Introduction

The dimensioning of injection heads for the extrusion of rubber profiles is exclusively based on empirical knowledge of the non-linear viscoelastic flow behaviour of elastomers especially of the so-called die swell. Thus, the design of injection heads is carried out subject to the used rubber blend, whereas the geometry of the appropriate profile is achieved by empirical adaptation of the extrusion die. This adaptation process is mainly influenced by the swelling after the extrusion of rubber and is carried out in several steps. The disadvantages of the current production process are the process technology which is non-reproducible over a long timeframe and the design of dies which is connected with long development time. This relevantly affects the capacity of the running production. This fact was one of the motivations for a research project concerning the characterisation of the die swell properties of rubber blends. The research work is performed in cooperation with Semperit Technische Produkte Ges.m.b.H. & Co. KG.

Experimental Investigations

The tested materials are nonvulcanised rubber blends used in industry, containing mainly EPDM and carbon black in different compositions. So far four different rubber blends were investigated. These are used for window sealings, pipeline constructions and various parts of cars.

As basis serves an experimental investigation of the viscous properties of rubber blends by means of a capillary-viscometer. This test apparatus was manufactured by Göttfert Ges.m.b.H. (Germany). A laser scanning swell value measuring unit was installed at the end of the circular capillary. It measures the diameter of the strand immediately after the exit of the capillary by means of a laser beam with a wavelength of approximately 700 nm. In this contribution the dimensionless swell value represents a strand cross-sectional area ratio. It is defined as:

\[ \chi = \left( \frac{d_i}{D} \right)^3, \]

where \( D \) is the diameter of the capillary and \( d_i \) is the diameter of the measured strand for the investigated duty point \( i \).

Rheological Characterisation

Capillary rheometry simulates polymer extrusion in a simplified way. It allows the characterisation of polymers and rubber blends, respectively, by determination of the viscosity function. For the description of the coupling between viscosity \( \eta \) and shear strain rate \( \dot{\gamma} \) the power law published in [6] is applied. Its usage for the investigated rubber blends is possible for a common interval of the shear strain rate.

The constitutive characterisation is done by means of an iteration method published in [7]. Because of consideration of wall slippage, the temperature effects and the die swell, the resulting parameter identification is represented by a coupled system of nonlinear equations. Describing their solution requires a numerical integration algorithm. For this purpose a generalised Newton-Raphson procedure has been adopted. The developed procedure avoids various application problems, which occur when common correction methods are used.

Die Swell Phenomenon of Rubber Blends

It is well-known that jets of viscoelastic liquids expand upon exiting from capillary tubes. This swelling of the extrudate on emerging from a capillary is typical for non-Newtonian viscoelastic liquids. This phenomenon is also called Barus effect [2]. With the measured swell values various statements about the elastic behaviour of the investigated rubber blends are possible [9]. From the rheological point of view the die swell occurs as a result of the recovery of the elastic deformation imposed in the capillary [4].

In this contribution all figures are related to rubber blend EPDM 1 which has compared to the other blends a small filler degree and a pronounced die swell behaviour. In order to compare the results from experiments and simulations a regression analysis for the swell value similar to [5] is performed:

\[ \chi_{\text{calc}} = \beta_1 + \beta_2 \log \dot{\gamma}, \]

where \( \beta_1 \) and \( \beta_2 \) are constants characterising the elastic properties of the material. The parameters \( \beta_1 \) and \( \beta_2 \) are obtained by means of a Gaussian quadrature. \( \beta_1 \) corresponds to the swell value at a shear strain rate of \( \dot{\gamma} = 1 \text{ s}^{-1} \).

Figure 1 shows the approximation of the swell value ex-
emplarily for capillary experiments with different ratios of capillary length to diameter, $L/D$, and a constant capillary diameter of $D = 2 \text{ mm}$ at a temperature of 120 °C. From Figure 1 it can be seen that the swell value increases with increasing shear strain rate in a logarithmic scale. According to [4] this behaviour is attributed to the increasing of the recoverable elastic energy.

In Figure 2 measured swell values versus a corresponding geometry ratio $L/D$ are plotted. In the figure, exemplarily, the results of the circular capillaries $L/D = 40/1, 20/1, 10/1,$ and $5/1 \text{ mm}$ for a temperature of 100 °C are depicted. It shows that the value of 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 duty point at a constant shear strain rate $\dot{\gamma}$. This behaviour of the die swell indicates that the elastic behaviour of the melt depends on the residence time of the melt in the capillary. According to [1], this can also be interpreted as a decaying memory of the melt.

In Figure 3 the measured swell value versus a geometry ratio $D_K/D$ is plotted, where $D_K$ denotes the diameter of the reservoir. In the figure, exemplarily, the results of circular capillaries $L/D = 20/2, 10/1,$ and $5/0.5 \text{ mm}$ for a temperature of 100 °C are depicted. It shows that the swell value increases at first with $D_K/D$ and then levels off as $D_K/D$ is increased further.

The dependence of the swell value on the ratio of reservoir diameter to capillary diameter is particularly interesting because it indicates that the amount of stored elastic energy in the melt depends on the diameter of the reservoir up to a certain value, at which the conical zone of the converging flow streamlines into the capillary becomes unaffected from the reservoir wall. This fact certifies that extrudate swell results from the recoverable elastic energy stored in the melt which originates from the large pressure drops at the entrance to the capillary. Both statements concerning Figure 2 and 3 have been observed for polymers, too [4].

The dependence of the die swell on the melt temperature has been investigated, too. Rubber blends and polymers have always the same behaviour. With increasing temperature the die swell decreases. Only for EPDM 1 a different behaviour has been investigated. Because of the amorphous structure the measured swell values increase with increasing melt temperature.

Figure 4 shows a plot of die swell ratios for EPDM 1 versus shear strain rate $\dot{\gamma}$ at various temperatures, measured during an experiment with a ratio $L/D = 40/2 \text{ mm}$. A decrease of die swell with increasing melt temperature has not been detected.
As regards the temperature dependence of the swell value, Figure 5 shows a plot of die swell ratio versus the shear stress $\tau$ at the wall of the capillary.

According to [4] a temperature dependence of this relationship is not evident. This statement has been proven for all investigated rubber blends except EPDM capillary $L/D = 40/2$ mm.

The unusual behaviour of EPDM 1 can be explained by the amorphous structure and the existence of a critical temperature [8]. When the temperature is increased the efficiency of the few crystalline crosslinks are decreased and the swelling deformation is higher causing an increase of die swell [8].

Empiric Relationship for Swell Values

As noticed in Figures 2 and 3 the die swell phenomenon depends significantly on the geometry of the used capillary as well as of the melt temperature. The goal of this chapter is to present a relationship which considers the influence of the length $L$ and the diameter $D$ of the capillary, respectively, and the melt temperature $T$.

As basic relationship equation (2) is used, where the parameters $\beta_1$ and $\beta_2$ are identified by means of the experimental swell value data:

$$\beta_1 = \beta_1(L,D,T,\mu)$$
$$\beta_2 = \beta_2(D,T,\mu).$$

where $\mu$ denotes a dimensionless material parameter. As the die swell is caused by the molecular constitution of the blends, both parameters must depend on the material parameter.

The length of the capillary $L$ is a parameter for the resistance time of the material in the capillary. The longer the melt stays in the capillary, the smaller the resulting die swell. This behaviour is often called as memory-effect. $L$ affects the absolute value of the swell value, therefore, only $\beta_1$ is influenced by $L$.

The diameter of the capillary $D$ is a parameter for the applied strain of the material. The heated material in the reservoir is pressed through the capillary. The smaller $D$, the higher the material is deformed. Due to the viscous properties the material never reaches the original state after the exit of the capillary.

In order to obtain dimensionless parameters $\beta_1$ and $\beta_2$ a reference experiment is used. Reliable results for the determination of the viscosity curve are obtained with the capillary $L/D = 10/1$ mm. Therefore this experimental configuration will be used as basis.

Using a linear regression the parameters of equation (2) are determined as:

$$\beta_1 = \left[1 + \frac{D}{L}\right] \left[1 + \frac{D}{D}\right] \left[1 + \frac{T}{T}\right]^{\mu}$$

and

$$\beta_2 = \left[1 + \frac{D}{D}\right] \left[1 + \frac{T}{T}\right]^{\mu},$$

where $D = 1$ mm and $T = 373$ K. The temperature $T$ is in [K]. The influence of the material is considered as all terms in (4) and (5) are raised to the power of the material parameter $\mu$. The exponent of the term concerning the temperature dependence in (4) and (5) leads to an increase of swelling for rising temperatures. For this behaviour values for $\mu > 0.25$ are obtained. Thus, the interaction between chemical structure and temperature dependence is considered.
Exemplarily, in Figure 6 the measured swell value for EPDM 1 for all performed capillary experiments are depicted. For clarity only the regression lines of the respective experiments are shown.

![Fig. 6: Measured swell values at $T = 120 \, ^\circ C$ for all investigated ratios of length to diameter of the capillary $L/D$](image)

Figure 7 shows the corresponding results for EPDM 1 at $T = 120 \, ^\circ C$ when using the empiric equation according to equation (2) and the parameters $\beta_1$ and $\beta_2$ according to (4) and (5).

![Fig. 7: Predicted swell values at $T = 120 \, ^\circ C$ for all investigated ratios of length to diameter of the capillary $L/D$](image)

An exact determination of the swell value for a single experiment is not possible with the present approach. The empiric equation can only consider the macroscopic behaviour of rubber blends. The influence of the parameters of the experiment $L$, $R$, $T$, and $\dot{\gamma}$ on the die swell ratio has been detected accurately.

Comparing the scale of the die swell on the ordinate of Figures 6 and 7 a value of $\Delta d = 0.1$ corresponds with the changing of the strand diameter $\Delta d = 0.023$ mm for capillaries with $D = 0.5 \, \text{mm}$, $\Delta d = 0.046$ mm for capillaries with $D = 1.0 \, \text{mm}$, and $\Delta d = 0.092$ mm for capillaries with $D = 2.0 \, \text{mm}$.

Furthermore, it was one goal of this investigation, to consider the influence of the material on the die swell by only one material parameter $\mu$. With one material parameter a better comparison of different melts is possible. With unknown $\mu$ the usage of the empiric equation is possible, too, as the length $L$ and the radius $R$ of the capillary has always the same quantitative influence on the swell value. The swell value increases for all investigated rubber blends with increasing shear strain rate $\dot{\gamma}$. For large diameters of the capillary this increase is stronger as for capillaries with small diameters.

### Influence of Extrusion on the Die Swell

In addition, the influence of extrusion of rubber on the die swell has been studied. An extrusion process leads to a rigorous reduction of the die swell [3]. In fact, extrusion yields to a molecular change of the material. Experiments with standard extrusion parameters were performed in order to prove the empiric relationship for applicability.

Analysing the experimental data for extruded materials leads essentially to the result that all principal dependencies ($L$, $R$, $T$, and $\dot{\gamma}$) keep valid. According to [3] the die swell ratio for all investigated rubber blends decrease in comparison with the original material. Due to the change of the material structure the material parameter $\mu$ must change, too.

In case for EPDM 1 a change of the material parameter to $\mu = 0.233$ occurs. Thus, the crystalline properties of the material increase and the die swell decrease. In fact, due to the mentioned change the dependence on the temperature altered, too. The swell value for extruded EPDM 1 decreases for increasing temperature.

### Conclusions

The parameter of a capillary experiment, length $L$ and radius $R$ of the used circular capillary, the melt temperature $T$ and the corresponding shear strain rate $\dot{\gamma}$ have a certain influence on the die swell of non-Newtonian fluids. In this contribution an empiric relationship for the consideration of these parameters was presented. The condition for application of this relationship is a capillary-experiment with measurement of the swell value. Such experiments are required to determine the viscosity function of rubber blends. With the empiric relationship an evaluation of performed experiments is possible. With the material parameter $\mu$ are qualitative appraisal of the experiment is achieved.

The knowledge of the influence of these parameters is required to evaluate numerical simulations of die swell phenomenon and its influence on profile geometry. This will lead to better results for the simulation of injection heads.
References
