

Blending of LLDPE with SiO₂ via Extrusion

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Abstract—Environmental pollution problems are becoming severe nowadays especially at the dumpsite. Thus it is imperative to develop new materials such as producing plastic that is ecological friendly to environment yet it has been found that there are limited research on the blending of LLDPE with inorganic material such as SiO₂. This work is done to investigate the synthesis of LLDPE/SiO₂ composite at different ratio and to investigate the characterization of composite by using FTIR and the mechanical properties of tensile and elongation at break. The composite is produced through extrusion process by using twin-screw extruder and being analyzed by using FTIR and the universal tester machine. The result demonstrated that the composite with lowest SiO₂ filler content which is 4% has the best mechanical properties. The production of this composite will be useful to the industry as the mechanical properties of the composites has been enhanced hence producing a better plastic materials.

Keywords— Extrusion, LLDPE, Plastic, Mechanical properties, SiO₂

I. INTRODUCTION

Biocomposites are made up of from an individual polymer matrix and fibre reinforcement component which remain bonded together by physical or chemical interactions, but retain their individual physical or chemical identities. Natural fibres are used as reinforcement that provides strength and rigidity imbedded in matrix polymers (Moliner et al., 2018). Matrix polymers are generally thermoplastic polymers derived from petroleum such as polypropylene (PP), low-density polyethylene (LDPE), high-density polyethylene (HDPE), and also polystyrene (PS). But these polymers are not easily degrading in natural environment thus become the problems to environment. One of the powerful method in producing new materials are by blending polymer with inorganic materials that is called polymer composites or filled polymers (Chaichana et al., 2007). It is known that blending is important to obtain polymer materials with best properties and also improve their processing ability and also their costs. Based on previous research in this field, most polymer are not in a homogenous system anymore but in a state, which is multiphase complex systems that are obtained through blending process.

Polymers that had been reinforced by the inorganic fillers have attracts industrial interest because of the modification on the important mechanical and physical properties (Urrego Yepes et al., 2017). Silicon Dioxide (SiO₂) is recognized as an inert diluent and is used as an enhancing agent for thermoplastic polymers (Wei et al., 2006). This filler has expanded the rangeability of applications in the usage of polymers. It is known that LLDPE has taken much interest in the manufacture of plastics. These plastics can be derived to be made as pipes, food wrapper and 9so forth. China has taken the

initiative to replace their conventional plastic micro-irrigation pipes with LLDPE plastic for the purpose of saving the water in the rural area, specifically in the northwest China (Liu *et al.*, 2005). Hence, it is vital to design a high performance compound to be used as the structure of the micro-irrigating pipes. However, LLDPE comes with the shortcomings such as poor flexibility and processability. The addition of some inorganic particles such as nano-SiO₂ has been found that it is able to enhance the physical and mechanical properties, processability, resistance to environmental stress cracking and aging behaviour of various polymers (Liu *et al.*, 2005).

The linear low-density polyethylene (LLDPE) is an attractive polymer because of its excellence in mechanical property, good resistance for chemicals and also high thermal stability. LLDPE is a copolymer of ethylene and another longer olefin. LLDPE consists of long chains of ethylene and α -alkene (Tokiwa et al., 2009). Hence, it has been widely used for several applications, especially in film production and packaging industries. LLDPE provides better processability, lower manufacturing costs, and improved mechanical properties such as toughness for the packaging applications. LDPE were blended into form of LLDPE where the film has more improved properties such as more tensile strength, impact strength, resistance to environment unfavourable conditions, flexibility, and more conformability which is much flexible and softer. In order to get more flexible and extra in strength, LLDPE is used for pond liners or blended into other film. The tearing and puncturing of films are prevented by the absorbance of impacts that are needed because of having huge amounts of strength by using LLDPE.

An initiative is taken by blending LLDPE with additives in order to enhance the specific properties of these polymers. The blending of polymer with filler, especially the inorganic material is proven to be one of the most effective method to produce the polymer composites (Jongsomjit *et al.*, 2005).

There are many study has been done on the blending of polymers with filler addition such as LLDPE/LDPE being filled with SiO₂ and TiO₂ in developing the polymer composite yet the not many research can be found on the study of the blending of SiO₂ with LLDPE only (Liu *et al.*, 2005). Many application of composite that can be found nowadays for example in the manufacture of automotive field, sports equipment and implemented in aircraft area. Plastics are one of the example major product from the application of polymer composites. The application of polymer composite also can be premised for the structural use as there are many research that can be found focusing on the mechanical properties. The interest of the research is to make the improvement in stiffness and toughness simultaneously (Sirocic *et al.*, 2009). The mechanical properties mentioned are as such tensile strength and elongation rate at break. However, there are several factors that can affect the stiffness, strength as well as the toughness which are the size of

the particle, range of filler size and so forth (Liu *et al.*, 2005). In this study, the SiO₂ with different percentage of ranging from 2%, 4% and 6% was used as the filler for the LLDPE. The influence of filler percentage on the mechanical properties and the characterization of the LLDPE/SiO₂ composites are investigated. The mechanical testing done for this research according to the standard of ASTM D 638, which are the standard test method for the tensile properties of polymer composite materials.

II. METHODOLOGY

A. Materials

The polymer used in this study was LLDPE with the melting temperature in the range of 120 to 125°C and density of 0.92 kg/cm³. The SiO₂ was used as the filler which has the molar mass of 60.083g/mol and temperature of ignition of 350°C.

B. Preparation of LLDPE/SiO₂ Composite

The LLDPE and SiO₂ were mixed after being weight according to the sample size. The mixture of LLDPE and SiO₂ was then extruded on a Thermo HAAKEE twin-screw extruding machine. Thus, the composite of formed from the extrusion process. The previously extruded mixture was grinded by using a grinder and then the sample of composite was sent for analysis. The analysis done for the sample were FTIR and TGA. Next, the grinded composite of LLDPE/SiO₂ was spread down onto the mould of the hot press for the next step. Thus, a block of LLDPE/SiO₂ composite was formed. Lastly, after undergoing the process sequentially the block of LLDPE/SiO₂ composite was cut by using a cutter. The composite of LLDPE/SiO₂ was prepared with different weight ratio, which were 98:2, 96:4 and 94:6 in order to determine the most ideal composition of LLDPE/SiO₂ blend.

C. Evaluation of the Properties of Composite

Mechanical Properties

The mechanical properties of the LLDPE/SiO₂ composite were tested in this research. The properties that encompasses were the tensile strength and elongation rate at break. These mechanical properties were tested by using the Tinius Olsen H50KT universal testing machine according to the standard of ASTM D638. The tensile measurements were being performed at room temperature approximately about 25°C. Subsequently, the samples of the rectangular were being stretched at the speed of 5 mm min⁻¹ with the contact of the cell load of 1000N and gauge length of 25 mm (Kuriakose *et al.*, 2012). The shape of the samples were in dog-bone which a cut by using the cutter with the measurement 6mm x 25mm x 2.5mm.

FTIR Analysis

FTIR analysis was used to identify the chemical bonds in a molecule by producing an infrared absorption spectrum. It is an analytical instrument used to detect the functional group and characterize the covalent bond available in the raw materials as well as the sample of LLDPE/SiO₂ composite. By using this instrument, the functional groups and the chemical of the silicon dioxide composite can be determined (Motaung & Luyt, 2010). The samples were tested by using Perkin Elmer Spectrum One FTIR spectrometer over a 400–4000cm⁻¹ wavenumber range at a resolution of 4 cm⁻¹, which is available at the instrumentation laboratory at Faculty of Chemical Engineering, UiTM Shah Alam.

III. RESULTS AND DISCUSSION

A. Characterization of LLDPE/SiO₂ Composite

The function of Fourier Transform Infra-Red Spectroscopy (FTIR) analysis is to determine the presence of certain functional groups in a molecule. The Fig. 1 shown below are showing the result of FTIR sample analysis done on the raw materials and the samples of composite of LLDPE/SiO₂ with different ratio. Firstly, it can be observed that the pattern of graph for the LLDPE and SiO₂ can be differentiated at certain band width. The graph for SiO₂ starts to develop a peak from 1470-950 cm⁻¹, whereby C-O bond is detected within the band width. As for LLDPE, there is no pattern of peak in its graph. Hence after the FTIR test, it can be seen that the pattern of the LLDPE/SiO₂ composite follows the trend of both LLDPE and SiO₂ graph. This is because the LLDPE/SiO₂ composites have undergone some changes of properties from the raw materials to a new composite. The FTIR spectrum of the LLDPE/SiO₂ composites have numerous peaks which will determine their origin of the group. The LLDPE/SiO₂ composites that have different ratio are sharing some similar traits as they share the similar peaks. The peaks at 2916 cm⁻¹, 2848 cm⁻¹, 1738cm⁻¹, 1466 cm⁻¹ and 719 cm⁻¹ indicate the presence of polyethylene. Meanwhile, as for the peaks at the range of 1092-1106 cm⁻¹ point out the presence of SiO₂ (Cheng *et al.*, 2011). Subsequently, the Fig.1 demonstrated as the filler SiO₂ increases, the peak at bond C-O are decreasing. This is due formation of the composite that has developed at the bond which interrupt the original spectrum of SiO₂.

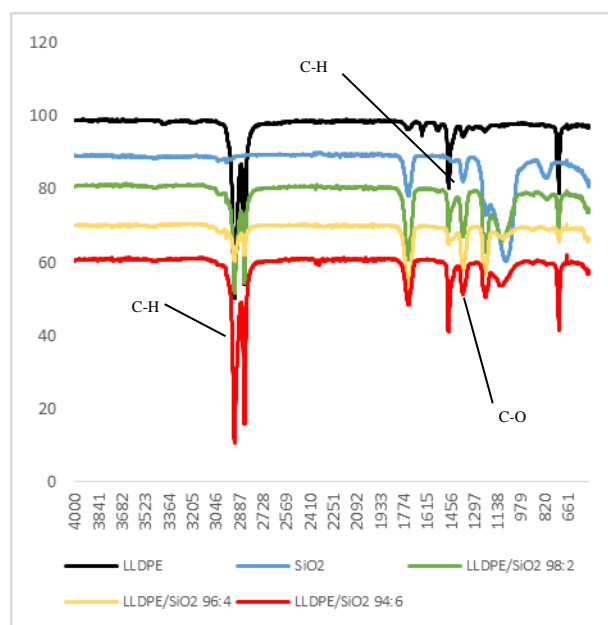


Fig. 1: FTIR spectrum the raw materials and the composites.

B. Effect of the SiO₂ Filler Content on the Mechanical Properties

For the mechanical test, the process done for each ratio is repeated for 5 times for each mechanical properties in order to get accurate results. The data for each ratio is then taken at its average. The stress-stain curve can be obtained from the tensile testing. Fig.2 presents the tensile strength for the composite against percentage of filler SiO₂ by the data at average for each ratio. The Fig.2 demonstrated that among those three different ratio of composite, the composite with 4% of filler SiO₂ records the highest tensile strength value. Tensile strength also commonly referred as yield strength. The elongation at break is interrelated with the tensile strength, which also can be obtained from the

stress-strain graph. According to the Fig.2, it can be observed that the yield strength increases with the increase in SiO₂ filler content which obeys the general findings. But at certain value, it will decrease thus show that the right amount of filler are affecting the yield strength of materials. The previous research suggested that at higher content of SiO₂, the mechanical properties of the composites declines (El-Tonsy *et al.*, 2014). The data obtained is then plotted together with elongation at break is as shown in Fig.3.

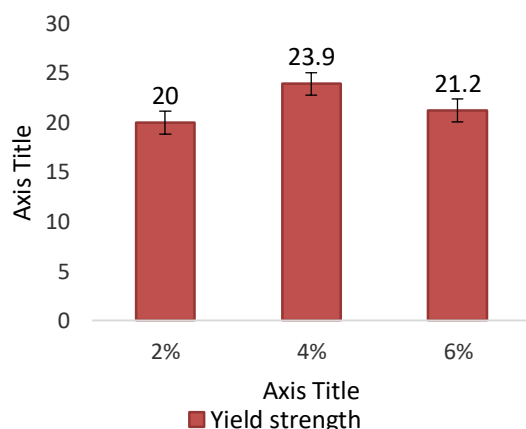


Fig.2: Tensile strength of the LLDPE/SiO₂ composite.

The elongation at break can be found in the stress-strain diagram, where it is the end point or being said at maximum strain. The trend of the graph shows that it increases with 4% filler content percentage which is similar with the yield strength graph pattern. But it decreases as the filler content is at 6%. The factor contributing to this is the capability of the polymer to chemically interact with the filler added. LLDPE, which derived from the polyethylene backbone is classed as a nonpolar polymers. The nonpolar polymers such as polyethylene and polystyrene are being distinguished as nonpolar due to the electronegativity between the elements forming the bond, whereby the electronegativity values of C-H bond is close. Hence, the net polarization does not exist. The polymer that is nonpolar usually have weaker interface especially with inorganic fillers as SiO₂ (El-Tonsy *et al.*, 2014). The yield strength of the LLDPE/SiO₂ composites decreases with the increment of SiO₂ 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 *et al.*, 2014). Hence from this interaction, the growth of cracks can be underdeveloped. This also explains the percentage of elongation at break is higher as if it is suitable with the percentage of filler content.

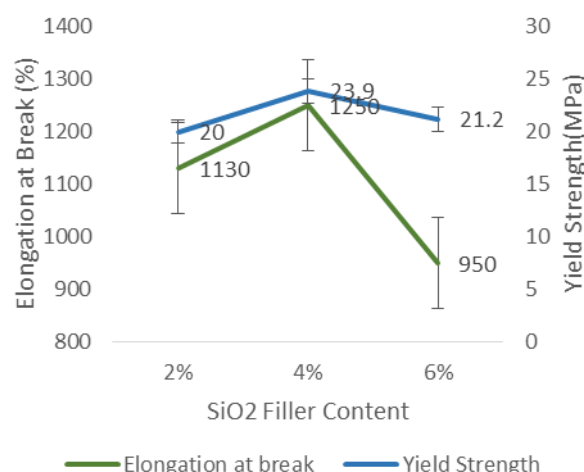


Fig.3: Combination of graph for yield strength and elongation at break for LLDPE/SiO₂ composites.

IV. CONCLUSION

LLDPE material reinforced with varying inorganic filler content of SiO₂ has been prepared for the study of mechanical and characterization study by using FTIR analysis. The FTIR spectrum for the LLDPE/SiO₂ composites projected that successful formation for the composite. The incorporation of 2% until 6% of SiO₂ filler content has shown the effect on the mechanical properties of tensile strength and elongation at break. From this research, it can be concluded that the addition of 4% of SiO₂ filler effectively improved the tensile and elongation at break of the LLDPE/SiO₂ composites. The incorporation of more or less than 4% of SiO₂ filler resulted to the agglomeration and irregular distribution of the particles through-out the LLDPE/SiO₂ composites. Thus at suitable percentage of filler, the composites shows it best properties.

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