

RELATION BETWEEN INTERFACIAL WAVE INSTABILITY AND TENSILE STRENGTH OF TWO-LAYER POLYMER STRUCTURE: AN EXPERIMENTAL STUDY

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Abstract The existence of interfacial instability at the interface of multilayer polymeric flows is well known. This article is designed mainly to provide guidelines for the development of experimental techniques for the improvement of two-layer polymer products in the polymer processing industry. This is done by performing a series of tensile tests on extrudate two-layer polymer melts in order to get insight into the relation between interfacial wave and mechanical properties of LLDPE/HDPE polymer system. Observed variations of the mechanical properties have been related to the conformation of the interfacial wave so that a relationship between interface morphology corresponding to extrusion instabilities and mechanical characteristics of the interfacial strength for polymer melts extrudate is established.

Keywords: Interfacial; Instability; Strength; Wave number; Extrusion.

INTRODUCTION

The experimental studies have shown that interfacial instability of two-layer Poiseuille flow plays an important role in polymer processing operation^[1–4]. These studies have qualitatively measured that interfacial waves in the multi-layer structure of polymer melt provided a mechanism for interlocking at the interface^[5, 6]. There have been many investigations dealing with polymer-polymer instability. They have studied interfacial instabilities parameters such as shear stress, layer thickness, viscosity, elasticity, interfacial pressure, interfacial wave, die geometry, molecular weight (MW) and molecular weight distribution (MWD). In co-extrusion of four types of LDPE, the broad-MWD materials have shown to have a great tendency to have instability due to layer depth ratio. In contrast, interfacial instabilities in narrow MWD materials are related to the stress at the interface^[7–9]. The main driving force for developing constitutive equations is the need for solving the polymer processing problems. During the past decade, the multilayer flow of viscoelastic fluids has been of interest in many polymer processing applications, such as the co extrusion of films, fibers, coating and multilayer co-extrusion technology. Since the co-extrusion process is associated with combining materials with specific properties into a single structure and due to the fact that the coextrusion process is more economical than the conventional laminating process, the interfacial stability in the multilayer flow of Newtonian polymeric fluids has been examined by a number of investigations. An unstable interface will possess interfacial waves similar in nature to surface waves in bodies of water. In general, these interfacial waves can occur at extremely low Reynolds number and are due to the combined effects of viscosity and elasticity differences in the respective layer, the relative thickness of the layers, wave number and geometry of the flow. Other factors such as density differences and interfacial tension can also play a role. In contrast to the relatively large number of theoretical

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investigations, very few experimental studies have addressed the problem of interfacial stability and interfacial strength. In the co-extrusion industry for describing the functioning of the polymer flow containing instabilities and for developing and understanding the behavior of various interrelationships between interfacial instabilities and interfacial strength of materials it has been pointed out that interfacial waves play a considerable role in determining the mechanical properties of the final product^[10–12]. There has been an aggressive demand in the polymer processing industry to use adhesive material between multilayer products to improve their mechanical interfacial strength. The aim of this work was to investigate the interfacial strength of extrudate at different layer depth ratios and wave numbers. Also an experimental set-up was designed in order to introduce a controlled amount of interfacial instability (wave amplitude) in the co-extrusion process to study variation of tensile toughness of the final extrudate products.

EXPERIMENTAL

The main part of experimental setup is shown in Fig. 1. The assembly includes three components: two laboratory extruders co-extrusion die, assorted fittings, adapters, and optical system camera and digital image processing equipment. Extruders No. 1 and No. 2 are 3/4 inch, 25:1 ratio. Both extruders are single-flight, constant-taper screw. The optical components and video camera are mounted on a standard optical bench. Two melt pressure transducers are mounted flush with the wall in both the upper and lower melt streams prior to the exit of die blade. The pressure signals from these pressure transducers are used to measure amplitude and frequency of the input pressure disturbances, and are recorded by data acquisition system. The temperature in each part of die is individually controlled by digital temperature control loops. An important part of the equipment is the experimental co-extrusion die. A schematic of the die is shown in Fig. 2. The die has three separate sections. The feed block connects both extruders and serves to secure the die blade in the main block. The flow channel is formed in the main block and has a channel depth of 2.5 millimeter and width of 25 millimeter. That results in a channel aspect ratio of 10:1 which meets those of requirements for two dimensional flows in a slit. In order to have a uniform cross section sample of extrudate polymer melts as required for any mechanical testing, an aluminum mold was designed to fabricate required samples for mechanical testing. Figure 3 shows the overall sketch of aluminum die which is attached to existing stainless steel co-extrusion die.

Procedure

Using the experimental apparatus described above, the interface can be viewed directly at different die positions (windows 1 to 4) and by using the digital image processing capabilities images can be enhanced and clarified.

By digital image processing the interfacial wave variation is detected. The image processor converts signal containing the image data in the form of digital “picture elements” or “pixels”. The digital image processing setup consists of a digital image board installed in personal computer. The most important use of the imager involves the compression of several seconds of video into a single “composite” image. Using composite image of the moving interface one is able to easily measure both the amplitude and frequency of the evolving interfacial wave. The channel pressure measurements and composite images are used in order to evaluate the frequency content of the interfacial wave under investigation. The experiments are performed either by relying on the background disturbances or by introducing a regular (periodic) disturbance with a known frequency and amplitude to study the stability of the interface. The controlled disturbances are introduced by forcing the screw to move forward by a prescribed amount once each screw revolution. This will create a regular pressure pulse of the form $P = A \sin \omega t$ where the amplitude A is determined by screw displacement and the frequency is identical to the screw rotation rate (RPM). In order to vary the disturbance frequency while maintaining a constant layer depth ratio, a bleed valve (Damper, see Fig. 1.) is installed at the path where the flow is directed from the extruder to the inlet die.

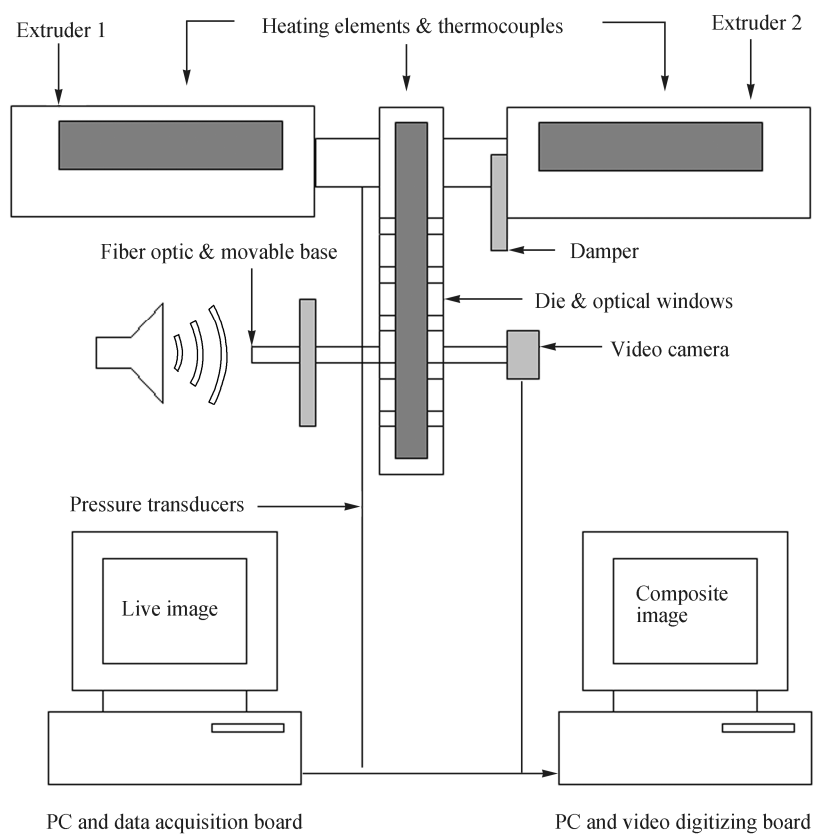


Fig. 1 Experimental set-up

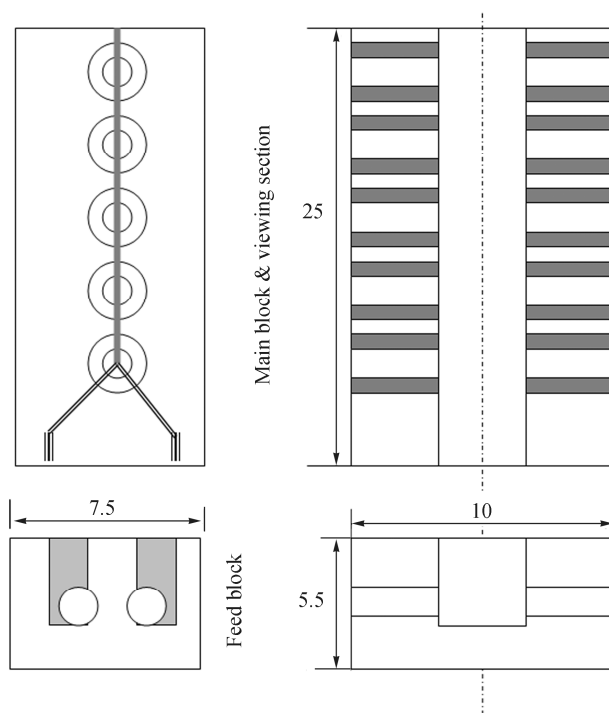


Fig. 2 Two layer parallel channel experimental co extrusions die (dimensions in centimeter)

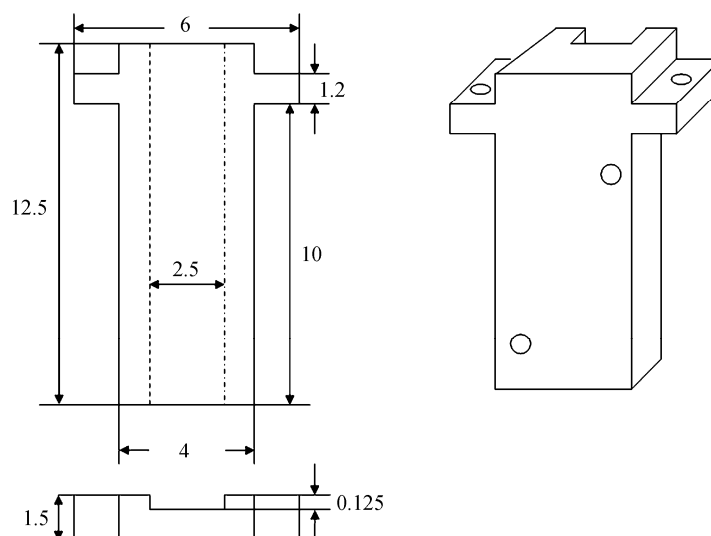


Fig. 3 Schematic of the aluminum mold for mechanical testing (dimension in centimeter)

Material Properties

The materials used in this study were consisting of high density polyethylene (HDPE) resin (Quantum LS556) from Quantum Chemical Company and linear low density polyethylene (LLDPE) resin (Dow 2045) from Mobile Chemical Company. These polymers were chosen because of their use in industrial packaging films and because of the rheological properties. Table 1 contains polymer properties at 204°C, the power-law coefficients, m , n and density.

Table 1. Polymer properties at 204°C

Polymer type	$\dot{\gamma}$ range (s^{-1})	m (poise)	n	ρ melt (g/cm^3)
HDPE	$0.3 \leq \dot{\gamma} \leq 9$	1.15×10^4	0.950	0.755
LLDPE	$0.1 \leq \dot{\gamma} \leq 8$	7.22×10^4	0.775	0.760

RESULTS AND DISCUSSION

In order to get an idea of mechanical properties of base material, in the first, flow direction tensile tests have been performed using single layer extrudate samples. The samples were fabricated using die geometry shown in Fig. 3. All samples for this experiment have been collected at the same temperature and are cooled from the die temperature (204°C) to room temperature. The Instron screw driver type testing machine was used at load cell with maximum range of 2500 N. The strain rate (cross head velocity) of 1.25 cm/min was chosen for all tests. This gives the requirement of having all test conditions constant while varying layer depth ratio, wave number and wave amplitude. The stress/elongation curves for these base materials are shown in Fig. 4. As figure indicates, HDPE has maximum tensile stress value of 19000 kPa and elongation of 0.7 cm. One can see from the figure that LLDPE has less tensile stress and much more elongation than HDPE. In the initial stage of this investigation the spectrum of disturbances which are in the coextrusion system was considered. The layer depth ratio is defined as d_2/d_1 (LLDPE/HDPE). In order to examine relation between polymer thickness and mechanical strength of the interface for each set of experiment, ratio of d_2/d_1 has been changed and ten samples are collected. Samples were cooled and best of them in terms of uniform cross section has been chosen for mechanical testing. The tensile test experiments have been performed as mentioned before for seven different layer depth ratios. The results are shown in Fig. 5. This figure shows effects of layer depth ratio on the mechanical strength of the interface on extrudate products in absence of a prescribed disturbance. As one can

see from the figure, layer depth ratio is a determinate factor for maximum tensile stress versus elongation in this polymer system. As thickness of LLDPE increases tensile stress of two layer structure decreases. This result is in agreement with tensile stress of single layer structure LLDPE and HDPE (see Fig. 4). The interfacial wave velocity of 2 layer polymer system is measured by observing and recording movement of a particle in the flow with respect to time (particle tracking). The interfacial wave velocity is measured in a region where instability has affected the flow substantially as well as near to the blade. Using the measured interfacial wave velocity at position nearest to the blade and the disturbance frequencies, the wavelength is calculated ($v = \lambda \times f$), where v is velocity, λ is wavelength and f is frequency of the interfacial wave.

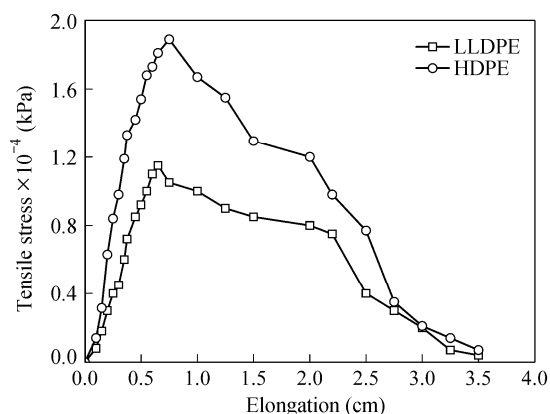


Fig. 4 Stress versus elongation for LLDPE, HDPE

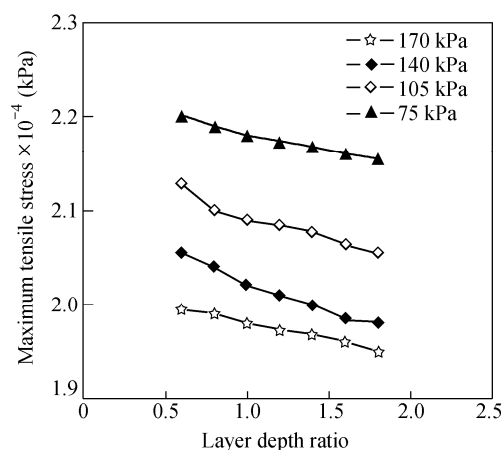


Fig. 5 Layer depth ratio versus tensile stress for LLDPE/HDPE at four different die pressure

If a disturbance is introduced into an undisturbed basic flow, the disturbance could either die out; persist at similar magnitude or grow until the basic flow has changed. In order to see how a known wave (wavelength and amplitude) affects interface and instability of the flow a known amount of disturbances were prescribed then flow instabilities were investigated. In these experiments ratio of LLDPE/HDPE has been changed. Following the procedure described before, stability contour was established. The growth or decay of the wave is calculated using the equation: $\frac{dA}{dx} = \frac{A_2 - A_1}{x_2 - x_1}$ where A_i is the amplitude at position i and x_i is downstream distance at position

i . A point was considered stable if a negative growth rate was observed, and unstable if a positive growth rate was observed or indeterminate if the growth rate was too small to make a precise determination (as low as 0.00015 cm/cm). For each layer depth ratio, frequency of the interface from 0.20 Hz to 1.20 Hz has been varied by extruder No. 2 and bleed valve or damper (see Fig. 1). Bleed valve is used imposing different frequency while maintaining a constant layer depth ratio. Typical composite images of interfacial wave at different die positions are shown in Fig. 6. In this figure a stable interface (top) and an unstable interface (bottom) with growing interfacial wave are shown. The images are generated using 30 seconds of flow video recording data. From a known frequency and interfacial wave velocity, the dimensionless wave number (α) was calculated with knowledge of the interfacial wavelength using the following equation:

$$\alpha = \frac{2 \times \pi \times d_2}{\lambda}$$

where d_2 is the thickness of the less viscous or elastic fluid (LLDPE) and λ was calculated based on the measured wave velocity at the interface. The results of these investigations have been used to establish stability/instability contour based on the layer depth ratio and dimensionless wave number. The Fig. 7 clearly shows the region in which flow is stable ($d_2/d_1 < 0.45$) and unstable region of the interface ($d_2/d_1 > 0.45$). For a given pressure amplitude (35 kPa), at five layer depth ratio which covers both the stable and unstable region of stability contour, the samples have been collected. The samples were cut for interfacial wave observation. When

flow is stable, as Fig. 8 shows there is no growing wave at the interface. As it is shown in Fig. 9, when flow is unstable interfacial wave is observed. Since, the growing interfacial wave obtains energy from the materials imbalances at the interface, when mixing occurs at the interface, some energy will be lost to the mixing process which will results in less energy being available for growing wave. The material at lower layer (HDPE) moves to the upper layer and clearly signifies an oscillatory current at the interface (Fig. 8). This current acts to transport material to and from the interface thereby enhancing the diffusion process. This process is responsible for small scale mixing which results low growth rate as well as low interfacial strength.

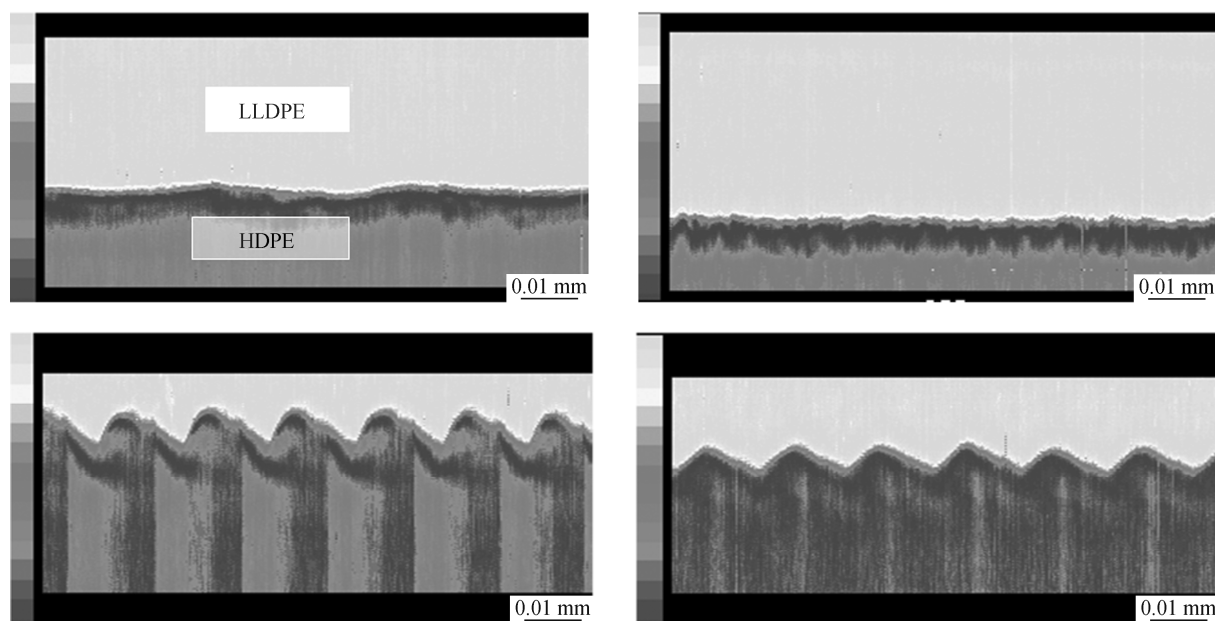


Fig. 6 Typical composite images of stable (top) and unstable (bottom) interfaces

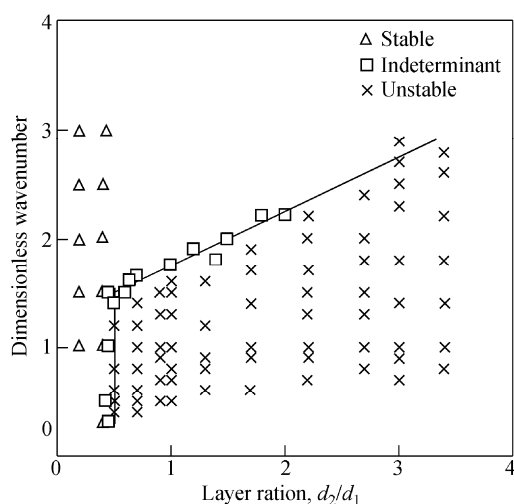


Fig. 7 Experimentally determined stability diagram for LLDPE/HDPE

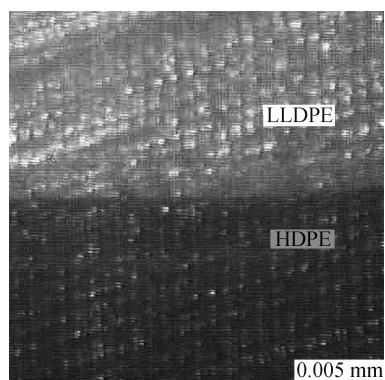


Fig. 8 Interfacial mixing (stable flow)

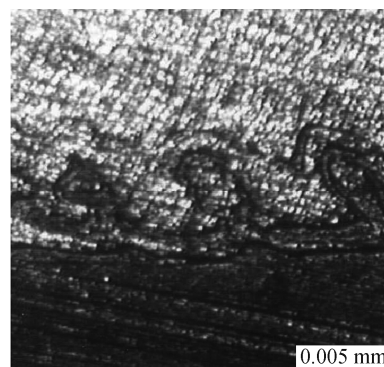


Fig. 9 Interfacial interlocking (unstable flow)

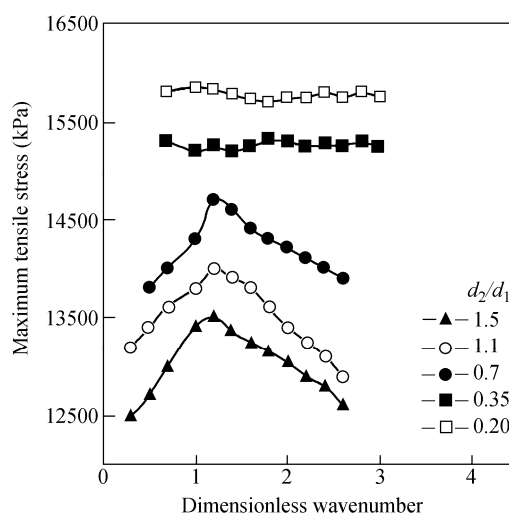


Fig. 10 Tensile stress versus dimensionless wave number at different layer depth ratio



Fig. 11 Photograph of extrudate cross-section wave interlocking

Among the all collected samples those selected for testing possessed uniform cross sections. The tensile tests have been performed. Based on the results, a relationship between interfacial wave and interfacial strength has been found. The results are shown in Fig. 10. The figure clearly shows effects of the stable/unstable interface on the mechanical properties of extrudate products. As this figure shows when the flow is stable that is $d_2/d_1 < 0.45$ mechanical strength of the interface does not change for all attainable frequency or wave numbers. In contrast, when the flow is unstable that is $d_2/d_1 > 0.45$ tensile stress values and in turn interfacial strength

changes due to different wave numbers. As figure indicates when the flow is unstable for all layer depth ratios there is a maximum interfacial strength exists at a frequency of close to unity. These results show that maximum rate of instabilities are corresponded to maximum interfacial strength. As it is shown, mechanical strength of interface increases when flow is unstable. Typical sample with growing interfacial wave is shown in Fig. 11. As this figure indicates wave bending and interlocking was observed at the range of dimensionless wave number from 0.8 to 1.1. This range of dimensionless number is happened to be related to maximum tensile stress. So, it is believed that this phenomenon is responsible for maximum tensile stress.

CONCLUSIONS

Interfacial strength of the LLDPE/HDPE has been investigated experimentally in two layer slit die geometry. It has been shown that instabilities are associated with interfacial waves, and it turns out that interfacial wave amplitude is known as a mechanism for controlling the strength of the interface of multilayer polymer products. An experimental apparatus device as a mold has been design and fabricated in order to collect proper extrudate sample for mechanical testing. It has been shown that for random disturbance experiments stress versus elongation relation and mechanical strength of the interface are controlled by layer depth ratio which is due to the mechanical properties of each individual polymer materials. It was not possible to find a relation between interfacial random frequency content and interfacial tensile stress. Based on these results, prescribed disturbance was introduced, and then regions of stable and unstable flows were determined. The mechanical testing results have been correlated to stability/instability of this polymer system. This experimental results show that, when the flow is stable, due to the interfacial mixing and diffusion there are no changes in interfacial strength, that is the tensile stress remains almost constant for all wave numbers. It is shown that maximum interfacial strength of the extrudate polymers occurs in the unstable region when the wave number approaches unity. It is shown experimentally that the mechanism of interfacial strength is related to interfacial instabilities as well as interfacial wave number. The results show that the interlocking phenomenon is controlling the over-all maximum strength of the two layer extrudate products in these polymer systems.

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