

# Elastic Behavior of LDPE/HEPE Blend Melts in Capillary Extrusion

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Received 2 September 1999; accepted 10 December 1999

**ABSTRACT:** The studies of the elastic behavior in the capillary flow of LDPE/HDPE blend melts were carried out at a test temperature range from 180 to 200°C and at an apparent shear rate of about 25–120 s<sup>-1</sup>. The end-pressure drop ( $\Delta P_{\text{end}}$ ) increased nonlinearly with increasing wall shear stress ( $\tau_w$ ) and achieved a minimum value at a weight fraction ( $\phi_{\text{HD}}$ ) of HDPE of 50%. The die-swell ratio ( $B$ ) increased basically linearly with increasing  $\tau_w$  or  $\Delta P_{\text{end}}$  and achieved a maximum value at  $\phi_{\text{HD}}$  of 50%. With the addition of the die length–diameter ratio, the values of  $B$  were decreased linearly. At a low shear rate, the temperature sensitivity of the melt die-swell was more significant than at a high shear rate. With increasing  $\phi_{\text{HD}}$ ,  $B$  increased when  $\phi_{\text{HD}} < 50\%$ , then decreased.  $B$  reached a maximum value at  $\phi_{\text{HD}}$  of 50% and a fixed apparent shear rate. This phenomenon may be explained by using the theory of viscoelastic competition between components of polymer blend melts. Furthermore, the first normal stress difference ( $N_1$ ) of the sample melts was estimated by using an equation published in a previous work. The results showed that  $B$  increased linearly with increasing  $N_1$ . © 2000 John Wiley & Sons, Inc. *J Appl Polym Sci* 78: 759–765, 2000

**Key words:** polyethylene; blending; melt; die-swell; capillary; extrusion

## INTRODUCTION

To obtain good comprehensive performance, polymer materials blending at a molten state is one of the effective, simple, and economic methods which are widely used in industry. High-density polyethylene (HDPE) has a series of advantages in higher hardness, tensile strength, heat soft point, as well as good chemistry and water resistance. The flexibility and ductility of low-density polyethylene (LDPE) are quite outstanding, but its heat soft point and tensile strength are lower than those of HDPE. Obviously, the application scope of these PEs will be extended, when LDPE is blended with HDPE in melt in a suitable mixing proportion.

It is a basic prerequisite for material processing and shaping as well as the machinery design that the rheological properties of polymer melts are well understood. In the recent two decades, the viscoelastic behavior of HDPE/LDPE blend melts during elongation and shear flow and the applications in extrusion pipes were investigated.<sup>1–4</sup> La Mantia et al.<sup>2</sup> studied the extensional flow of HDPE/LDPE blend melts using a constant-rate rheometer and obtained melt-extension viscosity using the converging flow analysis method proposed by Cogswell.<sup>5</sup> Curto et al.<sup>3</sup> also investigated the end effects and shear viscosity of the blend melts. The synergistic effects were evidenced when the relevant properties of the homopolymer parents were not too different from one another. Using a kinetic network model, Manero et al.<sup>4</sup> analyzed the melt-flow behavior of HDPE/LDPE blends and found that the shearing

**Table I Part Physical Properties of the Raw Materials**

Resin Trade Mark	Supplier	<i>MI</i> (g/10 min)	$\rho$ (g/cm <sup>3</sup> )
LD160 AS	Exxon Co., Belgium	4	0.921
HMN-55180	Daelim Co., Korea	20	0.955

history influenced the rheological properties of the pure homopolymers to a measurable and significant degree. Therefore, this had to be taken into account when these properties were used to predict the flow behavior of these blends. Garcia-Rejon and Alvarez<sup>6</sup> performed a complete rheological characterization of HDPE/LDPE blends. The experimental results showed that there were important differences in the mechanical properties (transverse and longitudinal) of the sheets from the blends. These differences were explained on the basis of the processing conditions (thermo-mechanical history) and the rheological properties of the molten blends. In previous work,<sup>7</sup> the authors studied the melt-flow properties of LDPE/HDPE blends in capillary extrusion. It was found that the melt shear flow obeyed the power law and that the melt shear viscosity achieved the maximum at the weight fraction of HDPE of 20%.

Die-swell, end-pressure losses, and the first normal stress difference are important characteristics of the melt elasticity for polymers. The objectives of this work were to investigate the elastic behavior and the factors affecting it during capillary flow of LDPE/HDPE blend melts on the basis of the previous work stated above.

## EXPERIMENTAL

### Materials

An HDPE and an LDPE, with different melt flow indexes (*MI*), were selected as the raw materials in this test. The part physical properties are sum-

marized in Table I. These *MI* values were measured under standard test conditions (190°C, 2.16 kg). It can be seen that the *MI* value of the HDPE melt is twice as high as that of the LDPE melt.  $\rho$  is the density at the solid state.

### Sample Preparation

A simple mixing of the HDPE and LDPE was conducted at room temperature. Then, the mixtures were blended in a molten state of the resins using a single-screw extruder. Finally, the extrudates were palletized to produce the samples. The blending ratios (weight percentage) of LDPE/HDPE were 0/100, 20/80, 50/50, 80/20, and 100/0. The basic technical parameters of the extruder screw and die as well as the blending operation conditions are listed in Table II.

### Apparatus and Methodology

The main instrument used in this work was a constant-rate type of capillary rheometer (Model Rheovis 2100, Ceast Co., Italy). A set of capillary dies with diameter of 1 mm was selected. The die length–diameter ratios (*L/D*) were 10, 20, 30, and 40, and the entry angle was 180°.

The rheometer reservoir temperature scope was 180–200°C. The piston speed (*V*) was from 2 to 10 mm/min (relevant apparent shear rate was about 25–120 s<sup>-1</sup>). The end pressure drop ( $\Delta P_{\text{end}}$ ) and the die swell ratio (*B*) were measured in these test conditions. The end-pressure drop was determined using the Bagley's plotting method.<sup>8</sup> *B* was measured using a method of constant-length-extrudate weighing at room temperature, which was estimated by

$$B = \left( \frac{4000W}{\pi \rho_m L_e D^2} \right)^{1/2} \quad (1)$$

where *W* is the weight of the extrudate with a constant length (*L<sub>e</sub>* in millimeters) in *g*. The

**Table II Screw and Die Parameters and Blending Operation Conditions**

Screw		Die		Barrel Temperature (°C)		Die Temperature (°C)	Screw Speed (r/min)
<i>D<sub>s</sub></i> (mm)	<i>L<sub>s</sub>/D<sub>s</sub></i>	<i>D<sub>d</sub></i> (mm)	<i>L<sub>d</sub>/D<sub>d</sub></i>	Rear	Front		
19.05	20/1	1.6	25/1.6	150–160	160–170	170–200	40

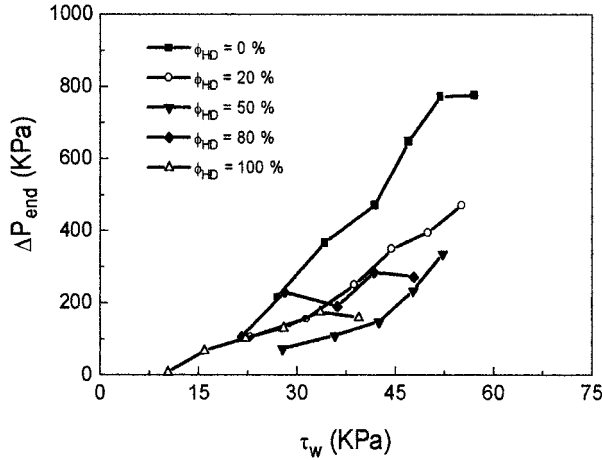


Figure 1  $\Delta P_{\text{end}}$  as a function of  $\tau_w$  (180°C).

blend melt density  $\rho_m$  can be approximately calculated by

$$\rho_m = \rho_{\text{LD}}(1 - \phi_{\text{HD}}) + \phi_{\text{HD}}\rho_{\text{HD}} \quad (2)$$

where  $\phi_{\text{HD}}$  is the weight fraction of HDPE, and  $\rho_{\text{LD}}$  and  $\rho_{\text{HD}}$ , the density of LDPE and HDPE, respectively.

## RESULTS AND DISCUSSION

### End-pressure Losses

When a polymer melt enters a capillary from a reservoir, the converging flow is formed at the front of the die entrance due to its viscoelasticity and the abrupt contraction of the channel. This entry flow concludes the elongation and shear flow. Thus, relevant extension and shear deformation will be produced, and the elastic strain energy will be stored in the melt, resulting in obvious end-pressure losses. It is generally believed that the end-pressure losses are important characteristics of the melt viscoelasticity, which are usually represented by using  $\Delta P_{\text{end}}$ . Figure 1 shows the plots of  $\Delta P_{\text{end}}$  against the wall shear stress ( $\tau_w$ ) for the sample melts at 180°C. The values of  $\Delta P_{\text{end}}$  are nonlinearly increased with increasing  $\tau_w$ . Under the same shear stress level,  $\Delta P_{\text{end}}$  achieves a minimum value at  $\phi_{\text{HD}}$  of 50%.

Because the end-pressure drop and die-swallow ratio are related to the melt elasticity, there should be some form of correlation between them. Figure 2 illuminates the relationship between  $B$  and  $\Delta P_{\text{end}}$  of the sample melts at 180°C. At the

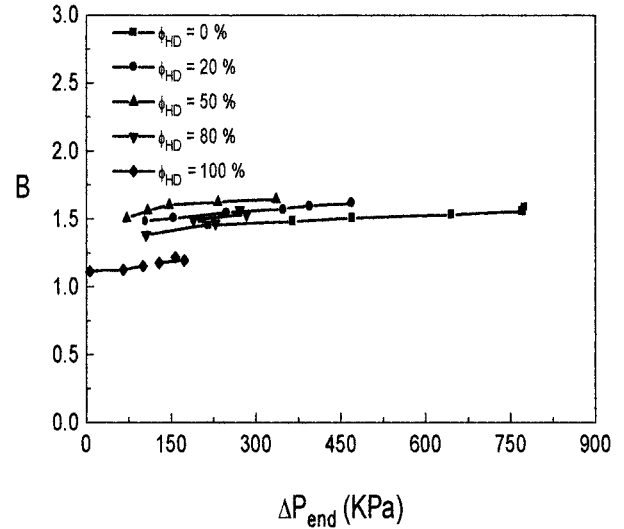


Figure 2  $B$  as a function of  $\Delta P_{\text{end}}$  (180°C,  $L/D = 30$ ).

same shear stress level, the values of  $B$  for the blend melts are greater than those of the pure LDPE and HDPE melts. The  $B$  increased lightly with the addition of  $\Delta P_{\text{end}}$ . Basically,  $B$  is a linear function of  $\Delta P_{\text{end}}$ , which is given by

$$B = A_0 + A_1\Delta P_{\text{end}} \quad (3)$$

where  $A_0$  and  $A_1$  are the constants related to the melt elasticity.

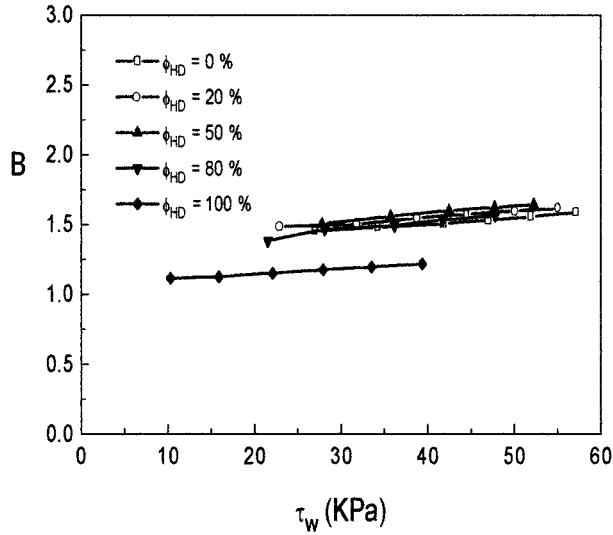
The values of  $A_0$  and  $A_1$  can be determined using a numeric analysis method. Table III lists the values of  $A_0$  and  $A_1$  for the sample melts calculated from the results as shown in Figure 2. It can be seen from Table III that the corresponding coefficients  $R$  are greater than 0.9, indicating that eq. (3) is basically suitable for the blend melts under the test conditions.

### Dependence of $B$ on $\tau_w$ and Temperature

The dependence of the die-swallow ratio for the sample melts on the wall shear stress is shown in

Table III  $A_0$  and  $A_1$  Values of the Blend Melts

$\phi_{\text{HD}}$ (wt %)	$A_0$	$A_1$	$R$
0	1.4035	0.0002	0.97302
20	1.4490	0.0004	0.99441
50	1.5006	0.0005	0.89854
80	1.2923	0.0009	0.92418
100	1.0991	0.0006	0.93617



**Figure 3**  $B$  as a function of  $\tau_w$  (180°C,  $L/D = 30$ ).

Figure 3 when the test temperature is 180°C. The values of  $B$  are linearly increased with increasing  $\tau_w$ . Similarly, the values of  $B$  for the blend melts are greater than those of the pure LDPE and HDPE melts at the same shear stress level. The correlation between  $B$  and  $\tau_w$  can be described as follows:

$$B = C_0 + C_1\tau_w \quad (4)$$

and

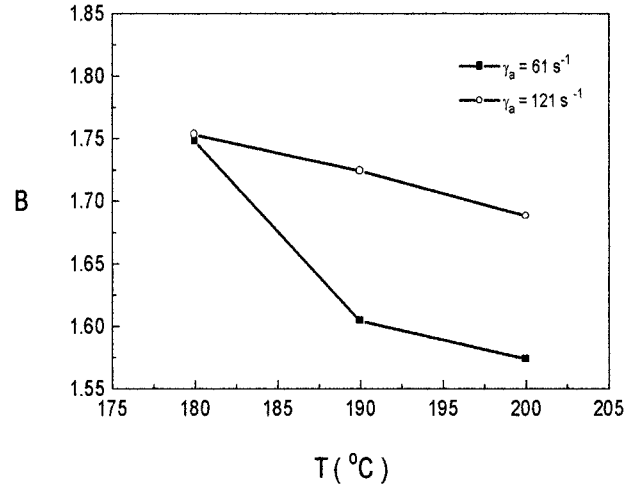
$$\tau_w = (\Delta P - \Delta P_{\text{end}})D/4L \quad (5)$$

where  $C_0$  and  $C_1$  are the constants related to the melt elasticity.  $\Delta P$  is the total pressure drop in the die extrusion of the sample melts.

The values of  $C_0$  and  $C_1$  for the melts can also be determined using a numeric analysis method such as linear apparent fitting from the experimental results shown in Figure 3. The results are listed in Table IV. It can be seen that the values of

**Table IV**  $C_0$  and  $C_1$  Values of the Blend Melts

$\phi_{\text{HD}}$ (wt %)	$C_0$	$C_1$	$R$
0	1.3278	0.0044	0.99401
20	1.3781	0.0043	0.99032
50	1.3499	0.0057	0.99418
80	1.2580	0.0065	0.97588
100	1.0719	0.0037	0.99707



**Figure 4** Dependence  $B$  on temperature ( $L/D = 20$ ).

$R$  are greater than 0.99. It suggests that the die-swell ratio of the melts is a linear function of  $\tau_w$ .

Figure 4 displays the influence of the test temperatures on the melt die-swell ratio. It can be observed that  $B$  decreased linearly at a higher apparent shear rate while it decreases rapidly at lower apparent shear rate, with increase of the test temperatures. It suggests that the effect of the shear rate on the temperature sensitivity of  $B$  for the blend melts is evident. The apparent shear rate ( $\dot{\gamma}_a$ ) is given by

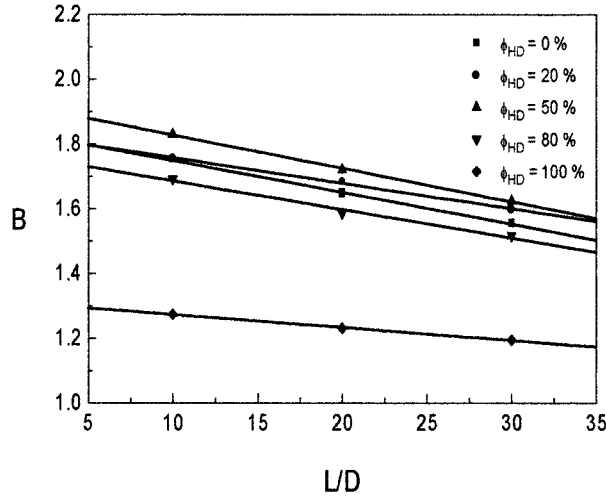
$$\dot{\gamma}_a = \frac{32Q}{\pi D^3} \quad (6)$$

where  $Q$  is the volumetric flow rate.

From the viewpoint of macrorheology, the elastic recovery of deformation produced during the die extrusion of polymer melts is an important origin, leading to the extrudate swell. This deformation is formed not only in the entry flow, but also in the die flow. Hence, the values of  $B$  for the sample melts and the remainder normal stress in the melts are increased relevantly with increasing  $\tau_w$  and  $\Delta P_{\text{end}}$  when the test temperatures are constant (see Figs. 2 and 3).

#### Correlation Between $B$ and $L/D$

When the test temperature is 180°C and the fixed apparent shear rate is 101 s<sup>-1</sup>, the correlation between  $B$  and  $L/D$  is as displayed in Figure 5. The values of  $B$  are linearly decreased with the addition of  $L/D$ . In other words,  $B$  is a linear function of  $L/D$  as follows:



**Figure 5**  $B$  as a function of  $L/D$  ( $180^{\circ}\text{C}$ ,  $\dot{\gamma}_a = 101 \text{ s}^{-1}$ ).

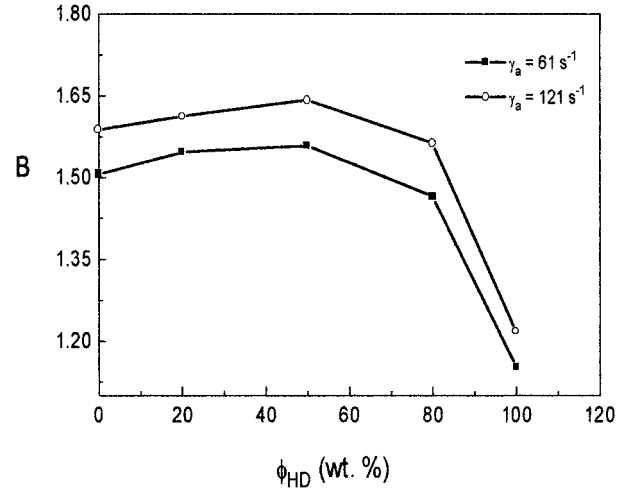
$$B = E_0 - E_1 L/D \quad (7)$$

where  $E_0$  and  $E_1$  are the constants related to the melt elasticity.

Table V lists the values of the  $E_0$  and  $E_1$  sample blend melts calculated from the experimental results as shown in Figure 4. The values of  $R$  for the melts exceed 0.99. When  $\phi_{\text{HD}}$  is 50%, the sensitivity of  $B$  to  $L/D$  of the blend melts is the most significant of all the melts. Generally, the extrudate swell degree depends mainly upon the quantity and releasing speed (stress relaxation) of the elastic strain energy stored in the melts in the flow. For a short die, the elongation and shear deformation produced in the inlet flow are the main factors for affecting the melt die-swell behavior due to a relatively small residence time of the melts in the die. Therefore,  $B$  increases relevantly under the same operation conditions (see Fig. 5). With increase of the test temperatures, the melt viscosity decreases and the melt relaxation time is shortened, resulting in weakening of

**Table V**  $E_0$  and  $E_1$  Values of the Blend Melts

$\phi_{\text{HD}}$ (wt %)	$E_0$	$E_1$	$R$
0	1.8460	0.0098	0.99938
20	1.8352	0.0079	0.99848
50	1.9324	0.0103	0.99943
80	1.7731	0.0088	0.99310
100	1.3113	0.0040	0.99869



**Figure 6**  $B$  versus of  $\phi_{\text{HD}}$  ( $L/D = 30$ ,  $180^{\circ}\text{C}$ ).

the melt elasticity. Thus, the die-swell ratio of the blend melts decreases correspondingly (see Fig. 4).

#### Effect of $\phi_{\text{HD}}$ on $B$

When the test temperature is  $180^{\circ}\text{C}$  and the apparent shear rates are fixed, the effect of the weight fraction of HDPE on the melt die-swell ratio of the samples is as shown in Figure 6. It can be observed that the values of  $B$  are increased at  $\phi_{\text{HD}} < 50\%$ , while they are decreased at  $\phi_{\text{HD}} > 50\%$ , with the addition of  $\phi_{\text{HD}}$ .  $B$  achieves a maximum value at  $\phi_{\text{HD}} = 50\%$  and a fixed  $\dot{\gamma}_a$ , suggesting that the synergistic effect in the die-swell for the blend melts is evident within a range of  $\phi_{\text{HD}}$  from 20 to 60%. In addition, when the test temperature is fixed, the melt  $B$  reaches a maximum value at the same end-pressure drop and blending ratio of 50/50 (see Fig. 2).

For polymer blend melts, the rheological behavior depends, to a great extent, upon the mixing and distribution among the components, the morphology of the interface between the phases, and the viscoelastic difference among the components in addition to the factors stated above. When  $\phi_{\text{HD}} < 50\%$ , the LDPE melt, as a continuous phase, contacts the channel walls and results in a relatively strong shear effect, leading to producing a large shear deformation and storing greater elastic energies in the melt flow. On the other hand, the HDPE melt, as a dispersed phase, is distributed in the LDPE melt in the form of droplets to produce a so-called sea-island structure. Because the HDPE melt droplets do not come into contact directly with the wall, the shear

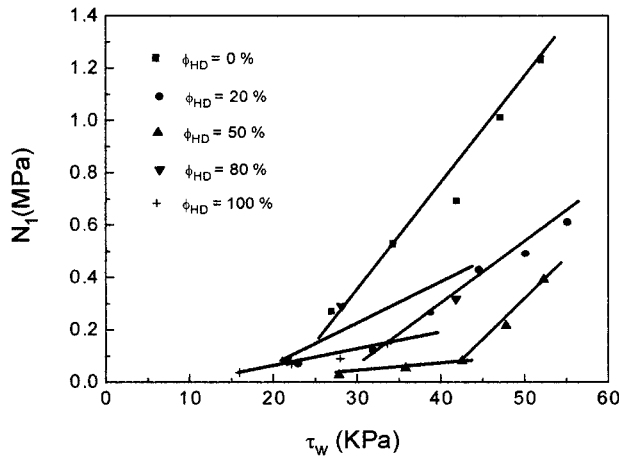


Figure 7  $B$  versus  $\tau_w$  ( $L/D = 30$ ,  $180^\circ\text{C}$ ).

deformation is relatively less, leading to small viscous dissipation. In addition, for a two-phase polymer-blending system, the synergistic effects will be evident when the relevant properties are not too different from one another.<sup>3</sup> Thus, the extrudate swell increases with increasing  $\phi_{\text{HD}}$ . It is contrary to this when  $\phi_{\text{HD}} > 50\%$ . As a result, the melt  $B$  achieves a maximum value at a blending ratio of 50/50 (see Fig. 6). Acierno et al.<sup>9</sup> blended LDPE in a molten state with three kinds of linear LDPEs and investigated the relationship between  $B$  and the component content at a constant shear stress. The results also showed that  $B$  achieved a maximum value at a blending ratio of 50/50, indicating that there is a viscoelastic competition mechanism between the components for the rheological behavior of polymer blends. When the component with stronger elasticity is dominant, the melt-swell increases. Otherwise, it decreases.

#### First Normal Stress Difference

The first normal stress difference,  $N_1$ , is an important parameter for characterizing the viscoelasticity of polymer melts. During the past three decades, several scientists related it to the die-swell ratio for viscoelastic fluids and proposed some well-known equations.<sup>10</sup> More recently, the authors<sup>11</sup> proposed an expression for describing the relationship between  $N_1$  and  $\Delta P$  as follows:

$$N_1 = 2\Delta P[\xi - n_c(1 - \xi)D/2L] \quad (8)$$

where  $\xi = \Delta P_{\text{end}}/\Delta P$ , and  $n_c$  is the Couette correction factor, which is about 1.5–2 for the polymer melts.

Figure 7 shows the dependence of  $N_1$  on the  $\tau_w$  of the sample melts at  $180^\circ\text{C}$ , which is estimated by using eq. (8) from the experimental data. It can be seen that  $N_1$  increases linearly with the increase of  $\tau_w$ . This is consistent with the results reported in previous work.<sup>11</sup> It is interesting that the values of  $N_1$  for the blend melt at a blending ratio of 50/50 are the smallest. The first normal stress difference reflects the nonisotropy in the flow of viscoelastic fluids. It may be, therefore, be that the viscous dissipation of energy for the blend melt is not too obvious in the die extrusion in this case, especially at the inlet flow (see Fig. 1).

Figure 8 illustrates the relationship between  $B$  and  $N_1$  of the sample melts at  $180^\circ\text{C}$ . Similarly,  $B$  increases linearly with increasing  $N_1$ . It can be also seen that  $B$  reaches a maximum value at  $\phi_{\text{HD}} = 50\%$ . As stated above, the viscous dissipation of energy is relatively less while the elastic energy stored in the melt is relatively more in this case. Hence, the elastic behavior for this melt is relatively high during extrusion flow, leading to the maximum value of the die-swell ratio.

#### CONCLUSIONS

Under the experimental conditions, the die-swell ratios of LDPE/HDPE blend melts were shown to be approximately linear functions of  $\tau_w$  and  $\Delta P_{\text{end}}$ , while  $\Delta P_{\text{end}}$  was a nonlinear function of  $\tau_w$ .  $B$  decreased linearly with increasing  $L/D$  and decreased with increase of the test temperatures. The temperature sensitivity of  $B$  is weakened with the addition of  $\dot{\gamma}_a$  when  $\phi_{\text{HD}}$  is constant.

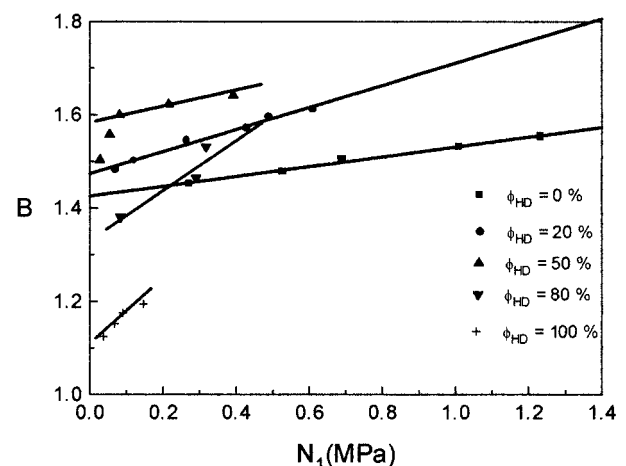


Figure 8 Relationship between  $B$  and  $N_1$  ( $L/D = 30$ ,  $180^\circ\text{C}$ ).

When the test temperature was fixed, the melt  $B$  achieved the maximum value at a blending ratio of 50/50, suggesting that the synergistic effect in the extrudate swell is significant for the melts within a  $\phi_{HD}$  scope from 20 to 60% under these test conditions.

The first normal stress difference increased linearly with increasing  $\tau_w$ . Similarly,  $B$  also increased linearly with the addition of  $N_1$  and reached a maximum value at a  $\phi_{HD}$  of 50%. This phenomenon may be explained by using the theory of viscoelastic competition between components of the polymer blend melts.

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