Numerical simulation of a viscous flow in a time dependent domain

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Abstract

The injection molding process is a flexible production method to fabricate plastic parts in series, characterized by the ability to realize complex shaped, highly integrated products in small cycle times. To enlarge the flexibility of the process, a multicomponent injection molding technique is being developed. Multiple extruders are connected to a hydraulic valve that controls the flow of the components into the mold. If more than two components have to be injected in combination with complex product geometries, this distribution device is incapable of switching fast enough between the different components to realize the desired product geometry. Therefore, the possibility is investigated of injecting the polymer with the aid of a cylindrical buffer. Then, the use of numerical tools is necessary to predict the configuration to be injected.

In this study, a simulation method has been set up and evaluated to simulate the injection with a piston/cylinder system. The time dependent piston/polymer interface is modeled, using a fixed mesh, in such a way that the correct velocity at the interface is obtained. This was done by applying a stick/slip-condition at the wall and representing the piston material by an artificial high viscosity.

Numerical tests showed that a viscosity difference of three decades between piston and polymer is sufficient to impose the piston velocity at the interface. Several ways of modeling the discontinuity in the velocity boundary conditions and in the viscosity-field at the interface have been tested. To prescribe the interface conditions, the location of the interface has to be determined. The possibility to follow the piston interface by convection of labels (particle identities) has been examined. The strategy proved not to be successful as a consequence of the distortion of the interface by numerical diffusion. In the label-field steep gradients did develop and convection of such a field suffers from numerical diffusion or dispersion. To reduce the influence of the diffusion, a correction of the label-field has been applied. The method is used to simulate the piston driven flow of polymer melts, modeled as generalized Newtonian fluids, through a contraction. Simulation results proved to be in good agreement with experimental results: all characteristics in the deformation patterns were predicted well. Disadvantage of the method appeared to be the use of a fixed mesh. Local mesh refinements can not be applied whereas they are necessary to describe the fountain flow phenomena at the moving interfaces more accurately.

Finally, to demonstrate its capabilities, the method has been used to simulate a non-isothermal reactive capillary flow.
Chapter 1

Introduction

The numerical simulations and experiments that come up for discussion in this report have been carried out within the framework of the development of a multicomponent injection molding technique. With the aid of numerical simulations some drawbacks of this technique can be overcome, making the multicomponent injection molding process more flexible.

1.1 Multicomponent injection molding

The injection molding process is a flexible production method for the manufacturing of plastic parts in series. The flexibility of the process is the production of complex shaped, integrated products with repetitive accuracy in small cycle times. Although a wide range of processes is available for producing plastic products, about 30 percent of all polymers is processed by an injection molding technique, indicating the importance of the process.

In figure 1.1 a schematic representation of an injection molding machine is given. An injection molding machine consists mainly out of two parts: an injection unit (containing the extruder) and a clamping unit (containing the mold). Granulated polymer is supplied by a hopper to the feeding part of the rotating extruder screw. The rotating of the extruder screw causes the granulate to move through the heated extruder barrel. Heat, generated by the heating elements and by the screw rotation, causes the granulate to plasticize. The constant

![Injection molding machine](image)

Figure 1.1: Injection molding machine
transport of polymer melt to the front of the screw (shot chamber) generates a pressure that moves the screw backwards (the nozzle is sealed by solidified polymer and the melt can not flow backwards due to a non-return valve). As soon as the screw reaches a preset limit, the shot size is met and the screw stops rotating. Next, the screw acts as a ram and pushes the polymer melt into the mold. After filling of the mold-cavity the pressure is maintained. In this way extra material can flow into the mold, in order to compensate for shrinkage. At the moment the gate is sealed, compensation for shrinkage is no longer possible. The pressure can be taken off. When the temperature has decreased below the ejection temperature, the mold is opened and the product can be ejected. In figure 1.2 an example is given of a realistic molding cycle. The whole cycle can be divided into three stages: the injection stage, the cooling stage and the ejection stage. At the moment the gate is sealed, the cooling stage starts, followed by the ejection stage.

The high flexibility of the injection molding technique can be extended with the possibility of combining different polymers within one product, each with its own specific properties. Therefore, a multicomponent (or multilayer) injection molding (MCIM) technique is being developed. With a MCIM-technique a layered structure of two or more components can be realized within a thin-walled product. Characteristic of the process is that the product is realized in one shot, pointing out the difference with a two- or multi-shot technique, by which the product is created in two or more shots. With respect to applications, there can be thought of a wide range, depending on the properties of the materials that are used. For example, realizing conductive layers in a product for EMI-shielding (Electro Magnetic Interference) or impermeable layers (to gases or fluids) in barrier products. The geometry of the layers in the product depends mainly on the position of the gate, the geometry of the nozzle and the sequence of injection. In figure 1.3 examples are given of a sequential, a simultaneous and a combined method of injection.

The first developments of a multicomponent injection molding technology date back from 1967. ICI [12] developed a multicomponent process for the manufacturing of stiff, large, and light-weight products. These products consisted of a characteristic structure of three layers; two outer-layers of material A and an inner-layer of material B. Material B consisted of ma-
Figure 1.3: Two component injection molding: sequential, simultaneous and combined injection

Material A, containing a foaming agent. The three layers were realized with sequential injection. These sandwich-products had hard, flat and glossy outer-layers and a light-weight foamed inner-layer. High clamping-forces were not necessary, since the foam structure of inner-layer B compensated for shrinkage by expanding. Applications, however, were restricted to thick-walled products (> 4 mm.); thin-walled, stiff and light-weight products can be made more economically using ribs. Further developments did lead to the Battenfeld two-channel technique [10]. By making use of two injection units and a distribution unit, not only sequential but also simultaneous injection became possible. Thin-walled products can be produced with three (ec)centric layers consisting out of two components. Formerly, this could only be achieved with a multi-shot method.

However, although a MCIM-technique seems to be very promising, some important limitations of the technique have to be overcome in order to make it competitive with existing processes. One important limitation is that only combinations of two materials are allowed. Being restricted to combinations of two components only, does not give the MCIM-process much of advantage with respect to the existing techniques. Interesting combinations of materials can not be used: apolar and polar polymers are not compatible, a product would fall apart. In this situation a third polymer is necessary, which acts as a glue, having both polar and apolar functional groups. Therefore, a three-component injection molding method is being developed at the Centre for Polymer and Composites of the Eindhoven University of Technology. Using a three component technique, polymer melt is supplied by three injection units. These units end up in one distribution device, which controls the flow of the different components into the mold. Another limitation is that only simple geometries can be realized, whereas the flexibility of an injection molding process should be the easy way of producing complex geometries. This drawback can be overcome by numerical simulations of the filling of the mold-cavity. With numerical simulations not only the flow of polymer into the mold can be calculated, but also the reverse situation, i.e. which component at what time had to be injected in order to realize the desired product. In this way, even for complex geometries, the injection sequence of the different components can be predicted.
1.2 Piston driven flow

During the development of a three component injection molding technique, another drawback appeared. Three injection units end up in one distribution device. This device is a hydraulic valve and it controls the flow of the different components into the mold by switching between the three units. The filling of the mold, however, has to be done within a very short period (about 5 seconds, see figure 1.2), otherwise solidification of the melt occurs before the mold has been filled completely. It is here where the problem arises: within the short period of mold-filling the hydraulic valve can not switch fast enough between the different components to create complex geometries in the mold. The valve's rate of switching is too slow with respect to the requested short period of mold-filling in case of complex product geometries. Injecting in this way, applications are restricted to simple geometries. To overcome this drawback, more time should be available to create the layers. Therefore, a method of injection is being developed where a buffer is used to store the melt that has to be injected. In this way the correct configuration can be created inside the buffer during the cooling stage of the previous product. Then, more time is available to create the layers (figure 1.2). As soon as the previous product has been ejected, the melt in the buffer can be injected into the mold, within the requested filling-time. As basic design for the buffer, a cylinder with a piston is used (figure 1.4). A study for the use of the cylinder/piston system as a buffer is given by Vos [23]. In this study it is investigated to what extent the original flow is being disturbed by the storage of the melt in the buffer. The fountain flow, that occurs when a piston is driving a fluid or a fluid is driving a piston, has been simulated by moving a polymer melt in between two pistons through a cylindrical cavity. By positioning tracers in the melt, deformation patterns were visualized. The use of a buffer in the injection-process makes numerical simulations of this piston driven flow necessary. Given the product geometry, the configuration to be injected has to be determined. With the aid of numerical simulations this configuration can be predicted.

1.3 Piston driven contraction flow

In his study, Vos simplified the buffer to a flow between two pistons in order to get a well-defined test problem. In this way, the influence of the fountain flow and the reverse fountain flow on the deformation of patterns was investigated. Injecting polymer melt into a mold with the buffer, however, implies a flow through a contraction. Therefore, experiments have been done with a piston driven flow through a contraction (see Oosterling [16]). These
experiments are analogous to the experiments done by Vos: again two pistons are used, but between the pistons a contraction is inserted. In this way, deformation patterns could be visualized of both filling and emptying a cylindrical buffer through a contraction. A short description of these experiments is given in appendix A. In order to set up a reliable method for predicting the injection configuration in MCIM, it must be possible to simulate these experiments accurately. The predictive quality can be checked by comparing the calculated deformation patterns and the experimental obtained patterns. Essential difference between the simulation of a flow between two pistons and a flow through a contraction between two pistons, is the time-dependency of the domain. The flow between two pistons can be simulated with a constant grid, modeling the flow as a driven cavity (Vos [23]). In case of a piston driven flow through a contraction, the idea of a driven cavity does not result in a time-independent domain. A possibility for modeling the time-dependent domain is the application of re-meshing techniques. The mesh definition is adapted to follow the deformation of the medium of interest and solutions are interpolated. Disadvantages of re-meshing techniques are application to 3D-simulations, extra cost and complicated programming. Therefore, a method without re-meshing could provide a flexible and economic way of simulating the contraction flow. Subject of the study, is to set up a flexible and economic method to simulate numerically the piston driven contraction flow on a fixed grid.

In chapter 2, a method is presented to simulate the contraction flow on a constant grid. Governing differential equations, modeling the isothermal flow of polymer melts (ABS, PS and PE) and the reactive flow (PU) are presented. In chapter 3, this method is tested and applied to the isothermal piston driven flow of thermoplastic melt through a contraction. Simulations with one material as well as simulations with combinations of materials have been done. The predictive quality of the simulation method is checked by comparing numerical results with experimental results. In chapter 4, the simulation method is applied to a non-isothermal, reactive flow. A capillary flow of a reactive polymer has been simulated. Finally, in chapter 5, conclusions and possibilities of the simulation method are discussed.
Chapter 2

Modeling the piston driven contraction flow

In this chapter a numerical model is presented to simulate a highly viscous, piston driven flow through a contraction. The method is characterized by an Eulerian formulation and a fixed computational grid. Equations, governing the simulations, are discussed.

2.1 Modeling a time-dependent domain

For a numerical simulation of a viscous flow with moving boundaries, several algorithms are available. These algorithms can be classified as mainly Eulerian, mainly Lagrangian or mixed Eulerian-Lagrangian. Most widely used in flow problems are Eulerian methods, characterized by a computational grid that is not related to the fluid. Advantage of these methods is that the fluid can be subjected to arbitrarily large deformations, without loss of accuracy. Disadvantage can be the difficulty to calculate accurately the position of a moving interface and to introduce correct boundary conditions at the interface.

A Lagrangian method is characterized by a computational grid that is related to the fluid. Therefore, a Lagrangian method is most suitable to define and to follow a material interface in time. Boundary conditions can be applied correctly at the interface in an easy way and nonlinear convective terms are absent. Disadvantage of a Lagrangian method is, that in case of large deformations of the fluid the irregular mesh (with fixed topology) can result in numerical inaccuracy. Due to severe distortions of the mesh, further calculations can even be obstructed.

A mixed Eulerian-Lagrangian method can be a good alternative to one of the previous methods. Advantages of both methods can be combined and disadvantages can be avoided. Although a mixed method can be very useful, additional cost or complicated programming can restrict its applications. A review of the different algorithms for viscous flows with moving boundaries is given in Floryan and Rasmussen [11].

The choice of a specific algorithm depends mainly on the flow problem of interest. If, for instance, position and curvature of a moving interface, together with an accurate surface tension is required, or when surface effects at a moving interface dominate flow phenomena, an appropriate method would be a Lagrangian-, a mixed- or an Eulerian-method with an adaptive grid. Otherwise an Eulerian-algorithm with a fixed grid could be used. Most of
the moving boundary problems (also referred to as free boundary problems) describe the behavior of an interface between two immiscible fluids. In these situations, the surface tension and correct boundary conditions at the interface can be important. In case of the piston driven flow, however, the time-dependent boundary is the interface between a steel piston and a polymer melt. Essential boundary condition at this interface is, that it has to move with the piston-velocity. Surface tension and other surface effects are of no importance to flow phenomena. If conditions of the contraction flow can be modeled in such a way, that the piston-velocity is obtained at the interface piston/melt, then it is possible to simulate numerically the piston driven flow with an Eulerian formulation and a fixed computational grid. Cost and complicated programming of re-meshing are avoided. Moreover, a rather straightforward implementation would result in a flexible simulation method, even in case of 3D-simulations. Therefore, a numerical method has been set up, based on an Eulerian approach and a constant finite element grid. Numerical simulations have been carried out with the finite element package SEPRAN [18].

2.2 Simulation method

To obtain useful results of a numerical simulation of a flow problem with a moving interface, a good understanding of the physical phenomena, that occur at the interface, is necessary. In case of two immiscible fluids (with constant surface tension), the interface conditions are identical normal and tangential velocity-components of both fluids at the interface and a jump in the normal stress-components over the interface, due to a surface tension, generated by the surface curvature (see Batchelor [2]). As mentioned in the previous section, the interface piston/melt is a special case of an interface between two immiscible fluids. This interface remains straight (i.e. if the piston is straight) and its position and velocity are known, whereas in most of fluid/fluid-interfaces the boundary is unknown and has to be determined as part of the solution algorithm. Moreover, surface tension can be neglected. The only important boundary condition having influence on flow phenomena, is the kinematic condition, that the interface moves with piston-velocity. Modeling the piston driven flow in an Eulerian way on a constant grid, this kinematic condition has to be realized. Two methods, that seem capable of realizing this kinematic condition, are a fictitious domain method (or imbedding method), as described in Glowinski et al. [13], and a method of pseudo-concentrations, as described in Thompson [21].

In case of the fictitious domain method, the actual computational domain is embedded in an extended, auxiliary domain. Incompressibility and the velocities at the boundaries of the original domain are taken into account as constraints of the balance of momentum. Essential part of the imbedding is, that there is no requirement for a strong coupling between the original domain and the new, extended domain. Therefore, this method could be very useful to simulate the piston driven flow in an Eulerian way on a fixed grid. The principle of imbedding, by which a domain can be made time-independent, applied to a piston driven flow, is visualized in figure 2.1. The time-dependent fluid domain in situation A is made time-independent in situation B by modeling both the fluid and the piston on one mesh.

With the method of pseudo-concentrations, values are assigned throughout the mesh in such a way, that they serve as material markers, i.e. they indicate the type of material that is present. Based on these pseudo-concentrations, the appropriate viscosity model and boundary conditions can be applied. The concentrations are being transported by convection
Modeling the piston driven contraction flow

Only, by which it is possible to use them as material markers. In Thompson [21], this method of pseudo-concentrations is used to calculate the deformation of transient creeping viscous fluids. At positions where material markers indicate presence of material, an appropriate value of the viscosity is used during the assembly of the stiffness matrix. On the other hand, if concentrations indicate absence of material, an artificial low value of the viscosity is assigned, as not to affect the velocity of the high-viscosity material.

The simulation method, set up for the piston driven contraction flow, is based mainly on the idea of material markers and the use of an artificial viscosity. Though a fictitious domain method seems to be a suitable method to simulate the piston driven flow with a constant grid, if the use of an artificial viscosity produces satisfactory accuracy of the velocity at the interface piston/melt, a more economic and flexible simulation method can be obtained. Implementation of a fictitious domain method requires additional programming in element subroutines, in order to take into account the constraint of the piston-velocity at the time-dependent interface. A method with material markers and artificial viscosity can be implemented in a rather straightforward way.

To be able to simulate the total deformation of the polymer melt, injecting it from one side of the contraction to the other, the mesh has to be made large enough to represent the total area that will be occupied by the melt during injection. On this mesh, both the driving and the driven piston, together with the melt have to be modeled. Therefore, throughout the mesh, each point is given a unique set of labels. These labels are transported by convection only and are used to distinguish between the piston material and the polymer melt. Where polymer melt is present, a realistic value for the viscosity is assigned. If labels indicate the...
presence of piston material, an artificial high value for the viscosity is assigned. Due to
this relatively high viscosity, the influence of melt-velocities is strongly reduced, realizing
the kinematic condition at the time-dependent piston interface. The remaining kinematic
boundary conditions are a no-slip condition at the melt/wall boundaries ($\vec{v} = \vec{0}$) and a slip
condition at the piston/wall boundary ($\vec{v} = \vec{V}_{\text{piston}}$). This principle of modeling the piston
driven contraction flow shown schematically in figure 2.2.

2.3 Governing differential equations

2.3.1 Equations of balance

Thermo-mechanical behavior of a fluid on macroscopic scale is described by continuum me-
chanics. Balance equations of mass, momentum, moment of momentum and energy, com-
pleted with the necessary constitutive equations, have to be solved. In a local, Eulerian
formulation, the equations of balance can be written as:

- balance of mass:
  \[ \rho \dot{\vec{v}} + \rho \vec{\nabla} \cdot \vec{v} = 0 \]  \hspace{1cm} (2.1)

- balance of momentum:
  \[ \dot{\rho} \dot{\vec{v}} = \rho \dot{\vec{f}} + \vec{\nabla} \cdot \sigma^e \]  \hspace{1cm} (2.2)

- balance of moment of momentum:
  \[ \sigma = \sigma^e \]  \hspace{1cm} (2.3)

- balance of energy:
  \[ \rho \dot{\epsilon} = \sigma : D - \vec{\nabla} \cdot \vec{q} + \rho r_e \]  \hspace{1cm} (2.4)

where $\rho$ denotes the density, $\vec{v}$ the velocity field, $\vec{f}$ the specific bodyforces, $\vec{\nabla}$ the gradient
operator, $\sigma$ the Cauchy stress tensor, $\epsilon$ the specific energy, $D$ the deformation rate tensor
($D = \frac{1}{2}(\vec{\nabla} \sigma + (\vec{\nabla} \sigma)^T)$), $\vec{q}$ the heat flux vector and $r_e$ a specific internal heat source. An
overdot on a variable indicates the material derivative:

\[ \dot{a} = \frac{Da}{Dt} = \frac{\partial a}{\partial t} + \vec{v} \cdot \vec{\nabla} a. \]  \hspace{1cm} (2.5)

Expressing the fluid behavior in terms of $\rho$, $\vec{v}$ and $T$ (independent variables), additional
constitutive equations for the Cauchy stress tensor, the internal energy, the heat flux vector
and the internal heat source are necessary, to relate the dependent variables to the independent
variables. Then, with appropriate initial and boundary conditions, the set of equations can be
solved.
2.3.2 Constitutive equations

Cauchy stress tensor $\sigma$

Material behavior of the polymer melt is described by a generalized Newtonian fluid model. Splitting the Cauchy stress tensor into a hydrostatic part ($-pI$) and a deviatoric part (extra stress tensor $\tau$):

$$\sigma = -pI + \tau,$$

(2.6)
a generalized Newtonian model is given by (see Sitters [19]):

$$\tau = 2\eta(p_0, T, I_1(D^d), I_2(D^d), I_3(D^d))D^d$$

(2.7)

where $\eta$ denotes the shear viscosity, dependent on the thermodynamic pressure $p_0$, temperature $T$ and the first, second and third invariant of $D^d$ (deviatoric part of $D$). The hydrostatic pressure $p$ consists out of two parts: $p = p_0 - \mu \text{tr}(D)$, with $\mu$ the bulk viscosity. The first invariant can be substituted by $\dot{\rho}$, using the balance of mass (eq. 2.1). In case of incompressible, isothermal flow, $\rho$ can be chosen as independent variable, instead of $p_0$. Otherwise, a constitutive relation $p = p(p_0, T)$ has to be given ($p, \nu, T$-diagram).

Simulating an isothermal flow of polymer melt, the melt is assumed to be incompressible. Then, shear viscosity is no longer dependent on $I_1(D^d)$. Dependency on the third invariant is neglected.

Experiments with the piston driven flow through a contraction have been done with polystyrene (PS), polyethylene (PE) and acrylonitrilebutadienestyrene (ABS) (see appendix A). A constitutive model for the shear viscosity of these materials can be obtained by fitting the parameters of a specific model on experimental data (see Zoetelief [24]). The viscosity models that are used in the simulations are given in appendix B. Note, that the use of a generalized Newtonian model implies that elastic effects are neglected. Although polymers melts are visco-elastic fluids, the use or a generalized Newtonian model can be justified if the Deborah-number $De < 1$. This number is defined as the ratio of a characteristic time of the fluid and a characteristic time of the flow. At the cavities of the contraction flow, the De-number is of order $O(1)$, which is the maximum value for which the use of a generalized Newtonian model can be justified. At the contraction, however, the De-number is of order $O(10)$, what can be a possible cause of differences between the numerical and experimental results.

Thermal properties

In order to solve the balance of energy (eq. 2.4), constitutive relations are required for the internal energy $\varepsilon$ and the heat flux vector $\overline{q}$. Frequently used constitutive relation for the internal energy $\varepsilon$, ignoring the elastic effects, is given by (see Bird [3] and Sitters [19]):

$$\dot{\varepsilon} = c_{p_0}\dot{T} + \frac{p_0}{\rho^2} \dot{\rho} + \frac{T}{\rho^2} \left( \frac{\partial p}{\partial T} \right) p_0 \dot{p}_0$$

(2.8)

where $c_{p_0}$ is the heat capacity at constant thermodynamic pressure. The constitutive relation for the heat flux vector often is given by Fourier's law:

$$\overline{q} = -\Lambda \cdot \nabla T$$

(2.9)

where $\Lambda$ is the conductivity tensor. In case of isotropic conduction, tensor $\Lambda$ reduces to $\lambda I$, with $\lambda$ a constant heat conduction coefficient.
The piston driven flow of thermo-plastic melt is simulated isothermally. Heating up and cooling down the experiments has not been simulated. Then, the energy equation does not have to be taken into account. Temperature dependency of the coefficients disappears. Influence of viscous dissipation \((\eta \dot{\gamma}^2)\) is then neglected. This can be justified by the low piston velocity \((O(10^{-3}))\). In case of reactive flow, discussed further on in this chapter, the energy equation has to be taken into account, since variables and parameters then are functions of the extent of reaction of the material, which in its turn is highly dependent on temperature. For reactive materials the internal heat source can not be neglected. It represents the reaction heat. The formulation of the heat source is given in section 2.3.4, where the governing equations of reactive flow are discussed.

### 2.3.3 Particle tracking

In order to realize the desired product geometry in the mold, knowledge of the time and duration of injection of the different components is required. Using a buffer, given the desired geometry, the initial configuration has to be determined. A possible method to calculate this configuration, is the reconstruction of flow paths of particles. Reconstruction is based on the fact, that a velocity vector \(\vec{V}(\vec{z}, t)\) is tangential to the flow paths. Given the initial position \(\vec{z}_0 = \vec{z}(\vec{z}_0, t_0)\) of a particle, reconstruction of its flow path is an initial value problem that can be solved adequately by a Runge-Kutta integration method (see Caspers [5]). Disadvantage of this method is, that at every time-step the velocity field has to be stored. This requires a large memory capacity.

Another method of particle tracking is the "conservation of identity", as described in Zoetelief [24]. Particles are given a unique set of labels \(\vec{i}(\vec{z}, t)\), representing the identity of particles, that are transported by convection only. Following critical contours of the labels, the deformation of layers due to the flow can be calculated. The conservation of identity can be calculated by setting the material derivative of the labels to zero:

\[
\dot{i} = \frac{D\xi}{Dt} = \frac{\partial \xi}{\partial t} + \vec{v} \cdot \nabla \xi = 0 \tag{2.10}
\]

where \(\xi\) denotes the labels. It shall be clear, that the labels, used for particle tracking, also can be used to distinguish between piston material and polymer melt, for labels and the so-called pseudo-concentrations are modeled identical. Therefore, they can serve as indicator in assigning values of the viscosity (see section 2.2).

### 2.3.4 Reactive flow

The numerical model for a simulation of a piston driven flow, can also be applied to thermosetting polymers. Then the flow is reactive. During heating and processing, the components of a thermoset react and the molecular weight increases. When the molecular weight approaches infinity the gel point is reached. Before gelation, the polymer is completely soluble; after gelation, both soluble and insoluble materials are present. Once the gel point has been reached, flow is no longer possible.

Numerical simulation of a reactive flow can be of predictive value for processes like IC-packaging, reaction injection molding (RIM) or compression molding. In Corbey [8], a finite element analysis of a 2D reactive flow is presented. A piston driven reactive flow is simulated between two moving pistons. Analogous to the simulations of Vos [23], this flow is simulated on a constant grid, modeling the problem as a driven cavity. In Spoelstra [20], a study is
presented of the flow of a highly filled epoxy compound. This study fits into the framework of the development of a multilayer encapsulation of IC’s.

A complete model of a reactive flow requires a mass or mole balance for every component together with a model of the reaction kinetics for every occurring reaction. However, due to lack of information on all reaction kinetics, often one parameter is used to describe the overall reaction kinetics: the conversion. Then, a mass balance of the system as a whole together with a mole balance for the reactant is sufficient. The mole balance of the reactant, that has to be solved in addition to the balance equations of mass, momentum and energy, is given by (see Corbey [8] and Peters et. al. [17]):

$$\dot{C} = \frac{\partial C}{\partial t} + \nabla \cdot \vec{v} C = D \nabla^2 C - R_c(T, C)$$ (2.11)

where $C$ is the molar concentration of the reactant, $D$ is the diffusion coefficient (isotropic diffusion is assumed and substituted into the molar flux) and $R_c$ is the rate of polymer production (rate of conversion), dependent on temperature and concentration of reactant. Often, the mole balance is written in terms of conversion $X$, defined as:

$$X = \frac{C_0 - C}{C_0}$$

with $C_0$ the initial concentration of reactant. Then, the mole balance reads:

$$\frac{\partial X}{\partial t} + \nabla \cdot \vec{v} X = -D \nabla^2 X + R_c(T, X)$$ (2.12)

As a constitutive relation for the reaction kinetics of PU (polyurethane), a model for autocatalytic curing, originally proposed by Kamal and Sourour [14], is used (see also Castro and Macosko [6] and Peters et. al. [17]) that is given by:

$$R_c = \frac{R_c}{C_0} = \left( K_1 + K_2 X^n \right) \left( 1 - X \right)^m$$ (2.13)

where $n$ and $m$ represent reaction orders and $K_1$ and $K_2$ are constants, representing temperature dependency. $K_1$ and $K_2$ are defined by:

$$K_i = k_ie^{\left( \frac{-E_i}{RT} \right)}$$ (2.14)

with $k_i$ pre-exponential factors, $E_i$ activation energies, $R$ the gas constant and $T$ the temperature. Values of the model parameters for PU are given in appendix B. Due to dependency on conversion, the balance of mass reads as:

$$\frac{\dot{\rho}}{\rho} = -\alpha \left( \frac{\partial \rho}{\partial T} \right)_{p_0,X} \dot{T} + \kappa \left( \frac{\partial \rho}{\partial p_0} \right)_{T,X} \frac{\dot{p}_0 + C_0 \left( \frac{\partial \rho}{\partial X} \right)_{p_0,T}}{\beta} \dot{X}$$ (2.15)

with $\alpha$ the volume expansion coefficient, $\kappa$ the isothermal compressibility factor and $\beta$ the reaction-shrinkage coefficient. The constitutive equation for the internal energy (eq. 2.8) has to be expanded with a term, reflecting dependency on the reaction rate, to (see Corbey [8]):

$$\dot{\epsilon} = \frac{c_{p_0} \dot{T} + \frac{p_0}{\rho^2} \dot{\rho} + \frac{T}{\rho^2} \left( \frac{\partial \rho}{\partial T} \right)_{p_0,X} \dot{p}_0 + h_r \dot{X}}{-\frac{1}{\alpha T}}$$ (2.16)
Chapter 2

with \( h_r \) the specific heat of reaction. Using this expression to eliminate \( e \) from the balance of energy (eq. 2.4), the energy balance for a reactive material can be written as:

\[
\rho c_p \dot{T} = -\nabla \cdot \dot{q} + T\alpha \dot{p}_0 + \gamma : D^d + \rho h_r R_x.
\]  

(2.17)

The internal heat source \( \rho h_r R_x \) represents the reaction. The balance of momentum does not change, and is given by eq. 2.2. Modeling the reactive flow with a generalized Newtonian fluid, a constitutive relation for \( \eta \) is required that depends on the conversion. This model is given in appendix B where also the necessary values of properties and parameters are given.

2.4 Discretized equations

Problem formulations can be simplified by neglecting the physical effects that have little relative importance to the situation that is modeled. For isothermal flow the balance of energy does not have to be taken into account and temperature dependency of parameters and variables disappears. Further on, the flow is assumed to be incompressible and due to the high viscosities of the polymer melt, inertial and gravity forces can be neglected with respect to the viscous forces (the Reynolds-number Re is of order \( O(10^{-5}) \)). Then, including particle tracking, the describing differential equations are given by:

\[
\frac{\partial \varphi}{\partial t} + \nabla p_0 - 2 \nabla \cdot \eta D^d = 0
\]  

(2.18)

\[
\nabla \cdot \vec{v} = 0
\]  

(2.19)

\[
\dot{X} = R_x
\]  

(2.20)

The reactive flow is also assumed to be incompressible: density changes due to reaction are neglected (see Castro and Macosko [6]). The viscosity of PU, at low degrees of conversion, is of order \( O(10^{-1}) \). Resulting Reynolds numbers are of order \( O(10^{-1}) \) and therefore, inertial forces can be neglected. Influence of gravity is neglected. With respect to the balance of energy, thermal properties are assumed to be constant and temperature changes due to pressure changes are neglected. In the mole balance, molecular diffusion can be neglected (isotropic diffusion coefficient \( D \approx O(10^{-7} \text{cm}^2/\text{s}) \)). In simulations of reactive flow particle tracking has not been used. The resulting differential equations, modeling the reactive flow, are given by:

\[
\frac{\partial \varphi}{\partial t} + \nabla p_0 - 2 \nabla \cdot \eta D^d = 0
\]  

(2.21)

\[
\nabla \cdot \vec{v} = 0
\]  

(2.22)

\[
\rho c_p \dot{T} - \nabla \cdot (\lambda \nabla T - \eta \dot{\gamma}^2) - \rho h_r R_x = 0
\]  

(2.23)

\[
\dot{X} = R_x
\]  

(2.24)

These nonlinear, coupled differential equations are solved with a Galerkin finite element method. The momentum equations are solved with a Galerkin-Bubnov method, using modified Crouzeix Raviart elements (extended quadratic elements with 6 velocity nodes and 1 pressure node). Balance of mass and momentum are uncoupled, by application of a penalty function method (see Cuvelier et al [9]). The mole balance and the conservation of identity
are hyperbolic first order partial differential equations: the diffusive part is absent. If this kind of equations is solved with a Galerkin-Bubnov method, solutions become oscillatory (see Baaijens [1]). In order to suppress this behavior, temperature, conversion and labels are approximated with a Streamline Upwind Petrov Galerkin (SUPG) method (see Brooks and Hughes [4]). Quadratic triangles are used. Time derivatives are approximated with an Euler implicit scheme, defined as:

$$\frac{\partial \varphi}{\partial t} \approx \varphi(z, t^{n+1}) - \varphi(z, t^n)$$

and nonlinear terms are linearized, using a Picard iteration scheme (successive substitution). Applying the Galerkin approximation as discussed, the discretized equations for the isothermal flow are given by:

$$M_p \frac{\xi^{n+1} - \xi^n}{\Delta t} + S(\xi^n, \xi^n)\xi^{n+1} + \frac{1}{\epsilon} L_p M_p^{-1} L_p^{n+1} = F_v(\xi^n)$$

$$c M_p \frac{\xi^{n+1} - \xi^n}{\Delta t} = -L \varepsilon^{n+1}$$

$$M_\xi \frac{\xi^{n+1} - \xi^n}{\Delta t} + N_\xi(\xi^n)\xi^{n+1} = 0$$

with mass matrices, $S$ the stiffness matrix, $N_\xi$ represents the convective terms of the label transport and $F_v$ is the right-hand-side of the momentum equation. The indices $n$ and $n + 1$ indicate the time-level of a variable: $t^n$ or $t^{n+1}$; $\Delta t$ is the incremental time-step. Note that, if viscosity is assigned as function of labels, the stiffness matrix becomes dependent on labels. The term $\frac{1}{\epsilon} L_p M_p^{-1} L_p^{n+1}$ represents the pressure gradients. Using the penalty function method, $p$ is eliminated from the momentum balance. The velocity is considered as degree of freedom and the pressure is computed as a derived quantity (see SEPRAN [18]). The number $\epsilon$ is the penalty parameter. It must be chosen such, that $\epsilon p = O(10^{-6})$. The discretized equations, in case of reactive flow, are given by:

$$M_p \frac{v^{n+1} - v^n}{\Delta t} + S(T^n, X^n) v^{n+1} + \frac{1}{\epsilon} L_p M_p^{-1} L_p^{n+1} = F_v(v^n)$$

$$c M_p \frac{v^{n+1} - v^n}{\Delta t} = -L \varepsilon^{n+1}$$

$$M_T \frac{T^{n+1} - T^n}{\Delta t} + K T^{n+1} + N_T(v^n) T^{n+1} = F_T(T^n, X^n, v^n)$$

$$M_X \frac{X^{n+1} - X^n}{\Delta t} + N_X(v^n) X^{n+1} = F_X(X^n, T^n)$$

with $K$ the conduction matrix. Again, $M_i$ represent the mass matrices, $N_i$ represent the convective parts and $F_i$ are the right-handsides. Modeling a piston driven flow, appropriate constant values are assigned for the thermal properties of steel and polymer. Therefore, the temperature mass matrix and the conduction matrix are not constant, but dependent on the position of the piston.
Chapter 3

Simulation results of isothermal thermoplastic flow

In this chapter the simulation method, discussed in chapter 2, is applied to an isothermal thermoplastic flow. Numerical and experimental results are compared.

3.1 Interface conditions

Principal idea of the simulation method is to prescribe a stick/slip-condition at the wall of the cylinder and to model the piston material by means of an artificial high viscosity in order to impose the piston velocity at the piston/polymer interface (see figure 2.2). This implies that functions with a discontinuity at the piston/polymer interface have to be modeled on the constant mesh. To achieve this, boundary conditions and material properties must be prescribed such, that the step functions are approximated by the resulting combination of element shape functions. Two methods of imposing boundary conditions of the velocity are tested in combination with three methods of prescribing the viscosity.

In the SEPRAN FEM-package, boundary conditions can only be prescribed at predefined boundaries of the mesh. In order to prescribe the stick/slip-condition at the wall boundary, elements along this boundary are considered as quadratic line elements. In figure 3.1 the

The quadratic shape functions \( N_i(\xi) \) as function of local coordinate \( \xi \) are given by:

\[
N_1(\xi) = \frac{\xi}{2}(\xi - 1) \\
N_2(\xi) = (1 - \xi^2) \\
N_3(\xi) = \frac{\xi}{2}(\xi + 1)
\]

Figure 3.1: Shape functions on 1D quadratic element
shape functions of such elements are shown. The two methods of prescribing the velocity at the wall boundary are:

- **velocity prescription A:** If the piston/polymer interface is positioned between the first and the second node of an element, then, at node 1 the piston velocity $V_p$ is prescribed and at node 3 the velocity is set to zero. At node 2, the velocity is prescribed in such a way that at the position of the interface the velocity equals $V_p$ (see figure 3.2). This value can be calculated with aid of the shape functions. At remaining upstream nodes of the wall boundary, the piston velocity is prescribed (slip condition of the piston) and at downstream nodes, where the polymer sticks to the wall, the velocity is set to zero. Once the piston has passed the midpoint of an element, at all three nodes the piston velocity is prescribed. This implies that at the first node of the next element downstream also the piston velocity is prescribed. Since at remaining downstream nodes the velocity is set to zero, backflow occurs, due to the definition of the shape functions. Therefore, when the third node of an element gets a non-zero velocity, the velocity at the midpoint of the next element is chosen such that the interpolation polynomial becomes positive everywhere (see figure 3.3; bold vertical lines represent the interface). This velocity value is determined by the shape functions.

![Figure 3.2: Velocity as function of interface position](image1)

![Figure 3.3: Velocity prescription, avoiding backflow](image2)

- **velocity prescription B:** If the piston/polymer interface is positioned between the first and the second node of an element, node 1 and 2 get the piston velocity and at node 3 the velocity is set to zero. This results in the constant boundary function of the velocity as shown in figure 3.4. If the interface has passed node 2, the same method of prescription is applied as in case of method A.

![Figure 3.4: Constant velocity function](image3)

![Figure 3.5: Methods of viscosity prescription](image4)

Also in case of prescribing the viscosity, values must be assigned such that the resulting interpolation polynomial approximates the jump at the interface and that negative values...
Isothermal thermoplastic flow

are avoided (physically impossible). In order to avoid negative values, the viscosity values at the midpoints are calculated by linear interpolation of the values at the vertices. This appeared to be necessary: due to high viscosity gradients in the polymer, negative values of the interpolation polynomial can be created. Yet, three different ways of prescribing viscosity are tested in order to investigate the influence on the velocity at the interface and to verify whether the use of a linear viscosity definition is justified. The three ways of prescribing viscosity across the interface are visualized in figure 3.5.

- **viscosity prescription I**: at node 1 the high piston viscosity is prescribed and at node 3 the viscosity of the polymer. At node 2, a linear interpolation of the values at node 1 and node 3 is assigned (line I in figure 3.5).

- **viscosity prescription II**: again, at node 1 the high piston viscosity is prescribed and at node 3 the polymer viscosity. At node 2, a value is assigned in such a way that the interpolation polynomial at all positions on the element becomes positive (line II in figure 3.5).

- **viscosity prescription III**: at node 1 the piston viscosity is assigned and at node 3 the polymer viscosity. Now, at node 2 the value is chosen such that the interpolation polynomial does not exceed the piston viscosity (line III in figure 3.5).

Note that the viscosity definitions are constant during the time the interface is passing the element. Obviously, at the remaining upstream nodes the piston viscosity is assigned and at downstream nodes the appropriate polymer viscosity.

Before the several combinations of prescribing velocity and viscosity are tested, the magnitude of the viscosity jump is determined that results in a good approximation of the piston velocity at the interface. Therefore, a piston driven Newtonian flow over a 1:3-contraction has been simulated (see figure 3.6). Influence of the magnitude of the jump was investigated by plotting the velocity at the interface for various values of the jump, when the interface is positioned between node 2 and 3 at the boundary (see figure 3.7). Reason for choosing this position is, that here both methods of prescribing the velocity are identical and the different ways of prescribing viscosity appeared to give the same results for the velocity solution.

The viscosity was prescribed by method I. Piston velocity was 1 mm/sec (constant) and the interface velocity has been calculated for viscosity jumps of 0, 10, 10² and 10⁵. Results are shown in figure 3.8. Values of the viscosity jump, corresponding to the 4 velocity profiles are

![Figure 3.6: Piston driven Newtonian flow over 1:3 contraction](image)

![Figure 3.7: Moving interface on mesh](image)
Figure 3.8: Interface velocity as function of
the jump in the viscosity

given by:

1 → Δη = 0
2 → Δη = 10
3 → Δη = 10^2
4 → Δη = 10^3

Interpretation of the profiles must be done carefully. Values, calculated with the postprocessing part of SEPRAN are linearly interpolated between the nodal values. Considering the deviation from the exact piston velocity \( \dot{V}_p \) (straight line), it can be concluded that a viscosity jump of \( 10^3 \) is sufficient to impose the piston velocity at the interface. This conclusion has been verified by the calculation of the velocities with the quadratic interpolation polynomials on the centerline (see figure 3.7). The resulting interface velocities as function of the viscosity jump are shown in figure 3.9. Indeed, these results indicate that modeling the piston material with a viscosity value three orders higher than the melt viscosity results in a good approximation of the piston velocity at the interface.

Testing of the two ways of prescribing the velocity boundary conditions, in combination with the three ways of prescribing the viscosity, has been done by investigating the resulting interface velocities for an interface position between node 1 and 2 on the boundary of element at the wall. Here, the methods of the velocity prescription are different. Results of prescribing the velocity by method A (velocity at node 2 is determined by the interface position) in combination with prescribing viscosity by method I, II and III (figure 3.5) are shown in figure 3.10. Results of prescribing the velocity by method B (at node 2 a constant value \( \dot{V}_p \) is prescribed) in combination with the different ways of the viscosity prescription are shown in figure 3.11. Also in figure 3.10 and 3.11, values must be interpreted carefully, due to the linear interpolation of nodal values. Effect of this interpolation is demonstrated by the
interface velocity at the wall boundary: in case of method A, the velocity should be exactly \( \bar{V}_p \) (prescribed as such) and in case of method B, the velocity should be higher than \( V_p \). These values are in contrast with the values that are shown in figure 3.10 and 3.11 respectively. For both methods of prescribing the velocity, the effect of the viscosity prescription shows an identical behavior: the lower the viscosity at node 2 (see figure 3.5), the better the piston velocity at the interface is approximated. This is also what could be expected. With viscosity prescription II, relatively more emphasis is given to velocity gradients inside the polymer region. With respect to the way of velocity prescription, results appear to be much better in case of method B. In order to verify these conclusions, given the calculated nodal velocity values, the quadratic velocity interpolation has been used to calculate the interface velocity on the centerline. Results for the velocity prescription according to method A are shown.

Figure 3.10: Interface velocities: velocity prescribed by method A

Figure 3.11: Interface velocities: velocity prescribed by method B

Figure 3.12: Velocity on centerline: velocity prescribed by method A

Figure 3.13: Velocity on centerline: velocity prescribed by method B
in figure 3.12 and for method B in figure 3.13. In figure 3.12, the position of the interface is indicated by a dashed line. In case of method B this position is not relevant: then, the prescribed velocity at the boundary is not a function of the relative position of the interface in an element. The quadratic velocity at the centerline shows the same trends for the different combinations of velocity prescriptions and viscosity prescription as can be concluded from the interface velocities in figures 3.10 and 3.11.

From the tested possibilities, it can be concluded, that the best approximation of the piston velocity at the interface is obtained by prescribing the velocity by method B and the viscosity by method II. However, the results with a linear approximation of the viscosity jump within an element are about the same accuracy, by which the use of this strategy is also justified (in combination with prescribing the velocity according method B).

3.2 Labels as material markers

For a numerical simulation of a piston driven flow, modeled by a stick/slip-condition at the boundary and a viscosity jump at the piston/polymer interface, the position of the interface has to be determined in order to prescribe conditions at the right nodes. The most general method to determine this position is to use labels (ξ) as material markers. In case of a piston driven flow there is another, less general, possibility: if the piston velocity $V_p$ is known in time, then the exact position of the interface $Z_p(t)$ can be calculated. In order to investigate the applicability flow of both possibilities, a Newtonian flow between two moving pistons has been simulated in two ways:

- **case 1**: prescribing conditions as function of labels: $\vec{v} = f(\xi)$ and $\eta = f(\xi)$;
- **case 2**: prescribing conditions as function of the exact interface position: $\vec{v} = f(Z_p(t))$ and $\eta = f(Z_p(t))$.

The flow is shown schematically in figure 3.14. Labels were defined as the initial $z$-coordinate of the particles. Labels entering the mesh (inflow of piston material) were given the value zero. Inside the fluid, 4 tracers are used to visualize the deformation patterns, caused by the flow. The two axi-symmetric problems were simulated with a uniform rectangular mesh, consisting out of 480 quadratic triangles (see figure 3.15). A constant piston velocity $V_p = 5$ mm/sec was prescribed during a time-lapse of 7 seconds. The piston material was modeled by a viscosity of $10^6$ Pa·s and the fluid by a value of $10^3$ Pa·s. The incremental time step was 0.05 sec. Prescribing the conditions as function of the labels, the velocity becomes dependent on the labels and a Picard iteration-scheme has been applied to calculate solutions at the new time level. Simulation results of case 1 are shown in figure 3.16 and results of case 2 in figure 3.17.

Examining the simulation results of both cases, it can be observed that the piston/polymer interfaces are distorted whereas they should remain straight. Furthermore, although both the interfaces were modeled in the same way, the upper interfaces are less distorted compared to the lower interfaces. At both simulation results, fountain flow and reverse fountain flow phenomena can be recognized. To obtain more information about the deformation of the interfaces, at several time steps the label values of case 2 at the wall boundary (figure 3.18) and at the centerline (figure 3.19) have been plotted. In these figures, the upper horizontal line is the label value of the upper piston, the lower horizontal line is the label value of the lower piston and the straight vertical lines represent the exact interface positions at the different times. In figure 3.18, the reason of the deformation of the lower piston interface is
clearly demonstrated. Due to the stick/slip-condition at the wall, polymer-labels stick to the wall and piston-labels are transported. With that, a growing jump is created in the label values. As time proceeds, this discontinuity increases. The theoretical transport of labels at the wall is visualized in figure 3.20. Numerically, the convection of a solution, containing a discontinuity, suffers from diffusion (or, even worse, dispersion). Consequence of this diffusion is, that during the progress of the flow the positions of the label values that indicate the lower piston interface, lag behind with respect to the exact positions. This is illustrated by the simulation results.

The phenomenon that the upper interface is less distorted than the lower interface can be explained by the essential difference between both pistons: the lower piston is driving the fluid whereas the upper piston is driven by the fluid. Therefore, at the intersection point upper interface/centerline the gradients in the label-field are less steep compared to the gradients at the intersection point lower interface/wall. Consequently, the upper interface is less distorted by numerical diffusion. The reason why the gradients at the upper interface are less steep is that here the gradients are determined by the label-values that are transported by the flow, while at the lower interface the gradients are determined by the values at the wall (no-slip condition). This implies that the gradient at the upper interface is dependent on the accuracy of the calculated velocity-field. Here a disadvantage of the use of a constant mesh appears: fountain flow phenomena occur at a certain distance of the interfaces, due to the coarseness of the mesh. Local mesh-refinements can not be applied whereas they are necessary to describe the flow at the interfaces more accurately. As a consequence of the inaccuracy in the calculated fountain flow the gradients at the upper interface are less steep. Note that, in contrast to the lower interface, the upper interface overtakes its exact position (see figure 3.19).
Figure 3.16: Simulation results of case 1

Figure 3.17: Simulation results of case 2
Figure 3.18: *Labels during transport at wall boundary at t=0, 1.75, 3.5, 5.25 and 7.0 sec.*

Figure 3.19: *Labels during transport at centerline at t=0, 1.75, 3.5, 5.25 and 7.0 sec.*

Figure 3.20: *Theoretical transport of labels at wall*
Prescribing conditions on basis of the exact interface position \( \xi_p(t) \) gives better results. If the conditions are prescribed on basis of the label values, which suffer also from diffusion and this is the reason why the lower interface in case 1 has been deformed more than in case 2. Besides diffusion, at convection of a solution containing a discontinuity, dispersion can occur. In that case, the label values at the discontinuity can oscillate. Then, the label value that indicates the interface is no longer unique. Therefore, the simulations of a piston driven flow have been carried out with prescribing the conditions on basis of the exact interface positions (known from experiments, or prescribed).

Occurance of diffusion is highly dependent on the definition of labels. To avoid diffusion, labels have to be defined by smooth functions: steep gradients have to be avoided. Initial smooth functions are relative easy to prescribe. However, as the flow progresses, the label-field is deformed, by which the steep gradients in the solution field are introduced. Advantage of the use of labels which have no physical meaning is, that they can be smoothed. Smoothing requires that contourlines are kept in place. Thompson [21] applied in numerical simulations of creeping viscous flows, a smoothing algorithm, based on the distance of nodes to the contourlines. In case of the piston driven flow, fountain flow phenomena bring about such complicated deformation patterns, that it is not straightforward to define an appropriate smoothing algorithm.

In order to verify whether the cause of the distortion of the piston interfaces is mainly the numerical diffusion of labels values of the simulation of case 2 has been repeated with a correction on the velocity-field. At nodes, situated in the region of the lower piston, the velocity was set to the piston velocity \( \vec{V}_p \), before the transport of labels took place. Results of this simulation proved to be identical to the simulation results of case 2, shown in figure 3.17. Therefore, it can be concluded, that the main cause of distortion of the interfaces is due to the numerical diffusion of label values and not as a consequence of an inaccurate velocity-field. In order to reduce the influence of diffusion, a correction of the label-field has been tested. In fact, the label-field at the piston region is known, since particles at this region are only
translated by the piston velocity. This information has been used to correct the label-field. Case 2 has been simulated again, but now the label solutions were corrected at the nodes inside the piston region by substitution of the exact values of the translated labels. This has been done each incremental time step and only at the region of the driving piston. Results were improved a lot by the correction (figure 3.21). A disadvantage of the applied correction method may be that substitution of values at nodes, without paying attention to values at neighboring nodes, can introduce oscillations. Therefore, a correction by a redefinition of the whole label-field may be numerically more stable and accurate.

### 3.3 Isothermal contraction flow

In this section, the results are presented of the numerical simulations of the experiments with the piston driven contraction flow (see appendix A). Experiments with one material as well as some of the experiments with a combination of two materials have been simulated. With all simulations of the contraction flow, the velocity boundary conditions were prescribed according method B and the viscosity was prescribed according method I (see section 3.1). The piston material was given an viscosity value $10^6 \text{ Pa}\cdot\text{s}$. The exact piston positions $Z_P$ were used to locate the positions of the interfaces. Also in case of the piston driven contraction flow a correction of the labels has been applied in order to reduce the influence of the numerical diffusion at the interfaces. Simulations without correction have been done to show the effect of correction to this flow. The correction is slightly different compared to the correction that has been applied in the previous section. As a consequence of the presence of the contraction and the no-slip condition at the wall boundary of the lower cavity, the exact label solution at this boundary is known. Therefore, label values were not only corrected at both piston regions but also at the wall boundary of the cavity from where the polymer is injected. The contraction flow has been simulated isothermally at 393 K during a time lapse of 11.9 seconds. The piston velocity $V_p$ was 5 mm/sec. The cavity diameter is 16 mm and the contraction diameter is 5 mm. The length of the contraction is 20 mm. Label values were defined by the initial z-coordinate. The incremental time step was 0.02 seconds and the mesh that has been used is shown in figure 3.22. The viscosity of the materials is dependent on the velocity field (shear thinning behavior). Therefore, a Picard iteration scheme has been applied to calculate solutions at the new time level. In the iteration loop, solutions at the old time level were used as initial estimation of the new solutions.

![Mesh used in simulations of the contraction flow](image-url)
3.3.1 Simulations with one material

First, the contraction flow of LDPE has been simulated without any corrections. The results of this simulation are shown together with the experimental results in figure 3.23. Hatched areas in this figure represent the position of the contraction.

Comparing the experimental and numerical results, it can be concluded that the results are qualitatively in good agreement. All characteristic deformations of the patterns are described, as well as at the upper cavity as at the lower cavity. The fountain flow at the driven piston and the reverse fountain flow at the driving piston can be observed. Also the deformation of patterns at the in- and outflow of the contraction are described well.

In spite of the good resemblance of the results, three essential points of difference can be observed. First, the length of the fluid region differs between the experimental and numerical configurations. The cause is the shrinkage of material. Experimental configurations are shown at room temperature (solidified), whereas the numerical configurations are shown at 393 K (melt). The shrinkage, caused by the cooling down of the melt, did result not only in the observed length differences but also in an unknown deformation of the tracer patterns. This deformation can be one of the causes of quantitative differences between the numerical and experimental results. The second important difference is, that, in contrast with the experimental results, in the numerical results fountain flow occurs at a certain distance from the interfaces. The cause of this phenomenon is the coarseness of the mesh. To simulate the fountain flow at the interfaces more accurate, a local mesh-refinement at the interfaces is required. Using a constant mesh in combination with the time dependency of the interfaces, such a mesh-refinement is not possible. This phenomenon was also observed with the simulations of the Newtonian flow between two moving pistons and is the reason why the upper interface becomes less distorted by diffusion than the lower interface (see section 3.2). The third phenomenon that can be observed is the distortion of the interface by numerical diffusion. Also this effect was present at the simulations of the Newtonian flow between two pistons. To reduce the influence of diffusion, a correction of the label-field has been applied. Results of the LDPE-simulation with correction of label values are shown in figure 3.24. Clearly, the results are improved by the correction. Since substitution of values is only justified at the region of the pistons, at nodes inside the melt region the diffusion is still present. Consequently, the solution is not corrected within a nodal distance a the interfaces. A more effective way of reducing the effects of diffusion may be a re-definition of the total label-field or the use of a more accurate numerical scheme to handle the convection (a Time Discontinuous Galerkin Least Squares method or a spectral finite element method method, for instance).

Simulations with ABS and PS have been done analogous to the LDPE-simulations with correction of the label-field. Results of the simulation with PS are shown in figure 3.25 and the results with ABS are shown in figure 3.26. Note that at all simulations the same initial configuration has been used, although the materials had different initial configurations at T = 393 K. However, these exact configurations were unknown due to the expansion during the heating of the polymer. Therefore, the initial numerical configuration that is shown at the results has been used as estimation of the initial experimental configuration. Advantage of using the same initial configuration is that the differences between the viscosity models can be observed better. The overall material behavior at the simulations is in good agreement with the experimental results. Numerical simulations show the smallest displacement of material at the center of the upper cavity for PE. This is also observed at the experiments.
Further on, the Cross model appears to describe the behavior of PS rather well. In case of ABS, the modification of the Cross model (see appendix B) can be recognized in the somewhat more flattened contourlines at the lower cavity. At the upper cavity, however, the deformation patterns are almost equal to the patterns of PS, whereas experiments show differences. The differences can be explained by the different scales of shrinkage and by an insufficient way of modeling the Bingham-like behavior of ABS by the modification of the Cross model. Moreover, the fluid has been modeled viscous whereas it is visco-elastic. Again, the same remarks can be made with respect to diffusion and the necessity of mesh-refinement at the interfaces.

3.3.2 Simulations with material combinations

Requirement of a simulation method with a predictive quality for a simulation of a multicomponent injection with a buffer, is that it has to be possible to simulate the flow of multiple components. Therefore, several experiments have been done with material combinations of LDPE, PS and ABS (see appendix A). The flow of ABS/PS combinations and of PE/PS combinations has been simulated. The simulations have been done analogous to the simulations with one component. However, an extension had to be made. The position of piston interfaces still can be prescribed by their theoretical position \( (z_p(t)) \). The position of the interface between the components, on the contrary, can not be determined in this way. In order to trace this position, labels have been used as material markers. Since at this interface there are no steep gradients in the labels, convection of this interface does not suffer from diffusion or dispersion. The use of labels as material markers makes the velocity-field label dependent and therefore the convection of labels had to be included in the iteration loop (Picard-scheme). The initial configurations were adjusted to the initial configuration of the simulated experiment in question. The results of the ABS/PS simulations are presented in the figures 3.27 and 3.28 and the results of the PE/PS simulations are presented in the figures 3.29 and 3.30.

Numerical and experimental results of the PS/PE combinations are in good agreement. As well in the simulations as in the experiments, material at the center of the upper cavity has a larger displacement when PE is the upper material. Considering the results of the ABS/PS simulations, larger differences can be observed. Especially at the lower cavity the experimental and numerical patterns are different. Here, the flow resembles a plug flow, when ABS is the lower material. This effect is not described by the simulations. Causes of these differences in the flow of ABS can be that the Bingham-like behavior (existence of a yield stress) is insufficiently described by the modified Cross model and that elastic effects have been neglected. In the PE/PS simulations these differences do not occur and the use of labels as material markers seems to be suitable to follow the PE/PS interface.
Figure 3.23: Piston driven contraction flow of LDPE
Figure 3.24: Piston driven contraction flow of LDPE with correction of labels
Figure 3.25: Piston driven contraction flow of PS
Figure 3.26: Piston driven contraction flow of ABS
Figure 3.27: Material combination: lower material: ABS, upper material: PS
Figure 3.28: Material combination: lower material PS, upper material: ABS
Figure 3.29: Material combination: lower material PE, upper material: PS
Figure 3.30: Material combination: lower material: PS, upper material: PE
3.4 Summary

In order to test the method to simulate a piston driven contraction flow on a constant grid some numerical investigations have been made. A viscosity jump between the polymer viscosity and the piston viscosity of three decades appeared to be sufficient to impose the piston velocity at the moving piston interface. To approximate the discontinuity in the viscosity and the velocity boundary conditions at the interface with the shape functions of the quadratic triangular elements, several possibilities were tested. The best results were obtained by prescribing the piston velocity at the first node in the melt region, with a correction to avoid "backflow" on elements in combination with prescribing the viscosity such that negative values in the quadratic viscosity interpolation are avoided. As a consequence of the stick/slip-condition and the typical deformation of the fountain flow a discontinuity in the label values at the wall and high gradients in the label-field at the interface are created. These steep gradients suffer from numerical diffusion. Effect of this diffusion is a distortion of the initially straight interfaces and the inability to determine the position of the interface positions by label values. To reduce the influence of the numerical diffusion on the results, a correction of the label-field has been applied. At nodes inside the piston regions, label values were substituted by the exact label values. The correction did improve the simulation results. Disadvantages of such a correction method are the possibility to introduce oscillations and the inability to correct label values across the interface. More effective methods to reduce the influence of numerical diffusion could be a redefinition of the whole label-field by which the critical contours are kept in place or the use of a more accurate numerical scheme to handle convection. Drawback of the use of a constant grid appeared to be the impossibility to apply local mesh-refinements at the interfaces. Consequently, fountain flow phenomena occur at a certain distance of the interfaces in contrast with the experimental results. Nevertheless, the simulation results proved to be in good agreement with the experimental results. All the characteristic deformation patterns in the experimental results are described by the numerical simulations.
Chapter 4

Simulation results of a reactive capillary flow

In this chapter the results are presented of a numerical simulation of a reactive capillary flow. The method to simulate a piston driven flow on a constant mesh has been applied to simulate the behavior of a thermosetting polymer (PU) in a capillary rheometer.

4.1 Capillary viscometry

A capillary flow is a typical example of a piston driven flow through a contraction. Most widely used applications of capillary flow systems are the Melt Flow Indexer and the capillary rheometer. The Melt Flow Indexer is used as a quality control test to assess information about the fluidity of a melt at standard conditions. The capillary rheometer is a widely used instrument in the rheology of polymer melts. This apparatus uses the constant pressure drop of a fully developed pipe flow (Poiseuille flow) to determine the stationary viscosity. A fluid in a cavity is forced through a capillary with a constant diameter. Consequence of the small diameter of the capillary is a fully developed pipe flow and thereby a constant pressure drop. With the pressure drop and the flow rate through the capillary it is possible, using the Rabinowitsch equation, to calculate the shear stress and shear rate at the wall of the capillary, by which the viscosity becomes known (see Cogswell [7]). Application of capillary viscometry to reactive materials, on the contrary, does not give always useful results. The use of a capillary rheometer requires a homogeneous material all over the capillary. In case of reactive materials this requirement can not always be met, since the curing of the reactive compound is time and place dependent. Consequently, the fluid will be inhomogeneous. In order to show that the developed tools give the opportunity to study the behavior of a thermoset in a capillary rheometer, a numerical simulation has been made of this reactive piston driven flow through a contraction.

4.2 Reactive flow problem

The capillary rheometer has been modeled by a 1:3-contraction. The physical flow situation is depicted in figure 4.1. The length and diameter of the capillary are respectively 22.5 mm and 1.5 mm; the length and diameter of the cavity are 20 mm and 45 mm. With the aid
of the piston, the curing thermoset is forced through the capillary with a constant piston velocity. The moving piston/polymer interface has been modeled in the same way as the moving interfaces of the contraction flow in chapter 3. The piston material was given a viscosity value $10^3$ Pa-s. The initial polymer viscosity is of order $O(10^{-1})$ Pa-s. During the curing of the thermoset, this value increases towards infinity. In the simulations, however, the high viscosity values were truncated at a value $10^3$ Pa-s (piston viscosity). The corresponding degree of conversion was about 0.8, which implies that the no-flow condition was not reached at the gel point $X_{gel} = 0.85$, but somewhat earlier. As soon as the thermoset had reached the no-flow condition, the viscous dissipation was set to zero.

The heating of the material has been modeled by a convective boundary condition of the temperature at the wall (equilibrium between heat transfer at the wall and conduction in the polymer). This boundary condition is given by:

$$-\lambda \frac{\partial T}{\partial n} = h_c (T_{\infty} - T)$$

(4.1)

with $\lambda$ the conductivity of the polymer and $h_c$ the heat transfer coefficient polymer/steel ($\approx 3500$ W/m$^2$K). Temperature $T_{\infty}$ was set to 350 K and the initial temperature of piston and polymer was prescribed 300 K. At the interface piston/polymer there is a discontinuity in the conductivity values of steel and polymer. This jump has been approximated with the quadratic shape functions in the same way as the velocity boundary conditions were prescribed. The $\rho c_p$-values of steel ($\approx 3.7 \cdot 10^6$ J/K-m$^3$) and polymer ($\approx 1.2 \cdot 10^6$ J/K-m$^3$) are about the same. Therefore, the averaged value has been used for both materials.

A point of attention is the conversion of the materials. Physically, the piston does not react. This implies that, during the reaction of the thermoset, a discontinuity in the conversion is created at the interface. Convection of this discontinuity will suffer from diffusion (or dispersion). The result will be a distorted conversion-field at the interface. To avoid this
distortion, the piston has been modeled such, that it did react without having influence on temperature and velocity. Then, the conversion at the interface is defined by a smooth function that does not suffer from numerical diffusion. At the inflow of the piston an averaged value of the conversion at the wall was prescribed.

The coupled system of equations (see chapter 2) has been solved, using a Picard iteration scheme. The sequence in which equations have been solved is shown in figure 4.3. Convergence of solutions was tested on velocity, temperature and conversion. In the iteration loop, solutions at the old time level were used as initial estimations of the new solutions. The mesh that has been used is shown in figure 4.2.

4.3 Numerical results

Three different situations of the capillary flow have been simulated:

- **simulation 1**: the piston velocity was set to zero;
- **simulation 2**: the piston velocity $V_p = 0.5$ mm/sec;
- **simulation 3**: the piston velocity $V_p = 1.0$ mm/sec.

In this way, the influence of convection on temperature and conversion could be observed. In simulation 1, convection is absent by which only the mutual influence of temperature and
conversion is seen. Results of the three simulations are shown in the figures 4.6, 4.7 and 4.8 respectively. The exact piston positions are indicated by straight horizontal lines. Since experimental results are not available, a qualitative check on the results has been made on the basis of dimensionless numbers. The dimensionless numbers that were used, are the four Damköhler-numbers, the Graetz-number, the Brinkman-number and the Nusselt-number. These numbers are defined by:

\[
\begin{align*}
\text{DaI} &= \frac{R_0 L}{V X_0} = \frac{\text{molar change by reaction}}{\text{molar change by convection}} \\
\text{DaII} &= \frac{R_0 \rho_0^2}{D X_0} = \frac{\text{molar change by reaction}}{\text{molar change by diffusion}} \\
\text{DaIII} &= \frac{h_r R_0 L}{c_p V^2 T \rho_0} = \frac{\text{heat change by reaction}}{\text{heat change by convection}} \\
\text{DaIV} &= \frac{r_f^2 h_r \rho_0}{\lambda_0 t_0 T} = \frac{\text{heat change by reaction}}{\text{heat change by conduction}} \\
Gz &= \frac{\rho_0 c_p r_0^2}{\lambda_0 t_0} = \frac{\text{heat change by convection in flow direction}}{\text{heat change by conduction normal to flow direction}} \\
\text{Br} &= \frac{n_0 V^2}{\lambda_0 T} = \frac{\text{heat change by dissipation}}{\text{heat change by conduction}} \\
\text{Nu} &= \frac{h_c L}{\lambda} = \frac{\text{convection heat transfer}}{\text{conduction heat transfer}}
\end{align*}
\]

Characteristic values of material properties are given in appendix B and characteristic lengths are given in the problem definition; R_0 is of order \(O(10^{-1})\) and X_0 = 1. The characteristic time t_0 is defined as L/V. Note that the very small diffusion coefficient (\(O(10^{-7})\)) results in a very high DaII-number and consequently, the effects of molar diffusion can be neglected. Further on, since Br \(\leq 10^{-4}\), also the viscous dissipation could be neglected.

The results of simulation 1 (figure 4.6) clearly demonstrate the effect of the jump in the conductivity at the interface. Compared to the polymer, the piston material reaches very quickly a steady temperature of 350 K. Moreover, the Nusselt-number of the polymer in the cavity is of order \(O(10^2)\) and, consequently, the temperature gradient does occur within the polymer. Other phenomenon that can be observed in the results of simulation 1 is a relative homogeneous temperature distribution inside the capillary, compared to the cavity. Considering the Damköhler IV-number, also this effect is physically realistic. At the capillary, DaIV\approx0.65 whereas at the cavity DaIV\approx5.81. This implies that temperature changes at the cavity are mainly determined by reaction and at the capillary temperature changes are levelled by conduction. This temperature distribution is reflected in the conversion of material: in the capillary changes in conversion are relatively small compared to temperature distribution in the cavity. Further on, the absence of convection and the absence of molecular diffusion the conversion is completely determined by reaction (DaII\(\rightarrow\) \(\infty\)), which is a function of temperature and conversion. Besides the physical realistic contourlines of temperature and conversion that are predicted by the simulation, also the first time at which the gelpoint is
reached is in agreement with the isothermal curing time of the compound. This curing time is defined (at constant temperature) by:

\[ t_x = \int \frac{X_{end}}{X_0} \frac{1}{R_x} \, dX. \]  

(4.2)

At temperature \( T = 350 \text{ K} \), \( t_x = 8.5 \text{ s} \), which compares well with the predicted \( t = 8 \text{ s} \) of the simulation.

The results of simulation 2 and 3 are shown in figure 4.7 and 4.7 respectively. Both simulations show qualitatively an identical behavior. As a consequence of the lower velocity value of simulation 2, the Graetz-number in the capillary is of order \( O(1) \), whereas in simulation 3 the Gz-number in the capillary is of order \( O(10^1) \). Consequently, the conduction has more influence, which explains the difference between the temperature profiles at \( t = 4.0 \text{s} \). Both flow simulations show an essential non-homogeneous distribution of temperature and conversion at the capillary, caused by convection (DaI \( \approx 10^{-1} \) and DaIII \( \approx 10^{-3} \)). Characteristic illustration of the time dependent inhomogeneity of the curing thermoset is given by the velocity profiles that are plotted on the cross-section at the middle of the capillary (figure 4.4). The numbers 1-10 indicate the time (in seconds). As a result of the viscosity changes that are generated by the curing, the velocity profiles alter from parabolic profiles to flat profiles (plug flow). The inhomogeneity of the material is also reflected in the pressure distribution: the pressure drop through the capillary is not constant. This is visualized in figure 4.5, where the pressure contours at the wall of the capillary are plotted.

Considering the simulation results of the capillary reactive flow, it can be concluded, that the simulation method or basis of a constant grid, is capable to predict a piston driven reactive flow in a realistic way. Application illustrates that the capillary rheometer is not suitable to obtain information about the fluidity of a thermoset in a trivial way, since the pressure drop along the capillary is not constant.
Figure 4.6: Simulation 1: contourlines of temperature and conversion
Figure 4.7: Simulation 2: contours of temperature and conversion
Figure 4.8: Simulation 9: contourlines of temperature and conversion
Chapter 5

Conclusions and recommendations

A numerical method has been set up and tested to simulate a piston driven flow, which is characterized by a time dependent fluid domain, on a constant mesh. Principle ingredients of the method are the use of a stick/slip-condition at the wall and a jump in the viscosity at the moving interface.

Considering the results, it can be concluded that the use of a viscosity jump and the stick/slip-condition is adequate to impose the piston velocity at the interface with reasonable accuracy. The particle tracking algorithm proved to be a capable tool to predict the deformation patterns, caused by the flow. All characteristic deformations of the fluid are described well. Cause of differences between numerical and experimental results can be the effects of expansion and shrinkage that are not taken into account and the elastic effects that have been neglected. At the piston/polymer interface, the particle tracking algorithm becomes inaccurate as a consequence of the discontinuity in the label-solution. Convection of a function that contains a discontinuity or steep gradients suffers from numerical diffusion or dispersion. This makes the particle algorithm incapable to follow the piston interfaces. Interfaces in the fluid region, on the contrary, can be traced adequately, since here no discontinuities or steep gradients are present. Disadvantage of the method is the use of the constant mesh. Local mesh-refinements at the interfaces are required to describe the fountain flow phenomena here more accurately. Otherwise, the fluid near the interfaces is not convected accurately. The method has a high flexibility: implementation is rather straightforward and 3D-simulations are easy to realize. In this study only the situation solid/liquid has investigated. The situation liquid/air is qualitatively identical, with the difference that now the velocity field at the interface is determined by the fluid. Therefore, the method can be a flexible way to simulate the filling of a cavity.

In order to improve the predictive quality of the method, in further investigation the attention has to be focused on the improvement of the accuracy of the velocity field near the interfaces and on the problem of the numerical diffusion. Attractive possibilities to improve accuracy are the use of more advanced numerical scheme, such as the Taylor-Galerkin Discontinuous Finite Element Method (TG-DFEM), and the use of higher order elements (spectral method). Thereby, also the numerical diffusion may be reduced. Other possibility to reduce effects of diffusion is to smooth steep gradients that are created in solutions. An appropriate smoothing algorithm has to be developed to achieve this. Then, a flexible numerical method is obtained to simulate the deformation of a driven fluid in an arbitrary 3D geometry.
Bibliography


Appendix A

Experiments

Experiments with a piston driven contraction flow through have been done with three materials:

- ABS: Ronfalin FX50 (DSM)
- PS: Styron 678E (DOW)
- LDPE: Stamylan 2101TN47 (DSM).

Principle of the experiments is to visualize deformation patterns of the viscous flow through the contraction by using tracers. A cylindrical cavity is filled with a stack of polymer counters (see fig. A.1). Some of the counters have a contrasting color, but consist of the same material as the remaining counters. These counters serve as tracer: they make it possible to visualize deformation patterns of the flow, without disturbing the flow (same material). After the cylindrical cavity has been filled, the polymer counters are melted with the aid of heating elements. Once an isothermal situation has been reached (473 K, after 20 minutes), with the aid of the piston, the melt is injected from one cylindrical cavity into another, with a constant piston-velocity of 5 cm/sec. The upper cavity is created by the melt, that is driving the upper piston. At several defined displacements of the piston, the experiments are cooled down. Then, by cutting the experimental results in two, along the axi-symmetric centerline, deformation patterns become visible. The experimental setup, that is used for the experiments, is shown in figure A.2. The contraction has a diameter of 5 mm. and a length of 20 mm. Values for the length L and the diameter D of the initial configurations are given in table A.

Figure A.1: Initial configuration: polymer counters containing some tracers
Main problem in obtaining well-defined experimental results, is the expansion and shrinkage of the polymer at respectively heating up and cooling down the material. Values for expansion and shrinkage are given in table A. The relative large value of the shrinkage of PE with respect to the value of PS and ABS is the result of the semi-crystalline structure of PE. ABS and PS are amorphous polymers. Due to the relatively large expansion of material, the straight lines of the initial configuration are already deformed. Cooling down the experiments isochoric, again the patterns are severely distorted, now by the effect of shrinkage and due to the enclosing of air bubbles. In order to avoid the distortion of patterns by air-bubbles and to get a controlled expansion and shrinkage, experiments have been done in such a way, that expansion is defined with respect to the top of the lower piston and shrinkage is defined with respect to the center of the contraction. This was realized by cooling down isobaric at

<table>
<thead>
<tr>
<th>Material</th>
<th>L (mm)</th>
<th>D (mm)</th>
<th>Expansion (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>ABS</td>
<td>60.0</td>
<td>16.0</td>
<td>6.0</td>
</tr>
<tr>
<td>PS</td>
<td>60.0</td>
<td>16.0</td>
<td>4.0</td>
</tr>
<tr>
<td>PE</td>
<td>59.0</td>
<td>16.0</td>
<td>10.0</td>
</tr>
</tbody>
</table>

Table A.1: Dimensions and values of expansion
12.2·10^5 \text{ Pa}. Due to the small size of the contraction diameter, melt solidified here first and therefore shrinkage is defined with respect to the center of the contraction. Effect of this way of doing experiments on the results was, that they already show severe deformations at a zero displacement of the piston. The effects of this experimental method should be kept in mind, comparing the numerical and experimental results.

Material combinations

Besides experiments with only one material, also experiments with combinations of materials have been done. Process conditions of these experiments are the same as in case of experiments with one material: a constant piston-velocity of 5 mm/sec, temperature = 493 K, and pressure $p = 12.2 \cdot 10^6 \text{ Pa}$. The different initial configurations are shown in figure A.3; in table A. Experimental results are not given here, but together with numerical results (chapter 3) in order to get a check on the predictive quality of the simulation method.

<table>
<thead>
<tr>
<th>Combinations</th>
<th>L (mm)</th>
<th>D (mm)</th>
<th>Expansion (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>ABS/PS</td>
<td>58.0</td>
<td>16.0</td>
<td>4.5</td>
</tr>
<tr>
<td>PS/PE</td>
<td>57.0</td>
<td>16.0</td>
<td>7.0</td>
</tr>
<tr>
<td>PE/ABS</td>
<td>57.0</td>
<td>16.0</td>
<td>7.5</td>
</tr>
</tbody>
</table>

Table A.2: *Dimensions for combinations of materials*

![Initial configurations for material-combinations](image)
Appendix B

Generalized Newtonian viscosity models

B.1 PS - Styron 6783 (DOW)

As constitutive model for the shear viscosity of PS, a 7-constant Cross model is used. This model reads as:

\[ \eta(\dot{\gamma}, T, p) = \frac{\eta_0(T, p)}{1 + \left(\frac{\dot{\gamma}}{\tau_0^*}\right)^{1-n}} \]  \hspace{1cm} (B.1)

where \( \dot{\gamma} \) is the shear rate, defined as \( \dot{\gamma} = \sqrt{2D^d : D^d} \), \( T \) the temperature and \( p \) the pressure; \( \eta_0(T, p) \) is given by:

\[ \eta_0(T, p) = D_1 e^{-A_1(T-T^*)}, \]  \hspace{1cm} (B.2)

with:

\[ T^*(p) = D_2 + D_3p \quad \text{and} \quad A_2(p) = A_2 + D_3p. \]  \hspace{1cm} (B.3)

The values of the model parameters are:

- \( D_1 = 4.76 \cdot 10^{10} \) [Pa·s]  
- \( D_2 = 373 \) [K]  
- \( D_3 = 5.1 \cdot 10^{-7} \) [K/Pa]  
- \( \tau^* = 3.08 \cdot 10^4 \) [Pa]

\( A_1 = 25.742 \) [-]  
\( A_2 = 61.056 \) [K]  
\( n = 0.252 \) [-].

B.2 LDPE - Stamylan 2101TN47 (DSM)

As a constitutive relation for the shear viscosity of PE a Carreau-Yassuda model is used. This model is given by:

\[ \eta(\dot{\gamma}, T) = a_T \eta_0(1 + (\lambda a_T \dot{\gamma})^\alpha)^{(1/\alpha - 1)} \]  \hspace{1cm} (B.4)

with \( a_T \) a temperature correction parameter, defined as:

\[ a_T = e^{\frac{E_a}{K}} \left( \frac{1}{T - T_{ref}} \right) \]  \hspace{1cm} (B.5)

55
the shear rate (see PS), $T$ the temperature and $R$ the gas-constant. Values of the model parameters are given by:

\[
\begin{align*}
\eta_0 &= 3.455 \cdot 10^4 \text{ [Pa-s]} \\
\lambda &= 3.039 \text{ [-]} \\
\alpha &= 0.45836 \text{ [-]} \\
n &= 0.32107 \text{ [-]} \\
T_{ref} &= 463 \text{ [K]} \\
E_A &= 66 \text{ [kJ/mol]}
\end{align*}
\]

### B.3 ABS - Ronfalin FX50 (DSM)

ABS has different viscosity behavior, compared with the behaviour of PS and LDPE. Where the viscosities of PS and LDPE approach constant values as the shear-rate decreases, the viscosity of ABS increases continuously. Therefore, ABS shows a Bingham-like behavior, resulting in a plug flow at very low shear-rates. This viscosity behavior has been modeled with an adapted Cross model. At moderate to high shear-rates, a 7-constant Cross model has been fitted on experimental viscosity behavior. At moderate to low shear-rates, a power-law is used to model the increasing viscosity. For very low shear-rates, the viscosity is given a constant value. This adapted Cross model is given by:

\[
\eta(\dot{\gamma}) = \begin{cases} 
4.5 \cdot 10^4 & \text{if } \dot{\gamma} < 10^{-2} \\
4.5 \cdot 10^2 \dot{\gamma}^{-0.5} & \text{if } 10^{-2} \leq \dot{\gamma} \leq 10^0 \\
\text{7-const. Cross model} & \text{if } \dot{\gamma} > 10^0 
\end{cases}
\]  

(B.6)

The definition of the Cross model is given at the viscosity model of PS. Values of the model parameters for ABS are given by:

\[
\begin{align*}
D_1 &= 5.2 \cdot 10^3 \text{ [Pa-s]} \\
D_2 &= 473 \text{ [K]} \\
D_3 &= 5.1 \cdot 10^{-7} \text{ [K/Pa]} \\
\tau^* &= 7.5 \cdot 10^4 \text{ [Pa]}
\end{align*}
\]

$$A_1 = 7.039 \text{ [-]}$$

$$A_2 = 119.7 \text{ [K]}$$

$$n = 0.25 \text{ [-]}$$

Note that, in case of isothermal simulations the temperature corrections only have to be computed once. Pressure dependency on the viscosity has been neglected.

### B.4 Polyurethane

In this study, reactive flow is simulated for polyurethane (PU). PU is a widely used material in RIM-processes. The reaction of this material is highly exothermal. PU is modeled with the constitutive relations, given in Castro and Macosko [6]. Important aspect with respect to the viscosity of PU, is the absence of shear-thinning behavior. The viscosity of PU is only a function of the temperature $T$ and the conversion $X$. The constitutive relation for the viscosity is given by:

\[
\eta(T, X) = A_\eta e^{(E_\eta/R \cdot T)} \left( \frac{X_{gel}}{X_{gel} - X} \right)^{(A + B X)}
\]

where $A_\eta$ is a pre-exponential factor, $E_\eta$ an activation energy, $R$ the gas constant and $A$ and $B$ constants. Values of the parameters are given by:
A\eta = 4.1 \cdot 10^{-8} \text{ [Pa-s]} \quad X_{gel} = 0.85 \quad [-]
E\eta = 38.3 \cdot 10^3 \text{ [J/mol]} \quad A = 4 \quad [-]
\mathcal{R} = 8.314 \quad \text{[J/(mol-K)]} \quad B = -2 \quad [-].

The constitutive relation for the reaction rate is given by equation 2.13. The constants $K_i$ are defined by Arrhenius-like functions of the temperature (eq. 2.14). The values of the parameters for PU are given by:

\begin{align*}
k_1 &= 9.36 \cdot 10^7 \quad [1/s] \\
E_1 &= 57.8 \cdot 10^3 \quad [\text{J/mol}] \\
k_2 &= 0 \quad [1/s] \\
E_2 &= 0 \quad [\text{J/mol}] \\
C_0 &= 2.6 \cdot 10^3 \quad [\text{mol/m}^3] \\
\eta &= 2 \quad [-] \\
m &= 0 \quad [-].
\end{align*}

Values for the heat-capacity $c_p$, density $\rho$, heat conductivity $\lambda$ and the specific reaction heat $h_r$ are given by:

\begin{align*}
c_p &= 1880 \quad [\text{J/(kg-K)]} \\
\rho &= 1000 \quad [\text{kg/m}^3] \\
\lambda &= 0.17 \quad [\text{W/(m-K)]} \\
h_r &= 83 \cdot 10^3 \quad [\text{J/mol}].
\end{align*}