

## Flow Behaviour and Viscoelasticity of Polypropylene-Kaolin Composites Extruded at Different Temperatures

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### ABSTRACT

A study of kaolin addition in polypropylene (PP-kaolin) melt was carried out to characterize its flow behaviour and viscoelasticity at different temperatures. The compound of 20 wt% kaolin was prepared by melt mixing using two roll-mill heated at 185°C, while the compounded composites were put through a single screw extruder to evaluate its melt flow properties. The prepared PP-Kaolin composites exhibited a shear thinning behaviour and appeared to be strongly dependent on temperature. Moreover, it was also found that the power law index was constantly increased as the temperature increased. Meanwhile, a similar trend was observed for swelling ratio, whereby it also increased with increasing temperature. It was also observed that changes in the die temperatures would result in the formation of obvious bubble like surface morphology, and it became more prominent when the temperature was lowered.

**Keywords:** Composites, polyolefins, processing, rheology, swelling

### INTRODUCTION

Flows of molten polymer are greatly affected by the heat generation due to viscous dissipation. This heat generation could cause large temperature rises in the region of high shear rate, such as near the wall of capillaries and die. Moreover, significant flow rearrangement may also occur due to the large temperature dependence of viscosity of most polymer melts. The occurrence of viscous dissipation or shear heating may harm and generate significant error in the processing of polymer melt, particularly for data taken at high shear stress or shear rates (Gang Yang, 1998; Gupta, 2000). The effect of shear heating or sometimes interpreted as non-Newtonian effect may cause reduction in polymer viscosity as the shear rate increases. Therefore, initiatives are taken in this study to determine the effect of flow and viscoelastic behaviour of PP-kaolin composites at different test temperatures.

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## EXPERIMENTS

### Materials

The thermoplastic used was Polypropylene (PP) copolymer, grade Pro-Fax SM240, supplied by Titan PP Polymer, Malaysia, and with the melt index of 25g/10 min. Kaolin was provided by Finn Chemicals with the density of 2.59 g/cm<sup>3</sup> and the mean particle diameter of 3.0 μm. PPgMA was supplied by the Exxon Mobil Chemical. The compound formulations contained 75 wt% PP, 5 wt% PPgMA, whereas 20 wt% Kaolin was chosen as several difficulties in extrusion process was confronted when exceeding 20 wt% of kaolin loading.

### Rheological Test

The rheological behaviour evaluation of the prepared sample was performed using a Brabender stand-alone single screw extruder KE 19/25 D. The test was conducted using a fabricated die with the L/D ratio of 22/3. The die temperature used in this study was in a range of 190°C - 230°C. Meanwhile, the volumetric flow rates (Q) were calculated from the weight of the extrudate cut in grams for 15 sec ( $W_{ext}$ ), which was divided with the melt density ( $\rho_m$ ) obtained from the MFI, as expressed in equation 1.

$$Q = \frac{W_{ext}}{\rho_m} \quad (1)$$

The apparent shear rate ( $\dot{\gamma}_{app}$ ) and the apparent shear stress ( $\tau_{app}$ ) were calculated from the known length (L) and radius (R) of the fabricated die and the pressure (P) generated at the die during the extrusion. These parameters are expressed in equations 2 and 3. The apparent shear viscosity is given by the relation of  $\tau_{app}$  over the  $\dot{\gamma}_{app}$ , as shown in equation 4 below.

$$\tau_{app} = \frac{RP}{2L} \text{ (apparent shear stress)} \quad (2)$$

$$\dot{\gamma}_{app} = \frac{4Q}{\pi R^3} \text{ (apparent shear rate)} \quad (3)$$

$$\eta_{app} = \frac{\tau_{app}}{\dot{\gamma}_{app}} \quad (4)$$

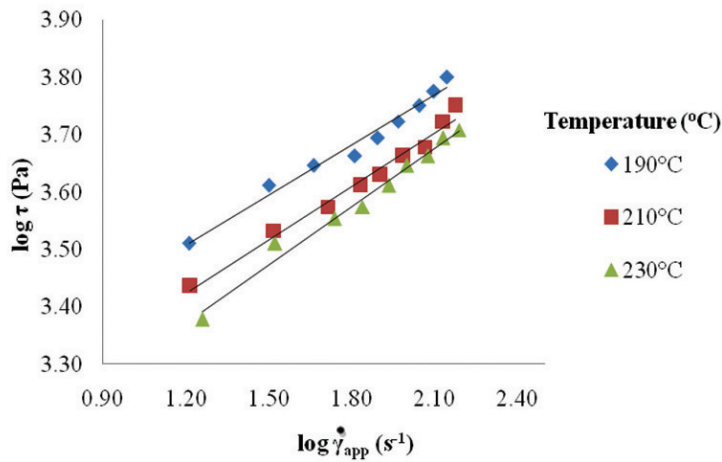
The extrudates were carefully cut at 3 cm long strand from the die and directly quenched in water. The ratio of the extrudate diameter to the die diameter was calculated using equation 5.

$$B = \frac{D_e}{D_d} \quad (5)$$

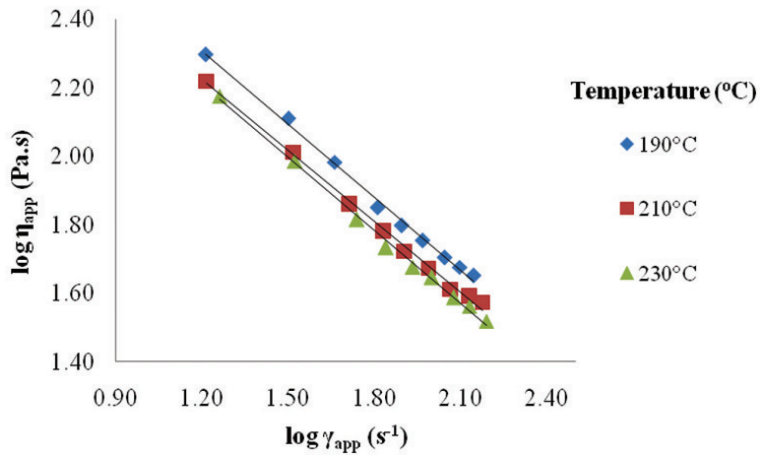
where  $D_e$  and  $D_d$  are the diameters of the extrudate and the die, respectively.

**RESULTS AND DISCUSSION**

The results gathered for the effects of temperature on the flow behaviour of the PP-kaolin composites are shown in *Fig. 1*. It has been clearly demonstrated that  $\tau_{app}$  decreases with the increase in temperature. Initially, the decreasing value in  $\tau_{app}$  adhered to the common trend previously observed by many researchers for other polymeric materials (Gupta, 2000; Brydson, 1989; Sombatsompop & Thongsan, 2001). This could be explained by with the rise of the temperature, while the flow resistance decreased with the increasing amount of free volume. *Fig. 2* illustrates the correlation between the  $\eta_{app}$  of the melt and the  $\dot{\gamma}_{app}$  at test temperature ranging from 190°C to 230°C for the PP-kaolin composites. As expected, the melt  $\eta_{app}$  reduced with the increase in  $\dot{\gamma}_{app}$  and test temperatures respectively (Liang *et al.*, 2000; Liang *et al.*, 1998).



*Fig. 1. Melt flow curve of PP-kaolin composites at various temperatures*



*Fig. 2. Viscosity curve of the PP-kaolin composites at various temperature*

Table 1 shows the power law flow indices ( $n$ ) of the power law relation which was obtained from the slope of the linear variation of  $\log \tau_{app} - \log \dot{\gamma}_{app}$  plot. It was also observed that the value decreases with temperature, indicating that the polymer melt became more pseudoplastic as the temperature increased; therefore, signifying the ease of flow for the polymeric chain segment.

TABLE 1  
Power law index ( $n$ ) for PP-kaolin composites at different temperatures

(°C)	Power law index, $n$
190°C	0.3389
210°C	0.3053
230°C	0.2892

Extrudate swell is often related to the elastic recovery of the materials at the inlet of the die where the polymer usually swells to a much greater diameter than that of the orifice. Fig. 3 illustrates the swelling ratio of the PP-kaolin composites at different  $\dot{\gamma}_{app}$  for various processing temperatures. It also shows that the increase in temperature causes a decrease of the swelling ratio. This could be explained by the decrease in the polymer melt strength with the increase in temperature (Samsudin *et al.*, 2006).

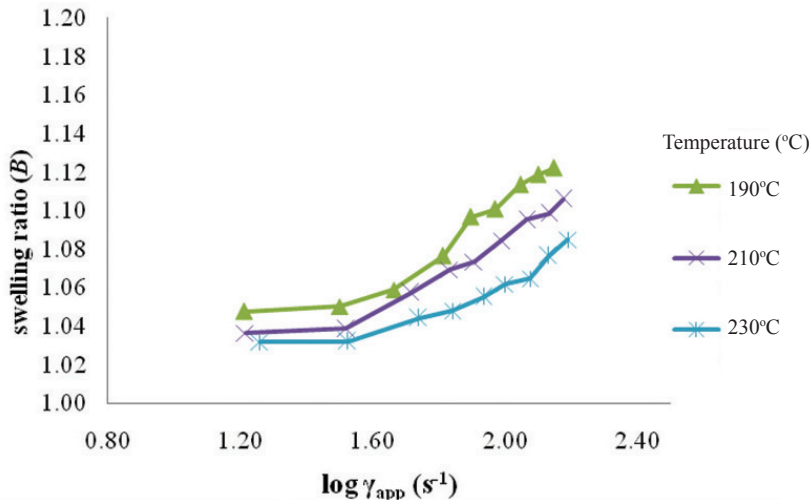


Fig. 3: Swelling ratio vs. shear stress of PP-kaolin composites at various temperatures

The extrudates collected were examined, whereas the extrudate surface textures of the composite at a constant speed of 35 rpm for the temperature of 190°C and 230 °C are presented in Fig. 4. It can be observed that the bubble size for the extrudates at lower temperature of 190°C is bigger compared to the bubble size generated for the extrudates at higher temperatures. This phenomenon is closely related to the decreasing value of  $\eta_{app}$  at higher processing temperatures. At the same amount of filler loading,  $\eta_{app}$  plays a vital role in altering the surface tension of the polymer. In

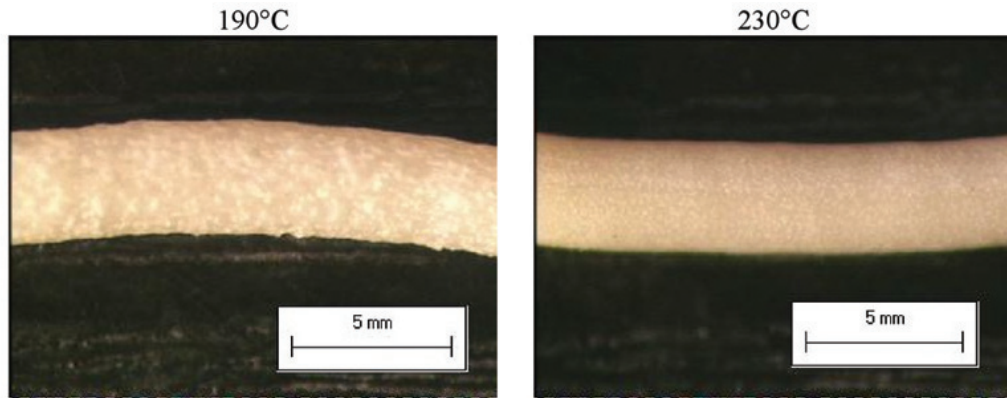


Fig. 4. The extrudate surface texture of PP-kaolin composites at various temperatures

the molten state, lower  $\eta_{app}$  will initially decrease the polymer melt surface tension, and produce smaller bubbles like surface texture which are obtained when high shear forces are applied with respect to the melt with higher  $\eta_{app}$  (Brydson, 1989; Beloshenko *et al.*, 1999) at lower temperature. Therefore, this finding provides an indication that it is important to optimize the extrusion speed of the PP-kaolin composites as smooth extrudate surface texture was obtained below 35 rpm at the temperature of 190°C to 230°C.

### CONCLUSIONS

$\tau_{app}$  has been shown to decrease, while the melt  $\eta_{app}$  reduces with the increase in  $\dot{\gamma}_{app}$ , as the temperature increases. This is caused by the decreasing flow resistance with the increasing amount of free volume (i.e. with a rise in the temperature). On the other hand, the increase in the processing temperature was observed to have caused a decrease of the swelling ratio due to the decrease in the polymer melt strength. Meanwhile, the size of bubbles on the extrudate surface at lower temperature of 190°C was found to be bigger compared to the size of the bubbles generated for the extrudates produced at higher temperature. This phenomenon could be closely related to the decreasing value of that creates lower melt surface tension at higher processing temperatures.

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