

# MODELING RESIN FLOW OF REACTIVE RESINS IN LIQUID COMPOSITE MOLDING

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**ABSTRACT:** Standard approach to LCM infusion modeling assumes that the injected resin does not significantly change its viscosity during the filling process. This is not necessarily true as the resin viscosity is a function of temperature and cure which may both change during the process. A comprehensive model of the reactive process, which simultaneously solves for the temperature and the conversion field is formulated and efficient multivariate numerical treatment is suggested to simulate the reactive flow. However, for a wide family of resins, the cure may progress during the filling without significant heat release, and there is more efficient approach to model such an infusion as the process remains virtually isothermal. The paper describes how, in such cases, the flow model of constant viscosity infusion is scaled with time or pressure, to incorporate the viscosity change with time but without spatial changes or temperature variations.

**KEYWORDS:** Liquid Composite Molding, Flow Modeling, Numerical Simulation

## INTRODUCTION

In Liquid Composite Molding (LCM), fibrous preforms are placed in a mold and injected with a thermoset liquid resin. Once the preform is fully saturated and the resin cures, the part is demolded. It is necessary to fully saturate the preform without any dry spots or voids for acceptable mechanical properties. To ensure successful filling, numerical analysis of the flow process has been widely applied.

The numerical simulation of resin flow in LCM has been around for more than two decades [1-5]. The aim of the flow modeling is to provide the resin flow patterns and to determine the pressure or time necessary to fully infuse the preform. Advanced applications of simulation include process optimization and control [6-8]. Others include physics that is usually neglected but might be significant for particular cases - such as dual scale flow, acceleration (gravity) forces and fabric deformation. Number of implementations have addressed the variable temperature and/or resin reaction [4, 9-10]. As the latter comes at a significant performance cost, it is beneficial to examine the conditions under which the effects of resin reaction and variable temperature become important.

The mathematical model that describes the resin flow velocity - usually volume averaged  $\langle v_f \rangle$  - uses Darcy's law to relate it to the resin pressure  $p$  gradient through preform permeability  $\mathbf{K}$ , and resin viscosity  $\eta$  as shown below:

$$\langle v_f \rangle = -\frac{\mathbf{K}}{\eta} \cdot \nabla p \quad (1)$$

This is well justifiable for the low resin velocity. Governing equation is then obtained using mass conservation in filled domain and solved quasi-statically to obtain resin pressure field at any instant:

$$\nabla \cdot \left( \frac{\mathbf{K}}{\eta} \cdot \nabla p \right) = 0 \quad (2)$$

The flow front of the saturated domain is advanced, either after the pressure is solved (explicitly) or during the solution step (implicitly). Then, the process is repeated. The local temperature and resin reaction enter this equation through a single term, the resin viscosity  $\eta$ . Depending on the resin,  $\eta$  may strongly depend on both the temperature and the degree of cure. Thus, for such cases one needs to evaluate the degree of cure  $\alpha$  and temperature  $T$  through the solution domain. This requires one to describe the energy equation and the reaction equation and in addition provide thermal and reaction material parameters.

The energy equation,

$$(\rho c_p)_{ef} \frac{\partial T}{\partial t} + (\rho c_p)_f \langle v_f \rangle \cdot \nabla T = \nabla \cdot (\mathbf{k} + \mathbf{K}_D) \cdot \nabla T + \eta \langle v_f \rangle \cdot \mathbf{K}^{-1} \cdot \langle v_f \rangle + \phi R \frac{\partial \alpha}{\partial t} \quad (3)$$

balances the change in internal energy and heat convection with heat conduction and dispersion, dissipation and heat generation due to the chemical reaction. One needs to provide porosity  $\phi$ , the effective fluid density and heat capacity  $((\rho c_p)_{ef})$ , the fluid density and heat capacity  $(\rho c_p)_f$ , heat conductivity and dispersion tensors respectively ( $\mathbf{k}$  and  $\mathbf{K}_D$ ), reaction heat  $R$  and the reaction rate  $\partial \alpha / \partial t$ . The reaction equation presents the species preservation in the form:

$$\phi \frac{\partial \alpha}{\partial t} + \langle v_f \rangle \cdot \nabla \alpha = \nabla \cdot \phi (\mathbf{D} + \mathbf{D}_D) \cdot \nabla \alpha + \phi \dot{R}_\alpha \quad (4)$$

Note that, besides of reaction rate  $\dot{R}_\alpha$  one needs to specify the diffusion tensor  $\mathbf{D}$  which also has the dispersion component. In non-isothermal reactive case, equations (3) and (4) should be solved coupled with the equation (2) to provide proper viscosity value for movement of the resin flow front.

## THE IMPACT OF TEMPERATURE AND CURE MODELING

The effect of reformulating from mathematical description in equation (2) to the system of equations (2)-(4) is significant. The impact can be summarized into two points as follows:

1. The single linear governing equation is replaced by three non-linear coupled governing equations. The computational cost will be significant (orders of magnitude higher).
2. A number of material parameters introduced by the governing equations (3) and (4) that need to be characterized. Even if the molecular diffusion is neglected, one must still characterize the material parameters to describe the heat dispersion and the reaction rate in the reactive resin flow model.

To address the dispersion, one needs to first determine if and when the individual terms in Eq. (3) and (4) are significant and then retain only the terms that may have an impact. A simple example of this will be presented in this paper. Once the system is simplified, an effective formulation is needed to solve it. This will require significant changes to the numerical formulation that was designed for efficient solution of Eq. (2). If the dispersion term is retained in equation (3), experimental or analytic approach is necessary to characterize the dispersion coefficient. These topics

are, however, beyond of scope of this paper. In this work we will only address reactive resins which do not influence the temperature of the resin during the filling stage.

## REACTIVE RESINS WITHOUT SIGNIFICANT EXOTHERM OR EXTERNAL HEATING

Equations (3) and (4) provide some guidance as to when the coupled model can be by-passed. In many cases (such as polyurea resins) the resin reacts during the infusion stage and hence its viscosity changes due to the conversion. However, no significant heat ( $R$ ) is generated in the reaction. If the resin is pre-mixed which is the usual case and in the absence of external heating (as is typical in VARTM and VIP processes) the solution of Eqns. (3) and (4) reduce to  $T=const$  and  $\alpha=\alpha(t)$ . One can still easily solve the Eq. (4) for  $\alpha$  if the reaction rate is described - only the leftmost and rightmost terms remain and there is no spatial difference.

The more efficient alternative to such an approach is to characterize viscosity instead of the reaction rate, obtaining an explicit relation  $\eta=\eta(t)$ . Since Eq. (2) is solved for pressure under quasi-steady state assumptions, the solution can be performed just as in the isothermal case. Depending on the boundary conditions applied, the solution may be actually obtained with pre-set viscosity and consequently, the time or pressure can be scaled. This provides a very efficient solution method without resorting to the coupled problem using the same isothermal solver.

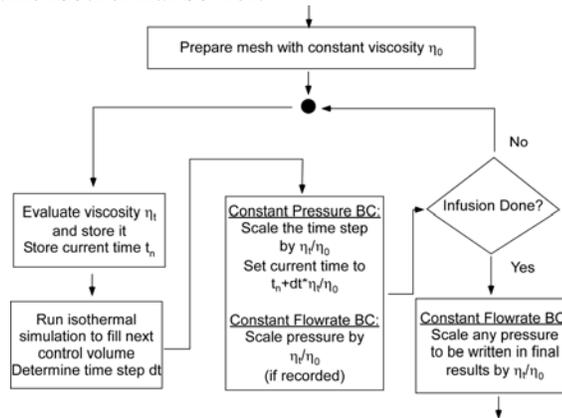


Fig. 1 Flow chart to model the reacting resin infusion using constant viscosity/temperature solver.

The algorithm utilizing LIMS simulation package [11] is as follows (Fig. 1). The system is solved with constant initial viscosity  $\eta_0$ . After each step, proper values are adjusted for the different viscosity  $\eta(t)$  at current time  $t$ , depending on the infusion boundary conditions. For more common constant pressure the time step size is scaled by the ratio of current viscosity over the initial one  $\eta(t)/\eta_0$  and time values in system and currently filled nodes are updated. For constant flow rate, only the pressure values evaluated must be scaled by  $\eta(t)/\eta_0$ .

For such cases, the incremental solver preserves the efficiency of the isothermal, constant viscosity approach. For mixed boundary conditions the condition itself has to be modified, as it includes the viscous losses within injection hardware, and the efficiency would be lost as the equation system would need to be re-assembled with each step. However, it is generally better to model the tubing using one dimensional

elements [11] and this case can be easily avoided. For the same reason the direct modification of viscosity with each step - although feasible - should be avoided, at least within LIMS.

As an example one might consider the VARTM infusion of glass panel (Fig. 2) with a resin system which exhibits increasing viscosity with time. We will present an example that will estimate how important the resin reaction is for predicting the infusion time and flow patterns. The viscosity change will be expressed by a simple relation

$$\eta(t) = \eta_0 e^{\frac{t}{t_0}} \quad (5)$$

in which  $\eta_0$  is the viscosity at time  $t_0$ . The typical values for in-plane permeability and through the thickness permeability were selected to be  $5.10^{-11} \text{ m}^2$  and  $2.85.10^{-12} \text{ m}^2$  respectively based on our permeability measurement experience with various fabrics. The mold length was selected to be 610 mm and thickness is 6 mm. We will study the flow in cross section only. The distribution media on top is 0.9 mm thick with permeability of  $2.10^{-9} \text{ m}^2$  and it is cut short by 51 mm. Initial resin viscosity  $\eta_0$  is set to 0.5 Pa.s and  $t_0$  is varied. Its value is described by non-dimensional factor  $\eta(3600)/\eta(0)$ , which describes the number of times the viscosity increases at the end of an hour as compared to the start of infusion.

Using the initial resin viscosity (Figure 2(a)) predicts the infusion time at roughly 1,800 s. Providing the solution with variable viscosity (Figure 2(b)) changes the estimate to almost 6,000 s. Note that the flow-front patterns do not change significantly, but the infusion rate and flow front progression speed decreases abruptly with the time for the variable viscosity resin model.

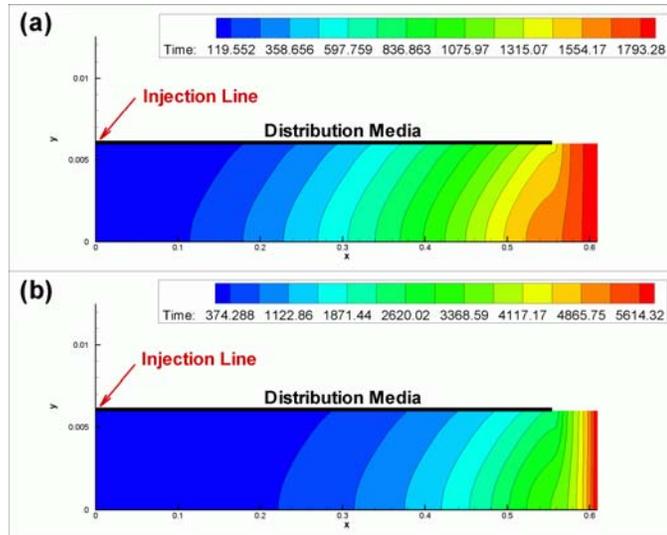


Fig. 2 Predicted flow progression with (a) constant viscosity (b) viscosity changing significantly with time due to the reactive flow ( $\eta(3600)/\eta(0)=6$ )

The impact of resin reactivity on fill time may be easily summarized by plotting the time to infuse the panel with reactive resin relative to the time it takes with the resin of constant viscosity  $\eta_0$ . The resin reactivity will be described by the factor  $\eta(3600)/\eta(0)$  again. This is plotted in Fig. 3.

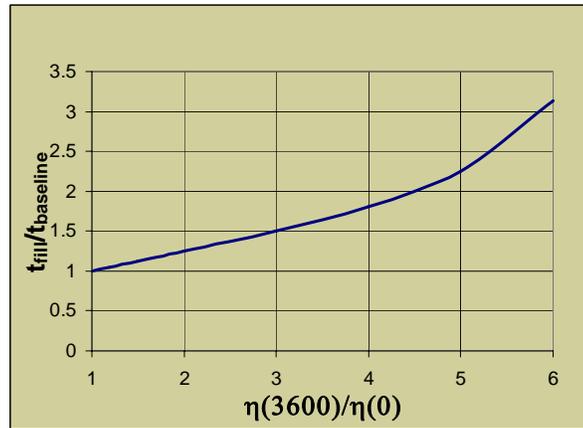


Fig. 3. Infusion time depending on resin reactivity  $\eta(3600)/\eta(0)$ .

It is quite obvious that, for the small changes of viscosity,  $\eta(3600)/\eta(0)$  less than 2, the effect on predicted time remains limited to about 25% and, as the flow patterns do not change, the isothermal solution with constant viscosity may be good enough as an estimate. As the reactivity increases (say  $\eta(3600)/\eta(0)$  about 4 to 5 as in polyureas, it is necessary to include this effect as described herein as the infusion time may double.

## CONCLUSIONS

The general description of the resin infusion flow of reacting resin requires solution of coupled system of equations; the mass conservation equation for pressure, energy equation for temperature and species conservation for degree of conversion. The system is non-linear and tightly coupled. Consequently, the cost of solution is much higher than for the isothermal, non-reacting cases. This will have particularly hamper our ability to use simulations for process optimization and control.

In some cases, the reacting resin does not generate significant energy and the problem description may be reduced to mass conservation with transient viscosity. This is important because this case has practical use and the numerical modeling is as efficient as that with no resin reaction. The solution can be implemented on top of isothermal, non-reactive solver, as described above. The actual impact of reacting resin on infusion time depends on the rate of viscosity change and for some resin systems it may be acceptable to ignore the reaction, but in cases it is not, the described approach provides efficient predictive capability for infusion modeling.

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