Reaction Injection Molding: Analyzing the Filling Stage of a Complex Product With a Highly Viscous Thermoset*

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Although numerical simulation has proved to be a useful tool to predict the filling behavior of injection molding products, few applications in reaction injection molding (RIM) have been reported. This is mainly due to the lack of appropriate descriptions of the material behavior, and to the lack of accurate material data. The analysis of the filling stage of the RIM process requires the determination of the cure kinetics and the rheological, pvT, and thermal behavior of the material, which in this study is a highly filled copolymer of ethylene and vinyl-acetate (EVA). Since this copolymer yields some viscosity anomalies, due to the amount of filler, the rheology of a similar EVA compound with low filler content was also investigated. The viscosity of this compound can be described by a Carreau model at low conversions levels (1%), whereas a conversion-dependent Power-Law model proves appropriate for higher conversion. Moreover, the viscosity behavior of both compositions is similar in the processing range. The combined Carreau/Power Law viscosity model is used for the numerical simulations of a complex product. The simulation results agree well with experimental molding results. The analysis shows that a combination of material characterization and modeling, numerical simulation, and experimental verification is necessary to make simulation programs a useful design tool for RIM products and molds.

INTRODUCTION

With the current tendency in industry to integrate several functions into one product—resulting in products with complex geometries—the prediction of process and product properties from manufacturing experience has become more difficult. In such cases, numerical simulation may be a useful tool in the design of products and molds (1). Therefore, the injection molding process of thermoplastics has been extensively analyzed over the past decades.

Since injection molding is generally limited to thin-walled products, a thin-film approximation is commonly applied to model the flow during the filling stage of injection molding processes. These models have been implemented in computer codes to simulate injection molding processes and to enable product designers to predict flow front advancement, pressure buildup, temperature distribution, and, in some cases, thermal and flow-induced stresses (2–5).

Reaction injection molding (RIM) is characterized by in-mold polymerization of monomers or oligomers. This causes an increase in viscosity with increasing conversion, and eventually results in the formation of a polymer network, which can flow no longer; viscosity increases towards infinity, and the material becomes a gel. Thus, compared with thermoplastic injection molding, the thermosetting or curing reaction causes an extra complication in modeling the RIM process.

Although RIM has been implemented in several injection molding simulation programs, monitoring the filling stage of the RIM process by numerical simulation has appeared to be difficult, especially when products with complex shapes and filled materials are involved. Typical problems frequently encountered in the process are: weld lines, short shots due to preliminary gelling, air entrapment, and viscoelastic behavior. Moreover, the materials used in RIM can give rise to complications in material characterization, e.g., in determination of the gel point and the exact viscosity

behavior (6). This was already stated by Kamal and Ryan [7, p. 359]: “Application of numerical simulation of injection molding] to thermosetting systems has not been extensively evaluated due to inadequacies in material characterisation and process models incorporating chemical reaction.”

A major aim of developing a numerical simulation program is to obtain a design tool for products and molds for RIM, in order to reduce the need for expensive mold prototypes and time-consuming experimental molding, and to understand product performance and failures. This investigation will show that the filling stage of thermoset products of a complex shape can be analyzed successfully by combining material characterization, modeling of material behavior, numerical simulation, and experimental verification.

**MODELING THE RIM PROCESS**

The modeling of RIM has been described previously (e.g., 3, 4, 7, 8). Since we do not add anything to this modeling, it will only be outlined briefly in this section.

**Equations of Conservation**

Reaction injection molding is governed by the equations of conservation of mass (Eq 1), momentum (Eq 2), moment of momentum (Eq 3), energy (Eq 4), and moles (Eq 5):

\[ \dot{\rho} + \rho \text{tr}(\mathbf{D}) = 0 \]
\[ \nabla \cdot \mathbf{\sigma}' + \mathbf{\tau}^f = \rho \mathbf{\dot{v}} \]
\[ \mathbf{\sigma} = \mathbf{\sigma}' \]
\[ \rho \dot{e} = \mathbf{\sigma} : \mathbf{D} - \nabla \cdot \mathbf{\dot{h}} + \rho r + \rho h, R_c \]
\[ \dot{X} = \nabla \cdot (\mathbf{D} \dot{\mathbf{X}}) + R_c \]

**Constitutive Equations**

To describe material behavior, constitutive models have to be developed for the Cauchy stress tensor, the density, the heat flow, the internal energy rate and the reaction rate:

- **Cauchy stress tensor** \( \mathbf{\sigma} \) (generalized Newtonian):
  \[ \mathbf{\sigma} = -p \mathbf{I} + \mathbf{\sigma}'^d \]
- **Density** \( \rho \):
  \[ \frac{\dot{\rho}}{\rho} = -\alpha \dot{T} + \kappa \dot{p}_0 + \frac{\beta}{C_v} \dot{X} \]
- **Heat flow** \( \dot{h} \) (Fourier’s Law):
  \[ \dot{h} = -\lambda \cdot \nabla T \]
- **Internal energy rate** \( \dot{e} \):
  \[ \dot{e} = c_p \dot{T} + \frac{p_0}{\rho} \dot{\rho} + \frac{1}{\rho} \lambda \dot{p}_0 \]
- **Reaction rate** \( R_c \) (\( n \)th order kinetics):
  \[ R_c = k_1 e^{-E_a/RT} (1 - X)^n \]

**Assumptions and Simplifications**

The basis of further RIM modeling is the thin-film approximation, which limits the use of the model to thin-walled products (4). As we will deal with such products only, this limitation is of minor importance. Moreover, full 3-dimensional RIM simulations on complex products are not realistic, because they would—at this time—lead to excessive memory usage and computing times.

The thin-film approximation implies the following simplifications:

- The pressure is assumed to be constant over the product thickness.
- Velocity gradients parallel to the product midplane (i.e. the plane parallel to and halfway between the mold walls of each product section) can be neglected compared with velocity gradients perpendicular to the midplane.
- Velocity components in thickness direction can be neglected compared with velocity components in midplane directions.
- Heat conduction in the midplane can be neglected compared with heat conduction in thickness direction.

This model has been implemented in an injection molding simulation program called VIp, which employs the finite element method (8, 9). Further simplifications used are:

- Inertia and body forces can be neglected compared with viscous forces (\( Re \ll 1 \) and \( Re \ll Fr \)). Although inertia forces do play an important role in most RIM processes and should therefore be accounted for in the model (see, e.g., 10), the high viscosity of the material that we consider in this study justifies this assumption.
RIM: Analyzing the Filling Stage

- The influence of abrupt changes of thickness on pressure is neglected, as well as the influence of mold intersections and sharp corners.
- At the flow front, a fountain flow front model is employed (9, 11, 12), but front residence times are not taken into account.
- Slip at the mold walls is not taken into account.
- Elongational flow is neglected compared with shear flow when determining the deformation rate tensor $D$.

As a consequence of this last simplification, $tr[D] = 0$ in Eq 7, and the shear rate $\dot{\gamma}$, defined as $\dot{\gamma} = \sqrt{2D^T \cdot D}$ reduces to:

$$\dot{\gamma} = \| \frac{\partial \mathbf{u}^*}{\partial x_i} \|$$

in which $^*$ indicates “with respect to midplane,” and $x_i$ is the coordinate in thickness direction. Specifically for the analysis in this investigation, some extra assumptions are made:

- no conversion occurs before the material enters the mold;
- reaction shrinkage is not taken into account ($\beta = 0$ in Eq 11);
- specific heat capacity does not depend on pressure;
- viscosity is not a function of pressure, since the pressures in RIM are generally too low to significantly influence viscosity;
- the molecular diffusion in Eq 5 can be neglected: $R_e = \dot{X}$.

### Implementation

Because of these simplifications, the equations of conservation of mass (Eq 1), momentum (Eq 2), and moment of momentum (Eq 3), together with the constitutive models, can be combined to give the pressure problem (9):

$$-\mathbf{v}^* \cdot (S\mathbf{v}^* p) + \int_{-h/2}^{h/2} \kappa d\chi \dot{p} + C \frac{\partial p}{\partial t} = \int_{-h/2}^{h/2} \alpha T \dot{\mathbf{x}}$$

in which $C$ is the mold compliance, and:

$$S = J_2 - \frac{J^2_2}{J_0}; \text{ fluidity}$$

$$J_i = \int_{s^*}^{s} \frac{\chi^i}{\eta} d\chi : i = 0, 1, 2$$

whereas the integral in Eq 20 is taken over the fluid layer thickness $s^* \leq \chi \leq s$. The boundary conditions usually consist of a prescribed pressure or flow at the injection area, no leakage at the mold edges, and ambient pressure at the flow front.

The equation of conservation of energy (Eq 4) and the constitutive equations on heat flow (Eq 12) and internal energy (Eq 14) combine to give the temperature problem:

$$-\frac{\partial}{\partial x_i} \left( \lambda \frac{\partial T}{\partial x_i} \right) + \rho c_p \dot{T} + \alpha T \dot{p} = \eta \dot{\gamma}^2 + \rho h R_e$$

The boundary conditions for the temperature problem prescribe temperatures or heat fluxes at the mold walls, and the injection temperature (which in RIM is generally lower than the wall temperatures).

Combining Eq 5 and the constitutive equation for the reaction rate (Eq 16) yields the conversion problem:

$$\dot{X} = k_1 e^{-E_a/RT}(1 - X)^n$$

whereas conversion is supposed to be zero upon mold entrance (owing to lack of appropriate data).

The simulation of the filling stage of a RIM process globally proceeds as follows (8, 9): for every time step, the flow front is propagated; pressure, pressure gradient, temperature, shear rate, and conversion are extrapolated from the previous to the new filled domain; the coefficients for the pressure, temperature, and conversion problems are determined iteratively by solving these problems and recalculating their coefficients until they converge. Then the next time step is taken, until the mold is filled.

For every simulation, a so-called background mesh is created that covers the entire mold’s geometry. This mesh serves as a “backbone” for the (separate) filled mesh, which covers the filled part of the mold. To solve the pressure, temperature, and conversion problems in the region filled with polymer, the flow front must coincide with edges of the finite elements of the filled mesh. Therefore, partially filled elements of the background mesh are subdivided into “unfilled” and “filled” elements, and the latter are added to the filled mesh. Thus, every time step, new temporary elements are created to cover the melt front, while the previous temporary elements are discarded.

The mesh consists of rectangular elements lying in the midplane direction. In each vertex of these elements, a grid line is defined over the local mold thickness; this grid line is covered by a (user-defined) number of grid points. The pressure and convection problem are solved on the mesh of rectangular elements, whereas the temperature and conversion problem are solved over the grid line in each vertex. The grid points are also used to perform the integrations in Eqs 18 and 20. The grid lines are discretized using 1-dimensional linear finite elements, whereas in many codes a finite difference mesh is used.

### Particle Tracking

Each particle entering the mold is identified by a unique label $\xi$, which usually consists of injection time and injection coordinates:

$$\xi = \xi(X_{nj}, t_{nj})$$

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Conservation of identity states that the identity of each particle does not change:

\[ \dot{\xi} = \frac{\partial \bar{\xi}}{\partial t} + \bar{\nu} \cdot \bar{\nabla} \bar{\xi} = 0 \quad (24) \]

The concept of particle tracking, which involves solving Eq 24, yields the time and position at which particles have entered the mold; or, conversely, particle tracking offers the possibility to see where particles, which were injected at a certain time and position, ultimately end up in the product (8, 9).

The particle tracking option has been implemented in the VIP program.

**MATERIAL CHARACTERIZATION**

**Material**

The material considered is a highly filled copolymer of ethylene and vinyl-acetate (EVA). The filler makes up 45 wt% (121 vol%) of the material and consists mainly of flame retardant and antioxidant. A small amount of peroxide is mixed in, initiating the curing reaction at 120°C. The material is used for its excellent insulating properties. However, its melting properties are poor, and material characterization is difficult: a gel point could never be determined properly.

To investigate the influence of the high filler content, an EVA compound with merely 7 wt% (3.5 vol%) filler has been characterized as well. This compound with low filler content will be referred to as EVA7, whereas the highly filled compound will be called EVA45.

**Reaction Kinetics**

Reaction kinetics have been determined by differential scanning calorimetry (DSC) using a Perkin-Elmer DSC 7. Heat flow was measured while samples were heated from 90°C to 230°C at a constant heating rate of either 3, 5, or 10°C/min. The resulting heat flow data were processed to yield reaction heat and reaction rate. It is assumed that conversion is equal to the fraction of the total reaction enthalpy that has been released (13). A reduced data set of all six experiments (two samples for each of the three temperature rates) was used to fit the reaction rate to the nth order kinetics model given in Eq 16.

The results from the DSC experiments on kinetics are given in Fig. 1 and Table 1. The curing behavior of the EVA45 compound is comparable to that of the EVA7 compound; but the reaction heat \( h \) of the EVA7 compound is twice that of the EVA45 compound.

**Shear Viscosity**

The determination of shear viscosity of thermosets under stationary conditions gives rise to problems: the change of properties due to curing makes capillary viscometry unsuitable for shear viscosity measurements, and the large strains in rotational steady shear viscometry cause the material to be disrupted (14). Therefore, shear viscosity is generally determined indirectly by dynamic mechanical thermal analysis (DMTA), in which the dynamic viscosity is measured. The Cox-Merz rule (15) enables translation of this dynamic viscosity to a quasi-stationary situation—as exists in RIM.

\[ \eta_d(\omega) = \eta_s(\dot{\gamma}) = \omega \quad (25) \]

It has been shown that the Cox-Merz rule can fail for highly filled polymers, especially at high shear rates (14, 16). However, for the highly filled EVA (from which the curing agent was omitted to enable capillary viscometry) the Cox-Merz rule has been found valid even at a shear rate of 200 rad/s (17).

Dynamic viscosity measurements were carried out on a Rheometrics RDS-II using a 25-mm-diameter plate-plate geometry. The samples were heated from 90°C to 230°C at a constant heating rate of 3, 5, or 10°C/min, and were subjected to oscillations of 1, 10 and 100 rad/s at a strain of 0.25%.

The results for EVA45 (Fig. 2) show an unexpected viscosity decrease between 170°C and 190°C at a heating rate of 10°C/min, and, to a lesser extent, at 5°C/min. The high filler amount was held responsible for this decrease, which makes the EVA45 compound difficult to characterize and model.

**Figure 3** shows the viscosity of EVA7, whereas a comparison between corresponding viscosity curves of both compounds can be made from **Fig. 4**. The most striking difference appears between 100°C and 170°C, where the viscosity of the (highly filled) EVA45 com-
The minimum viscosities of both at 10°C/min are nearly equal; as temperature increases, these viscosities increase similarly. From these observations, it is suggested that the viscosity drop of EVA45 at 170°C is caused by breakdown of filler agglomerates, and that this effect is concealed at lower heating rates by the curing effect on viscosity (18).

Yet, no single viscosity model could be found to fit the EVA7 viscosity data over the entire relevant temperature and conversion range. Instead, the viscosity at low conversion levels (X < 1%) is described by a Carreau model (15), whereas for higher conversions a Power Law model has been adopted:

\[ \eta = \eta_0 \alpha_r \beta_T [1 + (\lambda \dot{\gamma})^n]^{\frac{n-1}{2}} \quad (X < 1\% \text{: Carreau}) \]  
\[ \eta = \eta_0 \alpha_r \beta_T \dot{\gamma}^{n-1} \quad (X \geq 1\% \text{: Power Law}) \]  

in which \( \dot{\gamma} \) is the reduced shear rate (\( = \omega \alpha_r \)), and \( \alpha_r \) and \( \beta_T \) are the horizontal and vertical shift factor defined by:

\[ \alpha_r = \frac{\eta_0(T)}{\eta_0(T_0)} \cdot \frac{T_0 \rho_0}{T \rho} \quad (28) \]
\[ \beta_T = \frac{T \rho}{T_0 \rho_0} \quad (29) \]

These shift factors can be described by:

\[ \log \alpha_r(T, T_0) = -c_1 (T - T_0) \left[ \frac{c_2}{c_2 + (T - T_0)} \right] \quad \text{[WLF-equation (15)]} \]  
\[ b_T(T) = e^{c_3 T + c_4} \quad (31) \]

The parameters \( T_0, c_1, c_2, a, b, \eta_0, \) and \( n - 1 \) are given in Table 2, the latter two being functions of conversion in the Power Law model. Figure 3 shows the fitted curves for this two-fold model.

The isoconversion lines, connecting points of equal conversion for every shear rate (Fig. 3), are approximately parallel to each other, implying that the viscos-

<table>
<thead>
<tr>
<th>Carreau (for X &lt; 1%)</th>
<th>Power Law (for X &gt; 1%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \eta = \eta_0 a_r \beta_T (1 + (\lambda \dot{\gamma})^{1-n})^{\frac{n-1}{2}} )</td>
<td>( \eta = \eta_0 a_r \beta_T \dot{\gamma}^{n-1} )</td>
</tr>
<tr>
<td>( \eta_0 = 4.38 \times 10^4 \text{ Pa.s} )</td>
<td>( \eta_0 = a_0 + a_1 X + \cdots + a_4 X^4 )</td>
</tr>
<tr>
<td>( \lambda = 1.97 \text{ s} )</td>
<td>( a_0 = 2.30 \times 10^3 )</td>
</tr>
<tr>
<td>( \beta_T = \frac{T \rho}{T_0 \rho_0} )</td>
<td>( a_1 = 4.09 \times 10^3 )</td>
</tr>
<tr>
<td>( \beta_T = \frac{105}{105^2} )</td>
<td>( a_2 = 4.07 \times 10^5 )</td>
</tr>
<tr>
<td>( T_0 = 105 \text{ K} )</td>
<td>( a_3 = -4.17 \times 10^7 )</td>
</tr>
<tr>
<td>( \beta_T = \frac{105}{105^2} )</td>
<td>( a_4 = 1.28 \times 10^6 )</td>
</tr>
<tr>
<td>( n - 1 )</td>
<td>( \beta_T = b_0 + b_1 X + \cdots + b_4 X^4 )</td>
</tr>
<tr>
<td>( c_1 = 3.38 \text{ [-]} )</td>
<td>( b_0 = -0.534 )</td>
</tr>
<tr>
<td>( c_2 = 157.8 \text{ [K]} )</td>
<td>( b_1 = -0.725 )</td>
</tr>
<tr>
<td>( T_0 = 410.2 \text{ [K]} )</td>
<td>( b_2 = -0.565 )</td>
</tr>
<tr>
<td>( c_3 = 6.92 \times 10^{-3} \text{ [K}^{-1}] )</td>
<td>( b_3 = 1.92 )</td>
</tr>
<tr>
<td>( b = -2.86 \text{ [-]} )</td>
<td>( b_4 = -1.08 )</td>
</tr>
</tbody>
</table>
ity curves can indeed be shifted with shift factors $a_T$ and $b_T$. The data obtained for 5°C/min and 1 rad/s, however, deflect the isoconversion lines considerably: comparing the curves for 5 and 10°C/min at 1 rad/s, it would seem that points of equal conversion have higher viscosities at higher temperatures, which is physically inconsistent. Therefore, the data at 5°C/min and 1 rad/s have been omitted in the fitting procedure.

### Table 3. pvT and Thermal Data on the EVA45 Compound.

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>specific volume</td>
<td>$0.76 \times 10^{-3}$ m$^3$/kg at $T_0 = 323$ K</td>
</tr>
<tr>
<td>thermal expansion</td>
<td>$7 \times 10^{-3}$ K$^{-1}$</td>
</tr>
<tr>
<td>isothermal compression coefficient</td>
<td>$8 \times 10^{-10}$ Pa$^{-1}$</td>
</tr>
<tr>
<td>heat capacity</td>
<td>$1.95 \times 10^3$ J/(kg·K)</td>
</tr>
<tr>
<td>thermal conductivity</td>
<td>$0.35$ W/(m·K)</td>
</tr>
</tbody>
</table>

Fig. 5. Measured pvT-behavior of EVA45 compound: ○ 0 MPa, * 5 MPa, □ 10 MPa, △ 15 MPa, ▲ 20 MPa.

Fig. 6. Measured specific heat capacity $c_p$ of EVA45 compound at different heating rates: ○ 10°C/min, ▲ 20°C/min.

Fig. 7. Mold cross section of the 200S high voltage insulator part by Raychem Ltd. Product dimensions: ~90 mm (cylinder height) × 90 mm (shed diameter). Pressure transducer positions with corresponding nodal point numbers are indicated by ●.

**Specific Volume**

The pvT-behavior of the EVA45 compound was measured in an experimental setup based on the one described by Zoller et al. (19). Since it is impossible to
control the curing reaction in this apparatus, a non-
curing compound was used. It is assumed that be-
cause of the high filler content, the effect of curing on
the \( pV/T \)-behavior can be neglected.

From the results of the \( pV/T \)-measurements (shown
in Fig. 5), the zero specific volume, and the linear
expansion coefficient \( \alpha \) and isothermal compression
coefficient \( \kappa \), defined by:

\[
\alpha = \frac{1}{V} \frac{\partial V}{\partial T} \bigg|_{p_0, X} \\
\kappa = -\frac{1}{V} \frac{\partial V}{\partial P} \bigg|_{T, X}
\]

(32) (33)
can be determined. Their values are given in Table 3.

**Specific Heat Capacity and Thermal Conductivity**

The specific heat capacity \( c_p \) of a noncuring EVA45
compound was obtained from DSC heat capacity mea-
surements on a Perkin-Elmer DSC-7 (20, 21). Heating
rates of 10°C/min and 20°C/min were used. A com-
parison between a curing and a noncuring compound
showed that conversion has no influence on heat ca-
pacity.

From these measurements (shown in Fig. 6), the
specific heat capacity has been assumed constant at
\( c_p = 1.95 \times 10^3 \text{ J/kg} \times \text{K} \).

Thermal conductivity was measured using Lee’s
disc, in which a known heat flux is passed through a
sample and a reference material in a serial configura-
tion (22). For EVA45, a constant value of \( \lambda = 0.35 \)
W/m/K has been obtained.

**NUMERICAL SIMULATIONS**

**Geometry and Conditions**

The VI\(_p\) program has been used to simulate the
filling stages of a complex mold, the 200S insulator
part from Raychem (Fig. 7), at critical conditions, i.e.,
high conversion and pressure levels.
The material is injected through a tapered sprue and a long runner into a ring gate, which is connected via a restriction to the main cavity. The main cavity consists of a cylinder with three tapered sheds. The ring gate and restriction are meant to induce axisymmetric filling of the mold. Because of mold symmetry, modeling only one half of the mold sufficed, thus reducing computation times.

The 200S mesh contains 704 elements, and 11 grid points cover half the mold thickness; owing to symmetry of the temperature problem with respect to the midplane, only half the mold thickness needed to be modeled. Computation takes 3 h of CPU-time on a Silicon Graphics Challenge computer (without parallel processing).

The injection flow rate is assumed to be constant at $1.64 \times 10^{-6} \text{ m}^3/\text{s}$, although it was reported to fluctuate during mold filling. The polymer enters the mold at 120°C; the mold itself is at 195°C.

Results

The advancement of flow fronts, as predicted from numerical simulation (Fig. 8), can be compared with short shots from experimental molding—under the assumption that the influences of compressibility and thermal expansion after injection stopped before complete filling are limited (Fig. 9). They appear to be in agreement.

Figure 10 shows that a plug flow prevails in the runner, indicating that flow resistance—and thus pressure buildup—is concentrated in a relatively narrow fluid layer near the mold wall. This layer’s temperature is close to the wall temperature of 195°C, at which the EVA45 and EVA7 compounds show comparable viscosity behavior. As a result, Fig. 11a reveals a striking resemblance between the runner pressures calculated for the EVA7 compound and the pressures measured for the EVA45 compound.

The cavity pressures (Fig. 11b) also show the progress of filling: every time the flow front reaches the outer shed edge, flow resistance increases and pressure increases rapidly, until a new shed is reached by the flow front. Such steep increases can be seen towards $t = 30 \text{ s}$, and near $t = 40 \text{ s}$.

Although weld lines are usually deduced from the advancement of flow fronts (as shown in Fig. 8), the particle tracking concept enables us to follow the advancement of weld lines by following the injection time labels of the material at the weld lines. Figure 12 shows that the weld line of the 200S insulator, which was formed in and just above the ring across the injection point, has been transported upward into the upper shed. In the molding samples, small defects were often found in the upper shed across the injection point. Unfortunately, no experimental data on conversion and temperature are available.

CONCLUSIONS

With the modeling of the RIM process and the implementation in computer programs, simulation of the filling stage of RIM products has become possible; the complex shape of a mold does not limit the use of numerical simulation. One major problem, however, that one faces when simulating RIM is material characterization and modeling. Many thermosets contain large amounts of fillers, which are known to have a major influence on viscosity. Even at low content, fillers can affect properties significantly.

The amount of filler in the original EVA compound used in this investigation was reduced to obtain a nearly unfilled compound. The viscosity behavior of this compound was described by a Carreau model (for conversion $<1\%$) and a Power Law model with conversion-dependent parameters (for conversion $\geq1\%$).
The recognition that the highly filled EVA and the “unfilled” EVA show comparable processing viscosities is the key to successful numerical simulations: it enabled us to use the viscosity model for the “unfilled” compound to simulate the filling of a complex product with the highly filled compound, which is more difficult to characterize. These simulations yielded results that agree with the experimental short shots and pressure measurements that were used for verification. Thus, this knowledge provides insight into the filling behavior, and is therefore necessary for meaningful numerical simulations of RIM mold filling. Although this conclusion may seem obvious, regrettably we have observed that adequate characterization of commercial thermosets is not always common practice. Therefore, we emphasize that thorough characterization is particularly important for commercial materials: it may disclose nontrivial behavior due to fillers and additives that affects the filling of the mold and the product end properties, which can only be analyzed by the combination with 2-D modeling.

No experimental data on conversion and temperature during mold filling were available. Although flow fronts and pressure profiles yield indirect indications of conversion, access to experimental conversion data will be necessary to test the conversion solutions of RIM simulations.

However, to further improve the results of numerical simulation of RIM, attention has to be focused on material characterization of (filled) thermosets. Other improvements may be obtained by focusing on the initial and boundary conditions of RIM: the material temperature history upon injection, and possible slip at the mold walls have been neglected entirely, although they could be important in mold filling.

ACKNOWLEDGMENT

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SYMBOLS

\( a_T \) = Horizontal shift factor.
\( b_T \) = Vertical shift factor.
\( c_p \) = Specific heat capacity (at constant pressure), \( \text{J/kg} \cdot \text{K} \).
\( C \) = Mold compliance, \( \text{Pa}^{-1} \).
\( C_0 \) = Initial concentration of reactive groups, \( \text{mol/m}^3 \).
\( D \) = Diffusion rate, \( \text{m}^2/\text{s} \).
\( D_f \) = Deformation rate tensor, \( \text{s}^{-1} \).
\( E_a \) = Activation energy, \( \text{J/mol} \).
\( f \) = Specific external forces, \( \text{m/s}^2 \).
\( F_r \) = Froude number = inertia forces/gravity forces.
\( h \) = Mold thickness, \( \text{m} \).
\( h_t \) = Heat flux, \( \text{W/m}^2 \).
\( h_r \) = Reaction heat, \( \text{J/kg} \).
\( I \) = Unit tensor.
\( k_1 \) = Pre-exponential factor, \( \text{s}^{-1} \).
\( n \) = Power law exponent or reaction order.
\( n \) = Normal vector.
\( p \) = Hydrostatic pressure, \( \text{Pa} \).
\( p_0 \) = Thermodynamic pressure, \( \text{Pa} \).
\( r \) = Radiation heat, \( \text{W/kg} \).
\( R \) = Universal gas constant = 8.314, \( \text{J/mol} \).
\( R_c \) = Reaction rate, \( \text{s}^{-1} \).
\( Re \) = Reynolds number = inertia forces/viscous forces.
\( S \) = Fluidity.
\( t \) = Time, \( \text{s} \).
\( T \) = Temperature, \( \text{K or °C} \).
\( \bar{v} \) = Velocity, \( \text{m/s} \).
\( X \) = Conversion.

Greek Symbols

\( \alpha \) = Thermal expansion coefficient, \( \text{K}^{-1} \).
\( \beta \) = Reaction shrinkage coefficient, \( \text{m}^2/\text{mol} \).
\( \dot{\gamma} \) = Shear rate, \( \text{s}^{-1} \).
\( \dot{\gamma}_r \) = Reduced shear rate = \( \dot{\gamma} \cdot a_T \), \( \text{s}^{-1} \).
\( \eta \) = Shear viscosity, \( \text{Pa} \cdot \text{s} \).
\( \eta_d \) = Dynamic viscosity, \( \text{Pa} \cdot \text{s} \).
\( \eta_s \) = Steady-state viscosity, \( \text{Pa} \cdot \text{s} \).
\( \eta_0 \) = Zero reduced viscosity, Pa \cdot s.
\( \kappa \) = Isothermal conductivity, W/(m \cdot K).
\( \lambda \) = Thermal conductivity, W/(m \cdot K).
\( \lambda_0 \) = Relaxation time, s.
\( \lambda \) = Thermal conductivity tensor, W/(m \cdot K).
\( \mu \) = Bulk viscosity, Pa \cdot s.
\( \xi \) = Label vector.
\( \rho \) = Density, kg/m\(^3\).
\( \sigma \) = Cauchy stress tensor, kg/m\(^3\).
\( \omega \) = Frequency, rad/s.

Other Symbols

\( \nabla \) = Gradient operator, m\(^{-1}\).
\( \dot{\alpha} \) = Material time derivative of \( \alpha \).
\( A^c \) = Conjugate of tensor \( A \).
\( A^d \) = Deviatoric part of tensor \( A \).
* = With respect to midplane.

REFERENCES


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