Malaysian Journal of Chemical Engineering & Technology 3(2) (2020) 25–29

Malaysian Journal of Chemical Engineering & Technology

Journal Homepage: http://myjms.mohe.gov.my/index.php/mjcet

Composite from high density polyethylene (HDPE) reinforced with rice husks (*Oryza sativa*): Preparation and mechanical property

Norin Zamiah Kassim Shaari*, Ahmad Redha Taha, Suhaiza Hanim Hanipah

[a]Faculty of Chemical Engineering, Universiti Teknologi MARA, Selangor, Malaysia

*Corresponding email: norinzamiah@uitm.edu.my

Abstract

In this study, the composite from HDPE polymer was formulated with the incorporation of *Oryza sativa* also known in general as rice husks or hulls with two different methods: with the incorporation of plasticiser (glycerol and citric acid) and without plasticisers. The ratio of glycerol to citric acid used in the experiment was 1:2. The aim of this study is to determine the effect of incorporating rice husk with different fibre sizes on the mechanical properties of the HDPE composite. The blending of HDPE with the rice husk was performed in a mixer in processing compounding polymer followed by the extrusion process. Results show that in the absence of the plasticiser, rice husk portrays good compatibility with HDPE polymer, where the composite possessed good tensile strength and elongation. The best plasticising effect was portrayed by filler with 100 µm as it yields the highest tensile strength and strain. In conclusion, the composite could be potentially used for suitable applications, which requires flexible material with better processing ability and would not be brittle.

Article Info

https://doi.org/10.24191/mjcet.v3i2.10941

Article history:

Received date: 7 October 2020
Accepted date: 1 December 2020

Keywords:

HDPE
Rice husk
Fibre size
Tensile strength
Elongation

1.0 Introduction

High density polyethylene (HDPE) is produced by the polymerisation of ethylene through catalytic process. HDPE is mostly used for piping systems due to its high chemical resistance property and can be moulded, shaped or welded. HDPE possesses good low temperature toughness, reasonable tensile strength, less moisture absorption, good resistance to corrosion and can be reinforced with natural fibre or filler to improve the properties (Daramola et al., 2015). The reinforcement with fillers was carried out to improve the HDPE's low toughness, weather resistance and environmental stress cracking resistance (Rabeh et al., 2010). Applications of HDPE polymer varies from pipe material to carry hazardous waste and potable water system, until production of bottle caps, food storage containers, backpacking frame and vehicles fuel tank. The fabrication of blended HDPE was carried out by mixing and moulding process.

HDPE is a thermoplastic widely known for its high tensile strength and can withstand high temperatures. The density of HDPE ranges from 930 kg/m^3 to 970 kg/m^3. The HDPE density is higher than LDPE with longer chain branching. This chain branching provides HDPE with stronger intermolecular forces that results in high tensile strength as compared to LDPE (Thakare et al., 2015).

The rice husk or also known as rice hulls is an inexpensive material which is obtained from milling process of rice as a by-product. Around 0.23 tonne of rice husks was produced for every tonne of rice production. In Asia region, which are Malaysia, India, China, Indonesia and Bangladesh, the rice milling process is one of the major productions. The silica content in rice husk is used to enhance the mechanical properties of composites (Arjmandi et al., 2015). Rice husk has been utilized as an insulating material in the production of organic chemicals, supplementary cementing material and activated carbons due to its high availability, low bulk density and abrasiveness in nature (Das et al., 2014). Some of the rice husks were converted into agricultural by-products (fertilizer) and industrial biomass material. Hence, rice husks are crucial for developing new economical feasible processes or pathways to fully utilise organic substances. The properties of rice husks such as light weight, biodegradability, and toughness lead to incorporation of rice husks into polymer matrices. In comparison to wood-based composites, the rice husks have high resistance to termite and provide good dimensional stability as a filler in enhancing polymer structure, therefore it can be used in construction such as interior panel and frames of windows or doors (Battegazzora et al., 2014).

Moulding processes such as injection, compression and extrusion are common techniques for natural fibre reinforced thermoplastic composites. The enhancement of thermoplastics is important where the rice husks act as additional material or modifier in reducing the flow properties of the composites by increasing the interaction between plastic matrices and natural fibre. Also, the incorporation of rice husks helps in reducing the viscosity of the molten composites. The rice husks are commonly incorporated with low density polyethylene (LDPE), high density polyethylene (HDPE), polylactic acid (PLA), polypropylene (PP), and polyvinyl chloride (PVC). As a result, the improved composites were produced with enhanced thermal, mechanical, chemical and physical properties due to the addition of natural fibres such as rice husks (Arjmandi et al., 2015).

Generally, plasticisers can be classified into primary and secondary. The primary plasticiser is soluble at high concentration and this kind of plasticiser used as a main element to enhance polymerisation of thermoplastics. Meanwhile the secondary plasticiser was stated to have low gelation capacity with polymer and typically blend with the primary plasticiser to improve the properties of composites. Glycerol, which has a low molecular weight and is non-volatile, is used as a plasticiser to improve the flexibility of the HDPE composite since HDPE alone is brittle. The addition of glycerol from range 0% to 50% increases the flexibility of composites without jeopardising the strength (Gozan and Noviasari, 2018).

Citric acid as another type of plasticiser could inhibit the degradation of polymer. It enhances both barrier and mechanical properties of the blend polymer, and also function as renewable and biodegradable biopolymer that interact with organic rice husks fillers. Cross-linking of matrices in polymer provides strong flexibility and tensile strength. A blend of citric acid with glycerol is used as a plasticiser with different ratios in formulations of polymer composite (Khan et al., 2017). Chemical modifications in cross-linking of citric acid in blend polymer may provide an alternative to enhance mechanical properties and stability. The matrices of polymer can be modified by cross-linking reactions, chemical and physical reactions. The cross-linking in matrices of polymer is an effective alternative to improve the tensile strength and stable matrix of blend polymer, where the citric acid has a capability to cross-link the blend polymer and reported occur at high temperatures range from 165 °C to 175 °C (Quiroz et al., 2018).

HDPE was characterised by its strain hardening response under tension, stress and strain along by multiple cracking. In general, a direct tensile test was a good alternative to determine the fundamental tensile behaviour of HDPE composites which precisely important in design and modelling. The tensile test strength was obtained by tensile response of thermoplastic composites, which are carried out on small size specimens in variable of full-scale structural elements and the possible scale effects (Chao et al., 2018).

The tensile strength of thermoplastics such as HDPE is the maximum tensile stress by performing the dog bone shaped specimen on tensile test. The tensile test for HDPE plastics is conducted by Standard Test Method for Tensile Properties of Plastics (ASTM D638) 2014. Typically, a dog bone shaped specimen was chosen for testing the plastic specimen. The stress-strain curve is the first point for the yield point and by increasing the strain not the stress. Both yield stress or yield strength are usually defined as yield point. The elongation of HDPE might be up to ten times of the HDPE original gauge length. The occurrence of failure happened by breaking the molecules strain of defects to cause failure of the sample (Raed & Lampson, 2011).

The tensile tests were relatively simple, fully standardised, and inexpensive as a primary method for material acceptance, design limit and quality control. The parameters that can be obtained from the test are elastic modulus (E), tensile strength (σ), and strain (Osoka & Onukwuli, 2018).

In this study, the composite HDPE was formulated by mixing HDPE with rice husk fibre with and without plasticiser (glycerol and citric acid), respectively. The plasticiser is formed by mixing glycerol and citric acid at certain weight ratio. For the rice husks, it was sieved into three different sizes that are 50, 75, and 100 µm and its effect on the mechanical property of the HDPE composite was evaluated.

2.0 Methodology

2.1 Material

High-density polyethylene (HDPE) with density 0.957 g/cm^3 and purity 99.9% was obtained from US Pharmacopeia Brand. Glycerol with 99.5% purity and citric acid with density, ρ is 1.665 g/cm^3 were purchased from Sigma Aldrich. The rice husk, *Oryza Sativa* was obtained from Universiti Putra Malaysia.

2.2 Methods

2.2.1 Sieving process

The raw rice husks at the first stage were ground into small size approximately around 1000 μm. Then the ground rice husks were placed into sieving machine to separate three different sizes of the fibres at 50, 75, and 100 μm.

2.2.2 Preparation of plasticiser

About 8 g of plasticiser mixture was prepared by mixing citric acid with glycerol at weight ratio 2:1 of citric acid: glycerol. The mixture was consistently stirred with magnetic stirrer at 200 rpm (Tisserat et al., 2012).

2.2.3 Mixing process

The mixing of blended polymer was performed by using Thermo Haake PolyLab Internal Mixer and the temperature was set to 190 °C and preheated for 5 mins. The mixer can occupy 40 g of sample. An amount of 30 g HDPE was poured into the mixer followed by 8 g of plasticiser mixture. Then, 2 g of each size of rice husks was poured into the mixer (Nordyana et al., 2013). The rotor speed was started with 5 min^{-1} and increased by 5 min^{-1} for every 40 s until it reached to 40 min^{-1} after 10 mins. After completion, the sample was cooled down to room temperature for crushing process. For the composite without plasticiser, 38 g HDPE was mixed with 2 g rice husk fibres. Table 1 shows the formulation of each composite.

Table 1 Formulation of composites

Composites	Composition
HDPE	Pure HDPE
HDPE P50	HDPE, plasticiser & 50 μm rice husk
HDPE P75	HDPE, plasticiser & 75 μm rice husk
HDPE P100	HDPE, plasticiser & 100 μm rice husk
HDPE 50	HDPE & 50 μm rice husk
HDPE75	HDPE & 75 μm rice husk
HDPE 100	HDPE & 100 μm rice husk

2.2.4 Sample preparation for tensile test

The dog bone specimen was produced by using Hot Press equipment where initially the compression machine was pre heated at temperature 190 °C for 3 mins. Then, the compression machine was set at 190 °C to melt and shape the blended HDPE polymer with 10 mins of compression time. Based on ASTM D638, the range of thickness for the dog bone specimen was 1-5 mm. (Komurlu & Kesimal, 2017).

2.2.5 Tensile test

The test was conducted using Universal Testing machine model: H50KT, brand: Tinius Olsen. The load cell used was 10 kN and crosshead speed was set to 500 mm/min. This test was conducted according to the technical standard method ASTM D638. The test specimen was gripped between two ends and elongate at a determined rate of breakpoint. From the analysis, elastic modulus, tensile strength and strain were determined. For each formulation, three samples were tested and the average value for elastic modulus, tensile strength and strain respectively was calculated.

3.0 Results and discussion

3.1 Effect of rice husk on elastic modulus

Figure 1 shows the elastic modulus for each composite. Pure HDPE has the highest elastic modulus than the composites. High elastic modulus shows that the material has high toughness that is attributed to orderly branch distribution which has enhanced crystallisation and more dense material that reduces

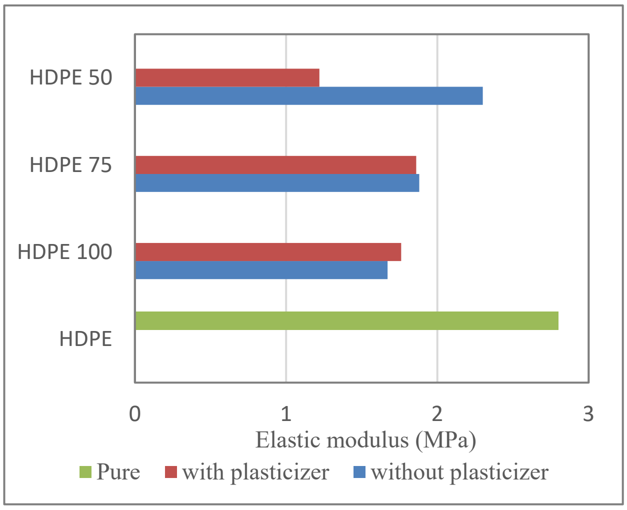

Fig. 1 Elastic modulus of the composites

chain mobility of the polymer matrix (Dikobe and Luyt, 2017). Fig. 1 shows that the presence of rice husk and plasticiser has lowered down the stiffness of the composites where the introduction of these materials has disturbed the order of the branch distribution in the HDPE polymer matrix.

3.2 Effect of rice husk on tensile strength and elongation

Based on Fig. 2 and 3, it was observed that with the presence of both plasticisers and rice husk in the HDPE matrix, the tensile strength and strain of the composites decrease regardless of the rice husk fibres' sizes. However, with the presence of only rice husk in the HDPE matrix, there was only slight increase in the tensile strength at 18.7 MPa with 75 µm rice husk fibre as compared to pure HDPE. Composites with 50 and 100 µm have almost similar strength with pure HDPE. However, there was an increase in the elongation of composites with the presence of rice husk without the plasticisers as depicted through the strain values. Larger elongation before fracture portrayed by these composites especially HDPE 100 was due to stronger interaction between HDPE and rice husk fibre that prevent the crack propagation through the composite's interface (Dikobe and Luyt, 2017). As stated by Salih et al. (2013), filler often gives negative effect on the tensile strength of materials if it is not compatible with the materials.

Based on Fig. 2 obviously the presence of rice husk as filler without plasticiser shows the good interaction between HDPE and rice husk as the tensile strength of composites was slightly increased. But opposite trend for composites HDPE P50, HDPE P75 and HDPE P100, where the presence of plasticiser has initiated the crack formation and then it was propagated along the interfaces between polymers. This situation was due to incompatibility of the mixtures (Dikobe and Luyt, 2017). As reported by Daramola et al., (2015), the incompatibility of polymer blend causes agglomeration of particle and formation of voids. Thus, the voids, which acts as stress concentration sites can magnify and amplify the stress even at low concentration leading to a fast fracture. Whereas the good compatibility of filler with the polymer matrix could be explained through an excellent load transfer from the polymer matrix to filler within the composite, good resistance to dislocation motion and no agglomeration of particles. These phenomena are known as stress transfer, interparticle space reduction and dislocation-interface (Daramola et al., 2015).

The reduction of elastic modulus for composite HDPE 50, HDPE 75 and HDPE 100 as depicted in Fig. 1, although the tensile strength and strain increases, could be explained through the disturbance on the branch distribution of HDPE polymer, which results in weaker, less stiff and freely chain rotation (Dikobe and Luyt, 2017). High elongation of the composites portrays the flexibility and ductility of the material. The resultant ductile composite will overcome problem of HDPE brittleness, which sometime restrict their applications.

4.0 Conclusions

Result shows that the composites consist of HDPE and rice husk fibre yielded higher tensile strength and elongation compared to that with the addition of plasticiser. High elongation of the composites portrays the flexibility and ductility of the material. Sizes of rice husk fibres also affect the mechanical property of the composite. The best plasticising effect was portrayed

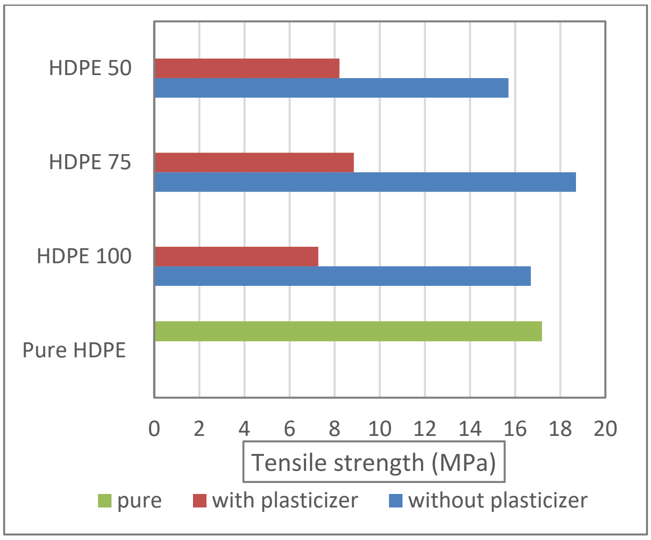

Fig. 2 Tensile strength of different composites

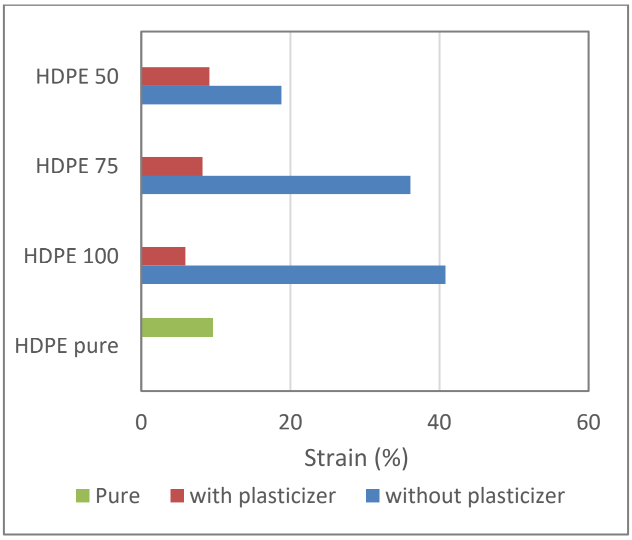

Fig. 3 Strains of different composites

by filler with 100 μm as it yields the highest tensile strength and strain. In conclusion, the composite could find the suitable applications, which requires flexible material with better processing ability and would not be brittle.

Acknowledgement

The authors would like to thank Mr. Mohd Idris Md Desah and Mr. Amin Fafizullah Omar for their assistances on the equipment handling during the fabrication process of the polymer composite.

References

A.M. Das, A. A. Ali, P. Hazarika, (2014). Synthesis and characterisation of cellulose acetate from rice husk: eco-friendly condition, *Carbohydrate Polymers. 11*2. 342–349.

A.Z. Raed & Lampson B. A. (2011, July). Tensile Testing. Retrieved from https://www.mse.iastate.edu/files/2011/07/polymer-lab-instron-procedure.pdf

B. Khan, M. B. K. Niazi, A. Hussain, Z. Jahan, (2017). Influence of carboxylic acids on mechanical properties of thermoplastic starch by spray drying, *Journal of Fibers and Polymers. 18(1)*. 44–73.

B. Tisserat, R.H. O'kuru, H.S. Hwang, A.A. Mohamed, R. Holser, (2012). Glycerol citrate polyesters produced through heating without catalysis, *Journal of Applied Polymer Science*. *125*. 3429–3437.

D. Battegazzore, S. Bocchini, J. Alongi, F. Marino, (2014). Rice Husk as Bio-source of Silica: Preparation and Characterization of PLA-Silica Bio-Composites, *RSC Advances*. 4.54703–54712.

D.G. Dikobe, A.S. Luyt, (2017). Thermal and mechanical properties of PP/HDPE/wood powder and MAPP/HDPE/wood powder polymer blend composites, *Thermochimica Acta*. 654. 40–50.

D.M. Gozan, C. Noviasari (2018). The effect of glycerol addition as plasticizer in spirulina platensis-based bioplastic, Web of Conferences 67. 03048; https://doi.org/10.1051/e3sconf/20186703048

E. Komurlu, A. Kesimal, (2017). Experimental Study on Usability of Friction Rockbolts with Plastic Bodies, *International Journal of Geomechanics* 1. *7(9)*. 04017058.

E. Osoka, O.D. Onukwuli, (2018). Determination of mechanical properties from stress-strain data: A new approach, *Journal of Engineering Research and Advanced Development. 4*. 61–70.

H. Rabeh, Elleithy, I. Ali, M.A. Ali, S.M. Alzahrani, (2013). High density polyethylene/micro calcium carbonate composites: A study of the morphological, thermal, and viscoelastic properties, *Journal of Applied Polymer Science, 117*. 2413–2421.

K.A. Thakare, H. G. Vishwakarma, A. G. Bhave, (2015). Experimental investigation of possible use of HDPE as thermal storage material in thermal storage type solar cookers, *Journal of Research in Engineering and Technology*. *4*. 92–99.

M.J.T. Quiroz, J.J.F. Diaz, A. Pinotti, (2018). Characterization of methylcellulose-based hydrogels by using citric acid as a crosslinking agent, *Journal of Applied Engineering Research. 13*.13302–13307.

O.O. Daramola, I.O. Oladele, B.O. Adewuyi, R. Sadiku, S.C. Awunca, (2015). Influence of Submicron Agro Waste Silica Particles and Vinyl Acetate on Mechanical Properties of High-Density Polyethylene Matrix Composites, *The West Indian Journal of Engineering*. *38(1)*. 96–107.

R. Arjmandi, A. Hassan, K. Majeed, Z. Zakaria, (2015). Rice husk filled polymer composites, *Journal of Polymer Science*. Article ID 501471; https://dx.doi.org/10.1155/2015/501471

R. F. Nordyana, A. Z. Romli, M. H. Abidin, (2013). Effect of Rice Husk Particle Size on Tensile and Density of Recycled PPVC Composite, *Advanced Materials Research*. *812*. 145–150.

S. E. Salih, A.F. Hamood, A.H. Abdalsalam, (2013). Comparison of the characteristics of LDPE:PP and HDPE:PP polymer blends, *Modern Application of Science*. *7*. 33–42.

S. H. Chao, W. C. Liao, T. Wongtanakitchareon, A. E. Naaman, (2016). Large scale tensile tests of high-performance fiber reinforced cement composites, *Journal of Materials and Structures. 30*. 259–2