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Experimental Study of Relationship between Interfacial Instabilities and Mechanical Strength of Two-layer PP/HDPE Polymer Melts

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ABSTRACT

The existence of interfacial instability at the interface of multilayer polymeric fluid is well known. The experimental studies have shown that interfacial instability of two-layer polymer melts plays an important role in polymer processing operation. This work is designed mainly to provide guidelines for the development of experimental techniques for the improvement of the two-layer polymer products in the polymer processing industry. This is done by performing a series of tensile tests on extruded two-layer polymer melts in order to get insight into the relation between interfacial instabilities and mechanical properties of polypropylene (PP) over high density polyethylene (HDPE) systems. Observed variations of the mechanical properties have been related to the conformation of the interfacial wave. Thus, a relationship is established between the interface morphology corresponding to extrusion instabilities and the mechanical characteristics of the interfacial strength of the extruded PP/HDPE.

Key Words:

interfacial;
instability;
wave number;
layer thickness ratio;
interfacial strength.

INTRODUCTION

The main driving force for developing constitutive equations is the need for solving the polymer processing problems [1-3]. 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, coatings and multilayer co extrusion technology [4,5]. Since the co-extrusion process is associated with combining materials with specific properties

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into a single structure and due to the fact that the co-extrusion process is more economical than the conventional laminating process, the interfacial stability in the multilayer flow of 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 [6]. There are so many investigations dealing with polymer-polymer instability in which 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) have been investigated.

In co-extrusion of four types of HDPE, the broad-MWD materials have shown a great tendency to ward instability due to layer depth ratio. In contrast, interfacial instabilities in narrow-MWD materials are related to the stress at the interface [7].

In general, these interfacial waves can occur at an extremely low Reynolds number and are due to the combined affects of viscosity and elasticity differences in the respective layer, the relative thickness of the layers, wave number, and geometry of the flow [1,8,9]. Other factors such as density differences and interfacial tension can also play role [1,3,10]. It is said that the large deviations between theoretical and experimental interfacial growth rates which cause instability for PP/HDPE are due to the interfacial mixing [11]. It has been found that orientation of the polymer chain near the interface will influence the rate of diffusion and consequently the thickness of interfacial diffusion layer [12].

Since the orientation angle of polymer chain is related to local state of stress, to a certain extend the interfacial diffusion is related to the position of the interface (layer thickness). In contrast to the relatively large number of theoretical investigations, very few experimental studies have addressed the problem of interfacial stability and interfacial strength of two layer PP/HDPE polymer products. The experimental investigations [5] of PP/HDPE have proven that interfacial instabilities exist. It has been pointed out that interfacial waves play a considerable role in determining the mechanical properties of the final product [3,9]. There has been an aggressive demand in the polymer processing industry to use adhesive material between multilayer products to improve their mechanical interfacial strength.

Since the large interfacial waves provide a mechanism for wave interlocking, if a controlled amount of interfacial instability is presented in the co-extrusion process, it would improve the tensile toughness of the final extrudate products. 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.

The aim of this work is to investigate the interfacial strength of two layer PP/HDPE polymer extruded. In this investigation, polymer parameters such as wave number (wave length) and layer thickness ratio (LTR) were considered.

Mechanism of Interfacial Instability

If one introduces a disturbance into an otherwise undisturbed flow (basic flow), the disturbance could die out, persist at similar magnitude or grow until the basic flow had changed all together. When the disturbance dies out the flow is termed stable, when it persists it is neutrally stable and when it grows it is referred to as unstable. From a theoretical view point, the introduced disturbance can be either related to linear instability theory or nonlinear instability theory.

Experimental Apparatus

The main part of experimental setup is shown in Figure 1. The assembly includes three components:

- Two laboratory extruders
- Co-extrusion die, assorted fittings, adapters, and optical system
- Optical windows, camera and digital image processing equipment

Extruder No. 1 is a 3/4 inch, 25:1 extruder controlled and powered by HBI system 90. Extruder No. 2 is 3/4 inch 25:1 extruder (Brabender Inc.). 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 end of die blade. The pressure signals from these pressure transducers are used to measure amplitude and frequency of the

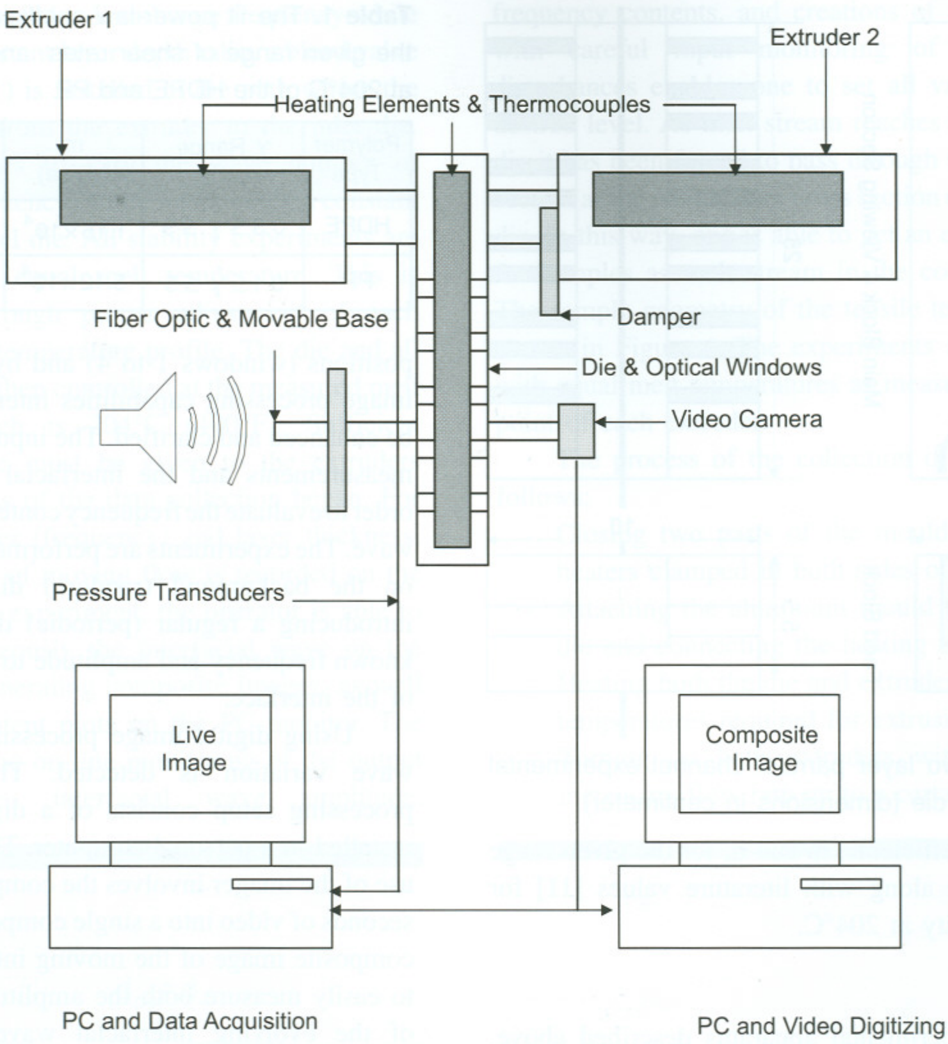


Figure 1. Experimental set-up.

input pressure disturbances, and are recorded by data acquisition system. The temperature in each part of the die is individually controlled by digital temperature control loops within the PC system. An important part of the equipment is the experimental co-extrusion die.

A schematic of the die is shown in Figure 2. The die has three separate sections. The feed block connects both extruders and servers 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 mm and width of 25 mm that results in a channel aspect ratio of 10:1 which meets those of requirements for two dimensional flows in a slit. An aluminum mould was designed to fabricate required samples for mechanical testing. Figure 3 shows the over-all sketch of aluminum die which is attached to stainless steal co-extrusion die.

Material Properties

Two polymer systems utilized in these experiments are HDPE and PP. Both resins (HDPE (LS556) and PP (PD 4252)) are from Quantum Chemical Company (U.S.A.). These polymers were chosen because of their use in industrial packaging, films, and multistructure products. Table (1) contains the fit

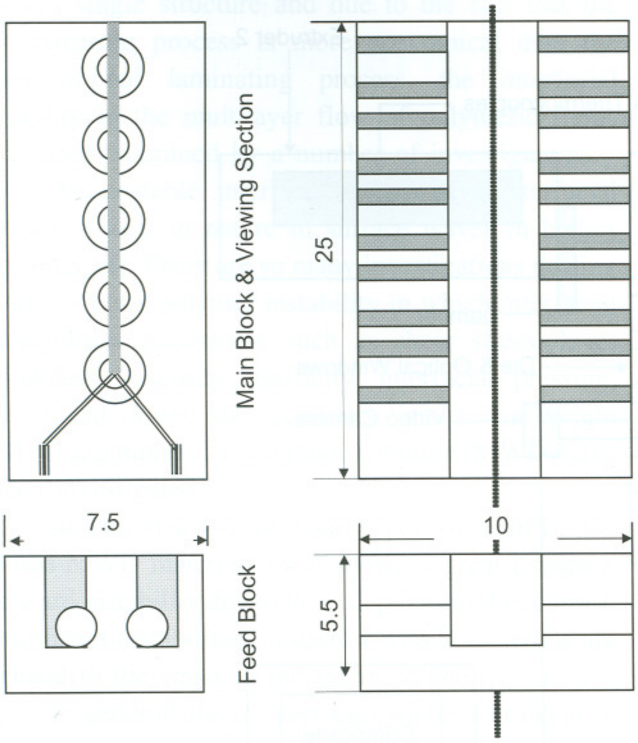


Figure 2. Two layer parralel channel experimental co-extrusions die (dimensions in centimeter).

power-law coefficients, m and n, for the given range of shear rates along with literature values [11] for the melt density at 204°C.

Table 1. The fit power-law coefficients, m and n, for the given range of shear rates and the melt density at 204°C of the HDPE and PP.

Polymer Type	$\dot{\gamma}$ Range (s^{-1})	m (poise)	n	ρ melt (g/cm^3)
HDPE	$0.3 \le \dot{\gamma} \le 9$	1.15×10^{4}	0.950	0.755
PP	$0.1 \le \dot{\gamma} \le 5$	5.12×10^{4}	0.715	0.760

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 interfacial images can be enhanced and clarified. The input channel pressure measurements and the interfacial data are used in order to evaluate the frequency content of the interfacial wave. The experiments are performed either by relying on the background (random) disturbances or by introducing a regular (periodic) disturbance with a known frequency and amplitude to study the stability of the interface.

Using digital image processing, the interfacial wave variation is detected. The digital image processing setup consists of a digital image board installed in a 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 controlled disturbances are introduced by forcing the screw to move forward by a prescribed amount once every screw revolution. This will create a regular pressure pulse of the form $P = A\,\mathrm{Sin}\,\omega t$ where the amplitude A is determined by screw displacement and the frequency is identical to the screw rotation rate (RPM).

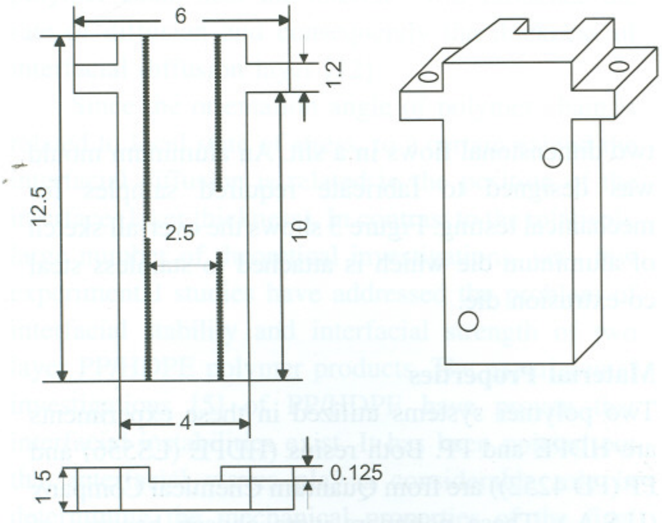


Figure 3. Schematic of the aluminum mould for mechanical testing (dimensions in centimeter).

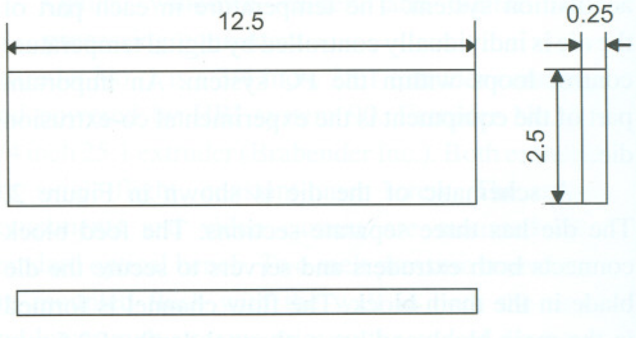
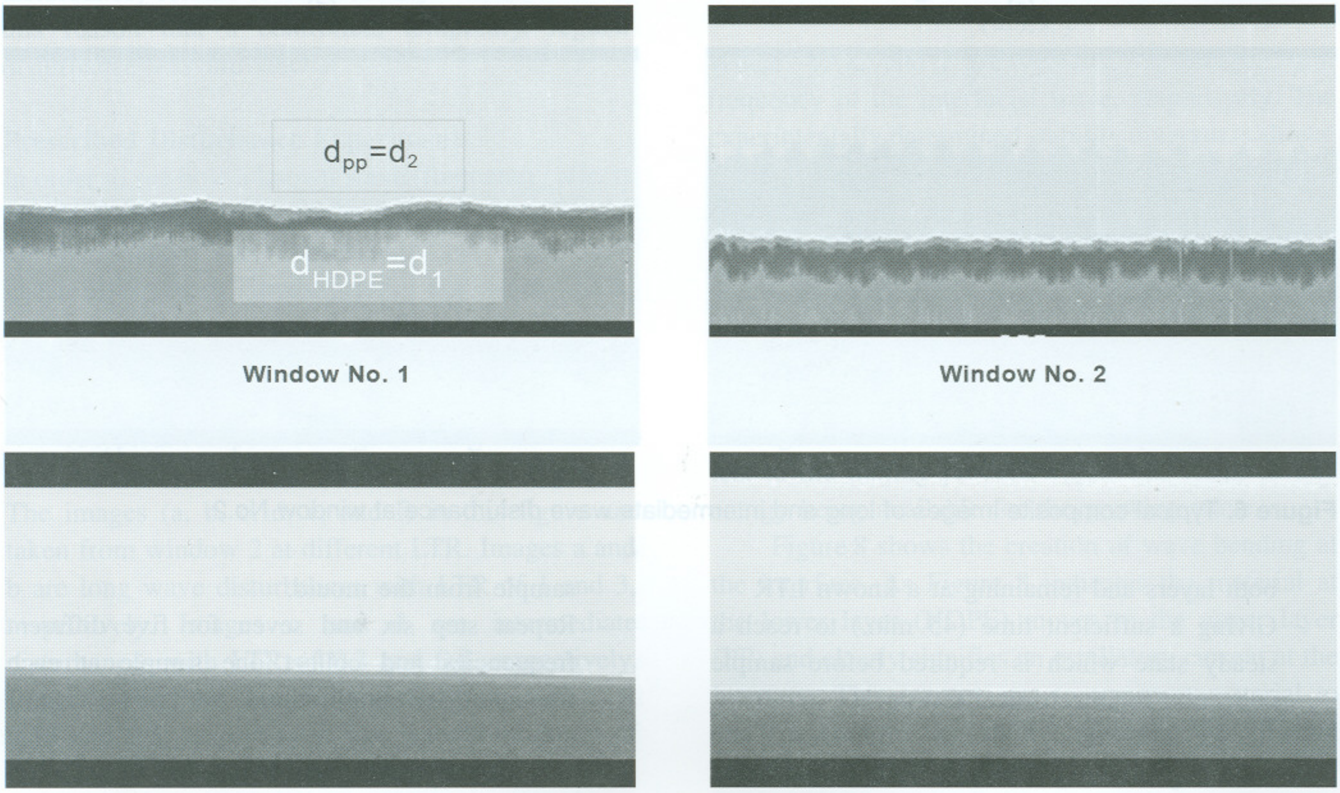


Figure 4. Geometry of samples for mechanical testing (dimensions in centimeter).

In order to vary the disturbance frequency while maintaining a constant layer depth ratio, a bleed valve (damper, Figure 1) is installed at the path where the flow is directed from the extruder to the inlet die. This set-up allows to vary the wave number of disturbances (frequency) while maintaining a constant LTR at the channel die. All stability experiments are carried out with equal melt temperature. This is accomplished through proper adjustment of each extruder's barrel temperature profile. The die and all other devices are then controlled at the measured melt temperature which is 204°C (400°F). Sufficient equilibration time must be given to the extruders before the process of the data collection began. For each wave number (frequency) and layer thickness, 30 to 60 seconds of moving flow is recorded on the VCR. During the experiment, the operator is able to observe simultaneously the interfacial wave on the monitor while generating composite images, as well as frequency content plots on the PC monitor. The combination of the on-line monitoring of the output information (i.e., interfacial wave amplitude, frequency contents, and creations of images) along with careful input monitoring of the pressure disturbances enables one to set all variables to the desired level. As melt stream reaches the end of the die it has been forced to pass through the same cross section as the melt stream cross section of co-extrusion die. In this way, one is able to get an exact geometry for samples as melt stream in the co-extrusion die. The sample geometry of the tensile test specimen is shown in Figure 4. The experiments are carried out with equal melt temperatures as measured at the end point of each extruder.

The process of the collection of samples is as follows:

- Closing two parts of the mould together with heaters clamped in both sides of the mould.
- Attaching the aluminum mould to co-extrusion die and connecting the heating elements.
- Heating both the die and extruders up to certain temperatures required for extrusion.
- Turning on both extruders with a gradually increasing flow rate up to a certain pressure for



Window No. 1

Window No. 2

Window No. 3

Window No. 4

Figure 5. Typical composite images of windows 1-4 (random disturbance).

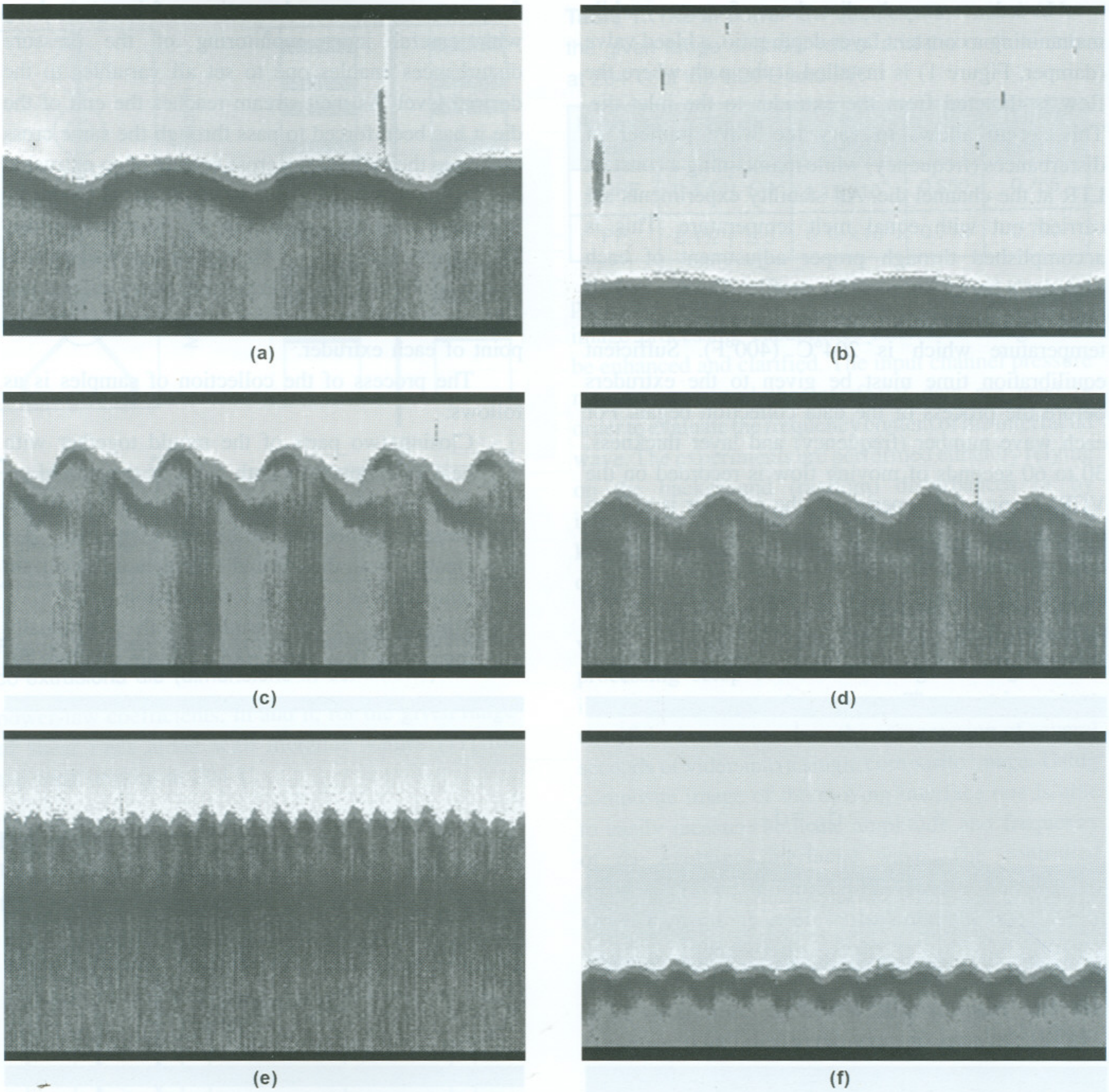


Figure 6. Typical composite images of long and intermediate wave disturbance at window No.2.

both layers and remaining at a known LTR

- Giving a sufficient time (45 min.) to reach a steady state which is required before sample collection for mechanical testing.
- At set layer ratio, collecting the sample by removing aluminum mould from co-extrusion die when two extruders are operating.
- Cooling the aluminum mould with extrudate sample to room temperature and removing the sample from the mould.
- Repeat step six and seven for five different frequencies and collect one sample at each frequency and amplitude.

RESULT AND DISCUSSION

Random Disturbance Experiment

An incompatible polymer system consisting of HDPE and PP was utilized in this experiment. Using experimental set-up and procedure, typical composite images of random disturbance experiments are taken and shown in Figure 5. LTR is defined as d_2/d_1 (d_{PP}/d_{HDPE}). In this figure, (windows 1 and 2) due to the background disturbances there are waves at the interface.

At the end of the die one can see (windows 3 and 4) that there is no wave at the interface which is indicating stable flow. This pattern was observed for all layer depth ratio and input pressure disturbances. As the image clearly shows, the interfacial wave arising from the spectrum of disturbances (pressure perturbations with a wide range of frequencies) within the apparatus gives rise to a similar spectrum of interfacial disturbances with a variety of wavelengths and amplitudes. It will provide relatively little insight into the behaviour of interfacial wave above and below the dominant value.

Thus, it is not possible to examine mechanical strength of the interface for a known wave (prescribed disturbance) related to different layer ratio. It is for this reason that a controlled, temporary, regular disturbance was introduced.

Prescribed Disturbance Experiments

In order to see how a known wave (frequency) affects interface and instability of the flow a known amount of disturbance was prescribed, and then flow instabilities were investigated. For each LTR, frequency of the interface from 0.20 to 1.20 Hz has been varied by extruder No. 2 and bleed valve (Figure 1).

A typical composite image for long intermediate and short wave disturbances are shown in Figure 6. The images (a, b, c, d, e, and f) in this figure are taken from window 2 at different LTR. Images a and b are long wave disturbances with LTR of 1 and 3, respectively. Images c and d are intermediate disturbances with LTR of 0.3 and 0.8, respectively. Images e and f are short wave disturbances with LTR 0.35 and 2.5, respectively. 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 and X_i are the amplitude and the downstream distance at the position i, respectively. A point was considered stable if a negative growth rate was observed, unstable if a positive growth rate was observed and indeterminate if the growth rate was too small to make a precise determination (as low as 0.00015 cm/cm).

Based on these investigations and dimensionless wave number (α) as well as LTR stability/instability diagram has been established. The dimensionless wave number was calculated with knowledge of the interfacial wavelength using the following equation:

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

Where, α and d_2 are the dimensionless wave number and the thickness of the more viscous or elastic fluid (PP), respectively. The interfacial wave velocity is measured in a region where instability has affected the flow substantially. Using the measured interfacial wave velocity at position nearest to the blade and the disturbance frequencies, the wavelength is calculated using

$$v=\lambda\times f$$

Where, v, λ, and f are velocity, wavelength, and frequency of the interfacial wave, respectively. The experimentally determined stability diagram is shown in Figure 7.

Interfacial Diffusion and Interlocking

The interfacial diffusion and mixing are observed with small scale due to the wave bending phenomenon. Since, the growing interfacial wave obtains energy from the material imbalances at the interface, when mixing occurs at the interface, some energy will be lost to the mixing process which will result in less energy being available for the growing wave.

Figure 8 shows the creation of wave bending at the interface. As Figure 8 indicates the material at the lower layer (HDPE) moves to the upper layer (PP) and clearly signifies an oscillatory current at the interface. This current acts to transport material to and from the interface thereby, enhancing the diffusion process.

Normally, co-extruded samples are easily delaminated since the incompatibility of polymers (PP/HDPE) results in poor interfacial adhesion.

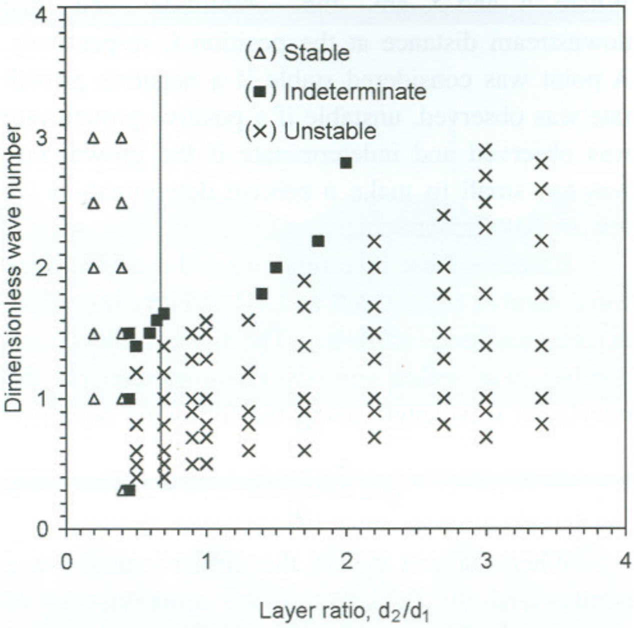


Figure 7. Experimentaly determined stability diagram for PP/HDPE.

However, large amplitude and highly bent waves are completely interlocked in the solid state which dramatically improves adhesion at the interface. It was observed that small interfacial waves had not undergone interfacial bending and there was no mechanical coupling at the interface.

On the other hand, the samples with large interfacial bending were given high degree of interlocking at the interface. The overall mechanical property of these samples improves since the stress in each layer will be coupled at the interface.

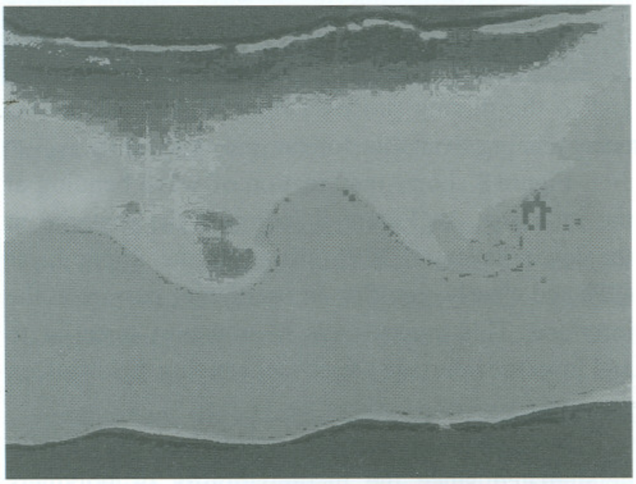


Figure 8. Wave interlocking.

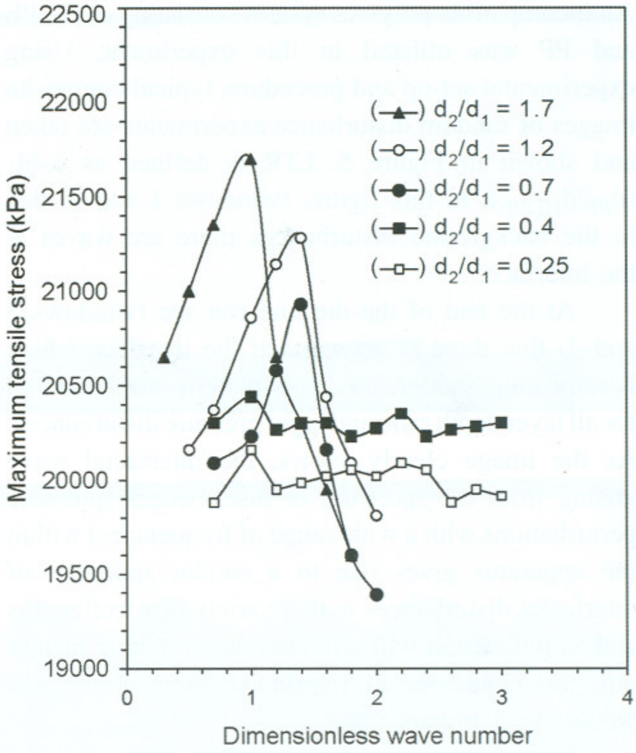


Figure 9. Tensile stress vs. dimentions wave number at diffrent layer depht ratios.

Mechanical Strength

Among all the collected samples those selected for testing possessed uniform cross sections. The tensile tests have been performed.

Based on the interfacial interlocking as described above and tensile test results, a relationship between interfacial wave and interfacial strength has been found. The results are shown in Figure 9. This 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 ($d_2/d_1 < 0.5$) due to the small interfacial waves and absence of interfacial wave bending there was no mechanical coupling at the interface. For this range of LTR mechanical strength, the interface does not change for all attainable wave numbers. In contrast, when the flow is unstable ($d_2/d_1 > 0.5$) due to the presence of interfacial wave interlocking, tensile stress values and in turn interfacial strength variations at different wave numbers occur.

A typical sample with growing interfacial wave is shown in Figure 8. As this figure indicates wave bending and interlocking was observed. Same pattern was_observed at the range of dimensionless wave numbers from 0.8 to 1.1. It was determined experimentally that variation of the wave number is related to variation of the interfacial strength.

As Figure 9 indicates, when the flow is unstable for all LTR a maximum interfacial strength exists at wave numbers of close to unity. These results show that the maximum rate of instabilities is corresponded to maximum interfacial strength. As it is shown, mechanical strength of interface increases when flow is unstable. The unstable range of dimensionless wave numbers happens to be related to maximum tensile stress. Thus, it is believed that unstable interface as well as presence of the interfacial wave interlocking phenomenon is responsible for maximum tensile stress.

CONCLUSION

Interfacial strength of the PP/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 mould has been designed and fabricated in order to collect proper extruded samples for mechanical testing.

It has been shown that for random disturbance experiments there is no relation between interfacial random frequency content and interfacial tensile stress. For a known amount of disturbances, the region of stable and unstable flow was determined. The mechanical testing results have been correlated to stability/instability of this polymer system.

These experimental results show that, when the flow is stable, 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 extruded 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 numbers. The results show that the maximum interlocking phenomenon is controlling the over-all maximum strength of the two layer extruded products in these polymeric systems.

REFERENCES

1. Rasmussen H. K., viscoelastic flow computations using a generalized molecular stress function model, *J. Non-Newtonian Fluid Mech.*, **106**, 107-120, 2002.
2. Sureshkumar R., Local linear stability characteristics of viscoelastic periodic channel flow, *J. Non-Newtonian Fluid Mech.*, **97**, 125-148, 2001.
3. Su Y.Y., Khomami B., Purely Elastic Interfacial Instabilities in Superposed Flow of Polymeric Fluids, *Rheal. Act.*, **31**, 413-420, 1992.
4. Poole R J,. Escudier M.P., Viscoelastic and shear-thinning liquids, *J. Non-Newtonian Fluid Mech.*, **109,** 193-230, 2003.
5. Khomami B., Hydrodynamic Stability of Microstructure Fluids, *Annual Conference, Progress, and Challenges*, Dept. of Fisica Fundamental, Madrid, Spain, 2003.
6. Zhang Y.L., Matar R.V., Thin weakly viscoelastic film, *J. Non-Newtonian Fluid Mech.*, **105,** 53-78, 2002
7. Perdikoulias J., Tzoganakis C., Interfacial instability in blown film co-extrusion of polyethylene, *J. plastic Eng.*, 41-43, 1996.
8. Wong A.C., Jiang L., Study on the melt entry flow characteristics in ax symmetric dies with abrupt contraction by a visualization technique, *J. Polym. Testing*, **8,** 861-867, 2001.
9. Han C.D., Shetty R., Studies on Multilayer Film Co extrusion. Part 2: Interfacial Instability in Flat Film Co- extrusion, *J. Polym. Eng. Sci.*, **18**, 180-186, 1978.
10. Olagunju L.P., Cook G.H., Effect of viscous heating on linear stability of viscoelastic cone-and-plate flow: ax symmetric case, *J. Non-Newtonian Fluid Mech.*, **102**, 321-342, 2002.
11. Gregory M.W., An Experimental Investigation of Interfacial in Superposed Flow of Polymer Melts, D.S dissertation, Washington University, St.Louis. 1993.
12. Zhao Liang J., Characteristics of melt shear viscosity during extrusion of polymers, *J. Polym. Testing*, **3**, 307-311, 2002.