Constitutive characterisation of rubber blends by means of genetic algorithms

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ABSTRACT: In this contribution a new procedure for parameter identification for the material characterisation of rubber blends is proposed which is based on genetic algorithms. Such a model has to consider the effect of wall slippage on the viscous properties as well as the identification of a new material parameter regarding die swell. As basis serves an experimental investigation of the viscous properties of rubber blends by means of a capillary-viscometer. Because of the consideration of wall slippage, the temperature and the die swell the resulting material characterisation is represented by a coupled system of nonlinear equations. Describing their solution requires a numerical integration algorithm. For this purpose a genetic algorithm has been adopted. The verification of the developed parameter identification was done by means of comparison with the classical material characterisation based on correction methods.

1 INTRODUCTION

Capillary rheometry simulates polymer extrusion in a simplified way. It allows the characterisation of polymers by means of determination of the viscosity function and extrudate swell. The task of the present material characterisation is to determine all required state variables which describe the flow situation of a die through a circular capillary.

For the description of the viscoelastic material behaviour of rubber blends the power law by (Ostwald 1925) is used. Its application to the investigated rubber blends is possible for a common interval $1\ \mathrm{s}^{-1} < \dot{\gamma} < 100{,}000\ \mathrm{s}^{-1}$ of the shear rate. The coupled state variables are:

- material parameters of the power law,
- pressure loss according to viscoelastic properties,
- shear stress at the wall of the capillary,
- shear rate at the wall of the capillary and
- wall slippage velocity along the capillary.

If these state variables are known, the determination of the shear rate dependent viscosity is possible.

Two types of rubber blends are used for the material characterisation, with Ethylene propylene diene rubber (EPDM) as the main component of both:

- EPDM 1: This blend has amorphous properties and is characterised by a strong die swell.
- EPDM 2: This blend has crystalline properties and is characterised by a small die swell.

The investigation of other rubber blends is planned for the near future.

This paper is structured as follows: In Chapter 2 a short description of the used measuring methods for the die swell is given. In Chapter 3 the classical material characterisation by means of correction methods is summarised.

Chapter 4 refers to the new material characterisation. Because of the coupling between viscosity and shear rate and the consideration of die swell the adaptation of an optimization method is required.

The verification of the new material characterisation is given in Chapter 5 by comparing the results of the different material characterisations methods. The paper will be completed by conclusions and some present research activities.

2 EXPERIMENTAL INVESTIGATION

In order to characterise the investigated materials various experiments by means of a capillary-viscometer with different capillary lengths L, capillary radii R and extrudate temperatures T were performed.

In addition to the extrusion pressure p and the appropriate stamp velocity v, the die swell area-ratio χ of all experiments was determined, too. In this chapter different methods for measuring die swell are explained and compared.

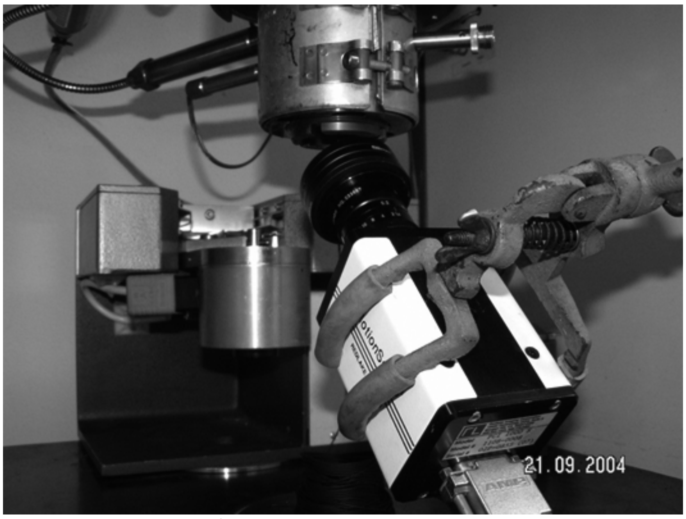

Figure 1. Position of the high speed camera.

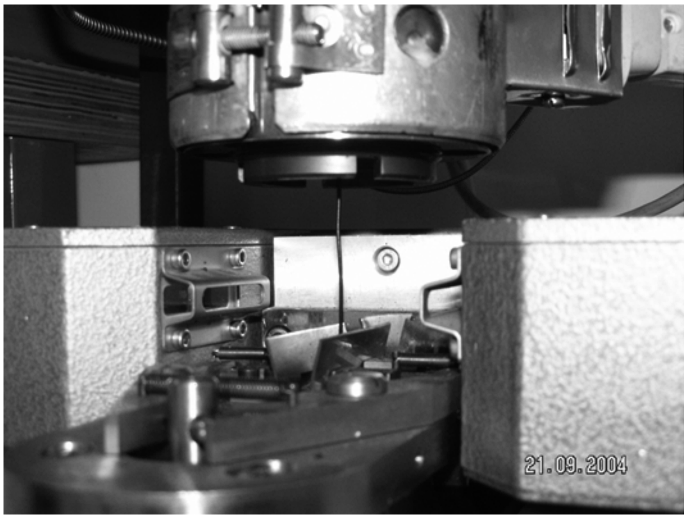

Figure 2. Swell value measuring unit.

2.1 *Adaptation with high speed camera PCI 1000*

Because the highest exit speed of a die through a capillary is up to 1000 mm/s, the shooting of the exit of capillaries cannot be investigated with common cameras. With the used high speed camera 1000 photographs per second can be taken. By using a reference wire with a known diameter the swell diameter can be determined by comparing the corresponding photographs. Figure 1 shows the situation of the camera in front of the exit of the capillary.

2.2 *Microscopical investigations*

An effective method to determine the diameter of a strand without consideration of time effects is the measurement with a microscope. Because no time dependent properties are considered this method is only for validation and not for characterisation.

2.3 *Swell value measuring unit*

The fastest method to determine the diameter of a strand is the usage of the so-called swell value measuring unit. It measures the diameter by means of a laser beam with a wave-length of approximately 700 nm. In this contribution the dimensionless swell value represents a strand cross-sectional area ratio. It is defined as follows:

$$\chi = \left(\frac{d}{D}\right)^2 \tag{1}$$

where d is the diameter of the measured strand and D is the capillary-diameter. There are two different ways of determining the swell value of a die:

- Dynamic swell value: After reaching a stationary state in the capillary the extrusion pressure p and the swell value χ_{dyn} are measured.
- Static swell value: After p is determined the stamp feed stops and the relaxation of the strand is noticed. After reaching a static value the so-called static swell value χ_{sta} is measured.

The influence of the dead weight of a die on the swell diameter is considered. By cutting the strand directly below the measuring point, the influence of the dead weight is minimized for the static swell value. Thus, it is used for the following investigations. Figure 2 shows the situation of the swell value measuring unit under the exit of the capillary.

2.4 *Comparison of the different methods*

Figure 3 collects the results of all measuring methods. Comparing these results leads to a successful validation of the swell value measuring tool. Furthermore, the die swell is independent of the shear rate and the stamp velocity respectively.

3 FORMER MATERIAL CHARACTERISATION

In order to determine viscous properties of rubber blends experiments with a capillary-viscometer are necessary. Due to the nonlinear coupling of the viscosity η and the shear rate $\dot{\gamma}$ correction methods are used to determine these material properties. First, Newtonian material properties are assumed. The resulting physical values are called apparent and marked with a subscript *"ap"*. Second, the apparent values will be transformed into true values using correction methods.

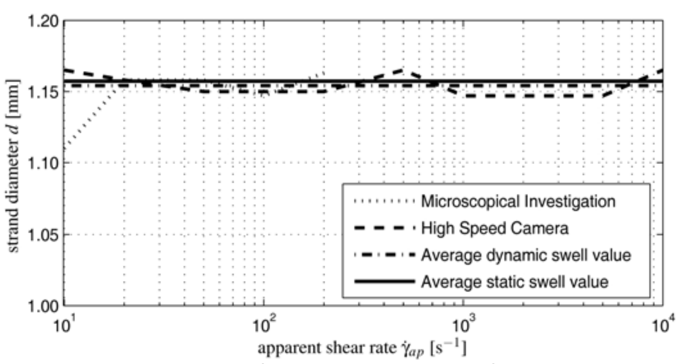

Figure 3. Comparison of measuring methods for the swell diameter d for an EPDM based rubber blend for the capillary-viscometer experiment with $L/D/T$ = 40 mm/1 mm/100 °C.

In Sections 3.1 and 3.2 various correction methods are briefly described:

3.1 *Pressure loss because of viscoelastic properties*

The measurement of the pressure is performed before the capillary entry. Therefore, entrance and outlet pressure losses are not considered by such a measurement. To overcome this problem (Bagley 1957) proposed to use an effective capillary length instead of the existing one.

In order to reduce the number of necessary experiments (Gleißle 1988) developed a method which related the pressure loss p_E with the normal-stress coefficient Θ. Thus, the advantage of using only one experiment has to be paid with the introduction of two more material parameters.

3.2 *Determination of the true shear rate*

In order to determine the true viscosity curve the calculation of the true shear rate is necessary. For this task, the empiric correction method by (Rabinowitsch 1929) is used.

The most important condition for using correction methods is the condition of adhesion of the flow on the wall of the capillary viscometer. However, wall adhesion did not occur in case of the investigated rubber blends. Therefore, the development of a new material characterisation method which considers wall slippage is required.

3.3 *Generalised Newton-Raphson procedure*

Because of the consideration of wall slippage and the coupling between viscosity and shear rate the resulting characterisation is represented by a coupled system of nonlinear equations. Describing their solution requires a numerical integration algorithm.

For this purpose (Müllner et al. 2005) adopted the Newton-Raphson-method. It requires for a certain number of unknown variables the same number of conditional equations. The variables are the coupled state variables listed in Chapter 1. A disadvantage of this method is the fact that the derivatives of the conditional equations are required.

4 ALGORITHMIC TREATMENT

4.1 *Basis concept of genetic algorithms*

Genetic algorithms are non-deterministic stochastic optimisation methods that utilize the theories of evolution and natural selection to solve a problem within a complex solution space.

They differ from other conventional optimisation approaches:

– They work with an encoding of the design variables set, not the design variables themselves.
– They search from a population of points in the problem domain, not a single one.
– They use payoff information as the objective function rather than the derivatives of the problem or other auxiliary knowledge.
– They use probabilistic transition rules based on fitness rather than deterministic one.

Genetic algorithms have demonstrated remarkable merits over gradient-based optimization methods in widespread engineering areas.

A complete description of genetic algorithms can be found in (Goldberg 1989). Genetic algorithms consist of the following most important elements:

4.1.1 *Representation*

Usually, individuals of genetic algorithms are represented as binary strings. In the context of the present material characterisation, an individual represents a set of parameters.

According to (Gupta & Sexton 1999) the basic sequence of a genetic algorithm is the same for binary-coded individuals and for individuals represented as a set of real numbers. Hence, the use of binary-coded individuals is not necessary. Following (Pichler 2004) the parameters of each individuals are converted in strings $a = [a_1, a_2, \ldots, a_k]$ of real numbers $a_j \in [0, 1]$. The used relation between a string a_j and the original parameter x is:

$$x = x_{\min} + \frac{x_{\max} - x_{\min}}{2^n - 1}\left(\sum_{j=1}^{n} a_j 2^{j-1}\right), \tag{2}$$

where n is the string length, while $x_{\min}$ and $x_{\max}$ denote the prescribed lower and upper limit of the parameter value.

4.1.2 *Reproduction*

Individuals are selected probabilistically and put in a so-called mating pool. Hereby, the probability to be selected to build up the new generation is higher for individuals characterised by an above-average fitness. Individuals with a below-average fitness will die out.

4.1.3 *Crossover and Mutation*

These basic operators are the most important two aspects to influence the performance of genetic algorithms.

In general, Crossover needs two individuals, called parents, to create two individuals of the new generation, whereas Mutation only needs one. The goal of Crossover and Mutation are diverse: Crossover makes the offspring inherit some genes from each parent; Mutation serves to introduce diversity in the generation. Figure 4 shows the operations by means of one generation with four individuals.

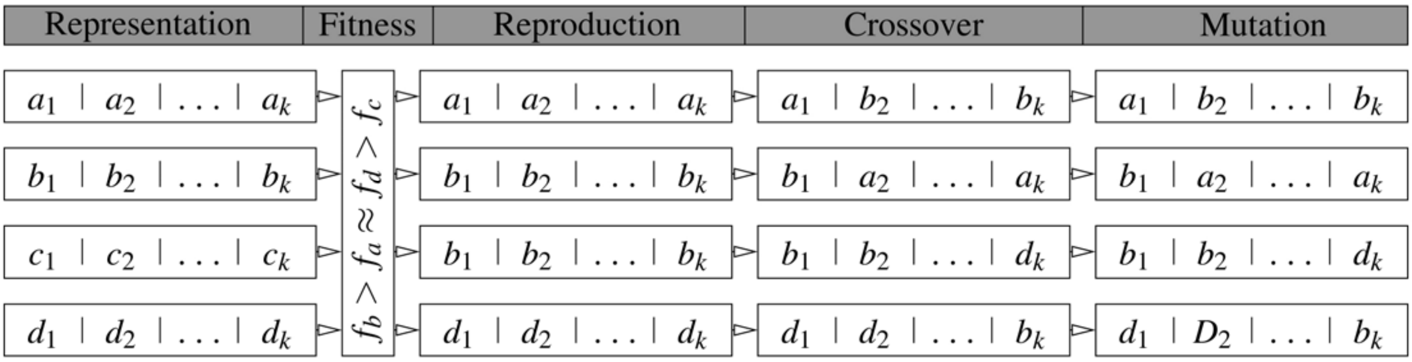

Figure 4. Basic operations of a genetic algorithm demonstrated by means of a generation with four individuals.

4.2 *Evaluation of the fitness*

The quality, i.e., the fitness of each individual of a certain generation is described by the respective value of the fitness function *f*. The fitness is the inverse value of the sum over the squared deviations between measured static swell values χ_{sta} and computed swell values χ. Let *j* denote the number of investigated stamp velocities per filling of the testing chamber of the capillary-viscometer. With this denotation, the fitness is computed as:

$$f = \left\{ \sum_{i=1}^{j} \left(\chi_i - \chi_{sta,i} \right)^2 \right\}^{-1} . \tag{3}$$

For the computed swell values the onset of (Tanner 1970) is used. This onset has been enlarged by (Han 1976) for finite-long capillaries. In order to make this law applicable for rubber blends, it is necessary to identify a new material parameter δ. For $\delta = 8$ the original onset is obtained. Hence, the swell value is computed as:

$$\chi = \left(\frac{d}{D} \right)^2 = \left(1 + \frac{D}{L} \right)^2 \sqrt[6]{1 + \frac{\Delta\sigma^2}{\delta \tau^2}} . \tag{4}$$

The normal-stress difference $\Delta\sigma$ according to (Gleißle 1988) depends on the normal-stress coefficient Θ which can be obtained by means of the viscosity curve:

$$\Theta = 2 \int_{\eta_\infty}^{\eta(\kappa/\dot{\gamma})} \frac{d\eta}{\dot{\gamma}} , \tag{5}$$

where κ denotes the so-called shift factor. In order to solve the integral (Leblans et al. 1985) accomplished a substitution and received the following equation for the normal-stress coefficient:

$$\Theta = -2 \int_{\dot{\gamma}/\kappa}^{\infty} \frac{1}{\dot{\gamma}} \frac{\partial \eta(\dot{\gamma})}{\partial \dot{\gamma}} \, d\dot{\gamma} . \tag{6}$$

Using the power law by (Ostwald 1925) for $\eta(\dot{\gamma})$ the partial derivative in (5) is obtained as:

$$\frac{\partial \eta(\dot{\gamma})}{\partial \dot{\gamma}} = \frac{\partial}{\partial \dot{\gamma}} \left(k \, \dot{\gamma}^{n-1} \right) = k \, (n-1) \, \dot{\gamma}^{n-2} . \tag{7}$$

The solution of (6) yields to a correlation between the normal-stress coefficient and the shear rate as:

$$\Theta_i = \frac{2\,k}{\kappa^{n-2}} \frac{n-1}{n-2} \dot{\gamma}_i^{\,n-2} . \tag{8}$$

Thus, the normal-stress difference is calculated as:

$$\Delta\sigma_i = \Theta_i \dot{\gamma}_i^{\,2} = \frac{2k}{\kappa^{n-2}} \frac{n-1}{n-2} \dot{\gamma}_i^{\,n} . \tag{9}$$

Using the power law for shear stresses and the force balance in a circular capillary $\dot{\gamma}^n$ is obtained as:

$$\tau_i = k \, \dot{\gamma}_i^{\,n} \Rightarrow \dot{\gamma}_i^{\,n} = \frac{\tau_i}{k} = \frac{\left(p_i - p_{E,i} \right) R}{2\,k\,L} . \tag{10}$$

Combining (9) and (10) with $p_E = q \, \Delta\sigma$ leads to:

$$p_{E,i} = \frac{R \, q \, (n-1)}{\kappa^{n-2} \, L \, (n-2) + R \, q \, (n-1)} \, p_i . \tag{11}$$

With this equation for the pressure loss p_E due to the viscoelastic behaviour of rubber blends the shear stress τ can be obtained as:

$$\tau_i = \frac{R}{2} \frac{\kappa^{n-2} \, (n-2)}{\kappa^{n-2} \, L \, (n-2) + R \, q \, (n-1)} \, p_i . \tag{12}$$

Using (9) and (12) the relation for the swell value (4) can be written as:

$$\chi_i = \left(\frac{d}{D} \right)^2 = \left(1 + \frac{D}{L} \right)^2 \sqrt[6]{1 + \frac{1}{\delta} \left(\frac{2}{\kappa^{n-2}} \frac{n-1}{n-2} \right)^2} \cdot \tag{13}$$

Thus, an equation for the evaluation for the fitness is obtained which exclusively depends on material parameters. Those will be explained in Section 4.3.

4.3 *Available material parameters*

For the proposed material characterisation by means of a genetic algorithm the following material parameters have to be determined (In addition, the used search intervals for the identification of the material parameters are given.):

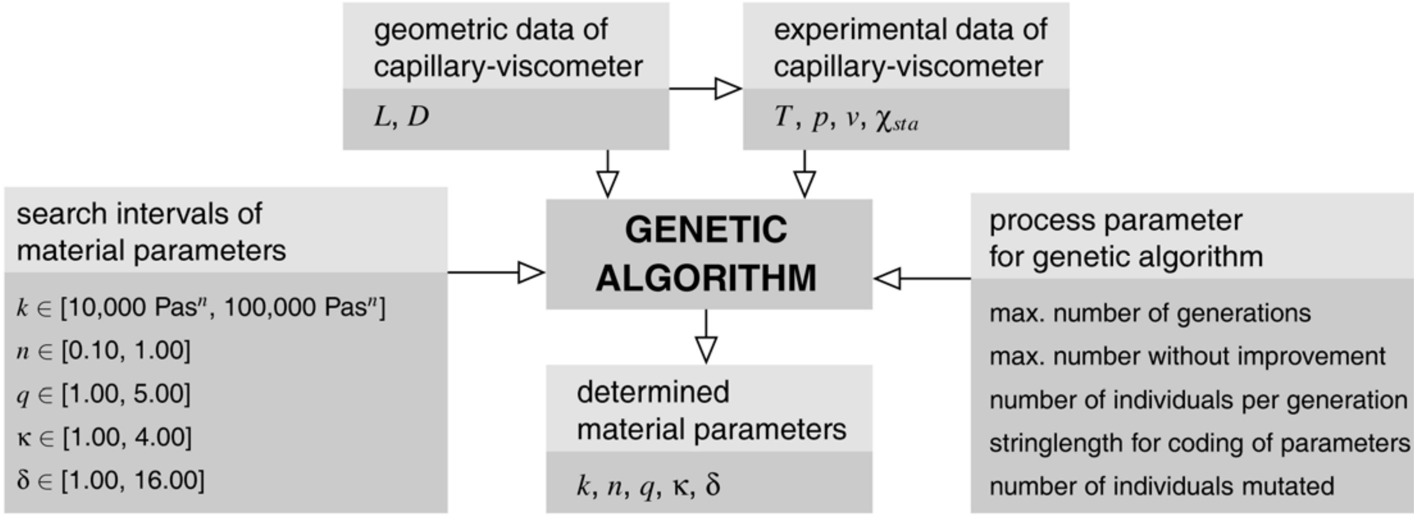

Figure 5. Required data for the usage of the genetic algorithm.

- Consistency factor k: The consistency factor is the parameter of the power law which determines the location of the viscosity curve in a viscosity-shear rate diagram. The limits of k according to the previous material characterisation are 10,000 Pas^n < k < 100,000 Pas^n.
- Viscosity exponent n: The viscosity exponent is the parameter of the power law which determines the gradient of the viscosity curve in a viscosity-shear rate diagram ($0.10 < n < 1.00$).
- Pressure factor q: The elastic pressure loss p_E is calculated by means of the normal-stress difference $\Delta\sigma$ and the pressure factor q. The latter is necessary when using the correction by (Gleißle 1988) instead of the correction by (Bagley 1957). Thus, only one experiment is required to determine p_E ($1.00 < q < 5.00$).
- Shift factor κ: The shift factor describes the adjustment of the shear rate $\dot{\gamma}$ when calculating the normal-stress coefficient Θ by means of the viscosity and the shear rate ($1.00 < \kappa < 4.00$).
- Swell parameter δ: The swell parameter makes the onset of (Tanner 1970) for the investigated rubber blends applicable ($1.00 < \delta < 16.00$).

By contemplating Newtonian properties ($n = 1$) the equations in Section 4.2 yields to the following well known relation for the wall shear stress:

$$\tau_i = \frac{R}{2\,L}\, p_i \,, \tag{14}$$

where other state variables are obtained as: $\Delta\sigma = 0$, $p_E = 0$. The swell value χ then only depends on the diameter to length ratio D / L of the used capillary.

4.4 *Application of the genetic algorithm*

Figure 5 shows the required data for the usage of the genetic algorithm, where as the process parameters are other input parameter which refer to the control of the genetic algorithm. For more details see (Goldberg 1989).

Contemplating (13) only three (n, κ, δ) of the five existing material parameters appear in the equation for the evaluation of the fitness. This fact challenges the uniqueness of the solution of the genetic algorithm characterised by reaching an absolute maximum of the fitness in the five-dimensional solution space $\{k, n, q, \kappa, \delta\}$.

Thus, after the material parameters are obtained a detailed validation of these parameters has to be done. By computation of all state variables and a following comparison with the results of the different material characterisations of Chapter 3 this validation is achieved.

Further studies on an optimal equation for the evaluation of the fitness for better parameter identification are in progress. This improvement can be achieved by means of an additional equation regarding other experimental data.

5 RESULTS AND COMPARISON

As the genetic algorithm is not able to improve the best fitness of an individual of the present generation, it stops after the specified number of generations without improvement of the best fitness. The obtained material parameters describe all coupled state variables listed in Chapter 1 for the investigated experiments.

As an example, Table 1 shows the results for one back-calculated capillary-viscometer experiment with a capillary length L = 10 mm, a capillary radius R = 0.5 mm and a temperature of T = 100 °C.

Table 1. Material parameters for capillary-viscometer experiment $L/D/T$ = 10 mm/1 mm/100 °C.

Material parameter		Determined value
Consistency factor	k	84,247 Pas$^{0.270}$
Viscosity exponent	n	0.270
Pressure factor	q	3.48
Shift-factor	κ	2.61
Swell parameter	δ	9.98

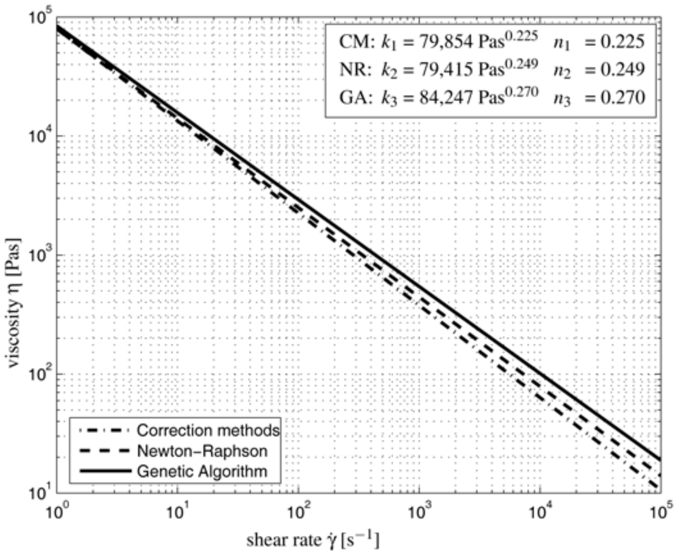

Figure 6. Comparison of the viscosity curve obtained with the different material characterisation methods for EPDM 1.

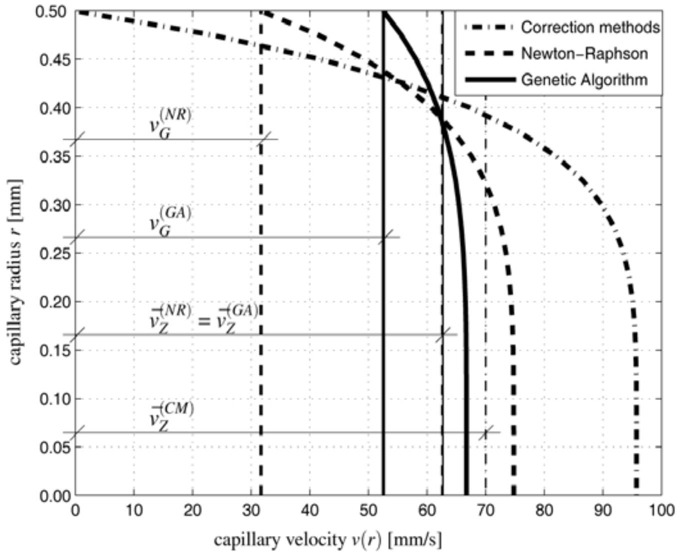

Figure 7. Comparison of the velocity profiles in a capillary obtained with the different material characterisation methods.

The validation of the results by means of the genetic algorithm is done by comparison with the results of the correction methods and the generalised Newton-Raphson procedure. The lack of the correction methods is that they do not consider wall slippage. In addition, the lack of both methods is that the parameters q and κ have to be pre-selected. Nevertheless, both methods do not consider die swell.

Figure 6 shows the viscosity curve for all three characterisation methods. Minor deviations are a consequence of considering the die swell of rubber blends. Consideration of wall slippage yields to smaller gradients of the viscosity curves.

Figure 7 shows the different obtained velocity profiles in a capillary, where $r = 0$ describes the middle of the circular capillary and $r = 0.50$ mm marks the wall of the capillary. The average velocity $\bar{v}_Z$ is equal for both the Newton procedure and the present version of the genetic algorithm. Remarkable are the flat gradients of the velocity profiles which consider wall slippage. The wall slippage velocity $v_G^{(NR)}$ represents approximately 40 % of the maximum velocity obtained by means of the Newton procedure. With the genetic algorithm a higher wall slippage velocity $v_G^{(GA)}$ is obtained. The computed swell values χ_i ($i \in \{1, \ldots, j\}$) for the mentioned example are 1.52 for all investigated stamp velocities v_i. The resulting swell diameters d_i are 1.23 mm.

6 CONCLUSIONS

Generally, the present form of the genetic algorithm can be used for both. On the one hand it can be used for back calculations of one capillary-viscometer experiment with a specified capillary length L, capillary radius R and a temperature T. On the other hand the influence of L, R and T on the present material parameter can be obtained.

The appearance of only three of the five material parameters in the main relation of the genetic algorithm leads to a challenging of the uniqueness of the obtained solution. This is because the measured pressure p has to be corrected. To eliminate this lack of the present form of the genetic algorithm viscoelastic properties have to be considered. Therefore the experimental results of a rubber process analyser will be introduced in the algorithm by means of extension of the fitness relation.

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