

# Effect of Vibration Extrusion on High-Density Polyethylene

Xueqin Gao, Jie Zhang, Changsen Chen, Kaizhi Shen

College of Polymer Science and Engineering, State Key Laboratory of Polymer Materials Engineering, Sichuan University, Chengdu, Sichuan 610065, People's Republic of China

Received 12 July 2006; accepted 14 March 2007

DOI 10.1002/app.26551

Published online 26 June 2007 in Wiley InterScience (www.interscience.wiley.com).

**ABSTRACT:** A new type of extrusion apparatus with a hydraulic vibration field was self-developed to study the effects of vibration on the rheological behavior, mechanical properties, and morphology of high-density polyethylene (HDPE) during the process of extrusion. In the research, the morphology and mechanical properties of specimens produced by conventional extrusion molding were used for comparison with specimens obtained via vibration extrusion molding. The research showed that the extrusion property of the HDPE melt was improved under a vibration field, and the apparent viscosity decreased. The maximum decrement of the apparent viscosity was 37.98%. The change in the apparent viscosity of the HDPE melt corre-

sponded to the vibration frequency and amplitude, die temperature, and rotational speed of the extruder screw. Both the longitudinal and transverse yield strengths of the HDPE specimens increased under the vibration field. The increment of the longitudinal yield strength (11.5%) was bigger than that of the transverse yield strength (6.9%). According to scanning electron microscopy micrographs, the vibration field made the crystallites orient and become smaller, and the crystal size became more homogeneous. © 2007 Wiley Periodicals, Inc. *J Appl Polym Sci* 106: 552–557, 2007

**Key words:** mechanical properties; morphology; rheology

## INTRODUCTION

During polymer molding and processing, the morphology of a polymer, which determines the macroscopic performance of the final products, is affected by the processing conditions. To control the morphology of a polymer and enhance the properties of the final products during the process of extrusion, the technology of vibration extrusion has been reported in several articles.<sup>1–6</sup> According to them, vibrations fall into four classes: mechanical vibrations, electromagnetic vibrations, ultrasonic vibrations, and hydraulic vibrations. The frequency of ultrasonic vibrations is very high, as much as 20 kHz, whereas the frequency of mechanical vibrations and the majority of hydraulic vibrations is generally low. To understand this technology deeply, especially in the case of medium-to-high vibration frequencies, which have been seldom studied, we developed a late-model hydraulic vibration extrusion apparatus with a frequency range of 0–93.3 Hz. With this equipment, the influence of the vibration field on the rheological behavior of the polymer melt and the changes in the mechanical properties and morphology of the

polymer products produced by vibration extrusion were studied.

## EXPERIMENTAL

### Equipment

A schematic drawing of the equipment is shown in Figure 1. Its vibration frequency, which was in the range of 0–93.3 Hz, and vibration amplitude, which was in the range of 0–30 mm, were continuously adjustable. Because of the high hydraulic pressure and direct action of the vibration ram on the polymer melt, the vibration intensity was high. The influence of the vibration field on polymer extrusion molding was studied with this equipment

### Material

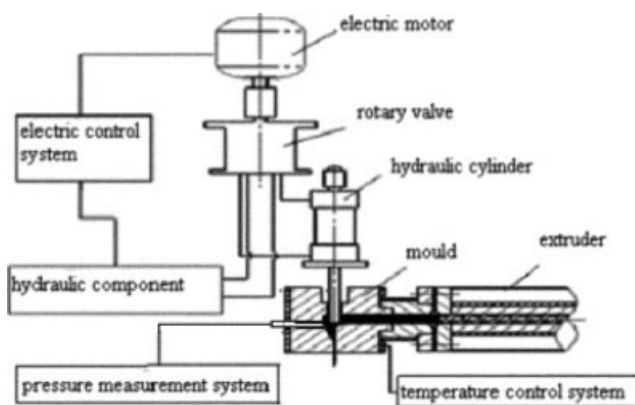
High-density polyethylene (HDPE) 6100M, with a melt index of 0.15 g/10 min (230°C, 2.16 kg) and a density of 0.954 g/cm<sup>3</sup>, was commercially obtained from Lanzhou Chemical Industry Co. (Lanzhou, China).

### Sample preparation

As shown in Figure 1, an extruder was used to plasticize the materials. A slit die was attached to the extruder. The vibration piston rod of the hydraulic

Correspondence to: S. Kaizhi (gxqlnw@yahoo.com.cn).

Contract grant sponsor: National Natural Science Foundation of China.



**Figure 1** Schematic drawing of the vibration extrusion system.

cylinder was put in the melt chamber. When the vibration rod did not act, the experimental device was equivalent to a conventional extrusion machine. Otherwise, when the vibration rod acted, this experimental device could implement vibration extrusion. During the course of vibration extrusion, the vibration amplitude (0–30 mm) and vibration frequency (0–93.3 Hz) were adjustable. The temperature and pressure of the melt in the melt chamber could be monitored at any time. The slit die, fixed on the extruder, was used to produce an HDPE sheet, from which specimens were prepared through the cutting of the sheet with a special cutting knife. The specimen for the tensile test was dumbbell-shaped.

### Tensile test

A Shimadzu model AG-10TA tensile machine (Japan) was employed for tensile testing at room temperature; the crosshead speed was 50 mm/min.

### Scanning electron microscopy (SEM) investigation

A Hitachi model S-450 scanning microscope (Japan) was used for the SEM measurements. The specimens were sputtered after being etched in a permanganate etching solution with a coating machine (model DMX-220A) (Beijing Instrument Factory, China). The temperature of the specimens was strictly controlled below 45°C during the entire process.

### Differential scanning calorimetry (DSC) investigation

DSC tests were conducted on a Netzsch DSC204 differential scanning calorimeter (Germany), which scanned at 10°C/min. The weight of the samples for each test was 5 mg. During the measurements, dried nitrogen gas was purged at a constant flow rate.

### Analysis of the apparent viscosity

The apparent viscosity could not be obtained directly in the experiment, but the mass of the polymer melt flowing out of the die in a certain interval could be measured. To compare the rheological properties of the HDPE melt under vibration extrusion and under conventional extrusion, the apparent viscosity was calculated with the volume flow rate.

A slit die [slit width ( $W$ ) = 60 mm, slit length ( $L$ ) = 30 mm, and slit height ( $h$ ) = 0.5 mm] was attached to the extruder in this experiment. With the die regarded as a slit rheometer because of  $W/h > 20$ , the mass of the melt and the differential pressure were recorded. Then, the apparent viscosity ( $\eta_a$ ) was calculated according to the following formula:

$$\eta_a = \frac{\tau}{\dot{\gamma}_a} = \frac{h\Delta P}{2L} \bigg/ \frac{6Q}{Wh^2} = \frac{Wh^3\Delta P}{12QL} = \frac{Wh^3\Delta P\rho t}{12LM} \quad (1)$$

where  $\tau$  is the shear stress,  $\dot{\gamma}_a$  is the rate of shear,  $Q$  is the volume flow rate,  $\Delta P$  is the differential pressure,  $\rho$  is the density of material,  $t$  is the interval of time, and  $M$  is the mass of the sample.

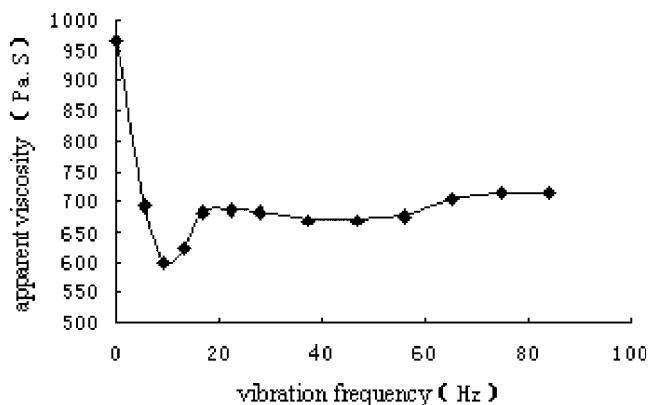
Because the purpose of analyzing the apparent viscosity was only to compare the differences between the apparent viscosities under vibration extrusion and under conventional extrusion, the formula with a non-Newtonian exponential was not adopted. It would not influence the comparative results.

## RESULTS AND DISCUSSION

### Rheological behavior

Rheological behavior affected by different vibration frequencies

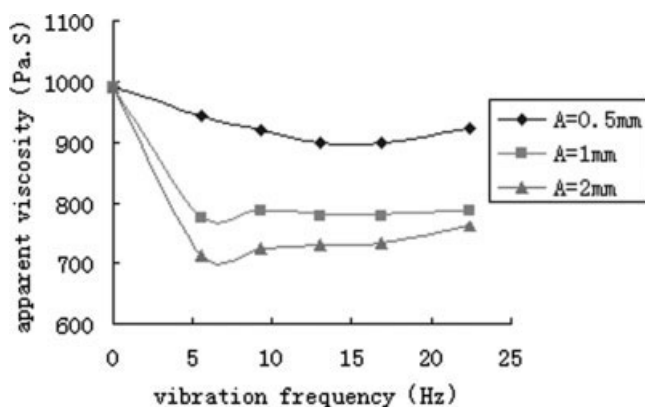
Under certain processing conditions (the die temperature is 180°C, the screw rotary speed is 5.56 rpm, and the vibration amplitude is 1 mm), the curve of the apparent viscosity of the HDPE melt versus the vibration frequency is shown in Figure 2. The apparent viscosity under the condition of dynamic extrusion decreases obviously compared with the apparent viscosity under conventional extrusion. When the vibration frequency is 0–9.3 Hz, the apparent viscosity decreases sharply with increasing vibration frequency. It reaches the minimum at 9.3 Hz, a reduction of 37.98% compared with that of the apparent viscosity under conventional extrusion. Then, it backs up. However, when the frequency is 20–84 Hz (the highest value studied), the viscosity, which is still much lower than the apparent viscosity under conventional extrusion, remains basically invariable. We discuss only the effect of the vibration frequency below 30 Hz in the rest of this article.



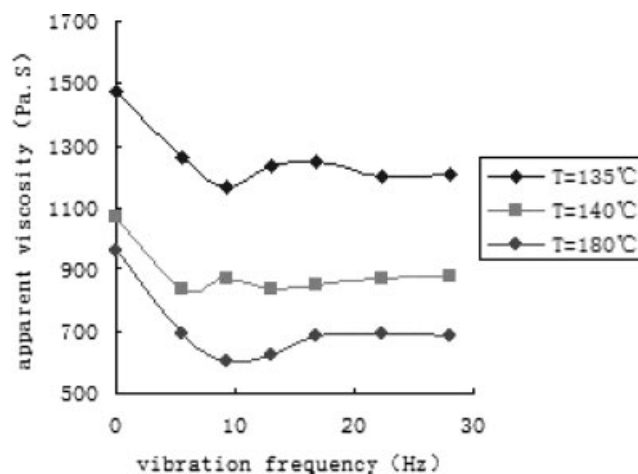
**Figure 2** Effects of the vibration frequency on the HDPE apparent viscosity.

#### Rheological behavior affected by different vibration amplitudes

The effect of the vibration frequency on the rheological properties of the HDPE melt at different amplitudes (0.5, 1, and 2 mm) was tested when the die temperature was 150°C and the screw rotary speed was 5.56 rpm. For the sake of prolonging the time of vibration action on the melt, the slow screw rotary speed was chosen. A periodic pressure fluctuation occurred in every short-time cycle under a large amplitude and frequency. However, the mean pressure in the long time was stable. In the experimental process, no melt slide at the wall of the die was found. We can see from Figure 3 that the apparent viscosity of the HDPE melt decreases under vibration, and the change tendency with an increasing vibration frequency at different vibration amplitudes is similar to what we discuss in a previous section, but the degree of the effect is different: a large amplitude makes the viscosity decrease more than a small amplitude. When the amplitude is 0.5 mm, the maximum decrement of the apparent viscosity is 9.22%. When the amplitude is 2 mm, the maximum decrement is 28.05%.



**Figure 3** Effects of the vibration frequency on the HDPE apparent viscosity at different amplitude ( $A$ ) values.



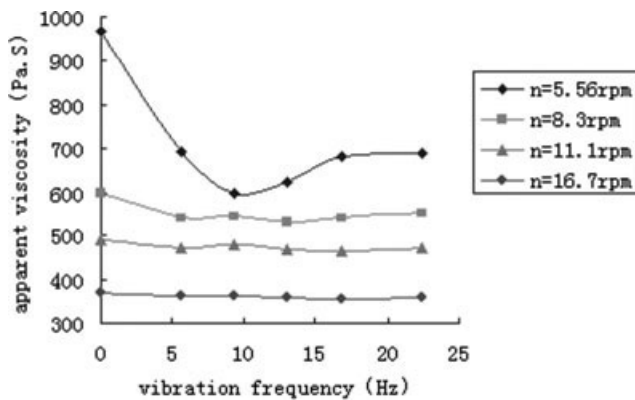
**Figure 4** Effects of the vibration frequency on the HDPE apparent viscosity at different die temperature ( $T$ ) values.

#### Rheological behavior affected by vibrations at different die temperatures

Figure 4 shows curves of the apparent viscosity versus the vibration frequency at different die temperatures under certain processing condition (the screw rotary speed is 5.56 rpm, and the vibration amplitude is 1 mm). The higher the die temperature is, the smaller the apparent viscosity is. The decrement of the apparent viscosity is different at different die temperatures: 20.64, 22.35, and 37.98% at 135, 140, and 180°C, respectively. This indicates that the apparent viscosity decreases slightly under vibration when the melt temperature is near the melting point (ca. 130°C). This might be because the activity of molecular chains is very weak at lower temperatures. Under the vibration field, it is difficult to make molecular chains disentangle accordingly. As the temperature rises, the disentanglement and orientation of the molecular chains become easier. Thus, the decrement value of the apparent viscosity at 180°C is bigger than those at 135 and 140°C.

#### Rheological behavior affected by vibrations at different screw rotary speeds

Figure 5 shows curves of the apparent viscosity versus the vibration frequency at different screw rotary speeds under certain processing condition (the die temperature is 180°C, and the vibration amplitude is 1 mm). We can see that the lower the screw rotary speed is, the higher the apparent viscosity is and the bigger the decrement of the apparent viscosity is. For example, the maximum decrement is 37.98% when the screw rotary speed is 5.56 rpm, whereas the maximum decrement is only 3.54% when the screw rotary speed is 16.70 rpm. This indicates that the effect of decreasing the apparent viscosity under vibration will be weakened with an increasing screw rotary speed.



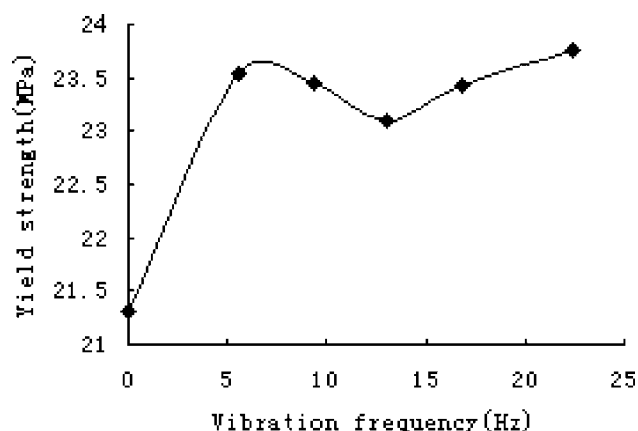
**Figure 5** Effects of the vibration frequency on the HDPE apparent viscosity at different screw rotary speed ( $n$ ) values.

This might be because the higher the screw rotary speed is, the shorter the residence time of the polymer in the die and the vibration action time on the polymer are. Moreover, when the screw rotary speed increases, the pressure in the die becomes higher. The effect of the vibration field on the polymer is relatively weakened under this condition. In another study about extrusion by vibration, such as ultrasonic vibration, although the vibration frequency was very high, the lower the screw rotary speed was, the more significant the apparent viscosity reduction was.<sup>6</sup>

## Mechanical properties

### Longitudinal mechanical properties

For HDPE samples prepared by vibration extrusion, we call the extrusive direction the longitudinal direction and the direction normal to the extrusive direction the transversal direction. Figure 6 shows the curve of the longitudinal yield strength under different vibration frequencies when the die temperature is 150°C, the screw rotary speed is 5.56 rpm, and the vibration



**Figure 6** Curve of the longitudinal yield strength at different vibration frequencies (die temperature = 150°C, screw rotary speed = 5.56 rpm, amplitude = 1 mm).

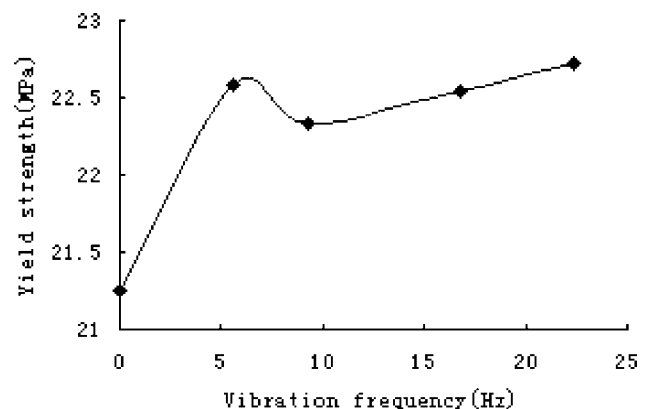
amplitude is 1 mm. We can see that the longitudinal yield strengths of the vibration samples are all bigger than those of the conventional samples. The maximum longitudinal yield strength of the samples prepared by vibration extrusion is 23.76 MPa, which is an increase of 11.5% versus the strength of the conventional samples, which is 21.31 MPa. This indicates that the longitudinal yield strengths of the HDPE6100M samples increase under the action of a vibration field.

### Transversal mechanical properties

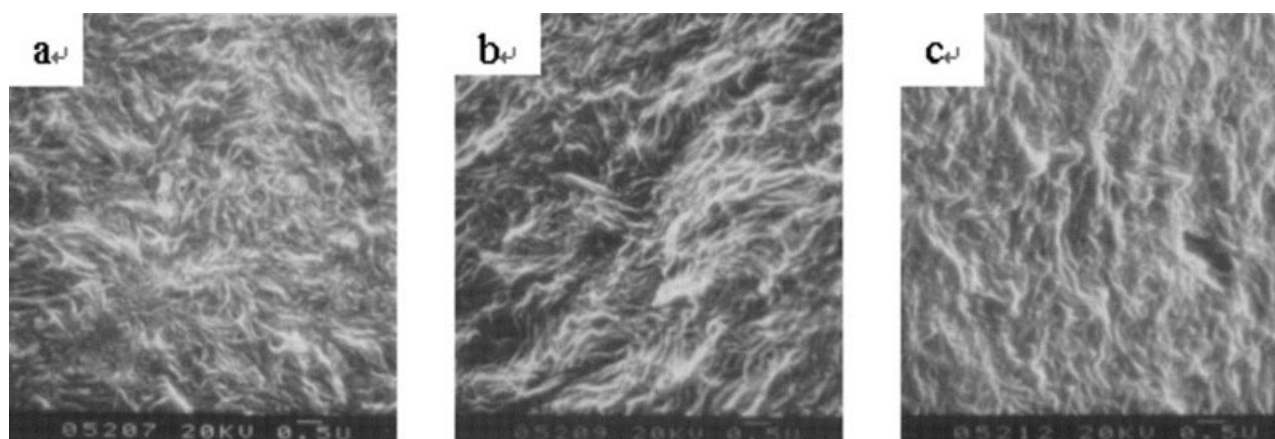
Figure 7 shows the curve of the transversal yield strength under different vibration frequencies under the same conditions used for the longitudinal samples. The change mode of the transversal yield strength under different vibration frequencies is similar to that of the longitudinal yield strength. The maximum increment of the transversal yield strength is 6.9%. From Figures 6 and 7, we can see that the longitudinal and transverse yield strengths of 5.6 Hz are higher than those of 9.3 Hz. This is perhaps because the molecular chains barely have time to respond when the vibration frequency is high. However, the longitudinal and transverse yield strengths of 22.4 Hz are highest. This should be because the strong vibration field can diminish the crystal grain size. Grain refinement will increase the mechanical properties of specimens to a certain extent. Anyway, both the longitudinal yield strengths and transversal yield strengths of the vibration samples increase versus those of conventional samples. The increment of the transversal strength is lower than that of the longitudinal strength. Thus, we can conclude that biaxial self-reinforced samples can be obtained via vibration extrusion.

### SEM analysis

Figure 8(a) is an SEM micrograph of an HDPE conventional extrusion-molded specimen. The horizontal



**Figure 7** Curve of the transversal yield strength at different vibration frequencies.

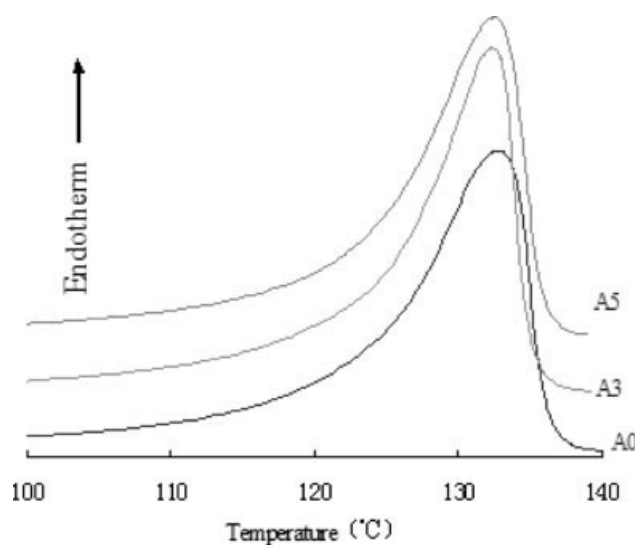


**Figure 8** SEM micrographs of etched HDPE extrusion-molded specimens produced at 150°C: (a) nonvibration specimen, (b) vibration specimen (frequency = 5.6 Hz, amplitude = 1 mm), and (c) vibration specimen (frequency = 9.3 Hz, amplitude = 1 mm).

direction is the extrusive and vibratory direction. The crystal structure of the conventional specimen is spherulites. The spherulites consist of many platelets that grow around their nuclei, reflecting a disorderly distribution in all directions. From Figure 8(b), we can see that the crystals orient obviously when the vibration frequency is 5.6 Hz. Platelets arrange in a parallel fashion toward the upper right. This is because the samples were placed slantways when we performed the SEM test. Figure 8(c) is an SEM micrograph of a specimen prepared when the vibration frequency was 9.3 Hz. We can see that the crystal orients along the extrusive direction more obviously.

### DSC analysis

Figure 9 shows DSC curves of conventional and vibration HDPE 6100M specimens produced at



**Figure 9** DSC curves of the HDPE 6100M specimen.

150°C and 5.56 rpm. DSC analysis could be used to obtain the crystal fusion heat of HDPE. The crystallinity of the specimen could be calculated with the following equation:

$$\alpha_c = \frac{\Delta H_f}{\Delta H_{f\alpha}} \times 100\%$$

where  $\alpha_c$  is the crystallinity of the specimen,  $\Delta H_f$  is the measured crystal fusion heat of the specimen from a DSC thermogram, and  $\Delta H_{f\alpha}$  is the perfect crystal fusion heat of HDPE (293 J/g). The temperatures of the melting peaks for the HDPE specimens prepared under conventional and vibration conditions and the calculated crystallinity values are shown in Table I. The melting peaks of the vibration specimens produced at 5.6 and 9.3 Hz both have small decrements compared with that of the conventional specimen. This indicates that melt vibration might reduce the lamellar thickness and make the crystal size smaller. Some increase in the fusion heat in the vibration specimen can be found in comparison with the conventional specimen. Therefore, the crystallinity of the vibration specimen increases about 3% versus that of the conventional specimen, as shown in Table I. This shows that the vibration field can make a polymer melt form a crystal nucleus more easily.

**TABLE I**  
DSC Test Results for the HDPE 6100M Specimen

Sample	Vibration frequency (Hz)	Oscillating amplitude (mm)	Melting peak (°C)	Enthalpy (J/g)	Crystallinity (%)
A0	0	0	133.2	182.2	62.18
A3	5.6	1	132.4	190.5	65.02
A5	9.3	1	132.52	191.7	65.42

Generally, mechanical properties increase obviously in the orientation direction and decrease in the direction normal to the orientation. Because the crystallites of the vibration samples orient along the extrusive direction (longitudinal direction of the samples), the longitudinal yield strength of the vibration samples increases. According to the aforementioned test, we know that the transversal mechanical properties of the vibration sample do not decrease but increase slightly. Maybe this is because the vibration field can make the crystallite smaller and make the crystallinity increase, and this can increase the strength. In the transversal direction, an increase in the mechanical properties caused by increased crystallinity and smaller crystallites is larger than a decrease in the mechanical properties caused by longitudinal orientation. Thus, the transversal mechanical properties increase in general. However, the effect of an improvement in the transversal mechanical properties is not more distinct than that of the longitudinal mechanical properties.

### CONCLUSIONS

On the basis of the results and discussion, the following conclusions can be drawn:

1. The extrusion property of the HDPE melt is improved under a vibration field. The apparent viscosity has a relationship with the vibration frequency, amplitude, die temperature, and screw rotary speed. With the vibration frequency increasing, the apparent viscosity decreases to the minimum and then backs up. However, when the frequency is 20–84 Hz (the highest value in our experiments), the viscosity,

which is still much lower than the apparent viscosity under conventional extrusion, remains basically invariable. The bigger the vibration amplitude is, the bigger the effect is on the apparent viscosity under the vibration field. When the die temperature exceeds the melting point, the effect on the apparent viscosity under the vibration field increases. The higher the screw rotary speed is, the lower the effect is on the apparent viscosity under the vibration field.

2. Both the longitudinal and transverse yield strengths of the HDPE specimen increase under the vibration field. The maximum increment of the longitudinal yield strength is 11.5%, whereas that of the transverse yield strength is 6.9%. According to SEM micrographs, the vibration field can make the crystallites orient, the crystal grain become refined, and the crystal size become more homogeneous.

Anyway, vibration extrusion can improve the rheological properties of polymer melts and the mechanical properties of specimens and change the morphology structure accordingly.

### References

1. Allan, P. S.; Bevis, M. J. *Plast Rubber Proc Appl* 1991, 16, 132.
2. Fridman, M. L.; Peshkovsky, S. L.; Vinogradov, G. V. *Polym Eng Sci* 1981, 21, 755.
3. Casulli, J.; Clermont, J. R.; Von Ziegler, A.; Mena, B. *Polym Eng Sci* 1990, 30, 1551.
4. Isayev, I.; Wong, C. M.; Zeng, X. *J Non-Newtonian Fluid Mech* 1990, 34, 375.
5. Peng, X.-F.; Qu, J.-P. *Polym Mater Sci Eng* 2001, 17, 129.
6. Chen, G.; Guo, S.; Li, H. *J Appl Polym Sci* 2002, 86, 23.