OBSERVATION OF THE CAVITATION PHENOMENA UPON THE GROSS MELT FRACTURE REGIME IN LLDPE EXTRUSION

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Abstract

We present observation of a cavitation phenomenon in LLDPE extrusion. In the gross melt fracture regime, cavitation was always observed in the first 0.5 ~ 1.5 mm upstream of the exit. We saw cavities at the wall form (seemingly out of nothing) grow to a length and width of about 150 μm, then shrink down and disappear. From velocity measurements of these structures, we conclude that their width in the radial direction is much smaller than those in the axial and lateral direction and that they are in contact with the wall. The process for the growth and disappearance is approximately 20 ~ 25 ms. The shape of the cavities is highly irregular. From several precise investigations, we concluded that the unstable flow and melt fracture in the entrance region is main source for the cavitation phenomenon, and the cavitation was initiated from the unstable flow by the extensional flow at the die exit.

Introduction

The rate of production of many polymer extrusion process such as fiber spinning, film blowing and coating process is limited by surface irregularity or melt fracture. In an extrusion process, when the throughput exceeds a critical value, small amplitude periodic distortions appear on the surface of extrudate (surface melt fracture or sharkskin melt fracture). These distortions have quite regular frequency and amplitude. As the throughput increases further, these take a more severe form of larger irregular distortions (gross melt fracture or wavy fracture). Gross melt fracture (GMF) typically involves diameter variations of 10% or more, which at high flow rates are extremely irregular, even chaotic.

So far, a great number of studies on the sharkskin melt fracture (SMF) has been carried out because the sharkskin begins to occur on relatively low throughput. It is now generally accepted that SMF originate in the die exit region. Contrary to SMF, GMF has been investigated much less. Major contributions about the features and the causes for GMF phenomena were already done in late 1950's and 1960's (1). GMF is known to be initiated at the entrance to die where the melt undergoes uniaxial extension due to the contraction. Important visualization studies (tracer technique and flow birefringence) in the entrance region were shown in earlier literatures (2-5). According to these results, at the onset of GMF the converging lamella flow pattern at the entrance becomes disturbed, the flow profile fluctuates and the axial symmetry of the streamline vanishes. As the flow rate increases further, the melt at the centerline of the entrance region fractures; the asymmetry and the fluctuation propagates to capillary die and consequently results in chaotic appearance of the extrudates. This behavior looks similar to the turbulent flow of a non-elastic liquid. However, this turbulent-like flow pattern for a polymer melt occurs at very low Reynolds number and is believed to be originated by the elastic nature of the polymer melt. This is why GMF is often called by 'Elastic Turbulence'(6). GMF seems to occur when the extensional stress at the entrance of die exceeds a critical condition (7) that seems to depend only on the polymeric fluid and little or not at all on the die diameter, length and die material (11). Kim and Dealy (12) showed that a critical tensile stress estimated from the entrance pressure drop can serve as a universal criterion for the onset of GMF. They also investigated the effect of molecular structure on the critical tensile stress with several polyethylenes. They found that the critical tensile stress is independent of molecular weigh for constant polydispersity, but increases with increase in long chain branching and polydispersity.

During the study of GMF, we observed an interesting phenomenon in LLDPE extrusion. The occurrence of cavitation was observed in the die exit region and this was always accompanied by GMF of extrudate. Here, we present several experimental results for this cavitation phenomenon and discussions for a possible cause.

Experimental

Materials

The polymer investigated is a linear low-density polyethylene, AFFINITY (13) (trademark of the Dow) EG8100 characterized by a melt index of 1.0 g/10 min and a density of 0.87 g/cm^3. This polymer is commercially available and metallocene catalyzed.

Apparatus and method

In a previous work, Migler et al. utilized a sapphire capillary die situated at the exit of a twin screw extruder. Same equipment was used in this study. A microscope with stroboscopic illumination was constructed at the die exit in order to image the flow inside the capillary. Particle tracking velocimetry with a high-speed video camera (1000 frames/s) attached to a microscope was utilized to measure the velocity profile of the polymer melt inside the capillary. The length and the radius of the capillary used are 32 mm and 0.8 mm, respectively. Details about the apparatus can be found elsewhere (14). A fluoropolymer-coated capillary was also used to investigate its effect on the GMF. Only the inner wall near the exit (from the die exit to about 2 mm upstream) was coated. The coating involves repetitive introduction of a dilute mixture (1% of fluoropolymer) of acetone and Dynamar^{TM} FX9613 into a hot die (140 ^oC) and the subsequent evaporation of the acetone. The coated fluoropolymer achieves good adhesion with the sapphire die upon heating to 160 ^oC for several minutes at the highest throughput. Experiments are conducted at 140 ^oC and 160 ^oC and various throughputs ranging from 1 g/min to 25 g/min.

Result and Discussion

The flow curve (ΔP versus flow rate) of LLDPE at two different temperatures is shown in Fig. 1, and Fig. 2 shows photographs of the extrudate identified by the numbers in Fig. 1. The arrows shown in Fig. 1 indicate the onset of GMF determined by appearance of the extrudate, i.e. severe and chaotic distortion. Because the focus of this study is on GMF regime, we did not show data for the stable flow regime, where the extrudate has a smooth and glossy surface. In fact, extrudate produced at a minimum flow rate available in our extruder and the temperature of 140 ^oC still showed the sharkskin defect. Onset of SMF for 160 ^oC was about 1 g/min. A stick-slip or spurt flow regime accompanied by a typical flow curve discontinuity was not observed in this study. A stick-slip behavior was observed in many studies (9, 15, 16), where a constant flow rate-device was used. In our study, a twin screw extruder was used and probably this equipment is neither constant pressure nor constant flow.

Below the throughput for the onset of GMF, the extrudate surface shows a typical sharkskin texture, i.e., a small amplitude and high frequency disturbance (number 1 & 5 in Fig. 2). Above the onset of GMF, the appearance of the extrudate changes dramatically, characterized by its wavy, rough and chaotic nature. In the GMF regime, a striking phenomenon was observed. We observed cavitation in the die exit region. Fig. 3 shows a typical example. The time interval between each photo is 1 ms. At first, only flowing LLDPE is observed in the capillary. Then all of sudden, a black spot appears and its size grows rapidly as can be seen in next photo. We believe that this is not an optical illusion (something like shadow) but a cavity. When we coated the lip of the die exit with black materials to prevent possible formation of an illusion, same thing was observed.

The size of the cavity reaches a maximum at about 5 ms after its creation and then decreases rather slowly until it vanishes completely. The lifetime of most cavities ranges from 20 ms to 25 ms. The shape of the cavities is highly irregular and its maximum size varies from several tens of microns to about 150 microns in width and length. From the velocity measurement of the melts as a function of the radial position of the capillary and the shape change of the cavities, we conclude that the size of the cavity in the radial direction is much smaller than its width and length.

Fig. 4 shows optical micrographs for cavities when the focal plane is varied from the wall to a plane 300 μm above the wall. The cavity on the focus of capillary wall is most clearly seen. As the focus departs from the wall, the image becomes dull and blurred. This implies that the cavity exists on the capillary wall or at least be very close to the wall. Fig. 5 is another proof for this fact. Comparing the velocity profile of the polymer melts and the velocity of the cavities, it can be concluded that the cavities are in contact with the wall or very close to the wall (within the range of 50 μm from the wall). The fact that cavity exists near the wall may implies that the shear stress of the melts plays an important role in the cavitation phenomenon. However, we observed that the radial position of the cavitation was not (or little) affected by an increase in the throughput or decrease in the temperature. If the shear stress were the dominant factor for the cavitation, the thickness of the zone, where cavitation occurs, would be wider with increase in throughput or decrease in temperature, i.e. the increase in the shear stress. This will be discussed in more detail, later on.

Fig. 6 shows the average size of the cavity at various processing conditions. We plotted the average area of the cavities projected to the optical microscope instead of real size of the cavity because it is impossible to measure real 3-D size with our experimental capability. It is clearly seen that the cavitation phenomenon becomes more severe as the throughput increases and the temperature decreases, which results in the increase in shear stress or extensional stress of the polymer melt. The cavitation frequency, i.e. how many times the cavitations occur per unit period of time, shows the same trend. Fig. 7 shows the average size of the cavities at various axial positions from the die exit. The cavitation occurs mostly at die exit region and becomes rare with the distance from the exit. The cavitation frequency shows the same trend. We observe that cavitation occurs at the upstream of the exit, 0.5 ~ 1.5 mm, depending on experimental conditions.

At this point, we wish to discuss the cause or mechanism for this cavitation phenomenon. The fact that the cavitation increases with increase in throughput and decrease in melt temperature, implies that the shear and/or extensional stress level plays an important role in this phenomenon. Both shear stress (17, 18) and extensional stress (19) can produce cavitation in a polymer melt when the induced stress is greater than cohesive force of the polymer melts (20). In this study, the role of shear flow was excluded because of the fact that thickness of the zone, where the cavitation occurs, was not or little affected by experimental conditions as mentioned earlier. It is clear that surface layer is accelerated from rest (stick at the wall) to the extrudate velocity at the die exit (21). This is main source for the generation of extensional flow field in vicinity of die lip of the exit region. It seems that the extensional flow at the die exit is very relevant to the cavitation phenomenon. However, the fact that the cavitation zone extends up to 1.5 mm upstream from the die exit, make us doubt that extensional flow at the die exit is a direct cause for the cavitation. A numerical calculation predicted that the extensional flow is significant within several microns from the die exit (22). Migler et al. observed that the polymer melt approaching the die exit near the capillary wall does not accelerate up to 30 μm upstream of the exit (23). This is also what we observed in this study.

In order to clear out whether the extensional flow in the die exit is a direct cause of the observed cavitation, we performed the following two experiements: 1) Addition of an obstacle next to the die exit, and 2) A fluoropolymer coating on the inner wall of the die exit (up to ~ 2 mm upstream). We found that no cavitation occurs even at very high throughput when we put the obstacle. In this case, the surface of the extrudates is much smoother than the extrudate upon GMF. They show only small-scaled roughness (ripple-like shape) on the extrudate surface. When we coated the die exit with the fluoropolymer, the onset of GMF and cavitation was not changed. A fluoropolymer is known to diminish polymer adsorption and allow significant wall slip and consequently decrease the extensional stress in the die exit region, significantly (13, 24). Therefore, the fact that the fluoroplymer coating at the die exit does not change the behavior of GMF and cavitation at all, suggests that the extensional stress at die exit does not play a direct or critical role. It is well documented that the entrance region plays a critical role on the onset of GMF. We observed the cavitation always occurs upon GMF regime. It is most unlikely that accordance of the cavitation and GMF regime is just a coincidence. Therefore, we speculate that most likely, unstable, chaotic and fractured flow at the entrance is a direct cause or seed of the cavitation at the die exit region though we do not know an exact mechanism at this point. As described in Introduction, the melts fracture at the entrance to die upon GMF regime. As this unstable polymer melt approaches the die exit, the extensional stress (even weak as in the case of the fluoropolymer coating) may initiate and propagate the cavitation or nucleate. However, if there is no extensional flow (We believed that presence of the obstacle eliminates completely the extensional flow since the velocity discontinuity at the die exit disappears with obstacle due to its expanding flow pattern.), the unstable melt may not be initiated to form the cavitation. These results cause us to speculate that cavitation events near the exit also contribute to GMF as a mean of initiator or nucleator.

Temblay (22) suggested that sharkskin may be the result of a cavitation event. He showed numerically that large negative pressure can exist at the die exit by simulating the flow of a linear polydimethylsiloxane melt using a finite element program. However, we observed the cavitation to occur only at throughput well beyond the onset of sharkskin. We conclude that his suggestion is not the explanation for our observation.

Conclusion

The cavitation phenomenon is observed in LLDPE extrusion process. We observe cavitation in the first 1500 microns upstream of the exit at our highest flow, where we observe gross melt fracture. We see cavities at the wall form (seemingly out of nothing) grow to a length and width of about 150 microns, then shrink down and disappear. From velocity measurements of these structures, we conclude that their width in the radial direction is very thin compared to its width in axial and lateral direction, and that they are in contact with the wall. The process for the growth and disappearance is approximately 20 ~ 25 ms. We observe the cavitation to occur only at throughputs where the gross melt fracture is observed. From these facts, we conclude that the cavitation phenomenon are closely related to the melt fracture at die entrance.

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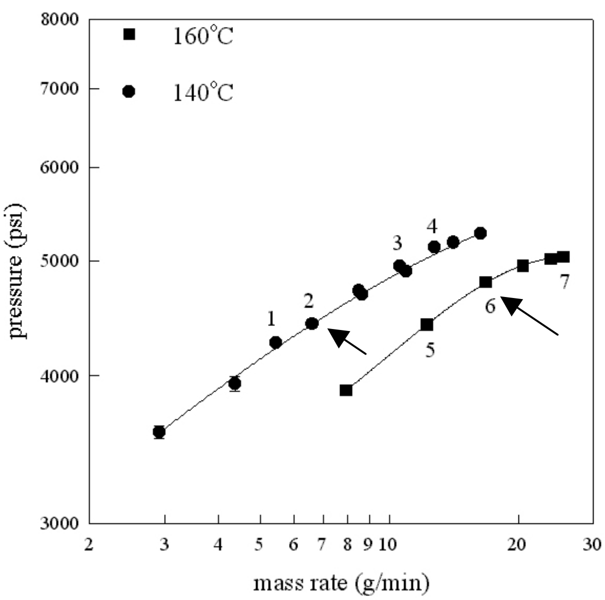

Fig. 1 Flow curves of LLDPE at two different temperature.

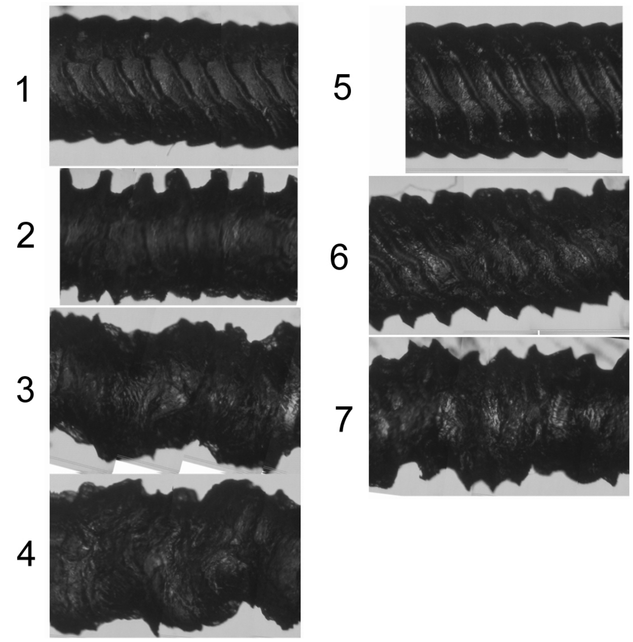

Fig. 2 Extrudate obtained under various processing conditions. Numbers in each photo correspond to those in Fig. 1.

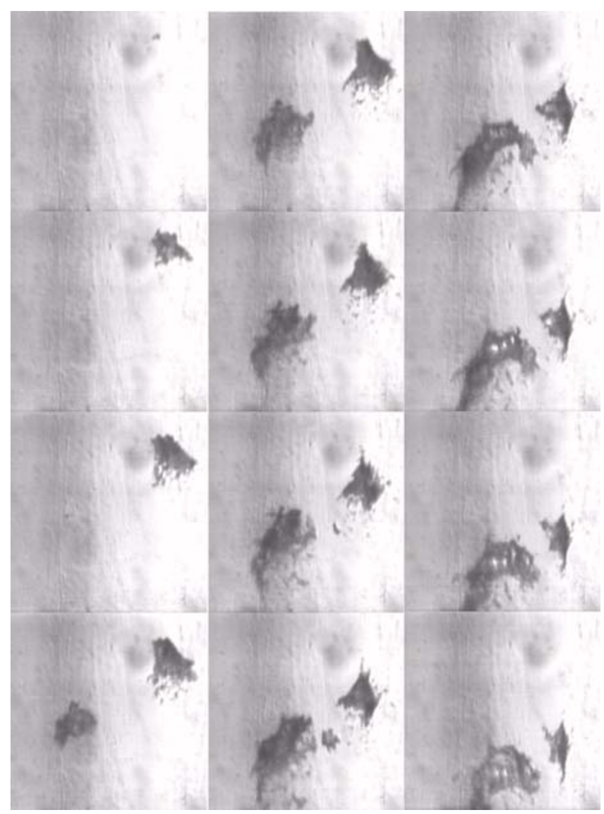

Fig. 3 Example of formation and extinction of the cavity Extrusion condition for this set of photograph corresponds to number 3 in Fig. 1.

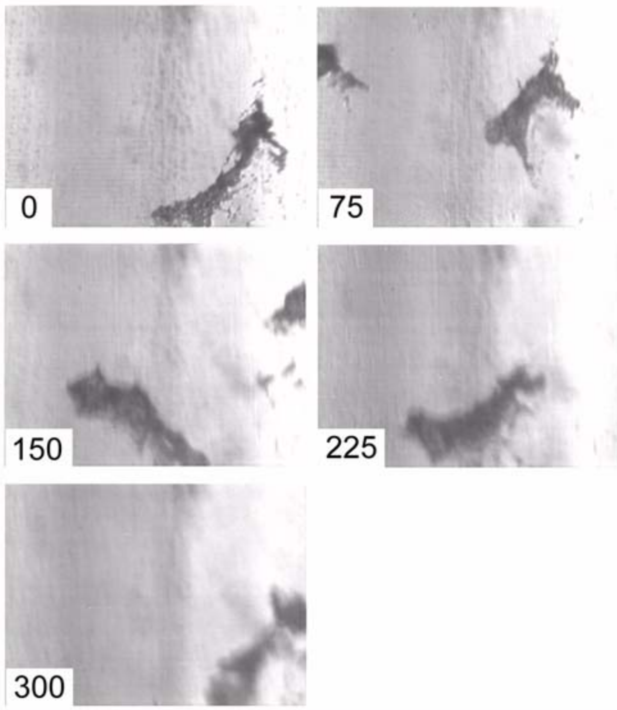

Fig. 4 Optical micrograph for the cavitation at various focus from the tube wall. Numbers in each photos are in micrometer. Extrusion temperature =160 °C, screw speed/ flow rate = 40 rpm/ 7.4 g/min

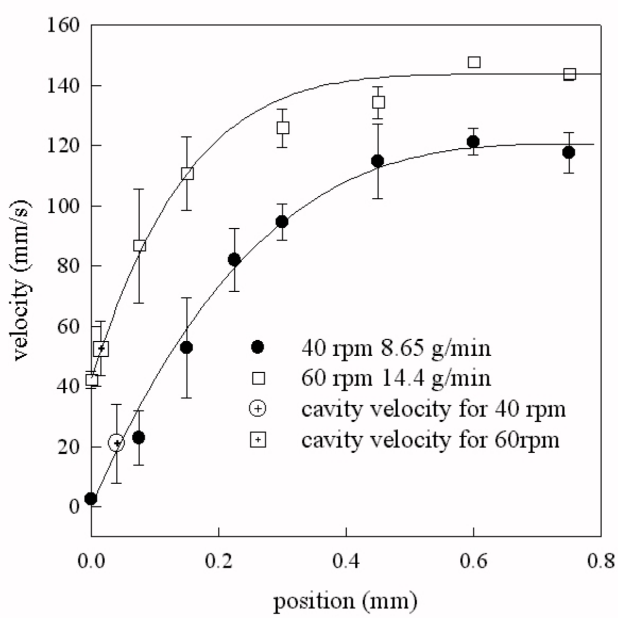

Fig. 5 Velocity profile of the polymer melt inside the capillary tube and cavity velocity at two different screw speed. Extrusion temperature = 140 °C,

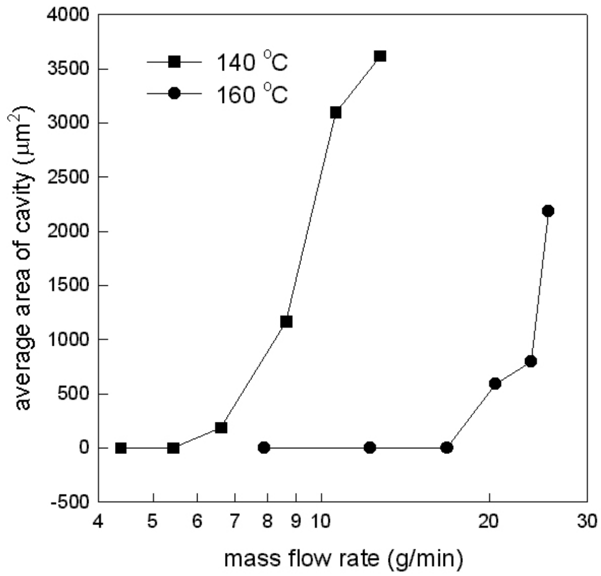

Fig. 6 Average area of the cavity versus mass flow rate.

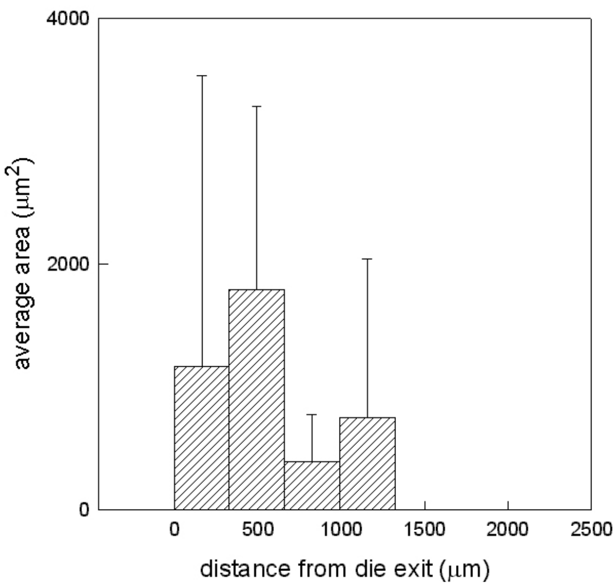

Fig. 7 Average area of the cavity versus distance from the capillary exit. Extrusion temperature