

Mechanical, Thermal Stability and Water Absorption Properties of Recycled Thermoplastic Blend Nanocomposites: Comparison of Nanoclay and Carbon Nanotube

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Abstract: This present paper investigates the influence of clay (Cloisite 10A) and multiwalled carbon nanotubes (MWCNT) loading for the tensile, flexural, thermal stability and water absorption properties of recycled thermoplastic blend made from recycled high-density polyethylene (rHDPE) and recycled polyethylene terephthalate (rPET). The nanocomposites were prepared via extrusion blending technique by using extruder and compressed using hot and cold press molding machine. The findings showed that the sample with 1 phr of Cloisite 10A and 4 phr of MWCNT showed the optimum of tensile strength at 13.9 MPa and 14.5 MPa as compared to other loadings. The flexural strength exhibited inconsistent trend with the increasing of Cloisite 10A and MWCNT content. The addition of Cloisite 10A and MWCNT-reinforced nanocomposite showed better thermal stability than pure polymer blend. As the increment of Cloisite 10A and MWCNT loading, the percentages of water uptake increased but the values were still lower than the neat thermoplastic blend.

Keywords: Nanocomposite, tensile strength, flexural strength, analysis thermogravimetric, water absorption

1. Introduction

In recent years, polymer nanocomposites (PNCs) containing organic and inorganic nanofillers have gained a good interest from many researchers and industry because of their unique characteristics [1]. Comparing to neat polymers, nanocomposites exhibit mechanical properties, low permeability for gaseous products, enhanced flame retardancy and chemical resistance, good thermal stability, enhanced thermal and electrical conductivity [2].

In the field of packaging industry, polyethylene (PE) and polyethylene terephthalate (PET) have been widely used and the annual rate of growth for production and consumption is always increasing [3] because of their valuable properties such as good mechanical properties [4], low density and cost and also proficiency of processing [5]. High-density PE (HDPE) has broad applications such as bottles, containers and consumers good [6]. Post-consumer HDPE can be washed, chopped and blended with other materials to make a reprocessed material for different applications [7]. The post-consumer or recycled HDPE (rHDPE) was an ideal polymeric matrix to manufacture plastic wood (lumber), garden products and automobile parts. It can behave as an effective moisture barrier and remains in solid state at room temperature [8]. Many industries have evaluated that the use of rHDPE could help to decrease cost and lower the environmental impact [9]. According to Mallakpour and Behranvand [10], PET exhibits deficient mechanical performance and thermal stability which have limited their valuable application in a broad range of industry. Recently, an interesting technique to enhance the mechanical and thermal performances of polymer was the blending method. The mechanical recycling that blending virgin PET or recycled PET (rPET) with other polymer offers economic profit and practicable solution for producing new polymer products [11]. As stated by Salleh, Ahmad [12], polymer blending combines the excellent properties from individual polymer component to produce a new polymeric material. For instance, rHDPE/rPET blends are less brittle when compared to PET but are stiffer and faster cooling in comparison to neat HDPE.

The incorporation of nanofillers into recycled polymers will produce a value-added and high-performance nanocomposite that is attractive in polymer and composites industries [13]. In the study of Müller, Bugnicourt [14], they have investigated that the addition of low content of nanofiller into the polymer enhanced the mechanical, thermal and barrier properties without affecting their processability. Researchers claimed that the use of carbon nanotubes (CNT) in various fields such as biomedical [15] and electrical [16] have gained a huge attention because of their unique structure, remarkable mechanical and electronic properties [17]. By adding multi-walled CNT (MWCNT) into polypropylene, the mechanical and viscosity properties have been improved [18]. Another reason why MWCNT was used for nano-strengthening is that the tube-shaped allotropes of carbon like rolled graphene has ultra-high aspect ratio and superior mechanical properties [19]. Fouad et al. investigated the HDPE/CNT nanocomposites using extrusion blending technique and the findings reported that 4 wt% of carbon nano-particles as optimal content to improve storage and Young's modulus and yield strength of the nanocomposites. However, the addition of CNT was not affected by the melting temperature in the thermal analysis [20]. Okolo, Rafique [21] has found the improvement in tensile strength and elastic stiffness of CNT filled PE nanocomposite. In the research of Ahmad, Al-Juhani [22], the rheological behavior of HDPE reinforced CNT was improved up to 7 wt%. While in the studies of Tang, Santare [23], they concluded that HDPE reinforced 5 wt% MWCNT had increased mechanical properties with the micrograph observation of uniform dispersion of MWCNT small aggregates.

Numerous literatures showed the investigation on single polymer (such as PE)/CNT, yet the research published on polymer blend/CNT is limited. In this study, the tensile, flexural, thermal stability and water absorption properties of rHDPE/rPET incorporated with various loading of nanoclay and MWCNT were investigated.

2. Methodology

2.1 Materials

Recycled high-density polyethylene (rHDPE) with a melt flow index of 0.72 g/10 min (190 °C, 2.16 kg load) and recycled polyethylene terephthalate (rPET) with an intrinsic viscosity of 0.68 dL/g were donated from BioComposites Extrusion Sdn. Bhd. The Cloisite 10A nanoclay (CEC of 125 meq/100 g; *d* spacing = 19.2 Å) was received by Southern Clay Products Co. Multiwalled carbon nanotubes (MWCNT) with a dimension of 9.5 nm (diameter), 1.5 µm (length), 90% (carbon purity) and 250-300 m²/g (surface area) were obtained from NanocylTM. The rHDPE pellets, rPET pellets, Cloisite 10A and MWCNT powder were oven dried overnight at 90 °C before using.

2.2 Methods

The rHDPE and rPET composition was fixed at a composition ratio of 75/25 (wt%)[4], whereas the Cloisite 10A and MWCNT loading were varied at 1, 2, 4 and 6 phr, respectively. The range of nanofiller content was determined based on the literature [24, 25]. The rHDPE, rPET and nanofiller were melt-blended via co-rotating screw extruder (Thermo Prism TSE 16PC) with the temperature profiles of 190 °C, 240 °C, 270 °C, and 250 °C. The fixed rotation speed was 30 rpm. The extruded granules were then compression molded via hot and cold pressing process (LP50, LABTECH Engineering Company LTD) to produce the specimen panels for characterization. The temperature of upper and lower platens was fixed at 200°C under 1000 psi. The hot pressing took for 10 min and cold pressing for 5 min.

For characterization, the tensile strength and flexural strength was examined from the tensile and flexural testing run based on ASTM D 638-03 (type I) and ASTM standard D790-03 by using the Testometric M350-10CT testing machine under ambient condition. The 5 mm/min crosshead speed and 5 kN load cell were used. For each composition, six specimens were tested to get the average values. Scanning electron microscope (SEM) Model SUPRA 55 VP at 5 kV was used to observe the fracture surfaces of the sample. The thermogravimetric analyses (TGA) of the specimen were performed using thermogravimetric analyzer (Mettler Toledo TGA/SDTA851^e). Approximately 10 to 15 mg of each specimen was tested from room temperature to 600 °C with a heating rate of 10 °C/min under atmospheric air flow condition. The water uptake test was conducted in distilled water. After immersion for 2 and 24 hours, the wet and dry surface of the specimens was weighed. The percentages of water absorbed (WA) were determined using equations:

$$WA (\%) = \frac{W_t - W_0}{W_0} \times 100$$

Where W_0 is the weight of the oven-dried specimen (the initial weight), and W_t is the weight of the water absorbed specimen.

3. Result and Discussion

3.1 Mechanical Properties

Table 1 shows the tensile and flexural strengths of rHDPE/rPET/Cloisite 10A and rHDPE/rPET/MWCNT with different loading. In absence of nanofiller, recycled thermoplastic blend exhibited tensile strength at 10.9 MPa. The

mixture of Cloisite 10A nanofiller on rHDPE/rPET showed the increment in tensile strength for the composition of 1 phr and then decrement trend for higher Cloisite 10A content from 2 phr to 6.0 phr. The optimal Cloisite 10A content was achieved at 1 phr with the highest tensile strength at 13.9 MPa. This reinforcing effect can be ascribed to the sludge between the silicate layers of Cloisite 10A which causes the spread of Cloisite 10A to become homogeneous inside the polymer matrix [26]. However, the exceeding loading of Cloisite 10A, i.e. 2 phr in this study is believed to cause the formation of agglomerate Cloisite 10A and poor dispersion of nanofiller in composites [27]. This is because of the higher energy of the Cloisite 10A surface which is due to the van der Waals interaction between the filler and the matrix [28]. On the other hand, the tensile strength was found to increase as the amount of MWCNT increased. The optimum tensile strength was obtained at 4 phr of MWCNT which was increased by about 14.5 MPa as compared to neat rHDPE/rPET. It is believed that a great interface between the MWCNT and rHDPE/rPET composites is necessary factor for the material to stand the stress [29]. As MWCNT loading was higher than the optimum level, the tensile strength started to decrease at 6 phr which could be because that the overload of MWCNT could not separate well in the matrix, as observed by the agglomeration or the non-uniform phases in Fig. 1. Without the proper dispersion, the agglomerated fillers tend to serve as the defect sites which limited the mechanical performance of the produced composites.

For flexural strength, the neat rHDPE/rPET without nanofiller showed the value at 29.8 MPa. With the addition of Cloisite 10A, the flexural strength reduced to 27.9 MPa at 1 phr and 24.1 MPa at 2 phr, and increased to 28.2 MPa and 30.7 MPa at 4 phr and 6 phr, respectively. For the MWCNT system, the flexural strength showed a similar trend as Cloisite 10A where the results were lower at the lower MWCNT content, i.e. 27.8 MPa (1 phr), 26.6 MPa (2 phr), 25.9 MPa (4 phr) and 28.8 MPa (6 phr). These results are inconsistent with the tensile strength because of different load working modes in both tensile and flexural testings where the failures are result of one-direction stress (extension), and a combined extension-compression at the convex and concave sides of the specimen, respectively. In addition, filler alignment is the major contribution to the flexural strength [30]. Therefore, it is believed that at the lower content of nanofillers, the filler is insufficient to uniaxially align and thus led to weaker flexural strength. At higher nanofiller content, the improvement of flexural strength was due to the better alignment of nanofillers and the restriction of frictional slippage of the filler-matrix interface against applied load [31].

Table 1 - Effect of nanofiller type and loadings on tensile strength and flexural strength of rHDPE/rPET composites

	Loading of nanofillers (phr)									
	Cloisite 10A					MWCNT				
	0	1	2	4	6	0	1	2	4	6
Tensile Strength (MPa)	10.9	13.9	13.5	11.5	10.8	10.9	13.5	13.9	14.5	13.8
Flexural Strength (MPa)	29.8	27.9	24.1	28.2	30.7	29.8	27.8	26.6	25.9	28.8

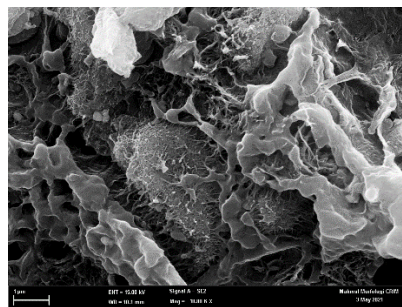


Fig. 1 - SEM micrograph for 6 phr MWCNT nanocomposite

3.2 Thermal Properties

Fig. 2 shows thermogravimetric analysis (TGA) curves of rHDPE/rPET, rHDPE/rPET/Cloisite 10A and rHDPE/rPET/MWCNT. Table 2 lists the TGA results of the three investigated samples. The TGA curves showed that both rHDPE/rPET/Cloisite 10A and rHDPE/rPET/MWCNT nanocomposite samples experienced a significant weight loss through the degradation process in one step. From the observation of the TGA curves, the addition of nanofillers into rHDPE/rPET nanocomposites shifted the TGA curves to the right side which was located at higher temperature values. The right shiftment of the mass loss curves indicates the improved thermal stability of the composite [32]. For the addition of Cloisite 10A at the optimal content of 1 phr, it showed higher onset temperature and maximum decomposition temperature at 391.9 °C and 455.9 °C, respectively, as compared to the neat blend. After decomposition at 500 °C, the percentage of residue for Cloisite 10A nanocomposite was 10.1 %, which was about double of the neat blend. This

phenomenon can support the increased thermal stability with the presence of Cloisite 10A layer that reduces the permeability of volatile degradation on a material and improves thermal insulation [33].

Similar observation was shown for the MWCNT nanocomposite with higher onset temperature of 396.7 °C and maximum decomposition temperature of 460.7 °C than that of the neat rHDPE/rPET blend (384.9 °C and 449.4 °C, respectively). The possible reason for the enhanced thermal stability is the barrier effect of the good-dispersed MWCNT which could avoid the transportation of the combusted products [34]. With the presence of MWCNT, the number of residues remaining after 600 °C increased to 15.1%, which is contributed by the carbon yield of MWCNT. The improved thermal stability of MWCNT nanocomposite is because of the physical barriers action of MWCNT to obstruct the transport of volatile decomposed products during thermal decomposition of polymer nanocomposite [35].

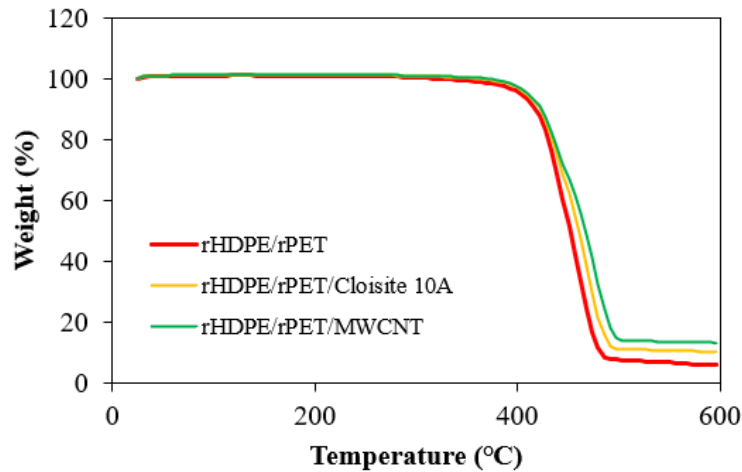


Fig. 2 - TGA curves of rHDPE/rPET, and rHDPE/rPET with Cloisite 10A (1 phr) and MWCNT (4 phr)

Table 2 - Data obtained from TGA curves of rHDPE/rPET without and with 4 phr MWCNT

Sample	Onset temperature (°C)	Maximum degradation temperature (°C)	Residue (%)
rHDPE/rPET	384.9	449.4	5.8
rHDPE/rPET/Cloisite 10A	391.9	455.9	10.1
rHDPE/rPET/MWCNT	396.7	460.7	15.1

3.3 Percentages of Water Absorbed

Fig. 3 shows the percentages of water absorption of rHDPE/rPET/Cloisite 10A and rHDPE/rPET/MWCNT composites with different loading after 2 and 24 hours' immersion in distilled water. The addition of 1 phr is shown to reduce the water absorption, while the higher content of Cloisite 10A for 2, 4 and 6 phr has increased the water absorption of rHDPE/rPET/Cloisite 10A nanocomposite compared to 0 and 1 phr. In fact, natural Cloisite 10A has hydrophilic properties and naturally tends to absorb water [36]. The lower water absorption at 1 phr Cloisite 10A can be attributed to a good dispersion of Cloisite 10A on the low content of Cloisite 10A nanoparticles and layer of silicate arranged in a polymer matrix [37]. Meanwhile, higher content of Cloisite 10A particles will cause to agglomeration and existence of a loose structure that allow the free spread of water molecules into the composite, as in agreement with the results of the study by Abd El-Fattah and Abd ElKader [38].

Another observation was shown that the percentages of water uptake were in different samples with different MWCNT loadings. For the water absorption at 4 phr of MWCNT, the percentage of water absorbed were decreased to 0.05 % for 2 hours and 0.06 % for 24 hours, as compared to neat thermoplastic blend. These observation indicated that the addition of MWCNT in a small amount has improved the water absorption stability [39]. However, incorporation of 6 phr of MWCNT, the percentage of water absorption of composites increased to about 0.09 % at 2 hours while 0.10 % for 24 hours. In this case, the increment in water absorption could be related to the existence of hydrophilic groups, i.e. -OH and -COOH, on the MWCNT [40]. When the immersion period increased from 2 to 24 hours, all the rHDPE/rPET/MWCNT nanocomposites were become physically unstable as indicated by the increased water uptake up to ~ 0.06-0.10 %. This increase is actually low where it is negligible and expected for hydrophobic polymers such as polyethylene.

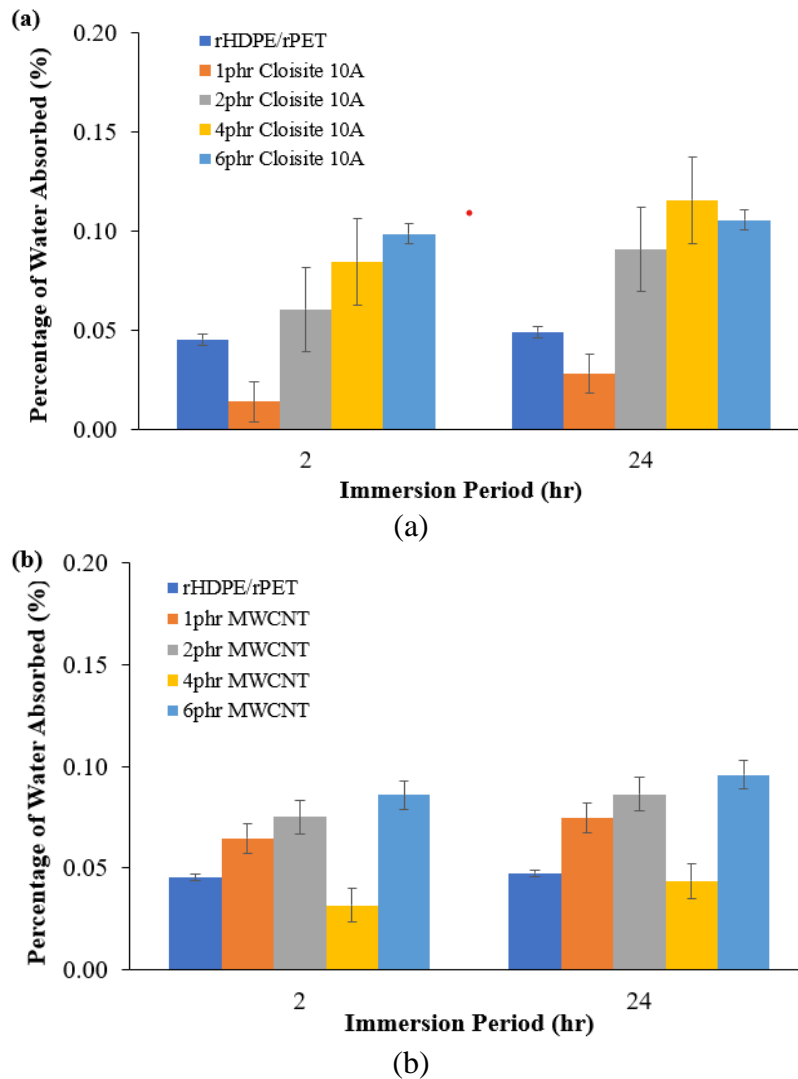


Fig. 3 - The percentage of water absorbed of the rHDPE/rPET composites filled with (a) Cloisite 10A and; (b) MWCNT after 2 h and 24 hrs of immersion in distilled water

4. Conclusion

The rHDPE/rPET nanocomposites containing Cloisite 10A and MWCNT as reinforcement were prepared via melt blending. It can be concluded that tensile strength was optimum at 1 phr of Cloisite 10A and 4 phr of MWCNT loading. In the presence of Cloisite 10A and MWCNT nanofillers, the thermal stability of nanocomposites was enhanced. The improvement in water absorption resistance of nanocomposites was shown only at the optimum loading of each nanofiller. The findings showed the useful application of nanofiller with recycled polymers in producing new composite materials for various field of industry.

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