

# Back-Calculation of the Die Swell Phenomenon of Rubber Compounds by Means of Torsional Rheometry

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**ABSTRACT:** The die swell phenomenon of capillary experiments with various ratios of length to diameter of capillary dies is investigated. This knowledge is important for the design of injection heads for the extrusion of rubber profiles. To predict the die swell of capillary experiments, additional tests with a torsional rheometer has been performed. For polymers, the combination of such rheological data is possible under usage of the empirical relationship by Cox and Merz. With the introduction of a scaling factor this rule is also applicable for rubber compounds. With the relationship of Laun and the new scaling factor a back-calculation of the first normal stress difference is done.

Finally, the die swell of arbitrary capillary experiments for industry-used rubber compounds can be computed without introduction of new material parameters. For this task a relationship of Tanner is used, which connects the die swell and the first normal stress difference. An adaptation of the capillary viscometer allows the experimental investigation of the die swell and, thus, a validation of the presented approach. © 2008 Wiley Periodicals, Inc. *J Appl Polym Sci* 110: 76–82, 2008

**Key words:** viscoelastic fluids; capillary viscometer; rubber process analyzer

## INTRODUCTION

Extrusion heads for the fabrication of rubber profiles are up to now designed on the basis of empirical knowledge of the nonlinear viscoelastic flow behavior. Therefore, the extrusion dies were designed for every specific rubber compound. This task is mainly influenced by the swelling of the extruded material. Thus, the geometry of the sealing profile is obtained by empirical adaptation of the extrusion die. This time-consuming adaptation process affects the capacity of the running production relevantly. The disadvantages of the current process are the nonreproducible process technology and the design of dies, which is connected with long development times.

To improve this situation, a research project devoted to characterization of the die swell properties of rubber compounds has been started. The main goal of the project is the realistic consideration of die swell phenomenon in the frame of numerical simulations of injection heads and extrusion tools. The research work is jointly carried out in cooperation with Semperit Technische Produkte Ges.m.b.H., providing the material and the experimental equipment.

Although rubber compounds have similar rheological behavior, the methods for the material characterization of polymers are not always applicable for rubber compounds.<sup>1</sup> The goal of this study is to use common polymer methods for the characterization of the die swell phenomenon of rubber compounds without introducing new empiric material parameters.

For studying the swelling behavior of rubber compounds, the die swell during capillary experiments was measured. To predict the die swell, additional experiments with a rubber process analyzer was performed. For correlation between experiments with capillary- and torsional-rheometer, the Cox-Merz rule<sup>2</sup> as well as the empiric relationship for determination of the first normal stress difference according to Laun<sup>3</sup> is used. A similar approach has been used by Wen et al.<sup>4</sup> for polystyrene solutions. To predict the die swell of capillary experiments, the relationship by Tanner<sup>5</sup> is used.

## EXPERIMENTAL

### Materials

The tested materials are nonvulcanized rubber compounds used in industry, containing mainly ethylene-propylene-diene-monomer (EPDM) and carbon black and chalk as filler material. Thereby, melts with different chemical conditions of EPDM, amorphous and crystalline, and different filler degrees were investigated. Such materials are used for window sealings, pipeline constructions, bridge dilations, and various parts of cars.

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**TABLE I**  
Main Properties of the Presented Rubber Compound

<i>ML</i> (1 + 4)	41.2 ME
$t_5$	16.8 min
Shore-grade	60 ShA
Density	1.075 g/cm <sup>3</sup>
Tensile strength	12 N/mm <sup>2</sup>
Elongation at break	600%
Molecular structure	Amorphous

To allow the comparison with former research work,<sup>6</sup> the results of one low-filler content rubber compound characterized by strong die swell are presented being representative for all investigated rubber compounds.

The main material properties of the investigated materials are summarized in Table I. *ML*(1 + 4) denotes the Mooney viscosity after 4-min and 1-min preheating at a temperature of 100°C.  $t_5$  is the Scorch time at 130°C. Both quantities were determined using a Mooney viscometer MV 2000 E (manufactured by Mosonto, Missouri, MO, USA) according to the German standard DIN 53523. The hardness values are given in Shore A according to DIN 53505. Melt densities were determined according to DIN 53479. In addition, tensile tests were performed according to DIN 53504, giving access to tensile strength and elongation at break.

### Instruments and methodology

#### Experiments with capillary viscometer

The major test apparatus was a capillary viscometer manufactured by Göttfert Ges.m.b.H. (Germany). A laser-scanning swell value measuring unit was installed at the end of the capillary die. It measures the diameter of the strand immediately after its exit of the die by means of a laser beam with a wave-length of ~ 700 nm. A successful validation of this method was performed by means of a high-speed camera.<sup>7</sup>

In this contribution, the dimensionless swell value  $\chi$  represents a strand cross-sectional area ratio. It is defined as follows:

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

where  $D$  is the diameter of the capillary die and  $d$  is the diameter of the measured strand for the corresponding working point. One capillary experiment allows various piston velocities. Furthermore, a set of capillary dies with various ratios of length to diameter  $L/D$  and diameters  $D$  were used (see Table II). Experiments were carried out at temperatures of 80, 100, and 120°C.

**TABLE II**  
Geometry of the Capillary Dies Used

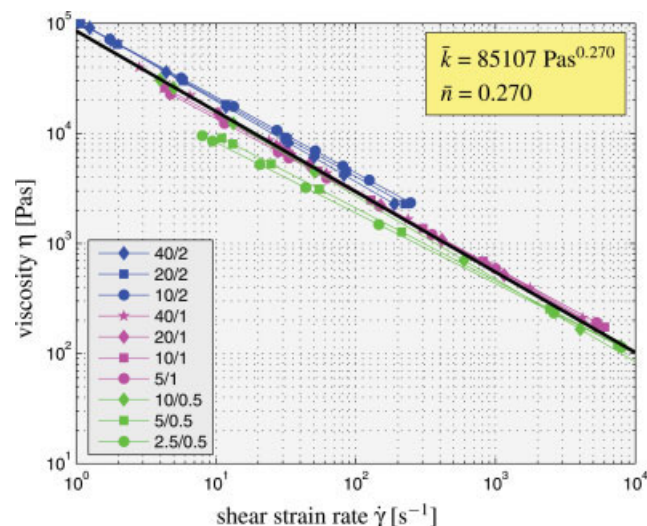
$L$ (mm)	$D = 0.5$ mm	$D = 1.0$ mm	$D = 2.0$ mm
2.5	X		
5.0	X	X	
10.0	X	X	X
20.0		X	X
40.0		X	X

#### Experiments with torsional rheometer

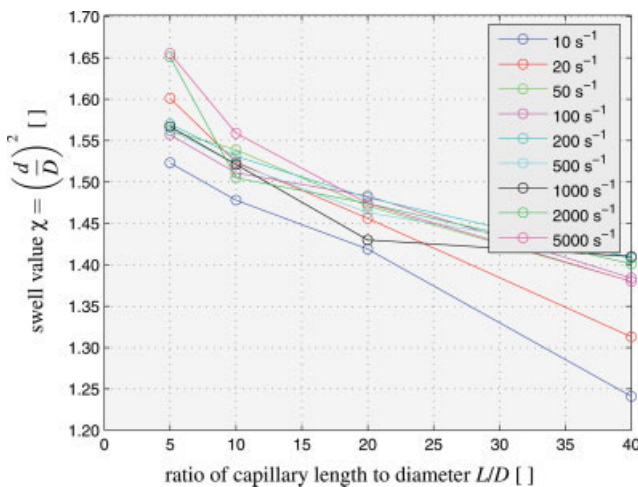
Additional experiments were performed by means of a rubber process analyzer from Alpha Technologies (Ohio, OH, USA). This cone–cone rheometer strains a sample in shear by oscillating the lower die sinusoidally. Oscillation frequency has been set from 0.1 to 30 Hz. The rubber process analyzer is designed to measure both elastic and viscous properties of elastomers and compounds. A complete description of this testing device has been summarized by Dick et al.<sup>8</sup> After a certain procedure, the storage modulus  $G'$  and the loss modulus  $G''$  are provided.

### RHEOLOGICAL CHARACTERIZATION

Because of the shear-thinning material behavior of rubber compounds, the shear rate dependence of the viscosity  $\eta$  is described by the power law after Ostwald<sup>9</sup> and de Waele.<sup>10</sup> Its application to the investigated rubber compounds is possible for a common range of the shear strain rate  $\dot{\gamma}$ . Figure 1 shows the obtained viscosity curves for all employed capillary dies according to Table II and gives the average values of the power law constants at 100°C. The power law reads as follows:



**Figure 1** Viscosity and flow curve for low-filler content rubber compound at a temperature of 100°C. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]



**Figure 2** Swell value versus ratio  $L/D$  at a temperature of  $100^{\circ}\text{C}$ . [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

$$\tau = \eta(\dot{\gamma})\dot{\gamma} \quad \text{with} \quad \eta = k\dot{\gamma}^{n-1}, \quad (2)$$

where  $k$  is the consistency factor in  $[\text{Pas}^n]$  and  $n$  is the dimensionless viscosity exponent.  $\tau$  is the shear stress at the wall of the capillary die.

Without die swell investigations, the capillary experiment is used to determine the shear stress  $\tau$  and the shear strain rate  $\dot{\gamma}$ . Till now, correction methods<sup>11</sup> are used to obtain the viscosity curve for a non-Newtonian fluid. Because of the application problems of these methods for the investigated rubber compounds, the constitutive characterization is done by means of an iteration method.<sup>12</sup> Because of the consideration of wall slippage, temperature effects, and die swell, the resulting parameter identification is represented by a coupled system of nonlinear equations. For their solution, a generalized Newton-Raphson procedure has been adopted. To minimize the number of coupled variables, a new method<sup>13</sup> was developed, which determines the pressure loss-correction due to measurement of the melt pressure in front of the die. A successful validation for this characterization method based on a genetic algorithm is summarized in Ref. 1.

All state variables in this contribution such as shear stress and shear strain rate are, therefore, related to the true nonlinear material behavior.

### DIE SWELL PHENOMENON

With the measured swell values, statements about the elastic behavior of the investigated rubber compounds are possible. From the rheological point of view, the die swell occurs as a result of the recovery of the elastic deformation imposed in the capillary die. In this chapter, the die swell of rubber compounds is described briefly. A detailed characteriza-

tion of the die swell phenomenon of rubber compounds can be found in Müllner et al.<sup>6</sup>

In Figure 2 measured swell values versus the geometry ratio  $L/D$  of the corresponding die are plotted. In the figure, exemplarily, the results of the capillary dies  $L/D = 40 \text{ mm}/1 \text{ mm}$ ,  $20/1$ ,  $10/1$ , and  $5/1$  at  $100^{\circ}\text{C}$  are depicted. This figure shows that the swell value decreases rapidly at low values of  $L/D$  and then levels off as  $L/D$  is increased further. Each line represents one working point at a constant shear strain rate  $\dot{\gamma}$ . According to Bagley et al.,<sup>14</sup> this behavior indicates that the elastic behavior of the melt depends on the residence time of the melt in the die.

For the numerical prediction of the die swell, information concerning the normal stress state in the capillary are required. As reported by Han,<sup>15</sup> the die swell depends on the recoverable shear strain as follows:

$$\gamma_{\text{el}} = \frac{N_1}{2\tau} = \frac{\sigma_{11} - \sigma_{22}}{2\tau}, \quad (3)$$

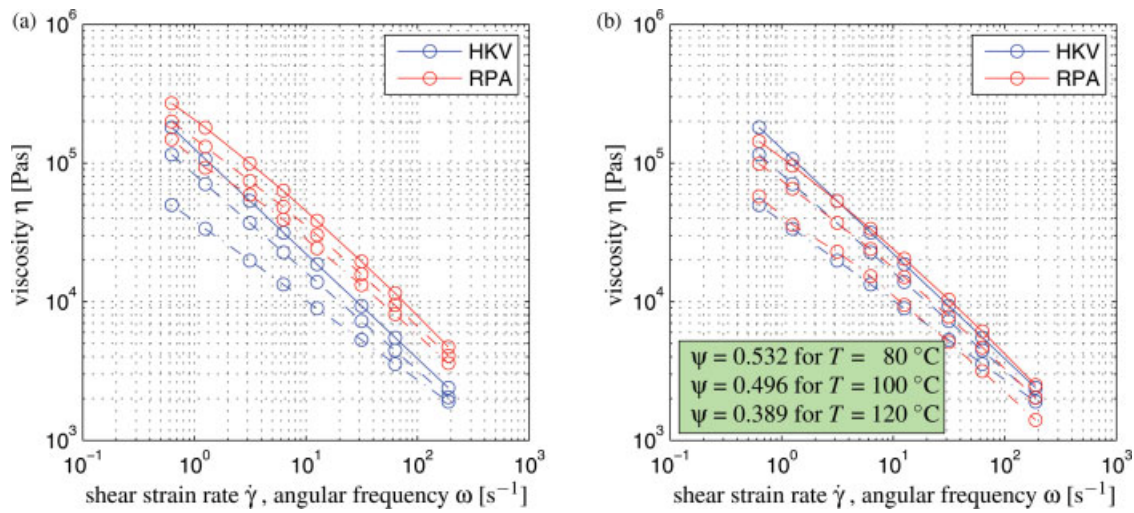
where  $N_1$  is the first normal stress difference in the die. It is well-known that for describing the die swell only the difference between the first and the second component of the normal stress tensor must be known. In this context,  $\sigma_{11}$  represents the normal stress in the flow direction and  $\sigma_{22}$  represents the normal stress in the direction of velocity change.<sup>16</sup> For determination of the recoverable shear strain, the determination of the shear stress  $\tau$  with capillary experiments is only possible. The first normal stress difference has to be determined by means of additional experimental data, e.g., experiments with a rubber process analyzer.

### CONDITION FOR AN ADMISSIBLE COMBINATION OF EXPERIMENTAL DATA

To use the experimental results from a torsional rheometer for the prediction of die swell data from capillary experiments, a comparison of the viscosity functions obtained from both experiments is required.<sup>3</sup> Therefore, an assumption concerning the investigated working points of both experiments is necessary. Working points of the capillary experiment are characterized by piston velocities, and working points of the experiments with the rubber process analyzer by angular frequencies.

For the described objective, a rule by Cox and Merz<sup>2</sup> can be used. This condition establishes a relationship between the absolute value of the complex viscosity  $|\eta^*(\omega)|$  and the stationary dynamic viscosity  $\eta(\dot{\gamma})$  obtained from continuous shear. The Cox-Merz relationship is defined as follows:

$$|\eta^*(\omega)| = \eta(\dot{\gamma}) \quad \text{for} \quad \dot{\gamma} = \omega. \quad (4)$$



**Figure 3** Comparison of viscosity curves obtained by means of a capillary experiment as well as an experiment with a rubber process analyzer (a) without and (b) with scaling factor  $\psi$  for different temperatures. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

This means that the absolute value of the complex viscosity corresponds with the stationary viscosity, when the numerical value of the angular frequency  $\omega$  is equal to the shear strain rate  $\dot{\gamma}$ . For many polymers, the validity of this relationship has been proven.

The validity of this relationship for the investigated rubber compounds is proven in this chapter. The absolute value of the complex viscosity is defined as follows:

$$|\eta^*| = \frac{|G^*|}{\omega}, \quad (5)$$

where

$$|G^*| = \sqrt{G'^2 + G''^2} \quad (6)$$

is the complex shear modulus obtained using storage modulus  $G'$  and loss modulus  $G''$  of an experiment with a rubber process analyzer of the same material at the same temperature.

Figure 3(a) shows the viscosity curves obtained using storage and loss modulus,  $|\eta^*(\omega)|$ , as well as the viscosity curves obtained by means of a capillary experiment,  $\eta(\dot{\gamma})$ . The full lines were obtained at a temperature of 80°C, the dashed lines at 100°C, and the dash-dotted lines at 120°C. In the figure, the results of the capillary die  $L/D = 40/2$  are shown.

Figure 3(a) shows that the Cox-Merz relationship is not applicable to the investigated rubber compounds. However, the slope of the viscosity curves is portrayed correctly. Therefore, the different viscosity curves can be correlated by introducing a scaling factor  $\psi$ . This factor is defined as

$$\psi = \frac{1}{s} \sum_{r=1}^s \frac{\eta(\dot{\gamma}_r = \omega_r)}{|\eta^*(\omega_r)|}, \quad (7)$$

where the dynamic viscosity is determined for the corresponding value of the angular frequency as follows:

$$\eta(\dot{\gamma} = \omega) = k \omega^{n-1}. \quad (8)$$

Using (8) for the dynamic viscosity and (5) for the complex viscosity, one can simplify eq. (7) as

$$\psi = \frac{k}{s} \sum_{r=1}^s \frac{\omega_r^n}{|G_r^*|}. \quad (9)$$

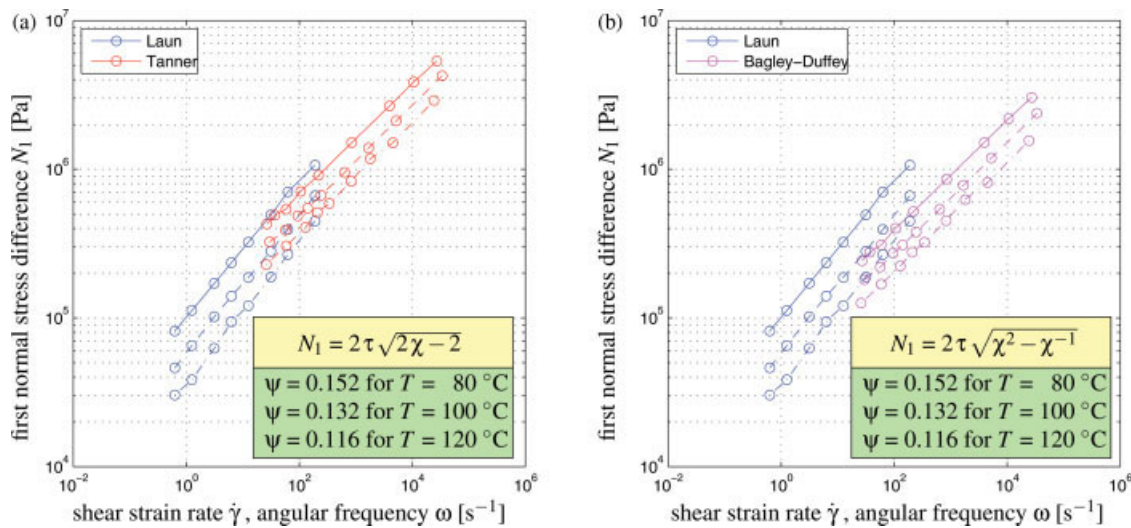
The power law constants  $k$  and  $n$  have already been obtained using the data of the corresponding capillary experiment and the material characterization according to Müllner et al.<sup>6</sup> The scaling factor is constant for all investigated working points. Under usage of the scaling factor, the Cox-Merz relationship is applicable for rubber compounds as

$$|\eta^*(\omega)|\psi = \eta(\dot{\gamma}) \quad \text{for } \dot{\gamma} = \omega. \quad (10)$$

The obtained correspondence between the viscosity curves is shown in Figure 3(b), e.g., for the capillary die  $L/D = 40/2$ . The obtained scaling factors  $\psi$  decrease with increasing melt temperature. The respective scaling factor for each investigated material is obtained by performing both experiments with capillary viscometer and torsional rheometer and by evaluating eq. (9).

### CALCULATION OF THE FIRST NORMAL STRESS DIFFERENCE

For various polymers, Laun<sup>3</sup> found an empirical equation for determination of the first normal stress



**Figure 4** Validation of the relationships by (a) Tanner<sup>5</sup> and (b) Bagley and Duffey<sup>17</sup> by means of comparison between empirical equation by Laun<sup>3</sup> and swell value measurements. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

difference. With a proven validity of the Cox-Merz relationship for any material, the normal stress state can be obtained with this empirical equation.

By introducing the scaling factor,  $\psi$ , the Cox-Merz relationship can be used for the investigated rubber compounds. Under consideration of the scaling factor according to (9), the first normal stress difference in a capillary die can be gathered from

$$N_1 = 2\psi G' \left[ 1 + \left( \frac{G'}{G''} \right)^2 \right]^{0.7}. \quad (11)$$

Wen et al.<sup>4</sup> have chosen another approach of using the original version of eq. (11). By variation of the power law index 0.7 in (11), the application of this empirical equation has been proven for their polymers. This approach is not used for the investigated rubber compounds to minimize the number of unknown material parameters.

The validation of (11) for the herein considered rubber compounds is done by means of a comparison with the results of the experimental determination of the die swell. For this purpose, a connection between die swell measurements and first normal stress difference is required. With the relationship by Tanner<sup>5</sup>

$$N_1 = 2\tau\sqrt{2\chi^3 - 2} \quad (12)$$

and the relationship by Bagley and Duffey<sup>17</sup>

$$N_1 = 2\tau\sqrt{2\chi^2 - \chi^{-1}} \quad (13)$$

the first normal stress difference is obtained using the experimental data of the die swell of the corresponding capillary experiment. As basis for these observations eq. (3) is used.

In Figure 4 the results of the validation of the first normal stress difference are shown, under usage of the experimental data for the capillary die  $L/D = 2.5/0.5$ . As in Figure 3, the results of different temperatures are shown. The empirically determined first normal stress difference correlates better with the relationship of Tanner<sup>5</sup> as shown in Figure 4(a). However, this comparison leads to a sufficient validation for the combination of capillary experiments and experiments with a rubber process analyzer.

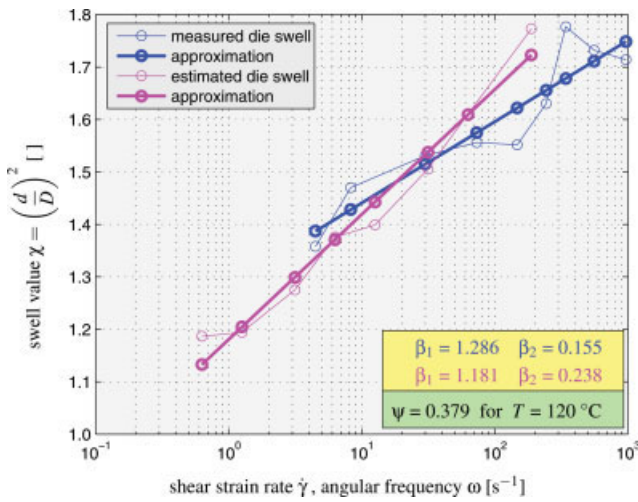
With the described approach no further material parameters are required. The scaling factor  $\psi$  is automatically determined when comparing the experimental data of both experiments. According to Müllner et al.,<sup>7</sup> the relationship of Tanner<sup>5</sup> can be used for the characterization of rubber compounds.

#### CALCULATION OF THE DIE SWELL USING RPA-EXPERIMENTS

It has been shown that the first normal stress difference  $N_1$  of rubber compounds can be calculated by means of the empirical equation by Laun.<sup>3</sup> With the relationship of Tanner,<sup>5</sup> the required connection between  $N_1$  and the die swell of capillary experiments is achieved. Dissolving (12) for the swell value  $\chi$  leads to

$$\chi = \sqrt[3]{1 + \frac{1}{2}\gamma_{el}^2} = \sqrt[3]{1 + \frac{1}{2}\left(\frac{N_1}{2\tau}\right)^2}, \quad (14)$$

where the dependence of the die swell on the recoverable shear strain is considered according to (3). The relationship by Tanner<sup>5</sup> is originally valid for infinite-long capillary dies and has been enlarged by Han<sup>15</sup> for finite-long dies as



**Figure 5** Calculated and measured swell values, shown exemplarily for the capillary die  $L/D = 20/1$  at  $120^\circ\text{C}$ . [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

$$\chi = \left(1 + \frac{D}{L}\right)^2 \sqrt[3]{1 + \frac{1}{2} \left(\frac{N_1}{2\tau}\right)^2} \quad (15)$$

Using (11) for  $N_1$  and  $\tau$  according to the material characterization mentioned in Chapter 3, one can calculate the die swell for the corresponding capillary experiment without swell value measurements.

To compare the results from different approaches, a regression analysis for the swell value similar to Liang<sup>18</sup> is done as follows:

$$\chi_{\text{calc}} = \beta_1 + \beta_2 \lg \dot{\gamma}, \quad (16)$$

where  $\beta_1$  and  $\beta_2$  are constants characterizing the elastic properties of the material. Similar to the power law constants, the parameters  $\beta_1$  and  $\beta_2$  are

obtained by means of a Gaussian quadrature.<sup>6</sup>  $\beta_1$  corresponds to the swell value at a shear strain rate of  $\dot{\gamma} = 1 \text{ s}^{-1}$ .

Figure 5 shows the comparison of measured and calculated swell values for the capillary experiment  $L/D = 20/1$  at  $120^\circ\text{C}$ . The figure also contains the parameters  $\beta_1$  and  $\beta_2$  of both the measured and the back-calculated swell values according to (15).

The agreement between measured and back-calculated swell value is good, especially in the overlapping range of both approaches. The different ranges for the shear strain rate  $\dot{\gamma}$  and the angular frequency  $\omega$  occur because of the comparison of different experiments.

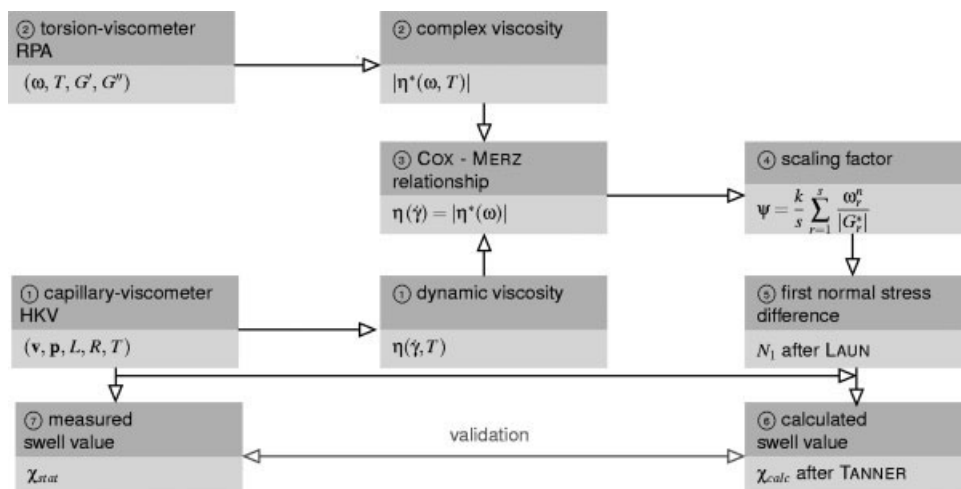
Comparing the scale of the die swell on the ordinate of Figure 5, a value of  $\Delta\chi = 0.1$  corresponds with the changing of the strand diameter  $\Delta d = 0.046 \text{ mm}$  for the employed capillary die  $L/D = 20/1$ .

The investigations of other rubber compounds have shown that the described procedure is not applicable for all capillary experiments. If the scaling factor reaches a value of  $\psi < 0.1$ , no satisfying agreement for the first normal stress difference is obtained.

However, it was one goal of this contribution to introduce no new material and process parameters, respectively, for the prediction of the measured die swell of the investigated rubber compounds.

### CONCLUSIONS

The published methods for the constitutive characterization and the identification of the swelling behavior are applicable for polymers. For rubber compounds used in industry, these methods cannot be used always. With a scaling factor, application of the Cox-Merz relationship for rubber compounds is possible. In the context of die swell phenomenon,



**Figure 6** Used approach for the back calculation of the die swell of one capillary experiment and its validation.

this contribution deals with the combination of experimental data of two standardly used experimental devices. The scaling factor allows the computation of the die swell of rubber compounds under usage of the relationships of Laun<sup>3</sup> for the calculation of the first normal stress difference and of Tanner<sup>5</sup> for the prediction of swell values. The chosen approach is shown in Figure 6.

The application of these equations for the investigated rubber compounds was successfully proven by means of the measurements of swell values after the exit of a capillary die. These measurements allow prediction of the swelling behavior of the used rubber compounds for fabrication of rubber profiles made by extrusion.

The presented method does not require any material parameters, which are unknown for other materials. The scaling factor is automatically computed when using the Cox-Merz relationship. The obtained results will be used to improve the numerical simulation of die swell phenomenon and its influence on the geometry of rubber profiles.

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