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Development of a numerical model for the filling stage of Reaction Injection Molding (RIM)

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ABSTRACT

Reaction Injection Molding processes, commonly called RIM processes, are processes for the rapid production of complex plastic parts using low viscosity monomers or oligomers. One of the main differences with thermoplastic injection molding is that RIM uses polymerization in the mold rather than cooling to obtain a solid polymer. A great advantage is that, due to the low initial viscosity, relatively low temperatures and pressures are needed to fill a mold. Some of the disadvantages of reaction injection molding, however, also arise from its low viscosity, like mold surface penetration and problems to seal a mold. Another significant disadvantage is that gas bubbles may get trapped during filling, due to instability of the flow front. It would be of great interest to numerically and experimentally analyze this phenomenon in order to obtain a better understanding and eventually a mold design tool.

This project starts with a literature study on the current knowledge of RIM-processes. It is also seen that currently available mathematical models have not allowed a good enough simulation of the process. Therefore, the main goal of the project is to develop a three-dimensional numerical model for the filling stage of RIM. Since remeshing for three-dimensional problems is complex and time- and memory-consuming, this project focusses on examining the possibilities of fixed-grid methods for modelling mold filling flows. Inertial effects and surface tension effects play a significant role.

The numerical model developed is based on solving the governing Navier-Stokes equations with a standard Galerkin finite element method, using the penalty function approach. Non-linear terms are linearized by a Newton-Raphson linearization. The Euler-implicit time integration scheme is applied. In order to determine the flow front position, the passive scalar advection equation is used to advect a ‘discontinuous’ scalar function representing the different materials. The viscosity of the computational domain is a function of this scalar field. The whole model is developed as a fixed mesh method. In flows with moving free boundaries, stress singularities at the contact lines appear. This problem is dealt with by relaxation of the no-slip boundary condition.

Test problems show that the model provides qualitative good simulation of three-dimensional mold filling processes. However, using discontinuous functions meets with several difficulties. First of all, it often appears to be a source of numerical instabilities. This is the main reason that, for the time integration, the Euler implicit scheme is used rather than the Crank-Nicholson time integration scheme. The Euler implicit scheme has great damping properties, necessary to avoid these numerical instabilities. Furthermore, flow front instabilities could be damped as well. The second difficulty is that the position of the flow front is not exactly determined. Having no grid line that coincides with the interface, applying surface tension forces is not simple. Following Brackbill et al. (1991) who developed a continuum surface force model for a finite difference approach, here this
model has been studied and tested in a finite element context. Major problems occurred, since second order differentiation requires smoother basis functions than those available in the present SEPRAN-package. Extra diffusion steps to smooth the interface gave no resolution of the problem.

Finally, using a jump in viscosity, the occurrence of turbulent flow structures has to be taken into consideration. Using a low viscosity as for air, high Reynolds’s numbers occur and near e.g. sharp edges turbulent flow structures are generated, leading to divergence of the numerical process.

As a conclusion it can be stated that the three dimensional model developed is certainly useful for gaining qualitative information on mold filling processes. Another potential application of the model developed is for example the simulation of gas-assisted injection molding processes. However, when small scale flow front instabilities have to be analyzed and when surface tension effects have to be modelled, remeshing strategies or mesh adaptation may be essential.

**Keywords:** Reaction Injection Molding, FEM method, fixed grid method
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1 Introduction

Reaction Injection Molding processes, commonly called RIM processes, are processes for the rapid production of complex plastic parts using low viscosity monomers or oligomers. For RIM mainly polyurethanes, formed by the reaction of isocyanates with active hydrogen compounds, are used. These different monomers and oligomers are combined by impingement mixing just as they enter the mold. A solid polymer is then formed by cross-linking or phase separation. One of the main differences with thermoplastic injection molding (TIM) is that it uses polymerization in the mold rather than cooling to obtain a solid polymer. In PUR-RIM monomer temperature and mold temperature are not so different, as the reaction is activated by the impingement mixing. Because the mixture is initially at a low viscosity, low pressures, less than 10 bar, are needed to fill the mold. As low viscosity and low pressures during filling translate into lighter weight and more complex molds, and hereby lower costs, it opens up lots of other possibilities and applications.

Some of the disadvantages of Reaction Injection Molding, however, also arise from its low viscosity. It is difficult to seal molds and the resulting flash can increase post-molding labor costs. Mold release has been a major problem as low viscosity liquids penetrate mold surfaces and urethanes adhere well to metals and an internal or external mold release agent has to be used.

Another significant disadvantage is that gas bubbles can become trapped during filling the mold, due to instability of the flow front. It therefore would be of great interest to accurately analyze the filling stage of RIM both numerically and experimentally, having special attention for the stability of the flow front.

Although fairly accurate predictions can be made for injection molding, currently available mathematical models have not allowed a good enough simulation for the RIM process. In this project, a three-dimensional numerical model is developed to simulate the filling of a mold. Inertial effects are taken into account and some research has been done on including surface tension effects. The possibility of numerical methods based on a fixed grid method are investigated, both for filling processes and imposing surface tension forces.

This report gives an overview of the current knowledge on RIM processes in section 2. The governing equations are derived in section 3, resulting in a simplified numerical model for the filling stage of RIM. In section 4 the currently available numerical methods for viscous flows with special emphasize to moving boundaries are summerized. In section 5 the finite element formulation of the set of governing equations is given. Some calculations and results are given in section 6. A final discussion, including the conclusions, is presented in section 7.
2 Reaction Injection Molding and mold filling

In this section, the Reaction Injection Molding process will be described shortly. Since accurately analyzing the mold filling stage of RIM is the main goal of this research project, special attention is paid to the current knowledge on this subject.

2.1 Reaction Injection Molding

Reaction Injection Molding (RIM) is a method for the rapid production of complex plastic parts from low viscosity monomers and oligomers. Figure 1 gives a schematic diagram of a typical RIM machine. Two or more liquid reactants are stored and blended in supply tanks. These supply tanks also maintain the level in the conditioning tanks. The conditioning tanks control temperature and degree of dispersion of the reactants by low pressure recirculation. This recirculation loop is also used for dispersing dry air or nitrogen into the reactants, to compensate for shrinkage during curing. Next the reactants are metered, under high pressure, 100 - 200 bar, to the mixhead at a sufficient flow rate for good mixing and at the proper ratio to give correct stoichiometry. From the mixing chamber the reactive mixture flows into the mold under a pressure less than 10 bar, filling the mold in only a few seconds. There it polymerizes and solidifies sufficiently to take the stresses of demolding. After postcuring some finishing steps like cleaning and painting take place.

![Figure 1: Schematic of a typical RIM machine (Macosko, 1989)](image)

Polyurethanes

Polyurethanes and polyureas constitute over 95 % of RIM production. In the last ten years nylon-6 has been used as well. Urethanes and ureas are respectively formed by the
reaction of isocyanates \((R - NCO)\) with important active hydrogen compounds like alcohols \((HO - R')\) or amines \((H_2N - R')\), see the equations 1 and 2. The polymers are now formed by stepwise polymerization, also called condensation. Together with polymerization crosslinking, phase separation occurs resulting in the right mechanical properties of the polymer for industrial use. More details on the reaction mechanisms can be found in Macosko's book: RIM fundamentals of Reaction Injection Molding (1989).

\[
R - NCO + HO - R' \rightarrow R - NH - CO - O - R' \tag{1}
\]

\[
R - NCO + H_2N - R' \rightarrow R - NH - CO - NH - R' \tag{2}
\]

In Reaction Injection Molding polymerization occurs in the mold simultaneously with setting the polymer shape. As mentioned before, the solid structure has to result from either crosslinking or phase separation. This causes rheological changes. The viscosity and the modulus and tear strength build up as the monomers react. To understand these changes, a lot of investigation has been done on the polymerization kinetics and the molecular structure of polyurethanes.

As RIM polymerization must reach completion within a few minutes or even less, one of the important parameters is the reaction rate. Thanks to reaction kinetic data and numerous studies on crosslinking and phase separation, a simplified kinetic expression could be suggested, neglecting diffusion (Macosko, 1989):

\[
\frac{d\alpha}{dt} = A e^{\left(\frac{E_a}{RT}\right)}(1 - \alpha^2) \quad \alpha(0) = 1 \tag{3}
\]

with \(\alpha\) the conversion of the isocyanates, \(E_a\) the activation energy, \(T\) the temperature, \(R\) the gas constant and \(A\) a material constant.

As mentioned before, the dynamic viscosity was found to depend on the conversion of the isocyanate groups. Castro et al. (1984) have correlated this viscosity with the temperature dependence of the initial viscosity \(\eta_0\) and the conversion according to

\[
\eta = \eta_0(T) \left(\frac{\alpha_g}{\alpha_g - \alpha}\right)^{c_1 + c_2 \alpha} \quad \text{for} \ \alpha < \alpha_g
\]

\[
\eta \rightarrow \infty \quad \text{for} \ \alpha \geq \alpha_g
\]

where \(\eta_0(T) = A_0 e^{(E_0/RT)}\), \(\alpha_g\) the gel conversion and \(c_1\) and \(c_2\) constants.

Mixing
The success of RIM initially depends on good mixing of the reactants in the mixing chamber, since the polymerization is activated by contact between the reactants. Therefore,
Numerical simulation of Reaction Injection Molding

high jet velocity impingement mixing is often called the heart of RIM. The mixing quality of a RIM machine is significant, however usually judged only by visual appearance. A more quantitative measure for the mixing quality is the Reynolds number defined for the flow of component A (or B) through its inlet nozzle in terms of the density $\rho$, flow rate $Q$, viscosity $\eta$ and diameter $d$.

$$Re_A = \frac{4\rho_A Q_A}{\pi \eta_A d_A}$$ (5)

It is concluded that inertial forces help mixing while viscous forces reduce mixing. Good mixing is achieved for most low molecular weight fluids roughly for $Re$ between 200 and 500. Chella and Ottino (1983) explain the influence of the Reynolds number in terms of a lamellar diffusion and reaction model. The mixing flow generates lamella with a distribution of striation thicknesses. When the striations are thin, the monomers diffuse and form polymer. Large striations will cause local stoichiometric imbalance slowing down the polymerization process. Increasing $Re$ reduces the number of too large striations and, therefore, improves the polymerization process until a critical Reynolds number is reached above which all the monomer can react and no further improvement is seen.

Visualization studies, however, show that the mixing created by impingement mixing itself is insufficient for the fast polymerization in RIM (Macosko, 1989). The fluid mechanics of impingement reduces the scale of segregation from the diameter of the inlet nozzles to a scale of order 100 $\mu$m, decreasing with $Re$ up to the critical value. Further reduction in scale results from fluid mechanical mixing during flow in runner and mold, but particularly from physiochemical effects at the interfaces between the two reactant streams. This is called micromixing. Finally, molecular diffusion takes place, see figure 2.

![Figure 2: The three mechanisms active in impingement mixing (Macosko, 1989)](image)

Filling
For mixing low viscosity is an advantage. For molding low viscosity is both advantageous
as a disadvantage. The reactants will wet and penetrate the mold surfaces and air entrainment can occur. To prevent air entrainment, the mold should be filled slowly. However, for fast reacting systems this can lead to incomplete filling. In the next section it will be investigated how process parameters and the reaction rate should be controlled to provide stable, laminar filling of the mold before the reactants gel. Several numerical simulations will be evaluated. It is concluded that unstable filling is due to inertial effects, surface tension effects, viscous forces and the geometry of the mold.

Curing and demolding
After the mold has been filled, curing takes place. During curing, the modulus and strength must build up quickly for rapid demolding and for obtaining optimal final use properties, potentially aided by a postcure step. While curing, the part shrinks due to polymerization and cooling. This shrinkage is compensated for by foaming, also known as gas nucleation (Macosko, 1989). Dry air or nitrogen is used. Foaming and mold temperature therefore affect the final properties of the product. For example no uniform density results and it decreases from the mold gate to the vents, typically by 10%.

The first step in demolding is mold release. Successful release requires that the adhesive forces between mold and polymer are less than the strength of the polymer. Adhesive can be reduced by using external and internal release agents. Internal release agents are preferable since external release agents need extra application time. To be demolded in one piece, the product must have developed enough strength to survive considerable bending stresses. Finally, the modulus has to be large enough to hold products shape, to prevent warpage during cooling.

2.2 Mold filling
In this section, a detailed overview of research on the analysis of RIM mold filling will be given. In general, the filling of thin molds can be characterized by two flow regions:

- a parabolic velocity profile throughout most of the mold
- a fountain flow region close to the front

Fountain flow occurs in flows with free boundaries and a no-slip condition at the walls. The fluid near the center of a channel moves at a higher velocity than the local average velocity across a channel. When it reaches the front it spreads towards the walls. This complex fountain flow plays an important role in the residence time distributions in the reactive molding process, see figure 3 and figure 4, and hence in the conversion and temperature distributions (Blake and Macosko, 1987).

A two-dimensional description of the fountain flow was introduced first by Castro and Macosko (1982). In their modelling of the main flow inertia is neglected in the momentum equation, which combined with the mass balance gives rise to explicit expressions for
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Figure 3: Filling of a vertical rectangular cavity. (Coyle, Blake and Macosko, 1987)

Figure 4: Calculated tracer position corresponding to the visualisation experiments including gravity effects. (Coyle, Blake and Macosko, 1987)

The pressure drop and the velocity profile. Garcia (1991) states, which may be unjustified, that, as filling is done normally under laminar conditions, neglecting inertia is reasonable. Finally, in the energy and mole balance the transverse convection terms are neglected.

The model and experiments by Castro and Macosko (1982) are described here in somewhat more detail, since often other papers refer to these results and compare them with new results. In their model, the momentum balance is simplified by using the lubrication approximation and assuming quasi-static conditions. The following additional assumptions are made: There is a constant flow rate and constant density, thermal properties, the thermal conductivity $k$ and heat capacity $C_p$, are constant, molecular diffusion can be ignored, as well as the sidewall effects and the hydrodynamic entrance length. The fluid can be considered as Newtonian, since shear has been shown to have no effect on viscosity until the conversion reaches the gel point (Macosko, 1989). Second order polymerization kinetics are assumed with an Arrhenius type of temperature dependence

$$ R = A_0 e^\frac{-E}{R_T} c^2 $$

with $c$ the concentration of the reactive species, $R$ the reaction rate, $R_T$ the gas constant, $E$ the activation energy and $A_0$ a material constant.

A laminar unidirectional flow is assumed, with negligible viscous heating and gravity forces. The analysis is thus restricted to thin rectangular molds. This is known as the
Mold filling

Hele-Shaw approach. This approach is also used in injection molding simulation programs like V1P (L. Caspers, 1995; W. Zoetelief, 1995). Explicit expressions for the velocity field \( \vec{v} = (v, w) \) and the pressure \( P \) are obtained by integrating the momentum and continuity equations. The temperature and conversion fields are described with the energy and species balance.

\[
\frac{\partial P}{\partial x} = \frac{\partial}{\partial z} \left( \eta \frac{\partial \vec{v}}{\partial z} \right)
\]

\[
\frac{\partial P}{\partial z} = 0
\]

\[
\rho C_p \dot{T} = k \frac{\partial^2 T}{\partial z^2} + \eta \frac{\partial \vec{v}^2}{\partial z} + R \Delta H_r
\]

\[
\dot{c} = \frac{R}{c_0}
\]

(7)

with \( \eta \) the viscosity, \( \vec{x} = (x, y, z) \) the position vector and \( H_r \) the heat of reaction.

For the flow front, the momentum balances were solved using a stream function formulation, neglecting inertia and assuming slip at the contact line and vanishing tangential stress at the free surface. The flow front was assumed to be flat. Eventually the velocity field is described with an approximate solution (Bhattacharji and Savic, 1965). Comparison with experimental results shows rather good agreement. It is concluded that little reaction and heat transfer occur during filling and the measured pressure rise basically coincides with the model when a constant viscosity is assumed. Castro and Macosko conclude that ignoring the flow front during filling results in an underestimated pressure rise. Moreover, the extent of reaction near the flow front differs considerably.

Lelakou and Richardson (1986) developed a finite difference model using a more complete fluid dynamical description and also considered non rectangular molds. In the main flow, only the momentum balance in the flow direction is solved, assuming that the transverse velocity is negligible. The transverse velocity obtained from the continuity equation, is included in the convection terms, as the derivatives can be non-negligible. Garcia (1991) mentioned that it was found by Charbonneau that when the transverse convection is not considered in the energy and mole balance, the model underestimates the pressure rise at long flow times. In order to avoid iterative evaluation of the pressure gradients of the front, Lelakou and Richardson (1986) solved the vorticity transport equation, taking the inertia terms into account and allowing the viscosity to vary within the flow front region. At the front, the interface is assumed to be flat and perpendicular to the main axis of the flow. For complex geometries this method is clearly too complicated.

Taking into account the convection along and across the flow is the most important feature of this model. It is observed that the convection interacts to contribute to the creation of the temperature and conversion profiles which control the curing stage and hence the properties of the final product. For this reason it is evident that careful consideration of
the main flow and front flow is justified and preferable. Lelakou and Richardson (1986) have used a moving, changing mesh, doing well for two-dimensional situations.

Sahay and Reible (1992) have developed a two-dimensional model for mold filling in RIM, governed by a coupled system of non-linear, partial differential balance equations. They used a finite element program FIDAP, remeshing the entire liquid domain to follow the advancing front. Surface tension effects, represented as boundary conditions, have been taken into account to determine the location of the free surface. Results show that liquid near the wall stays in the mold for the longest time and, consequently, conversion and temperature are higher in that region. The rheological and kinetic models of Castro and Macosko (1982) were used. The results are shown below in figures 5, 6, 7 and 8.

Anturkar (1994) developed a two-dimensional model for mold filling using a Petrov Galerkin finite element method with free surface parametrization. No a priori assumptions are made with respect to the flow front. Its position is computed simultaneously along with other variables using a kinematic boundary condition at the interface, relating the position of the interface with the local velocity. The elements of the mesh are stretched in axial direction, proportionally moving with the flow front. When elements become too far stretched, the mesh is regenerated. Although this method is more time-consuming then models with simplifying assumptions, it provides more refined predictions. These predictions are better suited in estimating flow front effects as bubble growth.

The fountain flow shown in the above figures is stable. However, it is known that at high velocities, the free surface of the flow front no longer moves smoothly into the mold, as is illustrated in figure 9. Waves appear and fingers shoot out from the front. As mentioned in the previous section instabilities occur due to surface tension effects or due to the presence of viscosity gradients across the mold and the mold geometry. Large air bubbles particularly seem to occur when reactants flow around inserts or from the gate into the mold.

Defining the capillary number

$$Ca = \frac{v \eta}{\gamma}$$

where $\gamma$ is the surface tension, the ratio of viscous to surface tension effects is given. For a typical RIM system (2D) $\gamma = 20 \cdot 10^{-8} N/cm$ and $Ca \approx O(10^4)$. Viscous forces are dominant and more likely to cause instabilities (Macosko, 1989). The ratio of the inertial forces to surface tension forces can now be given by the Weber number

$$We = (Ca)_e (Re) = \frac{\rho v^2 H}{\gamma}$$

with $H$ the thickness of the mold.

Cohnen and Osswald (1992) show results of experimental studies on the influence of inertial forces and surface tension on mold filling of RIM systems. They used a rectangular
Figure 5: Streamlines at various times during filling. (Sahay and Reible, 1992)

Figure 6: Free surface position with time (Sahay and Reible, 1992)

Figure 7: Species concentration contours during filling. (Sahay and Reible, 1992)

Figure 8: Temperature isotherms during filling. (Sahay and Reible, 1992)
Numerical simulation of Reaction Injection Molding

Figure 9: Filling as the average velocity is increased from a) 0.4 to b) 0.5 and c) 0.8 m/s. Fingers begin to shoot out from the flow front. Mold dimensions: 457 by 305 by 3.2 mm. RIM 2200 with initial viscosity 600 mPa.s (Castro et al., 1980)

mold of dimensions: 200 by 400 by 2 mm. Several assumptions are made. There is no dependence on temperature and pressure for the viscosity. There is no reaction during filling. The liquid is incompressible and isotropic. The effects of gravitation and fountain flow are negligible. Water at room temperature is used for the experiments. Due to a long inlet tube the flow is completely developed at the inlet of the cavity. The inner walls had to be very smooth, as the least disturbance at the wall have visible consequences for the development of the flow front.

To judge the influence of the inertial forces figure 10 shows the computed flow front positions with time for a Newtonian fluid filling the cavity at constant temperature. The inertia and capillary effects are neglected. The fronts are completely smooth. Figure 11 shows the experimental results for different inlet pressures. A higher inlet pressure results in a higher inlet velocity and therefore a higher Reynolds number, capillary number and Weber number.
Due to the inertial forces the region farthest from the inlet has not the longest residence time anymore. Mold venting has to take place now at the same side as the inlet of liquid. At higher Reynold's numbers air entrapment takes place, also due to the inertial forces. The Reynold's number is defined as:

$$Re = \frac{\rho v H^2}{\eta L}$$

with $\rho$ the density, $v$ a characteristic velocity, $\eta$ the viscosity, $H$ the thickness (2 mm) and $L$ the length (200 mm).

For $Re > 26.9$ fingers shoot out from the front. This could be an indication for the generation of local turbulences at high velocities. The stabilizing influence of the surface tension decreases more rapid (quadratic) with increasing velocity than the influence of the inertial forces. For $We > 49.8$ the influence of the surface tension is that small that the smallest
disturbances at the surfaces of the mold can lead to finger generation. Entrapment of larger air pockets can be avoided by properly locating the venting holes. More critical, of course is the air entrapment around inserts, due to a combination of both inertia and capillary effects. This has been investigated by Cohnen and Osswald (1992) as well.

They also developed and tested a numerical model (Mohammed, Cohnen and Osswald, 1994) to simulate high Reynolds' number flows in thin cavities present during reaction injection mold filling. The model, using the momentum equation for two dimensions and a penalized continuity approach, was tested by computing the flow in a simple sheeting die, as in their experimental studies. Including the shear stresses acting through thickness, the effect of changing the die thickness can be simulated. The velocity gradients through thickness for the stress terms are evaluated by assuming a parabolic velocity profile in the third direction. The governing equations are solved using a standard Galerkin finite element method. The advection of the flow front is simulated using a control volume approach. It was shown that increasing entrance velocity and die thickness the effect of inertia is increased. As inertia plays a more important role, recirculation zones begin to form. It was concluded from the results that inertia caused the material, entering the mold, to spread out less, when the entrance velocities are increased. Air pockets are formed at higher injection speeds. In this numerical model surface tension effects are not taken into account.

Lo and Reible et al. (1994) developed a three-dimensional numerical model based on a control volume approach, to evaluate and optimize RIM processing especially in thick complex molds. This model is capable of predicting the position of the dynamic flow front in three dimensions. The governing equations include inertial and gravitational effects. The SIMPLE-algorithm is used to determine the position of the flow front (Patankar, 1980). This position is determined after each timestep, using the maximum velocity among the control volumes on the advancing front. The maximum velocity is chosen as a basis velocity to normalize other velocities. The fluid in this specific control volume with the maximum velocity is advanced one grid cell, effectively determining the length of a timestep. The motion of the rest of the fluid is according to the local velocity. The calculated position of the interface is therefore not forced to conform to the gridlines. All other fluid and flow properties, however, are mapped onto the approximated grid. Comparison of the simulation results, using this method, and the limited experimental data currently available show rather good agreement.

From the above discussed experimental and numerical results, it can be concluded that for stable filling an upper bound is set to the mold filling velocity. However, to avoid short shots, thus to prevent gellation before complete filling of the mold, the filling time can not be too large. Gel time can be decreased by decreasing reactivity, i.e. by lowering the average reactant temperature or, when used, by lowering the catalyst level. However, decreasing reactant temperature may lead to mixing problems. Therefore, good mixing is one of the criteria to be considered.
Mold filling

For some reactants also thermal stability is important. This sets an upper limit to the storage temperatures of the reactants. All criteria can be summarized in a moldability diagram to achieve a well mixed, completely filled mold without air entrainment or degradation. An example is shown in figure 12.

Figure 12: *An example of a moldability diagram to achieve good filling.* (Macosko, 1989)

Such a moldability diagram can be readily constructed using experimental data for a new formulation or mold form, from the mold dimensions, the initial reactant viscosities as a function of temperature, the reaction kinetics and the adiabatic gel time or conversion.

Nearly all currently available numerical models are two-dimensional models, using front models and remeshing techniques.

2.3 Discussion

In the previous sections, the problems that occur during Reaction Injection Molding have been mentioned. It is noted that mold filling simulations become important, since the mechanical properties of the final products depend highly on parameters existing during their production. The stability of the flow front plays an important role. A detailed knowledge of the process may result in process optimization. Currently available mathematical models have not allowed a good enough simulation for the RIM process. Nearly all available simulations are two-dimensional simulations. Especially, because it is complex to determine the position of the flow front during the filling process. The discussed models, therefore, make use of front models and regeneration of the computational grid. These methods are inconvenient and unsatisfactory for simulating the filling of complex molds and analyzing flow front instabilities. A three-dimensional model is developed by Lo and Reible (1994) based on a control volume approach.

The current project will focus on developing a three-dimensional finite element model. The goal is to prevent remeshing techniques and front modeling.
Numerical simulation of Reaction Injection Molding

The literature survey resulted in the expectation that inertia and surface tension effects are of importance and have to be taken into account simulating high Reynolds' number flows as in reaction injection molding. Therefore, both inertia and surface tension effects will be considered modelling the filling stage of Reaction Injection Molding.
3 Modelling the RIM process

In this section, the filling stage of the RIM process is modelled. The molten polymer is considered as a Newtonian fluid, with physical properties independent of temperature, pressure, shear rate and conversion. First the fundamental equations of balance will be presented. The constitutive equations will be considered such that simplifications are allowed. Finally, a numerical model is provided.

3.1 Equations of balance

The fundamental equations governing the motion of a fluid, considered as a continuum, are described in this section. A continuum approach implies that the molecular structure of the material is ignored and that physical variables such as velocity and density can be defined at an arbitrary point in the fluid. To derive the governing balance equations, Reynold’s transport theorem for a material volume and the transport theorem for a surface are used.

3.1.1 Reynold’s transport theorem

Let $V(t)$ be a material volume with $A(t)$ the closed boundary and let $\phi(\vec{x}, t)$ be a smooth function in $V(t)$. The rate of change of the volume integral

$$\Phi(t) = \int_{V(t)} \phi(\vec{x}, t)dV$$

can be written as

$$\frac{d\Phi(t)}{dt} = \int_{V(t)} \frac{\partial \phi(\vec{x}, t)}{\partial t}dV + \int_{A(t)} \phi(\vec{x}, t)\vec{v} \cdot \vec{n} dA$$

where $\vec{v}$ is the velocity of the surface as well as the material velocity.

Using Gauss’s divergence theorem and the material derivative

$$\dot{\phi} = \frac{\partial \phi(\vec{x}, t)}{\partial t} + \vec{v} \cdot \nabla \phi$$

this can be written as

$$\frac{d\Phi(t)}{dt} = \int_{V(t)} \left[ \frac{\partial \phi(\vec{x}, t)}{\partial t} + \nabla \cdot (\vec{v} \phi) \right] dV = \int_{V(t)} \left[ \dot{\phi} + \phi \nabla \cdot \vec{v} \right] dV$$

This relation is only valid if $\phi(\vec{x}, t)$ is continuous in $V(t)$.

Let now $V(t)$ be a material volume consisting of two volumes $V_A(t)$ and $V_B(t)$, separated by a singular surface $S(t)$, see figure 13. Let $\Psi$ be an additive quantity associated with
volume $V(t)$, containing a discontinuity across this surface $S(t)$. $\Psi_V$ is the amount of $\Psi$ in $V(t)$ and may be written as:

$$
\Psi_V = \Phi(t) + \Phi_S(t) = \int_{V_A(t)+V_B(t)} \phi(\vec{x}, t) dV + \int_{S(t)} \phi_S(\vec{x}, t) dA
$$

(15)

with $\phi(\vec{x}, t)$ and $\phi_S(\vec{x}, t)$ the volume and surface densities of $\Psi$ respectively. The quantity $\phi_S(\vec{x}, t)$ and others defined on $S(t)$ are functions of the surface coordinates $U$. Figure 13: $V(t) = V_A(t) \cup V_B(t)$ is a material volume with $S(t)$ a discontinuity surface and $\vec{n}$ is the outward normal on $V_A(t)$ and $V_B(t)$ and $\vec{n}_{AB}$ on the surface $S(t)$ from $V_A$ to $V_B$ and $\tau$ are the tangential unit vectors on $S(t)$.

The rate of change of $\Phi(t)$ becomes

$$
\frac{d \Phi(t)}{dt} = \int_{V_A(t)} \frac{\partial \phi(\vec{x}, t)}{\partial t} dV + \int_{V_B(t)} \frac{\partial \phi(\vec{x}, t)}{\partial t} dV + \int_{A_A(t)} \phi(\vec{x}, t) \vec{v} \cdot \vec{n} dA + \\
+ \int_{A_B(t)} \phi(\vec{x}, t) \vec{v} \cdot \vec{n} dA - \int_{S(t)} \left[ \phi(\vec{x}, t) \vec{v} \cdot \vec{n} dA + \right] \vec{n}_{AB} \cdot \vec{v} dA
$$

(16)

where $\vec{v}$ is the velocity of the singular surface and

$$
[\phi(\vec{x}, t)]_S = \phi_B(\vec{x}, t) - \phi_A(\vec{x}, t)
$$

(17)

Using Gauss’s divergence theorem for both volumes $V_A$ and $V_B$ separately, and the material derivative the Reynold’s transport theorem is written as:

$$
\frac{d \Phi(t)}{dt} = \int_{V_A(t)+V_B(t)} \left[ \frac{\partial \phi(\vec{x}, t)}{\partial t} + \nabla \cdot (\vec{v} \phi) \right] dV - \int_{S(t)} \left[ \phi(\vec{x}, t)(\vec{v} \cdot \vec{n}) - \phi_S(\vec{x}, t) \vec{n} \cdot \vec{v} dA \right]
$$
Equations of balance

\[ \int_{V(t)} \nabla \cdot \mathbf{u} \, dV - \int_{\partial V(t)} \mathbf{u} \cdot \mathbf{n} \, ds = \int_{\Omega(t)} \mathbf{f} \, dV + \int_{\partial \Omega(t)} \mathbf{g} \cdot \mathbf{n} \, ds \]

where

\[ \mathbf{f} = \begin{cases} \mathbf{F} & \text{in } \Omega(t) \subseteq \mathbb{R}^3 \\ \mathbf{g} & \text{on } \partial \Omega(t) \end{cases} \]

and \( \mathbf{n} \) is the outward unit normal to \( \partial \Omega(t) \).

3.1.2 Transport theorem for a surface

The rate of change of the mass conservation equation can be written in a similar way as the rate of change of the momentum equation in the previous section (Müller, 1985).

\[ \frac{d}{dt} \int_{\Omega(t)} \rho \mathbf{u} \, dV = \int_{\partial \Omega(t)} \rho \mathbf{u} \cdot \mathbf{n} \, ds - \int_{\Omega(t)} \rho \mathbf{u} \cdot \nabla \mathbf{u} \, dV + \int_{\partial \Omega(t)} \rho \mathbf{g} \cdot \mathbf{n} \, ds - \int_{\Omega(t)} \rho \mathbf{f} \cdot \mathbf{u} \, dV 

\]

\[ \dot{\phi} = \frac{d}{dt} \phi(t) + \nabla \phi \cdot \mathbf{u}(t) \]

\[ \dot{\phi} = \frac{d}{dt} \phi(t) + \nabla \phi \cdot \mathbf{u}(t) \]

\[ v_{\Delta} = v_{\Delta} \mathbf{u} + v_{\Delta \perp} \mathbf{b} \]

The covariant derivative, which is a projection of the usual derivative on the surface, is defined as

\[ \nabla_{\Delta} \mathbf{v} = \nabla \mathbf{v} - \Gamma_{\Delta \gamma}^\nu \mathbf{v}^\gamma \]

where \( \Gamma_{\Delta \gamma}^\nu \) are the Christoffel symbols defined by:

\[ \Gamma_{\Delta \gamma}^\nu = \frac{\partial U^\Gamma}{\partial x^\Delta} \frac{\partial^2 x^\delta}{\partial U^\Delta \partial U^\gamma} \]

\[ \Gamma_{\Delta \gamma}^\nu = \frac{1}{2} g_{\Delta \gamma} \left( \frac{\partial g_{\delta \Delta}}{\partial U^\gamma} + \frac{\partial g_{\delta \gamma}}{\partial U^\Delta} + \frac{\partial g_{\Delta \gamma}}{\partial U^\delta} \right) \]

The mean surface curvature \( K \) is defined by

\[ K = \frac{1}{2} g^{\Gamma \Delta} b_{\Gamma \Delta} \]

with \( b_{\Gamma \Delta} \) the components of the curvature tensor \( b \).
3.1.3 General equation of balance

Let $\Psi$ again be an additive quantity associated with volume $V(t)$, as assumed in the previous sections, and $\Psi_V$ the amount of $\Psi$ in $V(t)$ such that:

$$\Psi_V = \Phi(t) + \Phi_S(t) = \int_{V_A(t)+V_B(t)} \phi(\vec{x}, t)dV + \int_{S(t)} \phi_S(\vec{x}, t)dA$$

The rate of change of $\Psi_V$ may have three different causes. First, $\Psi_V$ may change because surfaces $A_A, A_B$ and $\delta S$ are penetrated by a flux $F_V$. Second, a production $P_V$ of $\Psi$ within $V(t)$ will change $\Psi_V$. Finally $\Psi_V$ may change by supply $S_V$ from the outside. The general form of the equation of balance reads:

$$\frac{d}{dt} \left( \int_{V_A(t)+V_B(t)} \phi(\vec{x}, t)dV + \int_{S(t)} \phi_S(\vec{x}, t)dA \right) = F_V + P_V + S_V =$$

$$- \int_{A_A(t)+A_B(t)} \psi \Phi_V \bar{n}dA - \int_{A_A(t)+A_B(t)} \psi \Phi_S \bar{v}dl + \int_{V_A(t)+V_B(t)} (\psi p_V + \psi s_v)dV + \int_{S(t)} (\psi p_S + \psi s_S)dA \quad (28)$$

where $\psi \Phi_V$ and $\psi \Phi_S$ are the flux densities per element of area and per line element respectively, $\psi p_V$ and $\psi p_S$ are the production densities in the volume and on the singular surface respectively and $\psi s_v$ and $\psi s_S$ are the densities of supply.

The use of the transport theorems for volume and surface and the divergence theorem on the surface provide a more explicit expression for the equation of balance:

$$\int_{V_A(t)+V_B(t)} \left[ \frac{d}{dt} \right] \left[ \phi + \phi \nabla \cdot \vec{v} \right] dV + \int_{A_A(t)+A_B(t)} \psi \Phi_V \bar{n}dA - \int_{V_A(t)+V_B(t)} (\psi p_V + \psi s_v)dV =$$

$$- \int_{S(t)} (\phi_S + \phi_S(\vec{v}_S \cdot \vec{n}_{AB}) - 2K \vec{v}_S \cdot \vec{n}_{AB}) + \psi \Phi_S \Delta -$$

$$\left[ \phi(\vec{x}, t)(\vec{v}_S - \vec{v}) \right] \bar{n}_{AB}dA + \int_{S(t)} (\psi p_S + \psi s_S)dA \quad (29)$$

From this equation, the balance equations for mass, momentum, energy etcetera can be derived.

3.1.4 Mass balance

Conservation of mass means that the mass $m$ contained in a material volume $V(t)$ does not change. That is:

$$\frac{dm}{dt} = \frac{d}{dt} \int_{V(t)} \rho(\vec{x}, t)dV + \int_{S(t)} \rho_S(\vec{x}, t)dA$$

where $\rho$ and $\rho_S$ are the volume and surface mass densities respectively.
For the conservation of mass, the production densities as well as the fluxes and supply
densities are zero. For a material volume with a discontinuity surface it now follows:

$$\int_{V_{A(t)+V_{B(t)}}} [\dot{\rho} + \rho \nabla \cdot \vec{v}] dV =$$
$$- \int_{S(t)} \left( \rho_S \dot{\vec{v}}_S + 2K \vec{v}_S \cdot \vec{n}_{AB} \right) - \left[ \rho(\vec{x}, t)(\vec{v}_S - \vec{v}) \right]_{S} \cdot \vec{n}_{AB} dA \quad (31)$$

Since the material volume is arbitrary, this results in a local continuity equation and its
jump relation

$$\dot{\rho} + \rho \nabla \cdot \vec{v} = 0 \quad \text{in } V_{A(t)} \text{ and } V_{B(t)} \quad (32)$$
$$\dot{\rho}_S + \rho_S \left( \vec{v}_S \cdot \vec{n}_{AB} \right) + \left[ \rho(\vec{x}, t)(\vec{v}_S - \vec{v}) \right]_{S} \cdot \vec{n}_{AB} = 0 \quad \text{on } S(t) \quad (33)$$

### 3.1.5 Balance of momentum

The rate of change of momentum for a material volume is equal to the sum of external
forces acting on that volume $V(t)$. The external forces include both bodyforces $\vec{F}_b$, acting
as supply densities on the interior of $V(t)$, and traction forces $\vec{F}_t$, acting as flux densities
on the surface of the volume, given by

$$\vec{F}_b = \int_{V(t)} \rho \vec{f} dV \quad (34)$$
$$\vec{F}_t = \int_{A(t)} \vec{t} dA = \int_{A(t)} \sigma \vec{n} dA \quad (35)$$

with $\vec{f}$ the total body force per unit mass, $\vec{t}$ the stress vector acting on an element $dA$ of
the boundary surface $A(t)$ and $\sigma$ is the stress tensor.

Using the general equation of balance and application of the transport theorems, Gauss’s
divergence theorem and the mass balance equation leads to,

$$\int_{V_{A(t)+V_{B(t)}}} \left[ \rho \dot{\vec{v}} - \rho \vec{f} - \nabla \cdot \sigma \right] dV +$$
$$\int_{S(t)} \rho_S \dot{\vec{v}}_S - \sigma^A_\Delta - \rho_S \vec{f}_S - \left[ \rho(\vec{v} - \vec{v}_S)(\vec{v}_S - \vec{v}) + \sigma \vec{n}_{AB} \right]_{S} \cdot \vec{n}_{AB} dA = 0 \quad (36)$$

Since the material volume is arbitrary, this results in a local momentum equation and its
jump relation

$$\rho \dot{\vec{v}} - \rho \vec{f} - \nabla \cdot \sigma^c = 0 \quad \text{in } V_{A(t)} \text{ and } V_{B(t)} \quad (37)$$
$$\rho_S \dot{\vec{v}}_S - \sigma^A_\Delta - \rho_S \vec{f}_S - \left[ \rho(\vec{v} - \vec{v}_S)(\vec{v}_S - \vec{v}) + \sigma \vec{n}_{AB} \right]_{S} \cdot \vec{n}_{AB} = 0 \quad \text{on } S(t) \quad (38)$$
In a non-polar body, i.e. in the absence of spin, the stress tensor $\sigma$ is symmetric.

$$\sigma = \sigma^c$$

This relation follows from the balance of angular momentum.

### 3.2 Constitutive equations

In this section, the governing constitutive equations are given. The polymer will be considered as an isothermal viscous fluid. In the general Navier-Stokes theory a linear relation of stress and deformation is postulated. In that case the Cauchy stress tensor can be split into two contributions. First, the thermodynamical pressure $p_0 I$, as in a fluid at rest only normal components of stress on a surface appear and the stress does not depend on the orientation of the surface. A moving fluid develops additional components of stress due to viscosity. This is accounted for in the extra stress tensor. The Cauchy stress tensor becomes

$$\sigma = -p_0 I + \sigma^e = -p_0 I - p_1 I + \sigma^d = -p I + \sigma^d$$

where $\sigma^d$ is the deviatoric part of the Cauchy stress tensor and $\sigma^e$ is the extra stress tensor.

Considering generalized Newtonian fluids the extra stress tensor can be written as

$$\sigma^e = \mu \text{tr}(D) I + 2\eta D^d$$

with

$$D = \frac{1}{2}(\nabla \dot{v} + (\nabla \dot{v})^T)$$

$\mu$ and $\eta$ represent the bulk and shear viscosity.

### 3.3 Conditions at the discontinuity surface

The normal velocity component has to be continuous across a boundary for purely kinematical reasons. The velocity (both the normal and tangential component) is continuous across the discontinuity surface, whenever the two media are in equilibrium. However, a fluid in relative motion cannot be in exact thermodynamical and mechanical equilibrium, and, it is then necessary to wonder whether the absence of equilibrium may be accompanied by a discontinuity in velocity. According to Batchelor (1967) we may expect that under common conditions of moving fluids, velocity is continuous across a material boundary between a fluid and another medium. Since molecular migration and interaction are likely to be as effective in equalizing the local velocities of two different media at an interface as in equalizing the velocities at two neighbouring points in a fluid, an approximate equilibrium will be established everywhere.
The same considerations hold for surface tension effects. Equilibrium conditions are supposed, also for fluids in motion. It is assumed that the discontinuity surface does not have properties of its own except for the surface tension. Body forces are ignored. Consider a membrane at rest. The momentum balance over the interface then reads

$$-\sigma_{S,\Delta}^{A} - [\sigma, \vec{n}_{AB}]_{S} \cdot \vec{n}_{AB} = 0$$

(44)

The surface stress $\sigma_{S}^{A}$ ($\Delta, \Gamma = 1, 2$) can be split into normal and tangential stress components. As there is no spin on the surface, the normal surface stress component is zero. When $S^{\Gamma \Delta}$ is the tangential component of the surface stress, the jump relation becomes

$$[\sigma, \vec{n}_{AB}]_{S} \cdot \vec{n}_{AB} = -S^{\Gamma \Delta} \tau_{T} - S^{\Gamma \Delta} b_{\Delta \Gamma}$$

(45)

The second term on the right-hand side is the surface tension times curvature $\gamma \kappa$. The first term on the right-hand side presents local changes in surface tension at the surface $\Delta \gamma$.

The stress balance over the interface can be written as (Subbiah, 1995)

$$\left( -p_{1} + 2\eta_{1} \frac{\partial u_{n}}{\partial n} \right) - \left( -p_{2} + 2\eta_{2} \frac{\partial u_{n}}{\partial n} \right) = \gamma \kappa$$

(46)

for the normal component, and

$$\eta_{2} \left( \frac{\partial u_{n}}{\partial s} + \frac{\partial u_{s}}{\partial n} \right) - \eta_{1} \left( \frac{\partial u_{n}}{\partial s} + \frac{\partial u_{s}}{\partial n} \right) = \frac{\partial \gamma}{\partial s}$$

(47)

for the tangential components, with $\eta_{i}$ and $\rho_{i}$ (i = 1, 2) the viscosities and densities of the two different fluids.

3.4 Recapitulation

In the volumes $V_{A}(t)$ and $V_{B}(t)$ the following balance and constitutive equations are valid:

$$\dot{\rho} + \rho \nabla \cdot \vec{v} = 0$$

(48)

$$\rho \ddot{\vec{v}} - \rho \ddot{\vec{f}} - \nabla \cdot \sigma = 0$$

(49)

$$\sigma = -p_{0} I + \sigma^{e}; \quad \sigma^{e} = \mu \text{tr}(\mathbf{D}) I + 2\eta \mathbf{D}^{d}$$

(50)

The jump relations are:

$$\dot{\rho}_{S} + \rho_{S} \left( \vec{v}_{S,\Delta} - 2K \vec{v}_{S} \cdot \vec{n}_{AB} \right) + \left[ \rho(\vec{z}, t)(\vec{v}_{S} - \vec{v}) \right]_{S} \cdot \vec{n}_{AB} = 0 \quad \text{on } S(t)$$

(51)
Numerical simulation of Reaction Injection Molding

\[
\rho_S \ddot{v}_S - \sigma_S^A - \rho_S \ddot{v}_S - [\rho(\ddot{v} - \ddot{v}_S)(\ddot{v}_S - \ddot{v}) + \sigma_{AB}]_S \ddot{u}_AB = 0 \quad \text{on } S(t) \tag{52}
\]

On the discontinuity surface the following conditions determine the flow

\[
[v]_S = 0 \tag{53}
\]

\[
\left( -p_1 + 2\eta_1 \frac{\partial u_n}{\partial n} \right) \left( -p_2 + 2\eta_2 \frac{\partial u_n}{\partial n} \right) = \gamma \kappa \tag{54}
\]

\[
\eta_2 \left( \frac{\partial u_n}{\partial s} + \frac{\partial u_s}{\partial n} \right) - \eta_1 \left( \frac{\partial u_n}{\partial s} + \frac{\partial u_s}{\partial n} \right) = \frac{\partial \gamma}{\partial s} \tag{55}
\]

With sufficient initial and boundary conditions the solution of the problem can, in principle, be determined. The conditions at the discontinuity surface have already been discussed. Two types of boundaries are left: the inlet and the walls of the mold. At the inlet and at the walls the velocity need to be given.

3.5 Magnitude estimation of significant terms in governing equations

In several situations it is not necessary to solve the complete set of Navier-Stokes equations, since the motion of the fluid can be described with sufficient accuracy by a simpler set of equations. By introducing dimensionless groups an order of magnitude estimation of the various terms in the governing equations can be given. The magnitude estimation ascertain whether terms in the Navier-Stokes equations may be neglected or not.

In this section the governing equations for the flow in the filling stage as it exists in Reaction Injection Molding are simplified combining some geometrical conditions with flow assumptions. Here, e.g. a cylindrical geometry is considered, with \( r_0 \) the radius of the cylinder and \( L \) a characteristic length in the \( z \)-direction, that is the flow direction. The cavity is assumed to get filled by a polymeric liquid, polyurethane, while pushing air into the open.

The following characteristic values are defined:

\[
\begin{align*}
p_0^* &= \frac{p_0}{\eta_0 V} \\
\kappa^* &= \frac{\kappa}{\kappa_0} \\
\eta^* &= \frac{\eta}{\eta_0} \\
\rho^* &= \frac{\rho}{\rho_0} \\
t^* &= \frac{t}{t_0} \\
t_0 &= \frac{r_0}{V}
\end{align*}
\]
Equations of balance

\[
\tilde{f}^* = \frac{\tilde{f}}{f} \quad \nabla^* = \nabla r_0
\]

The dimensionless numbers and coefficients are now defined as follows:

\[
\kappa = \frac{1}{\rho} \left( \frac{\partial P}{\partial \rho_0} \right)_{T,X} \quad \text{isotherm compressibility factor}
\]

\[
Re = \frac{\rho_0 V r_0}{\eta_0} \quad \text{Reynold's number}
\]

\[
Ga = \frac{1}{\alpha_0 (T_W - T_0)} \quad \text{Gay-Lussac number}
\]

\[
Ca = \rho_0 V^2 \kappa_0 \quad \text{Cauchy number}
\]

\[
Fr = \frac{V^2}{f r_0} \quad \text{Froude number}
\]

\[
Ma = \frac{V}{c} \quad \text{Mach number, with c the speed of sound waves}
\]

The governing equations can accordingly be written as:

The continuity equation:

\[
\frac{V}{r_0} \nabla^* \cdot \tilde{v}^* = -\frac{Ca}{Re} \kappa^* \tilde{p}_0^*
\]

(56)

As will be shown later, incompressibility will be dealt with and therefore, the stress tensor \( \sigma \) reduces to (Batchelor, 1967):

\[
\sigma = -\rho_0 I + 2\eta \mathbf{D}^d
\]

(57)

The momentum equation becomes in dimensionless form:

\[
Re \rho^* \tilde{\dot{\mathcal{V}}}^* - \frac{Re}{Fr} \rho^* \tilde{\mathcal{V}}^* + \nabla^* \tilde{p}_0^* - 2
\]

\[
\nabla^* \eta^* \mathbf{D}^d = 0
\]

(58)

3.5.1 Volume \( V_A(t) \), the polymeric liquid

Using the characteristic values in table 2 at the end of this section, the balance equations can be simplified for the polymeric liquid, volume \( V_A(t) \), to:

\[
\nabla \cdot \tilde{\mathcal{V}} = 0
\]

(59)

\[
\rho \tilde{\mathcal{V}} - \rho \tilde{\mathcal{V}} + \nabla \tilde{p}_0 - 2 \nabla \eta \mathbf{D}^d = 0
\]

(60)
3.5.2 Volume $V_B(t)$, air

For volume $V_B(t)$, air, the balance equations become, using table 3 at the end of this section and the assumption of incompressibility:

\[ \nabla \cdot \mathbf{\tilde{v}} = 0 \quad (61) \]

\[ \rho \mathbf{\tilde{v}} - \rho \mathbf{\ddot{f}} + \nabla p_0 = 0 \quad (62) \]

The assumption of incompressility is correct as

\[ \left| \frac{1}{\rho} \frac{\partial \rho}{\partial t} \right| \ll \frac{r_0}{V} \quad (63) \]

or, by introducing the dimensionless Mach number $Ma^2 \ll 1$. This declares that air flows with relatively low velocities can be assumed as effectively incompressible. In case of filling a mold with polymer, while pushing the air out of that mold, and assuming a stress free outflow, the assumption of incompressibility holds.

3.5.3 Discontinuity surface

The jump relations are simplified to, as discussed in section 3.3:

\[ \left[ \mathbf{\tilde{v}} \right]_S = 0 \quad (64) \]

\[ \left( -p_1 + 2\eta_1 \frac{\partial u_n}{\partial n} \right) - \left( -p_2 + 2\eta_2 \frac{\partial u_n}{\partial n} \right) = \gamma \kappa \quad (65) \]

\[ \eta_2 \left( \frac{\partial u_n}{\partial s} + \frac{\partial u_s}{\partial n} \right) - \eta_1 \left( \frac{\partial u_n}{\partial s} + \frac{\partial u_s}{\partial n} \right) = \frac{\partial \gamma}{\partial s} \quad (66) \]

3.6 Rheology and kinetics

In order to simulate the reactive RIM flow, constitutive relations are needed to describe the chemical kinetics and the dynamic viscosity. Here, the constitutive relations used by Castro and Macosko (1982) will be used. Second order kinetics with Arrhenius temperature dependence is assumed

\[ R = A_0 e^{\left( \frac{-E_a}{RT} \right)} (1 - \alpha)^2 \quad (67) \]

with $R$ the reaction rate, $X$ the conversion, $T$ the temperature, $R_g$ the gas constant, $E_a$ the activation energy and $A_0$ a material constant. The following equation was found to give a good description of the dynamic viscosity for RIM systems

\[ \eta = \eta_0(T) \left( \frac{\alpha}{\alpha_g - \alpha} \right)^{\alpha_1 + \alpha_2 X} \quad (68) \]
where $\eta_0(T) = A_\eta \exp(E_\eta / R_g T)$ and $\alpha_\eta$ the gel conversion.

The kinetic and rheological parameters are given in table 4 and 5 at the end of this section.

As most of the conversion takes place after the mold has been filled, a Newtonian viscosity with no temperature and conversion dependence is assumed for the modelling in this project.

Table 2. Characteristic values, polyurethane (Macosko, 1982)

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$T_0$</td>
<td>$333 , ^\circ\text{C}$</td>
</tr>
<tr>
<td>$T_W$</td>
<td>$338 , ^\circ\text{C}$</td>
</tr>
<tr>
<td>$\rho_0'$</td>
<td>$10^1 \frac{N}{m^2}$</td>
</tr>
<tr>
<td>$c$</td>
<td>$0.0066$</td>
</tr>
<tr>
<td>$\lambda_0$</td>
<td>$0.17 \frac{J}{mKs}$</td>
</tr>
<tr>
<td>$\kappa_0$</td>
<td>$1$</td>
</tr>
<tr>
<td>$\rho_0$</td>
<td>$1000 \frac{g}{mol}$</td>
</tr>
<tr>
<td>$C_0$</td>
<td>$2410 \frac{mol}{m^3}$</td>
</tr>
<tr>
<td>$t_r$</td>
<td>$10^2 , [s]$</td>
</tr>
<tr>
<td>$\tau_0$</td>
<td>$0.0032 , [m]$</td>
</tr>
<tr>
<td>$D$</td>
<td>$10^{-11} \frac{m^2}{s}$</td>
</tr>
<tr>
<td>$\gamma$</td>
<td>$2.10^{-2} \frac{N}{m}$</td>
</tr>
</tbody>
</table>

Table 3. Characteristic values, air

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$T_0$</td>
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</tr>
<tr>
<td>$T_W$</td>
<td>$338 , ^\circ\text{C}$</td>
</tr>
<tr>
<td>$\rho_0'$</td>
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<tr>
<td>$\epsilon$</td>
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<tr>
<td>$\lambda_0$</td>
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</tr>
<tr>
<td>$\kappa_0$</td>
<td>$10^{-5} \frac{m^2}{N}$</td>
</tr>
<tr>
<td>$\rho_0$</td>
<td>$1.2 \frac{g}{m^3}$</td>
</tr>
<tr>
<td>$\tau_0$</td>
<td>$0.0032 , [m]$</td>
</tr>
<tr>
<td>$C_{P0}$</td>
<td>$1000 \frac{J}{kgK}$</td>
</tr>
<tr>
<td>$L$</td>
<td>$0.457 , [m]$</td>
</tr>
<tr>
<td>$\gamma^c$</td>
<td>$1.401$</td>
</tr>
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Table 4. Thermal and kinetic parameters

<table>
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<th>Value</th>
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</thead>
<tbody>
<tr>
<td>$E_\alpha$</td>
<td>$53.2 \frac{kJ}{mol}$</td>
</tr>
<tr>
<td>$A_0$</td>
<td>$10560 \frac{m^3}{mol^2}$</td>
</tr>
<tr>
<td>$R_g$</td>
<td>$8.31 \frac{J}{molK}$</td>
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Table 5. *Rheological parameters*

<table>
<thead>
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<tr>
<td>$E_\eta$</td>
<td>41.3 $kJ/mol$</td>
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<tr>
<td>$A_\eta$</td>
<td>10.3$10^{-8}$ [Pas]</td>
</tr>
<tr>
<td>$\alpha_g$</td>
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<td>$c_1$</td>
<td>1.5</td>
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<tr>
<td>$c_2$</td>
<td>1.0</td>
</tr>
</tbody>
</table>
4 Numerical methods for free surface flows

In this section, a short review is given on the currently available numerical methods for viscous flows with moving boundaries. First, a short introduction is given, followed by a summary and description of the various methods. Finally, a method is discussed to model surface tension.

4.1 Introduction

A free surface problem is a problem where the domain of interest has an unknown boundary, which has to be determined as part of the solution procedure. Two approaches to the modelling of the interface can be distinguished (Floryan and Rasmussen, 1989). First, interface capturing methods exists, that attempt to resolve the detailed structure of the interface. No special modelling, other than finer gridding, is necessary in the vicinity of the interface. The interface is captured in the sense that the actual physical discontinuity is someplace near the middle of the gradient. The second approach is the interface tracking method, where the interface is treated as a discontinuity, requiring a special model to account for the interface effects. The location of the interface has to be known a priori in order to be able to apply the interface model. In this section, only the second approach will be discussed. Floryan and Rasmussen (1989) together with Wang and Lee (1989) give a complete review of all methods.

The interface is assumed to be isothermal and to have only the equilibrium property of a uniform surface tension. No contamination is present, since for nonisothermal interfaces the mechanical properties are not well understood and additional material constants could be involved. At the interface the following equations hold:

\[ \vec{v}_A \cdot \vec{n} = \vec{v}_B \cdot \vec{n} = \vec{v} \cdot \vec{n} \]  \hspace{1cm} (69)

\[ \vec{v}_A \cdot \vec{t} = \vec{v}_B \cdot \vec{t} \]  \hspace{1cm} (70)

\[ \vec{n}.(\sigma_A - \sigma_B) = \kappa \gamma \vec{n} \]  \hspace{1cm} (71)

with \( \vec{v}_A \) and \( \vec{v}_B \) the velocities of fluids \( A \) and \( B \) at the interface, \( \vec{n} \) is a unit vector normal to the interface pointing from fluid \( A \) to \( B \), \( \sigma_A \) and \( \sigma_B \) are the stress tensors, \( \gamma \) is the surface tension and \( \kappa = -\nabla_s \cdot \vec{n} \) is the mean surface curvature, where subscript \( s \) denotes the surface operator. When the interface is non-isothermal or when a surface contamination is present additional terms should be added to account for variation in surface tension. Another problem that complicates the modelling process is that the physical conditions occurring along contact lines, where the interface comes in contact with a solid surface, are not completely understood. Choices that can be made for modelling purposes will be discussed in section 4.3.

4.2 Interface tracking methods

The currently available algorithms for interface tracking methods are generally split into three categories:
Numerical simulation of Reaction Injection Molding

- Eulerian methods
- Lagrangian methods
- Mixed, i.e. Eulerian-Lagrangian methods

All these methods work well under a specified set of conditions and do not have totally general purpose. The Eulerian methods are most widely used and most accurate. In the next sections, the first two categories will be discussed. The mixed Eulerian-Lagrangian methods will not be discussed separately but will be proposed in the context of either Eulerian or Langrangian simulations, since they nearly always can be characterized as being mainly Eulerian or Langrangian.

4.2.1 Eulerian methods

Eulerian methods are characterized by a coordinate system that is either stationary in the laboratory reference frame or is moving in a certain prescribed manner in order to accommodate the continually changing shape of the solution domain. The fluid travels between different computational cells even when the grid moves. Grid movements are not related to the motion of the flow. The main advantage is that the fluid can undergo large distortions without loss of accuracy. Four different types can be distinguished:

- fixed grid methods
- adaptive grid methods
- mapping methods
- special methods
  that are used for simplified flow situations, like the Hele-Shaw flow.

Fixed grid methods

In the fixed grid method, the moving interface most generally does not coincide with a grid line. Special procedures have to be used to handle these situations. This leads to the main disadvantage of the fixed grid method. Calculating the position of the interface accurately, is very difficult. The main advantages are that the interface may undergo large deformations without loss of accuracy and that it is relatively straightforward to handle multiple interfaces. This method suffices for large scale flows where the flow is not dominated by the surface effects. In that case, the treatment of the surface need not be very accurate.

There are two ways of tracking the interface using fixed grids:

- Surface tracking

The interface is represented as a series of interpolated curves through a discrete set of points on the interface. At each timestep the information about the location of the points and the sequence in which they are connected is saved. The points are then moved
Numerical methods for free surface flows

According to an interface evolution equation. In the simplest methods for surface tracking for two dimensions, the points (interface) are determined as a sequence of heights above a given reference line or as a series of points with a parameter, see figure 14. The advantage of surface tracking methods is that features smaller than the cell spacing of the grid can be resolved. The high storage requirements are a main disadvantage. It is also difficult to handle merging and folding interfaces. Because of accuracy consideration, it is necessary to limit the maximum distance between neighboring points. For increasing interfaces, this means that points have to be added along the interface, or conversely deleted whenever required. The best way to do this still has to be found.

![Figure 14: Surface tracking of an interface by: (a) a sequence of heights above a reference line; (b) a series of points and a parameter (Floryan and Rasmussen, 1989)](image)

Surface tracking methods are generally used in one- or two-dimensional calculations for non-interacting interfaces.

- **Volume tracking**

Volume tracking methods are based on reconstructing the interface whenever necessary. This method does not require high storage requirements. The reconstruction is done cell by cell and is based on marker quantities within the cell. The interface is given by those cells that contain both fluids. The accuracy with which the reconstructed interfaces approximate the real interface is difficult to judge.

The simplest algorithm is the Marker-And-Cell method (MAC). This finite difference method marks different fluids with massless marker particles. The interface is defined as somewhere in the cells that contain both marker particles. No details are known about the exact location, orientation and curvature of the interface. After the velocity field of the fluid flow is calculated, marker particles are then moved with a weighted average of the four nearest cell velocities. One of the disadvantages of this method is the fact that the resolution is limited by the grid size, viscous stress is not included, a double grid system is required and it is difficult to impose boundary conditions on the interface. This is the basic MAC method and it has been extended in several directions. This method has been applied for TIM and RIM filling to analyze the fountain flow phenomenon.
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The Flow Analysis Network (FAN) approach is an extension of the MAC concept using the lubrication theory instead of the Navier-Stokes equations. FAN uses the flux calculations at the boundary of each control volume to determine the fraction of fill in order to trace free surfaces. This allows the presence of partially filled cells, improving the accuracy of the calculation of the advancing front, without having to refine the grid.

The Volume Of Fluid (VOF) method is essentially based on the same concept. It defines a function $F(x, y, t)$ that is equal to unity at any point occupied by fluid and zero elsewhere. The discontinuity in $F$ propagates according to

$$\frac{\partial F}{\partial t} + \overline{u} \cdot \nabla F = 0$$  \hspace{1cm} (72)

The interface is a material line and is approximated by a straight line through the cell. The location and slope of the line are determined by the average value of $F$ in the cell (the fractional volume) and by the gradient of $F$. Since standard finite difference approximations to the equation above would lead to smearing of $F$, a flux approximation is used called the donor-acceptor method (Floryan and Rasmussen, 1989, refer to Ramshaw and Trapp, 1976). An iterative pressure adjustment is included so that the continuity equation is satisfied. This approach is a finite difference method, but can also be extended for finite element use.

In figure 15 the reconstruction of an interface based on both the MAC and VOF technique is given.

![Figure 15: Reconstruction of Interface using Volume Tracking Procedures](image)

Volume tracking methods cannot resolve details of the interface that are smaller than the mesh size. The location, orientation and curvature of the interface can not be determined accurately, which affects the accuracy with which e.g. surface tension effects can be accounted for.

Assume, for simplification reasons, the basic mathematical model of a single incompressible viscous fluid:

$$\nabla \cdot \overline{u} = 0$$  \hspace{1cm} (73)
Numerical methods for free surface flows

\[
\frac{\partial \vec{u}}{\partial t} + \vec{u} \cdot \nabla \vec{u} + \frac{1}{\rho} \nabla p - \nu \nabla^2 \vec{u} = 0
\]  

(74)

Where \( \rho \) is the density, \( \nu \) the kinematic viscosity \( \vec{u} = (u, v) \) the velocity and \( p \) the pressure. Let \( y = f(x, t) \) be the free surface. The boundary conditions at the interface can then be expressed in the form of a complex second order differential equation.

The MAC-method can e.g. be used to determine the position of the free boundary. Determining the surface cells, which are cells partially filled or adjacent to an empty cell, a simplification of the boundary conditions is used in these cells. In the simplest form

\[
p = p_a
\]  

(75)

is used. No analytical justification is given. This is the basic MAC method and it has been extended in several directions (Floryan and Rasmussen, 1989).

Adaptive grid methods

In the adaptive or moving grid method the grid is altered so that the moving interface always coincides with one of the grid lines. Therefore the interface is a well defined, continuous curve and information on its location, orientation and curvature is readily available. Steady and unsteady flows need to be treated separately (Floryan and Rasmussen, 1989). The main disadvantage is the difficulty of adjusting the grid to follow highly deformed interfaces.

- **Steady flows**

The first algorithms used for solving steady free boundary problems were based on a Picard type iteration. First, the shape of a moving boundary is assumed and an appropriate grid is constructed. The Navier-Stokes equations are solved using two of the three boundary conditions on the moving interface. The third boundary condition is used to adjust the moving boundary. Finally the grid is adjusted. In case of no convergence, the last three steps are repeated. The choice of the boundary conditions that are used to adjust the interface, strongly depends on the relative strength of the surface tension.

More recent studies use a Newton type iteration to calculate simultaneously the location of the moving boundary and the fields. Both finite difference and finite element methods are developed. Silliman (1979) proposed a method for simultaneous solution for the location of the interface and the field variables. The finite element grid is parametrized and these parameters are considered to be unknown and selected in such a way that one of the grid lines coincides with the interface, see figure 16. A Galerkin method is used to formulate the field equations and the kinematic condition provides the equations necessary to close the system of equations for the grid parameters. This method is referred to as a method of spines.
Unsteady flows

In unsteady flows, the problem arises how to treat the time dependence. This can be done by choosing finite elements in both time and space or finite differences in both time or space. Also a combination can be chosen, finite elements in space and finite difference in time. For example, the method of spines can be extended (Kheshgi and Scriven, 1982; 1984) to unsteady flows, using information available at the previous timestep to estimate all the dependent variables for the new timestep. Newton iteration is employed for simultaneous determination of their actual values.

An additional problem left for unsteady flows is the determination of the moving contact line, since there arise stress singularities, see section 4.3. To avoid this singularity, the no-slip condition at the wall near the contact line can be replaced with some type of slip condition. In the case of static contact lines and small slip, the flow field is modelled well by the corresponding flow field with no-slip at all. Near a dynamic contact line, the situation is quite different as the singularity is much stronger. So far, the models assume that the fluid at the contact line and the contact line move with same velocity. Discretization of the problem leads to numerical slip.

Mapping methods

In the mapping method, the unknown irregularly shaped mesh is transformed onto a fixed regularly shaped computational domain. The mapping function appears as one of the unknown functions and has to be determined together with the fields. This method allows sharp resolution of the interface. However, its applicability is limited to a class of geometries that do not lead to singular mapping.

Special methods

Very often it is not necessary to solve the complete set of Navier-Stokes equations, since the motion of the fluid can be described with sufficient accuracy by a simpler set of
Numerical methods for free surface flows

equations. For example, a moving grid scheme has been used to model the compression molding process as a generalized Hele-Shaw flow. This scheme can be considered as a special free boundary method as it is used for a simplified set of equations. The geometry of the advancing flow front is predicted using a mesh expansion scheme. The expansion of the mesh is done by moving each nodal point to a certain distance in an explicit manner. A variable timestep procedure is used to assure that the flow front advancement does not pass the mold boundaries. As the mesh is expanding during each time step some distorted elements with high aspect ratios need to be corrected. When the total volume of the charge remains constant and the initial shape covers a major portion of the mold, the same mesh can be used throughout the whole simulation without adding elements at the front. However, during the injection molding process the volume of fluid increases. Elements have to be added at the front resulting in a changing size of the stiffness matrix, the element connectivity and the bandwidth.

4.2.2 Langrangian methods

Langrangian methods use coordinate systems that move with the fluid. Each computational cell always contains the same fluid elements. This method results in difficult to handle governing equations. The main advantages of the Langrangian methods are that material interfaces are allowed to be specifically delineated and precisely followed and the interface boundary conditions are easily applied. Another advantage is that a refined mesh can be used in regions that require this refinement. Numerical smearing is absent. The main disadvantages are mesh tangling and numerical inaccuracy. Furthermore Langrangian methods are limited to small deformations (Subbiah, 1995).

When the mesh becomes severely distorted it is necessary to produce a new mesh and transfer information from the old to the new mesh. This is called rezoning or remapping. The interpolation between the meshes may be quite arbitrary and the meshes may be quite arbitrary as well, as far as the number of cells, their geometry and topology are concerned. The rezoning may occur at every time-step (continuous) or after many time-steps (general). Continuous rezoning is preferable as it takes the advantage of the simplifications offered by relative small mesh displacement. A possible algorithm is Arbitrary-Langrangian-Eulerian (ALE). This algorithm has the full interfacial boundary conditions incorporated on the interfaces. Fauchon et al. (1991) have presented a computation method based on the ALE approach coupled with an automatic partially remeshing technique triggered only when the mesh becomes distorted. They state that their method can simulate the flow features of mold fillings inside any polygonal domain, even in the presence of multiple filling fronts.

For highly complex flows, rezoning is not sufficiently capable to avoid mesh scrambling and to provide an accurate representation of the flow. Therefore, the free Langrangian technique is introduced. Mesh points are not tied together for the duration of a calculation and can be deleted, added or reconnected. Reconnecting results in changing the
Numerical simulation of Reaction Injection Molding

mesh topology such that the mesh points acquire new neighbors. The main disadvantage of this technique is that diffusion occurs, having its effects on the accuracy of localizing the interface position.

A different group of methods, particle methods, is based on the concepts adopted from molecular dynamics. The interparticle forces simulate the true pressure and other body forces (Floryan and Rasmussen, 1989).

4.3 Contact lines

The hydrodynamics in the vicinity of the contact line and the factors affecting the contact angle are still open questions. The continuum approach, along with the no-slip boundary condition, results in a discontinuous or unbounded stress tensor at the contact point, resulting in a singularity. Singular stresses, although non-physical, can be tolerated as long as they are integrable. For a static contact line, the singularity is integrable. For a moving contact line, the singularity is non-integrable when the contact angle is less than 180° (Mavridis, 1988). One usually has a choice for modelling purposes, combined with the no-slip hypothesis, of: 1) a fixed contact line condition, for static contact lines, 2) a fixed contact angle condition, for moving or dynamic contact lines as done by Mavridis (1988) and 3) a mixed condition involving the previous cases. An other attempt to avoid stress singularities at the contact points is a relaxation of the no-slip boundary condition. Kamal et al. (1988) use a slip boundary condition to alleviate these singularities in the flow structure. They divide the boundary at the top of the mold wall into two regions: a no-slip region and an apparent slip region where the fountain effect plays a role (Kamal et al., 1988). Experimental investigations as well as analytical solutions (Bhatacharji and Savic, 1965) have shown that the fountain effect is limited to a distance that is approximately equal to one thickness of the cavity behind the melt-wall contact line. In the slip region, a dynamic condition for the tangential component of velocity, that allows slip at the wall, may be used:

$$\frac{\partial u}{\partial y} = -L\beta(x')u$$

(76)

where \(u\) is the tangential component of velocity, \(L\) the height of the cavity and \(\beta\) a parameter of slip, a suitable exponential function. \(\beta\) equal to zero, corresponds to unimpeded tangential slip, whereas the limit \(\beta \to \infty\) corresponds to imposing the no-slip boundary. \(x'\) is the positive distance from the contact point to a point on the mold wall behind the flow front. Simulations have shown that the slip boundary condition was necessary not only to alleviate the singularity at the contact point, but also to maintain an equilibrium shape of the melt front interface (Kamal et al., 1988). The velocity at the interface reaches a constant value.
Numerical methods for free surface flows

4.4 Continuum surface force model, CSF

As previous methods have suffered from difficulties in modelling topologically complex interfaces having surface tension, the continuum surface force model will be discussed. This method is applicable at fixed grids and probably relatively easily to implement for three-dimensional problems.

Brackbill et al. (1991) have presented this numerical finite difference method for modelling surface tension that alleviates the interface topology constraints. The model is called the Continuum Surface Force model (CSF) and interprets surface tension as a continuous, three-dimensional effect across an interface rather than as a boundary value condition on the interface. This model has been applied successfully to model incompressible fluid flow in low-gravity environments, capillarity and droplet dynamics.

The surface stress boundary condition at an interface between two fluids $A$ and $B$ is:

$$ (p_A - p_B + \gamma \kappa) \hat{n}_i = \left( \sigma_{Aik} - \sigma_{Bik} \right) \hat{n}_k + \frac{\partial \gamma}{\partial x_i} $$

with $\gamma$ the surface tension coefficient, $p$ the pressure, $\sigma_{ik}$ the viscous stress tensor and $\kappa = \frac{1}{R_A} + \frac{1}{R_B}$ the local surface curvature, where $R_A$ and $R_B$ are the principal radii of the surface curvature, and $\hat{n}_i$ is the unit normal at the interface into fluid 2.

Assume that the fluids on both sides of the interface are incompressible. The normal and tangential projections to the interface of the above equation lead to scalar boundary conditions at the interface

$$ (p_A - p_B + \gamma \kappa) \hat{n}_i = 2\mu A \hat{n}_k \left( \frac{\partial u_k}{\partial n} \right)_A - 2\mu B \hat{n}_k \left( \frac{\partial u_k}{\partial n} \right)_B $$

$$ \mu B \left( \hat{t}_i \frac{\partial u_i}{\partial n} + \hat{n}_k \frac{\partial u_k}{\partial s} \right)_B - \mu A \left( \hat{t}_i \frac{\partial u_i}{\partial n} + \hat{n}_k \frac{\partial u_k}{\partial s} \right)_A = \frac{\partial \gamma}{\partial s} $$

with $\mu$ the viscosity, $\hat{n}$ the unit normal, $\hat{t}$ the tangent and $\frac{\partial}{\partial s} = \hat{t} \cdot \nabla$ and $\frac{\partial}{\partial n} = \hat{n} \cdot \nabla$. The normal stress boundary condition can be satisfied when the fluid is in rest. The tangential stress boundary condition requires that the fluid is in motion. Brackbill et al. (1991) consider inviscid incompressible fluids with a constant surface tension coefficient. This is presumably done as for finite difference schemes stress boundary conditions are complicated. The interface boundary condition now simplifies to

$$ p_s \equiv p_B - p_A = \gamma \kappa $$

In its standard form surface tension contributes a surface pressure $p_s$ that is the normal force per unit interfacial area $A$ at points $\vec{x}_s$ on $A$, $p_s(\vec{x}_s) = |\mathbf{F}_{sa}(\vec{x}_s)|$. $\mathbf{F}_{sa} = \mathbf{F}_{sa}^{(n)} + \mathbf{F}_{sa}^{(t)}$. Here it is assumed that the total surface force equals the normal component. Then

$$ \mathbf{F}_{sa}(\vec{x}_s) = \gamma \kappa (\vec{x}_s) \hat{n}(\vec{x}_s) $$
Consider the two fluids to be distinguished by a characteristic function \( c(\vec{x}) \)

\[
c(\vec{x}) = \begin{cases} 
  c_A & \text{in fluid A} \\
  c_B & \text{in fluid B} \\n  \langle c \rangle = \frac{c_A + c_B}{2} & \text{at the interface}
\end{cases}
\]  

(82)

that changes discontinuously at the interface. Now replace this discontinuous characteristic function by a smooth variation of fluid color \( \tilde{c}(\vec{x}) \) from \( c_A \) to \( c_B \) over a distance of \( O(h) \), with \( h \) a length comparable to the resolution afforded by the computational mesh. The discontinuous change of fluids at the interface is therefore replaced by a continuous transition. Surface tension should now act everywhere within the transition region in stead of an induced pressure jump. Figure 17 illustrate the properties of the smoothed variation produced by interpolation.

![Figure 17: Contours of the color function \( \tilde{c} \) separate fluids with color values \( c_A \) and \( c_B \). The transition region (unshaded) has width \( h \). Normals given by \( \hat{n} = \nabla \tilde{c} / |\nabla \tilde{c}| \), are calculated at vertices of computational cells lying in the interface region. Surface tension forces, \( \mathbf{F}_{sv} \) are calculated at cell centers from the divergence of \( \hat{n} \). (Brackbill et al., 1991) Consider a volume force, \( \mathbf{F}_{sv}(\vec{x}) \), that gives the correct surface tension force per unit interfacial area, \( \mathbf{F}_{sv}(\vec{x}_s) \) for \( h \to 0 \)

\[
\lim_{h \to 0} \int_{\Delta V} \mathbf{F}_{sv}(\vec{x}) d^3x = \int_{\Delta A} \mathbf{F}_{sv}(\vec{x}_s) dA
\]

(83)

and

\[
\mathbf{F}_{sv}(\vec{x}) = 0 \text{ for } |\hat{n}(\vec{x}_s). (\vec{x} - \vec{x}_s)| \geq h
\]

(84)

The area integral is over portion \( \Delta A \) of the interface lying within the small volume \( \Delta V \) that is constructed so that its edges are normal to the surface and \( h \ll \kappa(\vec{x}_s) \). The Lagrangian fluid momentum equation for an inviscid fluid in the presence of surface tension becomes

\[
\rho \frac{d\vec{u}}{dt} = -\nabla p + \mathbf{F}_{sv}
\]

(85)
To formulate the volume force the color function \( \tilde{c}(\vec{x}) \) has to be defined by convolving the characteristic function \( c(\vec{x}) \) with interpolation function \( I \)

\[
\tilde{c}(\vec{x}) = \frac{1}{h^3} \int_V c(\vec{x'}) I(\vec{x'} - \vec{x}) d^3\vec{x'}
\]  

(86)

For the interpolation function \( I \) a normalization is required and bounded support

\[
\int_V I(\vec{x}) d^3x = h^3 \quad \text{and} \quad I(\vec{x}) = 0 \quad \text{for} \quad |\vec{x}| \geq \frac{h}{2}
\]  

(87)

One example of an interpolation function with according properties is the B-spline, which is described by De Boor (1978). Furthermore it is required that \( I \) is differentiable and decreases monotonically with increasing \( |\vec{x}| \).

The interpolation function is defined such that

\[
\lim_{h \to 0} \tilde{c}(\vec{x}) = c(\vec{x})
\]  

(88)

\[
\lim_{h \to 0} \nabla \tilde{c}(\vec{x}) = \nabla c(\vec{x})
\]  

(89)

This, eventually, leads to (Brackbill et al., 1991)

\[
\textbf{F}_{sv}(\vec{x}) = \gamma \kappa(\vec{x}) \frac{\nabla \tilde{c}(\vec{x})}{[c]}
\]  

(90)

where \([c]\) is the jump in color, \([c] = c_B - c_A\) and the curvature can be written as

\[
\kappa = -(\nabla \cdot \hat{n}) \quad \text{with} \quad \hat{n}(x) = \frac{\nabla \tilde{c}(\vec{x})}{|\nabla \tilde{c}(\vec{x})|}
\]  

(91)

The CSF model has been validated by Brackbill et al. on both static and dynamic interfaces having surface tension, using MAC and ALE methods. They conclude that it alleviates previous topology constraints on modelling interfaces having surface tension without sacrificing accuracy. They suggest that extension of the model to include, spatially varying surface tension, dynamic contact angle treatment of wall adhesion and the implementation of the model in three dimensions would result in possibilities to simulating many new and physically interesting problems.

One of their test problems is shown here. In the absence of viscous, gravitational, or other external forces, surface tension causes a static liquid drop to become spherical. Brackbill et al. have chosen the density \( \rho \) as the initial color function. Figure 18 shows the color function choosing A the drop density and B a smoothed drop density. It is seen that the drop curvature is smoother and more accurate when the smoothed drop density is used. It can be concluded that smoothing is essential.
4.5 Discussion

In this section, the available numerical methods for viscous flows with moving boundaries have been described. It is seen that Eulerian methods are most widely used and most accurate, sustaining large distortions. Using fixed mesh methods is most convenient regarding three-dimensional modelling. However, for these methods accounting for surface tension effects is more severe. The Continuum Surface Force (CSF) model will probably help to describe the influence of surface tension on the flow. Several options to alleviates stress singularities at contact lines are discussed. For three-dimensional modelling relaxation of the no-slip boundary condition is probably most efficient.
5 Finite element formulation of the set of equations governing a mold filling flow

As described in the section 3, the mold filling flow is governed by the set of non-linear differential equations in both volumes containing air and reactive species respectively, coupled by the surface tension at the flow front. Therefore, the, in first instance isothermal non-reacting mold filling flow is governed by the Navier-Stokes equation and the continuity equation as described below, neglecting surface tension.

Assume a volume $\Omega \subset \mathbb{R}^n$ ($n = 2$ of $3$) with smooth boundary $\Gamma = \Gamma_0 + \Gamma_1$ and spatial points given by $x \in \overline{\Omega} = \Omega \cup \Gamma$. The velocity field in $\Omega$ is given by $\vec{u}(x,t)$, where $t \in [0,T]$ and $[0,T] \subset \mathbb{R}_+$. The governing equations can now be written as:

$$\frac{\rho}{\partial t} \frac{\partial \vec{u}}{\partial t} + \rho \vec{u} \cdot \nabla \vec{u} - \nabla \cdot \eta \nabla \vec{u} + \nabla p = \vec{f}$$

$$\nabla \cdot \vec{u} = 0$$

Or, using a penalty function approach:

$$p = -\frac{1}{\epsilon} \nabla \cdot \vec{u}$$

where $\eta$ is the dynamic viscosity, $\rho$ is the density, $p$ represents the pressure field and $\vec{f}$ represents the external forces. The corresponding boundary conditions may be for example:

$$\vec{u} = \vec{0} \text{ on } \Gamma_0$$

$$\sigma \cdot \vec{n} \cdot \vec{u} = 0 \text{ on } \Gamma_1$$

with $\sigma$ the Cauchy stress tensor:

$$\sigma_{ij} = -p\delta_{ij} + \eta \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right)$$

with $\delta_{ij}$ the Kronecker delta.

The position of the moving front is determined by solving an advection equation, where $c(x,t)$ is a scalar. This method is known as passive scalar convection. The scalar field is also used to determine the viscosity.

$$\frac{\partial c}{\partial t} + \vec{u} \cdot \nabla c = 0$$

$$\eta = \eta(c)$$

In the next section, the finite element formulations of the equations above will be given.
5.1 The Navier-Stokes equation

Provided that all functions are smooth, the Navier-Stokes equation and continuity equation can be written in a weak form, imposing lower order differentiability requirements on the solution than in the original strong form.

The weak formulation is:

\[
\int_{\Omega} \left\{ \rho \frac{\partial \tilde{u}}{\partial t} + \rho \tilde{u} \cdot \nabla \tilde{u} - (\nabla \cdot \eta \nabla \tilde{u}) + \nabla p \right\} \cdot \tilde{w} d\Omega = \int_{\Omega} f \cdot \tilde{w} d\Omega 
\]

\[
\int_{\Omega} \left\{ p + \frac{1}{\epsilon} \nabla \cdot \tilde{u} \right\} q d\Omega = 0
\]

with \(\tilde{w}\) and \(q\) test functions. Using partial integration, including the Gauss divergence theorem, these equations can be written as:

\[
\int_{\Omega} \rho \frac{\partial \tilde{u}}{\partial t} \cdot \tilde{w} d\Omega + \int_{\Omega} \rho \tilde{u} \cdot \nabla \tilde{u} \cdot \tilde{w} d\Omega + \int_{\Omega} \eta \nabla \tilde{u} \cdot \nabla \tilde{w} d\Omega
\]

\[
- \int_{\Gamma} \eta \nabla \tilde{u} \cdot \tilde{n} \cdot \tilde{w} d\Gamma - \int_{\Omega} p \tilde{w} \cdot \tilde{n} d\Gamma - \int_{\Omega} f \tilde{w} d\Omega = 0
\]

\[
\int_{\Omega} \left\{ p + \frac{1}{\epsilon} \nabla \cdot \tilde{u} \right\} q d\Omega = 0
\]

For the discretization of the equations, the standard Galerkin method is applied.

\[
\tilde{u}_h = \sum_{i=1}^{3} \sum_{n=1}^{N} u_{in}(t) \phi_{in}(\bar{x})
\]

\[
p_h = \sum_{k=1}^{K} p_k(t) \psi_k(\bar{x})
\]

\(\tilde{w}_h\) is chosen in the subspace spanned by the basis functions \(\phi_1, \ldots, \phi_N\) and \(q_h\) in the subspace spanned by \(\psi_1, \ldots, \psi_K\).

\[
\tilde{w}_h = \sum_{i=1}^{3} \phi_{im}(\bar{x}) \tilde{e}_i \quad \text{for} \ m = 1, \ldots, N
\]

\[
q_h = \psi_t(\bar{x}) \quad \text{for} \ t = 1, \ldots, K
\]

The terms in the equations above can now be written as:

\[
\int_{\Omega} \rho \frac{\partial \tilde{u}}{\partial t} \cdot \tilde{w} d\Omega = \sum_{i=1}^{3} \sum_{n=1}^{N} \int_{\Omega} \rho \frac{\partial u_{in}}{\partial t} \phi_{in} \phi_{im} d\Omega \tilde{e}_i
\]
Finite element formulation

Now, $M$, being the mass matrix, is defined as:

$$
M^{ij} = \rho \int_{\Omega} \phi_{i,m} \phi_{j,n} d\Omega \delta_{ij} \quad (109)
$$

$$
\int_{\Omega} \rho \vec{u} \cdot \nabla \vec{u} \cdot \vec{\omega} d\Omega = \sum_{i=1}^{3} \sum_{j=1}^{3} \sum_{n=1}^{N} \sum_{l=1}^{N} \int_{\Omega} \rho u_{i,n} \phi_{j,n} u_{i,l} \frac{\partial \phi_{i,m}}{\partial x_{j}} \phi_{i,m} d\Omega \vec{e}_{i} = N(\vec{u})\vec{u} \quad (110)
$$

$N(\vec{u})\vec{u}$ is the non-linear convection term.

$$
\int_{\Omega} \eta \nabla \vec{u} \cdot \nabla \vec{\omega} d\Omega = \sum_{i=1}^{3} \sum_{j=1}^{3} \sum_{n=1}^{N} \sum_{l=1}^{N} \int_{\Omega} \eta u_{i,n} \nabla \phi_{i,n} \cdot \nabla \phi_{j,m} d\Omega \vec{e}_{i} \quad (111)
$$

The stiffness matrix $S$ is defined as:

$$
S^{ij} = \int_{\Omega} \eta \nabla \phi_{i,n} \cdot \nabla \phi_{j,m} d\Omega \quad (112)
$$

$$
- \int_{\Omega} p \nabla \cdot \vec{\omega} d\Omega = - \sum_{i=1}^{3} \sum_{k=1}^{K} p_{i} \psi_{k} \frac{\partial \phi_{i,m}}{\partial x_{i}} d\Omega \vec{e}_{i} \quad (113)
$$

The divergence matrix $L$ is defined as:

$$
L^{i} = - \int_{\Omega} \psi_{k} \frac{\partial \phi_{i,m}}{\partial x_{i}} d\Omega \quad (114)
$$

$$
\int_{\Omega} \vec{f} \cdot \vec{\omega} d\Omega = \sum_{i=1}^{3} \int_{\Omega} f_{i} \phi_{i,m} d\Omega \vec{e}_{i} \quad (115)
$$

The force vector can be defined as:

$$
f^{i} = \int_{\Omega} f_{i} \phi_{i,m} d\Omega \quad (116)
$$

The surface integrals will vanish due to the applied boundary conditions. On $\Gamma_{0}$, where $\vec{u} = \vec{0}$, the testfunction $\vec{\omega}$ is chosen equal to zero. On $\Gamma_{1}$, where $(\sigma \cdot \vec{n}) \cdot \vec{n} = 0$, the surface integrals will become zero as well.

$$
\int_{\Gamma} \eta \nabla \vec{u} \cdot \vec{n} \cdot \vec{\omega} d\Gamma + \int_{\Gamma} p \vec{\omega} \cdot \vec{n} d\Gamma = \int_{\Gamma} (\sigma \cdot \vec{n}) \cdot \vec{n} d\Gamma = 0 \quad (117)
$$

The terms in the continuity equation can be written as:
Numerical simulation of Reaction Injection Molding

\[ \int \{ p + \frac{1}{\varepsilon} \nabla \cdot \vec{u} \} \varrho d\Omega = \int \sum_{i=1}^{3} \sum_{k=1}^{K} \sum_{n=1}^{N} \{ p_k \psi_k \psi_i + \frac{1}{\varepsilon} \mu \frac{\partial \phi_{in}}{\partial x_i} \psi_i \} d\Omega \] (118)

The pressure matrix \( M_p \) is defined as:

\[ M_p = \int \psi_k \psi_i d\Omega \] (119)

Both the Navier-Stokes equation and the continuity equation can be rewritten, leading to a set of non-linear ordinary differential equations:

\[ \begin{cases} M \vec{u} + S \vec{u} + N(\vec{u}) \vec{u} + \frac{1}{\varepsilon} L^T M_p^{-1} L \vec{u} = \vec{f} \\ p = \frac{1}{\varepsilon} L^T M_p^{-1} L \vec{u} \end{cases} \] (120)

For \( \varepsilon \) in the limit to zero, the solution obtained with the penalty function approach converges to the solution of the direct method. The advantage of the penalty function approach is that the pressure can be eliminated, resulting in a smaller set of equations. Applied boundary conditions different from homogeneous Dirichlet boundary conditions as imposed in the above derivation of the set of non-linear ordinary differential equations give similar variational problems. E.g., inhomogeneous Dirichlet boundary conditions lead to an extension of the right hand side vector.

5.1.1 Time integration

The time derivative can be approximated by a finite difference \( \theta \)-method, resulting in:

\[ M \frac{\vec{u}^{n+1} - \vec{u}^n}{\Delta t} + \theta[S + N(\vec{u}^{n+1})] + \frac{1}{\varepsilon} L^T M_p^{-1} L \vec{u}^{n+1} = \vec{f}_{n+\theta} \]

\[ - (1 - \theta)[S + N(\vec{u}^n)] + \frac{1}{\varepsilon} L^T M_p^{-1} L \vec{u}^n \] (121)

\[ p^{n+1} = \frac{1}{\varepsilon} L^T M_p^{-1} L \vec{u}^{n+1} \]

where \( \vec{u}^n \) and \( \vec{u}^{n+1} \) are the velocity fields on the respective timesteps \( t^n \) and \( t^{n+1} \).

A Newton-Raphson linearization is used to linearize the non-linear advection term \( N(\vec{u}^{n+1}) \vec{u}^{n+1} \):

\[ N(\vec{u}^{n+1}) \vec{u}^{n+1} = N(\vec{u}^n) \vec{u}^{n+1} + N(\vec{u}^{n+1}) \vec{u}^n - N(\vec{u}^n) \vec{u}^n = J(\vec{u}^n) \vec{u}^{n+1} - N(\vec{u}^n) \vec{u}^n \] (122)

where \( J(\vec{u}^n) \) is the Jacobian of \( N(\vec{u}^n) \vec{u}^n \).
This leads to a one-step solution procedure:

\[
\left\{ \frac{M}{\Delta t} + \theta [S + J(u^n) + \frac{1}{\epsilon} L^T M_p^{-1} L] \right\} \vec{u}^{n+1} = \vec{f}^{n+\theta} \\
+ \left\{ \frac{M}{\Delta t} + (1 - \theta) [S + \frac{1}{\epsilon} L^T M_p^{-1} L] \right\} \vec{u}^n - \frac{(1 - 2\theta)}{\Delta t} N(u^n) \vec{u}^n
\]

As this solution procedure leads to worse pressure approximations as a result of undamped oscillations, an equivalent two-step procedure is applied, replacing \(u^{n+1}\) by \(\frac{1}{\theta} \{ \vec{u}^{n+\theta} - (1 - \theta) \vec{u}^n \} \):

- **step 1:**

\[
\left\{ \frac{1}{\theta \Delta t} M + S + J(u^n) + \frac{1}{\epsilon} L^T M_p^{-1} L \right\} \vec{u}^{n+\theta} = \vec{f}^{n+\theta} + \left\{ \frac{1}{\theta \Delta t} M + N(u^n) \right\} \vec{u}^n
\]

\[
p^{n+\theta} = \frac{1}{\epsilon} L^T M_p^{-1} L \vec{u}^{n+\theta}
\]

- **step 2:**

\[
\vec{u}^{n+1} = \frac{1}{\theta} \{ \vec{u}^{n+\theta} - (1 - \theta) \vec{u}^n \}
\]

This two-step procedure is less time- and space-consuming and simpler to implementate.

From a detailed study on the unconditional stability and long-term behaviour of transient algorithms for the incompressible Navier-Stokes and Euler equations (Simo and Armero, 1994), it can be concluded that the \(\theta\)-method as described in this section is unconditionally stable for \(\theta \geq \frac{1}{2}\). The best choice, when short-term transient simulations are considered and accuracy is an important factor, is the Crank-Nicholson scheme with \(\theta = \frac{1}{2}\).

### 5.1.2 Contact lines.

In section 4.3 the problem of stress singularities at contact points, modelling flows with moving boundaries, is discussed. It was concluded that several options are available to evade this problem. Most options concern conditions for the contact angle. Since in this project a three-dimensional model will be developed, these options are not adequate. Therefore, it is chosen to alleviate the stress singularities by a relaxation of the no-slip boundary condition in the neighbourhood of the contact line. Naturally, in the mold, filling polymer, the no-slip condition is preserved. For the air, pushed out of the mold, free tangential slip is supposed.
5.2 The advection equation, front tracking.

As mentioned in the introduction, the position of the moving front is determined by solving an advection equation. All 'particles' or points in the fluid domain are given a unique set of scalars, where $c(\vec{x}, t)$ is the scalar field. These scalars represent the identity of the 'particles' and are transported by convection only. It may be evident that this scalar field can be used as an indicator in assigning values for the viscosity. Solving the advection equation can be done both with a Langrangian or an Eulerian approach. Numerical experiments have showed that the Eulerian approach is the most flexible and the most efficient approach (Caspers, 1991). The advection equation is:

$$\frac{\partial c}{\partial t} + \vec{u} \cdot \nabla c = 0$$  \hspace{1cm} (126)

In the weak formulation this equation becomes:

$$\int_{\Omega} \left( \frac{\partial c}{\partial t} + \vec{u} \cdot \nabla c \right) \omega d\Omega = 0$$  \hspace{1cm} (127)

with boundary condition $c(\vec{x}, 0) = c_0(\vec{x})$ in $\Omega$. Again the standard Galerkin method is applied:

$$c_h = \sum_{j=1}^{N} c_j(t) \phi_j(\vec{x})$$  \hspace{1cm} (128)

$$\omega_h = \phi_i$$  \hspace{1cm} (129)

The advection equation is written as:

$$\int_{\Omega} \left( \frac{\partial c}{\partial t} + \vec{u} \cdot \nabla c \right) \omega d\Omega = \sum_{j=1}^{N} \frac{\partial c_j}{\partial t} \int_{\Omega} \phi_j \phi_i d\Omega + \sum_{j=1}^{N} c_j \left[ \int_{\Omega} (\vec{u} \cdot \nabla \phi_j) \phi_i d\Omega \right]$$  \hspace{1cm} (130)

The mass matrix $M$ is defined as:

$$M = \int_{\Omega} \phi_j \phi_i d\Omega$$  \hspace{1cm} (131)

The stiffness matrix $N$ is now defined as:

$$N(\vec{u}) = \int_{\Omega} (\vec{u} \cdot \nabla \phi_j) \phi_i d\Omega$$  \hspace{1cm} (132)

The advection equation can then be written as:

$$M \dot{c} + N(\vec{u}) c = 0$$  \hspace{1cm} (133)
5.2.1 Time integration

The time derivative in the advection equation can be approximated by a finite difference \( \theta \)-method as for the Navier-Stokes equation, resulting in:

\[
M \frac{c^{n+1} + c^n}{\Delta t} + \theta N(\mathbf{u}^{n+1})c^{n+1} + (1 - \theta)N(\mathbf{u}^n)c^n = 0 \tag{134}
\]

where \( c^n \) and \( c^{n+1} \) are the scalar fields on the respective timesteps \( t^n \) and \( t^{n+1} \).

This leads to a one-step solution procedure:

\[
[M + \theta \Delta t N(\mathbf{u}^{n+1})]c^{n+1} = [M - (1 - \theta)\Delta t N(\mathbf{u}^n)]c^n \tag{135}
\]

Again an equivalent two-step solution procedure can be obtained by replacing \( c^{n+1} \) by \( \frac{1}{\theta} c^{n+\theta} - (1 - \theta)c^n \) and recalling that \( N \) is a linear operator and \( \mathbf{u}^{n+1} = \frac{1}{\theta} \{\mathbf{u}^{n+\theta} - (1 - \theta)\mathbf{u}^n\} \).

Therefore:

\[
N(\mathbf{u}^{n+1}) = \frac{1}{\theta}[N(\mathbf{u}^{n+\theta}) - (1 - \theta)N(\mathbf{u}^n)] \tag{136}
\]

\[
\frac{1}{\theta \Delta t} M c^{n+\theta} - \frac{1}{\theta \Delta t} M c^n + \left\{\frac{1}{\theta} N(\mathbf{u}^{n+\theta}) - \frac{1 - \theta}{\theta} N(\mathbf{u}^n)\right\}c^{n+\theta}
\]

\[
- (1 - \theta)\left\{\frac{1}{\theta} N(\mathbf{u}^{n+\theta}) - \frac{1 - \theta}{\theta} N(\mathbf{u}^n)\right\}c^n + (1 - \theta)N(\mathbf{u}^n)c^n = 0 \tag{137}
\]

This can be written as:

\[
\frac{1}{\theta \Delta t} M c^{n+\theta} - \frac{1}{\theta \Delta t} M c^n + N(\mathbf{u}^{n+\theta})c^{n+\theta}
\]

\[
+ \frac{1 - \theta}{\theta} \{N(\mathbf{u}^{n+\theta}) - N(\mathbf{u}^n)\}c^{n+\theta} - \frac{1 - \theta}{\theta} \{N(\mathbf{u}^{n+\theta}) - N(\mathbf{u}^n)\}c^n = 0 \tag{138}
\]

\[
\frac{1}{\theta \Delta t} M c^{n+\theta} - \frac{1}{\theta \Delta t} M c^n + N(\mathbf{u}^{n+\theta})c^{n+\theta}
\]

\[
+ \frac{1 - \theta}{\theta} \{N(\mathbf{u}^{n+\theta}) - N(\mathbf{u}^n)\}\{c^{n+\theta} - c^n\} = 0 \tag{139}
\]

As \( N \) is a linear operator, it follows that:

\[
N(\mathbf{u}^{n+\theta}) - N(\mathbf{u}^n) = N(\mathbf{u}^{n+\theta} - \mathbf{u}^n) \tag{140}
\]

The order of accuracy for Taylor expansions is e.g.:

\[
c^{n+\theta} = c(t^{n+\theta}) = c(t + \theta \Delta t) = c(t) + \theta \Delta t \frac{\partial c}{\partial t} + O((\Delta t)^2) \tag{141}
\]

This leads to a two-step solution procedure for the advection equation of \( O((\Delta t)^2) \):

- step 1:

\[
\frac{1}{\theta \Delta t} M c^{n+\theta} - \frac{1}{\theta \Delta t} M c^n + N(\mathbf{u}^{n+\theta})c^{n+\theta} = 0 \tag{142}
\]
step 2:

\[ c^{n+1} = \frac{1}{\theta} [c^{n+\delta} - (1 - \theta)c^n] \quad (143) \]

For simplicity reasons and time and space consuming considerations, the two step solution procedure is preferred.

### 5.2.2 Upwinding

Convection dominated flows result in the numerical solution of hyperbolic equations. During the last ten years several finite element methods have been developed to solve this kind of equations, using various upwinding techniques to increase numerical stability. Amongst these techniques is the Streamline Upwind Petrov Galerkin (SUPG) method, which is widely used. In this method the test function \( \tilde{\omega} \) consists of two parts

\[ \tilde{\omega} = \omega + \alpha \bar{u} \cdot \nabla \omega \quad (144) \]

in which \( \alpha \) is the upwind parameter, which weights the information upstream more heavily than downstream. The weak form is now given by

\[ \int_\Omega \left\{ \left( \frac{\partial c}{\partial t} + \bar{u} \cdot \nabla c \right)\omega + \left( \frac{\partial c}{\partial t} + \bar{u} \cdot \nabla c \right)(\alpha \bar{u} \cdot \nabla \omega) \right\} d\Omega = 0 \quad (145) \]

For the pure convection equation, two choises of the parameter \( \alpha \) can be made (Zoetelief, 1995; Vreugdenhil and Koren, 1993):

- the classical upwind scheme: \( \alpha = \frac{h}{2||\bar{u}||} \)
- a time dependent parameter: \( \alpha = \left\{ \left( \frac{2}{\Delta t} \right)^2 + \left( \frac{2||\bar{u}||}{h} \right)^2 \right\}^{1.5} \)

where \( h \) is the element size and \( \Delta t \) the timestep. The time dependent parameter is originally derived by Shahib (1988).

From results of the Molenkamp test, it is concluded (Zoetelief, 1995) that the SUPG method with Shahib's approximation of the upwind parameter gives the best approximation. This upwinding technique is used in this project.

### 5.3 Surface tension effects

In this project, research is done on the possibility of implementing surface tension effects on interfaces that do not coincide with grid lines. The approach is based on the continuum surface force model, CSF, from Brackbill et al. (1991), that gives satisfying results for their finite difference computations. As discussed in section 4.4, this model interprets surface tension as a continuous, three dimensional effect across an interface instead of a boundary value condition on the interface. For this project, a uniform isothermal surface
tension is assumed, with no surface contamination present. The surface stress boundary conditions at the interface between two fluids A and B is now simplified to

\[ p_A - p_B - 2\mu_A \hat{n}_k \left( \frac{\partial u_k}{\partial n} \right)_A + 2\mu_B \hat{n}_k \left( \frac{\partial u_k}{\partial n} \right)_B = \gamma \kappa \]  

(147)

The tangential component is assumed to be zero. Therefore, the total surface force is

\[ \mathbf{F}_{ss}(\vec{x}_s) = \gamma \kappa(\vec{x}_s) \hat{n}(\vec{x}_s) \]  

(148)

Using the CSF model this can be written as a volume force

\[ \mathbf{F}_{sv}(\vec{x}) = \gamma \kappa(\vec{x}) \frac{\nabla \hat{c}(\vec{x})}{[c]} \]  

(149)

where \([c]\) is the jump in color, \([c] = c_B - c_A\) and the curvature can be written as

\[ \kappa = -(\nabla \cdot \hat{n}) \quad \text{with} \quad \hat{n}(x) = \frac{\nabla \hat{c}(\vec{x})}{|\nabla \hat{c}(\vec{x})|} \]  

(150)

The viscosity, a step-function for which a linear interpolation is used to obtain continuity, is used as the color function. Since Brackbill et al. (1991) showed the importance of smoothing, a diffusion step is applied to the viscosity field.

The curvature can be calculated by taking the divergence of the unit normal using the derivatives of the basis functions. An other method, for two-dimensional problems, is to calculate a mean unit normal per computation cell at the cell center, that is per element. For each nodal point of that element the curvature can be computed by calculating the point of intersection between the elements mean unit normal and the unit normal of that nodal point. The distance \(R_1\) between the concerning nodal point and the point of intersection as well as the distance \(R_2\) between the cell center and the point of intersection are measures for the local radius. The curvature and the concerning nodal point is defined as

\[ \kappa = \frac{1}{\overline{R}} \quad \text{with} \quad \overline{R} = \frac{R_1 + R_2}{2} \]  

(151)

As test problem a static liquid drop is used as was done by Brackbill et al. (1991), see section 4.4.

### 5.4 Implementation in FEM-package SEPRAN

The essential parts of the numerical process are presented in the next flow scheme.
Numerical simulation of Reaction Injection Molding

input data

initialization

new time step

new iteration step

calculation velocity field, N.S.

calculation scalar field, F.T.

calculation new viscosity

convergence ?

Possible calculation surface tension

last time step ?

exit
• Input data

This includes the input of the geometrical data, the process parameters and the filling of the coefficients.

• Initialization

The initial solutions are created, especially the initial scalar field, the initial velocity field and, if necessary, the surface tension force.

• Calculation velocity field, N.S.

The calculation of the velocity field is done according to the two-step \( \theta \)-method as discussed in section 5.1.

• Calculation scalar field, F.T.

The scalar field is calculated using the two-step solution procedure as discussed in section 5.2.

• Calculation new viscosity

The viscosity is determined by the scalar field.

• Convergence?

The convergence is checked with respect to the velocity and scalar fields. 
\( \bar{u}^{i+1} - \bar{u}^i < 10^{-5} \) and \( c^{i+1} - c^i < 10^{-5} \)

• Possible calculation surface tension

To do some research on modelling surface tension effects, the surface tension force can be calculated as described in section 5.3.

The computational domain for the finite element calculations has to be subdivided into elements. For two-dimensional problems, a 7-noded Crouzeix-Raviart element is used (Cuvelier et al., 1986), an extended isoparametric quadratic triangle. In the three-dimensional case, a 27-noded Crouzeix-Raviart element is used, a tri-quadratic hexahedron. Velocity unknowns are defined in all the nodal points of the element. Pressure unknowns are only defined in the center node.

5.5 Test flow problems

The problems described in this section are used to test the numerical finite element model. The first problem described concerns a Laser Doppler Anemometry experiment.
5.5.1 Laser Doppler Anemometry experiment

To simulate experimentally the filling stage of an injection molding process, a quasi two-dimensional measuring section is constructed as shown in figure 19. The inner dimensions of the measuring section are 14.8 x 91 x 300 mm. In stead of injecting the fluid into the mold the walls of the mold are given a velocity of 0.04 ms⁻¹ while the fluid is at 'rest'. This is realized by moving two thin plates along the walls into the channel. The section was filled with a mixture of water and glycerol, having a viscosity of 5.56 mPas and a density of 1125 kgm⁻³. Measurements are made when a fully-developed flow has been established. Only the axial velocities are measured along paths parallel to the initial flow front in the middle of the channel.

![Figure 19: Schematic representation of the measuring section](image)

The quantitative velocity measurements have been performed by means of an optical dual beam Laser Doppler Anemometry system in forwardscatter mode. The principle of LDA is based on detecting the Doppler frequency shift of laser light that is scattered by small particles moving with the fluid. From this Doppler frequency shift the velocity is determined. A 5 mW Helium-Neon laser was used generating a laser beam of wavelength λ = 623.8 nm. This beam is splitted, later, into a principle beam and a reference beam. A front lens with a focal length of 80 mm focuses the principle laser beam and the reference beam to describe a measuring volume of 1132 x 78 x 78 μm.

5.5.2 Bifurcation

Most plastic molds are no straight cavities but have bifurcations. The melt flow then has to split in order to fill all branches. In most simulation codes used for simulating mold filling processes, nearly all 2½-D codes, this splitting leads to problems. In most codes all properties (like e.g. temperature) are equalized in the intersection points, except for the pressure gradients, which naturally differ for each branch. This leads to unrealistic distribution of properties of material particles. The origin of erroneous distributions stems
from the fact that the mass balance is only globally satisfied. Further, particles are being cloned, and appear unphysically at different sites at the same time. Zoetelief (1995) has used a more realistical model. The product geometry is split into parts that contain no bifurcations and that can be considered as two-dimensional flow geometries. In case of e.g. a T-junction as in figure 20, the conditions at the outflow boundary of part A are mapped as inflow conditions at the inflow boundaries of parts B and C with regard of the height of both parts and the pressure gradients in the branches. Figure 21 shows a time-label distribution at times that one of the branches is filled. The polymer has a viscosity of 1000 Pa.s and a density of 1000 kgm^{-3}. The computations have been made for isothermal filling.

Figure 20: Mapping outflow boundary conditions part A on inflow boundary of part B and C as done by Zoetelief

With the numerical model described in section 4 the same calculations have been made.
Figure 21: Time label distribution for a bifurcation with equal sized branches (top) and one with branches of different dimension (bottom) done by Zoetelief.
5.5.3 Gas-assisted injection molding

Gas-assisted injection molding is a low-pressure variation of conventional injection molding, that can be used to produce hollow thermoplastic parts. This molding process offers a variety of potential benefits. It provides a more uniform packing pressure, reducing thermal stresses and, consequently, warpage. Machine pressure and product cycle times can be reduced as well. However, the fundamental physics of the process seems not to be completely understood. Therefore more research is done. Good numerical simulation of the gas-assisted injection molding process might be essential to obtain a better understanding of the process and eventually, to obtain a mold design tool. In most simulation attempts, the modeling of the second flow front in the gas behind the polymer is a major problem. Especially, since most simulation programs use front models, which implies application of remeshing techniques. With two fronts present, remeshing gets even more complicated. Besides, front models are very unsatisfactory in simulations to analyze flow front instabilities. Furthermore, gas-assisted injection molding is in essence three-dimensional.

For testing purposes and to show the possibilities of the developed numerical model first a two-dimensional cavity is filled with polymer followed by gas injection. Next gas-assisted filling of a three-dimensional mold is simulated. The 3D-computational domain is shown in figure 22. The material viscosity is 1000 Pa.s and the density is 1000 kgm\(^{-3}\).

![Figure 22: Computational domains](image)
6 Results

6.1 Numerical stability and slip boundary conditions

The test problem concerns the filling of a two-dimensional cavity, see figure 23. The computational domain is subdivided into 1600 elements (40 x 40). The viscosity of the 'polymer' was chosen 11 Pa.s, the density 1000 kgm\(^{-3}\). The 'air' was given a viscosity of 1.7\(\times\)10\(^{-5}\) Pa.s, the density is chosen the same as for the 'polymer'.

\[ \Gamma_1 \]
\[ \Gamma_2 \]
\[ \Gamma_3 \]
\[ \Gamma_4 \]

Figure 23: Numerical conditions for the filling of a 2D cavity

- \( \vec{v} = \vec{v}_0 \) on \( \Gamma_1 \), inflow condition
- \( (\vec{v} \cdot \vec{n}) = 0 \) on \( \Gamma_2 \), symmetry condition
- \( \vec{v} = \vec{v}_0 \) on \( \Gamma_4 \), no-slip in 'polymer' and free slip in 'air'
- \( \vec{\sigma} \cdot \vec{n} = 0 \) on \( \Gamma_3 \), stress free outflow

The major strength of the developed numerical model is that remeshing is avoided. Therefore, the position of the flow front is determined by the jump in a label field related with the viscosity. This discontinuity is convected with the local velocity, calculated from the governing Navier-Stokes equations. However, this approach generates problems with regard to the contact lines. The no-slip condition, which is valid at the walls, hinders the flow front to go up along those walls. As shown in figure 24, a layer of air keeps stucked at the walls. In this figure, the position of the flow front for a test problem is given as well as contour plots of the time label distribution visualizing fountain flow.

To prevent this phenomenon, one can think of relaxation of the no-slip boundary condition. In the 'polymer', the no-slip boundary condition is preserved. The 'air' is given a tangential slip boundary condition. The slip-velocity is based on the mean velocity of the polymer, since a flow front moves with the mean velocity of the fluid. The slip boundary condition can be applied by two methods. In the first method a so called 'free' slip is given at the walls

\[ \vec{\sigma}_f = \vec{n} \cdot (\vec{n} \cdot \vec{n}) \vec{n} = \vec{0} \]  \hspace{1cm} (152)
Figure 24: Position of the flow front, the color yellow represents the 'air' and contour plots of the time label distribution, the color blue represents the 'air', with no-slip boundary conditions.

Figure 25: Position of the flow front, yellow represents the 'air', and contour plots of the time label distribution, blue represents the 'air', with free-slip boundary conditions in air.

Figure 26: Position of the flow front, yellow represents the 'air', and contour plots of the time label distribution, blue represents the 'air', with slip boundary conditions in air based on stationary lubrication layer problem.
In figure 25 it is seen that this method leads to instabilities, which are probably due to the discontinuity of viscosity at the interface.

In the second method, the slip velocity is computed by first solving a stationary problem without a discontinuous viscosity. At the walls a lubrication layer is present, in which the fluid is given a free tangential slip. The viscosity of the polymer is used. The computed velocities in the lubrication layer are used as the slip velocity boundary condition of the 'air', providing more satisfactory results, figure 26. However, this method of determining the slip velocity for air is physically incorrect. Better ways to determine the slip velocity have to be looked for, investigated and experimentally validated. Nevertheless, since the results seem in first instance not too bad, this approach of applying slip boundary conditions is used in the rest of this study.

A discontinuity in the computational domain is very often the source of numerical instability. In figure 27 it is seen that using a Crank-Nicholson time integration scheme many instabilities occur. However, when the Euler implicit scheme is used, all instabilities caused by the discontinuity are damped.

The time integration scheme, as discussed in section 5.1.2 is unconditionally stable for $\theta \geq \frac{1}{2}$. For $\theta = \frac{1}{2}$ this scheme is $O(\Delta t^2)$ accurate. For $\theta = 1$, this scheme reduces to the Euler implicit scheme, which is of accuracy $O(\Delta t)$. However, the damping properties of this scheme are excellent, comparing figure 26 and figure 27. Therefore, the Euler implicit time integration scheme is used further.

![Figure 27: Contour plots of the time label distribution with slip boundary conditions in air base on stationary lubrication layer problem for $\theta = \frac{1}{2}$, blue represents the 'air'](image)

### 6.2 Three dimensional cavity flow

A cavity flow is relatively easy to verify through experiments. In future, these verification experiments probably will be made. Computations have already been performed to compare two-dimensional and three-dimensional modelling. The computational domain for the cavity flow as shown in figure 28 is subdivided into 360 elements in the three-dimensional and into 1600 elements in the two-dimensional case. The viscosity is 11 Pa.s and the density is 1000 kgm$^{-3}$. The inflow velocity is $v_0 = 0.04 \text{ m s}^{-1}$. 
Results

Figure 28: Boundaries of a cavity flow problem

- \( \vec{v} = \vec{v}_0 \) on \( \Gamma_1 \)
- \( (\vec{v} \cdot \vec{n}) = 0 \) on \( \Gamma_2 \) and \( \Gamma_3 \), symmetry condition
- \( \vec{v} = \vec{v}_0 \) on \( \Gamma_4 \) and \( \Gamma_5 \), no-slip in 'polymer' and free tangential slip in 'air'
- \( \sigma \cdot \vec{n} = \vec{0} \) on \( \Gamma_6 \), strain free outflow

Figure 29 shows the timel label distribution at \( t = 4s \) for two intersections. The first intersection is in the middle of the channel \( (z = 44.9 \, \text{mm}) \) and can be compared with figure 30. The second intersection is at 3 mm from the side wall and is in essence three-dimensional. Figure 30 shows the time label distribution at \( t = 4s \) for the equivalent two-dimensional problem for a mesh subdivided into 1600 elements. Differences are mainly due to the coarseness of the three-dimensional mesh. This is seen in figure 31, where a 2D-grid as coarse as the 3D-grid is used, 120 elements. Coarse meshes are not capable to resolve the fountain flow phenomenon. Nevertheless, a rather good agreement is seen between 2D and 3D simulations.
Figure 29: Time label distribution at $t = 1.5s$ for (top) $z = 44.9$ mm and (bottom) $z = 3$ mm for the three-dimensional computations, blue represents 'air'

Figure 30: Time label distribution at $t = 1.5s$ for the two-dimensional computations on a fine mesh, blue represents 'air'

Figure 31: Time label distribution at $t = 1.5s$ for the two-dimensional computations on a coarse mesh, blue represents 'air'
6.3 Test flow problems

6.3.1 Laser Doppler Anemometry results

The velocity data obtained with the LDA system are presented in figure 32 and 33. In figure 32 the velocity profiles for the different measuring sites behind the flow front are presented. It is clearly seen that, close to the flow front, the velocity profile is flattened. More downstream a parabolic velocity profile is developed. Figure 33 gives the axial velocity for each site respectively, with \( y \) the distance to the flow front. The plates moved with velocity \( v = 0.04 \text{ ms}^{-1} \) into the fluid.

![Velocity profiles for different sites downstream the flow front](image)

Analyzing the experimental results the following considerations have to be taken into account. First, the exact position of the flow front is not precisely known as the fluid level rises a little while pushing the plates into the channel, notwithstanding that a constant head tank is used. Second, stowage occurs when the plates move through the fluid domain. In the discussion the experimental results will be compared with some numerical results.
Figure 33: Measured velocities for 8 different sites with z the position along the width of the channel and y representing the distance to the flow front.
6.3.2 Bifurcation

Simulating mold filling processes in bifurcations, leads for most simulation codes to problems as described in section 5.5.2. The present numerical model is used to compute the complete velocity field and time label distributions in T-junctions. The computational domain is subdivided into 1704 elements. T-junction 1 consists of three equal-sized branches of $2.5 \times 40$ mm. T-junction 2 consists of three branches with dimensions: A: $2.5 \times 40$ mm; B: $1.5 \times 40$ mm; and C: $2.5 \times 30$ mm. The inflow velocity is $0.094 \text{ ms}^{-1}$.

Having a computational domain containing sharp edges turbulent flow structures occur in the 'air'. This is probably due to the high Reynold's number in the air region, $Re = 10000$. Therefore, the viscosity of 'air' is replaced by a higher viscosity such that turbulent flow structures are avoided. It has also been tried to close one of the branches when it was filled, which resulted in strong numerical instabilities.

Figure 34 shows the time label distributions for T-junction 1 for two different moments in time. At $t = 1.5$ s the branches have been filled completely. It is seen that the fluid is splitted equally, not favouring one of the branches. Figure 35 shows the time label distributions for T-junction 2 for two different moments in time. At $t = 1.5$ s the top branch has been filled completely. Looking at the results it is seen that the particles are favoritely choosing the channel with the smallest resistance. It is also seen that in the middle section there is still 'air' left in one of the corners. Although slip boundary conditions are given in 'air', the sharp edges present practical programming problems. Therefore, these points are given a no-slip boundary condition, even in 'air', and are probably acting as sources of instability. These instabilities are damped later on.
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Figure 34: Time label distribution of T-junction 1 at (top) $t = 0.375s$ and (bottom) $t = 1.5s$, blue represents 'air'
Figure 35: Time label distribution of T-junction 2 at (top) $t = 0.365s$ and (bottom) $t = 1.5s$, blue represents 'air'
6.3.3 Gas-assisted injection molding

In simulation programs using remeshing techniques and front models, the presence of a second front leads to major problems. Using the developed numerical model, a two-dimensional cavity of 0.038 x 0.4 m is filled with polymer at a velocity $v_0 = 0.06 \text{ms}^{-1}$. At $t = 4 \text{s}$ 'air' is injected. The viscosity of the polymer is 11 Pa.s and of the 'air' is $1 \cdot 10^{-2} \text{Pas}$. The uniform density is 1000 kgm$^{-3}$. The computational domain, half of the cavity, is subdivided into 1600 elements. Figure 36 shows at $t = 6 \text{s}$, a well developed second flow front. The injected 'air' leaves a layer of polymer along the walls of constant thickness. This layer is expected regarding industrial gas-assisted injection molding.

Figure 36: Time label distribution of two-dimensional gas-assisted injection molding of a cavity, green represents 'air'

As an example of further application possibilities for the numerical model, mold filling of a more complex three-dimensional mold is simulated. Again gas-assisted injection has been simulated. The computational domain as shown in figure 22 is subdivided into 332 elements. The mold is filled through surface $\Gamma_1$ with velocity 0.1 ms$^{-1}$. Two situations are distinguished. In the first case, the boundary surfaces $\Gamma_2$ and $\Gamma_3$ are the outlets. In the second case, the boundary surface $\Gamma_4$ is the outlet and $\Gamma_2$ and $\Gamma_3$ are closed. The time label distributions of the first case are shown in figure 38 and 39. Intersections are presented for $x = 0.0002 \text{ m}$, $y = 0.002 \text{ m}$, $z = 0.005 \text{ m}$ and $z = 0.015 \text{ m}$, see figure 37.

Figure 37: The intersections as presented in figure 38 and 39

The irregular form of the contour lines is due to the coarseness of the mesh. The results of the second case are shown in figure 40 and figure 41. As expected the fluid flow chooses the way of the least resistance towards the free outlet. Although the mesh is coarse, a
Results

good qualitative impression is given by these simulations.

Figure 38: Time label distribution of case 1 at (top) $t = 1.6s$ and (bottom) $t = 3.2s$ for (left) $x = 0.002$ and (right) $y = 0.002$, green represents 'air'
Figure 39: Time label distribution of case 1 at (top) $t = 1.6s$ and (bottom) $t = 3.2s$ for (left) $z = 0.005$ and (right) $z = 0.015$, green represents 'air'
Figure 40: Time label distribution of case 2 at (top) $t = 1.6s$ and (bottom) $t = 3.2s$ for (left) $x = 0.002$ and (right) $y = 0.002$, green represents 'air'
Figure 41: Time label distribution of case 2 at (top) $t = 1.6s$ and (bottom) $t = 3.2s$ for (left) $z = 0.005$ and (right) $z = 0.015$, green represents 'air'
6.4 Modelling surface tension; a static liquid drop

Research on modelling surface tension on interfaces that do not coincide with a grid line, has been made. In section 4.4 the continuum surface force model of Brackbill et al. (1991) is discussed. It is concluded that for finite difference computations this model gives good results. In this section, the model is tested using a finite element method on a static liquid drop. For the color function, the viscosity is used. As is shown by Brackbill et al. (1991) smoothing of this function is essential. The viscosity is smoothed using a diffusion step with a diffusion coefficient of $10^{-4}$. The unsmoothed and smoothed viscosity functions for the test problem of a static liquid drop, are plotted in figure 42 together with the computed unit normals. The computational domain is subdivided into 2500 elements. The viscosity of the background fluid is 11 Pa.s, the drop viscosity is $1 \cdot 10^{-2}$ Pa.s and the density is 1000 kgm$^{-3}$ uniformly. The surface tension is taken to be $1 \cdot 10^{-2}$. The radius of the drop is 0.01 m.

![Figure 42: The unsmoothed (topleft) and smoothed (bottomleft) viscosity function for a static drop resulting in respectively a unit normal field (topright) and (bottomright)](image)

As described in section 5.3 two methods to compute the curvature are investigated. The results are shown in figure 43, using the smoothed viscosity function. The method that uses the standard SEPRAN calculation of the divergence (Segal, 1995) is referred to as div1. The method computing the intersection of unit normals with the mean unit normal of the computational cell to compute the curvature, is referred to as div2. The second
method provides the best results. Although the curvature shows no negative values for this approach, the calculated curvature is still too high.

Figure 43: The curvature computed with (left) div1 and (right) div2

A plot of the curvature on the whole computational domain, using div2, shows irregularities, see figure 44. The resulting surface tension forces, force the drop to deform in a non-physical way, as shown in figure 45. This implies that even better smoothing is required.

Figure 44: The curvature computed with div2

Figure 45: The surface force vectors computed with div2 for several time steps resulting in deformation of the drop. (left) $t = 0$ s; (right) $t = 2$ s
7 Discussion and conclusions

A major advantage of Reaction Injection Molding processes is that, due to the low initial viscosity, relatively low temperatures and pressures are needed to fill a mold. Some of the disadvantages of reaction injection molding, however, also arise from its low viscosity. Gas bubbles may get trapped during filling, due to instability of the flow front. To analyze the mold filling stage of RIM processes is, therefore, of great interest to obtain a better understanding and eventually a mold design tool.

From literature study it is concluded that inertial and surface tension effects play a significant role in Reaction Injection Molding processes. It is also seen that currently available mathematical models have not allowed a good enough simulation of the process. Therefore, a three-dimensional numerical model for the filling stage of RIM is developed. Since remeshing for three-dimensional problems is complex and time and memory-consuming, the numerical model is based on a fixed grid method.

The governing Navier-Stokes equations are solved with a standard Galerkin finite element method, using the penalty function approach. Non-linear terms are linearized by a Newtonian-Raphson linearization. In order to determine the flow front position, the passive scalar advection equation is used to advect a discontinuous scalar function. The viscosity of the computational domain is a function of this scalar field.

In the following paragraphs, the numerical stability and the strategy for solving the contact line problem are discussed. Furthermore, the experimental verification as well as the eventual applications of the three-dimensional modelling is discussed. The surface tension model is examined. And finally, the main conclusions are drawn.

Numerical stability

Using discontinuous functions meets with several difficulties. One difficulty is that these discontinuities often appear to be a source of numerical instabilities. This is the main reason that for the time integration the Euler implicit scheme is used rather than the Crank-Nicholson time integration scheme. It is seen that by application of the Crank-Nicholson time integration scheme the numerical solution becomes unstable. The Euler implicit scheme has strong damping properties, necessary to avoid these numerical instabilities. This results in more stable numerical solutions. However, flow front instabilities of physical origin could be damped as well. Therefore, this model might not be good enough to analyze flow front instabilities.
Contact line treatment, no-slip relaxation

The modelling process for flows with free boundaries is complicated by the presence of contact lines. At these lines the continuum approach results in a discontinuous or unbounded stress tensor, resulting in a singularity. In the numerical model developed, this problem is solved by relaxation of the no-slip boundary condition. The 'air' preceding the polymer is given a tangential slip velocity at the walls, pulling the polymer along these walls. The slip velocity is determined by computing a stationary problem with a continuous viscosity field of the polymer. At the walls a lubrication layer is present with free tangential slip. The computed velocities in this layer are used as the slip boundary condition for the real problem. This approach is physically incorrect. Better ways to determine the slip velocity have to be looked for, investigated and experimentally validated. Nevertheless, this approach seems to provide rather good results.

Turbulent flow structures

Whenever a discontinuous viscosity function is used, with a polymer viscosity on the one hand and the viscosity of air on the other hand, the occurrence of turbulent flow structures has to be taken into consideration. Low viscosities as for air result in high Reynolds numbers, $Re = 10000$. Near e.g. sharp edges in complex geometries this results in the generation of turbulent flow structures, leading to divergence of the numerical process. Therefore, the viscosity of air has to be replaced by a higher viscosity. This does not effect the results explicitly.

Test problems and three-dimensional modelling

The numerical program is written for application of three-dimensional modelling. Computations show great resemblance between two-dimensional and three-dimensional solutions of equivalent problems. The main differences appear mainly to be caused by a coarser three-dimensional grid. It is seen that the model provides qualitatively good simulation of three-dimensional mold filling processes. More quantitative results can be achieved by finer meshing. Since this is very (computing-)time and memory consuming, faster Navier-Stokes solvers could be helpful.

Other application possibilities are shown by some test problems. Computing the filling of a bifurcated mold, it is seen that the velocity field and time label distributions are easily calculated throughout the whole bifurcation. Most simulation programs are not capable of computing velocity fields in the whole bifurcated flow domain and, therefore, are missing some essential information. However, looking at the results it can be concluded that the approach of flow splitting as done by Zoetelief (1995) with the VIP simulation program is good enough for the specific situation.
Modelling of gas-assisted injection molding suffers difficulties with modelling a second flow front. With the present model, this second flow front has been computed without problems and possibly can be used for developing a front model for other simulation programs like VIP. Also for the more complex three-dimensional situation, the model opens many possibilities for qualitative simulations. However, it is still very (computing-)time consuming.

**Experimental validation**

Laser Doppler velocity measurements have been made in a quasi two-dimensional measuring section. Downstream the flow front, a parabolic velocity profile is measured. Near the flow front, this velocity profile is flattened. In Figure 46 numerical and experimental results are compared for two different sides. Near the front ($y = -2.0$ mm) differences are very large. The numerical model, as is seen in the figure, does not give accurate results near the walls, due to the relaxation of the no-slip boundary condition. Further downstream, rather good agreement is found between experiment and computation. Nevertheless, better validation experiments should be made, as the experimental setup has some deficiencies. First, the exact position of the flow front is unknown as the fluid level rises a little, while pushing the plates into the fluid, notwithstanding that a constant head tank is used. Second, stowage occurs when the plates move through the fluid. Nevertheless, a good qualitative agreement is found.

![Figure 46: Comparison of experimental and numerical results at two different sites, y is the distance to the flow front](image_url)

**Modelling surface tension**

Modelling surface tension effects is not simple since the position of the flow front is not exactly determined, using a fixed mesh numerical method. For this method, no grid line
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coincides with the interface. In this study, research is made on the possibilities of applying surface tension forces. Brackbill et al. (1991) have developed a continuum surface force model for a finite difference approach. This model has been studied and tested in a finite element context. Major problems occurred as second order differentiation requires smoother basis functions than available in the present SEPRAN-package. Extra diffusion steps to smooth the interface gave no resolution. Options to provide better results probably are fitting of the flow front using splines or a remeshing or adaptive meshing strategy.

Conclusions and recommendations

The numerical model developed is based on a fixed grid method. To determine the position of the flow front a discontinuous scalar function is convected with the local velocity of the flow. Because of numerical stability considerations, the Euler implicit time integration scheme is used, for numerical diffusion is related with this scheme. Instabilities at the flow front of physical origin may not be resolved. Therefore, the model may not be fit to analyze flow front instabilities.

The problems regarding contact lines are solved by a relaxation of the no-slip boundary condition. The 'air' is given a tangential slip velocity at the walls determined from an equivalent stationary lubrication layer problem. This method is seen to be a good alternative. Near the flow front, this method may lead to small errors as shown through experimental validation. Again, it must be concluded that the model is not fit for flow front analyses.

In the application of a viscosity jump, one has to be prepared for turbulence as high Reynolds' numbers occur in the 'air'. Containing a constant pressure field in the region with 'air', might be the solution to this problem. However, this region is not of much interest.

Since no remeshing has taken place and no front models are used, the developed numerical model is capable of computing velocity fields etcetera on the whole fluid domain for complex geometries like bifurcations and flows in which gas injection plays a role.

Three-dimensional modelling is very time- and memory consuming, since the present model requires fine grids. Three-dimesional computations can best be used to gain good qualitative information of the flow phenomena of mold filling processes. Experimental validation of the three-dimensional numerical model is essential.

Modelling surface tension effects seems extremely difficult on fixed grids. Major problems occurred as second order differentiation requires smoother basis functions than available in the present SEPRAN-package. Extra diffusion steps to smooth the interface gave no resolution. Local remeshing or mesh adaptation strategies may be essential.
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