Evaluation on Mechanical Properties of Silicon Dioxide (SiO_2) Biocomposite

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***Abstract* — Linear low density polyethylene (LLDPE) was blended with filler SiO_2 at a ratio up to 6% by extrusion to study the mechanical properties of the product. The Maleic Anhydride (MAH) was grafted into LLDPE-SiO_2 to determine the possible reaction. Dicumyl peroxide used as an initiator. The LLDPE-g-MAH composite filled with SiO_2 were extruded and melt-pressed in order to prepare samples for mechanical properties test. It showed 4% wt. of SiO_2 have the highest tensile strength and Young's Modulus while 6% wt of SiO_2 is the highest ductility. From this study, it is found that SiO_2 and MA can improve the strength of polymer but must be at the right amount.**

Keywords— Linear low density polyethylene (LLDPE), silicon dioxide (SiO_2), maleic anhydride, extrusion, mechanical properties

I. INTRODUCTION

Polymer is the most widely used materials in chemical industries. The development of polymers from renewable resources has received more attention in recent years, in particular due to the application of polymer composites as engineering materials has become state of art. It is possible to synthesize both thermoplastic and thermosetting polymer from bio-based chemical which usually derived from plants. In addition, by blending with bio-derived polymers can reduce the environmental impact of synthetic materials while utilizing abundant natural resources.

Biocomposites are the combination of natural fibre or biofibre with polymer matrix. Natural fibres acts as the reinforcement or filler that provides strength where the properties of the composite are controlled by properties of the fillers. Meanwhile, the matrix holds the fibres together and to transfer externally loads to the reinforcement. Fillers are employed to improve the desire properties of the polymer or simply reduce the cost. It also protects the reinforcement from environmental and mechanical damage. The advantages of bio-based products such as light weight and high specific strength make them dominant in various applications including automotive parts. Recent study has been focuses on the ability of it to improve the mechanical properties of various of polyolefins.

Silicon dioxide (SiO_2) is one of the most common inorganic fillers that have been used in polymer. Other than that, TiO_2, ZnO and $CaCO_3$ are one of the mostly used inorganic particles. Naturally, silica bodies in plants are essential for growth, mechanical strength, fungal pathogens and outside heat (Hanipah, Mohammed, & Baharuddin, 2016). For biocomposts degradation process, silica bodies provide protection by restricting microbial activities on the fibre's surface (Omar et al., 2016). Silicon dioxide (SiO_2) which also known as silica is an inorganic amorphous oxide formed by polymerization processes within plants. Some researchers suggested that the inclusion of silica bodies in composites prevent a sliding motion between filler and matrix. In contrast, by excluding the silica bodies can increase and improved contact area between the filler and matrix. Therefore, this study is trying to mimic the contain of silicon dioxide (SiO_2) in plants.

In order to blend the filler onto the polymer, it need to undergo a process called extrusion. Extrusion is a process where the pressure will be applied as force to the plastic or molten material to make it flow to a shaped die (Verbeek & S. H. Hanipah, 2009). The polymer which at certain cross section will be forced to flow through a die or smaller cross-sectional area to form a new product at new cross section. Extruder machine will be used for the extrusion process. Extruder machine that have been used to form thermoplastics products by using a uniform cross-section such as pipe, hose, wire and cable. The section of the extruder can be divided into three main section which are melting, compression and metering section. Depending on the number of screws in the barrel, extruder is classified into two types which is single screw or twin screws extruder. Grafting process also have been conducted in this study. Grafting can be defined as process of combining a new monomer onto polymer chain by bonding it using covalent bond (Jin-hua, Guo-qin, Huang, & Lin-jian, 2012). The surface of polyethylene will be modified in order to add maleic anhydride onto it. Grafting help to control, structure and determine maximum potential of the polymer.

II. METHODOLOGY

A. Materials

Linear low-density polyethylene (LLDPE) was obtained in powder form from R&M Chemicals that was used directly as the raw material for this study. Silicon dioxide (SiO2) was obtained from R&M Chemicals which is chemically pure and has molecular weight of 60.08g/mole to be used as a filler. Maleic anhydride was obtained from Merck KGsA with molecular weight of 98.02g/mole and it will be grafted with LLDPE-SiO2. Dicumyl Peroxide with molecular weight of 270.37g/mole was obtained from R&M Chemicals. The function of dicumyl peroxide is as an initiator because of its effectiveness to introduce long chain branches in LLDPE.

Table 1: Summary of Materials Used

Material	Supplier	Density (g/cm^3)	Melting point (°C)
Linear low-density polyethylene (LLDPE)	R&M Chemicals	0.918	115 - 130
Maleic Anhydride (MAH)	Merck Millipore Corporation	1.32	51 – 53
Dicumyl Peroxide (DCP)	Sigma-Aldrich	1.56	39 – 41
Silicon Dioxide (SiO_2)	R&M Chemicals	2.21	1650 (±50)

B. Preparation of LLDPE-g-MAH/SiO_2 Biocomposite

The total weight of sample of solid mixture of LLDPE, dicumyl peroxide, maleic anhydride and silicon dioxide was 100g. The solid mixture was prepared in three different weight ratio which were 93:2:4:1, 91:4:4:1 and 89:6:4:1 of LLDPE, SiO_2, dicumyl peroxide and maleic anhydride respectively. The reaction product was extruded using Thermo HAAKEE twin-screw maintained at 170°C and speed screw of 20 rpm. The output was cooled in the open air. The extruded product was cut down to small pieces with the help of scissors and crushed by using crusher. The solid particles were formed into sheets by melt-pressed at 170°C under a pressure of 2000kPa for 20mins. The cooled sheets were cut into specific shapes for measuring tensile strength.

Table 2: Formulation of all sample of LLDPE-g-MAH/SiO2 biocomposite

Sample	Weight percentage (%)			
	LLDPE	MAH	DCP	SiO_2
1	93	1	4	2
2	91	1	4	4
3	89	1	4	6

C. Specimen Testing

The tensile tests were conducted based on American Standard Test Method (ASTM D638). The dogbone shaped specimens with a gauge length, thickness and width of 30mm, 6mm and 3mm were adhere with 1mm thick of sandpaper at the biocomposite end-tabs. The dimensions of the specimen are illustrated in Figure 1(a). At least three specimens were tested for each system. Prior testing, the actual width and thickness of the coupon at the gauge length was measure at 3 different points using a calliper. A Tinius Olsen H50KT universal testing machine with wedge type grips as shown in Figure 1 (b) was used for the tensile testing at a crosshead speed of 5 mm/min. A 25 kN load cell and a 25 mm gauge length clip-on extensometer were used to record the applied load and elongation data. These data were logged to a computer for analysis. The tensile properties such as elastic modulus, strength and failure strain were determine based on the standard.

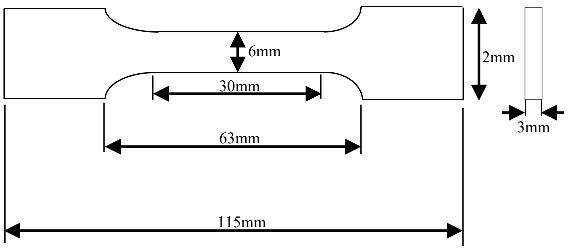

Figure 1(a): Illustrated of dimensions of the dogbone shape specimen

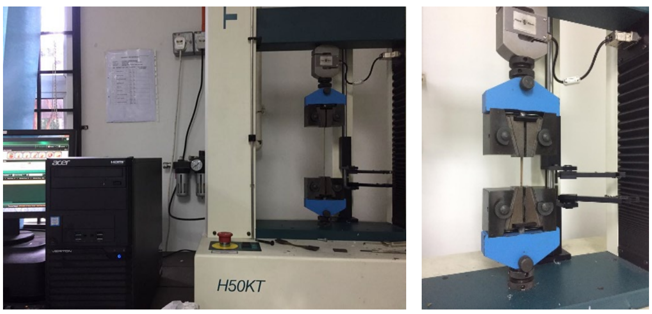

Figure 1(b): A Tinius Olsen universal testing machine with wedge type grips for tensile test

III. RESULTS AND DISCUSSION

The effects of SiO_2 filler content on the Mechanical Properties

Tensile Strength

It is to be noted that the testing done for each ratio is repeated for 3 times for tensile strength in order to get accurate results. The data for each ratio is then taken at average. The term tensile strength refers to the amount of tensile (stretching) stress a material can withstand before breaking or failing. Silicon dioxide (SiO_2) biocomposites loaded with various amount of silicon dioxide (SiO_2) were successfully fabricated by melt blending and their tensile strength were examined and reported. The amount of silicon dioxide (SiO_2) loaded was 2wt%, 4wt% and 6wt% with the presence of 1wt% of MA and 4wt% of DCP to improve the interfacial adhesion and reinforcement of the hydrophilic natural fibres with the hydrophobic polymers. The study was repeated with different weight percentages of LLDPE (89wt%, 91wt% and 93wt%).

Figure 2 shows the tensile strength of notched specimen optimum weight ratio of silicon dioxide (SiO_2) was 4wt% in which the tensile strength had its maximum value at 31.9 MPa whereas further increase in silicon dioxide (SiO_2) content results in decrease in the value of tensile strength. This is because filler particles act as barrier in transferring stress from one point to another. According to (Mallakpour & Naghdi, 2018), in the higher contents of silicon dioxide (SiO_2) will reduced the tensile strength as a result of intermolecular disturbance. Other than that, as the filler content increases, the bonding surface area increases and hence bonding strength decreases (Singh, Batra, & Chand, 2015). Due to the insufficient amount of bonding between three different constituents, the loads may not effectively be transferred from one end to another and hence there is reduction in tensile strength of the silicon dioxide (SiO_2) biocomposite. The yield strength of the LLDPE/SiO_2 composites decreases with the increment of SiO_2 filler content is the result of the cross-linking effect among the polymer chain. The interaction of the chain of polymer and the filler lead to enhance strength at localized regions ((El-Tonsy, Fouda, Oraby, Felfel, & El-Henawey, 2016).

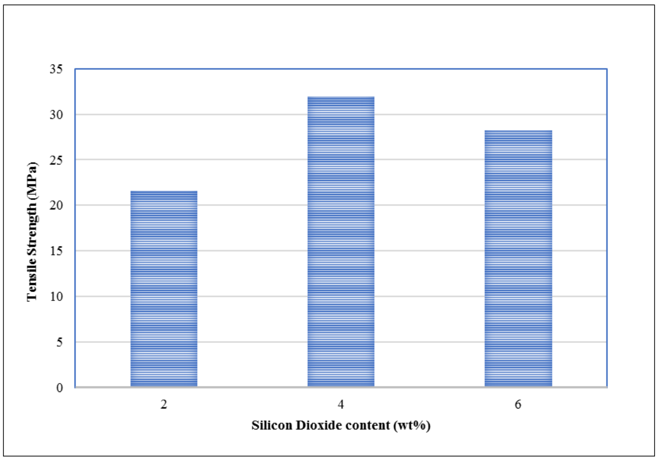

Figure 2: Tensile strength at different silicon dioxide (SiO_2) loading

Ductility (Elongation at break)

Ductility is a measure of the degree of plastic deformation that has been sustained at facture. Ductility may be expressed quantitatively as either percent elongation or as percent reduction area. In this study, the ductility of the polymer will be presents as percentage of elongation at break. The effect of different silicon dioxide (SiO_2) loading on the ductility of the biocomposite is shown in Figure 3.

From Figure 3, the ductility of the samples increased with the silicon dioxide (SiO_2) content and it reached the maximum value at 6% which is 2043%. As silicon dioxide (SiO_2) content is more than 2wt%, the elongation at break increases with increasing silicon dioxide (SiO_2) amounts.

The degradation was further supported by a gradual colour change after each extrusion. It was observed that with the increasing amount of silicon dioxide (SiO_2) specimens were getting darker, taken as indicative of greater degradation. Figure 4 shows the colour change due to the degradation of polymer by using DCP R&M Chemicals as the peroxide initiator. Polymers are oxidized, partially degraded and cross-linked when processed in an air atmosphere, reducing the mechanical properties and the service-ability of the polymer (Verbeek & S. H. Hanipah, 2009)

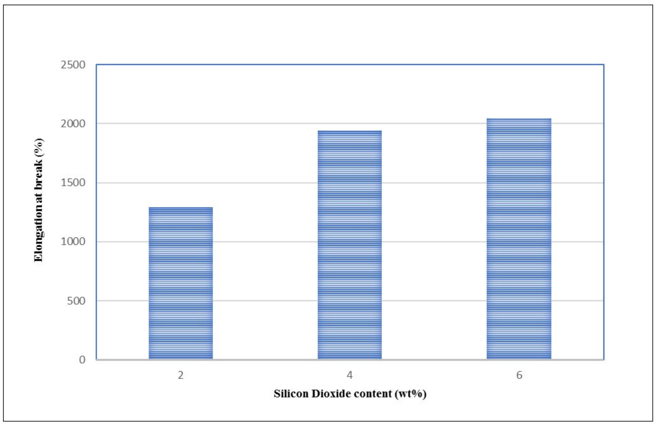

Figure 3: Elongation at break at different silicon dioxide (SiO_2) loading

Figure 4: Colour changes of the polymer with an increment of silicon dioxide (SiO_2) content

Young's Modulus

The Young's modulus of polymer is dependent on molecular mass. At high molecular mass, more force is required to align polymer chains because of chain entanglement. Degradation and cross-linking may therefore have opposing effects on modulus, since degradation reduces molecular mass and cross-linking reduces mobility.

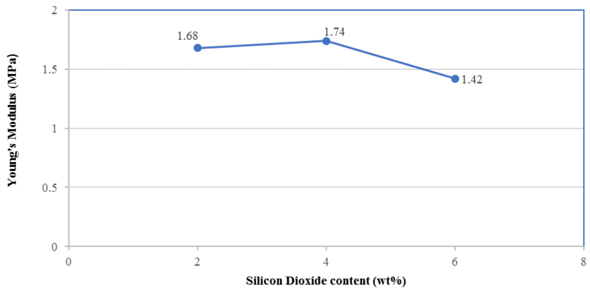

Figure 5: Young's Modulus at at different silicon dioxide (SiO_2) loading

In the case of Young's modulus, its variation for different silicon dioxide (SiO_2) loading is shown in Figure 5. An increase in modulus with filler loading is observed for LLDPE-g-MAH/SiO2 biocomposite, which reaches up to 1.74 MPa and indicates decreased stiffness due to the rising filler content. Modulus is typically less affected by a reduction in chain length, especially above the critical chain length of the polymer in question.

IV. Conclusion

It was concluded that the objectives were successfully achieved based on the results and all the samples were well prepared. Besides, most of the analysis in this study are following the theory. Most of the analysis showed that increasing the silicon dioxide (SiO_2) and maleic anhydride (MAH) content can improve mechanical properties of the LLDPE but must be at the right amount or otherwise it will decrease.

Analyzing the experimental results, mechanical tests of LLDPE-g-MAH/SiO2 biocomposite specimens were tested by conducting a tensile test. The results showed that increase in silicon dioxide (SiO_2) content up to 6wt% will decrease in the value of tensile strength and Young's modulus. However, the elongation at break increases with increasing of silicon dioxide (SiO_2) content.

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References

[1] ASTM D638. (2015). Standard test method for tensile properties of plastics. *ASTM International*, *08*, 46–58. https://doi.org/10.1520/D0638-14.1

[2] El-Tonsy, M. M., Fouda, I. M., Oraby, A. H., Felfel, R. M., & El-Henawey, M. I. (2016). Dependence of physical properties of linear low-density polyethylene on the silicon dioxide filler size. *Journal of Thermoplastic Composite Materials*, *29*(6), 754–767. https://doi.org/10.1177/0892705714533376

[3] Hanipah, S. H., Mohammed, M. A. P., & Baharuddin, A. S. (2016). Non-linear mechanical behaviour and bio-composite modelling of oil palm mesocarp fibres. *Composite Interfaces*, *23*(1), 37–49. https://doi.org/10.1080/09276440.2016.1091681

[4] Jin-hua, T., Guo-qin, L., Huang, C., & Lin-jian, S. (2012). Mechanical properties and thermal behaviour of LLDPE/MWNTs nanocomposites. *Materials Research*, *15*(6), 1050–1056. https://doi.org/10.1590/s1516-14392012005000122

[5] Mallakpour, S., & Naghdi, M. (2018). *Polymer/SiO2 nanocomposites: Production and applications*. *Progress in Materials Science* (Vol. 97). https://doi.org/10.1016/j.pmatsci.2018.04.002

[6] Omar, F. N., Hanipah, S. H., Xiang, L. Y., Mohammed, M. A. P., Baharuddin, A. S., & Abdullah, J. (2016). Micromechanical modelling of oil palm empty fruit bunch fibres containing silica bodies. *Journal of the Mechanical Behavior of Biomedical Materials*. https://doi.org/10.1016/j.jmbbm.2016.04.043

[7] Singh, G., Batra, N. K., & Chand, R. (2015). Effect of Silicon Dioxide (Sio2) On Physical and Mechanical Properties of Vinyl Ester Composite, *2*(7), 881–885.

[8] Verbeek, C. J. R., & S. H. Hanipah. (2009). Grafting Itaconic Anhydride onto Polyethylene Using Extrusion. *Polymer*, (November 2009). https://doi.org/10.1002/app