Improved Mechanical Properties and Structure of Polypropylene Pipe Prepared Under Vibration Force Field

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ABSTRACT: The polypropylene (PP) pipes were prepared using a self-made electromagnetic dynamic plasticating extruder, which introduced a vibration force field into the whole plasticating and extrusion process by the axial vibration of the screw. The effects of the vibration frequency and the vibration amplitude on the mechanical properties and microstructure of PP pipes were investigated using bursting pressure testing, tensile testing, impact testing, differential scanning calorimetry (DSC), and wide-angle X-ray diffraction (WAXD). The mechanical properties testing showed that the circumferential strength of PP pipes increased significantly, and the biaxial self-reinforcement pipes could be obtained. Also, the impact strength was improved. When compared with the conventional static extruded specimens, the maximum increase of bursting pressure, tensile yield strength, and impact strength were 27.03%, 7.3%, and 16.2%, respectively. DSC and WAXD analysis showed that the PP pipes obtained by vibration plasticating extrusion (VPE) had higher crystallinity, higher melting temperature, more perfect crystals, and smaller crystal sizes, but no new polymeric crystalline peak appeared. The improvement of mechanical properties of the PP pipes prepared by VPE was attributed to the higher crystallinity and the improvement of the molecular orientation and of the crystalline morphology under the action of the vibration force field. © 2009 Wiley Periodicals, Inc. J Appl Polym Sci 114: 3612–3620, 2009

Key words: polypropylene; pipe; mechanical properties; microstructure; vibration force field

INTRODUCTION

Polymer processing is an important part of macro-molecule material science and engineering in modern times. It is also one of the most crucial approaches to improve the properties of the polymer materials. In recent years, the vibration technique has been applied to polymer extrusion processing and gained a lot of detailed investigations.[1–11] The vibration force field introduced into polymer extrusion processing affects the molecular movement and the rheological behavior of the polymer melt, which in turn affects the morphological development of the final extruded products and then the final properties.

Till now, several types of melt vibration techniques have been investigated during extrusion. For instance, Casulli et al.[2] superimposed longitudinal and transverse oscillations on the die section at the exit of an extruder. The results showed that the die swell diminished and the mechanical properties of the extrudate increased. Wong et al.[3] developed a sound wave extrusion system and investigated the effect of the parallel superposition of large amplitude oscillation on the flow of polymer melts through an annular die, and found that the die pressure at the entrance reduced and the temperature for the melt exiting the die increased. Guo et al.[8] used a special ultrasonic oscillation extrusion system to greatly improve the surface appearance of the LLDPE extrudate, and the productivity of LLDPE extrudate increased in the presence of ultrasonic oscillation. Jiang et al.[10] successfully used a mandrel-rotating die to obtain high-hoop-strength HDPE pipe by self-reinforcement. Gao et al.[11] developed a new type of extrusion apparatus with a hydraulic vibration field to produce HDPE sheets, and found that both the longitudinal and transverse yield strengths of the HDPE specimens increased.

Unfortunately, most of the processing operations introduce the mechanical vibration, ultrasonic oscillation, or hydraulic vibration into the polymer melt only to the limited zone inside the die. Hence, the vibration effect is very limited because the affected area of vibration is local. Furthermore, it is very difficult to extend to industrialization production because every die for producing product needs special design, and some of the setup or apparatus are very complicated. On the contrary, the electromagnetic dynamic plasticating (EMDP) extruder for plastics invented by Qu[12–14] is another example using the vibration technique. In this kind of

extruder, the vibration force field is applied to the solid conveying, plasticating, melting, and melt flow of the entire extrusion molding process by the axial vibration of the screw. Because of the vibration of the screw, an additional stress is superimposed on the shear flow of the polymer melt, which alters the rheological behavior of the melt. Therefore, the inner microstructure of the product is also changed during forming and solidifying of the extrudate, and the product properties are improved significantly under the action of the vibration force field. The EMDP extruder can produce extruded products under the conditions of industrial production. Later researches showed that the EMDP extruder for plastics had significant advantages, compared with conventional extruder, such as low energy consumption, low melt apparent viscosity, high output, superior kneading and mixing effect for polymer melt, and excellent mechanical properties of the extruded products.[15–19] For example, in extruded HDPE/CaCO$_3$ nanocomposites through EMDP, it is found that the dispersion of nanoparticles in the HDPE matrix can be improved and the tensile and impact strengths of the products can be increased by the vibration force field.[18] Nevertheless, few investigations have been carried out on the effect on the relationship between microstructures and mechanical properties of plastic pipes.

Plastic pipes are widely used in many fields including building, municipal construction, hydraulic engineering, and farmland irrigation, etc. When compared with traditional pipes (cast iron pipe, steel pipe, concrete pipe, etc.), plastic pipes have multiple advantages such as low weight, small flow resistance, excellent corrosion resistance, good sealing performance, and long service life. So plastic pipes play a very important role among industrial materials and the consumptions every year during the world are very tremendous. However, the strength of plastic pipe is much lower than traditional pipe. In engineering applications, plastic pipes have to be reinforced to meet the high demands on strength. So how to improve the strength of the plastic pipe by self-reinforcement is our keen interest.

Based on the virtues of the EMDP extruder,[15–19] in this work, an attempt was made to improve the mechanical properties of polypropylene (PP) pipes by vibration extrusion process. We extruded PP pipes using an EMDP extruder, and investigated the effects of vibration force field on the microstructure and mechanical properties of the samples.

EXPERIMENTAL

Material

The material used in this work was PP grade EP240H, provided by Basel Company, with a melt
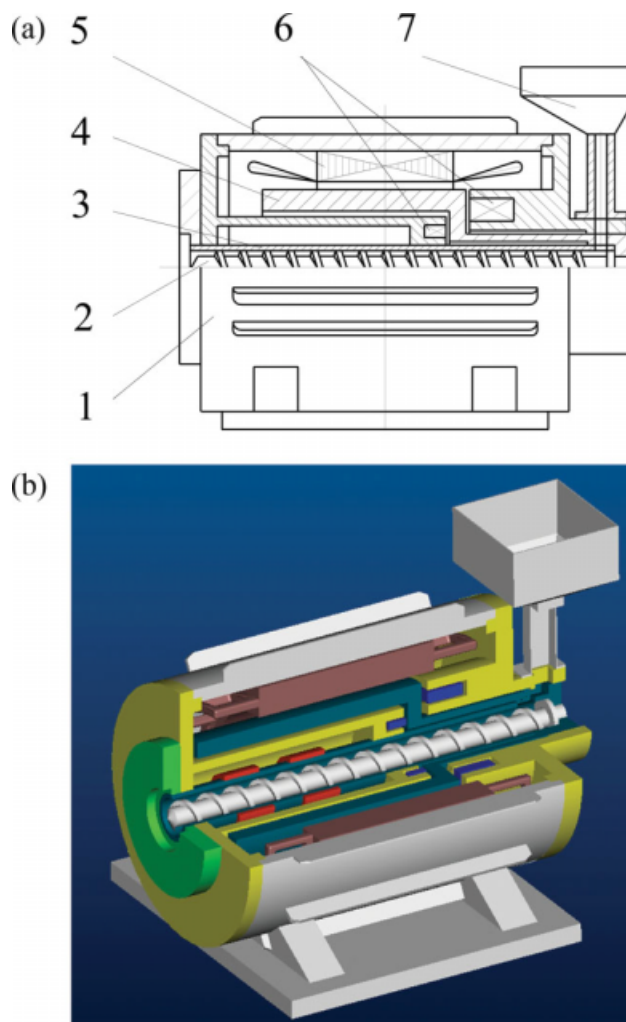

Figure 1 SJDD-260 electromagnetic dynamic plasticating extruder. 1, base; 2, screw; 3, barrel; 4, rotor; 5, stator; 6, electromagnetic support; and 7, hopper. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

flow index of 1.8 g/10 min measured at 230°C under 2.16 kg.

Apparatus

The apparatus used in this experiment consisted of a self-made EMDP extruder (model SJDD-260), a spiral mandrel die, and auxiliary equipments.

The EMDP extruder is different from traditional extruder. The schematic drawing of this extruder is shown in Figure 1. Its barrel and screw were inserted into a motor cavity, which was specifically designed, and the screw was fixed to the rotor coaxially. The screw can rotate periodically along with the rotor induced by the electromagnetic coil. At the same time, the back side of the screw was fixed to the metal plank of the vibration inducer, which made the screw vibrate in the axial direction
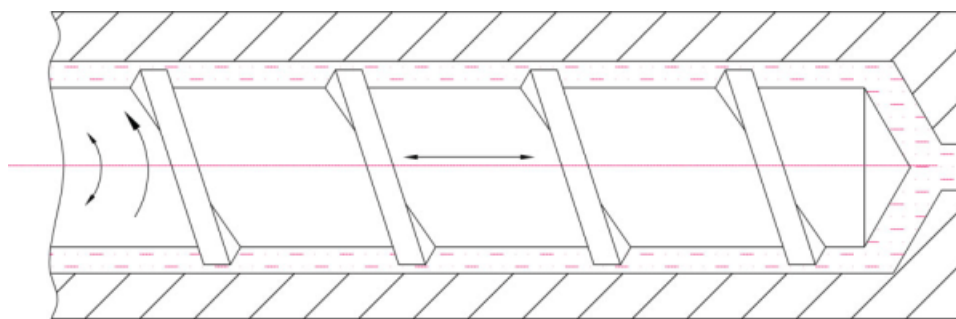
Figure 2 Schematic diagram of the rotation and axial vibration of the screw. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

along with the metal plank. The rotor rotation and plank vibration were controlled by different currents. The diameter of the screw was 30 mm and the L/D ratio was 23 : 1. The vibration frequency and amplitude could be adjusted by the vibration inducer independently. When the screw rotated and simultaneously vibrated axially in pipe extrusion, as shown in Figure 2, solid transportation, plasticating, melting, and extruding were all under the action of a vibration force field, implying that the vibration was superposed on the whole plasticating and extrusion process by the screw. This is called vibration plasticating extrusion (VPE). If the screw did not do axial vibration, the extruder was equivalent to a traditional single-screw extruder. In this case, it is mentioned as conventional static extrusion (CSE).

The die used in this work was a self-made spiral mandrel die. The reason for designing a spiral mandrel die is that it has many advantages[20–24]: the absence of mandrel support elements that disturb the melt flow, hence no weld lines or flow markings; uniform melt flow and good mixing performance; uniform wall thickness; low pressure drop; low mechanical stress and thermal stress; low melt temperature build-up at high output; and reasonable self-cleaning properties. The main parameters of the designed spiral mandrel die are listed in Table I.

The auxiliary equipments were composed of vacuum sizing box, traction device, and cutting device. The vacuum sizing box and traction device were made by Zhangjiagang Wansu Co., China.

A schematic diagram of the experimental setup is illustrated in Figure 3. The polymer was plasticized and extruded by the EMDP extruder, and a spiral mandrel die to form a hollow tube. The tube was passed through the vacuum sizing bush, in which it was sized and cooled preliminarily, then entered the vacuum sizing box that included spray water system and was cooled to form a pipe. Finally, the pipe was dragged by the traction device and cutted by the cutting device according to the specified length.

Sample preparation

The extrusion temperature profile used for PP was 190, 225, 225, and 220°C from hopper to die, the screw rotation speed was maintained at 65 rpm. The VPE specimens influenced by vibration frequency and vibration amplitude were prepared, respectively, with CSE specimens for comparison. For VPE, vibration frequency of the screw in the axial direction was varied from 2 to 14 Hz, and the amplitude was 50–200 μm.

The diameter and wall thickness of the final PP pipes obtained in this research were 32 mm and 2.4 mm, respectively.

Bursting pressure testing

The valid length of PP pipes used for bursting pressure testing was 600 mm. The pipes were tested in a press machine (40-SY, Changsha, China) at room temperature (23°C). First, the pressure was up to 2.6 MPa, and then remained for 1 h. Finally, the pressure was increased continuously until the pipes were broken. The average value of three samples was reported for each process condition.

Tensile testing

Dumbbell specimens were prepared along the axial direction (MD) of the extruded pipe samples according to GB/T 8804-2003. The valid geometry of the specimens is $25 \times 6 \times 2.4$ mm^3. An

TABLE I
The Main Parameters of the Spiral Mandrel Die

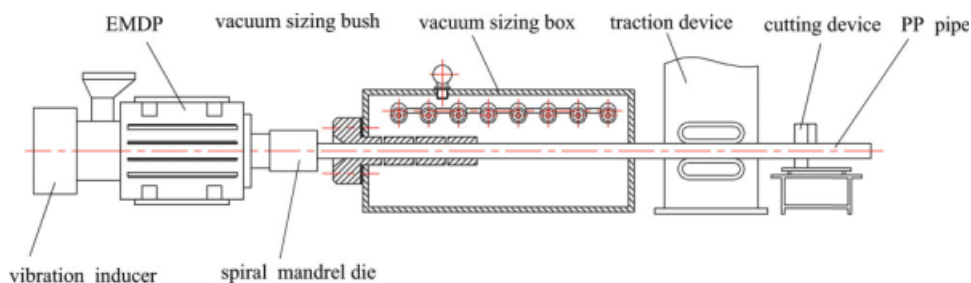| Main parameters	Parameters value
Mandrel diameter (mm)	90
Number of spiral channels	4
Taper angle (°)	3.54
Spiral run out angle (°)	4
Helix angle (°)	12.37
Initial width of spiral channel (mm)	14
Initial depth of spiral channel (mm)	13

Figure 3 Schematic diagram of experimental setup. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

INSTRON_Merlin Series 5566 Testing System produced by American INSTRON Company was used for tensile testing at room temperature, and the crosshead speed was 100 mm/min. The average value of five samples was reported for each processing condition.

Impact testing

The specimens were prepared along MD of the extruded pipe samples according to GB/T 1843-1996. The specimens were obtained by machining the pipes into $80 \times 10 \times 2.4$ mm^3 bars along MD. The bars were notched with a single-tooth cutter. The notch depth was 2 mm. An INSTRON_Merlin Series POE 2000 Charpy-type Impact Machine produced by American INSTRON Company was used for impact testing at room temperature. The average value of five samples was reported for each processing condition.

Differential scanning calorimetry investigation

Differential scanning calorimetry (DSC) studies were conducted on a NETZSCH DSC204 differential scanning calorimeter, which scanned at 10°C/min in the temperature range of 25–200°C under nitrogen atmosphere. Each sample was weighed 4–6 mg. All results were recorded for the first heating of the samples. The DSC peak separation technique was adopted using the Peak Separation software provided by NETZSCH Corp. in the DSC204 equipment.

DSC can be used to obtain the crystal fusion heat of PP. The crystallinity of the samples can be calculated from the following equation:

$$X_c = \frac{\Delta H_f}{\Delta H_{fc}} \times 100\%$$

where X_c is the crystallinity of the sample, ΔH_f is the measured crystal fusion heat of the sample from a DSC thermogram, and ΔH_{fc} is the perfect crystal fusion heat of PP, 209 J/g.

Wide-angle X-ray diffraction investigation

Wide-angle X-ray diffraction (WAXD) experiments were conducted using a RIGAKU D/MAX-IIIA X-ray diffractometer. A conventional Cu Kα X-ray tube at 40 kV and 30 mA was applied to obtain WAXD spectrum. WAXD intensities were recorded from $2\theta = 3°$ to $2\theta = 50°$ in transmission mode in samples at a scan step of 0.02°. The average crystalline sizes (crystalline domain size) were calculated by the Scherrer equation:

$$L_{hkl} = \frac{K\lambda}{B \cos \theta}$$

where L_{hkl} is the crystalline domain size (in nm), K is the shape factor of the crystalline ($K = 0.9$), λ is the wavelength (in this experiment $\lambda = 0.15406$ nm), θ is the diffraction angle (in degree), and B is the full width at half maximum of the hkl peak (in radian). L_{hkl} is considered as an average crystalline dimension perpendicular to the reflection planes (hkl).

RESULTS AND DISCUSSION

Mechanical properties

Figures 4 and 5 show changes of bursting pressure with vibration frequency (F) and vibration amplitude (A), respectively. The data points where vibration frequency is zero or vibration amplitude is zero represent the samples prepared by CSE. The experimental results show that the bursting pressure all increases obviously with increasing vibration frequency under the same vibration amplitude or increasing vibration amplitude under the same vibration frequency, and then, with further increase in frequency or amplitude, it decreases. However, it is higher than that of the CSE samples. When the vibration frequency is 10 Hz and the vibration amplitude is 100 µm, the bursting pressure reaches to a maximum value (4.7 MPa), a 27.03% increase compared with the CSE samples (3.7 MPa).
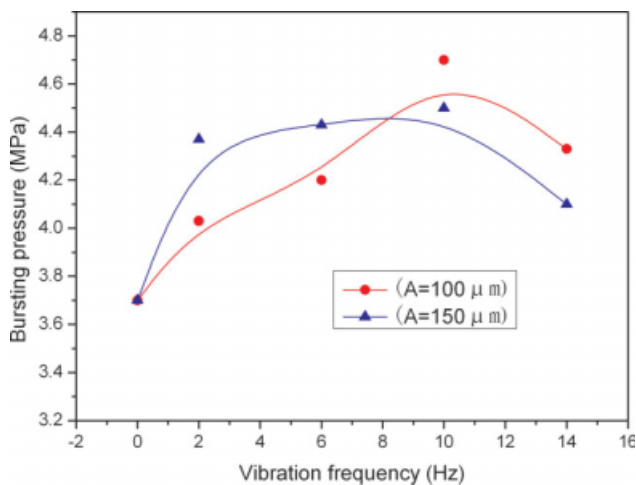
Figure 4 Bursting pressure of PP pipe under different vibration frequency. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]
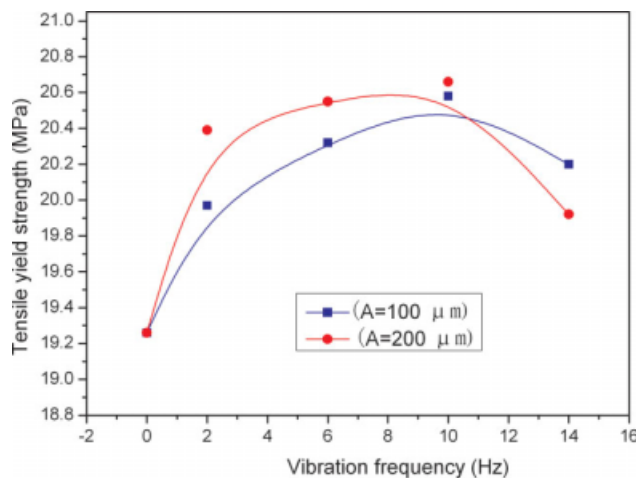

Figure 6 Tensile yield strength of PP pipe under different vibration frequency. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

Generally, the axial strength of plastic pipe prepared by traditional extrusion process is much higher than circumferential strength. This can be explained as follows. During CSE process, most of the macromolecular chains align along the flowing direction and few of them align along the circumferential direction (TD). After leaving the die, the polymer melt stretches greatly in MD because of the large drag force of the traction device. The molecular orientation in MD is much larger than that in TD, which results in higher pipe strength in MD than that in TD. In addition, the axial stress in pipe wall is half of the circumferential stress, when an internal hydraulic pressure is applied to thin-walled pipe.[10] So the strength in TD is poorer than in MD and pipes are very easily destroyed in that direction. To

improve the strength of the plastic pipes, we must increase the circumferential strength at first. In this work, the bursting pressure of the PP pipes obtained by VPE is enhanced dramatically, namely the circumferential strength is increased notably by self-reinforcement, and this is very important for the application of the plastic pipes which bear an internal hydraulic pressure.

Figures 6 and 7 show changes of tensile yield strength in MD with vibration frequency and vibration amplitude, respectively. The experimental results show that the tensile yield strengths of the VPE samples are all greater than those of the CSE samples. Furthermore, the tensile yield strength reaches to the maximum value (20.66 MPa) when the vibration frequency is 10 Hz and the vibration
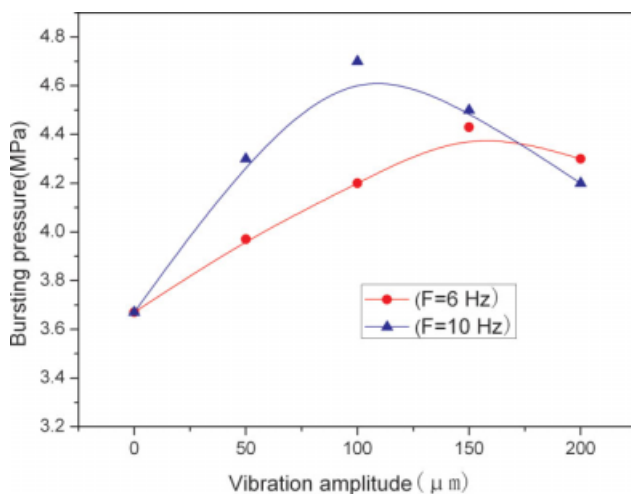

Figure 5 Bursting pressure of PP pipe under different vibration amplitude. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

Figure 7 Tensile yield strength of PP pipe under different vibration amplitude. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]
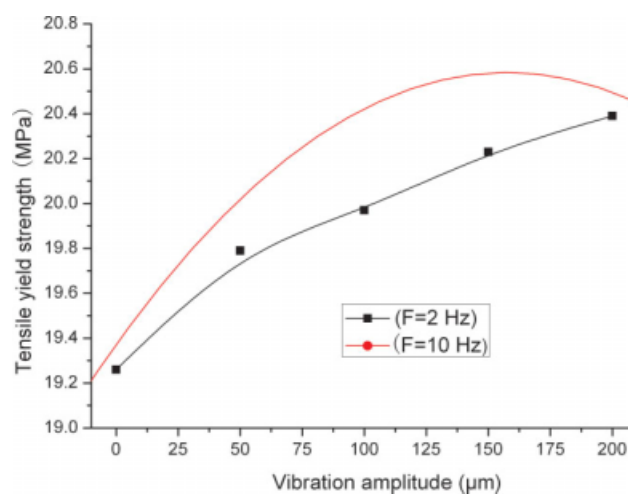
amplitude is 200 μm, which is an increase of 7.3% versus the tensile yield strength of the CSE samples (19.26 MPa).

Figures 8 and 9 show changes of impact strength in MD with vibration frequency and vibration amplitude, respectively. The experimental data show that most of the VPE samples have remarkable enhancement in impact toughness. When the vibration frequency is 6 Hz and the vibration amplitude is 100 μm, the impact strength of PP pipe is 35.61 kJ/m^2, a 16.2% increase compared with the CSE samples (30.64 kJ/m^2). However, under excessive vibration frequency (more than nearly 14 Hz) and amplitude (more than nearly 200 μm), the impact strength decreases compared with that of the CSE sample.

These experimental results show that the VPE process has an important effect on the mechanical properties of extruded PP pipes. The bursting pressure is increased dramatically and the tensile yield strength in MD is enhanced simultaneously, which means that the VPE has a considerable self-reinforcement effect on the circumferential strength of pipes without reducing the tensile strength in MD. Also, the impact toughness is improved dramatically. These effects are mainly owing to the occurrence of stress-induced crystallization and molecular orientation.[25] During VPE process, in fact, the polymer melt bears a complex shear force in the flowing direction and axial direction imposed by the screw rotation and axial vibration, as shown in Figure 2. Accordingly, the molecular chains are sheared both in circumferential and axial direction, the molecular chains and their segments can obtain instantaneous impulses[26–28] easily from the axial vibration force. Also, because of decreased chain entanglement under the action of the vibration force field, the

Figure 9 Impact strength of PP pipe under different vibration amplitude. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

mobility of the molecular chains and their segments can be enhanced, that is to say the ability of the chains and their segments to move to different directions can be increased. So the molecular chains are oriented not only in the flowing direction but also in TD. It is beneficial to crystallization, and those molecules oriented in the direction other than the flowing direction can form microcrystals or serve as strong connections between lamellas.[27] In this case, the mechanical strength of amorphous phase can be enhanced. All these are propitious to the improvement of the uniformity of pipes mechanical properties. However, when the instantaneous impulse is immoderate under the action of the vibration force field, that is to say under the excessive vibration frequency and amplitude; the molecular chains and their movement of segments cannot catch up with the vibration frequency, namely, the molecular chains and their segments barely have time to respond, when the vibration frequency is too high. Thus PP orientation and crystallization declines, which is probably the main reason why the impact strength is lower than the CSE sample, when the vibration frequency is >14 Hz, as shown in Figure 8.

DSC analysis

Figures 10 and 11 show DSC curves of PP samples prepared at different vibration amplitude when the vibration frequency is 10 Hz and at different vibration frequency when the vibration amplitude is 100 μm, respectively. The curves are vertically offset for clarity. It can be seen that the melting point of the sample obtained by VPE shifts toward the higher temperature. This means that, by VPE, more perfect crystals can be achieved. When compared with the CSE samples, the maximum increase of the melting
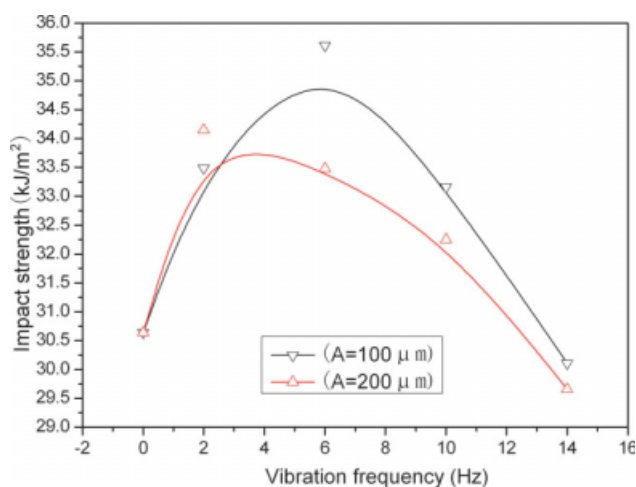

Figure 8 Impact strength of PP pipe under different vibration frequency. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]
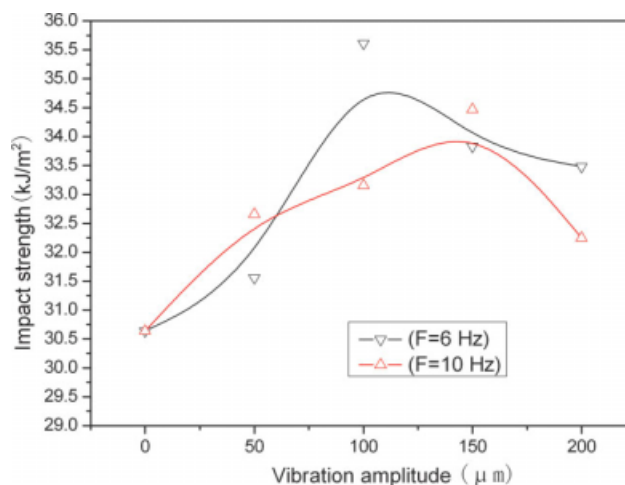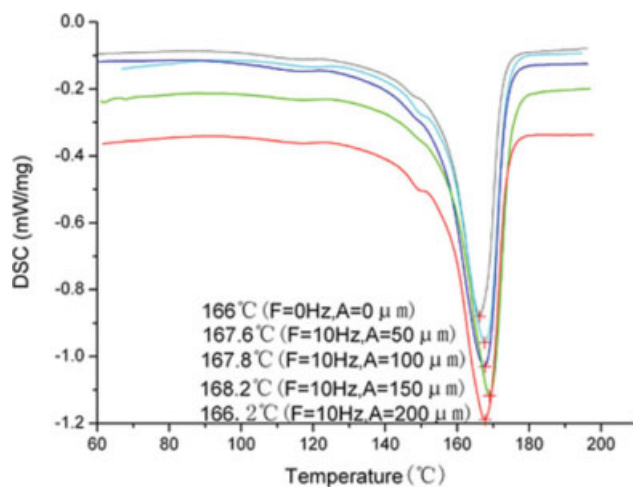
Figure 10 DSC curves of PP samples obtained at different vibration amplitude when the vibration frequency is 10 Hz. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

temperature is 3.2°C from 166 to 169.2°C, when the vibration frequency is 14 Hz and the vibration amplitude is 100 μm.

The calculated crystallinity values of PP samples prepared at different vibration amplitude when the vibration frequency is 10 Hz and at different vibration frequency when the vibration amplitude is 100 μm are given in Tables II and III, respectively. It can be found that the crystallinity of the VPE samples increases in comparison with the CSE samples. When the vibration frequency is 2 Hz and the vibration amplitude is 100 μm, the crystallinity increases 2.42% from 30.61 to 33.03%.
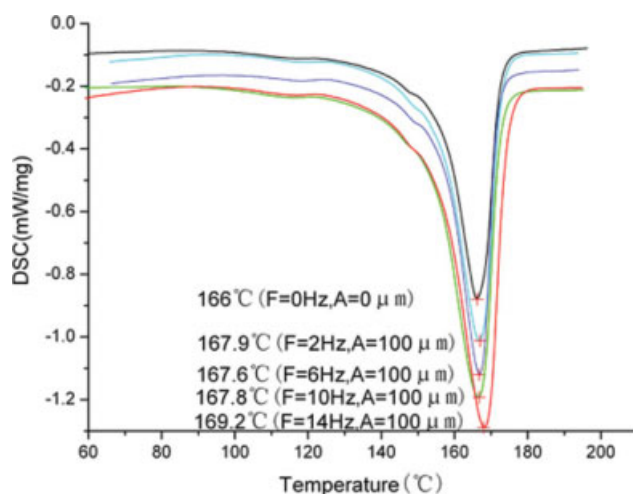

Figure 11 DSC curves of PP samples obtained at different vibration frequency when the vibration amplitude is 100 μm. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

TABLE II
DSC Results of the PP Samples Prepared at Different Vibration Amplitudes When the Vibration Frequency is 10 Hz

Vibration amplitude (μm)	Enthalpy, ΔH_f (J g^{-1})	Crystallinity (%)
0	63.97	30.61
50	69.00	33.01
100	67.48	32.29
150	64.81	31.01
200	66.82	31.97

The DSC results mean that the VPE process can make the PP melt form crystal nuclei more easily and facilitate the perfection of the crystals. This may be explained as follows. As discussed earlier, because of decreased chains entanglement and instantaneous impulses obtained associated with the vibration force field, the molecular chains and their segments are easily oriented under the inducement of the shear flow and elongational flow, which results in the form of more local ordered structures. These ordered structures facilitate nucleation when the melt is cooled, and more perfect crystals are formed, so the melting temperature shifts toward the higher temperature. At the same time, the vibration force field can promote the portion at the higher side of the molecular weight distribution melt uniform and form perfect crystals. Therefore, the increase in melting temperature may be owing to the vibration resulting in crystal perfection, which improves the mechanical properties of the PP samples. On the other hand, the molecular chains of PP are quite flexible with few branches. It is easy for the chains to rearrange themselves into the crystalline lattice under the vibration force field. So the crystallinity of PP is improved by the vibration force field, which is propitious to improve the tensile strength of the PP samples, and the crystalline structure is more thermally stable. Vibration also increases the glass transition temperature of the amorphous regions in the interspherulites, presumably as a result of orientation of the molecules in

TABLE III
DSC Results of the PP Samples Prepared at Different Vibration Frequencies When the Vibration Amplitude is 100 μm

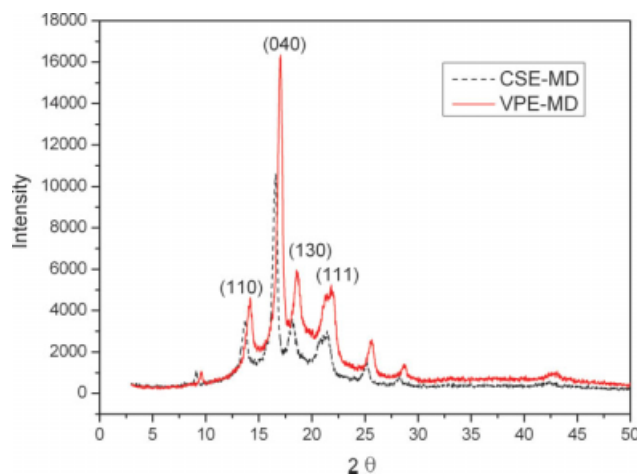| Vibration frequency (Hz)	Enthalpy, ΔH_f (J g^{-1})	Crystallinity (%)
0	63.97	30.61
2	69.03	33.03
6	65.50	31.34
10	67.48	32.29
14	65.02	31.11

Figure 12 WAXD curves of PP samples obtained by CSE and VPE ($F = 10$ Hz, $A = 100$ μm) in MD. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

this region,[6,27] and the orientation of the interlamellar and interspherulitic ties, which has a considerable impact on the mechanical properties.[6,27] All these facilitate the improvement of mechanical properties of the PP samples.

WAXD analysis

Figures 12 and 13 show WAXD curves for PP samples obtained by CSE and VPE (vibration frequency is 10 Hz, vibration amplitude is 100 μm) in MD and TD, respectively. As can be seen, these curves are very similar, no new polymeric crystalline peak has been observed in spectrum of the VPE samples compared with the CSE samples, which denotes that no
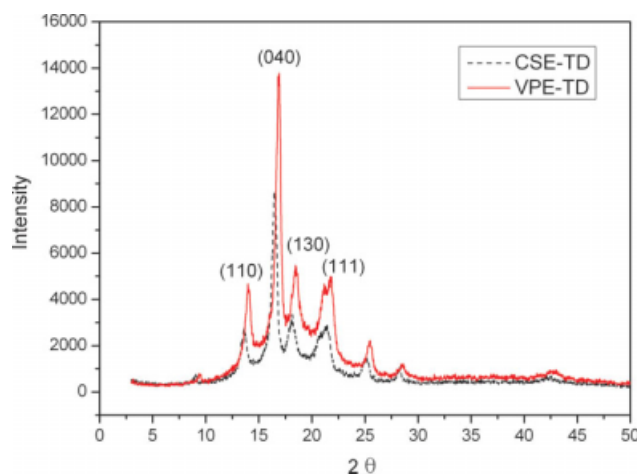

Figure 13 WAXD curves of PP samples obtained by CSE and VPE ($F = 10$ Hz, $A = 100$ μm) in TD. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

TABLE IV
The Average Crystalline Sizes of PP Samples Obtained by CSE and VPE in MD

Vibration frequency (Hz)	Vibration amplitude (μm)	hkl	L_{hkl} (nm)
0	0	110	15.94
		040	17.92
		130	15.27
6	150	110	15.78
		040	17.46
		130	15.18
10	100	110	14.94
		040	17.09
		130	14.69
14	200	110	15.15
		040	16.42
		130	14.74

change in the crystalline form in the VPE samples. Both VPE and CSE samples exhibit the same α crystal from the spectrum of PP. Furthermore, it is obvious that the diffraction intensity of the crystalline planes (110), (040), (130), and (111) of the VPE sample both in MD and TD are greatly increased. This indicates that the orientation degree of (110), (040), (130), and (111) planes of the VPE sample is increased both in MD and TD. This is one of the reasons why the the tensile strength in MD and circumferential strength of the VPE samples are improved.

The results of the average crystalline sizes for the CSE and VPE samples in MD and TD are summarized in Tables IV and V, respectively. It can be found that the average crystalline sizes of (110), (040), and (130) crystalline planes of the VPE samples decrease compared with those of the CSE samples, indicating that more small crystallites are formed in the VPE samples, that is to say the VPE can make the crystallite smaller, and this can increase the impact strength. It is in line with the test results of the mechanical properties.

TABLE V
The Average Crystalline Sizes of PP Samples Obtained by CSE and VPE in TD

Vibration frequency (Hz)	Vibration amplitude (μm)	hkl	L_{hkl} (nm)
0	0	110	15.60
		040	15.93
		130	14.65
6	150	110	14.99
		040	14.76
		130	13.55
10	100	110	14.63
		040	15.87
		130	13.37
14	200	110	14.96
		040	15.72
		130	13.04

CONCLUSIONS

On the basis of the results and discussions, the conclusions can be drawn as follows:

1. VPE can improve the overall mechanical properties of PP pipes effectively. By VPE, the circumferential strength of PP pipes increases significantly, and the biaxial self-reinforcement pipe can be obtained. Also, the impact strength is improved. When compared with the CSE samples, the maximum increment of the bursting pressure, tensile yield strength, and impact strength of the VPE samples is 27.03%, 7.3%, and 16.2%, respectively.

2. VPE is an effective processing approach for the microstructure development control of PP. By VPE, the PP pipes have higher melting temperature, higher crystallinity, smaller crystal sizes, higher orientation degree of crystalline planes, and more perfect crystals. All these are favorable for the improvement of the mechanical properties of the PP pipes.

In short, the improvement of mechanical properties of the PP pipes prepared by VPE is attributed to the increase of the degree of crystallinity and the improvement of the molecular orientation and of the crystalline morphology structure under the action of the vibration force field.

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