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Development of Graphene Oxide/ Polyethylene to enhance mechanical properties through melt mixing technique

M A Yusof¹, N H Nor Rahman¹, I Izirwan^{2*}, M S Z Mat Desa¹ and N F Ab Ghani¹

¹ Faculty of Chemical and Process Engineering Technology, Universiti Malaysia Pahang, 26300 Gambang, Pahang, Malaysia

² Faculty of Industrial Sciences and Technology, College of Computing and Applied Sciences, Universiti Malaysia Pahang, 26300 Gambang, Pahang, Malaysia

*Corresponding author: izirwan@ump.edu.my

Abstract. Graphene Oxide (GO) was added to Low-Density Polyethylene (LDPE) to test the mechanical properties of the compound. Compared to other types of polyethylene, LDPE provides a good balance of mechanical properties and processability, which leads to its being chosen as the research's matrix material. This experiment aims to prepare LDPE/GO through melt mixing technique and analyse mechanical properties of tensile strength/tensile elongation of LDPE/GO composite based on GO concentration (0, 0.5, 1.0 and 1.5 wt%). The compounding of LDPE/GO was blended using a five-stage twin-screw extruder under specific conditions before going through the moulding process at melting temperature. The composites were cut according to ASTM D638 specimen dimension. The tensile properties of LDPE composites were filled separately with different weight fractions of GO at a 50 mm/min rate. It was found that the values of Young's modulus of the composites increased, but the values of the tensile elongation at break decreased with increasing the GO weight fraction. The relatively big interfacial area and excellent interfacial adhesion between the matrix and the GO may be responsible for the composites' reinforcement. This study provided a basis for further development of GO-reinforced LDPE composites with desirable mechanical performance and good damage behaviour.

1. Introduction

In the automotive industry, polymer composites are becoming increasingly popular due to their lightweight and fuel-efficient properties. Some thermoplastics, particularly polyethylene, offer low cost and ease of processing through an extrusion and compression moulding. The mechanical strength of polymer composites can be increased by adding fillers, which can produce superior barrier characteristics [1]. The qualities of composite material are determined not only by the properties of its elements (matrix and filler) but also by the properties of the interface/interphase between them [2].

Polyethylene (PE) is one of the most important commodity polymers. PE is divided into groups based on branching and density. The extent and type of branching, crystal structure, and molecular weight all substantially impact the material's mechanical properties. Due to its superior processing flexibility compared to high-density PE and its excellent stiffness and strength balance, low-density polyethylene (LDPE) has had the fastest growth within the PE family [3].

Due to its unique properties, like high mechanical strength, graphene oxide (GO), a two-dimensional, single layer of sp^2 hybridised carbon atoms, has attracted the attention of researchers. Graphene is ideally



suited to be incorporated into various functional materials due to its excellent characteristics and ease of manufacturing and functionalization [4]. Additionally, because they have a better surface-to-volume ratio than carbon nanotubes (CNTs) and are less expensive, graphene and graphene polymer nanocomposites are of great interest to industry and academia [5].

In this study, LDPE/GO was undergoing an extrusion process. The mechanical energy produced by the barrel's heaters and turning screws eventually melted the material. It changed into a pipe that hardens during cooling. The polymer composite was cut to become pellets before undergoing the compression moulding process which a preheated polymer was placed into an open, heated mould cavity. The moulded LDPE/GO was cut into a dumbbell shape according to ASTM638 standard test method and analyzed in mechanical properties, including tensile strength and tensile elongation. Strength of materials, also called mechanics of materials, deals with the behaviour of solid objects subject to stresses and strains. Therefore, it is important to achieve good dispersion of filler material to enhance the mechanical properties. This work attempts to prepare LDPE/GO through melt mixing technique and analyze mechanical properties of the tensile strength/tensile elongation of LDPE/GO composite.

2. Material and methods

LDPE in solid form was supplied by Petlin (Malaysia) Sdn Bhd. In order to protect LDPE from degradation during the high temperature processing, 0.4 g of phenolic stabilizer was added for each 1 kg of LDPE. GO was purchased from XG Science. GO is easily dispersed in organic solvents, water, and other matrixes. This is a significant advantage when using the material to improve the mechanical properties of polymer or ceramic matrixes. Figure 1 shows the flowchart of overall process LDPE/GO [6].

The LDPE/GO was pre-mixed in the sealed containers and manually shaken. The mixing composites were fed into PRISM Eurolab 16 twin-screw extruder, with the barrel temperature profile used for compounding for all blends set at 110-170 °C, and the screw rotation was set at 50 rpm. The nanocomposites were pelletized by using pelletizer. GO contents in nanocomposites ranging from 0 to 1.5 wt% were developed. Afterwards, the composites were moulded using a Hot and Cold Moulding Press machine at 110 °C. LDPE/GO composites were cut with dumbbell shapes according to ASTM D638. The nanocomposites specimens were tested with a Universal tensile testing machine under ambient conditions to analyze mechanical properties.

3. Result and discussion

3.1. Relationship between tensile stress and tensile strain

The relationship between the tensile stress and strain of LDPE/GO composites is shown in Figure 2. From the graph, the value of tensile stress increases but tensile strain decreases with increasing amount of GO concentration. The LDPE/GO composite with the GO content of 1.5 wt% had the maximum tensile stress value of 10.8 N/mm², which is extremely higher than those composite with less amount of concentration. Compared to the tensile strain, pure LDPE which is 0 wt% of GO has the maximum value of elongation at break of 16.7%. These results were similar with other GO reinforced polymer composites.

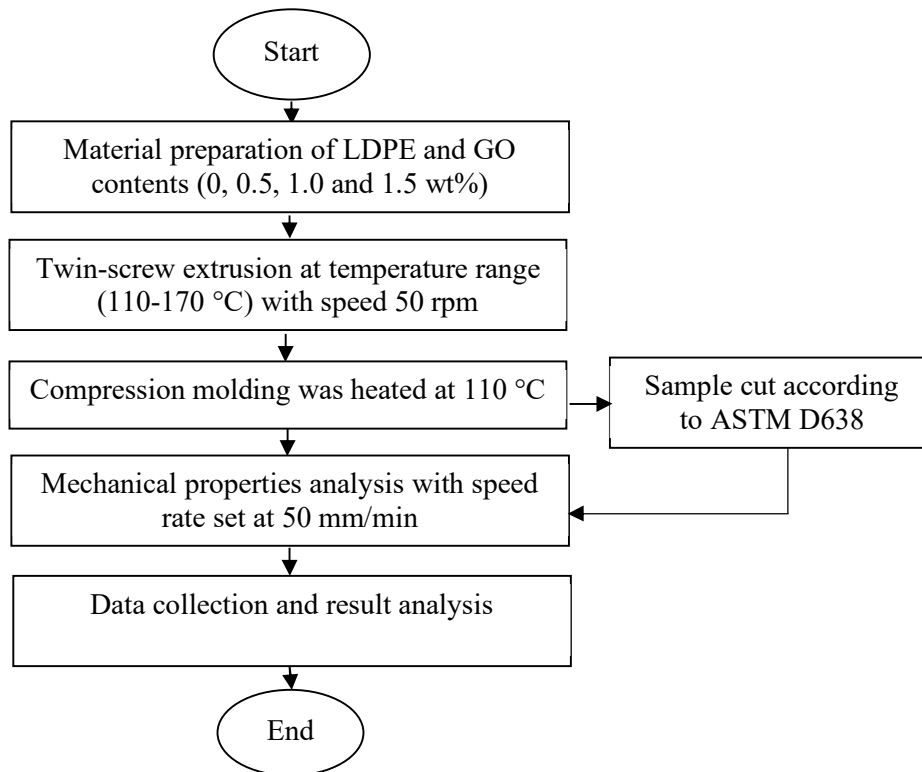


Figure 1. Overall process of LDPE/GO composites.

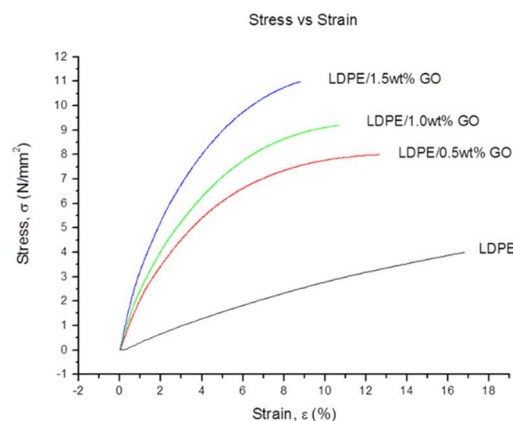


Figure 2. Relationship between tensile stress and tensile strain of LDPE/GO composites.

The strong interfacial adhesion caused by H-bonding between the GO and LDPE matrix and the molecular-level dispersion of GO in the LDPE matrix were cited as the causes of this [7]. The applied stress was anticipated to transfer from the matrix to the GO layers, improving their mechanical characteristics due to the high surface area of the GO nanolayers in the LDPE matrix. However, as the GO content was increased to 1.5 wt%, the composite's tensile strain dropped. These findings are comparable to the polymer reinforced fibre composite system. The typical stress-strain curve additionally demonstrates that the composite's tensile modulus increased at higher elongation compared

to the lower region. This indicates that the material's deformation behaviour becomes more elasto-plastic in higher elongation regions with increased GO loading.

3.2. Dependence of Young's modulus on GO content

The Young's modulus of the LDPE/GO composites is dependent on the GO concentration, as shown in Figure 3. The value of Young's modulus was obtained from slope of the stress-strain curve. As shown in the graph, Young's modulus increases drastically with an increase of GO concentration. The value of Young's modulus at 1.5 wt% GO content was reported the highest value of 1.1 N/mm² but pure LDPE has the lowest value of 0.23 N/mm².

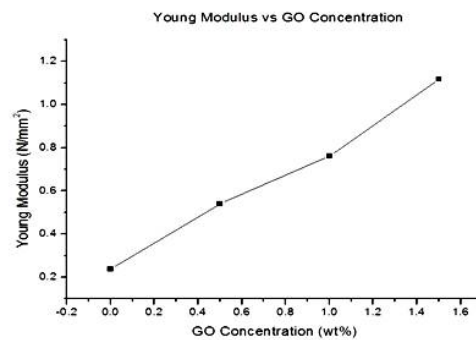


Figure 3. Dependence of Young's modulus on the GO concentration.

Figure 4 shows the idealized filler-reinforced composite material. Several physical crosslinking points between the filler and the matrix occur when GO particles are loaded to LDPE, acting as the skeleton in the matrix and limiting the mobility of molecular chains to increase the stiffness of the composite systems. Inclusions play a function in heterogeneous nucleation for crystalline polymers, increasing their degree of crystallinity or altering their crystal type, improving the stiffness of the composite systems in this situation. The stiffness of polymer composites will increase with the number of inclusions when the filler particles are evenly distributed throughout the matrix. The larger the number of particles under the same filler volume fraction, the smaller the particle size.

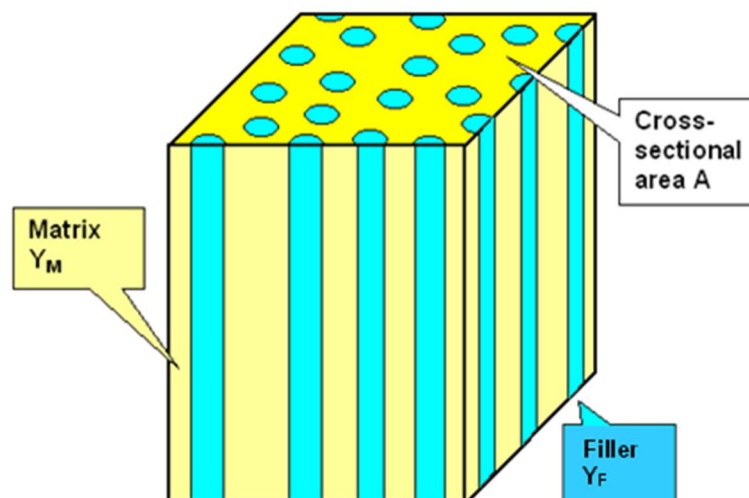


Figure 4. Idealized filler-reinforced composite material.

3.3. Dependence of tensile elongation at break on GNPs content

An important factor in determining a material's ductility (or tensile fracture toughness) is tensile elongation at break. The length ratio changed to the length before the test specimen was fractured. It describes a material's capability to withstand shape changes without developing cracks. Figure 5 shows the dependence of tensile elongation at break on various GO concentration. The value of tensile elongation at break has uniform decrease with an increase of GO concentration. Pure LDPE shows the highest value at 16.9% but 1.5 wt% GO concentration shows the lowest of with 9.5%.

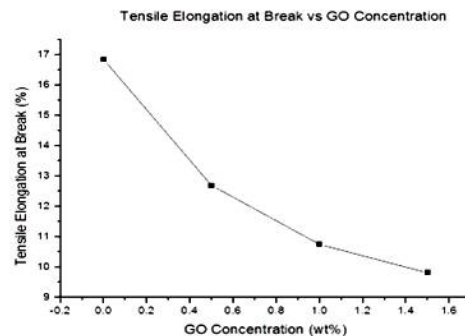


Figure 5. Dependence of tensile elongation at break on the GO concentration.

The reinforcing and toughening effects were improved by increasing the filler-specific surface area while maintaining the same interfacial adhesion status. Although less strong, more elastic or flexible material breaks at higher deformation. In this situation, the composite may become significantly weaker due to inadequate filler-matrix adhesion at the interface, and fillers may function more like matrix defects than reinforcements. The elongation at break will be higher because the strength will be lower in this situation.

Figure 6 presents the picture of tensile fractured specimen of LDPE/GO composite. It can be seen that the specimen has decrease in length with the increase of GO concentration. It shows that 1.5 wt% GO is brittle and easily to break but 0 wt% is ductile and able to be deformed without losing toughness. The tensile elongation at break the specimen may be increased due to these microcracks' ability to absorb tensile fracture energy and improve the ductility of PP composites.



Figure 6. Tensile fractures specimen of LDPE/GO composite.

4. Conclusion and recommendation

The content of GO in the filled LDPE composites affected their tensile properties. The findings showed that while the tensile elongation at break values decreased with increasing the GO weight fraction, the

composites' Young's modulus values increased. Because the inclusions are in the form of a thin sheet, the interfacial area between the GO and the matrix is relatively large. The strong interfacial adhesion and the relatively large interfacial area between the GO and the LDPE matrix may be responsible for the reinforcement of the composites.

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