

Rheological Properties and Melt Strength of LDPE During Coextrusion Process

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ABSTRACT: Rheological properties and mechanical strength of coextruded low-density polyethylene (LDPE) were studied in this work. The influences of die temperature, shear rate at skin and core layers, and number of extrusion passes at the core layer were of interest. The experimental results suggested that the viscosity and swelling behavior of LDPE coextrudate were more dependent on the shear rate of skin layer as compared with that of core layer. Increasing die temperature resulted in decreases in viscosity and die swell ratio. The reductions in viscosity and die swell ratio of the coextrudate for the reprocessing numbers 1 and 2 at the core layer arose from the disentanglement of long chain branching of LDPE molecules, while the increase

of viscosity and die swell ratio for the reprocessing numbers 3 and 4 mainly involved an occurrence of crosslinking structure. For mechanical strength results, the changes in elongational stress for different shear rates, die temperature, and number of extrusion passes were in line well with the rheological measurements. That was, the higher the stored energies in polymer melts due to higher shear rates, the greater the amount of force required to conquer the elastic resistance. © 2012 Wiley Periodicals, Inc. *J Appl Polym Sci* 125: 2187–2195, 2012

Key words: coextrusion; processing parameters; rheological properties; melt strength; number of extrusion passes

INTRODUCTION

Coextrusion is one of the most important processes that have the possibility of producing high functional products. By using appropriate combination of materials such as strength, barrier properties, color, and other features of different polymers, this technique allows the processor to combine the satisfactory properties of multiple polymers into a single multilayered structure with enhanced performance properties. It is often desirable for the skin material to have a superior appearance or scratch resistance, while the strength or permeability of the part is strongly dependent upon the core material. In addition, the foamed and/or recycled core materials can

be employed as the core layer, which yields the potential weight reductions and cost savings.¹ Pioneered investigations^{2,3} on the interfacial configurations and viscous properties of two molten polymers, flowing side by side either through a circular or rectangular die, so called bicomponent coextrusion, were reported by White et al.² and Han et al.³ Their findings indicated that the more viscous component tended to be convex at the interface while the less viscous component tended to preferentially warp around the more viscous component. It was found that the pressure gradient of the bicomponent was somewhere between the pressure gradients of the two individual components and the effect of stretching did not appear to influence the interface shape. White et al.² also suggested that the shape of the interface was influenced by the shape of the die cross-section, the component ratio of the two melts, the length-to-diameter ratio (L/D) of a capillary die, and the viscoelastic properties of the individual fluids concerned. The effects of polymer viscoelasticity and cross-sectional shape of die on the layer thickness uniformity of coextruded structures were investigated by Dooley et al.⁴ They concluded that the occurring secondary flow within different die geometries, due to the viscoelastic characteristics of the

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polymer, resulted in the interface rearrangement. The influences of processing variables on the layer deformation and interfacial instability during three-layer film coextrusion were studied by Martin and Averous.⁵ The plasticized wheat starch was chosen as the core layer and poly(esteramide) was used as the skin layer. They indicated that the stability of coextrudate was closely related to shear stress at the interface as a result of increasing volumetric flow rate and the die gap geometry. Recently work on coextrusion of polymer composites carried out by Yao and Wu⁶ focused on the effects of wood loadings, core quality, and thickness of skin layer on mechanical and water absorption properties of the coextruded composites of recycled polyethylene and wood flours. They found that the coextruded composites had significantly better flexural and impact strengths, long-term moisture resistance, and dimensional stability in comparison with the corresponding single structure individual.

Measurement of elongational melt strength provides very useful information for extrusion process, such as tubular blown film, blow molding, wire coating, and melt spinning in order to prevent the sagging and fracture of the extrudate during processing. The most widely used and important device for the melt strength and transient extensional viscosity measurements is so-called "Rheotens".⁷⁻⁹ Recently published works by Sombatsompop et al.¹⁰⁻¹² proposed an experimental device which was specially designed and constructed in a similar fashion to the Rheotens to determine the melt strength online in a single screw extruder using a wide range of polymer systems including homopolymers,^{10,11} polymer blends and composites¹² and processing and reprocessing conditions.^{10,11} They suggested that the draw-down force of molten LDPE was dependent on volumetric flow rate, die temperature, roller speed, take-up style, and the number of extrusion passes. For the rapid speed take-up method, the drawdown forces of the molten LDPE were approximately 40–60% greater than that of step-ladder take-up style. It was also found that the drawdown force was greatly affected by the number of extrusion passes and related to the chemical changes in the extrudates. Although a thorough search through the existing literature reveals many references to the uniaxial elongational flow properties of various polymer systems, these including homopolymer,^{8,13} polymer blends,¹⁴ and composite systems.¹⁵ However, as for the coextrudates, knowledge of melt strength and their dependencies on various process conditions remains elusive. Therefore, in this study, the melt strength of coextruded LDPE were investigated with respect to the processing parameters, including die temperature, shear rate of skin and core layers, and strain rate. Furthermore, the effect of number of reprocessing at the core layer

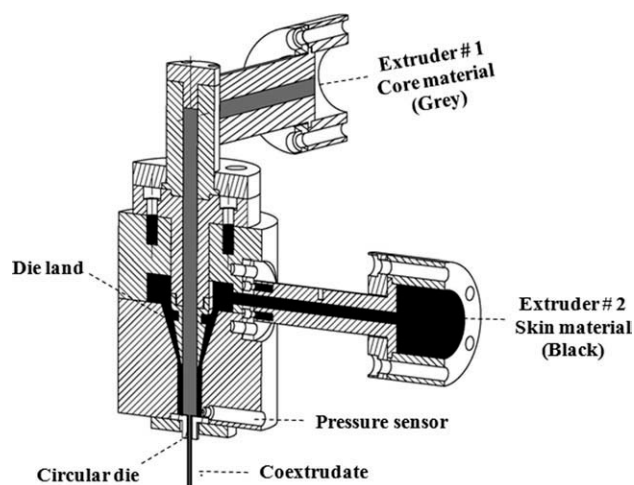


Figure 1 A feed-block unit on both of single screw extruders for coextrusion process of LDPE.

was also one of our main interests. The fundamental rheological properties, such as viscosity and swelling behavior, were also examined to understand the rheological changes in the LDPE coextrudates under the interested parameters.

EXPERIMENTAL

Raw materials

This study used commercially available grade of low density polyethylene with a melt flow rate of 5 g/10 min (LD1905FA-LDPE, supplied by Thai Polyethylene, Bangkok, Thailand). To investigate the effect of number of reprocessing passes at the core layer on the rheological properties and melt strength of coextruded LDPE, the as-received virgin LDPE was reprocessed several times (from 1 to 4) by one of the single screw extruders using a die temperature of 140 °C and a screw rotation speed of 11 rpm. The in-plant recycled granules of LDPE were designated as LDPE number 1, LDPE number 2, LDPE number 3, and LDPE number 4, respectively.

Experimental rig

An experimental coextrusion rig was designed and manufactured in this work in order to allow the measurements of rheological properties and mechanical strength of LDPE coextrudates. The schematic diagram of the coextrusion rig is shown in Figure 1. In the experiments, the feed-block unit was specially designed and constructed and employed for the rheological measurement of molten LDPE during coextrusion processes. The coextruded feed-block unit was fed by two separated single screw extruders (Extruders # 1 and # 2) with compression ratio of 4:1.2 and length-to-diameter of 20 ($\phi = 25$ mm). The feed-block unit had a straight-through

section for the mainstream of core material (gray) and the coextruded stream was introduced through a side port partway through the land length, which distributed the skin material (black) around the core material. The combined two-molten polymer, namely coextrudate, was then extruded through a circular die 5 mm in diameter (L/D ratio = 4). A pressure transducer (Dynisco, Model PT460E-2CB-6, Franklin, MA) was located near the die entrance for the pressure drop (ΔP) measurement.

Rheological characterization

The rheological properties of coextruded LDPE were characterized by measuring their true viscosity (η_{true}) and swelling ratio (B), according to the recommendation of ASTM D 3835-02 (Standard test method for determination of properties of polymeric materials by means of a capillary rheometer). The core material was colored by an addition of a small amount (about 2 wt %) of pigment in order to facilitate the identification of the interface between skin and core layer. All the tests were performed under isothermal condition where the melt temperatures from both extruders were equal and had attained a steady state in accordance with the preset temperatures. The three heating zones (from feed to die zones) were set at 130, 135, and 140°C, respectively. The die temperature was varied from 140 to 160°C.

In this work, the Bagley and Rabinowitsch corrections were applied for obtaining true shear stress (τ) and true shear rate ($\dot{\gamma}$), respectively. These two values were then calculated to give true shear viscosity (η_{true}) of the coextruded LDPE using eqs. (1)–(3). The entrance corrections can be determined by experimentally using a series of dies of the same diameter but different lengths (from 20 to 40 mm). These correction results were obtained at various shear rates of skin and core layers of LDPE. The total volumetric flow rate (Q_{total}) was measured by mass throughput of the individual extruder.¹⁶ The volumetric flow rate of skin (Q_{skin}) and core layers (Q_{core}) were varied from 1.5 to $4.6 \times 10^{-7} \text{ m}^3 \text{ s}^{-1}$, this being achieved by altering the screw rotation speeds.

$$\text{True shear stress} = \tau = \frac{R(\Delta P)}{2(L + eR)} \quad (1)$$

$$\text{True shear rate} = \dot{\gamma} = \left(\frac{3n + 1}{4n} \right) \left(\frac{4Q_{\text{total}}}{\pi R^3} \right) \quad (2)$$

$$\text{True viscosity} = \eta_{\text{true}} = \frac{\tau}{\dot{\gamma}} \quad (3)$$

where R is the radius of the die (m), ΔP is the pressure drop along the die (N m^{-2}), L is the length of the die (m), e is the extension of the imaginary

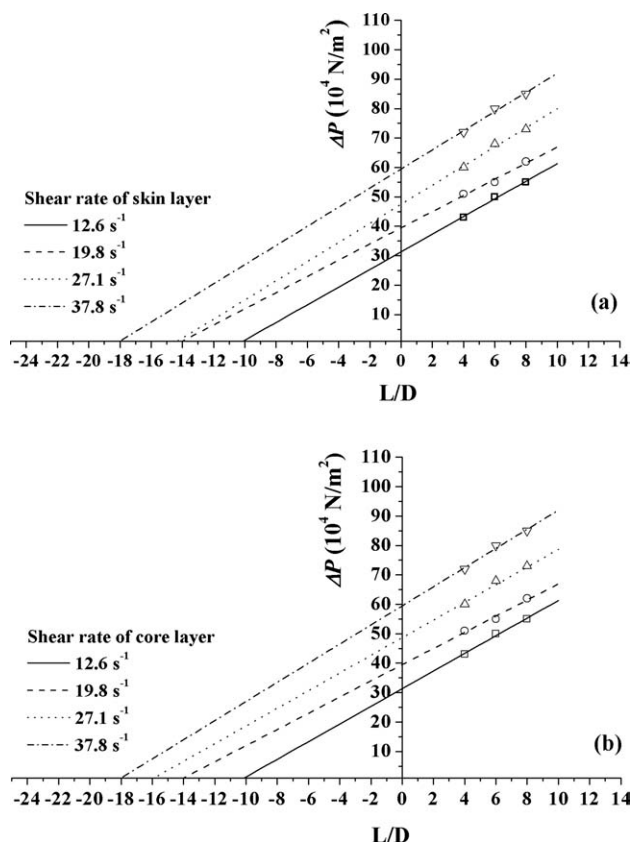


Figure 2 Plots of pressure drop and L/D ratio for LDPE coextrudates at various shear rate of (a) skin and (b) core layers.

length of the die, Q is the volumetric flow rate along the capillary die ($\text{m}^3 \text{ s}^{-1}$) and n is the power law index, which can be determined from the slope of the plot between $\log(\tau)$ against $\log(\dot{\gamma})$. The average value of n used in this work was determined to be 0.59.

The plots of ΔP against L/D for coextruded LDPE at different shear rates of skin and core layers are shown in Figure 2(a,b). From the results, it can be seen that the ΔP increased with increasing die length for a given shear rate of skin and core layers, and the relationship between ΔP and L/D was linear. The extrapolations were carried out using a linear least square best line fit in order to obtain the e values.¹⁷

Melt strength measurement

Figure 3 illustrates an experimental arrangement for melt strength measurement of the coextruded LDPE which used the coextrusion rig from Figure 1 coupled with an extension of the melt strength measuring unit, roller take-up device, visualization devices, and temperature control equipment. The experimental set-up and factions of these components were already detailed and given in previous

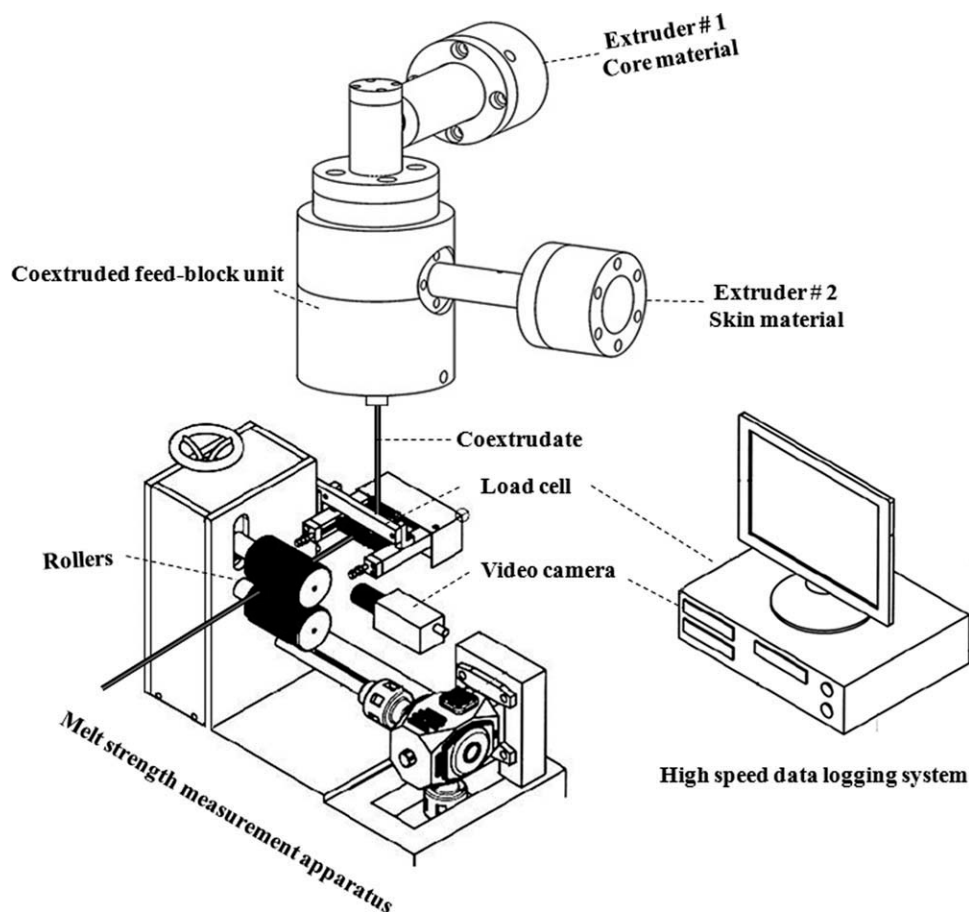


Figure 3 An experimental arrangement for melt strength measurement of coextruded LDPE.

works.^{10,11} The drawdown force (F) as a function of time was required for accurate measurements. The molten LDPE was coextruded through a capillary die in the form of continuous strand before being pulled down by the rollers and the take-up device until the coextrudate was failed. The drawdown force was recorded using a high speed data-logging and recording system. The rollers (take-up) speed was also varied from 50 to 325 rpm. The spin-line (distance from die to take-up rollers) was set at 150 mm and the coextrudate was cooled down by ambient cooling. In this work, the elongational stress (σ_e) and elongational strain (ϵ_e) of the coextruded filament of LDPE were reported to determine the mechanical strength of LDPE melt, the calculations being expressed by eqs. (4) and (5).

$$\sigma_e = \frac{Fv_1}{Q_{\text{total}}} \quad (4)$$

$$\epsilon_e = \ln \frac{v_1}{v_0} \quad (5)$$

where F is the drawdown force at the take-up rollers (cN), v_1 is the velocity at the rollers (m s^{-1}) and v_0 is the velocity at die (m s^{-1}).

RESULTS AND DISCUSSION

Rheological properties of coextruded LDPE

The influences of die temperature on the true viscosity (η_{true}) and swelling ratio (B) of coextruded virgin LDPE at a given core/total shear rate ($\dot{\gamma}_{\text{core}}/\dot{\gamma}_{\text{total}}$) are shown in Figures 4 and 5, respectively. The shear rate of skin layer ($\dot{\gamma}_{\text{skin}}$) was kept at 23.2 s^{-1} while the shear rate of core layer ($\dot{\gamma}_{\text{core}}$) was varied from

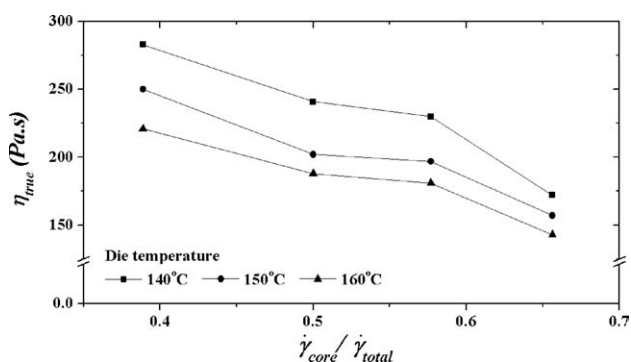


Figure 4 Variations in true viscosity for coextruded virgin LDPE as a function of $\dot{\gamma}_{\text{core}}/\dot{\gamma}_{\text{total}}$ at various die temperatures.

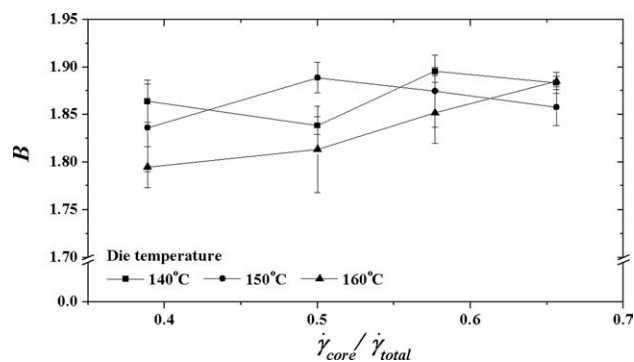


Figure 5 Variations in swelling ratio for coextruded virgin LDPE as a function of $\dot{\gamma}_{core}/\dot{\gamma}_{total}$ at various die temperatures.

14.7 to 44.2 s^{-1} . It was evident that the molten LDPE coextrudates exhibited a pseudoplastic non-Newtonian behavior, in that the viscosity of coextruded LDPE decreased with increasing $\dot{\gamma}_{core}$. Increasing die temperature also resulted in a significant decrease in the coextrudate viscosity. Figure 5 shows that die swell was a strong function of temperature and shear rate, as expected. The swelling ratio of the coextruded LDPE appeared to decrease with the increase of die temperature while tended to increase with increasing shear rate. The obvious latter effect was seen for the die temperature of 160°C. In the case die temperatures of 140 and 150°C, the swelling ratio was not significantly affected by increasing shear rate of core layer. This was due to higher shear heating effect for the melt flowing in the die with lower temperatures (i.e., higher material viscosity was expected).^{18,19} This finding appeared to agree with the viscosity results in Figure 4. That was, a significant reduction in the viscosity for coextruded LDPE at high shear rate of core layer was observed at the die temperature of 140°C as compared to that obtained at die temperatures of 150 and 160°C.

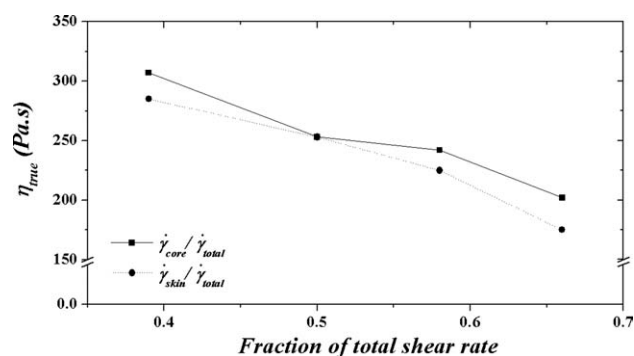


Figure 6 Flow curves for coextruded virgin LDPE at different shear rate of skin and core layers.

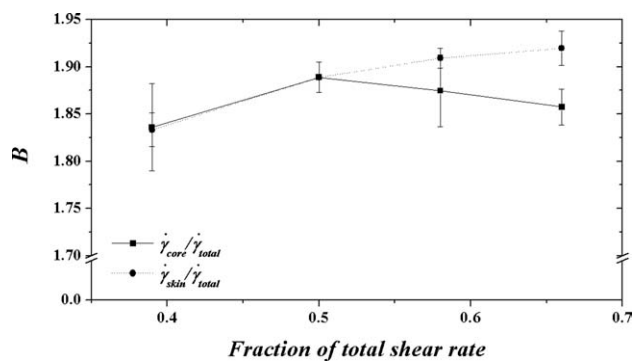


Figure 7 Die swell ratio (B) for coextruded virgin LDPE at different shear rate of skin and core layers.

The changes of $\dot{\gamma}_{skin}$ and $\dot{\gamma}_{core}$ on the true viscosity and swelling ratio of coextruded virgin LDPE are depicted in Figures 6 and 7. It should be noticeable in Figure 6 that as the $\dot{\gamma}_{skin}$ was increased, especially at high shear rate, the viscosity of coextrudate decreased sharply as compared with those obtained by increasing the $\dot{\gamma}_{core}$. This was probably associated with a considerable shear heating occurred at the skin layer during the shear flow in the die and its exit. The high shear heating at the skin layer could be explained by considering the development of velocity profiles of a polymer melt during shear flow as schematically illustrated by Figure 8. That was, when a fully developed melt was flowing in the die (Point A) it had a relatively high velocity at the center and relatively low near the die wall. On exiting the die (Point B), the velocities of the melt across the die diameter had to equalize to form a plug flow.¹ During the transition of velocity pattern the melt velocity near the wall, usually has relatively high shear rate, had to accelerate to compete with the melt at the center and this could generate a considerably high shear heating and thus affected the bulk viscosity of the melt.²⁰ On the contrary, if the $\dot{\gamma}_{core}$ was

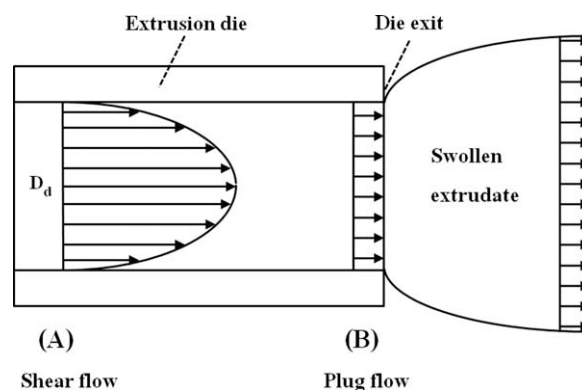


Figure 8 A schematic illustration of velocity profile development of flowing melt in and out of die.

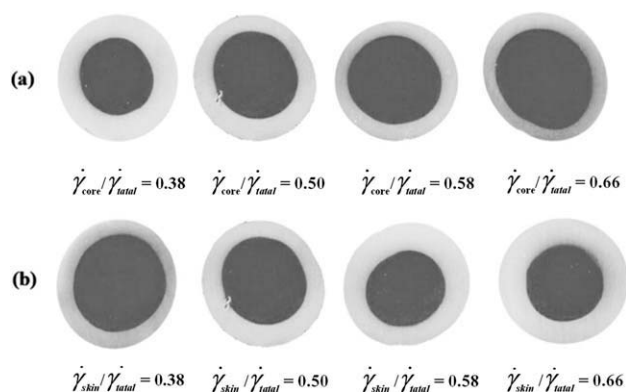


Figure 9 Optical micrographs showing the LDPE coextrudates at various shear rate of skin or core layers (a) Increasing shear rate of core layer and (b) Increasing shear rate of skin layer.

increased, this effect would expect to be very small as the shear rate of the melt at the centre was relatively low.

When considering the swelling ratio in Figure 7, it was found that the swelling ratio of the LDPE coextrudate progressively increased with increasing $\dot{\gamma}_{\text{skin}}$, whereas the increase of $\dot{\gamma}_{\text{core}}$ did not significantly affect the swelling behavior of coextrudate. The physical swellings of the coextrudates by varying shear rate of core or skin layers are given in Figure 9. The increase of swelling ratio with increasing the $\dot{\gamma}_{\text{skin}}$ was attributed to an increasing amount of the stored elastic energies that were put into the skin layer which had relatively higher shearing stresses generated during the flow in the die, as mentioned earlier. These stored elastic energies would then be released on exiting the die and thus increased swelling ratio of the coextrudate. On the other hand, the unchanged swelling behavior of coextrudate obtained by increasing $\dot{\gamma}_{\text{core}}$ was due to the relatively low shear stresses occurred in core layer.²¹

In this work, recycled LDPE was extruded as the core layer in LDPE coextrudate as usually employed

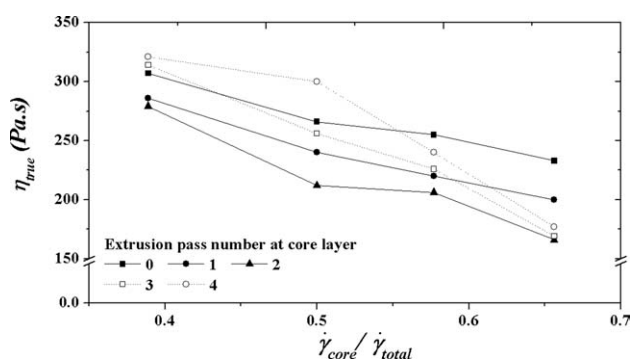


Figure 10 Influence of number of reprocessing at the core layer on viscosity for coextruded LDPE as a function of $\dot{\gamma}_{\text{core}}/\dot{\gamma}_{\text{total}}$.

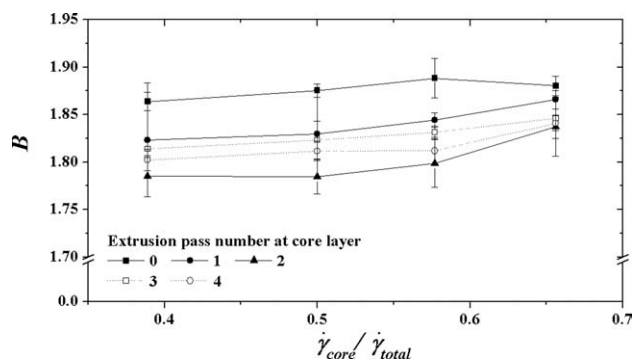


Figure 11 Influence of number of reprocessing at the core layer on swelling ratio for coextruded LDPE as a function of $\dot{\gamma}_{\text{core}}/\dot{\gamma}_{\text{total}}$.

in normal coextruded products for cost saving purposes. Figures 10 and 11 illustrate the influence of number of reprocessing at the core layer on the true viscosity and swelling behavior of coextruded LDPE. In comparison with the as-received virgin LDPE, it was clearly seen for the reprocessing numbers 1 and 2 that the true viscosity and die swell ratio of coextrudates decreased with increasing number of extrusion. The reductions in the viscosity and die swell ratio probably involved a shear modification of long chain branched LDPE at the core layer. That was, the molecular entanglement reduces as the melt experienced the shearing force. This shear modification theory was also stated by Han.²² However, the increments of viscosity and die swell ratio were observed for the reprocessing numbers 3 and 4. This was thought to result mainly from a partial cross-linking effect of LDPE after prolonged thermal history. These explanations can be substantiated using gel content experiment which was carried out by immersion of the reprocessed LDPE in xylene solvent and the determination of gel content was calculated in accordance with the experimental method by ASTM D 2765-01 (2001). The gel content results for coextruded LDPE after extrusion are given in Table I. It can be seen that the gel contents extracted from the reprocessed LDPE samples gradually increased with increasing number of reprocessing, especially for numbers 3 and 4. The significant increases in the insoluble LDPE at the extrusion

TABLE I
Gel Contents for Recycled LDPE Coextrudates as a Function of Number of Extrusion Pass

Number of extrusion pass	Gel Content (%)
1	1.37
2	2.3
3	7.25
4	12.48

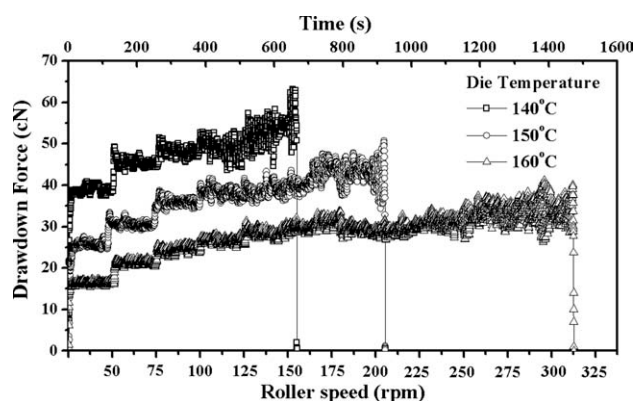


Figure 12 Relationship between measured drawdown forces against time with increasing roller speeds for coextruded virgin LDPE at various die temperatures.

numbers 3 and 4 indicated the occurring some cross-linked molecules. This explains why the viscosity and the swelling ratio of the LDPE coextrudate increased at the reprocessing numbers 3 and 4. A similar observation was also reported in previous studies.^{11,21,23}

Melt strength of coextruded LDPE

The effect of die temperature on the drawdown force of virgin LDPE coextrudate is given in Figure 12 at the $\dot{\gamma}_{\text{core}}/\dot{\gamma}_{\text{total}}$ ratio of 0.5. The experimental results were expressed in terms of drawdown force as a function roller speed and time. In general, at any given die temperature, it can be seen that the drawdown force sharply increased in the initial stage of increasing roller speed (0–50 rpm) and then slowly increased until to the final stage. The significant increase of drawdown force at the initial roller speed was associated with the branching characteristics of LDPE, resulting in the elastic resistances to the applied deformation. This behavior was also in line with previous investigations.^{7,9–11} At higher

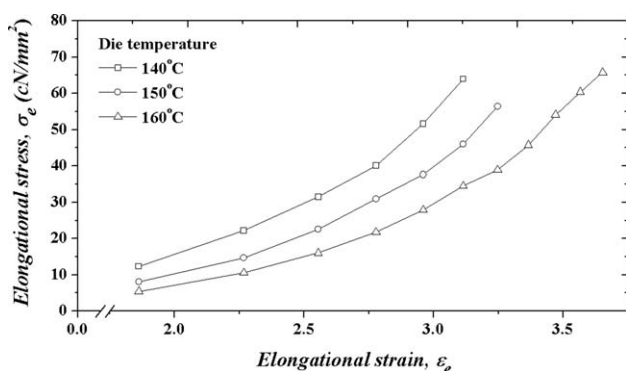


Figure 13 Plots of elongational stress and elongational strain for coextruded virgin LDPE at various die temperatures.

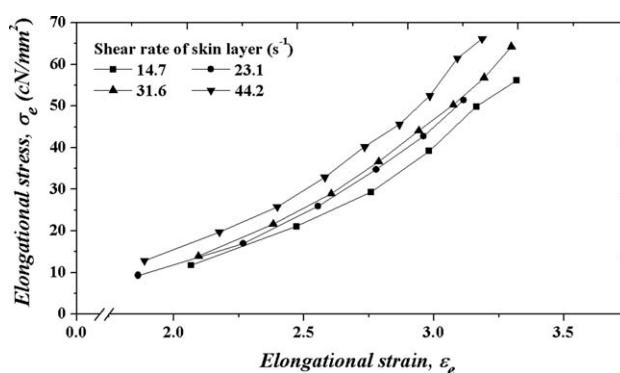


Figure 14 Effect of $\dot{\gamma}_{\text{skin}}/\dot{\gamma}_{\text{total}}$ on elongational stress of coextruded virgin LDPE at die temperature of 150°C.

roller speeds (greater than 50 rpm), the increases in the drawdown force were marginal. This was because the molecular chains of LDPE in this stage became completely disentangled and could easily slip past each other, no further forces being required. Moreover, fluctuation of drawdown force was also observed during the final stage of increasing the roller speed. This was a result of draw resonance effect, which was reported and found by a number of publications.^{7,10–13}

The plots between elongational stress (σ_e) and elongational strain (ϵ_e) for different die temperatures are shown in Figure 13 which could be explained in a similar fashion as given for Figure 12, in that the higher the die temperature, the lower elongational stress and the higher the elongational strain. Increasing the die temperature caused the increases in viscous deformation and chain mobility in the LDPE coextrudates which facilitated the deformation with a lower external stress. Figures 14 and 15 show the influence of shear rate of skin and core layers on the melt strength of virgin LDPE coextrudate. In all cases, especially for an increase of $\dot{\gamma}_{\text{skin}}/\dot{\gamma}_{\text{total}}$ as depicted in Figure 14, the higher the shear rate the

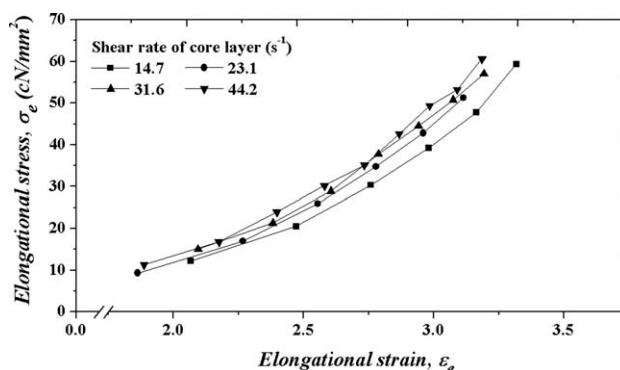


Figure 15 Effect of $\dot{\gamma}_{\text{core}}/\dot{\gamma}_{\text{total}}$ on elongational stress of coextruded virgin LDPE at die temperature of 150°C.

greater the elongational stress and the higher the elongation at break. This was contributed to the fact that the greater amount of stored elastic energy given to the molten polymer at the higher shear rate. The recoverable energies could then release as kinetic energies within the extrudate and thus resulting in the increment of stress at break. This result was in agreement with the work carried out by previous researchers^{7,8,10,11,24} in that the mechanical strength of molten polyolefin tended to increase with increasing extrusion rate. It was interesting to note that the changes in elongational stress appeared to be more sensitive and pronounced by altering the $\dot{\gamma}_{\text{skin}}/\dot{\gamma}_{\text{total}}$ as compared with by changing the $\dot{\gamma}_{\text{core}}/\dot{\gamma}_{\text{total}}$ as seen in Figure 14. Furthermore, Figure 16 shows a direct comparison of the elongational stress of coextruded LDPE using the $\dot{\gamma}_{\text{skin}}/\dot{\gamma}_{\text{total}}$ and $\dot{\gamma}_{\text{core}}/\dot{\gamma}_{\text{total}}$ of 0.66. It was found that the elongational stress obtained by using the $\dot{\gamma}_{\text{skin}}/\dot{\gamma}_{\text{total}}$ was also higher than that of $\dot{\gamma}_{\text{core}}/\dot{\gamma}_{\text{total}}$ for any given strain rates. The reason for this discrepancy was probably the same as that given to explain the rheological properties in Figures 6 and 7. The results in Figures 6, 7, 14, and 15 confirmed that the skin layer had greater effect on the rheological properties and mechanical strength of the LDPE coextrudates than the core layer.

As mentioned earlier that recycled LDPE was usually used as the core layer in coextruded products. The elongational stress obtained from different numbers of reprocessing LDPE at the core layer is given in Figure 17. It can be seen that the elongation stress for the reprocessing numbers 3 and 4 were higher than that of virgin LDPE, while no significant differences were found for the reprocessing numbers 1 and 2. These results can be explained by the gel content results (in Table I) which indicated the cross-linked structure of LDPE at the reprocessing numbers 3 and 4, this resulting in higher melt strength of coextruded LDPE.

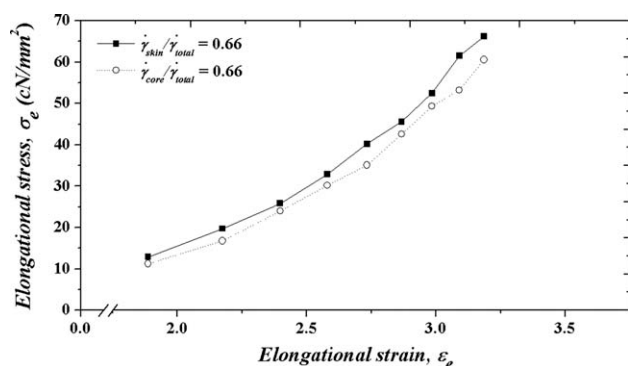


Figure 16 Comparison of elongational stress obtained using $\dot{\gamma}_{\text{skin}}/\dot{\gamma}_{\text{total}}$ total and $\dot{\gamma}_{\text{core}}/\dot{\gamma}_{\text{total}}$ of 0.66 at various elongational strain.

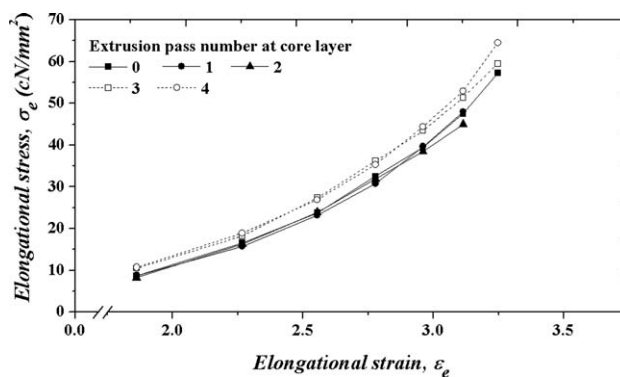


Figure 17 Effect of reprocessing number at core layer on elongational stress for coextruded LDPE at $\dot{\gamma}_{\text{core}}/\dot{\gamma}_{\text{total}}$ of 0.5.

CONCLUSION

In this study, the coextruded feed-block unit coupled with a mechanical strength measuring rig was specially constructed to determine the rheological properties and melt strength of coextruded LDPE. The effects of die temperature, shear rate, the number of extrusion passes at the core layer, and roller take-up speed were studied. As for the rheological properties of coextruded LDPE, it was found that the true viscosity and swelling ratio of the LDPE coextrudate decreased with increasing die temperature. The rheological properties and mechanical strength of LDPE coextrudates were more dependent on the shear rate of skin layer as compared to those of core layer. The reductions in viscosity and die swell ratio of coextrudate for reprocessing numbers 1 and 2 were probably caused by the disentanglement of long chain branching LDPE at the core layer. On the other hand, the increments of viscosity and die swell ratio for reprocessing numbers 3 and 4 resulted from crosslinking effect. The elongational stress of coextrudates was found to increase with increasing elongational strain, while tended to decrease with increasing die temperature. The changes in mechanical strength by the effects of shear rate and number of reprocessing corresponded well with the rheological measurements.

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