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Material behaviour

Melt flow behavior of polypropylene composites filled with multi-walled carbon nanotubes during extrusion

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ABSTRACT

Polypropylene (PP) composites filled with multi-walled carbon nanotubes (MWCNTs) were prepared using a twin-screw extruder. The melt flow properties of the composites were measured with a capillary rheometer in a temperature range from 180 to 230 °C and at various apparent shear rates varying from 100 to 4000 s^{-1}. The results showed that the melt shear stress increased almost linearly while the melt shear viscosity decreased almost linearly with increasing shear rates in a bi-logarithmic coordinate system. The melt shear flow followed the power law relationship and the dependence of the melt shear viscosity on temperature obeyed the Arrhenius equation. The relationship between the melt shear viscosity and the MWCNT weight fraction was roughly linear under the investigated range of temperature or shear rate.

1. Introduction

The carbon nanotube (CNT) is a new type of carbon filler, in the form of a cylinder with both ends sealed through crimping a graphene layer as a tube. In addition to the special seamless nanometer tube structure, CNT has some properties of general nanometer materials; it has excellent mechanical, electrical and thermal conductive properties due to its high specific surface area and length to diameter ratio. The properties of polymeric materials can be improved or some new properties can be obtained if they are filled with CNTs [1–3]. CNTs can be divided into two categories: single wall carbon nanotubes (SWCNTs) and muti-walled carbon nanotubes (MWCNTs). Owing to the lower cost of MWCNTs, the applications of the MWCNTs are more extensive than those of the SWCNTs.

Polypropylene (PP) is one of the five general plastics. It has become a widely used thermoplastic resin due to its advantages such as low cost, ease of production, recyclability and comprehensive good properties. However, PP has some disadvantages including high flammability, low strength, high notch sensitivity, etc. Thus, studies on modification of PP resin have been made to extend its application range. PP modified with CNTs has been an important new research area, and a number of research results have been obtained since CNTs were discovered. The PP/CNTs composites have been prepared by means of methods such as melt blending [4,5], shear blending [6], melt spinning [7], in situ polymerization [8] and solution blending [9].

Rheological properties reflect the processing and performances of polymeric materials. However, the rheological behavior of polymer/CNTs systems has not been much investigated [10]. Melt extrusion is one of the major processing routes for polymers. Melt shear viscosity is an important parameter for characterizing the melt flow behavior of polymers during extrusion, and is often

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required for determining processing conditions and for the design of extruders and runner systems of an injection mould tool [11–13]. Therefore, it is meaningful to conduct studies on the polymer processing and rheological behaviors of PP/CNTs composite systems, such as their shear flow behavior. The objectives in the present study are to investigate the influence of filler content and processing conditions on the melt flow behavior of the PP/MWCNTs composites during extrusion.

2. Experimental

2.1. Raw materials

The polypropylene with trademark CJS-700 serving as the matrix material was supplied by Guangzhou Petrochemical Works in Guangdong province (Guangzhou, China), and its density in the solid state and melt flow rate were 910 kg/m^3 and 10 g/10 min (230 °C, 2.16 kg), respectively.

Muti-walled nanotubes (MWCNTs) were selected as the filler to identify the influence of the filler content on the melt flow behavior of the composite systems. The MWCNTs were supplied by the Chengdu Organic Chemical Co., Ltd. of the Chinese Academy of Sciences (Chengdu, China), and prepared by means of the chemical vapor deposition method. The main properties include external diameter about 8–15 nm and length 30–50 μm, carbon content >90% and density 2.1 g/cm^3.

2.2. Composite preparation

The PP was mixed with MWCNTs in a high speed compounding machine, model GH-10, supplied by Beijing Plastics Machinery (Beijing, China). The PP/MWCNT blends were then melt-blended in a twin-screw extruder, model SHJ-26, supplied by Nanjing Chengmeng Machinery Ltd. Co. (Nanjing, China) at a screw speed of 100 rev/min and in a temperature range from 190 °C to 210 °C, to prepare the PP/MWCNT composite systems. The screw diameter was 26 mm, while the length to diameter ratio of the screw was 40. The weight fractions of the MWCNT were 1, 2, 3, 4 and 5 wt.%. The extrudate of the composites was granulated, and then the granules were dried at 80 °C for five hours before testing.

2.3. Instrument and methodology

The melt flow properties of the PP/MWCNT binary composites were measured using a capillary rheometer, model Rheologic 5000, supplied by the Ceast Co. Ltd (Italy). The melt flow behavior tests were carried out in temperature range from 190 to 230 °C and at different apparent shear rates from 100 to 5000 s^{-1}. The diameter and the length to diameter ratios of the capillary die were 1 mm and 40, respectively; and the die entry angle was 180°. It is generally believed that the effect of entrance pressure losses on extrusion flow behavior of polymer melts is insignificant when the length to diameter of die is greater than 30 [14]. Thus, the shear stress at the channel wall (τ_w) may be expressed as follows:

$$\tau_w = \frac{\Delta PD}{4L} \tag{1}$$

and the apparent shear rate can be given by

$$\gamma_a = \frac{32Q}{\pi D^3} \tag{2}$$

3. Results and discussion

3.1. Melt flow curves

A melt flow curve describes the relationship between shear stress and shear rate during extrusion flow of polymer materials. Fig. 1 shows the melt flow curves of PP/MWCNT composites at 200 °C. It can be seen that the value of shear stress at the die wall increases approximately linearly with increasing apparent shear rate in a bilogarithmic coordinate system. This means that the melt shear flow of the composite system roughly obeys the following power law equation under the experimental conditions.

$$\tau_w = K\dot{\gamma}_a^n \tag{3}$$

where the parameter n is the non-Newtonian index, and K is the consistency of polymer melt. The values of K and n under experimental conditions can be obtained by using a linear regression method. Table 1 lists the values of K and n of the PP/MWCNT composite melts at 200 °C. It was found that the values of the consistency of the composite melts increase while the values of the non-Newtonian index decrease with increasing filler weight fraction. Moreover, the value of linear correlation coefficient (R) is higher than 0.99.

Fig. 2 displays melt flow curves of the PP/MWCNT composites at 220 °C. Similar to the results shown in Fig. 1, the values of shear stress at the die wall increase approximately linearly with increasing apparent shear rate in a bilogarithmic coordinate system. This also means that the

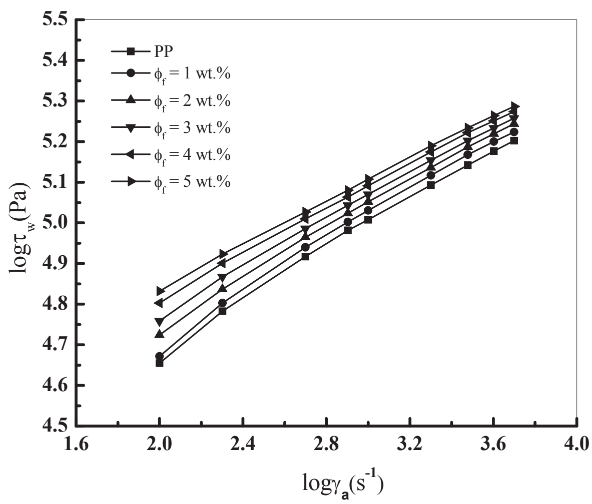

Fig. 1. Melt shear flow curves at 200 °C.