Die-Swell Behavior During the Short-Tube Flow of Rubber Compounds

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ABSTRACT: In this study, the flow properties and die-swell ratios (*B*'s) of two kinds of rubber compounds (SI was a calcium carbonate filled natural rubber compound, and SII was a carbon-black-filled natural rubber/butadiene–styrene rubber/cis-1,4-butadiene rubber compound) in a short-tube extrusion flow were measured by means of a capillary rheometer under test conditions with a temperature of 90°C and an apparent shear rate varying from 10 to 4000 s^{-1} to identify the effects of extrusion conditions on the rheological behavior of the materials and to estimate *B*. The shear flow roughly obeyed the power law, whereas *B* increased nonlinearly with increasing extrusion rate. Under the same shear rates, the viscosity of SII was higher than that of SI, whereas the values of *B* of SI were higher than those of SII. Furthermore, *B* of the rubber compounds was estimated by means of an extrudate swell equation published in a previous work. The results show that the predictions of *B* were close to the measured data from the experiments. © 2007 Wiley Periodicals, Inc. J Appl Polym Sci 104: 70–74, 2007

Key words: extrusion; rheology; rubber

INTRODUCTION

Extrusion is one of the commonly used methods for polymer processing. Rheological behavior during the extrusion flow of polymeric materials plays an important role in the selection and control of shape-processing parameters and in the design of the extruder screw and head; it is related directly to the product quality and production cost. When a viscoelastic fluid leaves a circle die, the extrudate diameter is usually greater to some extent than the channel diameter. This is the *die-swell phenomenon,* also called the *extrudate swell* or *Barus effect* and alternately termed *memory* or *puff-up.* The degree of extrudate swell is usually expressed by the die-swell ratio (*B*). For the origin of the extrusion swell of viscoelastic fluids, there have been various interpretations, such as a normal stress effect, elastic energy effect, entropy enlargement effect, orientation effect, and memory effect. In fact, these interpretations are related to each other.[1–3] It is generally believed that die-swell is an important characteristic of the fluid elasticity in flow. With regard to the macroscopic view of polymer rheology, the relevant stress and strain of the macromolecular chains of polymeric fluids will be produced under the action of the extension, shear, and compression. If these stresses cannot be relaxed completely in the channel flow, the corresponding elastic deformation will be recovered after the fluid leaves from the die and will lead to extrudate swell.

Fifty years ago, Spencer and Dillon[4] discussed the die-swell problem in connection with the flow behavior of molten polystyrene. In the early 1970s, a number of researchers, including Bagley and Duffey,[6] Graessley et al.,[7] and Tanner,[8] studied the die-swell behavior and its mechanisms during the extrusion flow of polymer fluids and proposed mathematical models for describing the relationship between *B* and the rheological parameters of the melts.[5] Most of these models were established on the basis of the analysis of long capillary flow, and *B* was connected with the recoverable shear strain (S_R) or elastic strain energy. However, there have been relatively few studies on the die-swell behavior of polymeric materials in short-tube flow.

Because of entrance flow patterns, the factors affecting rheological behavior during the short-tube flow of polymer melts are relatively numerous and complicated.[9,10] Rubber materials are a typical viscoelastic material. It is necessary for the design of extrusion dies that *B* be accurately predicted. However, there have been relatively few quantitative descriptions on die-swell behavior during the short-tube extrusion of polymer melts. Huang and White[11] investigated the influence of extrusion conditions on the extrudate swell in the short-slit and capillary die flow of polystyrene and polypropylene melts and proposed corresponding die-swell equations with an elongation rate parameter. In a

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TABLE I
Partial Physical Properties of the Rubber Compounds

Material	Hardness (Shore A)	Plasticity (Williams)	ρ (kg/m^3)
SI	51 ± 3	0.53 ± 0.04	1131
SII	63 ± 3	0.28 ± 0.05	1095

previous work,[9] I derived a short-die extrudate swell equation with a tenser analysis method. My objectives in this study were to investigate the influence of extrusion conditions on the viscoelastic properties, such as the flow properties and die-swell behavior, of rubber compounds in short-die extrusion and to compare the measurements from the experiments with the predictions of B by the application of this short-die extrudate swell equation.

EXPERIMENTAL

Materials

The materials used in these tests were two rubber compounds, which were named SI and SII, respectively. SI was a calcium carbonate ($CaCO_3$) filled natural rubber (NR) compound, and the content of $CaCO_3$ was 20 phr. SII was a carbon-black (CB)-filled NR/styrene–butadiene rubber (SBR)/cis-1,4-butadiene rubber (CBR) compound. The blending ratio of NR/SBR/CBR was 45/10/45, and the content of CB was 56 phr. In addition, both the rubber compounds included some additives, such as sulfur and stearic acid. The partial physical properties of these compounds are listed in Table I.[12]

Instruments and methodology

The major instrument used in this test was a Instron capillary rheometer (model 3211). Two capillary dies with different lengths were selected to measure the rheological properties of the sample materials. The length of the long die was 40 mm, the length of the short die was 0.2 mm, and the diameter (D) of these dies was 1 mm. In this case, the shear at the channel wall (τ_w) was determined by

$$\tau_w = \frac{\Delta P_l D}{4L} \tag{1}$$

where ΔP_l is the total pressure drop when the materials flow the long die with a length of L.

In this study, B was determined by means of a weighing method with a constant length of extrudate:

$$B = \left(\frac{400W}{\pi\rho l D^2}\right)^{\frac{1}{2}} \tag{2}$$

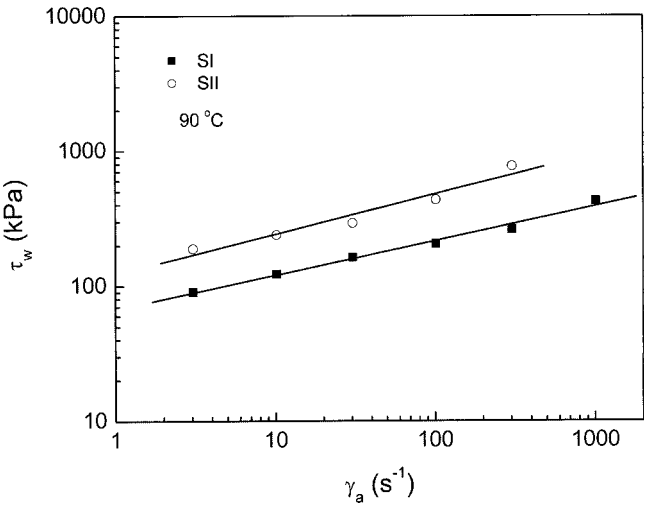

Figure 1 Shear flow curves of the materials.

where W, ρ, and l are the weight, density, and length of the extrudate, respectively. Weighing was carried out after the extrudate was cooled to ambient temperature.

The reservoir temperature range started at 90°C, and the piston speed (V) varied from 0.45 to 150 mm/min. The relationship between apparent shear rate ($\dot{\gamma}_a$) and V (mm/min) can be expressed as follows:

$$\dot{\gamma}_a = \frac{2V\beta^2}{15D} \tag{3}$$

where $\beta = D_R/D$ and D_R is the reservoir diameter (mm).

RESULTS AND DISCUSSION

Shear flow curves

Figure 1 shows the relationship between shear stress and $\dot{\gamma}_a$ when the rubber compounds flowed through the long die at a test temperature of 90°C. The wall shear stress increased linearly with increasing $\dot{\gamma}_a$ in a bilogarithm coordinate system. This means that the shear flow of the rubber compounds obeyed the power law under the extrusion conditions. That is

$$\tau_w = K\dot{\gamma}_a^n \tag{4}$$

where K and n are the power law constants.

From the results shown in Figure 1, the values of K and n could be determined with a linear regression analysis method. The results are listed in Table II. Under the same test conditions, the viscosity of SII was greater than that of SI.

TABLE II
***K* and *n* Values of the Rubber Compounds**

Material	K (kPa s)	n	R
SI	67.1707	0.2537	0.9932
SII	123.3360	0.2941	0.9736

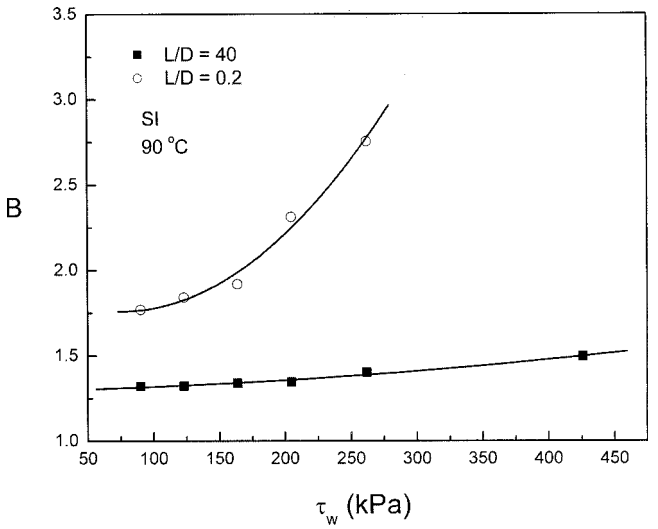

Figure 2 Dependence of B on τ_w of SI.

Dependence of B on shear stress

Figure 2 illustrates the dependence of B on shear stress at the tube wall during the capillary extrusion of SI at 90°C. For a long-die extrusion, B increased gently with increasing τ_w, and the relationship between them was approximately linear. For a short-die extrusion, B increases quickly with increasing τ_w, and the relationship between them was an index function.

Figure 3 displays the dependence of B on shear stress at the tube wall during the capillary extrusion of SII also at 90°C. It could be that B increases gently with increasing τ_w for a long-die extrusion, whereas for a short-die extrusion, B increases quickly with increasing τ_w. The relationship between B and τ_w, either in a long-die extrusion or in a short-die extrusion, was also an index function.

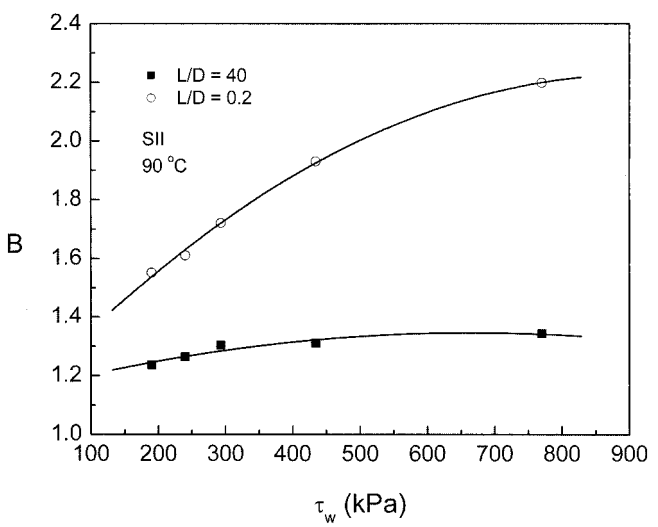

Figure 3 Dependence of B on τ_w of SII.

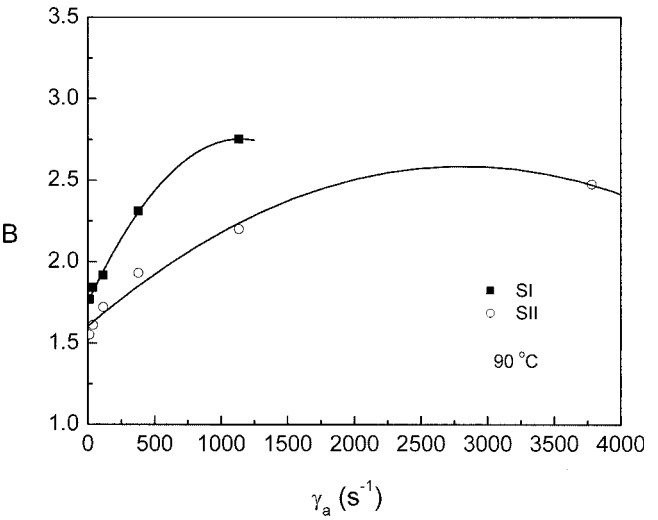

Figure 4 Dependence of B on $\dot{\gamma}_a$ ($L/D = 0.2$).

Relationship between B and $\dot{\gamma}_a$ in a short-die flow

Figure 4 shows the correlation between B and $\dot{\gamma}_a$ during a short capillary die extrusion flow of these two rubber compounds at 90°C. B increased nonlinearly with increasing $\dot{\gamma}_a$. With the same $\dot{\gamma}_a$, the values of B for SI were greater than that for SII. This indicates that the fluid elasticity in the extrusion of SI was higher than that of SII. One of the origins for this phenomenon might be that SII was a rubber compound with a high loading of CB and the fluid elasticity was weakened with the addition of inorganic fillers.

Comparison of predictions and measurements of B

Inlet flow includes shear flow and elongation flow; the effect of the entry flow pattern on die-swell behavior during the short-channel flow of polymeric materials is significant due to the fact that the stresses cannot be relaxed completely when the fluid leaves the tube. Therefore, the die-swell mechanisms of polymeric materials in a short tube are complicated, and the main factors affecting the extrudate swell should consist of shear flow and elongation flow. The entry natural convergence angle of viscoelastic fluids is an important parameter for the characterization of inlet converging flow. In a previous work,[10] I derived a die-swell equation on the basis of an investigation of polymer melts flowing through a short tube by means of a tensor analysis method, as follows:

$$B = [1 + 2\lambda S_R + \lambda^2 S_R^2]^{\frac{1}{4}} \tag{5}$$

where S_R is the recoverable shear strain and λ is the entry converging flow parameter, which is given by

$$\lambda = \frac{1}{2} tg\alpha_o \tag{6}$$

where α_o is the half-entry natural convergence angle of viscoelastic fluids.

Ma et al.[13] observed the entrance flow patterns in elastomers and their CB compounds during extrusion through dies with a trace mark technique, and the flow marker was titanium dioxide. They found that CB compounds all exhibited a converging streamline flow in dies with an entrance angle of 180°, and the natural convergent angles were from 100 to 150° under extrusion conditions. Sombatsompop and Wood[14] investigated the effects of test conditions on the flow patterns and entrance velocity profiles in the die extrusion of NR and also observed a similar variation in the natural convergent angles. In this study, the entry natural converging angle was taken as about 120° according to the previous experimental results and relevant test conditions.

Tanner[8] proposed an expression for describing a relationship between S_R and B based on a long-tube flow of viscoelastic fluids:

$$S_R = [2(B^6 - 1) - 2]^{\frac{1}{2}} \quad (7)$$

In this study, S_R was calculated from measured data of B during the long capillary extrusion of the rubber compound with eq. (7). Figure 5 displays the comparison between the estimations by means of eqs. (5) and (7) and the measured B from SI under the test conditions. The values of the estimations of B were relatively close to the measured data at lower $\dot{\gamma}_a$'s, and the difference between them increased gently with increasing $\dot{\gamma}_a$.

Figure 6 shows the comparison between the estimations and the measured B from SII under the test conditions. Contrary to the results shown in Figure 5, the difference in B between the calculations by means of eqs. (5) and (7) and the measured B from SII was relatively large at lower $\dot{\gamma}_a$'s, and the values of the estimations of B were gradually close to the measured data with increasing $\dot{\gamma}_a$. This indicates that eq. (5) was roughly suitable for estimating B during the short-tube extrusion flow of rubber compounds.

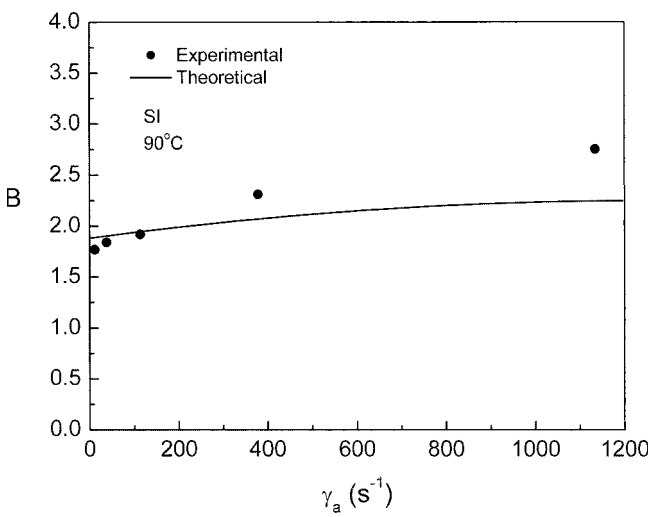

Figure 5 Comparison between calculations of B and measured data from SI.

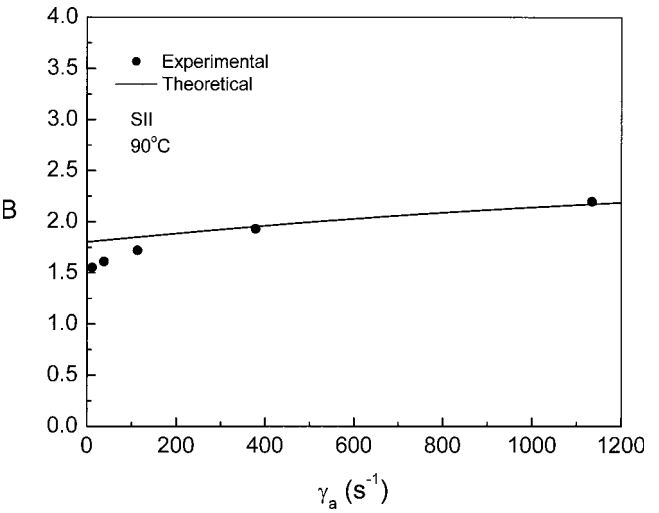

Figure 6 Comparison between calculations of B and measured data from SII.

Discussion

As the stated previously, the effects of entrance flow patterns on the flow and elastic behavior during the short-tube extrusion of viscoelastic fluids are very significant because when a viscoelastic fluid enters a small channel from a reservoir, it will generate an entry convergent flow. It is generally believed that inlet convergent flow includes shear flow and elongation flow and produces large shear and elongation stress and corresponding deformation, which leads to the formation of obvious entry pressure losses. These stresses produced in entrance flow might not be wholly relaxed when the extrudate leaves the tube because of the short time the fluid is in this channel, which will result in an increase in the die-swell degree.

The entry natural convergent angle is an important parameter for the characterization of converging flow patterns during the tube extrusion of viscoelastic fluids, which is closely related to the viscoelastic properties of the fluid and flow conditions, including channel geometry. Although there have been several equations for the estimation of the entry natural convergent angle during the extrusion flow of polymer melts reported in literature,[12,15] it is very difficult to accurately determine the entry natural convergent angle under generally experimental conditions, especially for some polymeric materials with strong viscoelastic properties and their blends or composites.

CONCLUSIONS

Entrance flow patterns directly affect the flow and die-swell behavior during the short-tube extrusion of

polymeric fluids, and the entry natural converging angle of the fluids is an important parameter for the characterization of the entry flow patterns. The flow and die-swell behavior of the NR/$CaCO_3$ compound (SI) and NR/SBR/CBR/CB (SII) compound in a short-tube extrusion flow were investigated by means of a capillary rheometer at 90°C and with $\dot{\gamma}_a$ varying from 10 to 4000 s^{-1}. The shear flow roughly obeyed the power law, whereas B increased nonlinearly with increasing shear rate. Under the same shear rates, the viscosity of the SII system was higher than that of the SI system, whereas the values of B of SI system were higher than those of the SII system. Furthermore, B of these rubber compounds was estimated by means of an extrudate swell equation [eq. (5)]. The results show that the predictions of B were close to the measured data from the experiments of these two rubber compounds, especially for the SII system.

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