Rotating Die Technique for Sharkskin Minimization in Highly Viscous Wood/PP Composite Melt in an Extrusion Die

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ABSTRACT: This study used a newly developed rotating die system for purposes of reducing entrance pressure drop and sharkskin fracture for molten polypropylene (PP) and wood/polypropylene (WPP) composites in a single-screw extruder. The sharkskin fracture characteristics of the PP and WPP composite surfaces were examined quantitatively via roughness profiles and relaxation time evaluations, and qualitatively through scanning electron microscopy under the effects of wood content, shear rate, die temperature, and die rotation speed. The experimental results suggested that the entrance pressure drop of PP increased with increasing wood content and shear rate. The die entrance pressure drop for WPP composite melt with 30 wt % wood content could be minimized by 20–50% by using a die rotation speed of 70 rpm. The roughness level (sharkskin) and relaxation time were found to increase with increasing wood content, but could be minimized by rotating the die—the die rotating effect being more meaningful for WPP when compared with neat PP extrudate. The rotating die system was found to be an effective technique for minimizing the extrusion load and fracture level of extrudate skins for high-viscosity materials such as the WPP composites used in this work. © 2012 Wiley Periodicals, Inc. J Appl Polym Sci 125: 2312–2321, 2012

Key words: polymer extrusion; thermoplastics; composites; melt

INTRODUCTION

Flow instabilities often occur during processing when polymer melts are extruded through a confined channel at a flow rate exceeding the critical shear stress. Three main instabilities of the flow are usually observed as a function of increasing throughput: sharkskin, stick-slip, and gross melt fracture. The sharkskin instability is the first instability to occur, and this affects not only the productivity during processing but also the quality of the polymer products. A number of studies have attempted to measure and control the sharkskin effect in polymers.[1–4] Burghelea et al.[1] investigated the sharkskin instability of low-density polyethylene and linear low-density polyethylene (LLDPE) through a slit die using laser Doppler velocimetry to monitor the melt flow rate, and found that the sharkskin occurred because the tensile stress around the die exit had exceeded the melt strength. This effect was obvious with LLDPE. Carreras et al.[2] examined the sharkskin of polystyrene-*block*-poly(ethylene-*co*-butylene)-*block*-polystyrene during the extrusion process using a video camera for visual observation. They stated that primary cracks initiated and propagated at the die exit at low shear rate, and more severe cracks developed further with increasing shear rate.[3,4] Allal et al.[3] found that the development of surface sharkskin for linear polymers was perpendicular to the flow direction, and that the period and amplitude of the sharkskin progressively increased with increasing flow rates. The surface instabilities during extrusion could also occur as a result of high extensional stresses.[4]

Although the importance of the sharkskin effect in polymer processing has long been realized, only a few methods have been available for moderating the sharkskin instability. The existing methods include using processing aids, redesigning die geometries, and adjusting die and processing temperatures.[5–20] The processing aids that have been widely used included fluoropolymer-based polymer processing aids,[5,6] maleated polyethylene,[7] and block copolymers

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of diisocyanates and polyols.[8] The selection of processing aids is dependent on the molecular structure of the polymer melt used; their functions are to delay shear stress development at the die exit where the melt emerges. Several methods have been proposed to improve die design to effectively reduce the surface roughness of the polymer extrudates for polyethylenes: increasing the die diameter, reducing the die entry angle, and using a divergent taper.[9–11] Increasing die temperature can also moderate the sharkskin effect[12] by reducing the relaxation time of the melt; this leads to a reduction in elastic recovery effects, such as stress development at the die exit, extrudate swelling level and, thus, sharkskin instability.[13–15]

The sharkskin effect is more pronounced and occurs more easily in polymer composite systems because of the high shearing stresses developed during processing.[16–18] Wood/plastic composites (WPCs) have recently attracted the attention of a number of scientists and engineers because of their advantages in cost savings, good mechanical properties,[19] better dimensional stability, low density, and environmental friendliness when compared with synthetic fiber/plastic composites. This also included with the WPC products with use of recycled thermoplastics.[20] Their applications include window and door profiles, decking, automobile paneling, panel inserts, flowerpots, and so on. However, a potential drawback is that the addition of wood fibers to polymeric materials causes a considerable increase in composite melt viscosity, creating difficulties during processing due to the high pressure drop buildup.[16,18] This can eventually lead to flow instabilities, shape distortions, and low qualities of WPC articles although proper design of dies may be selected and used.[21,22] Work by Hristov and Vlachopoulos[17] clearly suggested that the surface tearing and extrudate distortions of wood/metallocene-catalyzed polyethylene (mPE) composites increased with increasing wood content (from 30 to 50 wt %). Therefore, it would be essential to reduce or moderate the shearing stress development that may occur during extrusion of WPC materials.

Based on the existing literature,[1–17] few studies have focused on improving the flow instabilities of WPC materials. Hence, this became an area of interest and inspired this work. More recent work by Intawong et al.[23] was the original design and manufacture of a rotating die system for reduction of extrusion forces and entrance pressure drop for molten polypropylene and wood/polypropylene (WPP) composites under isothermal and nonisothermal extrusion processes. The experimental results have suggested that the rotation of the die could considerably moderate the extrusion loads and entrance pressure drop, especially with high wood content and at high shear rate. In the case of this work, it was hoped that the newly developed rotating die system could minimize the surface fracturing, or sharkskin effect, of highly viscous WPP composites during processing by single-screw extrusion at shear rates that exceeded their critical shear stresses. The developed die-rotating apparatus was connected to the end of a single-screw extruder that was used for production of molten WPP composites. In this work, the fracture characteristics of WPP composite surfaces were not only visualized qualitatively, via scanning electron microscopy (SEM) but also they were determined quantitatively, in terms of roughness profiles—using a surface roughness tester (SRT) that measured the period and amplitude of the fracture peaks on WPP surfaces—and relaxation time evaluation. The effects of wood content, shear rate, die temperature, and die rotation speed on the flow properties and sharkskin for molten WPP composites were the primary focuses of interest. These findings should result in great practical benefits, leading to increased productivity of the extrusion process for highly viscous wood composites without experiencing surface ruptures on the WPC products.

EXPERIMENTAL

Raw materials and preparation of wood/PP composites

Polypropylene (PP), grade 1100NK, having a melt flow rate (MFR) of 9.8 g/10 min was supplied in granule form by IRPC Public Co., Ltd. (Bangkok, Thailand). Wood particles obtained from carpentry and woodworking processes were supplied by V.P. Wood Co., Ltd. (Bangkok) and had an average size of 250 μm. The wood particles were dried to a constant weight to avoid any bubbles and voids during processing. The maximum moisture content allowed during processing was less than 5%; this being achieved using an oven at 80°C for 24 h.[24]

The dried wood particles were first dry-blended with PP granules using a high-speed mixer (LMXS; Labtech Engineering, Samut Prakan, Thailand) before being melt-blended in a twin-screw extruder (HAAKE PolyLab Rheomex CTW100p; Thermo Electron, Karlsruhe, Germany) to obtain wood/PP (WPP) composites. The wood content used varied from 0 to 30 wt % of the PP. The blending temperature profiles on the extruder were 170, 180, 190, and 200°C from hopper to die zones, and the screw rotating speed was 50 rpm. A three-strand die, each strand having a diameter of 3 mm, coupled with a pelletizing unit was used to produce the WPP composite pellets. These were kept in an oven for 24 h at 80°C to avoid moisture regain before further processing.

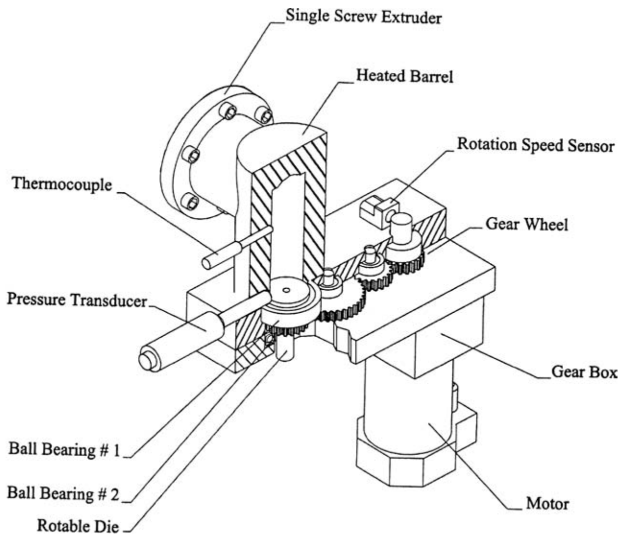

Figure 1 An experimental arrangement for flow property and sharkskin measurements for PP and WPP composite melts in a single-screw extruder.

Experimental apparatus and procedures

The rotating die system that was first developed by our previous work[23] was connected to the end of a single-screw extruder (HAAKE PolyDrive; Thermo Electron), which was used for extruding the WPP composites. The arrangement of the experimental rig is shown in Figure 1. The detailed descriptions for each component of the experimental rig can be found in our previous work.[23] The exact length-to-diameter (L/D) ratio of the barrel was 450/19 mm/mm, and the temperature profiles on the extruder from hopper to die zones were 170, 180, 190, and 200°C. Screw rotation speeds varied from 10 to 30 rpm to generate shear rates from 2.0 to 7.8 s^{-1}, respectively. A rotatable circular die, 65 mm in length and 5 mm in diameter, was located at the bottom of the heated barrel. The rotating speed of the die could be adjusted between 0 and 70 rpm and was controlled using a rotation speed sensor installed on the driving axis of the motor. The temperature was controlled using a DD6 temperature controller (Changchai Motor, Bangkok, Thailand); the die temperature used was 200°C throughout this work. The entrance pressure drop was measured using a pressure transducer (PT460E-2CB-6; Dynisco, Franklin, MA) situated at the base of the barrel just above the die face.

Flow properties and sharkskin measurements

The flow properties of molten WPP composites affected by the use of the rotating die system were considered in terms of changes in pressure drop values at the die entrance. The calculation of wall shear rates used in this work were the volumetric flow rate for any given screw rotation speed of the single-screw extruder. The sharkskin instabilities of WPP extrudate surfaces were assessed quantitatively using a SRT (Form Talysurf Series II; Taylor Hobson, Leicester, UK). The roughness of the WPP samples was measured using a small stylus tip, which moved automatically in a transverse direction along the extrudate surface. The stylus tip used was a standard diamond conical stylus with a cone angle of 90° and a tip radius of 2 μm. The roughness results were reported in terms of period (W_s) and amplitude (W_z) of the fracture peaks on the extrudate surfaces before being calculated into the relaxation time (θ) of the composite material, which can be expressed by eq. (1).[3] The reported experimental data were averaged from at least five independent determinations.

$$\theta = \frac{W_Z}{\bar{\nu}} \tag{1}$$

where W_Z is the amplitude of the surface roughness, and $\bar{\nu}$ is the average axial velocity of the extrudate at the die exit.

The sharkskin surfaces of WPP composites were also visualized using a scanning electron microscope (JSM-6301F; JEOL, Tokyo, Japan) at 10 kV of accelerating voltage. WPP composite fracture surfaces were obtained for examination after 2 min immersion in liquid nitrogen. The details of the experimental procedure and sample preparation for SEM investigations can be obtained elsewhere.[24]

RESULTS AND DISCUSSION

Figure 2 shows the real-time pressure drop profiles as a function of extrusion time and die rotation speed for neat PP [Fig. 2(a)] and WPP [Fig. 2(b–d)] composite melts at three different shear rates and a die temperature of 200°C. It should be noted that the shear rates used in this work were relatively low due to the large diameter of the die used. Therefore, the recorded entrance pressure drop was relatively low. The entrance pressure drop was measured while the die rotation speed was increased in a stepwise manner. Without the use of a rotating die system (at extrusion times of 0–120 s), the entrance pressure drop of PP increased with increasing wood content. This was because the addition of wood particles to PP resulted in an increase in bulk viscosity of the PP melt, which could generate more shear stress during the flow.[21] This behavior corresponded to the work by Muksing et al.,[13] who suggested that the apparent melt viscosity of polypropylene increased up to 70% by increasing the wood content. However, when rotating the die from 0–70 rpm (at extrusion times of 120–500 s), the entrance pressure drop appeared to change, the effect being most

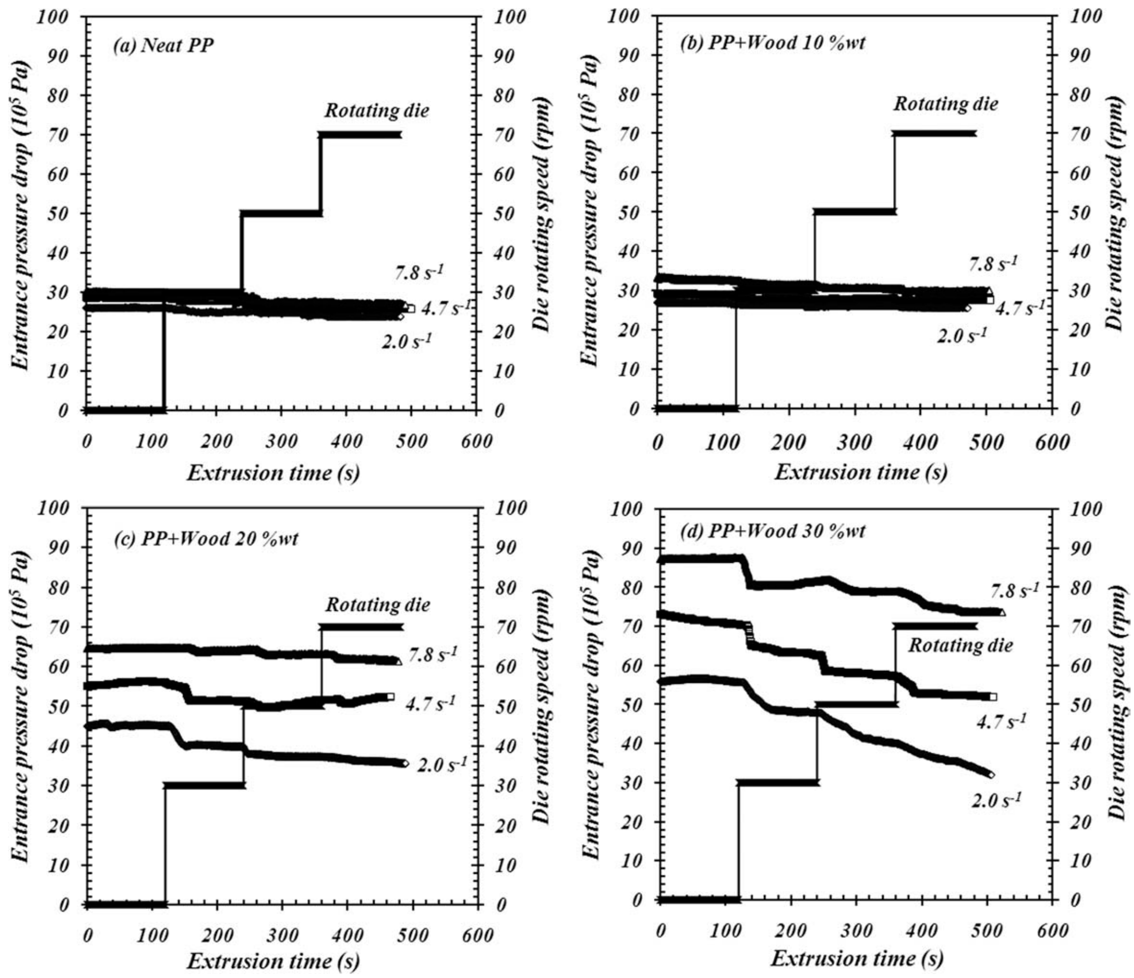

Figure 2 Entrance pressure drop vs. time for PP and WPP composite melts for different wood contents at three different shear rates and a die temperature of 200°C: (a) neat PP, (b) WPP with 10 wt % wood content, (c) WPP with 20 wt % wood content, and (d) WPP with 30 wt % wood content.

pronounced at a wood content of 30 wt %. The die rotation speed and apparent wall shear rate did not affect the entrance pressure drop for neat PP [Fig. 2(a)] and WPP composite with 10 wt % wood content [Fig. 2(b)]. For WPP with 20 wt % [Fig. 2(c)], the effect of shear rate on the entrance pressure drop was more pronounced than the effect of die rotation speed. For WPP composite with 30 wt % wood content [Fig. 2(d)], the entrance pressure drop progressively decreased, from 55×10^5 to 30×10^5 Pa (about a 50% reduction) for low shear rate (2.0 s^{-1}), and from 88×10^5 to 75×10^5 Pa (about a 20% reduction) for high shear rate (7.8 s^{-1}), by increasing the die rotation speed from 0 to 70 rpm. The reduction of entrance pressure drop by the action of die rotation speed was associated with two concurrent effects: torsional shear strain and additional shear heating (details of the experimental evidence were given in our previous work).[23] The former can be visualized as a twisting of the flow layers (helical or spiral flows), which is believed to produce torsional shear strain within the die.[25] This could lead to a circumferential slippage between the melt and die wall during the flow in the rotating die. The latter effect would be additional shear heating that had occurred to reduce the melt viscosity.[26] The pressure drop reduction could indirectly indicate higher extrusion outputs or productivities in polymer extrusion. It can, therefore, be concluded that the rotating die system was suitable and effective for moderating the pressure drop at the die entrance, especially in the case of materials with high bulk viscosity.

Figures 3 and 4 show exemplary roughness profiles for neat PP and WPP extrudates, respectively, for a 5 mm length of the extrudate emerging from the die exit. The PP and WPP (30 wt % wood content) extrudates were produced using different die rotation speeds of 0, 30, 50, and 70 rpm at a shear rate of 7.8 s^{-1} and a die temperature of 200°C. It can be observed that the shape patterns of the roughness profiles for neat PP extrudates with different die rotation speeds were very similar, whereas those for WPP extrudates differed with varying die rotation speeds. For the WPP extrudates, the periods,

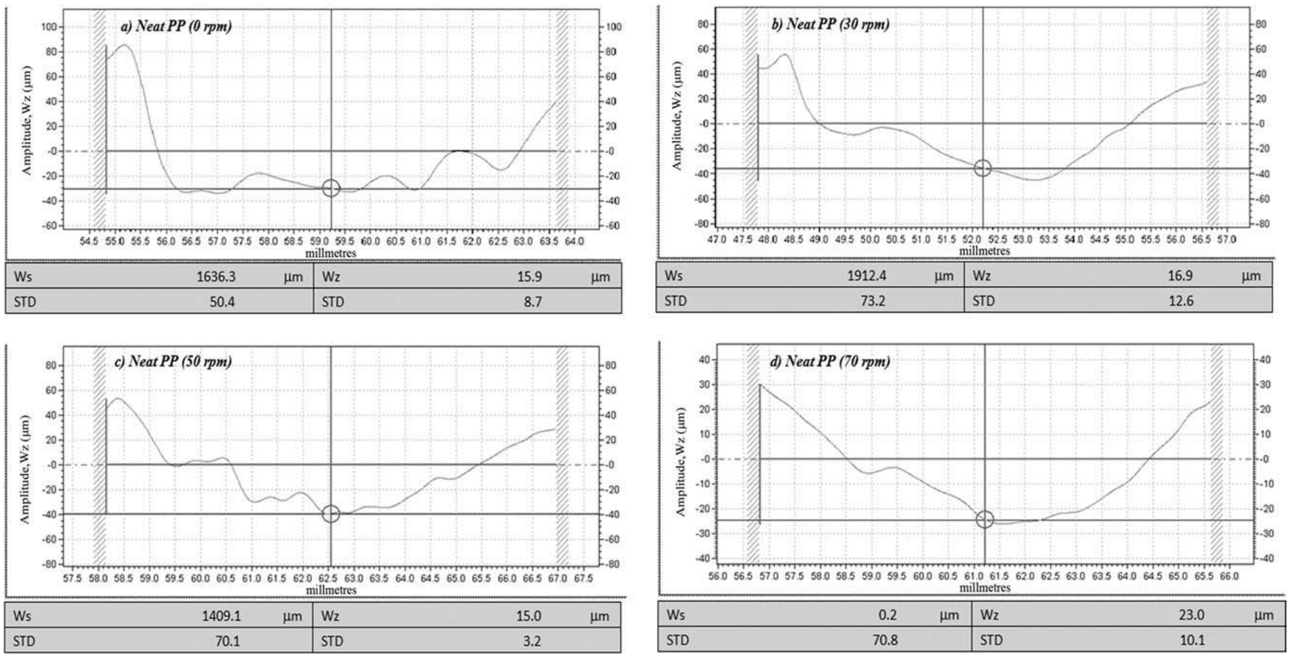

Figure 3 Roughness profiles for neat PP extrudates produced by different die rotation speeds at 7.8 s^{-1} shear rate and a die temperature of 200°C.

number of peaks, and amplitudes of the roughness peaks appeared to be minimized as the die rotation speed increased. Generally, it has been understood that the greater the amplitude and period of the roughness peaks, the higher the skin fracture of material.[27] If this is the case, the minimizations of the amplitude and period for WPP extrudates (Fig. 4) clearly indicate that the severity of the sharkskin was reduced by rotating the die. To be more understandable, the experimental results were given in terms of amplitude (W_z) and period (W_s) values for neat PP and WPP with various wood contents and wall shear rates, and a wide range of die rotating speeds (Figs. 5 and 6). It can clearly be seen in Figure 5 that the WPP extrudates exhibited much greater amplitudes than the neat PP. This indicates higher levels of sharkskin effect on the WPP composite extrudates, as one would expect, because the

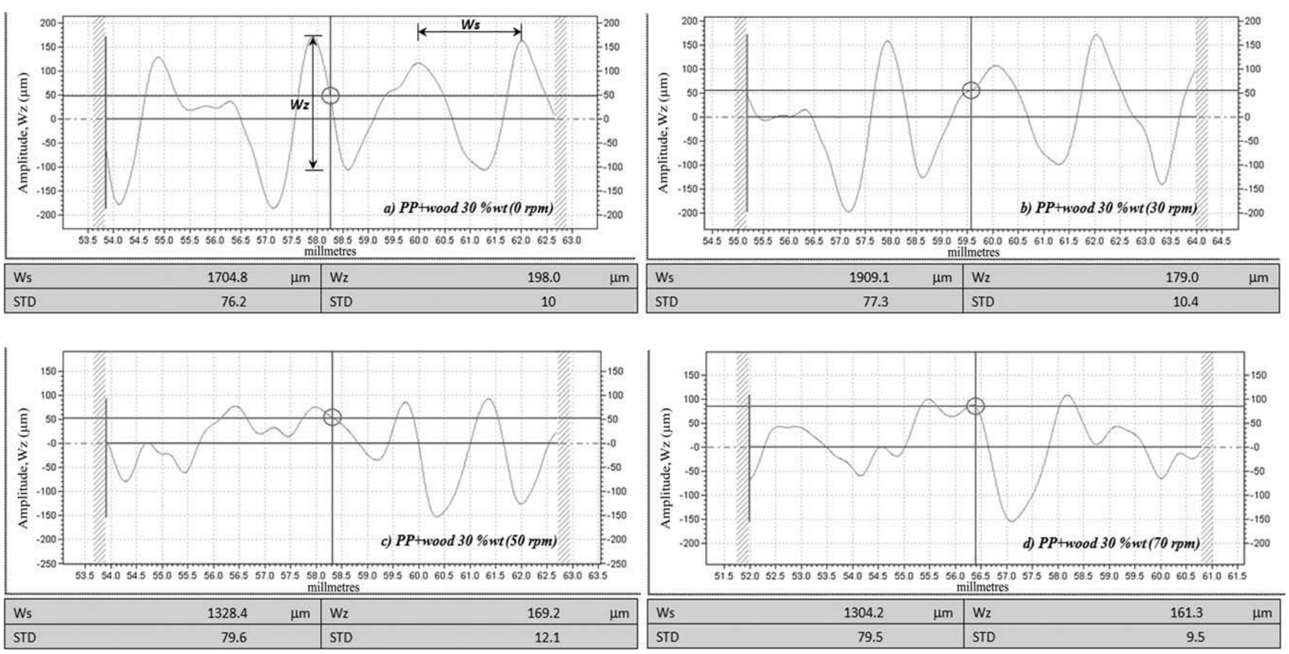

Figure 4 Roughness profiles for WPP composite extrudates with 30 wt % wood content for different die rotation speeds at 7.8 s^{-1} shear rate and a die temperature of 200°C.

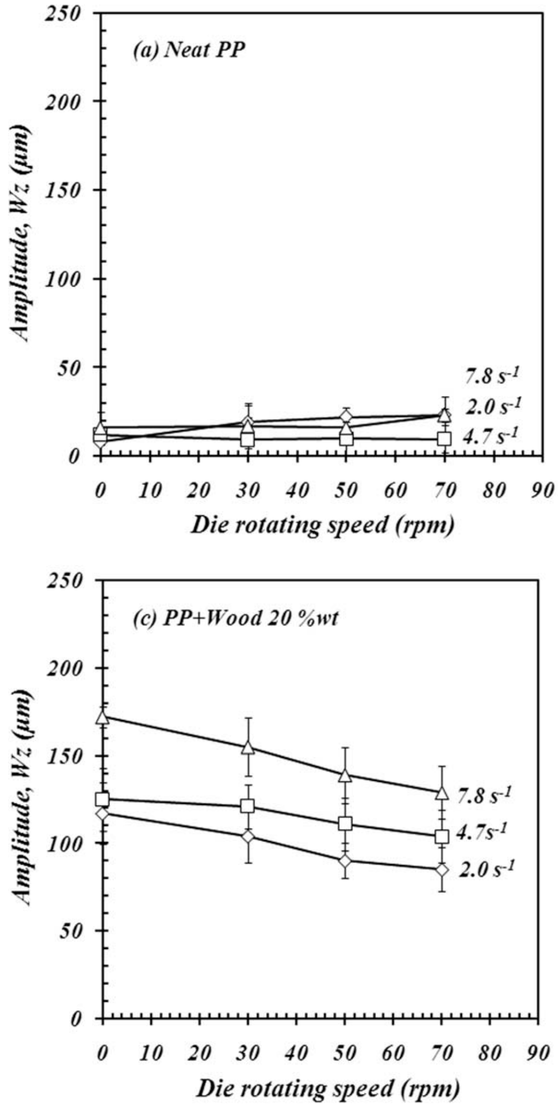

Figure 5 Effects of die rotation speed and wall shear rate on amplitude level of surface roughness for neat PP and WPP extrudates at a die temperature of 200°C.

WPP possessed higher viscosity, as already indicated by the higher entrance pressure drop in Figure 2. The physical evidence for the increased sharkskin levels with increasing wood content can be seen in SEM micrographs (Fig. 6), showing axial sectional views of the PP extrudates with various wood contents using a stationary die (not rotating the die). The surface fractures of the WPP were obvious at the wood loading of 30 wt % together with some voids within the composites. The presence of voids within the wood/polymer composites is usually expected if the composites were processed by extrusion or die based processing technique.[18]

Figure 5 also illustrates that the amplitudes for neat PP and WPP were affected by the shear rates used, the effect being more pronounced as the wood content was increased [Fig. 5(b–d)]—the higher the wall shear rate, the greater the amplitude. This is as one would expect, since the higher shearing stresses would be more likely to produce extrudates with skin fractures, especially for the highly viscous wood/PP composite extrudates in this work. It is interesting to note that the effect of die rotation speed on reducing the amplitude was obviously more pronounced for the WPP composite extrudates that exhibited the highest amplitude or sharkskin level; this was the case for the WPP extrudate with the highest wood content (30 wt %) and the highest wall shear rate (7.8 s^{-1}) [Fig. 5(d)]. The explanation for this, as mentioned earlier, is given in terms of the torsional shear strain and additional shear heating effects. The physical evidence for reducing the sharkskin levels by increasing the die rotation speed can be seen in SEM micrographs (Fig. 7) showing axial sectional views of the WPP composite extrudate (30 wt %wood content) at die rotation speeds

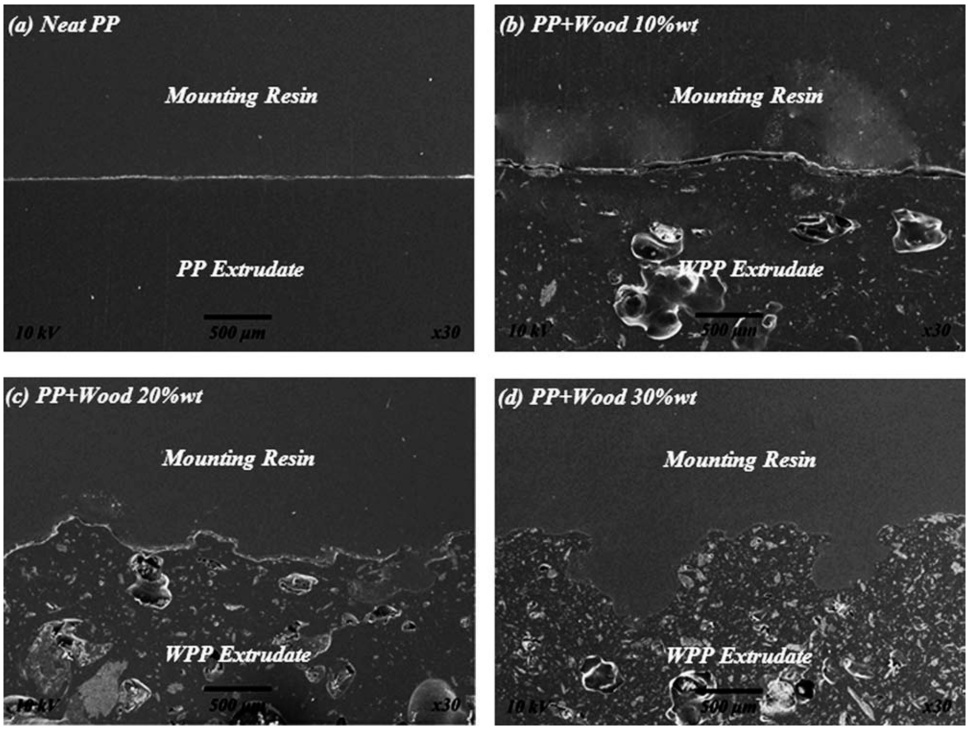

Figure 6 SEM micrographs of axial roughness for PP as a function of wood content, without the use of a rotating die system.

from 0 to 70 rpm. It can clearly be observed that the roughness or sharkskin lines became much minimized for extrudates produced using the rotating die system. However, the roughness of the WPP extrudate with the highest die rotating speed (70 rpm) was still greater than that of the neat PP extrudate. The experimental results shown in Figures 2, 5–7 suggest the practical benefits of reducing the

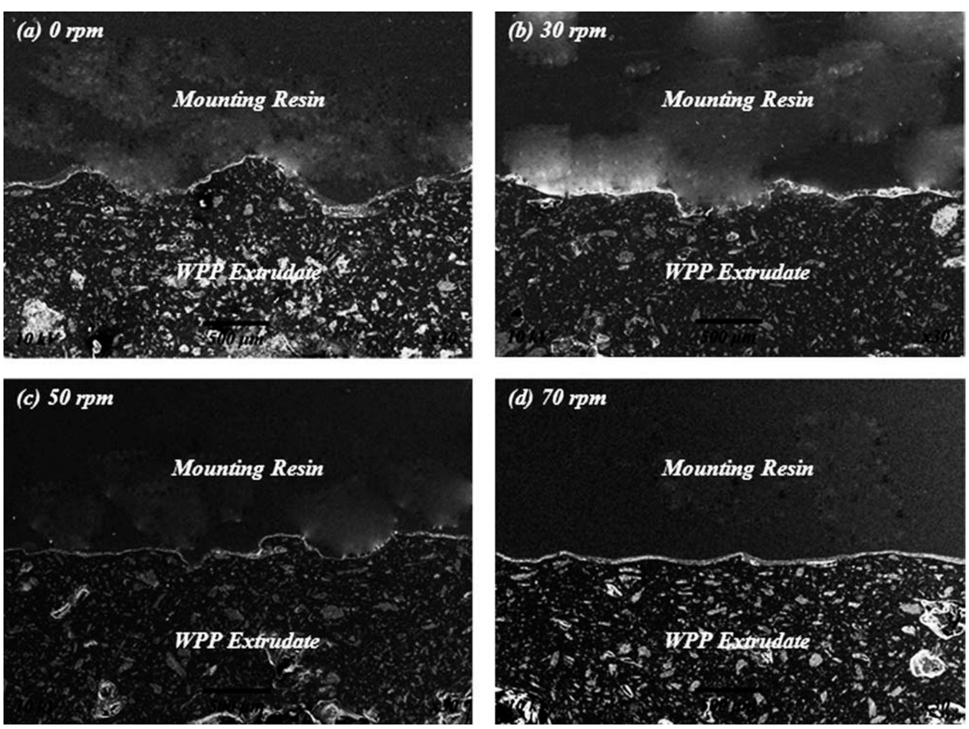

Figure 7 SEM micrographs of axial roughness for WPP with 30 wt % wood content at various die rotation speeds.

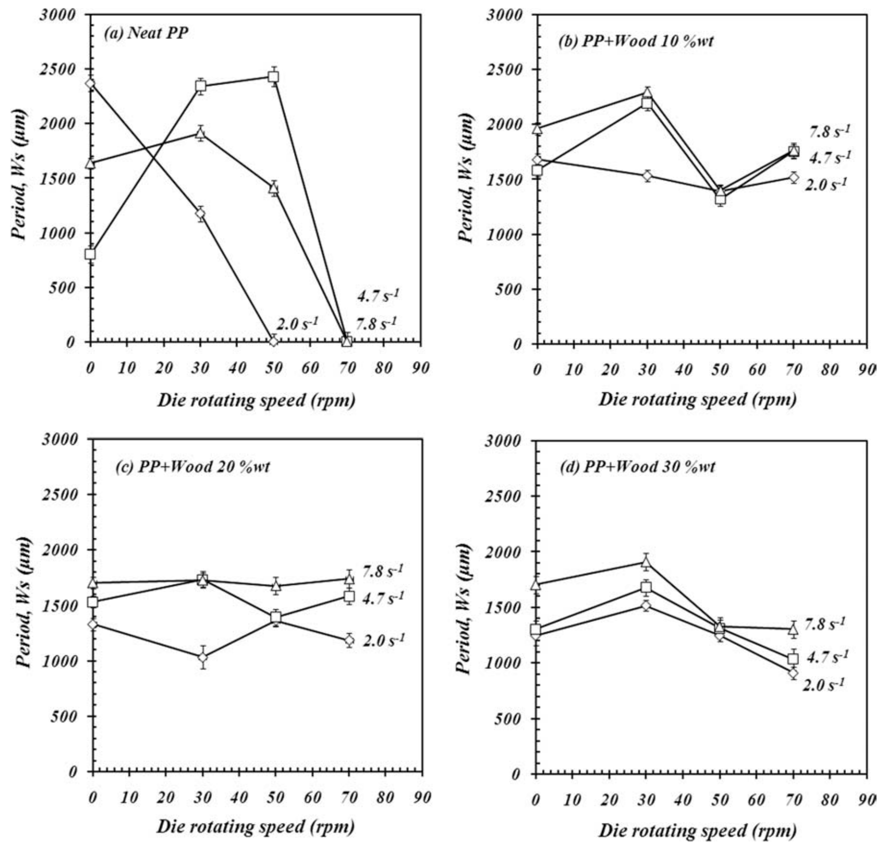

Figure 8 Effect of die rotation speed and wall shear rate on period (W_s) of surface roughness for neat PP and WPP extrudates at a die temperature of 200°C.

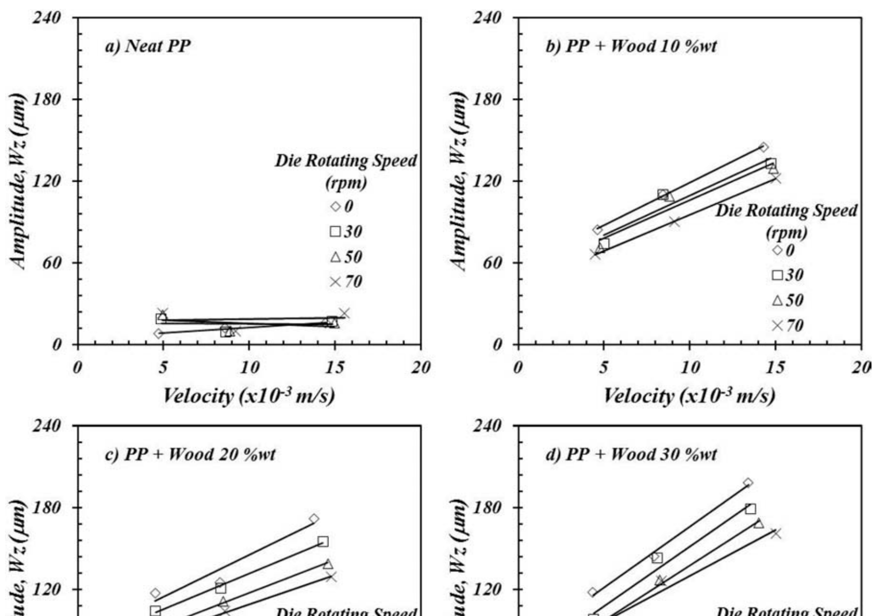

Figure 9 Plots of Wz-$\bar{v}$ for PP and WPP composite melts at various die rotation speeds.

TABLE I
Calculated Relaxation Time (θ) for Molten PP with Different Wood Contents and Die Rotation Speeds

Die rotating speed (rpm)	Relaxation time ($\times 10^{-3}$ s) Wood flour contents (wt %)			
	0	10	20	30
0	0.8	6.3	6.1	8.9
30	0.3	5.8	5.3	8.7
50	0.2	5.6	4.8	8.2
70	0.2	5.3	4.3	6.7

sharkskin level of high-viscosity materials by using this rotating die system, i.e., that the extrusion output or productivity could be increased.

Figure 8 shows the effects of wall shear rate and die rotation speed on the period of roughness peaks for neat PP and WPP composites with various wood contents. It can be seen that there were no definite trends from which to determine any further details or draw any further conclusions on the sharkskin character. Only more periods of fluctuation were observed for neat PP, but these were greatly reduced by increasing the die rotation speed.

Allal et al.[3] suggested that a plot of amplitude against average axial velocity ($\bar{v}$) of the melt flow at the die exit could produce a quantitative measurement of the elastic parameters in a molten polymer, i.e., the relaxation time (θ), as expressed in eq. (1). Figure 9 shows examples of plots of amplitude against melt velocity for neat PP and WPP composite extrudates at various die rotation speeds using a wall shear rate of 7.8 s^{-1} at a die temperature of 200°C. Similar to the results shown in Figure 5, the amplitude appeared to decrease with increasing die rotation speed but increased with increasing melt velocity at the die exit. Table I shows the results of relaxation times for PP with various wood contents at varying die rotation speeds. It can be seen that the relaxation time of PP was very low, and that of WPP composite melts ranged from 4.3×10^{-3} to 8.9×10^{-3}s under the experimental conditions used in this work. This relaxation time range was similar to the findings by Allal et al.[3] The relaxation time of PP increased considerably with the presence of wood particles (at 10 wt %), and slowly increased when increasing the wood content to 30 wt %. This was to be expected, as wood materials usually possess higher elastic response when compared with polymeric materials.[28,29] It was interesting to observe that the relaxation times of the PP and WPP composite extrudates progressively decreased with increasing die rotation speed. The lowering effect on the relaxation time was more pronounced for the WPP composite with the highest wood content (30 wt %). The reduction in relaxation time means that the WPP composite melt could be allowed to relax faster while flowing in the die, and hence had less stored elastic energy to cause skin fractures in the extrudate. In other words, the rotating die system was able to reduce the severity of sharkskin by shortening the relaxation time of the WPP composite materials.

CONCLUSIONS

This work used a rotating die system to extrude molten PP and wood/PP composites in a single-screw extruder. The entrance pressure drop and sharkskin level were investigated under the effects of various wood contents added to PP, wall shear rate, and die rotation speed. The results suggested that without the use of a rotating die system, the entrance pressure drop of PP increased with increasing wood content. In the WPP composite melts, the entrance pressure drop progressively decreased by 20–50%, depending on the shear rates used, by increasing the die rotation speed from 0 to 70 rpm. The sharkskin level and relaxation time were found to increase with increasing wood content but could be moderated or minimized by rotating the die. The relaxation times of the WPP composite melts were in the range of 4.3–8.9 $\times 10^{-3}$ s—the higher the rotation speed, the greater the magnitude of reduction of sharkskin and relaxation time. The rotating die effect was most pronounced for the PP extrudate with 30 wt % wood content. Based on the experimental results of this work, the rotating die system was found to be suitable and effective for moderating the entrance pressure drop and severity of sharkskin in wood/PP composites with relatively high viscosities.

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