

# Effects of Ultrasonic Oscillations on Rheological Behavior and Mechanical Properties of Novel Propylene-Based Plastomers

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**ABSTRACT:** The effects of ultrasonic oscillations on the die pressure, productivity of extrusion, melt apparent viscosity, melt surface appearance, and die swell of novel propylene-based plastomers were studied in a specially designed ultrasonic oscillations extrusion system developed in our laboratory. The effects of ultrasonic oscillations on molecular weights, tensile strength, and dynamic mechanical properties of extrudates were also studied. The experimental results showed that the presence of ultrasonic oscillations during extrusion could significantly increase the productivity of plastomers at the same die pressure, and reduce die swell and melt fracture such as sharkskin at a given screw rotation speed. The die pressure and apparent viscosity of plastomers remarkably

decreased with increasing ultrasonic intensity. Introduction of ultrasonic oscillations into plastomer melts can improve their processibility. The possible mechanism for ultrasonic improvement of rheological behavior was also proposed in this article. Under certain conditions, ultrasound-assisted extrusion could slightly decrease the glass transition temperature ( $T_g$ ) and storage modulus of plastomers due to the minor reduction in molecular weights, but showed no significant impact on yield strength and strength at break. © 2007 Wiley Periodicals, Inc. *J Appl Polym Sci* 106: 1725–1732, 2007

**Key words:** polypropylene; ultrasonic oscillations; rheology; molecular weight

## INTRODUCTION

Polypropylene (PP) is a semicrystalline polymer with very good mechanical and processing properties, chemical resistance, low density and relatively low cost.<sup>1</sup> VERSIFY™ (trademark of the Dow Chemical Company) Plastomers and Elastomers are a versatile family of propylene-ethylene copolymers synthesized by a revolutionary catalyst in combination with Dow's proprietary INSITE technology and solution process.<sup>2</sup> Polymer ethylene contents are between 5 and 15 wt %. With ethylene content increasing, the polymers show elastomeric properties. They have a

unique molecular architecture different from Ziegler-Natta catalyst-based and metallocene catalyst-based copolymers of propylene. Their narrow molecular weight distribution (2–3) and broad melting distribution (ca. 50–145°C) of the resins result in improved temperature performance against metallocene catalyst-based products of comparable olefin content. The unique molecular architecture of these new polymers provides films, fibers, and molded parts with an outstanding combination of excellent optics, sealing, and hot tack performance, plus elasticity, flexibility, softness, and compatibility in blends.<sup>3</sup> However, the narrower molecular weight distribution and more uniform comonomer incorporation also lead to higher die pressure, higher apparent viscosity and melt fracture, which are the key concern to some applications. As compared with ethylene-based materials, although propylene-based materials have better processibility due to their high shear thinning rheological behavior, their key weakness is the susceptibility to chain scission. Therefore, the improvement of processibility through increasing temperature and shearing may not be practical for propylene-based materials. The traditional routes for solving the above-mentioned problems are the addition of processing aids or plasticizers.<sup>4</sup> However,

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these methods are not efficient for these special polymers and are harmful to performance characteristics such as optics and mechanical properties. Accordingly, seeking innovative routes for improving the processibility of a narrow molecular weight distribution material continues to be very challenging. The recent development of melt vibration technology that gives relatively "mild" shearing condition seems to be an effective method and is proposed to be studied for improving the processing behavior of propylene-based materials.<sup>5-7</sup>

In addition, ultrasonic oscillations have been gradually introduced into polymer processing as a clean, efficient and attractive melt vibration technology in recent years. Isayev and coworkers<sup>8-12</sup> reported a novel ultrasonic technology for devulcanization of ground tire rubber and many other rubbers during extrusion. The devulcanized rubbers could be reprocessed, reshaped, and revulcanized in the same way as virgin rubber. In other applications, in our lab, high-energy ultrasonic oscillations were introduced to an extruder for the purposes of *in-situ* compatibilization of immiscible polymer blends.<sup>13,14</sup> The superposition of ultrasonic oscillations during extrusion could greatly decrease the apparent viscosity of polymer melts and eliminate unstable flow of polystyrene, linear low-density polyethylene (LLDPE), metallocene-catalyzed linear low-density polyethylene (mLLDPE), mLLDPE/LDPE blend, and Illite-filled high-density polyethylene composites.<sup>15-19</sup> In our previous work,<sup>15-19</sup> it was found that ultrasonic improvement of processibility of polymers was dependent on their molecular characteristics. In this article, superposition of ultrasonic oscillations in the extrusion of the plastomers was applied to decrease their viscosity and improve their processing rates. The plastomers used in this experiment are lower ethylene content grades of VERSIFY™ Plastomers and Elastomers. The effects of the ultrasonic oscillations on their processing behaviors including the die pressure, flow rate, apparent viscosity of the melts, appearance, and die swell of the extrudates were studied. The variation of molecular weight and molecular weight distribution of the plastomers as well as the effects of tensile and dynamic mechanical properties under ultrasound-assisted extrusion were also studied.

## EXPERIMENTAL

### Materials and equipment

Developmental plastomers (abbreviation: DP), DP-1 and DP-2, were supplied as pellets by Dow Chemical Co. (USA). Their properties are listed in Table I. The main equipment was a specially designed ultrasonic oscillation extrusion system developed in our

TABLE I  
DP Material Properties

Sample	Comonomer, wt %	Density (g/cm <sup>3</sup> )	Hardness, Shore A	Melt flow index (g/10 min)
DP-1	8	0.876	70	2
DP-2	5	0.888	75	5

laboratory, which included a lab-scale single screw extruder and a cylinder die connected to a probe of ultrasonic oscillations. The die included a special horn serving as a capillary (length/diameter = 8). The maximum power output and fixed frequency of the probe were 200 W and 20 kHz, respectively. The ultrasonic oscillations were in the direction parallel to the flow of the polymer melt. A pressure transducer and a thermocouple at the die entry were installed to continuously record the variation of die pressure and temperature during extrusion. Flow rate was monitored by the weight of the extrudate emerging from the die. A schematic diagram was described in Refs. <sup>15</sup> and <sup>16</sup>.

### Measurements and characterization

To study *in-situ* improvement of processing behavior of the plastomers in the presence of ultrasonic oscillations during extrusion, on-line measurements for die pressure and mass flow rate during ultrasonic extrusion processing were performed at melt temperatures, in the die, between 160 and 190°C in steps of 15°C, screw rotation speeds of 5, 15, 25, 35, and 45 rpm and superimposed ultrasonic intensities between 0 and 200 W in steps of 50 W. The shear rate,  $\dot{\gamma}$ , shear stress,  $\tau_w$ , and apparent viscosity,  $\eta_a$ , were calculated as follows:

$$\tau_w = \frac{PD}{4L} \quad (1)$$

$$\dot{\gamma} = \frac{32Q}{\pi\rho D^3} \quad (2)$$

$$\eta_a = \frac{\tau_w}{\dot{\gamma}} \quad (3)$$

where  $P$  is the die pressure,  $L$  is the length of capillary,  $D$  is the diameter of capillary,  $\rho$  is the density of plastomers and  $Q$  is the mass flow rate.

The surfaces of plastomers extrudates in the absence and presence of ultrasonic oscillations were observed using a JSM-5900LV SEM (Hitachi, Japan) to study the appearance and die swell of the extrudates. Before observation, the surfaces were coated with gold sputter.

The molecular weight and molecular weight distribution of plastomers extrudates in the absence and presence of ultrasonic oscillations were measured for the purposes of studying the potential ultrasonically-induced chain scission by PL-220 instrument (Polymer Laboratories Co.) at 150°C, using 1, 2, 4-trichlorobenzene as a solvent with polystyrene calibration standards.

To confirm the mechanism of apparent viscosity drop during ultrasonic extrusion processing, off-line rheological parameters of ultrasonic extrudates were measured at 160°C in a high-pressure capillary rheometer of RHEOGRAPH 2002 (Gottfert, Germany), with a capillary having a length to diameter ratio of 20 : 1.

The tensile tests were conducted at room temperature using dumbbell shaped samples obtained by compression molding on an Instron 4302 tension machine (Canton, MA), with a crosshead speed of 500 mm/min. The size of the narrow section of the samples was 25 × 4 × 1 mm<sup>3</sup>. The reported values of yield strength and strength at break are the averages of the five tests.

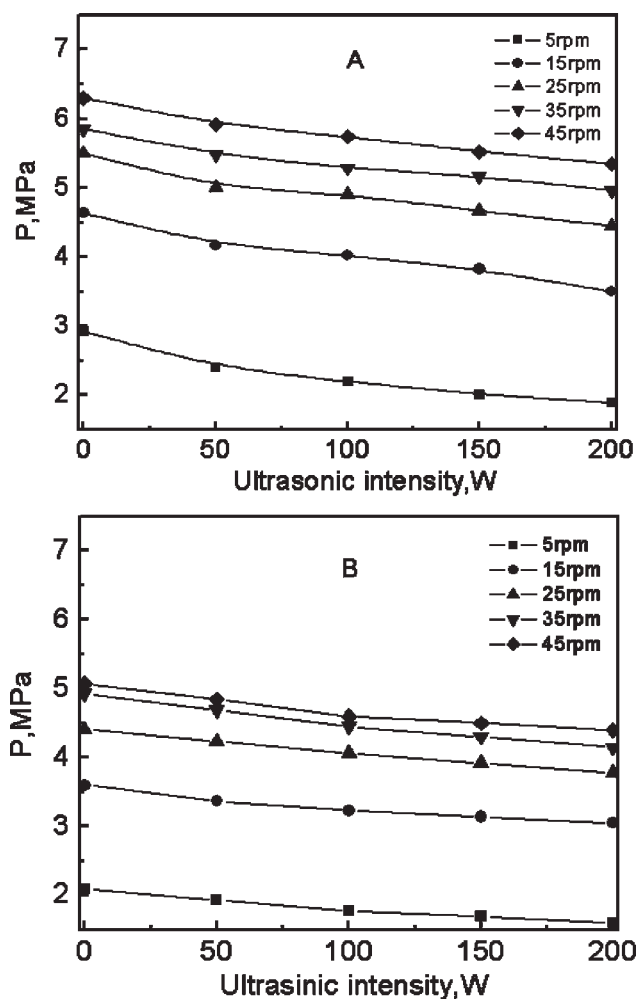
Dynamic mechanical analysis was performed with a DMA Q800 instrument (TA Co.) to measure tan δ and storage modulus over the temperature range -50 to 100°C at a constant frequency of 10 Hz and a heating rate of 5°C/min, using specimens of 17 × 7 × 1 mm<sup>3</sup> dimensions in the tensile mode.

## RESULTS AND DISCUSSION

### On-line rheological behavior

#### Die pressure

Die pressure changes of DP-1 and DP-2 with ultrasonic intensity were measured at melt temperatures in die of 160, 175, and 190°C, respectively. Figure 1 shows the dependence of die pressure  $P$  at different screw rotation speeds of the extrusion on ultrasonic intensity  $W$  at 160°C. The plots of  $P$ - $W$  of DP-1 and DP-2 are separated into two figures in the same scale to avoid congestion. The die pressures of DP-1 and DP-2 decrease not only with decreasing screw rotation speed but also with increasing ultrasonic intensity. The decreases of die pressure in the presence of ultrasonic oscillations at the same screw rotation speed indicates that DP-1 and DP-2 can be extruded under higher screw rotation speed by the aid of ultrasonic oscillations, as the die pressure remains constant. Moreover, the die pressure drop of DP-1 is higher than that of DP-2 at the same screw rotation speed and ultrasonic intensity. This implies that ultrasonic oscillations are more beneficial to decreasing die pressure of polymer with higher apparent viscos-

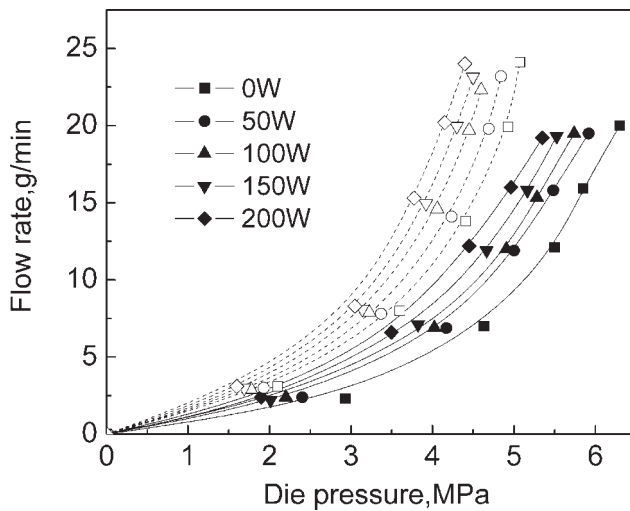


**Figure 1** Dependence of die pressure of plastomers on ultrasonic intensity at various screw rotation speeds at 160°C (A: DP-1; B: DP-2).

ity (lower melt index or higher molecular weight). This phenomenon will be confirmed later.

#### Productivity of extrusion

Mass flow rate versus die pressure for DP-1 and DP-2 is highly affected by ultrasonic intensities at above-mentioned melt temperatures in die. The data obtained at 160°C are shown in Figure 2. The mass flow rates of DP-1 and DP-2 increase with the rise of die pressure (screw rotation speed) at the same ultrasonic intensity, which is a typical route in industrial community to enhance productivity of extrusion. The curves of mass flow rate versus die pressure move to the left region in the graph with increasing ultrasonic intensity, indicating that mass flow rate of DP-1 and DP-2 increase in the presence of ultrasonic oscillations at the same die pressure. That is, ultrasonic oscillations can increase the pro-

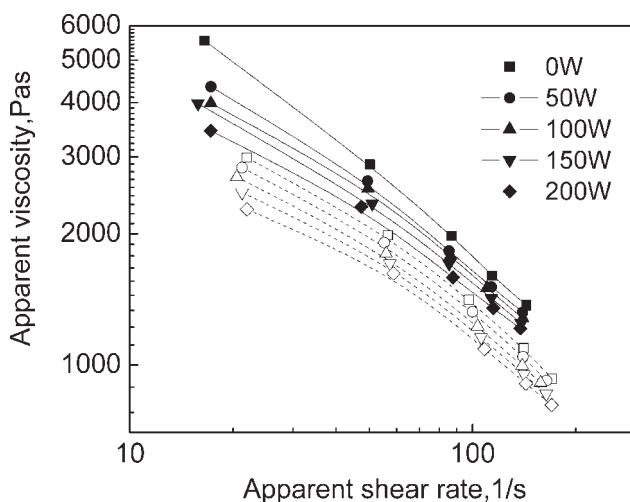


**Figure 2** Mass flow rate vs. die pressure for plastomers in presence of ultrasonic oscillations with various intensities at 160°C (real line: DP-1; dotted line: DP-2).

ductivity of extrusion. In other words, it also shows that ultrasonic oscillations can reduce die pressure at the same mass flow rate, which is more beneficial in improving appearance of extrudates (described in more detail later).

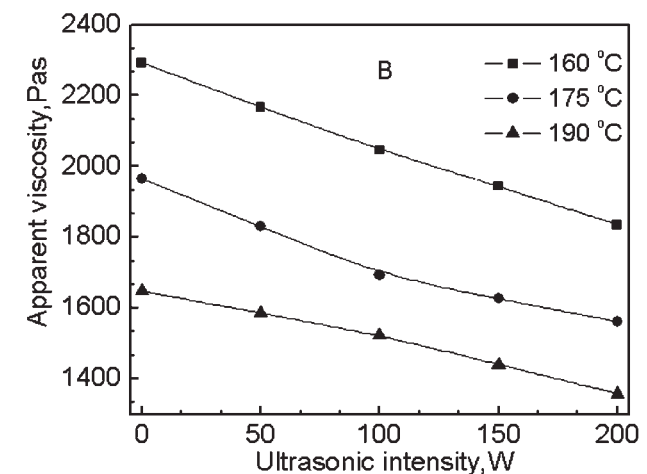
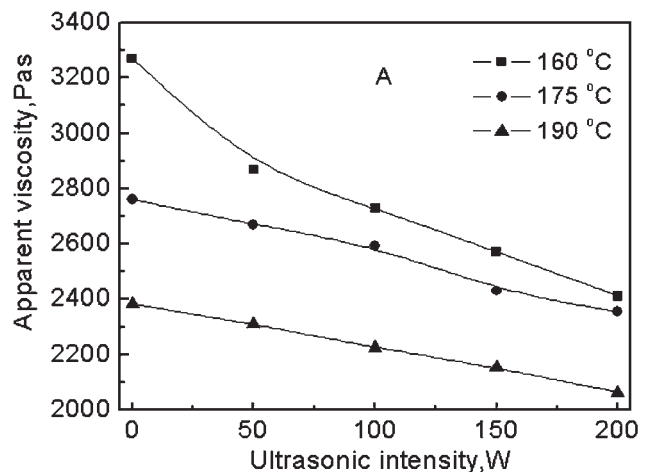
#### Melt apparent viscosity

Figure 3 shows viscosity curves of DP-1 and DP-2 during extrusion in the absence and presence of ultrasonic oscillations at 160°C, which are calculated according to eqs. (1)–(3). It shows that DP-1 and DP-2 melts still remain pseudoplastic. As expected, apparent viscosity decreases with increasing shear



**Figure 3** Apparent flow curves of plastomers in presence of ultrasonic oscillations at 160°C (real line: DP-1; dotted line: DP-2).

rate, regardless of the presence of ultrasonic oscillations. But apparent viscosity versus apparent shear rate has a nonlinear relation in experimental shear rate range of 15–175  $s^{-1}$ , indicating that the relationship between apparent viscosity of DP-1 and DP-2 and shear rate did not obey the simple power law equation. It was also observed that the apparent viscosity of DP-1 and DP-2 decreased as the ultrasonic intensity increased, especially in the lower shear rate range. At lower screw rotation speed, polymer melts have a longer residence time in the die. Therefore, DP-1 and DP-2 endured a longer ultrasonic irradiation in the lower shear rate range, causing the higher drop of apparent viscosity. In addition, it was also found that ultrasonic oscillations cause a greater reduction of apparent viscosity of DP-1, which has higher viscosity (lower melt index) compared with DP-2. This conclusion accords with above phenomenon of die pressure drop. Figure 4 shows that, at a constant shear rate of 40  $s^{-1}$ , the apparent viscosity of DP-1 and DP-2 decreased with increasing ultra-



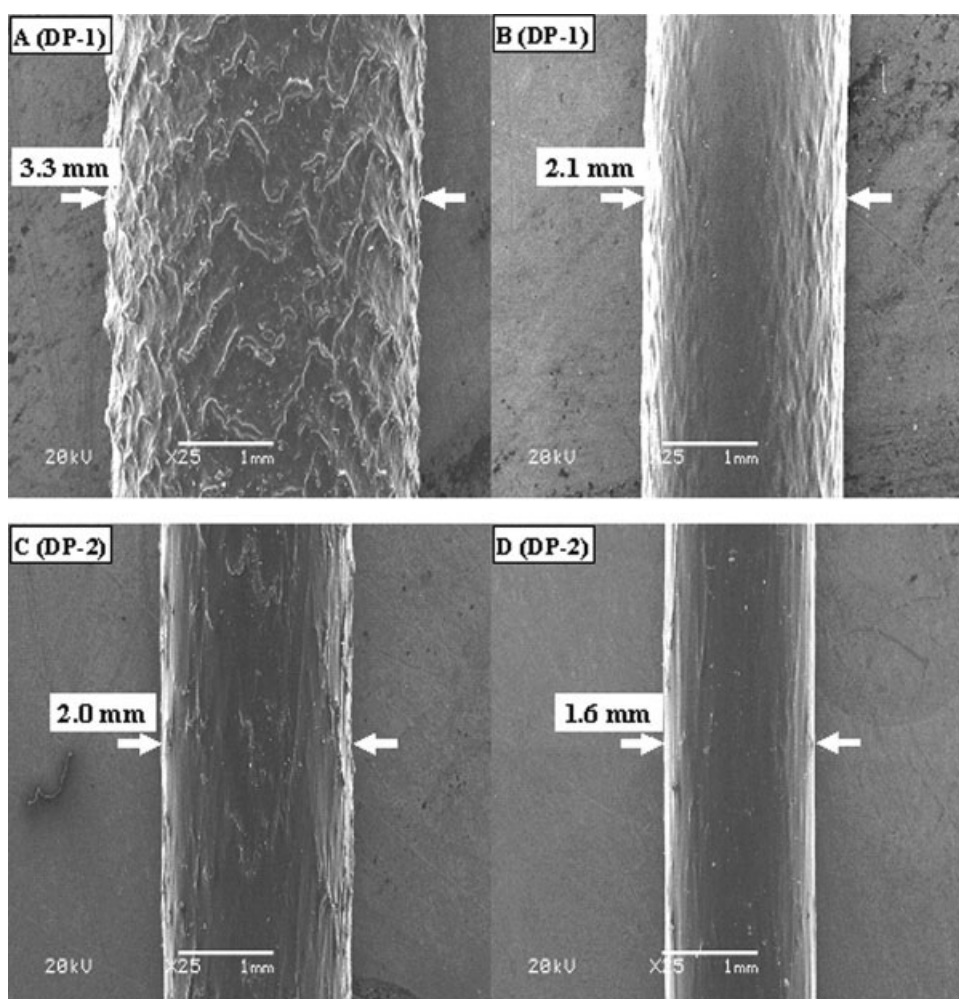
**Figure 4** Dependence of apparent viscosity of plastomers on ultrasonic intensity at various die temperature (A: DP-1; B: DP-2. apparent shear rate: 40  $s^{-1}$ ).

sonic intensity at different die temperatures (these are also plotted in two figures to avoid congestion). This result clearly indicates that the presence of ultrasonic oscillations can reduce the extrusion temperature of DP-1 and DP-2 if the apparent viscosity is maintained at the same level.

#### Appearance and die swell of extrudates

Melt fracture is one of significant problems in the processing operations of the plastomers such as extrusion, film blowing, etc, along with loss of specular gloss and more severe forms of "sharkskin". Considerable research to determine the origin of sharkskin melt fracture has been done over the past 40 years. Sharkskin appearance occurs at a critical shear stress and shear rate, mainly ascribed to both high local stress and breakdown of adhesion at the polymer/die wall interface.<sup>20</sup> Figure 5 reveals that DP-1 and DP-2 showed sharkskin appearance when

they were extruded at 160°C and 15 rpm [Fig. 5(A,C)]. However, the incidence of "sharkskin" was weakened or disappeared when DP-1 and DP-2 were extruded at 160°C and 15 rpm with superimposed 200 W ultrasonic oscillations [Fig. 5(B,D)]. In addition, it is well known that the deformation of polymer in die and the relaxation of the molecules at the exit of the die can lead to die swell of extrudates. It was also found from Figure 5 that DP-1 and DP-2 extrudates became thinner in the presence of ultrasonic oscillations at the same screw rotation speed (diameter of DP-1 and DP-2 extrudates was decreased by about 36 and 20%, respectively), suggesting that die swell of the plastomers gets weakened. These results indicate that ultrasonic oscillations, as a clean and efficient melt vibration technology, are a novel approach for weakening sharkskin melt fracture and die swell of the novel propylene-based plastomers, avoiding addition of processing aids such as fluoropolymers and organic silicone compounds.

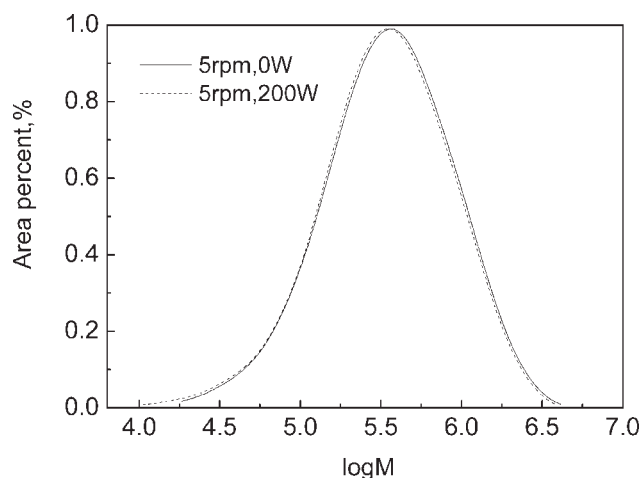


**Figure 5** Surface morphology of plastomers extrudates (A, C: without ultrasonic oscillations; B, D: ultrasonic intensity: 200 W).

### Mechanism for improvement of rheological behavior

Three research groups of Isayev (USA), Kim (Korea), and Guo (China) thought the occurrence of mechano-chemical chain scission during ultrasonic processing.<sup>21-24</sup> In our experiment, ultrasonically treated and untreated extrudates of DP-1 at 160°C and 5 rpm were chosen to measure the molecular weight and molecular weight distribution through high-temperature gel permeation chromatography. As shown in Figure 6 GPC curves, the introduction of ultrasonic oscillations leads to a slightly broadened curve with increased low molecular weight fraction as compared with the original sample. This indicates that slight chain scission of the elastomer can also occur during ultrasonic extrusion processing. The GPC data listed in Table II show that at very low screw rotation speed (5 rpm), 200 W ultrasonic oscillations would lead to decrease  $\overline{M}_n$  and  $\overline{M}_w$  of DP-1 by 8.1% and 3.3% respectively and increase its  $\overline{M}_w/\overline{M}_n$  by 5.6%. It is important to note that this chain scission due to ultrasound irradiation is likely to be a highly localized phenomenon. The extent of degradation is too small (3.3%  $\overline{M}_w$  change) to greatly affect the viscosity but significant enough to induce an *in-situ* grafting reaction.<sup>13</sup> The experimental results show that a permanent change of melt viscosity is less than 10% due to the change of molecular weight and its distribution (which will be described in detail later).

Is ultrasonically induced chain scission the main factor for apparent viscosities reduction during ultrasonic extrusion processing? To confirm this mechanism, off-line rheological behavior of DP-1 extrudates at 160°C and 5 rpm were measured through a high-pressure capillary rheometer. The experimental condition was the same as on-line extrusion, except



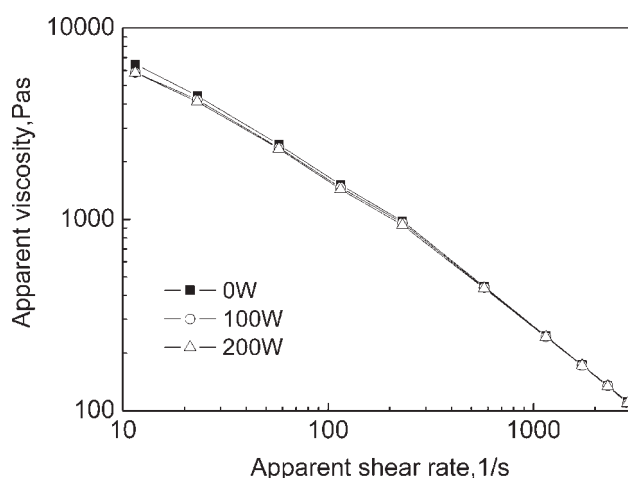
**Figure 6** GPC curves of DP-1 extrudates at 160°C with and without ultrasonic oscillations.

**TABLE II**  
GPC Results of DP-1 Extrudates at 160°C with and without Ultrasonic Oscillations

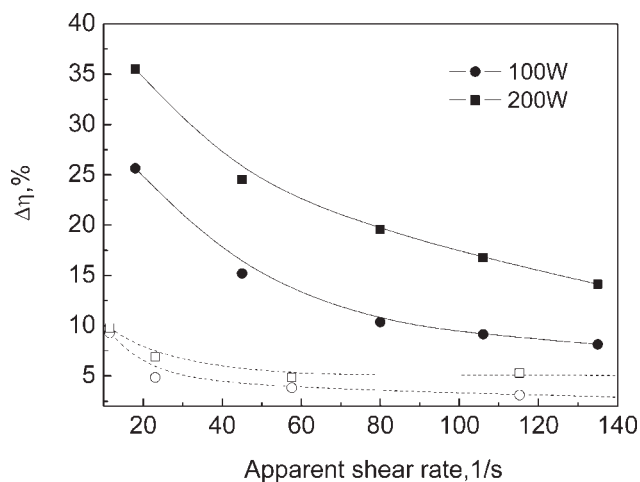
Extrusion conditions	$\overline{M}_n \times 10^{-4}$	$\overline{M}_w \times 10^{-4}$	$\overline{M}_w/\overline{M}_n$
5 rpm, 0 W	22.2	51.8	2.33
5 rpm, 200 W	20.4	50.1	2.46

for the length-diameter ratio of the capillary. It is clearly seen from Figure 7 that the off-line apparent viscosities of ultrasonic extrudate of DP-1 slightly decreased in the shear rate of 10–600  $s^{-1}$  owing to low molecular weights produced during ultrasonic processing. The relative apparent viscosity drop  $\Delta\eta$  can be used to characterize the extent of apparent viscosities reduction with introduction of ultrasonic oscillations, which is expressed as  $\Delta\eta = \frac{\eta_0 - \eta_u}{\eta_0} \times 100\%$ . There,  $\eta_0$  and  $\eta_u$  are the apparent viscosities in the absence and presence of ultrasonic oscillations, respectively. Figure 8 shows that off-line  $\Delta\eta$  of ultrasonically treated DP-1 in high-pressure capillary rheometer was less than 10%. However, on-line viscosity change,  $\Delta\eta$  of DP-1 in the presence of 200 W ultrasonic oscillations amounts to 35.5% at shear rate of 18  $s^{-1}$ , which is more than three times higher than the off-line viscosity change  $\Delta\eta$ . The results suggest that the apparent viscosities of the elastomers can be mostly recovered to initial viscosity value after ultrasonic oscillations are removed. Therefore, the reason for the significant drop for on-line measured apparent viscosities of the elastomers in the presence of ultrasonic oscillations is mainly due to a reversible physical cause and not due to chain scission.

On the basis of above-mentioned discussion and our previous work,<sup>15-18,25</sup> the improvement of rheological behavior of the polymer during ultrasonic

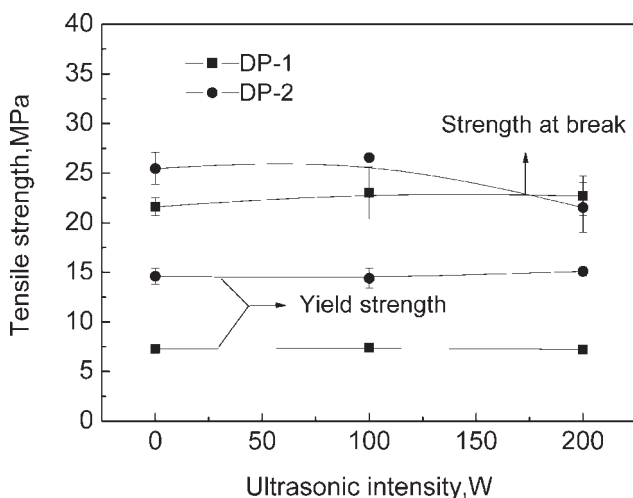


**Figure 7** Off-line apparent flow curves of DP-1 treated by ultrasonic oscillations at 160°C and 5 rpm.

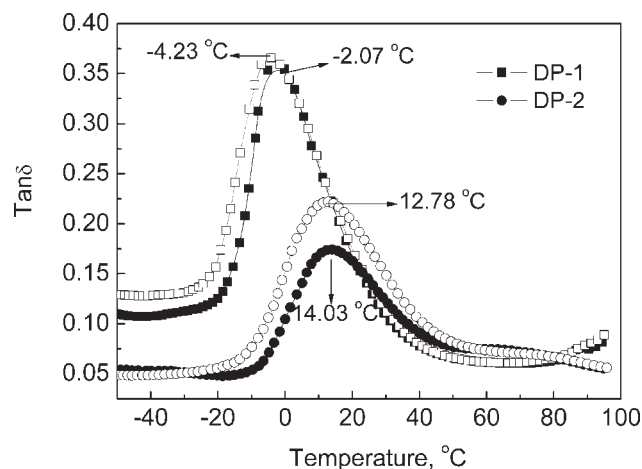


**Figure 8** Dependence of  $\Delta\eta$  of DP-1 on apparent shear rate at various ultrasonic intensities (real line: on-line; dotted line: off-line).

extrusion should be ascribed to the die pressure drop. Introduction of ultrasonic wave can drive the vibration of the extruder die,<sup>25</sup> which decreases the friction and surface stress at the polymer/die wall interface. Ultrasonic waves can be absorbed by the plastomer melts and partially translated into kinetic energy of molecular chain.<sup>23</sup> Hence ultrasonic waves can loosen the entanglements of polymer chains and enhance mobility of polymer chains, and even slightly induce molecular chain scission. The higher the viscosity of the plastomer, the more evident the effect of ultrasonic wave on the plastomer melts. As a result, it causes a reduction in melt viscosity and elasticity in the level of molecular microstructure. The apparent viscosities of the plastomers are ultimately decreased in the presence of ultrasonic oscil-



**Figure 9** Dependence of tensile strength of plastomers on ultrasonic intensity.



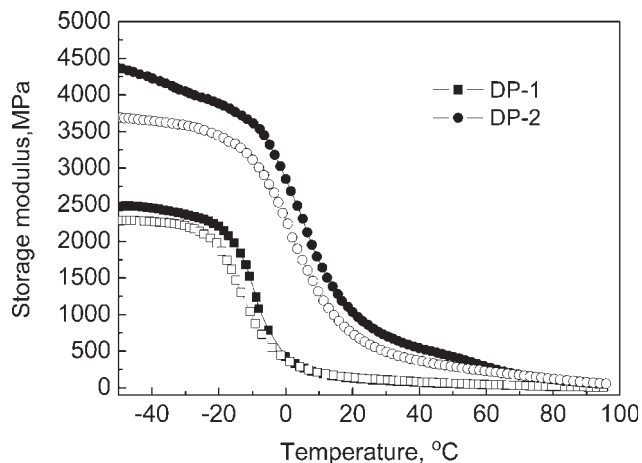
**Figure 10** DMA traces of loss tangent ( $\tan \delta$ ) vs. temperature for plastomers at 160°C and 5 rpm (real line: 0 W; dotted line: 200 W).

lations, along with weakening of the melt fracture and die swell of extrudates.

### Mechanical properties

The effect of ultrasonic oscillations on the yield strength and strength at break of the plastomers is showed in Figure 9. The yield strength of DP-1 and DP-2 and the strength at break of DP-1 show no distinct change with introduction of ultrasonic oscillations at 160°C and 5 rpm.

DMA traces of loss tangent ( $\tan \delta$ ) versus temperature for the plastomers are shown in Figure 10.  $\tan \delta$  of the plastomers increases with the introduction of ultrasonic oscillations, indicating the rise of internal friction of polymer chains. All samples present a dispersion peak in the temperature range studied, which is attributed to the micro-Brownian motion of



**Figure 11** DMA traces of storage modulus vs. temperature for plastomers at 160°C and 5 rpm (real line: 0 W; dotted line: 200 W).

the plastomers in the amorphous phase. The peak temperature is the glass-rubber transition temperature ( $T_g$ ), whose value is also marked in Figure 10. It is well known that  $T_g$  of the polymer is dependent on the mobility of chain segment. As shown in Figure 10, the addition of ultrasonic oscillations slightly decreased the  $T_g$  of the plastomers, which could be due to either slightly decreased molecular weight or disentangled polymer chains induced by ultrasonic oscillations, which in turn facilitates the motion of the entire chain segment of the plastomers. At the same time, the variation in storage modulus as function of temperature for the plastomers in the absence and presence of ultrasonic oscillations are shown in Figure 11. Evidently, the storage modulus of DP-1 and DP-2 in the presence of ultrasonic oscillations is also slightly decreased owing to a slight shift in molecular weights, especially in the glass state and glass-rubber transition region of the polymer. The same result has been confirmed in ultrasonically treated isotactic PP homopolymer.<sup>26</sup>

### CONCLUSIONS

Rheological behaviors of the novel propylene-based plastomers during ultrasound-assisted extrusion as well as the tensile and dynamic mechanical properties of their extrudates are studied. The die pressure and apparent viscosity of the plastomers in extrusion are greatly decreased in the presence of ultrasonic oscillations, and decrease as the ultrasonic intensity increases. The productivity of the plastomers extrusion increases with increasing ultrasonic intensity under the same die pressure. Ultrasonic oscillations can decrease the extrusion temperature of the plastomers, die swell, and tendency of melt fracture phenomenon. The processing behavior and appearance of the plastomers are greatly improved. Simultaneously, chain scission of the plastomers would occur

in ultrasonic extrusion processing. As a result, the glass-rubber transition temperature and storage modulus of the ultrasonically treated plastomers are slightly reduced due to a slight shift in molecular weight and its distribution.

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