Constitutive characterization of extruded rubber blends by means of an extrusion rheometer

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ABSTRACT: In this contribution the influence of an extrusion process on the viscosity function of rubber blends is identified. This knowledge is required for performance of realistic numerical simulations of injection heads and extrusion tools for rubber profile production. The experimental basis of this investigation are tests with both, capillary and extrusion rheometer. The latter is a combination of an industrial extruder and a circular cross-sectional extrusion tool, allowing the application of material characterization methods used for capillary experiments. Thus, determination of the viscosity function of extruded rubber blends is possible. Additional capillary experiments allow an identification of the influence of extrusion on the viscosity of rubber blends. For the material characterization a nonlinear iteration scheme is used, which is proven to be applicable for rubber compounds and rubber blends. Successful verification of extrusion rheometer tests are performed with numerical back-calculation of the corresponding pressure measurements.

1 INTRODUCTION

During rubber profile extrusion unvulcanized rubber blends are pressed through a jet with a specified geometry. For that purpose raw material is melted and homogenized due to heating and internal friction in a screw extrusion press. Before the extrusion tool is reached the melt is heated within an injection head up to the desired temperature. The production speed of this continuous process is controlled by increasing the revolutions of the screw press. After the heated rubber material leaves the extrusion tool the shape of the produced profile is guaranteed by an assembly line whose speed is insignificantly higher as the speed of the extruder. The vulcanization process is completed in a salt bath (Michaeli 2003).

Due to the viscoelastic behavior the heated rubber melt expands after exiting from capillary tubes and extrusion tools, respectively. This swelling of the extrudate on emerging from a die is typical for such materials. Thus, the shape of extrusion tools for the fabrication of rubber profiles has to consider the respective swelling phenomenon inversely. Up to now extrusion tools are designed on the basis of empirical knowledge of the non-linear viscoelastic flow behavior. Therefore, the injection heads were designed for every specific rubber blend. Thus, in rubber profile extrusion there is an increasing interest in predicting the deformation of profile shapes after leaving the tool of the extrusion line by means of numerical simulations.

Usage of finite element software allows drastic reduction of the time-consuming adaptation process for the design of extrusion tools. For this task, the influence of an extrusion process on the shear strain rate dependent viscosity has to be determined. This knowledge cannot be identified adequately by means of standard experiments, such as capillary and torsional rheometry. In order to determine the viscosity of extruded rubber blends, an extrusion tool with a cylindrical cross-section has been built. Pressure and temperature measurements within the cylindrical area are possible. Therefore, a comparison with capillary experiments is possible.

This paper is structured as follows: In Chapter 2 a short description of the used experimental devices is given. In Chapter 3 the used procedure for determination the viscosity function is briefly explained. This method is applicable for both, capillary experiments and extrusion rheometer tests.

Chapter 4 refers to a numerical validation of the extrusion rheometer. With the CFD software POLYFLOW a numerical simulation of the extrusion rheometer is performed.

In Chapter 5, on the basis of additional capillary experiments a comparison of viscosity functions of raw and extruded rubber blends is presented. It allows the identification of the influence of an extrusion process. By comparing capillary and extrusion rheometer tests, statements concerning the geometry dependence of the viscosity function are given in Chapter 6.

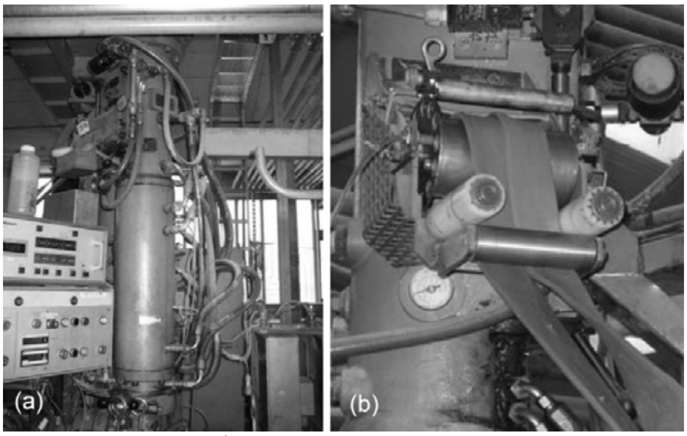

Figure 1. Extruder for industrial applications: (a) vertical extruder, (b) band supply of raw material.

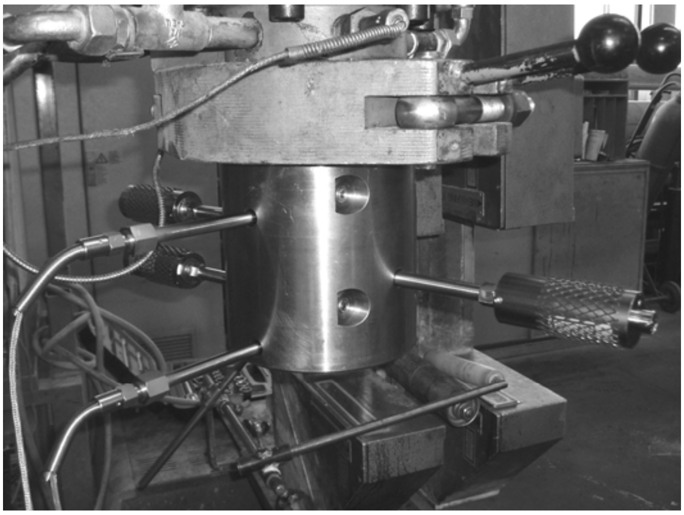

Figure 2. Mounted extrusion tool with three pressure transducers and two temperature sensors.

2 EXPERIMENTAL INVESTIGATIONS

2.1 *Materials*

The tested materials were non-vulcanized rubber blends used in industry, containing mainly EPDM (ethylene-propylene-diene-monomer), carbon black, and chalk as filler material. Such materials are used for window and pipeline seals, bridge expansion joints and various parts of cars.

2.2 *Capillary viscometer tests*

Rheological tests were performed with a capillary-viscometer (manufactured by Göttfert Ges.m.b.H., Germany) with capillary dies. One capillary experiment allows various piston velocities. In order to identify the influence of the geometry of the employed capillaries on the viscous material properties, a set of capillary dies with various ratios of length to diameter L/D and diameters D were used (Müllner et al. 2007). Experiments were carried out at temperatures of 80 °C, 100 °C, and 120 °C.

With a capillary viscometer raw blends or blends, which have been taken between extrusion tool exit and salt bath entry, can be investigated only. Therefore, relaxation effects can adulterate capillary viscometer measurements. With an extrusion rheometer these effects can be avoided.

2.3 *Extrusion rheometer tests*

In order to identify the influence of an extrusion process on the viscosity more precisely, an extrusion rheometer is used (Limper et al. 1999). It consists of a vertical extruder (see Fig. 1a) and a specified extrusion tool (see Fig. 2). The supply of raw material is shown in Figure 1b. In order to avoid the disadvantages of capillary viscometry (Müllner et al. 2006), an extrusion tool with a circular cross-section has been built. Thus, determination of the viscosity is possible under usage of characterization methods (Müllner et al. 2005a), which are standardly applicable for capillary experiments with rubber blends.

The extrusion tool and its dimensions are shown in Figure 2 and 3b. The elements of an extruder until the salt bath entry are shown principally in Figure 3a. The influence of salt bath vulcanization on the profile shape is not relevant, therefore is it not investigated in this contribution. Details regarding the numerical simulation of an air vulcanization process can be found in (André & Wriggers 1999).

The diameter of the cylinder is 14 mm, therefore positioning of pressure transducers within the cylinder is possible. The length of the cylinder is 133 mm. This length allows arrangement of three pressure transducers and two temperature sensors. With preliminary numerical studies (Petracek 2006) the distances of the pressure transducers have been fixed avoiding significant flow interaction. The usage of a vertical extruder allows the measurement of the die swell phenomenon of rubber blends in a similar manner according to (Müllner et al. 2005a).

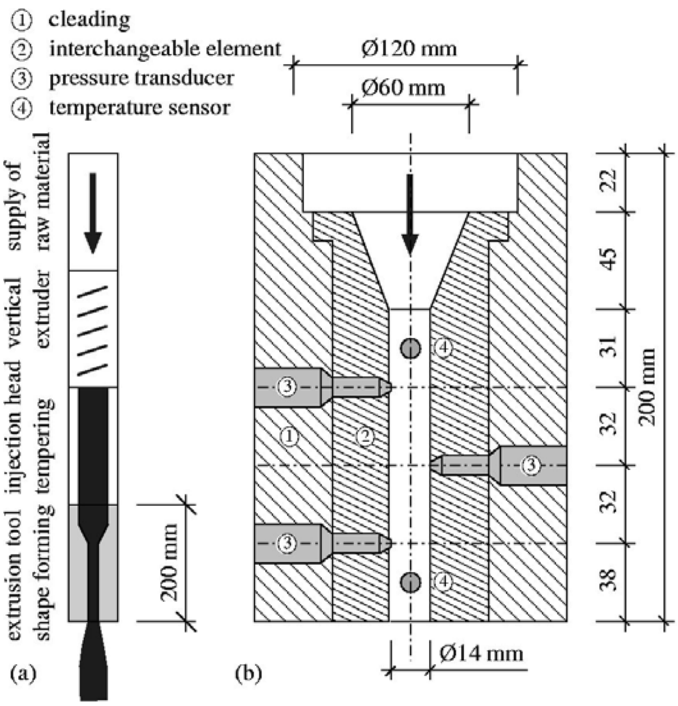

Figure 3. (a) Elements of extruder for rubber profile extrusion, (b) Dimensions of extrusion tool with circular cross-section.

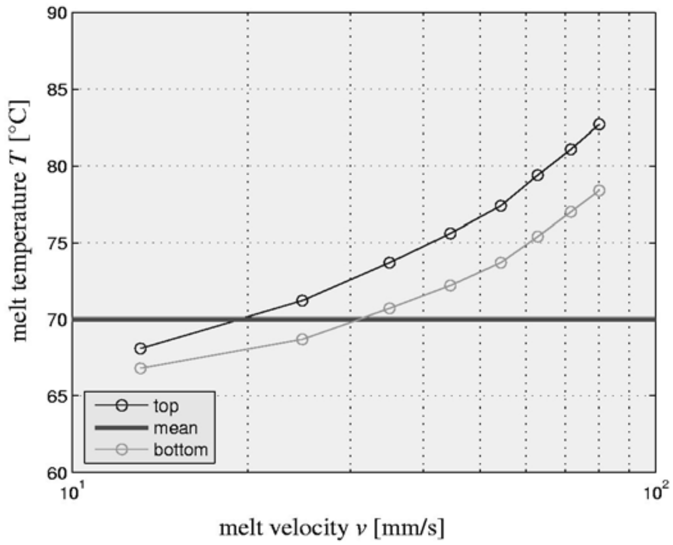

Figure 4. Increase of melt temperature during extrusion for a given temperature of the extrusion tool of 70 °C.

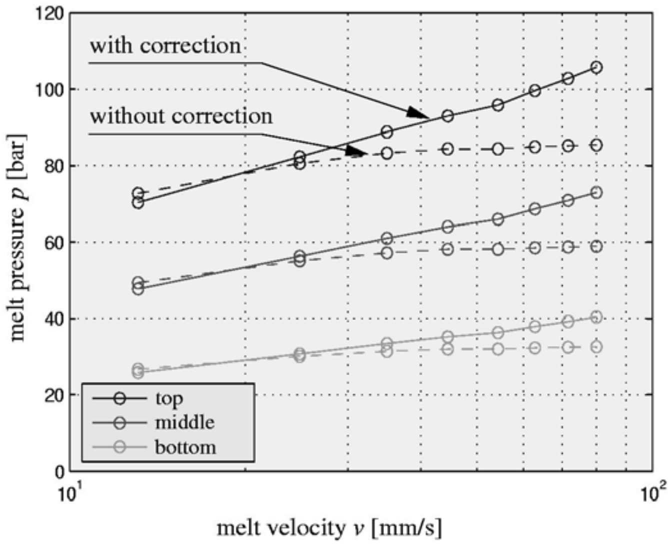

Figure 5. Consideration of energy dissipation due to increase of melt temperature by means of temperature law by Arrhenius.

3 MATERIAL CHARACTERIZATION

3.1 *Consideration of temperature increase*

As for capillary viscometry, during one experiment the investigation of different working points is possible by varying the speed of the screw press. This speed cannot be used as quantity for the melt velocity and the shear strain rate, respectively. Therefore, the extruded material has been weighted after three minutes for each working point. Applying the melt density of the raw material the melt velocity for each working point is obtained. The melt pressures and temperatures for every working point are the mean values over three minutes operating time. As regards pressure and temperature measurements, the operating time starts after reaching a steady flow state.

With increasing screw speed the melt temperature increases, too. As shown in Figure 4, the melt in the cylinder is not constant. The occurrence of energy dissipation has to be considered in the framework of material characterization. For that purpose, the pressure measurements have to be corrected by means of the temperature law by (Arrhenius 1889) as

$$p_V = p \exp\left[\frac{E_0}{R}\left(\frac{1}{T_0} - \frac{1}{T}\right)\right], \qquad (1)$$

where T is the measured temperature. E_0 is the material-specific activation energy. A detailed description of the thermodynamic behavior of rubber blends can be found in (Menges & Targiel 1982).

(Röthemeyer 1975) explains the interaction between rheological properties of rubber blends and the temperature within an extruder. To investigate this effect extrusion rheometer tests at 60 °C and 70 °C were performed. The temperatures of screw press and extruder were chosen as constant (see Tab. 1).

Figure 5 shows the results of one experiment with the extrusion rheometer. In the diagram both, the measured and the corrected pressure-velocity curves are shown. After consideration of energy dissipation a linear correlation of melt pressure and velocity in a double logarithmic scale is identified. This linear correlation is valid for a certain temperature, which also is constant during capillary viscometer tests.

3.2 *Determination of viscosity function*

Since rubber blends are shear-thinning, the viscosity η is a function of the shear strain rate $\dot{\gamma}$. The power law by (Ostwald 1925) and (de Waele 1923) was found to be useful for the description of the viscosity of rubber blends for a common range of shear strain rates. This model approximates experimental data by a straight line on a double logarithmic plot of shear stress versus shear strain rate:

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

where k is the consistency factor in [Pas^n] and n is the dimensionless viscosity exponent.

The constitutive characterization is done by means of an iteration method (Müllner et al. 2005b). Because of consideration of wall slippage, temperature effects and 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 generalized Newton-Raphson procedure has been adopted.

Table 1. Temperatures for extrusion rheometer tests.

screw	extruder			injection	tool
	zone 1	zone 2	zone 3	head	zone
50 °C	30 °C	40 °C	45 °C	65 °C	60 °C
50 °C	30 °C	40 °C	45 °C	65 °C	70 °C

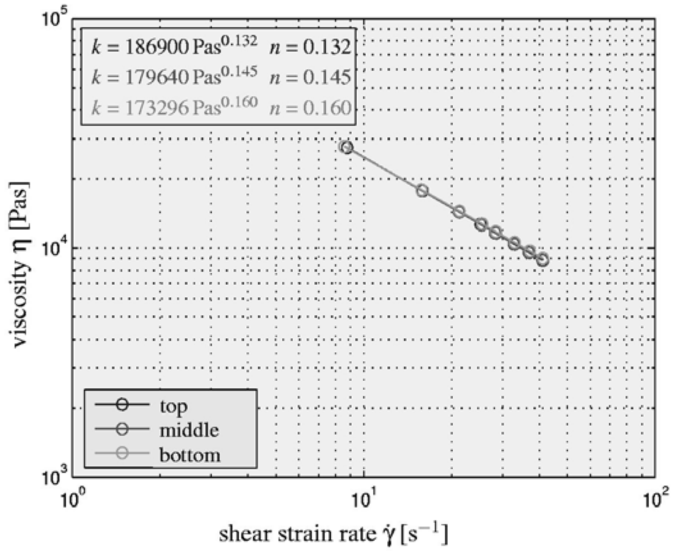

Figure 6. Viscosity functions of one experiment with the extrusion rheometer for all pressure measurements at 70 °C.

To minimize the number of coupled variables a new method was developed, which determines the pressure loss-correction due to measurement of the melt pressure in front of the capillary (Müllner et al. 2007). The developed procedure avoids various application problems, which occur when common correction methods are used (Michaeli 2003). The application of often used apparent quantities, mainly shear strain rate and shear stress, is not required. A successful validation for this characterization method based on a genetic algorithm is summarized in (Müllner et al. 2005a).

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

Figure 6 shows the viscosity curve at a tool temperature of 70 °C. The figure contains the results for all pressure measurements. It is remarkable that the extrusion rheometer covers a small range of shear strain rates only. However, the range of evolutions of the screw varies from 5 rpm to 40 rpm. Using capillary viscometry the range of shear strain rates is normally 1 s^{-1} < $\dot{\gamma}$ < 10,000 s^{-1}).

4 NUMERICAL VALIDATION

A numerical simulation of an extrusion rheometer test is performed using the CFD software POLYFLOW Version 3.92 (by Fluent Inc., Belgium). A working point at a tool temperature of 70 °C and a screw speed of 20 rpm is considered. Therefore, a comparison between experiment and simulation allows a validation of the present material characterization according to (Müllner et al. 2005b).

For the simulation wall adhesion is considered. The consideration of wall slippage in the framework of numerical simulations is still in progress. Preliminary studies on this topic can be found in (Petracek 2006). Due to symmetry reasons a quarter of the extrusion rheometer is considered.

As material description the power law with parameters according to Figure 6 is used (values of middle pressure transducer). In Figures 7 to 10 the distributions of various state variables are shown. The positions of the pressure transducers are indicated in the figures. A verification of pressure measurements is, therefore, possible (see Tab. 2).

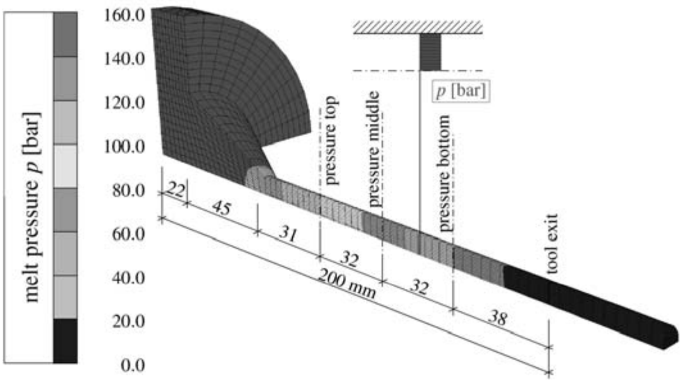

Figure 7. Pressure distribution during extrusion rheometer test.

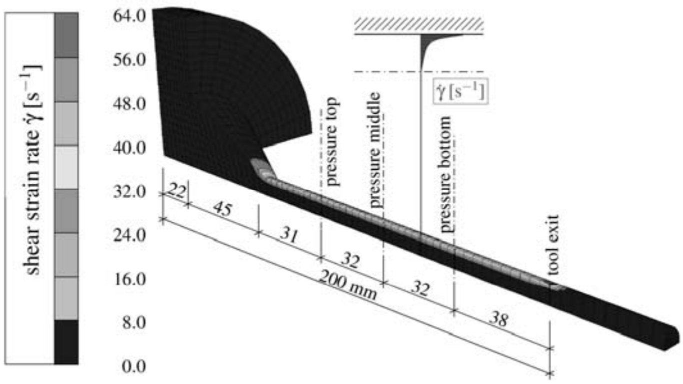

Figure 9. Shear strain rate during extrusion rheometer test.

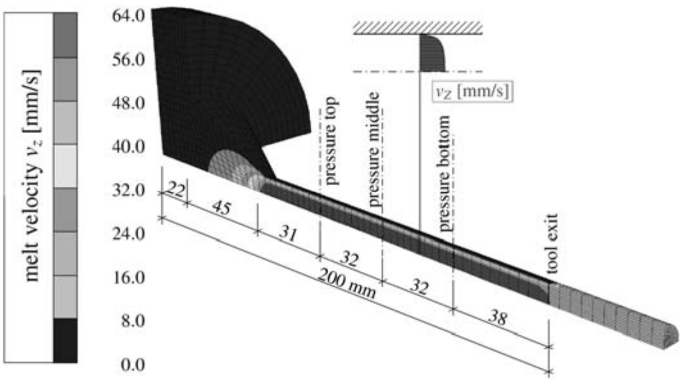

Figure 8. Velocity distribution during extrusion rheometer test.

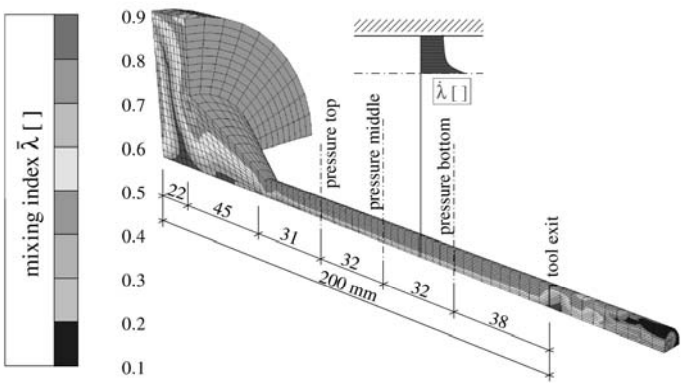

Figure 10. Flow conditions during extrusion rheometer test.

Table 2. Comparison of melt pressure.

	experiment	correction	simulation
injection head	136.0 bar	149.7 bar	149.8 bar
pressure top	84.4 bar	92.9 bar	94.1 bar
pressure middle	58.1 bar	64.0 bar	62.4 bar
pressure bottom	32.0 bar	35.3 bar	38.5 bar

The obtained numerical results show a good agreement with experimental data under consideration of energy dissipation according to equation (1). Therefore, a satisfactory validation for the material characterization according to (Müllner et al. 2005b) has been achieved.

However, the influence of an extrusion process on the viscosity function has not been identified. For this task, additional capillary experiments with raw and extruded material are required.

5 INFLUENCE OF EXTRUSION ON VISCOSITY FUNCTION OF RUBBER BLENDS

For identification of the interaction between extrusion and the rheological properties of rubber blends additional capillary experiments were performed. According to Section 2.2 different capillary dies and various melt temperatures were investigated.

Figures 11, 12 show all calculated viscosity functions for a typical EPDM rubber blend at 80 °C for both conditions, raw and extruded. The figures include the mean values of the power law constants. Comparing the mean values for both, consistency factor k and viscosity exponent n, a decrease of k, i.e. the absolute value of the viscosity function, of approximately 15 % is obtained. The slope of the viscosity curve, i.e. the shear-thinning behavior, remains nearly constant. Consideration of experimental data of all investigated rubber blends yields a viscosity reduction of 15 % to 35 %.

The reduction of viscosity due to extrusion is discussed in the open literature for plastics.

(da Costa et al. 2005) investigated the influence of multiple extrusion processes on the rheological properties of plastics. The higher the number of extrusion cycles, the higher the decrease of the complex viscosity, which has been determined by means of torsional rheometry. In addition, the elasticity of the material is reduced, too.

Investigations of (González-González et al. 1998) proved a change of molecular weight distributions of plastics. Due to extrusion molecular chain scissions were observed, leading to a reduction of viscosity. The latter was determined using the melt flow index.

The influence of extrusion on viscosity of plastics by means of capillary viscometer and single screw extruder was investigated by (Sombatsompop & Intawong 2005). Due to extrusion, an increase of apparent viscosity and extrudate swell was observed (Collyer & France 1980).

The absolute values of the viscosity function, i.e. the consistency factors k, of different experiments (Figs. 6, 12) cannot be compared. This fact can be explained, because of a geometry dependence of the viscosity function, which is standardly observed for rubber blends (Menges et al. 1983).

6 GEOMETRY DEPENDENCE OF VISCOSITY FUNCTION OF RUBBER BLENDS

The determination of viscoelastic properties of rubber blends by capillary viscometry is characterized by different viscosity functions due to different geometries of capillary dies. According to (Klüppel et al. 1992) the occurrence of wall slippage is one possibility for this phenomenon. However, due to different diameters of capillary dies (D = 0.5, 1.0, 2.0 mm) and extrusion tool (D = 14 mm) the viscosity functions of Figures 6, 12 cannot be compared.

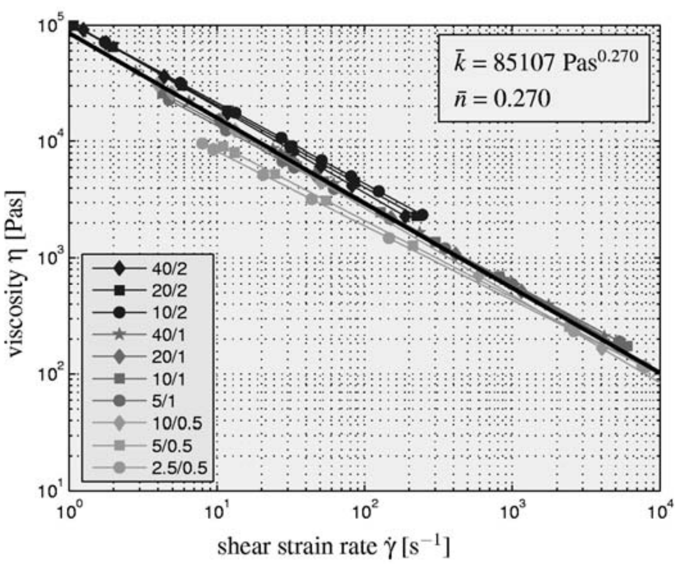

Figure 11. Viscosity of raw rubber blend at 80 °C.

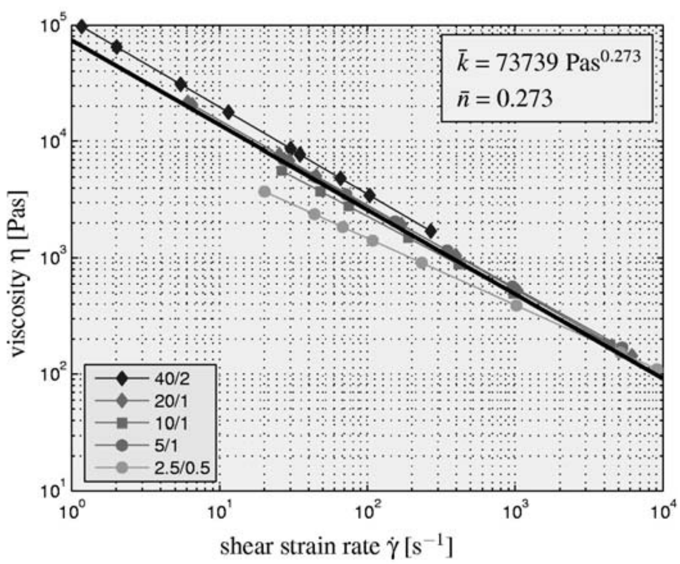

Figure 12. Viscosity of extruded rubber blend at 80 °C.

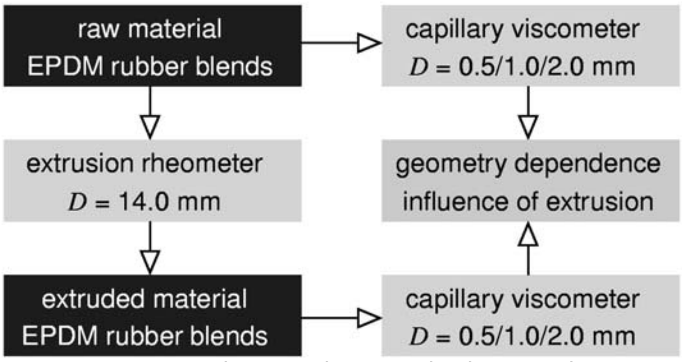

Figure 13. Strategy for identification of influence of extrusion process on viscosity function by means of two experiments.

The investigation of extruded materials under usage of capillary viscometry allows verification of both, interaction between extrusion process and viscosity as well as geometry dependence of viscosity functions of rubber blends. In Figure 13 the required approach for fulfilling both tasks is shown.

Because of usage of circular geometries for both experiments, capillary and extrusion rheometer tests, the consistency factors of different experiments can be compared. Figure 14 shows a plot of consistency factor k of raw and extruded material versus the diameter D of employed capillary dies and extrusion tool, respectively. For both, raw and extruded rubber blends, a clear relation between viscosity and geometry of the employed extrusion tool is identified.

7 CONCLUSIONS

The goal of this contribution was to identify the influence of extrusion of rubber blends on the viscosity function which has to be considered in the framework of numerical simulations of a rubber profile production. For this task, an extrusion rheometer was built. In combination with capillary experiments the interaction between extrusion and viscosity is clearly identified. Furthermore, the geometry dependence of the viscosity function of rubber blends is verified.

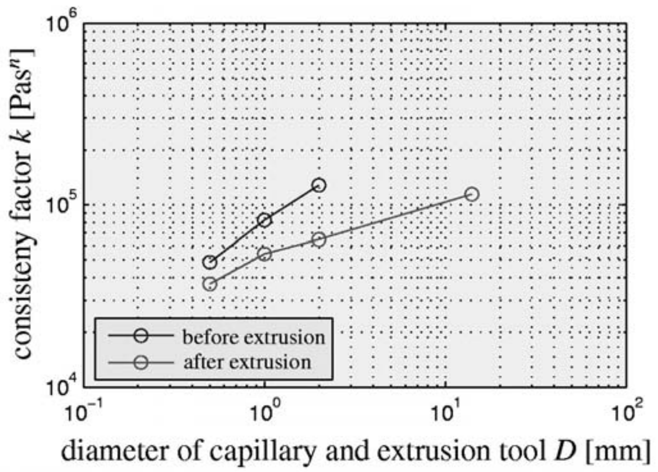

Figure 14. Identification of geometry dependence and influence of extrusion process on viscosity function.

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