Enhancement of Strength and Flexibility of Polymer Composites by Using Glycerol

Nurfatheen binti Abd Rahman[a] and Dr. Norin Zamiah binti Kassim Shaari[a]

[a]Faculty of Chemical Engineering, UniversitiTeknologi MARA, Selangor, Malaysia

Abstract

An attempt is made in this research to enhance the strength and toughness by fabricating fibre fillers (rubber leaves) reinforced polymer composites with the presence of glycerol as plasticizers. At first, size of the fibre filler is varying which are 200µm, 300µm and 500µm in order to select the best size to use for the experiment. The research is made by compression moulding of the mixture of polymer composites at 190°C and 4.826 x 10^6 Pa. The tensile properties of the polymer composite were tested using tensile machine. The aim of this research is to determine the toughness, strength and flexibility of the fabricated polymer composites. The analysis shows that the presence of fibre and plasticizer increased the strength of the polymer composites at filler size of 200 µm. In addition, from the Scanning Electron Microscopic (SEM) analysis, it was found that the filler and plasticizer filled in the porous of HDPE.

Article Info

Article history:

Received date: 6th December 2019
Accepted date: 6th December 2019

Keywords:

Glycerol
Polymer composites
Rubber leaves
Plasticizer
HDPE

1.0 Introduction

Since prehistoric, polymers have been widely used by the people in the world in the form of plastic containers, bottles, bags and plastic toys [1]. Nowadays, the production of polymers is increasing due to high demand. The polymer is the chemical that are bonds with the large molecules in a series of building blocks. The word of polymer itself has a meaning of “many parts” from Greek’s dictionary. Polymer can be classified as the synthetic and natural polymers which both can be used in the form of inorganic and organic polymers [2]. There are several types of polymers that are known to the world which are polyethylene, polypropylene (Herculon), polyvinyl chloride (PVC), polystyrene, and polytetrafluoroethylene (Teflon).

This research only focuses on high density polyethylene (HDPE). These polymer characteristics are tasteless or odourless, lightweight, nontoxic and relatively cheap. There are two type of polyethylene which are low-density polyethylene (LDPE) and high-density polyethylene (HDPE). Both of these have different properties and being used in different application [3]. The most common used by petrochemical industries is HDPE because of its high quality, highly versatile and affordable. The most famously use for HDPE include the pipe system, toys, shampoo bottles and chemical containers [4].

Over the past few years, the test on material toughness is an interesting topic by researchers because some of the polymers have lack in durability. The problem faced by the industries is about the fracture and leakage of the storage tank and garbage bin. Since most common polymer used in industry is HDPE due to its lightweight, cheap and nontoxic compare to other types of polymers, however, it still have a lack in toughness, strength and flexibility.

Thus, the study on polymer is carrying out to enhance the strength of polymer by incorporating fibre filler with glycerol. Since glycerol is known as plasticizer which it can help to improve the strength and flexibility of the polymer.

Due to the problem faced by the industries on polymers, the research on polymerise continuing by formulate polymer composites by varying the concentration of glycerol as plasticizer in order to determine the surface morphology, toughness, strength and flexibility of the fabricated polymer composites.

The toughness can be describe as a property of a material that have an ability to absorb and distribute relatively huge amount of energy of repeated impacts before it crack or fracture by deformation. A polymer that has low toughness and strength is not a good product. For instance, the ceramics which are strong but not ductile have a low toughness [5]. Brittle is hard but reliable to break easily because the brittle materials absorb little energy prior to fracture even at high strength with a snapping sound. When it comes to polymers, the cause of brittle on some of polymer is due to its nature brittle and some of it turn to brittle because of the environmental conditions or aging. A way to avoid the brittleness problem, the composite materials can change the mechanical properties of the polymers with reinforcement of fillers including fibres, nanotubes and others.

Some of the article said that the most usual filler used is natural fibre for polymer composites due to its environmental friendly and it’s lightweight. The idea of the combination of both fibre filler and polymer composites is to enhance the performance on the physical and chemical properties of the mixture. Also, natural filler do not increase the weight and cost of the polymer composites. There is a way to improve the filler is by treating the filler with chemical treatment. The Sodium Hydroxide (NaOH) is one of the chemical that can be used as chemical treatment because it has an ability to increase the strength and adhesiveness [6].

Other than that, there is a softening substance or a solvent called plasticizer that can decrease the intensification of intermolecular interaction and increase the mobility of polymer chains. Plasticizer is made by a reaction between alcohol and acid such as adipic acid, phthalic anhydride and others.

There is a situation when the chain is slides against each other in an opposing direction which can help the process easier, the plasticizer act as internal lubricant in this situation by reducing the friction between the chains. Plasticizer can be classified in two different polar groups which are known as primary plasticizer and secondary plasticizer. The primary plasticizer contains polar groups that are characterized by high salvation capabilities and it is compatible with PVC.

It is known that the most common plasticizers use for films and polymers is glycerol because of its ability to increase the strength properties of the materials. There is a result of research had proven that the presence of glycerol can increase the tensile strength, however, the amount of glycerol is important because if a large amount of glycerol added to the material it may lead to a decrease in the strength of polymer [7].

Thus, the best solution to use the glycerol as plasticizer is by combining it with other chemicals such as citric acid-glycerol (CG) and glycerol-sorbitol (GS) [8]. The citric acid is a nontoxic metabolic product with approval by FDA for its use in human and also known as nutritionally harmless which can help to prevent retro gradation [9]. The combination of citric acid and glycerol produce a various biodegradable polymers [10].

2.0 Methodology

2.1 Material

In this experiment, the materials that have been used are glycerol, citric acid, fibres filler which is rubber leaves and high density polyethylene (HDPE) as polymer. The glycerol and citric acid was used as a cross linking agent called plasticizers. Meanwhile, the fibres as a filler is use to hybrid with the polymer to test on the strength and toughness.

2.2 Methods

The experiment was begun by grinding the fibres into powder and sieve to few sizes in a range of 200µm, 300µm and 500µm, respectively. However, in this experiment the sizes of the fibre that being used was 200µm in order to make the fibre in powder form since the smaller the particle the bigger the surface area [11].

The plasticizer was prepared by dissolving the citric acid into glycerol at 90°C that it can be mixed by setting the mixing velocity to 100 rpm. The ratio of the combinations between glycerol to citric acid is 2:1 [10]. The ratios for the mixtures are 70% is HDPE and 30% is plasticizers. The quantity taken for powder fibres is 5% from the total plasticizer mass. The polymer was mixed with plasticizer and fibre filler.

In order to do comparison with the main sample, the mixture of polymer/plasticizer and polymer/fibre has prepared.

2.3 *Internal Mixing*

The mixture of polymer, plasticizer and fibre filler was mixed and stirred until perfectly mixed. The internal mixing take place at 200°C for the heater since the melting point temperature for HDPE is in a range of 120-180°C and the mixing velocity is increasing 5rpm in every 30 seconds until the rotation reached 55 rpm. This is based on the instruction table shown at the laboratory for the machine setup. The mixture was melt for 10-12 minutes to make sure that the polymer has completely melted. The sample was taken out and it been press slowly to make it thin before it become cold and hard. This can help to ease the experiment during crushing.

2.4 *Compact Crushing*

The experiment continues after the sample became warm at room temperature and can be touched barehanded. The compact crushing was used to crush the polymer composites. It is advisable to crush it twice or more to make sure that the polymer in small shape in order to get large surface area to avoid producing bubble during compression.

If the sample from internal mixing was huge and thick, it cannot be crushed using this crusher because the blades do not have enough power to crush the sample. Thus, the sample needs to be preheated and compressed for 5-10 minutes at 190°C. The sample was then being cut while it is still hot into small pieces. This sample can now be crushed using the crusher, however, it still need to be crushed more than twice.

2.5 *Compression Moulding*

The test was conducted by compression moulding by using hot press of the specimens for evaluation of mechanical. The set temperature for moulding the samples was at 190°C and being compressed at constant pressure of 4.826×10^6 Pa. The suitable shape for tensile test of these samples is in dog bone shapes. The samples were preheated for 5 minutes and 10 minutes of compression time.

The temperature was set at 30°C to proceed with cooling process. The compression continued during cooling process to make sure the sample stays in its shapes. The samples were cool using cooling water for about an hour until the temperature of the heater reached the set temperature. Took out the samples from the hot press and did the checking on the samples. The samples can be acceptable for the test if the samples had no bubbles, and fully melted on the samples.

2.6 *Tensile Test*

The tensile tests was used to measure the force that was applied in order to break a polymer composites specimen and the extent to which the specimen can actually stretches or elongates in its dimensions to its breaking point [12]. The load for the tensile test was 5kN. The specimens have the same width, thickness and length which are 13.7 mm, 3mm and 90.25 mm, respectively [13]. The speed was set at 500 mm/min for the specimens. The standard test method for tensile properties of plastics was chosen to use ASTM-D638 which suits the specimens [14].

2.7 *Scanning Electron Microscopic (SEM)*

The function of SEM is to generate high-resolution images the surface of the polymer composites and show spatial variations. The data was collected at the fracture area of the surface of the experimental samples and an image in two-dimension was generated to show the spatial variations [15]. The investigation of the morphology of the samples using

SEM will be done at an acceleration voltage of 10 kV [16].

3.0 Results and discussion

3.1 Tensile strength

The main objective of the experiment is to enhance the strength of the polymer by combining the HDPE with fibre filler and plasticizer or also called as polymer composites. In order to evaluate the ultimate stress between the polymer, polymer with plasticizers ,polymer with fibre filler and the polymer composites, the tensile test was tested by repeating the test three times to get more accurate data. The average data of those specimens was extracted from the tensile test and tabulated into table as shown in Table 1.

Table 1 Average data from the tensile test of the specimens

Specimen name	**Tensile strength (MPa)**	**Stress at break (MPa)**	**Break strain, %**
HDPE	17.2	15.5	22.6
Plasticizer	20.5	18.4	19.6
Filler (200µm)	20.4	18.3	12.5
Filler (300µm)	19.6	17.7	24.1
Filler (500µm)	19.2	17.2	20.4
Polymer Composites	22.1	19.8	21.6

The result in Figure 1, the result shows that the strength is increased when the polymer was mixed with fibre fillers. However, the highest strength of those samples was fibre powder of 200µm with 20.4 MPa of ultimate stress.

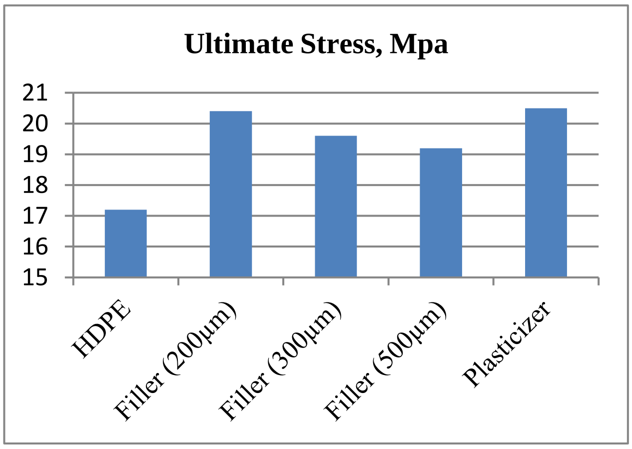


Figure 1 The Comparison on Ultimate Stress of Polymer, Polymer with Plasticizer and Polymer with Fibre Filler of 200µm, 300µm and 500µm.

It has the highest strength because of its small particle size which has a large surface area. The fibre powder fills the pores to make the polymer become more compact and strong. Thus, the highest ultimate stress of fibre was chosen to continue the experiment by mixing it with glycerol as plasticizer in order to check either the plasticizer can enhance the strength of the polymer or not.

Other than that, the bar chart shows that the plasticizer and highest fibre powder composition have the same value of ultimate stress. This shows that with the combination of these two materials, it may enhance the strength of the polymer.

Further testing was conducted by comparing the results between polymer and polymer composites. The 200µm of fibre powder was chosen or to the next section the composites formulation since it shows the best result for ultimate stress as compared to other formulations.

In Figure 2 above, the polymer composites shows the best results where it has the highest ultimate stress in compared to other with 22.1 MPa. The combination of plasticizer and fibre really helps the polymer to enhance its strength. Based on previous research, both of the fibre filler and plasticizer were used in other research to increase the strength of the materials [6].

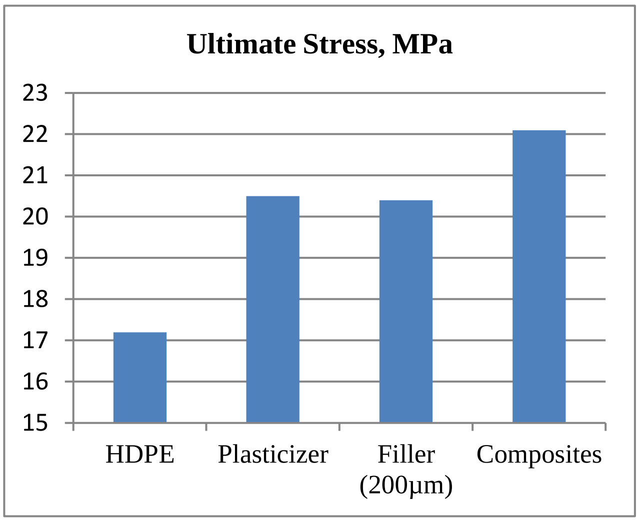


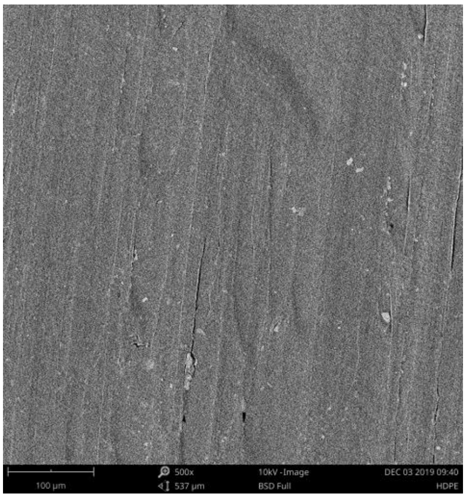
Figure 2 The Comparison on Ultimate Stress for Polymer, Polymer with Plasticizer, Polymer with Filler and Polymer Composites.

Thus, the idea of combining these two materials makes the polymer composites was stronger than other composition.

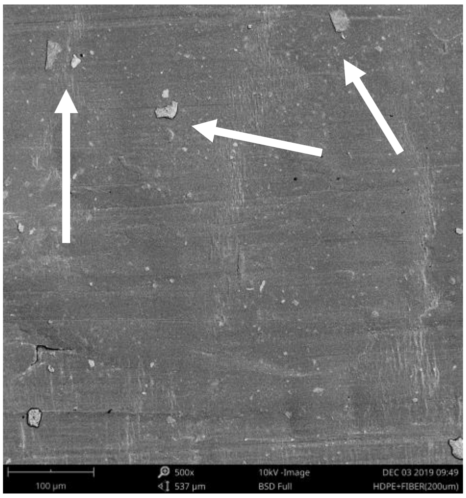
3.2 *Scanning Electron Microscopic (SEM)*

The SEM micrographs with pure HDPE and different composition of 200µm, 300µm and 500µm of rubber leave as the fillers for the polymer. The test was measured by cutting the cross-sectional of the specimens in order to observe inside of the polymer and polymer composites.

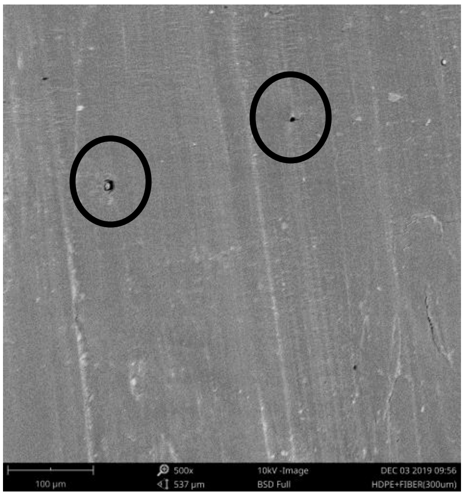
Based on the images in Figure 3, it shows that with the presence of fibre filler in the polymer, the porosity of the polymer is being occupied by the filler [17]. However, the filler of 500 µm in image (d) and 300 µm in image (c) did show a few porous on the cross-section of the specimens which can be the reasons that affect the strength of the polymer composite because the filler is not well exfoliated.



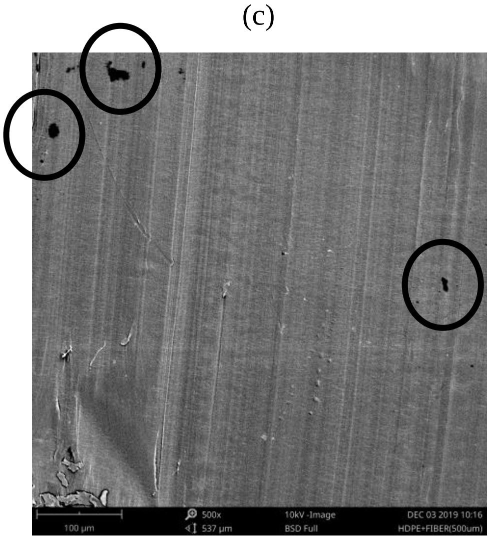
(a)



(b)



(c)



(d)

Figure 3 SEM Images of a) pure HDPE, b) 200µm of Filler, c) 300µm of Filler and d) 500µm of Filler.

The pores on the specimen made the specimens can easily be break at a low force. The smaller size of the filler do help to increase the strength since it has large surface are and it is enough to fill in the porous of the polymer.

In the images, the highest number of the filler that been embedded with the polymer is the smallest size of the filler which is 200µm in compared to 300µm and 500µm. It can be analysed by observing the number of dot appear on the images based on Figure 3.

Other than that, the morphology of the polymer composite with plasticizer also been tested. Based on Figure 4, the presence of the plasticizer and filler can be seen clearly inside the porous surface at the HDPE.

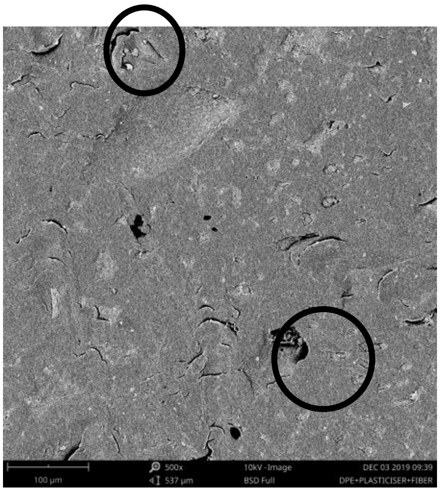


Figure 4 SEM image of polymer composite (HDPE + 200µm + Plasticizer)

The image proved that the porous is being occupied by the filler and plasticizer since the size of these two particles is smaller than HDPE particle and can fit the porous. However, the surface of the specimen may not perfectly mix with the filler and plasticizer during the preparation of the specimens. The surface may not be homogenized between those mixtures which may result to some porosity which can be seen on the surface.

4.0 Conclusions

From the result, the best composite produced is the one from HDPE with 200 µm of filler because it has the highest ultimate stress compare to the other two composite with 20.4 MPa. Based on the above result, it shows that the presence of the plasticizer helps the polymer composite to enhance its strength compares to pure HDPE and polymer composite with filler.

The SEM test analysis revealed that those results are obtained due to the presence of the fillers and plasticizer that filled the porous of the surface of the HDPE which increases the strength of the polymer. Among all the specimens on its strength, the best composition is polymer composites with 200µm by additional of glycerol as plasticizer since it have the highest ultimate stress with 22.1 MPa.

Acknowledgement

The authors feel grateful to the UiTM Shah Alam for financial support and the facilities provided in the campus to make this project successful.

References

1. Allen, G. (1975). Polymeric materials. *Contemporary Physics*, *16*(1), 35–49. https://doi.org/10.1080/00107517508228029

2. Namazi, H. (2017). Polymers in our daily life. *BioImpacts*, *7*(2), 73–74. https://doi.org/10.15171/bi.2017.09

3. Britannica, T. E. (2019, February 29). Polymer.

4. Katz, J. (2014, April 16). *Quality Plastic Film and Plastic Sheets*. Retrieved from What is difference between HDPE and LDPE plastic sheets?: https://www.grafixplastics.com/difference-hdpe-ldpe-plastic-sheets/

5. Brostow, W., Hagg Lobland, H. E., & Khoja, S. (2015). Brittleness and toughness of polymers and other materials. *Materials Letters*, *159*, 478–480. https://doi.org/10.1016/j.matlet.2015.07.047

6. Vignesh, P., Venkatachalam, G., Gautham Shankar, A., Singh, A., Pagaria, R., & Prasad, A. (2018). Studies on Tensile Strength of Sugarcane Fiber Reinforced Hybrid Polymer Matrix Composite. *Materials Today: Proceedings*, *5*(5), 13347–13357. https://doi.org/10.1016/j.matpr.2018.02.327

7. Yunos, M. Z. B. (2011). Effect of Glycerol on Performance Rice Straw/Starch Based Polymer. *Applied Scrience*, 2456–2459.

8. Sanyang, M. L., Sapuan, S. M., Jawaid, M., Ishak, M. R., & Sahari, J. (2015). *Effect of plasticizer type and concentration on physical properties of biodegradable films based on sugar palm (arenga pinnata) starch for food packaging*. https://doi.org/10.1007/s13197-015-2009-7

9. Shi, R., Zhang, Z., Liu, Q., & Han, Y. (2007). *Characterization of citric acid / glycerol co-plasticized thermoplastic starch prepared by melt blending*. *69*, 748–755. https://doi.org/10.1016/j.carbpol.2007.02.010

10. Halpern, J. M., Urbanski, R., Weinstock, A. K., Iwig, D. F., Mathers, R. T., & Recum, H. A. Von. (2013). *A biodegradable thermoset polymer made by esterification of citric acid and glycerol*. 1467–1477. https://doi.org/10.1002/jbm.a.34821

11. Ashik, K. P., & Sharma, R. S. (2015). A Review on Mechanical Properties of Natural Fiber Reinforced Hybrid Polymer Composites. *Journal of Minerals and Materials Characterization and Engineering*, *03*(05), 420–426. https://doi.org/10.4236/jmmce.2015.35044

12. Fibre, B., Polymer, R., Naik, P. K., & Londe, N. V. (n.d.). *Experimental Study on Fracture Toughness of Natural Fibres Reinforced Hybrid Composites Experimental Study on Fracture Toughness of Natural Fibres Reinforced Hybrid Composites*. https://doi.org/10.1088/1757-899X/376/1/012088

13. Essabir, H., Achaby, M. E., Hilali, E. M., Bouhfid, R., & Qaiss, A. (2015). Morphological, Structural, Thermal and Tensile Properties of High Density Polyethylene Composites Reinforced with Treated Argan Nut Shell Particles. *Journal of Bionic Engineering*, *12*(1), 129–141. doi: 10.1016/s1672-6529(14)60107-4

14. International, A. *Standard Test Method for Plastics*. West Conshohocken: ASTM International.

15. Utilizing, Q. D. (2016). *(12) United States Patent WITH A SURFACE PROFILER g : A*. *1*(12).

16. Sanjay, M. R., Arpitha, G. R., & Yogesha, B. (2015). Study on Mechanical Properties of Natural - Glass Fibre Reinforced Polymer Hybrid Composites: A Review. *Materials Today: Proceedings*, *2*(4–5), 2959–2967. https://doi.org/10.1016/j.matpr.2015.07.264

17. Rashidi, A. R., Muhammad, A., & Roslan, A. (2017). Morphology-Property relationship of high density Polyethylene/Hevea Brasiliensis Leaves/Imperata cylindrica hybrid composite: Impact strength. doi: 10.1063/1.5002423