EFFECTS OF INTERFACE SLIP ON FLOW SHEAR STRESS BETWEEN DIFFERENT MATERIALS

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Abstract

Interfacial instability is an unsteady-state process in which the interface located between layers varies locally in a transient manner. This instability develops in the co-extrusion mold, and can be correlated with a critical interfacial shear stress for a particular polymer. In this work, a multilayer film co-extrusion experiment was conducted to measure the flow pressure drops on the mold wall along the flow to evaluate the flow shear stresses. Two pressure sensors were set sequentially on the mold wall along the flow direction for measuring the flow pressure drops. Pressure drops in the flow would be used to calculate flow shear stress. Variations of shear stress are then used to analyze interface slipping occur in the multilayer film. Two combinations of Polycarbonate (PC) / Polymethylmethacrylate (PMMA) and Polypropylene (PP) / Polycaprolactam (PA) were used as the experimental materials. Extrusion rate was experimentally increased to raise the pressure within the flow. According to the experiment results, when shear stress reaches a certain threshold, a large number of interfaces cause interfacial slip, which leads to a decline in shear stress. Increasing the number of interfaces increases interfacial slip. Moreover, PP/PA co-extruded film is more likely to cause slippage than PC/PMMA co-extruded film, because of its poor binding force between PP/PA interfaces.

Key words: *co-extrusion, multilayer film, interfacial slip*

1. INTRODUCTION

Consumers increasingly demand plastic products with high added value (high functional) and high quality. Compound technology is essential for making highly functional products. Compound technology is technique in which various materials are mixed and melted to obtain required properties that a single material cannot provide. Compound technology can be broadly divided to two categories: Mixing Technology and Multilayer technology. Multilayer technology is layered by each material layer to achieve high functional purpose, such as multilayer co-extrusion process. In the multilayer co-extrusion process, different molten polymer materials are extruded in a single mold by using two or more different extruders. This simple process can produce multiple layers. Depending on different function required, the number of layer can be from two to more than hundreds of layers. The advantages of multilayer co-extrusion process are the reduced number of process steps, which can be performed continuously, and its low technical complexity. However, interfacial instability may occur and affect the product quality.

W. J. Schrenk noted that when the interfacial shear stress is too high, the consequently high shear rate cause trigger interface instability. He used mathematical models to deduce the occurrence of sharkskin or interface instability from the following key parameter; skin layer to that of the center layer, total extrusion rate and die geometry [1] . O. Martin investigated interfacial instability and found that lower viscosity of the layer close to the die wall resulted in higher stability, but could result in an excessive shear stress or an excessive ratio of the thickness of the skin layer to that of the center layer difference, inducing an unstable flow. With respect to the die geometry, he also found that when the die gap is too narrow, unstable flows are always produced [2] . M. Zatloukal studied double LDPE extrusion, and found that reducing the thickness of the layer near the die wall causes the molten interface to be closer to the die wall, making the interface unstable. As the layer thickness is reduced, the molten interface becomes more wave-like, and when the layer is very thin, it eventually adopts a zip-zag surface. M. Zatloukal found that instability in the co-extrusion is wave-like [3] . M.T. Martyn used the same material (LDPE) in co-extrusion experiment. The experimental results indicated that differences in channel thickness and in mold shape can cause instability. Particularly, the mold edges and corners or the

intersection of the melt, which have large stress would produce instability first. The elasticity of melt increased would also increase instability too [4] . E. Baer's reduced the thickness of multilayer film to nanoscale and heated to above the melting point. The multilayer film surface breaks and splits into droplets because of Reyleigh instability. The droplet diameter increased with film thickness, which resulted in a discontinuous film surface [6] . K. Lamnawar et al investigated ways to reduce interfacial instability of co-extruded PE and PA6. The findings show instability of PA_6/PE interface can be improved and make the interface more smoothly by increasing PA6 residence time in the mold and reducing extrusion rate [7] [8] .

Compared to generally blended composites, multilayer composites showed better mechanical properties. When the external loading was parallel to the interface, the interfacial stress between the adjacent layers was created due to the interfacial adhesion of layers. The interfacial slip would occur when the interfacial adhesion of layers was not strong enough to restrict the external loading. The ability to against the external loading increased with number of layers [9] .

Multilayered composites also showed the good soundproofing ability. Increasing the number of layers would increase the chances of sound reflection at interfaces. More sound reflections enhance the soundproofing performance. Furthermore, the multilayer composites demonstrated larger tensile strength and elongation compared to those blended composites [10] [11] .

Recent studies have attempted to create new properties of multilayer film by increasing the number of interfaces or adding nanoscale materials to the multilayer film. However, as the number of layers in a multilayer film increases, interfacial forces begin to have a large effect on multilayers. This work tried to analyze the shear stress inside the mold flow. Variations of flow shear stress with the number of layers were investigated.

2. MATERIALS AND METHOD

This study designed and fabricated a new mold for layer-multiplying the co-extruded multi-layer polymer films. The layer-multiplying unit was set behind the exit of 2 materials co-extrusion mold, as presented in Fig. 1. The layer-multiplying unit has the following features:

1. Melt thickness is reduced by half, and melt width is doubled.
2. The melt was divided into two parts and flowed to different directions.
3. Two melts to the same plane and stacked from top to bottom. The thickness and width in this step are identical to those in the first step. However, the layer number is doubled.

Figure 2 shows that, by repeating this process, the layer-multiplying units increase the number of layers in the co-extruded polymer multilayer films. Connecting n sets of layer-multiplying units gets 2n+1 layers in one multilayer. Figure 3 shows how the pressure drop along a co-extrusion flow is measured by attaching two pressure sensors to the mold wall in the flow direction. The materials used in the experiments were polycaprolactam (PA_6) from BASF (Germany), polypropylene (PP) from SUMITOMO (Japan), polycarbonate (PC) from SABIC and polymethylmethacrylate (PMMA) from Asahi Kasei.

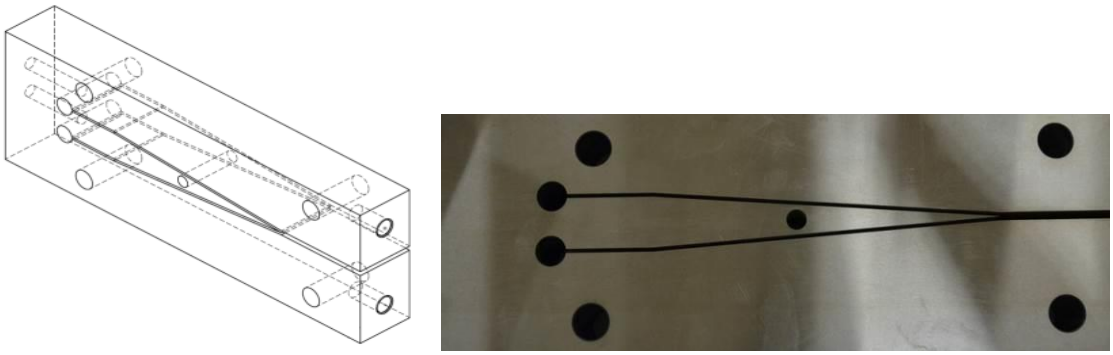


Figure 1. Two materials co-extrusion mold.

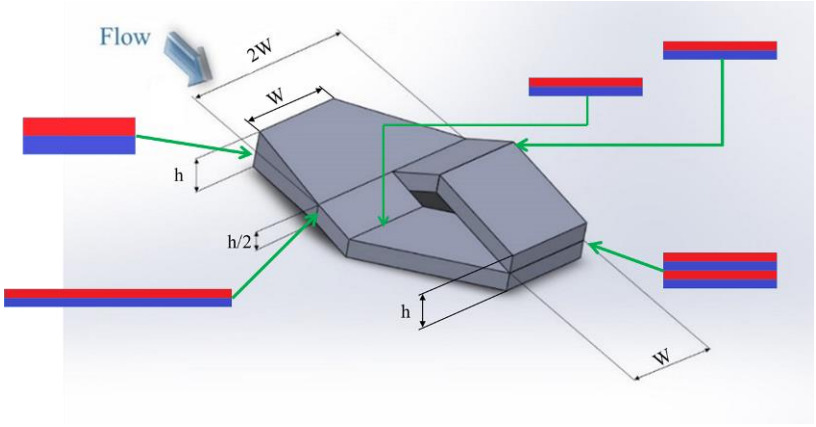


Figure 2. The schematic diagram of layer-multiplying unit.

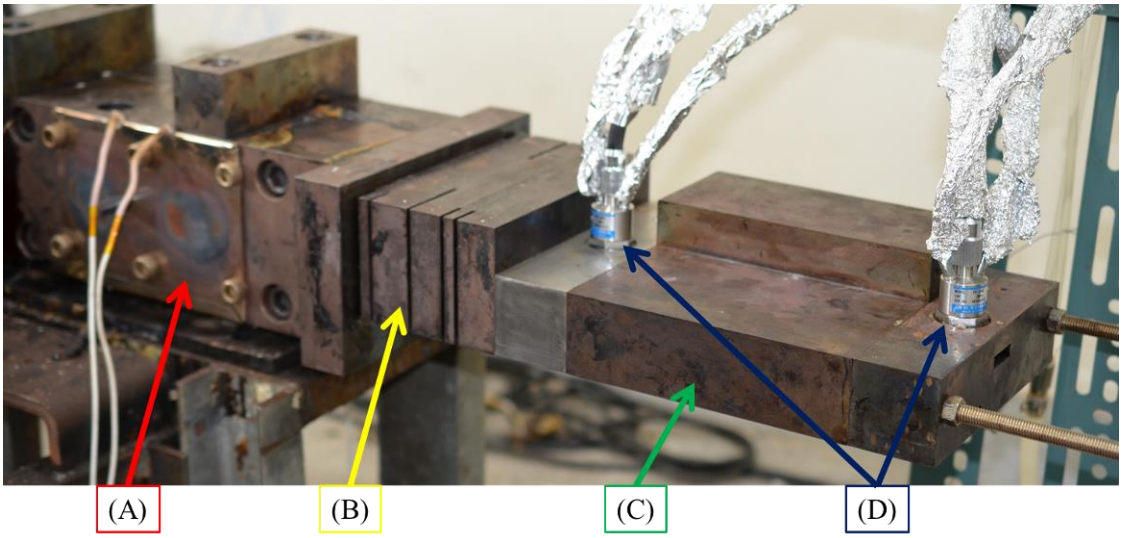


Figure 3. (A) Two materials co-extrusion mold, (B) layer-multiplying unit, (C) pressure drop measurement range, (D) pressure sensor.

3. RESULTS AND DISCUSSION

The material temperatures are set as follows: T_{PP}=190℃, T_{PA6}=240℃, $T_{PP/PA\ mold}$=220℃; T_{PC}=270℃, T_{PMMA}=260℃, $T_{PC/PMMA\ mold}$=260℃. Five groups of co-extrusion experiments were performed, and each co-extrusion experiment had a different number of layer-multiplying units (2, 4, 6, 8, and 10). The melt pressure drop on the die wall was measured along the flow to evaluate the flow shear stresses. In this process, we ignore the viscous fugitive from flow center to mold wall. Figure 4 and Eq. 1 show that, in a steady-state flow field, the pressure drop multiplied by the cross-sectional area of the flow channel equals the shear stress on the mold wall multiplied the area of flow channel wall.

$$\Delta p*a=\tau*2(A1+A2) \quad (1)$$

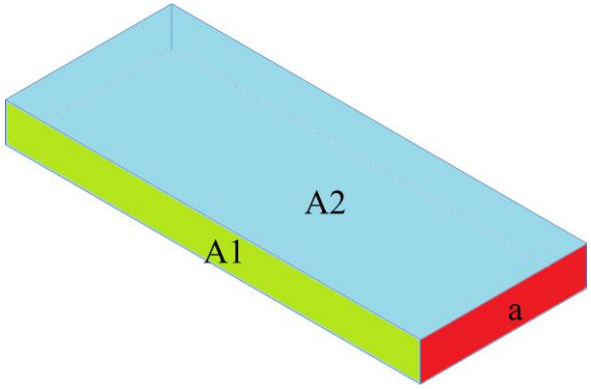


Figure 4. The Calculation of shear stress (τ) on the mold wall was defined by the following formula: $\Delta p*a=\tau*2(A1+A2)$.

In Eq. 1, "2(A1+A2)" is the area of flow channel wall, "a" is the cross-sectional area of flow channel, "Δp" is pressure drop in the flow, "τ" is the shear stress on the mold wall.

Figure 5 shows the calculated shear stress on the mold wall for different flow rates (screw rotation rate). The calculated shear stress for a mold wall with 4 and 8 layers is almost the same. When the number of layers is increased to 32, the shear stress gradient increased slowly as the flow rate increased. Additionally, when the number of layers is further increased to 512, the shear stress gradient increased very slowly as the flow rate increased. That is, with the increasing of the number of layers, the shear stress gradient increasing rate tends to slow. Therefore, for a given thickness, the potential for interfacial slip increases as the number of interfaces increases. When interfacial slip occurs, the shear stress on the mold wall is no longer increasing quickly with the flow rate, but increasing slowly even tends to a constant value.

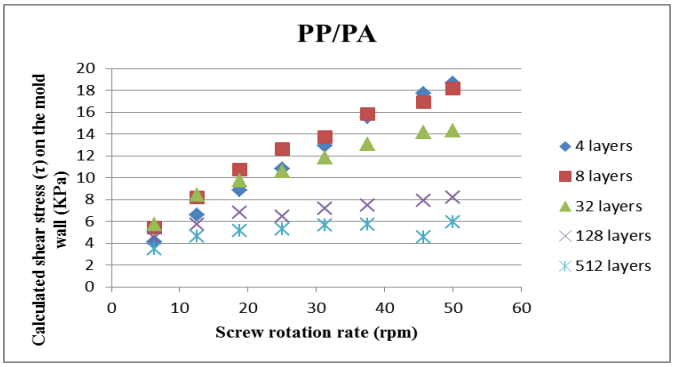


Figure 5. The calculated shear stress on the mold wall for different flow rates (PP/PA).

Figure 6 shows the calculated shear stress on the mold wall distributes in different layers. The shear stress values show that, in the low flow rate (low screw rotation rate), shear stress increases with the number of layers increases at first, but later shear stress is gradually decreased as the number of layers increases. Therefore, for a co-extruded multilayer with a low flow rate, interface of the multilayer had a tendency to increase the shearing resistance of the flow. However, when the rise in shear stress reaches a certain threshold, further increasing the number of interfaces causes interfacial slip and a decline in shear stress. When the flow rate is high (high screw rotation rate), shear stress decreases rapidly as the number of layers increases. In contrast, when the flow rate is low, shear stress increases first and then decrease later with the number of layers increases. This phenomenon occurs because a very high shear stress at the outset causes a high flow rate and makes the interface slip. Interfacial slip resulting in a decline of the shear stress. This downward trend increases as the number of layers increases. Figure 7 shows that, during the experiment, PP / PA co-extrusion products could be peeled off easily. Considering that PP is non-polarity material and that PA is strong-polarity material, interfacial bonding between these two materials was poor. This study showed that the two materials undergo interfacial slip during the co-extrusion process.

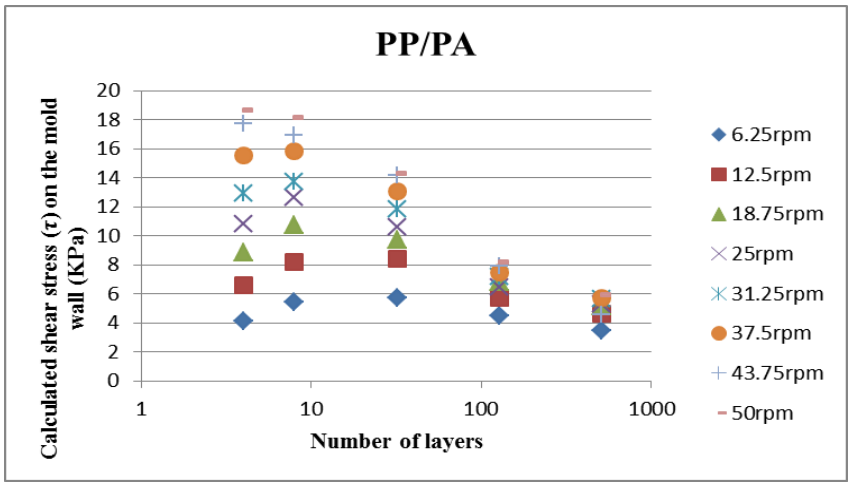


Figure 6. The calculated shear stress on the mold wall distribute in different layers (PP/PA).



Figure 7. The peeled off co-extrusion product.

Figure 8 shows the calculated shear stress on the mold wall distributes in different layers. Compared to the experiment using PP/PA, it did not have a significant decline of the shear stress in the experiment using PC/PMMA. At first, the shear stress rises with the number of layers and the extrusion rate increase. When the number of layers became more than 500, the shear stress decreased slightly. Figure 9 shows the cross section of PC/PMMA co-extruded product. Each layers overlapped smoothly and no interface slip or turbulent phenomenon been observed.

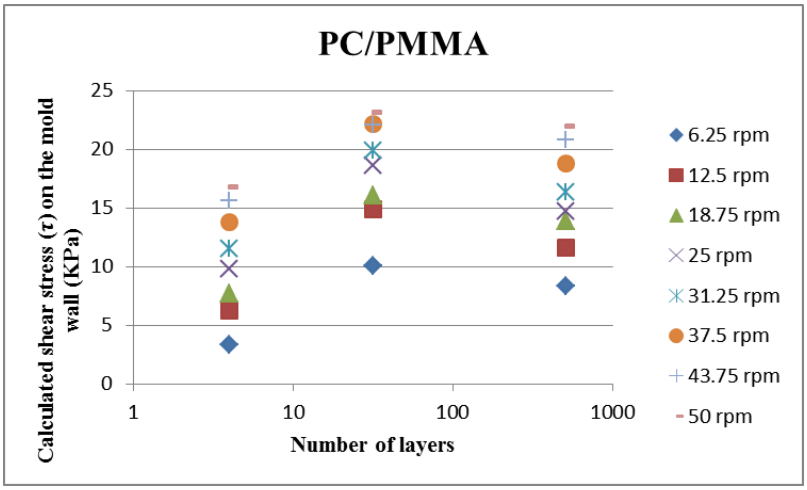


Figure 8. The calculated shear stress on the mold wall distribute in different layers (PC/PMMA).



Figure 9. The cross section of PC/PMMA co-extruded product.

4. CONCLUSION

This study used a specially designed mold in plastic co-extrusion experiments, and different numbers of layer-multiplying units were connected behind the exit of co-extrusion mold so that we can change the number of interface of multilayer. Interfacial slip was examined under varying co-extrusion rates and varying numbers of interfaces in the multilayer. The follow conclusions are drawn.

1. A multilayer interface tends to increase the shearing resistance of the flow with low co-extrusion rate. When the rise in shear stress reaches a certain threshold, further increasing the number of interfaces causes interfacial slip and a decline in shear stress.
2. When the co-extrusion rate is high, the initial pressure in the mold flow is high. The large internal pressure was more likely to result in interfacial slip.
3. The number of interfaces in the co-extrusion product can be increased by connecting more number of layer-multiplying units behind the exit of co-extrusion mold. The more number of interfaces could provide more significant interfacial slip. Severe interfacial slip results in much lower shear stress on the mold wall.
4. For the PC/PMMA layers with strong interfacial bonding, interface slip would hardly occur when the number of layers fewer than 500 and the extruded rate is low. When the number of layers over 500, the interfacial slip occurs between layers and causes shear stress slightly decreased.

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