Mechanical and Rheological Properties of High Density Polyethylene Reinforced Polyvinyl Alcohol Fiber Composites

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Abstract. In the current study, high density polyethylene filled polyvinyl alcohol fiber composites have been made via melt compounding process using a twin screw extruder. Four different fiber loadings (0, 5, 10, 20 wt%) together with HDPE matrix were mixed and melt blended with the extruder. The prepared composites were tested for their melt rheological properties, mechanical properties, FT-IR spectra, and water absorption behavior. Rheological test results exhibited that complex viscosity of the composites were higher than the neat HDPE and increased with the increase of PVA loadings. Moreover, the improvement of complex viscosity was more prominent at higher PVA loadings (i.e. PVAC-10 and PVAC-20) than at the lower one (PVAC-5). The flexural modulus and strength were higher for the all composites samples when compared to the neat HDPE, indicating that the incorporation of PVA fiber has successfully improved the mechanical (i.e. flexural) properties of the HDPE/PVA fiber composites. The FTIR analysis results prevailed the appearance of C=O spectrum at 2361 cm^{-1} that corresponding to carbonyl bond of PVA fiber on the whole composites. Additionally, from the water uptake test, the degree of water absorption of the composites increased with the fiber loadings.

Introduction

Poly(vinyl alcohol) (PVA) fiber is also known in Japan as "vinylon" or in United States as "vinal". This fiber has been used as reinforcing material for plastic since last century. This fiber has interesting features, e.g. good mechanical properties, good resistance to chemicals like acid and natural conditions, low/competitive price, widely available in the market. These will make PVA fiber will continuously become good reinforcing material for worldwide applications [1].

Research studies in the field of using PVA fiber as reinforcement in polymeric materials are still limited, which makes this research area is interesting. Most of research studies focused more in the use of PVA fiber as reinforcement on engineering Cementous composite (ECC) for construction, where cement/mortar acted as the matrix. Whereas, several literatures that reported about the use of PVA as reinforcement material in polymer composites are as follow: Chafidz et al. [2] studied the dynamic mechanical thermal analysis (DMTA) of PVA fiber reinforced HDPE composites. They found that storage modulus of the composites improved with the increase of PVA fiber concentrations, indicating that the stiffness of the composites has improved. Li et al. [3] studied about short PVA fiber reinforced geopolymer composites (SFRGCs) via melt extrusion. They reported that the SFRGCs could be prepared without any rheological modifier. The resulted composites also showed an increase in the ductility of SFRGCs, which changed the properties from brittle to ductile.

Wu and Shen [4] studied the effect of irradiated HDPE incorporation to the HDPE/PVA fiber composites. They found that the yield strength and impact strength of the HDPE/PVA fiber composites considerably improved. Additionally, it has been well-known that the melt rheological properties are the important factors to understand the melt processability as well as internal morphology of the polymer composites [5]. Therefore, it is important to study the melt rheological properties of the polymer composites studied. In this study, we have developed HDPE composites using PVA fiber as reinforcement. The composites were made by melt compounding/blending method utilizing a twin screw extruder and followed by injection molding machine to prepare standard samples. The effects of relative weight fraction of the PVA fibers on the mechanical properties (e.g. flexural strength), rheological properties, FT-IR spectra, and water absorption behavior of the composites have been investigated.

Experimental

Materials. High Density Polyethylene (HDPE) used as a matrix was an injection molding grade, which obtained from a local market in Saudi Arabia. It has melt index of 30 g/10min (at 3 kg and 190°C) and density of 954 kg/m3. Whereas, the poly (vinyl alcohol) (PVA) fiber was supplied by Kuraray, Japan. From the manufacturer's datasheet, the fiber length was 8 mm. Prior to the composites fabrication, the PVA fiber and HDPE pellets were manually mixed and put in the oven at 70°C for 24 hours to reduce the moisture content. Fig. 1 shows the physical appearance of HDPE pellets and PVA fiber.
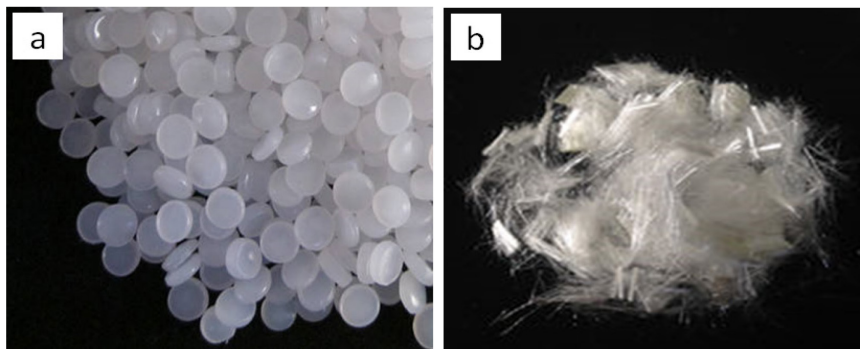

Fig. 1 Physical appearance of a) HDPE pellets, and b) PVA fiber.

Preparation of Composites. To prepare the composites, the mixed and dried HDPE matrix and PVA fiber were melt compounded with different contents of PVA fiber (ranging from 5% to 30%) by using a twin screw extruder (TSE), Farrel FTX-20, United Kingdom. The TSE was run at die temperature of 200°C and screw speed of 17 rpm. The molten composites that coming out from the die was directly immersed thorough cooling-water bath, then air dried and pelletized. The pellets were then put in oven at 100°C for 2 hours to reduce the moisture content. Afterward, they were fed into an injection molding machine, Super Master Series SM 120, Asian Plastic Machinery Co., China to make ASTM standard molded samples. The cooling-water temperature used to cool the mold was 15°C. The detail processing conditions for the injection molding machine are shown in Table 1. The composites samples were referred to as PVA-0, PVA-5, PVA-10, and PVA-20 for the addition of PVA fiber at 0, 5, 10, 20 wt% loadings, respectively. The PVA-30 was excluded from the characterization step since the composites failed to obtain as it very easy to cut and break when it came out from the die.

Table 1 Processing parameters of the injection molding machine.

Injection pressure (bar)	Temperature (°C)				Screw speed (rpm)	Cooling time (sec)	Total Cycle time
600	T 1	T 2	T 3	Feed	200	20	40

Characterization of Composites. The prepared composites were then characterized by melt rheological test, flexural test, FT-IR analysis, water uptake analysis.

Melt rheological analysis. The melt rheological properties of the HDPE/PVA fiber composites was characterized by a rheometer ARG2, TA Instruments, USA. The test was done on parallel plate geometry. The sample used in the test was a square plate of 25 mm x 25 mm with thickness of 3.2 mm. Additionally, the test was done in frequency sweep procedure at consant temperature of 150°C; Strain of 0.01%; and angular frequency range of 0.1 – 6228.3 rad/s. Before starting the melt rheological test, once the temperature was stable at 150°C for a while, the excess of molten sample (if any) was cleaned to make sure the measurement is validated. The result of the melt rheological test was the plot of complex viscosity, $|\eta^*|$ versus angular frequency, ω.

Flexural test. One of important mechanical properties is flexural (i.e. modulus, strength). The flexural test was done on Hounsfield H100 KS testing machine based on ASTM D-790 standard. The test specimens had dimensions of 3.2 × 12.7 × 119 (mm). The test was done at a loading speed 20 mm/min and span range 40 mm. Flexural strength was calculated from the records of the computer connected to the testing unit.

Fourier Transform Infrared (FTIR) Spectroscopy. Nicolet iZ-10 spectrophotometer was used to obtain FTIR spectra of PVA fiber composites from 600 to 4000 cm^{-1}.

Water uptake test. The water uptake test based on method ASTM D-570 was done to determine the amount of water absorbed by composites. Prior to test the sample was dried in an air convection oven at 100˚C for 2 h. Then the specimen was put in a dish of distilled water at ambient temperature for 1h, 6h, 24h, 48h, 96h, 120h, 144h, and 192h. The test specimen was a bar with 28 mm long, 12.7 mm wide and 3.2 mm thick. For measurement, the specimen was taken out from the water, wiped with a dry cloth, and immediately weighed.

Results and Discussion

Melt Rheological Properties. The complex viscosity, $|\eta^*|$ versus angular frequency, ω of HDPE/PVA fiber composites measured at fixed temperature of 150°C is shown in Fig. 2. As seen in the figure, the complex viscosity of the composites were higher than that of neat HDPE. Furthermore, the improvement of complex viscosity was more prominent at higher PVA loadings (i.e. PVAC-10 and PVAC-20) than at the lower one (PVAC-5). It was most likely due to the incorporation of PVA fiber in the HDPE matrix, which hindered the HDPE molecular chains mobility in the melt state. As the PVA fiber content increased, the interfacial area shared by the PVA fiber and the neat HDPE also increased, and thus the HDPE chains mobility of became more hindered, which led to the increase of $|\eta^*|$ of the composites. Additionally, as seen in Fig. 2, the increase of $|\eta^*|$ was more significant at low ω region (0.1-10 rad/s) rather than at high ω region (10 – 628.3 rad/s). It can be explained that at high ω region, the PVA fibers tend to align with the HDPE matrix, and thus the possibility of the collision between fiber to fiber was much lower, which resulted in less complex viscosity of the composites at high ω region [6].
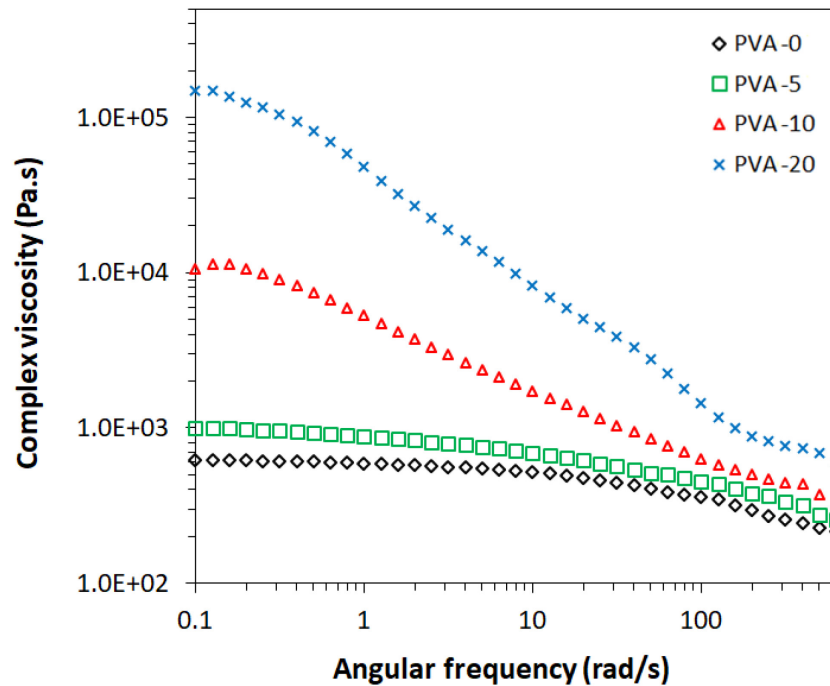
Fig. 2 Complex viscosity, $|\mu*|$ versus angular frequency (ω) of the composites.

Flexural Strength. The flexural modulus and strength of the HDPE/PVA fiber composites are listed in Table 2. As seen in the table, the flexural modulus was higher for the whole composites when compared to the neat HDPE, indicating that the addition of PVA fiber has successfully improve the mechanical (i.e. flexural) properties of the HDPE/PVA fiber composites. The percentage improvement in flexural strength, resulting from the reinforcement of the HDPE matrix by PVA fiber was 5%, 10%, and 33% for PVA-5, PVA-10, and PVA-20, respectively. Whereas, the percentage improvement in flexural modulus was 15%, 21%, and 73% for PVA-5, PVA-10, and PVA-20, respectively.

Table 2 Flexural properties of HDPE/PVA fiber composites.

Sample	Flexural Strength (MPa)	Flexural Modulus (GPa)
PVA-0	30.69	0.93
PVA-5	32.23	1.07
PVA-10	33.83	1.13
PVA-20	40.86	1.61

Fourier Transform Infrared (FTIR) Analysis. The spectra of neat HDPE, PVA fiber, and their composites are shown in Fig. 3. The broad absorption band in the region around 2916 cm^{-1} and 2848 cm^{-1} could be attributed to -CH$_2$ stretching. The peaks for the -CH$_2$ bending was observed at 1462 cm^{-1}, furthermore spectrum 718.9 cm^{-1} could be considered as -CH=CH- (cis). These bands are shown in neat HDPE and all of composites. These spectrums could be attributed to the hydrocarbon bonding from HDPE, in which as the fiber content increases the HDPE content will decrease. The spectrum peak on 2360 cm^{-1} is also shown clearly, which corresponding to C=O and appears only on the composites and the intensity also become larger with increasing on fiber content. This last peaks could be noticed as carbonyl bond of PVA fiber. Moreover, the sharpness of the peaks in composite spectrum showed regularity in polymer molecular chain. It can be noticed from IR spectra investigation that PVA fiber disperse well within the HDPE matrix.
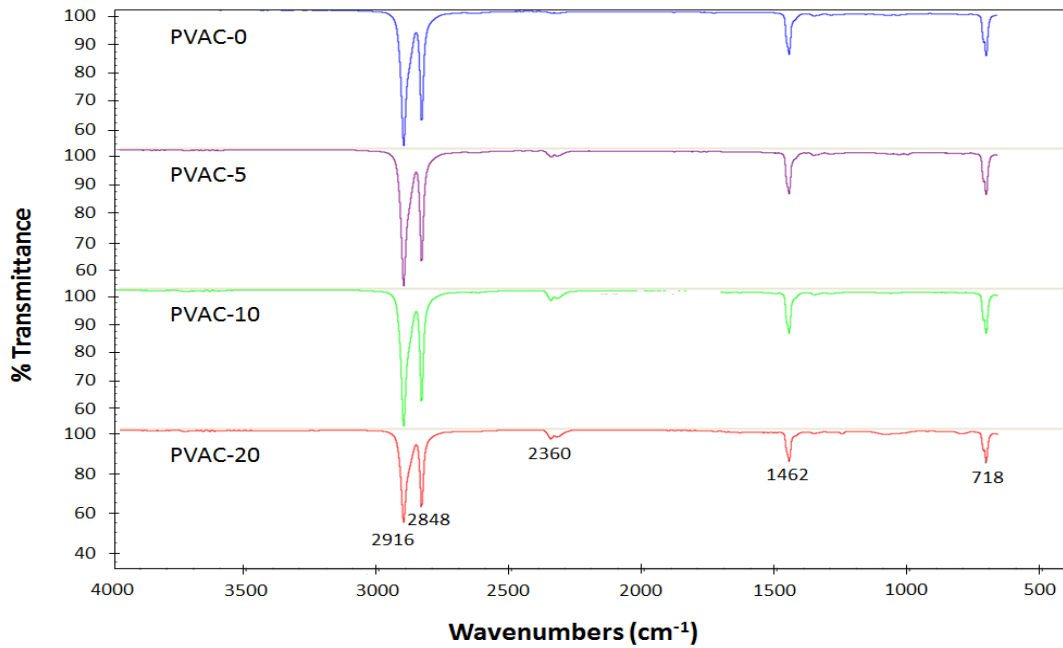
Fig. 3 FT-IR spectra of neat HDPE, PVA fiber, and HDPE/PVA fiber composites.

Water Uptake Test. The water uptake test in the form of water absorption behavior of HDPE/PVA fiber composites is exhibited in Fig. 4. As seen in the figure, the amount of water absorbed increased with increasing fiber loadings. From the water absorption curves in Fig. 4, it can be seen that the water absorption gradually increased and then it leveled off, which attribued to the equilibrium [7], which was taken at period of 144 hours. The significant increase was shown at composites with 20 wt% PVA loading, which was about seven times higher than the neat HDPE at equilibrium point. Since the PVA fiber is hydrophilic material, it was expected that amount of water absorbed by the composite increased with the increase of the fiber loading. The PVA fibers swolen due to the absorbed water, which led to the development of shear-stress throughout the interface between the matrix and the fiber. This resulted in the debonding and delamination of the composties. Additionally, water absorption could also be associated to the capillary phenomenon, where the presence of hydroxyl groups improves the water absorption by the formation of hydrogen bonds [7].
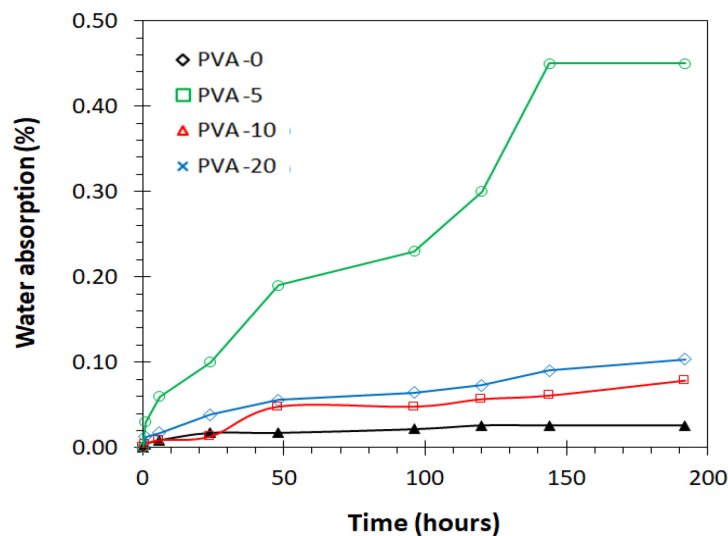

Fig. 4 Water absorption behavior of HDPE/PVA fiber composites

Conclusion

Rheological test results exhibited that the complex viscosities of the HPDE/PVA composites were higher than that the neat HDPE. Furthermore, the improvement of complex viscosity was more prominent at higher PVA loadings (i.e. PVAC-10 and PVAC-20) than at the lower one (PVAC-5). The flexural modulus and strength were higher for the all composites samples when compared to the neat HDPE, indicating that the incorporation of PVA fiber has successfully improved the mechanical (i.e. flexural) properties of the HDPE/PVA fiber composites. The percentage improvement in flexural strength, resulting from the reinforcement of the HDPE matrix by PVA fiber was 5%, 10%, and 33% for PVA-5, PVA-10, and PVA-20, respectively. Whereas, the percentage improvement in flexural modulus was 15%, 21%, and 73% for PVA-5, PVA-10, and PVA-20, respectively. The FTIR analysis results prevailed the appearance of C=O spectrum at 2361 cm-1 that corresponding to carbonyl bond of PVA fiber on the whole composites. Additionally, from the water uptake test, the amount of water absorbed by the composites increased with increasing fiber loadings.

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