. This condition is obtained by observing that, during filling, the resin is continuously mixed with the fibre preform, which remains at initial room temperature before being reached by the front. It is therefore easily shown that the front temperature decreases quasi-instantaneously to the initial mat temperature [1]. As can be seen in Fig. 2, the step between injection and front temperatures is convected at the velocity  $\bar{v}_{heat}$  which is lower than the front velocity. In addition, at those relatively low temperatures, no reaction occurs in a reasonable time scale.

The next example illustrates the effect of the heated walls on the general heat transfer. The thermal exchanges taking place between the two-phase medium and the walls are modelled according to the variable thermal penetration length approach developed by Dheur [9]. The injection and mat temperatures are the same as in Fig. 2, while the walls are at 423 K. Results are depicted in Fig. 3. The final temperature profile is tremendously influenced by the walls (Fig. 3.a). At the end of the filling, the part is indeed nearly at the wall temperature, except in the two zones located in the vicinity of the gate and the last flow front (where injection and mat temperatures are imposed). At this general filling temperature, the initiator has totally consumed the inhibitor in a large portion of the injected part, allowing for the curing reaction to begin (Fig. 3.b.c.d). In general, the walls are observed to play a complex role, which first tends to heat the medium, and then, when curing starts, to cool it down. Note that an adiabatic simulation with an injection temperature of 423 K would lead to a final temperature of about 490 K, which shows that the final temperature profile in Fig. 3.a is comparable to the one that would be obtained without taking the reaction heat into account. The thermal role of the walls is therefore more important in SRIM than in TIM.

## CONCLUSIONS

We have developed a simplified model for the non-isothermal simulation of SRIM. Mechanical dispersion is neglected in the mid-surface directions, and assumed to be perfect in the gapwise direction. However, in view of the results obtained in Fig. 3 (which demonstrate the thermal influence of the walls), both thermal conduction and mechanical dispersion should be taken into account in the process. A more accurate model should therefore incorporate both phenomena, together with a better thermal boundary condition at the interface between the sliding polymer and the steel wall. Recent experimental results will help to validate the model [10].

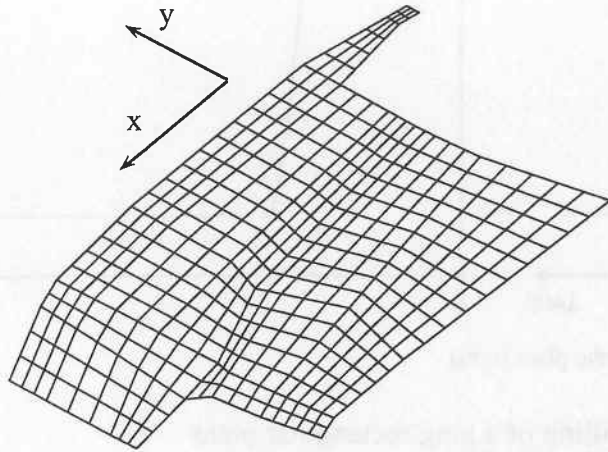
## Acknowledgements

The results presented in this paper have been obtained in collaboration with the Shell Research and Technology Centre in Amsterdam (the Netherlands), within the framework of the Belgian

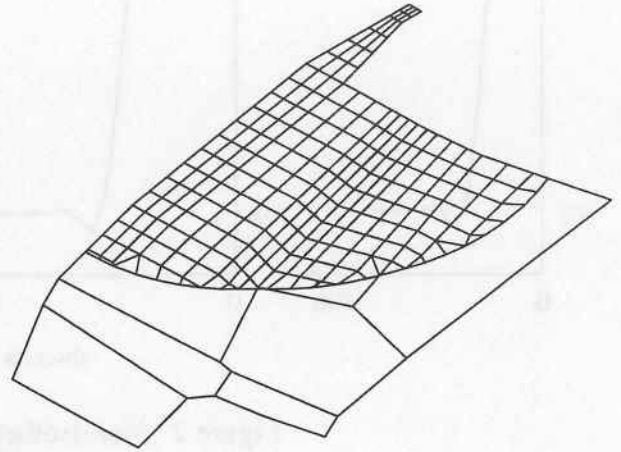
Program on Interuniversity Poles of Attraction, initiated by the Belgian State, Prime Minister's Office for Science, Technology and Culture, and within the frame of a Multimateriaux Project initiated by the Walloon Region of Belgium.

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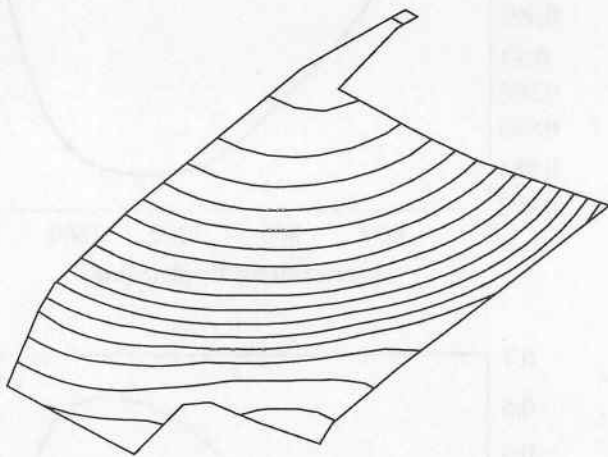
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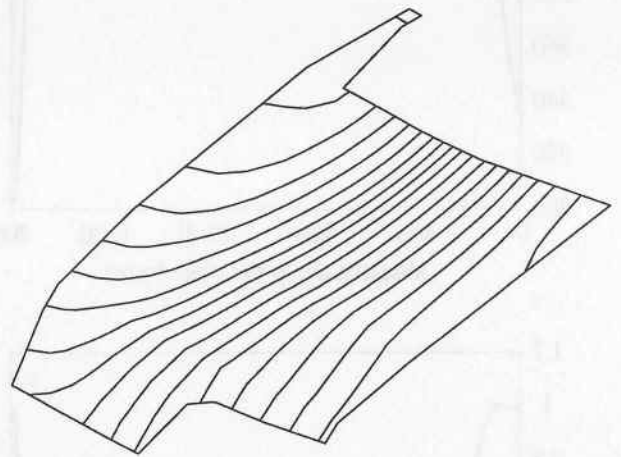
a.



b.



c.



d.

Figure 1 : Isothermal filling of an automotive bonnet.

- a. Finite element mesh;
- b. Example of temporary mesh;
- c. Flow front evolution for  $K_{xx}/K_{yy} = 1$ ;
- d. Flow front evolution for  $K_{xx}/K_{yy} = 5$ .

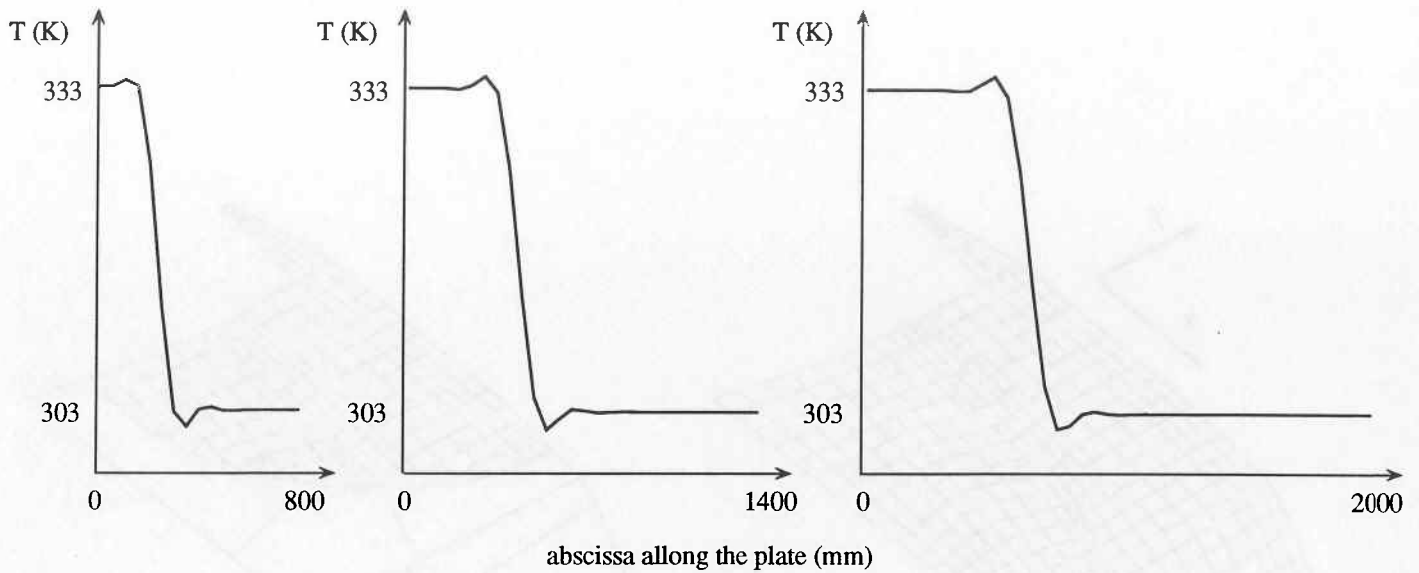


Figure 2 : Non-isothermal filling of a long rectangular plate with adiabatic walls.  
Temperature profile at several stages of the filling.

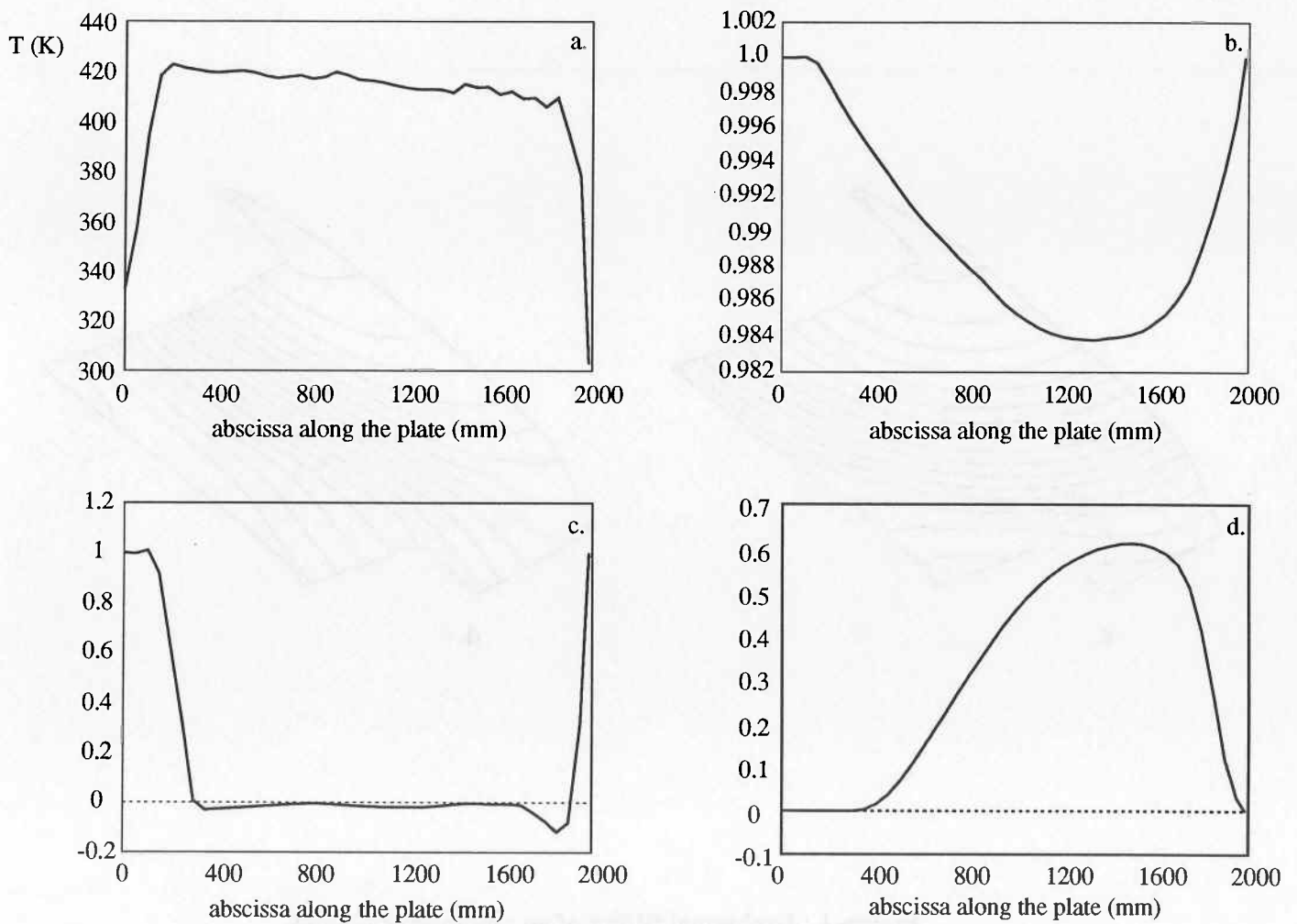


Figure 3 : Non-isothermal filling of a long rectangular plate, with the influence of the heated walls taken into account.  
Profile of different quantities at the end of the filling :

- a. Temperature;
- b. Initiator cc. ratio;
- c. Inhibitor cc. ratio;
- d. Reaction degree.