

# **Evaluation of Die Swell Behavior During Capillary Extrusion of Poly(lactic acid)/ High density polyethylene Blend Melts**

**Fadi Alzarzouri<sup>(\*)</sup> and Fawaz Deri<sup>(\*\*)</sup>**

(\*)Physics Department, Higher Institute for Applied Sciences and Technology, Damascus, Syria.

(\*\*)Chemistry Department, Laboratory of Materials Rheology, Damascus University, Damascus, Syria.

E-mail: [fadi.alzarzouri@gmail.com](mailto:fadi.alzarzouri@gmail.com)

**Abstract.** Poly lactic acid (PLA) and High density polyethylene (HDPE) have been blended in a Brabender plastograph in the molten state. The melt die swell of the blends has been studied by using a capillary rheometer. Effects of the capillary dimensions, shear stress, shear rate, temperature and blending ratio on die swell of PLA/HDPE blend melts were investigated. The results showed that die swell ratio decreased with increasing of capillary length and temperature while it increased with increasing of shear stress, shear rate and capillary diameter. It was also found that the plots of die swell versus blending ratio go through a maximum for blending ratio PLA/HDPE (60/40).

**Keywords.** PLA, HDPE, blend melts, die swell, capillary.

## **1. Introduction**

Swelling characteristics of a polymer melt is an important parameter for characterization of elastic properties in an extrusion process [1]. When the polymer melt flows through capillary or forming mold, the particles are released from the stresses affecting them which leads to the occurrence of the phenomenon of die swell or Barus effect [1]. Die swell must be controlled because the extruded polymer product must have specific dimensions that are compatible with the applications [2]. many studies utilized capillary rheometry to investigate effects of extrusion conditions on the die swell behavior of polymer blend melts [3-7]. Polylactic acid (PLA), as a biodegradable polymer has received particular attention due to the high impact of plastic daily waste on environment [8]. PLA is a linear aliphatic thermoplastic polyester and it has prospects to replace petroleum-based polymers [8]. Since PLA is biodegradable, biocompatible and nontoxic to the human body and the environment it has been widely used in biomedical, agricultural and industrial fields, but a few drawbacks such as high brittleness, low strength, low toughness and poor heat resistance limit the expansion and diversification of PLA's application [9]. In order to encounter this problem, PLA is blended with many thermoplastic polymers to obtain a biodegradable material, good performance in processing, improve the mechanical properties and reduce cost [9]. High density polyethylene is one of the cheaper synthetic polymers with a combination of outstanding physical, chemical, mechanical and thermal properties [10]. PLA was blended with High density polyethylene (HDPE) [11-15] to balance the cost effective issue of PLA and enhance the degradability of HDPE. however, no studies had been published about factors which effect die swell behavior of PLA/HDPE blend melts. The objectives in this work are to investigate the effects of the capillary dimensions, shear stress, shear rate, temperature and blending ratio on die swell of blends PLA/HDPE via capillary rheometry.

## 2. Materials and methods

### 2.1. Materials

Poly(lactic acid) (PLA) (ESUN™ A-1001) [density = 1.25 g/cm<sup>3</sup> (21.5°C), MFI = 12.5 g/10 min (190°C/2.16 Kg)] was supplied by Bright China Industrial Company. Ltd (Shenzhen, China). High density polyethylene (HDPE) (SABIC® HDPEM80064) [density = 0.964 g/cm<sup>3</sup>, MFI = 8 g/10 min (190°C/2.16 Kg)] was supplied by Sabic (KSA).

### 2.2. Blends Preparation

The PLA/HDPE blends were prepared with weight blending ratio of 0/100, 10/90, 20/80, 30/70, 40/60, 50/50, 60/40, 70/30, 80/20, 90/10 to 100/0. The PLA/HDPE blends were mixed in the molten state using Brabender plastograph. The blending time was 15 min at 190 °C and screw speeds of 30 rpm.

### 2.3 Methods

Elastic properties of the blends were studied in term of die swell ratio using capillary rheometer. Part of experiments were carried out at 180°C and using capillaries with constant diameters (2 cm) and different lengths where Length/Radius L/R = 8, 15, 25 and 36, where another part were carried out at 170°C, 180°C and 190°C and using capillaries with constant lengths (15 cm) and different diameters (1, 1.5, 2 and 3 cm). Apparent shear rate ( $\gamma_a$ ) was determined by the following equation [7]:

$$\gamma_a = \frac{4Q}{\pi R^3} \quad (1)$$

where Q- volumetric flow (cm<sup>3</sup>/s), R- radius of the used capillary die (cm). Apparent shear stress ( $\tau_a$ ) was determined by the following equation [7]:

$$\tau_a = \frac{P \cdot R}{2 \cdot L} \quad (2)$$

where P is the pressure at the capillary entrance (Pa), L is the capillary length (cm).

After measuring The extrudate diameter D (mm), the die swell ratio (B) was determined by the following equation [7]:

$$B = \frac{D}{D_0} \quad (3)$$

Where, D<sub>0</sub>- die diameter (mm).

## 3. Results and Discussion

### 3.1. Torque curves

Figures (1 to 3) show the torque curves as a function of blending time for some polymer blends obtained during processing in the Brabender mixer.

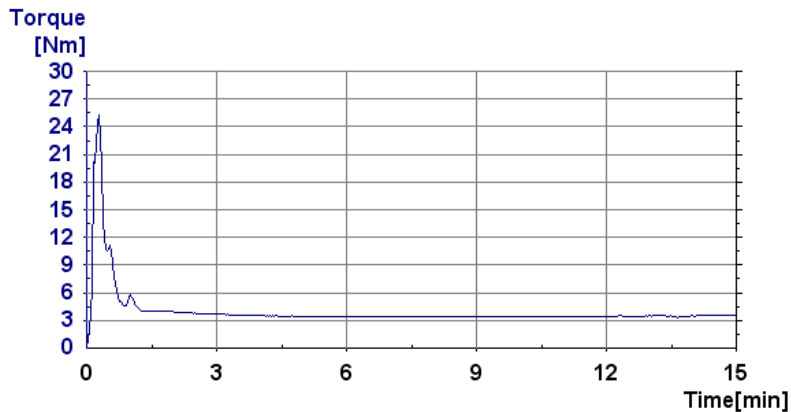


Figure 1. Torque curve of PLA/HDPE (20/80)

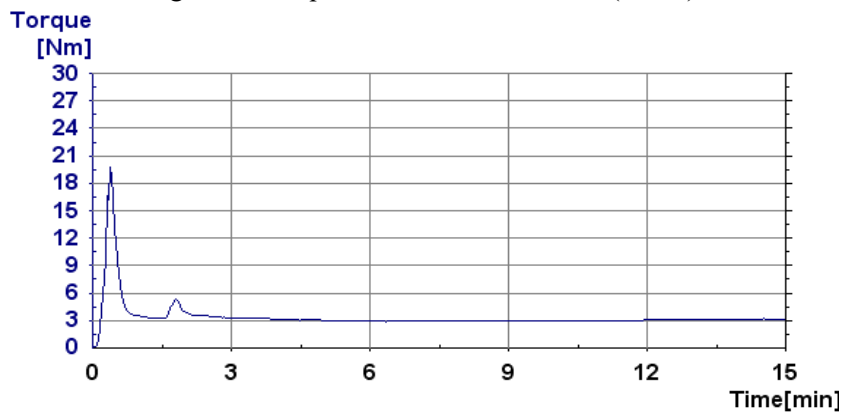


Figure 2. Torque curve of PLA/HDPE (50/50)

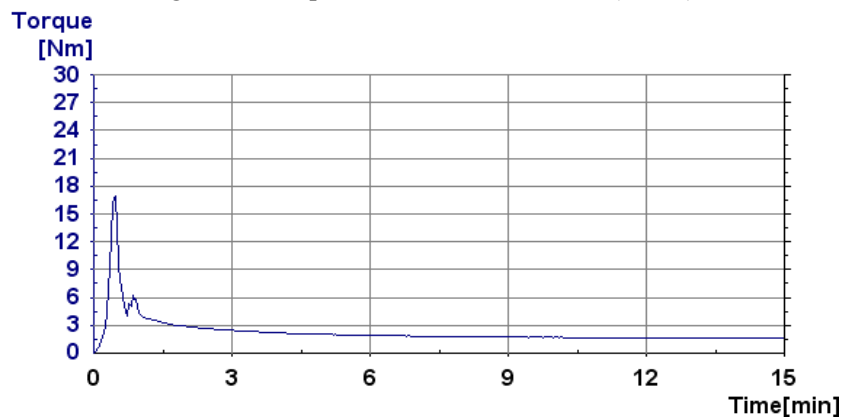


Figure 3. Torque curve of PLA/HDPE (80/20)

When placing PLA/HDPE blends into the blending chamber, the components are gradually aggravated by the heating temperature and the rotation of the screw, accompanied by an increase in torque, and then decrease and stability due to the homogeneity of the temperature and the polymer blend. Torque observation during the melt blending and homogenization of polymer blends allows continuous monitoring of the torque change in the blending system. It can be seen that all curves have nearly a similar profile. Increasing the torque at the beginning of the blending process indicates an increase in the viscosity of the polymer blend and the difficulty of the blending process and then the decrease and stability of the torque after a short time of 2-3 min, where the viscosity decreases with the continuity of the blending and homogenization process.

### 3.2. Relationship between B and L/R

Figures 4 and 5 show the relationship between B and L/R for PLA/HDPE blend melts at constant load (3 Kg) and when test temperature is 180 °C.

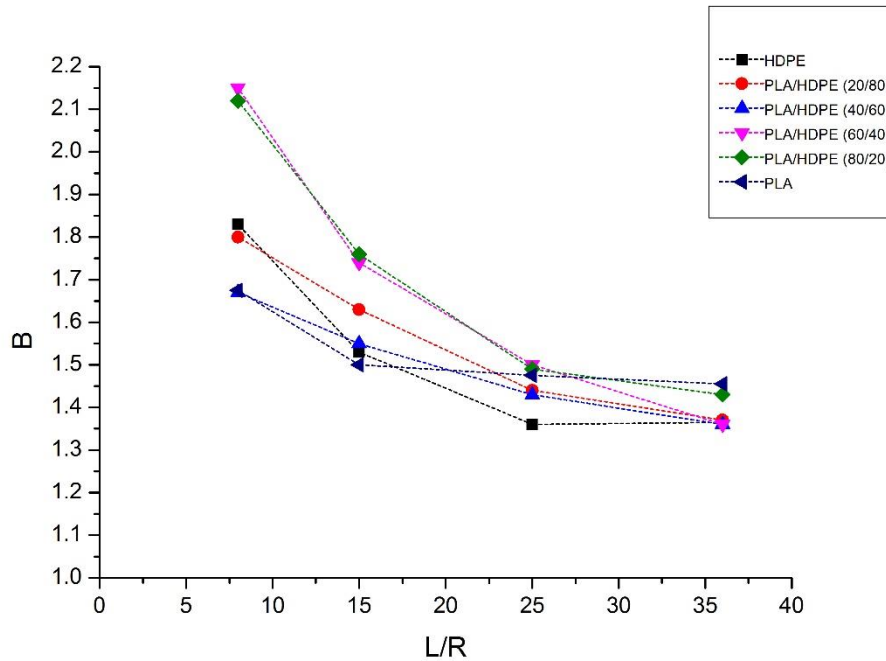


Figure 4. Relationship between B and L/R of (PLA, HDPE, PLA20%, PLA40%, PLA60%, PLA80)

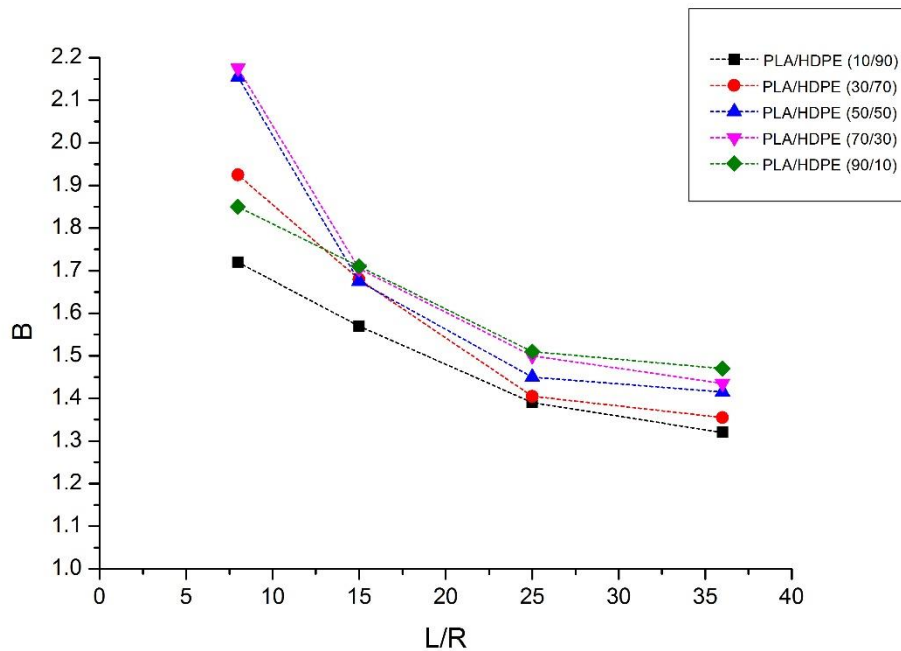


Figure 5. Relationship between B and L/R of (PLA10%, PLA30%, PLA50%, PLA70%, PLA90%)

It can be seen that, the values of B decrease with increasing L/R ratio of the capillary. It can also be seen that the sensitivity of the die swell to L/R for HDPE is much more obvious than that for PLA.

Increasing the die length/diameter ratio reducing the remaining stresses in the melts when they leave from the channel which leads to a decrease in the rate of melt swelling [4]. J.Z.Liang et al. [4] studied the elastic properties of LDPE/LLDPE blends in the molten state. The results showed that the swelling ratio (B) of these blends decreased with increasing capillary die length/diameter ratio.

M. Kaseem et al. [7] also studied the elastic properties of the PVDF/LDPE blends in the molten state. The results showed that (B) of these blends decreased with increasing L/R.

### 3.3. Relationship between B and die diameter

Figure 6 shows the relationship between B and capillary diameter for PLA, HDPE and PLA/HDPE (50/50) blend at constant load (3 Kg) and when temperature is 180 °C and capillary length is 15 cm.

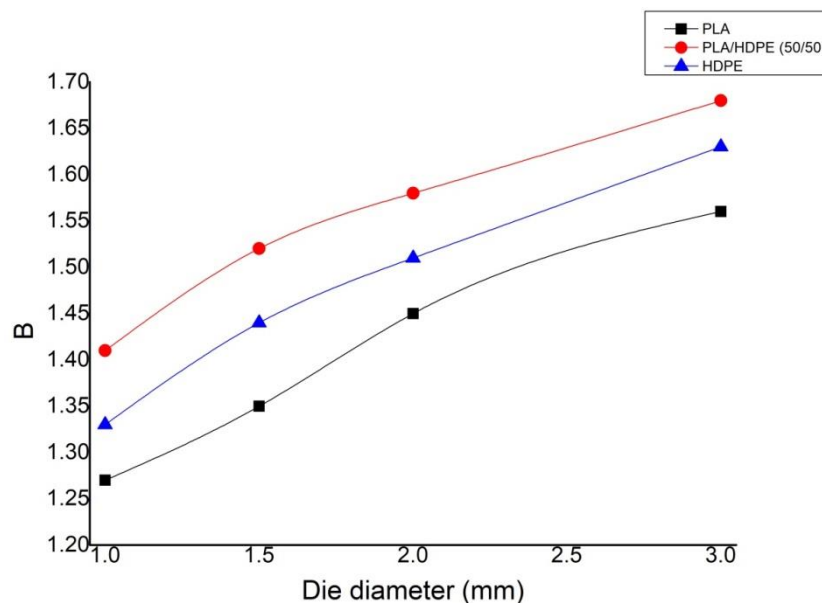


Figure 6. Relationship between B and capillary diameter

It can be seen that, the swelling ratio of PLA, HDPE and the PLA/HDPE (50/50) blend increases with increasing capillary diameter. This phenomenon is due to decreasing the time that the polymer melt resides inside the capillary channel with the increase in capillary diameter and that cause reducing the time of stress relaxation of the melt when leaving the capillary channel, which leads to an increase in the rate of swelling [6]. This result is similar to that has been reported by J.Z.Liang et al [6].

### 3.4. Dependence of B on shear stress and shear rate

Figures 7 and 8 display the relationship between B and both shear stress and shear rate respectively for PLA/HDPE blend melts when test temperature is 180 °C.

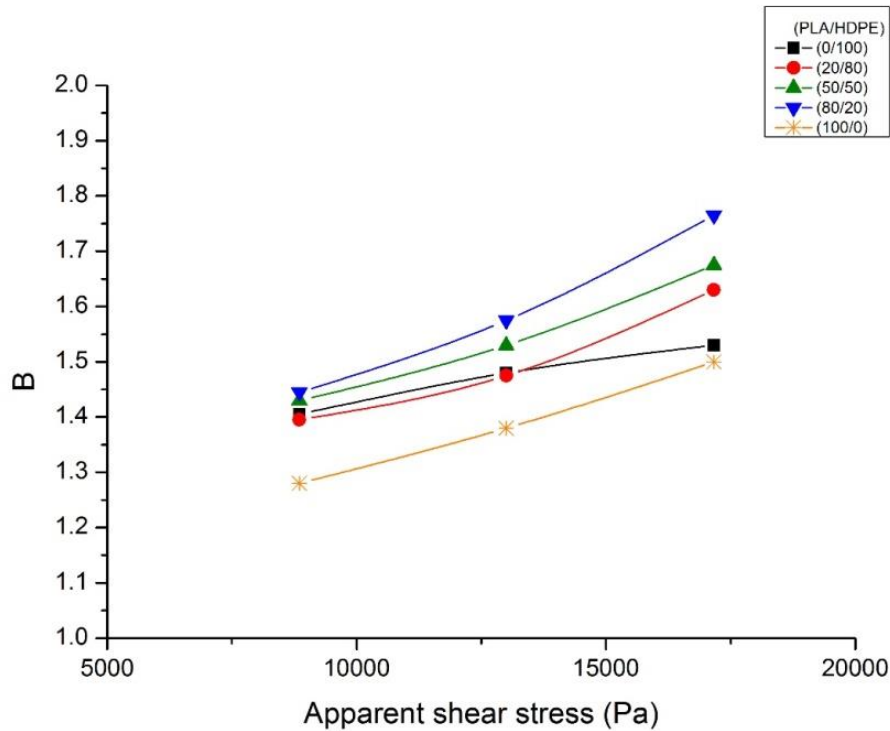


Figure 7. Relationship between B and *shear stress* of (PLA, HDPE, PLA20%, PLA50%, PLA80%)

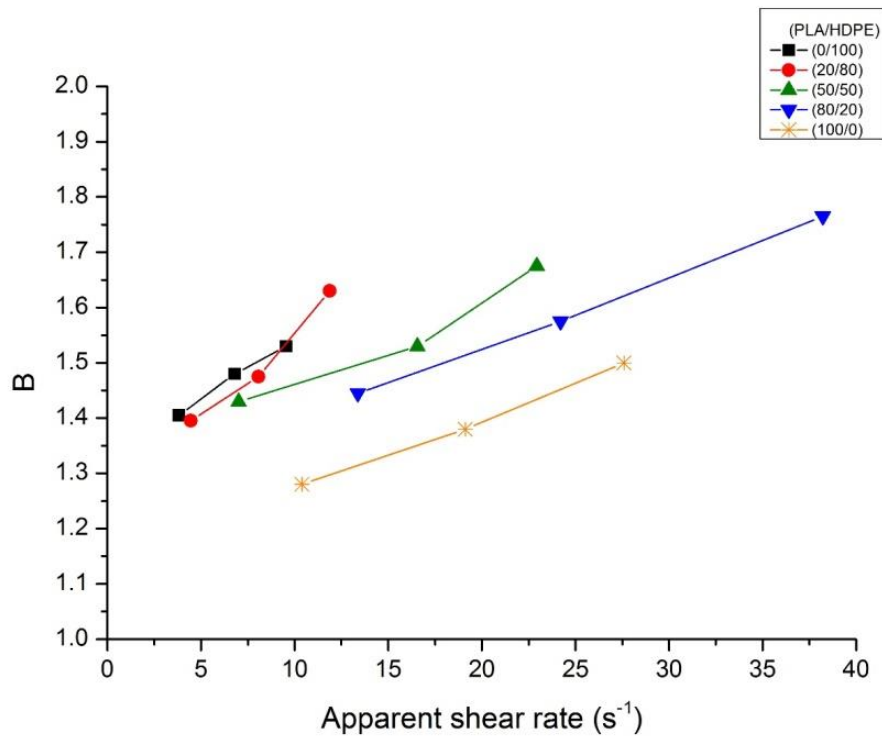


Figure 8. Relationship between B and *shear rate* of (PLA, HDPE, PLA20%, PLA50%, PLA80%)

It can be seen that die swell ratio increases with increasing both shear stress and shear rate. Deformational energy stored in the melts increases by increasing shear stress or shear rate which leads

to a increase in the rate of melt swelling. W. Sinthavathavorn et al. [5], studied the elastic properties of the PA6 / LDPE blends in the molten state, and it was found in this study that the rate of swelling (B) of these blends increased with both shear stress and shear rate.

### 3.5. Dependence of B on temperature

Figure 9 shows the plots of die swell versus temperature for PLA, HDPE and PLA/HDPE (50/50) blend at constant load (3 Kg) and using capillary with dimension Length/Radius L/R = 15/1.

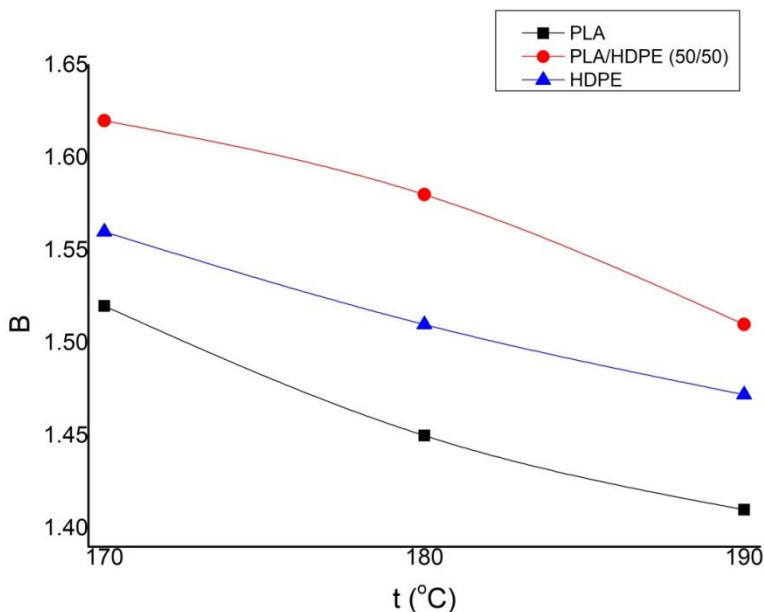


Figure 9. Relationship between B and temperature

It can be seen that, the swelling ratio of PLA, HDPE and the PLA/HDPE (50/50) blend decreases with increasing temperature. when the temperature increases the melt free volume increases which reduction interactions and friction force between polymer chains [16], which may leading to a decrease the remaining stresses in the melts and this leads to decreased swelling ratio. The same behavior was noted in LDPE/PP blend [3].

### 3.6. Effect of Composition on B

Figure 10 displays the effect of the blend composition on the melt die swell at constant shear stress when test temperature and L/R are 180 °C and 15, respectively. It can be seen that the curves of B versus blending ratio go through a maximum for blending ratio PLA/HDPE (60/40) and it may be due to the viscoelasticity difference between the two phases and a maximum value of dispersive phase in blending ratio PLA/HDPE (60/40) which be able to recover a larger fraction of the energy than the other blends.

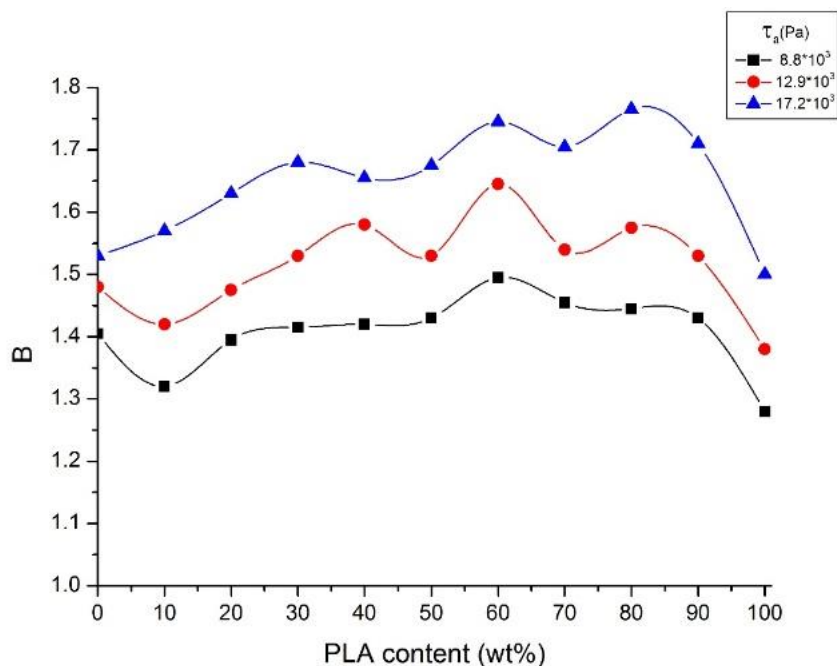


Figure 10. Effect of blending ratio on B (L/R =15, t =180°C)

#### 4. Conclusion

The present study has investigated the effect of many factors on melt die swell ratio of PLA/HDPE blends using a capillary rheometer. The die swell was found to decrease with an increase in the capillary length and with a rise of temperature. With increasing capillary diameter, shear stress and shear rate that die swell ratio increases. It was also found that the die swell of PLA/HDPE blends reaches a maximum at PLA = 60%.

#### References

- [1] A.R.Mendelson, L.F.Finger, B.E.Bagley: Die swell and recoverable shear strain in polyethylene extrusion, *Journal of Polymer Science Part C: Polymer Symposia*, 35(1), 177-188 (1971).
- [2] K. Wang: Description of extrudate swell for polymer nanocomposites, *Materials*, 3(1), 386-400 (2010).
- [3] J.Z. Liang, N.J. Ness: The melt die-swell behaviour during capillary extrusion of LDPE/PP blends, *Polymer testing*, 17(3), 179-189 (1998).
- [4] J.Z. Liang: The elastic behaviour during capillary extrusion of LDPE/LLDPE blend melts. *Polymer Testing*, 21(1), 69-74 (2002).
- [5] W. Sinthavathavorn, M. Nithitanakul, P.B. Grady, R. Magaraphan, R: Melt rheology and die swell of PA6/LDPE blends by using lithium ionomer as a compatibilizer, *Polymer bulletin*, 63(1), 23-35 (2009).
- [6] J.Z. Liang: Effects of extrusion conditions on die-swell behavior of polypropylene/diatomite composite melts, *Polymer testing*, 27(8), 936-940 (2008).
- [7] M. Kaseem, K. Hamad, W. H. Yang, H.Y. Lee, F. Deri, F, G.Y. Ko; Melt rheology of poly (vinylidene fluoride)(PVDF)/low density polyethylene (LDPE) blends, *Polymer Science Series A*, 57(2), 233-238 (2015).
- [8] A.R. Auras, T.L. Lim, E.S. Selke, H. Tsuji, H. (Eds.): Poly (lactic acid): synthesis, structures, properties, processing, and applications, John Wiley & Sons, Vol.10 (2011).
- [9] M. Nofar, D. Sacligil, J. P. Carreau, R.M. Kamal, C.M. Heuzey: Poly (lactic acid) blends: Processing, properties and applications. *International journal of biological macromolecules*, 125, 307-360 (2019).

- [10] C. Vasile, M. Pascu, M: Practical guide to polyethylene, iSmithers Rapra Publishing (2005).
- [11] X. Lu, J. Huang, B. Kang, T. Yuan, P.J. Qu: Bio-based poly (lactic acid)/high-density polyethylene blends as shape-stabilized phase change material for thermal energy storage applications. *Solar Energy Materials and Solar Cells*, 192, 170-178 (2019).
- [12] A. Machado, I. Moura, F. Duarte, G. Botelho, R. Nogueira, G.A. Brito: Evaluation of properties and biodeterioration potential of polyethylene and aliphatic polyester blends. *International Polymer Processing*, 22(5), 512-518 (2007).
- [13] G. Madhu, H. Bhunia, K.P. Bajpai, V. Chaudhary: Mechanical and morphological properties of high density polyethylene and polylactide blends. *J Polymer Eng*, 34(9),813-821 (2014).
- [14] G. Madhu, H. Bhunia, K.P. Bajpai: Blends of high density polyethylene and poly (L-lactic acid): mechanical and thermal properties. *Polymer Engineering & Science*, 54(9), 2155-2160 (2014).
- [15] I. Moura, A. Machado, F. Duarte, G. Botelho, R. Nogueira: Biodegradability Assessment of Aliphatic Polyesters-Based Blends Using Standard Methods. *J Appl Polymer Sci*, 119:3338-3346 (2011)
- [16] A.T. Saki, T. A. (2015). Reactive melt blending of low-density polyethylene with poly (acrylic acid). *Arabian Journal of Chemistry*, 8(2), 191-199 (2015).