An Investigation on the Rheological Properties of Ultra-high Molecular Weight Polyethylene Single-polymer Composites

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Abstract. Rheological behavior of Ultra-high molecular weight polyethylene single-polymer composites (UHMWPE SPCs) was studied. UHMWPE cloth was used to prepare polyethylene single-polymer composites (PE SPCs). SPCs samples were prepared by imitating hot pressing process with a rotational rheometer. Rheological data was collected by the rheometer during the preparation of the material. Rheological responses such as the storage modulus and complex viscosity to a dynamic strain sweep, and linear viscoelastic (LVE) frequency sweep were achieved. The relationship between rheological properties of PE SPCs and temperature was discussed. The LVE range of the PE SPCs was extended with increasing temperature. Both storage modulus and complex viscosity of the PE SPCs were decreased with increasing temperature. The composites showed pseudoplastic shear thinning behavior. Reinforcement effect of fiber was more obvious in the low-frequency range than in the high-frequency range.

1. Introduction
Traditional fiber reinforced polymer composites exhibit excellent mechanical properties so that they have been widely used. Conventional fiber reinforced composites are typically reinforced with glass fibers, carbon fibers, and natural fibers [1]. However, these composites almost cannot be easily recycled, as it is hard to find a suitable and economical method to separate reinforcement from matrix and meanwhile keep them intact [2]. SPCs made up of same chemical compositions but different physical properties of matrix and reinforcement, seem to can solve this problem. They also have superior interfacial properties and smaller weight. So SPCs have received more and more attention in recent years especially in new material and environmental fields. It is particularly important to study the performance of the SPCs.

Rheological analysis is a very important method in many studies of composites. It is considered an effective tool for the investigation of processing behaviour and microstructural features. The responses of the complex viscosity ($\eta^*$), storage modulus ($G'$), and loss modulus ($G''$) obtained from rheological analysis provides a better understanding of the process ability, dispersion of the filler in the matrix, matrix-fiber interactions, etc. [3, 4]. Most of the existing rheological researches focus on micro-and nanoparticle filled polymer composites and traditional fiber reinforced polymer composites. The effect of filler length, concentration, orientation and dispersion on $\eta^*$, $G'$, $G''$, yield stress and reinforcement network of composites is usually investigated. However, literatures are scanty regarding the rheological properties of SPCs. Fortunately, many investigations on the melt rheological behavior of
fiber reinforced thermoplastics and elastomers have been reported. The research methods they used are also applicable to SPCs.

Because ultra-high molecular weight polyethylene (UHMWPE) have various excellent properties and extensive application, we prepared and studied UHMWPE SPCs. In addition, rotational remoter is widely used in rheological testing due to its high accuracy, wide range of applications and easy to operate. In this investigation, the UHMWPE cloth was made into PE SPCs by the rotational remoter. The effects of operating temperature on the viscoelastic properties of PE SPCs were investigated by dynamic rheological analysis. Moreover, the fiber content and the enhancement effect of fiber were analyzed by the rheological data.

2. Experimental

2.1. Materials
The plain woven UHMWPE cloth (Barrday Corp., Canada) with warp density and weft density both of 90 threads/10cm was utilized to prepare the PE SPCs. There are 300 fibers in each bundle. The cloth has a strength of 300 MPa.

2.2. Differential Scanning Calorimetry
Melting point (Tm) of the samples were measured by a differential scanning calorimeter (DSC), Shimadzu Co., to guide the processing temperature setting. The samples, sealed in an aluminum pan, heated at a rate of 10ºC /min from 30°C to 240°C.

2.3. Preparation of the PE SPCs
We imitated hot pressing process [5] to prepare PE SPCs. They were formed by UHMWPE cloth in a Bohlin Gemini II rotational remoter (Malvern Instruments Inc., Worcestershire, United Kingdom) which provided temperature and pressure when preparing PE SPCs. The temperature in the sample chamber is controlled by the normal extended temperature cell (ETC) electrical heater. Samples will be heated by compressed air preheated in the ETC. The cloth was cut into a circular sample with a diameter of 25 mm, which was the same size as the parallel plate of rotational remoter. For all experiments, the normal force applied to samples by two 25-mm parallel plates was set to 200 g by auto-tension function, which equals 4074 Pa. The cloth could become the PE SPCs through the combination of surface melted fiber and internal unmelted fiber when it was sandwiched between two parallel plates. The temperature of 150, 155 and 160℃ were used respectively to investigate the influence of temperature on the rheology properties. It was determined by the DSC testing results.

2.4. Rheological Characterization of the PE SPCs
The dynamic rheological properties were measured using a stress and strain controlled remoter. The experiments were carried out in parallel plate mode with a plate diameter of 25 mm. We used a parallel disc oscillatory shear method, which did not change the material structure during the test as it is a small amplitude deformation test [6]. A dynamic strain sweep was conducted first to determine the linear viscoelastic region by varying the possible strain ranges from 0.1 to 50% at a constant frequency of 1 Hz at each temperature. Dynamic frequency sweeps were then carried out from 0.01592 to 15.92 Hz (0.1~100 rad/s) at a constant strain within the linear viscoelastic (LVE) region of the material. We have kept a constant strain of 1% for the frequency sweep study because all the samples at different temperatures show an LVE region within the strain range. We did frequency sweep on samples at 150, 155 and 160 °C respectively. Because a very thin sample results in a small plate gap, the normal force experienced by the sample will be so large that it may damage the instrument and the test results are not accurate when plates thermal expansion. We varied the gap of two plate with a fixed normal force in each testing.
3. Results and Discussion

3.1. Differential scanning calorimetry
In the heating curve of the UHMWPE fabric, there are two melting peaks, as shown in Figure 1. The first and second peak value are 144.60°C and 154.23°C, respectively, which is due to the different crystalline forms of UHMWPE fiber. In order to study the performance of different samples, we set 150°C, 155°C and 160°C temperatures in the processing and testing.

![Figure 1. DSC traces of the UHMWPE fabric.](image)

3.2. Rheological Characterization of the PE SPCs
Before a dynamic frequency sweep, it was necessary to determine the linear viscoelastic range of each sample. Once the linear viscoelastic range is exceeded, the structure of sample is destroyed. Figure 2 shows the storage modulus (G’) variation as a function of strain with a frequency of 1 Hz. At three temperatures, the curves followed the same trend. At low strain, G’ showed no effect by strain and then exhibited a drastic drop when a certain strain was reached. The samples showed LVE regions up to a strain level of 4%, 10% and 12% at 150°C, 155°C and 160°C, respectively. In this study, a strain of 1% was chosen for the dynamic frequency sweep to measure the linear viscoelastic properties of all samples. Figure 2 also shows that as the temperature increased G’ decreased but the linear range was extended. The effect of temperature on the performance of SPCs is critical. The higher the temperature, the more fiber melting and the lower fiber contents of the composites. In addition, the motion of molecular chains became fast at high temperature. Thus the performance of the sample would be similar to the fluid and the shear resistance of the sample would be low. As a result, the value of G’ was small and the sample could undergo greater strain at high temperature.
The changes of the complex viscosity of the PE SPCs are presented in Figure 3. The complex viscosity decreased with frequency increasing continuously at each temperature. The composites show pseudo plastic shear thinning, which implied that the PE SPCs exhibited non-Newtonian fluid behavior in the whole testing frequency range. We have already discussed the relationship between temperature and fiber contents of the composites. The figure indicated that the complex viscosity increased with increasing fiber contents, and the reinforcement effect was more obvious at low frequency than at high frequency. This phenomenon demonstrated that the fiber played a more dominant role at low oscillatory frequency than at high oscillatory frequency. Meanwhile, the viscosity difference became smaller in the high-frequency range.

The storage modulus values of the PE SPCs as a function of frequency are also shown in Figure 4. In these curves, the storage modulus at low frequency is lower than that at high frequency. This is due
to the fact that at low frequency time is large enough for unraveling of the entanglements, a large amount of relaxation occurrence results in small values of storage and loss modulus [7]. However, when the composite is deformed at large frequency, the entangled chains do not have time to relax so modulus goes up. The storage modulus difference became smaller in the high-frequency range. Also, like complex viscosity, the storage modulus of the samples increases as temperature decreases. This indicates that the modulus also increases with the fiber contents.

![Graph](image)

**Figure 4.** Variation of storage modulus with frequency sweep at different temperatures.

### 3.3. Morphology of the PE SPCs

Figure 5 shows the photographs of PE SPCs processed in different temperatures. The sample on the left is the original UHMWPE cloth with a diameter of 25mm. As shown in the picture, the samples had a great degree of shrinkage at different processing temperature. The sample, prepared at 150 °C, had a higher degree of regularity and could see the net structure of fiber woven, which revealed the fact that only a part of fiber woven melted. More fiber would melt with the increase of temperature. For example, a majority of fiber woven melted at 160 °C, and the degree of regularity of structure decreased. This was because the fiber woven melted forming a lot of fractionlets at high temperature and the viscosity of the sample decreased, which could not withstand the high shear force resulting in the degree of regularity of the sample decreased.

![Photograph](image)

**Figure 5.** Photograph of PE SPCs processed in different temperatures.

### 4. Conclusion

The preparation and rheological analysis of PE SPCs were realized simultaneously by rotational remoter. Studies on rheological properties of the PE SPCs showed that the linear viscoelastic range of the PE SPCs was extended with increasing temperature. Meanwhile, both storage modulus and
complex viscosity of the PE SPCs were decreased with increasing temperature. The composites showed pseudo plastic shear thinning behavior. The reinforcement effect of fiber was more obvious in the low-frequency range than in the high-frequency range. The storage modulus at low frequency is lower than that at high frequency. This is related to the time it takes to unravel the entanglements of molecular chains.

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