

Development of Biocomposite: The Influence of Natural Fillers

by

Noor Atilia binti Selamat

Dissertation submitted in partial fulfillment of
the requirements for the
Bachelor of Engineering (Hons)
(Mechanical Engineering)

JUNE 2010

Universiti Teknologi PETRONAS
Bandar Seri Iskandar
31750 Tronoh
Perak Darul Ridzuan

CERTIFICATION OF APPROVAL

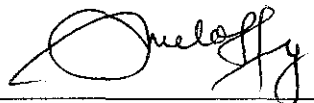
Development of Biocomposite: The Influence of Natural Fillers

by

Noor Atilia binti Selamat

A project dissertation submitted to the
Mechanical Engineering Programme
Universiti Teknologi PETRONAS
in partial fulfilment of the requirement for the
BACHELOR OF ENGINEERING (Hons)
(MECHANICAL ENGINEERING)

Approved by,



(AP. Dr. Puteri Sri Melor Megat Yusoff)

Dr Puteri Sri Melor Bt Megat Yusoff
Associate Professor
Mechanical Engineering Department
Universiti Teknologi PETRONAS

UNIVERSITI TEKNOLOGI PETRONAS

TRONOH, PERAK

June 2010

CERTIFICATION OF ORIGINALITY

This is to certify that I am responsible for the work submitted in this project, that the original work is my own except as specified in the references and acknowledgements, and that the original work contained herein have not been undertaken or done by unspecified sources or persons.



(NOOR ATILIA BINTI SELAMAT)

ACKNOWLEDGEMENTS

Alhamdulillah praised to Allah the Almighty for His love, guidance and strength until I completed this dissertation.

My deepest appreciation goes to my dedicated supervisor Assoc. Prof. Dr. Puteri Sri Melor binti Megat Yusoff for her guidance, valuable suggestion and constructive comments throughout the course of completing this research project.

I would also like to express my gratitude to all UTP lab technologists, Mr. Zairi, Mr. Faisal, Mr. Shairul, Mr. Irwan, Mr. Anuar and Mr. Omar for their kind cooperation and assistance in lab works and the technical support provided throughout the research period.

Special thanks to all my friends for the knowledge shared, motivation, support and companion through thick and thin. Not to forget, my utmost appreciations to my beloved parents Selamat bin Hj. Rahmat and Hasma binti Hj Wa'Gani and other family members for their prayers, continuous support and encouragement.

To all the people who have helped me directly or indirectly throughout this research, your contributions are very much appreciated. Thank you.

ABSTRACT

A study was carried out to investigate the influence of different types of natural fillers and their loadings on the mechanical properties of high density polyethylene (HDPE) based composites. Silane treated oil palm empty fruit bunch (EFB) and sawdust, varied from 10 – 30 %wt were used as the filler in the composites. The incorporation of fillers was found to reduce the tensile strength of EFB and sawdust-filled HDPE by 25% and 24% respectively whereby the flexural strength increased up to 11% for EFB composite and 14% for sawdust composite. Insignificant change was observed on the tensile strength of both EFB and sawdust composites as the filler loading was varied. However, the flexural strength of EFB composite increased up to 10% while sawdust composite impart no significant improvement on flexural strength as the effect of filler loading was concerned. The micrographs from scanning electron microscope (SEM) revealed the interaction between the fillers and the HDPE matrix through the presence of fibre pull out, small gap and matrix tearing that weakened the composite materials.

TABLE OF CONTENTS

CERTIFICATION OF APPROVAL	i
CERTIFICATION OF ORIGINALITY	ii
ACKNOWLEDGEMENTS	iii
ABSTRACT	iv
TABLE OF CONTENTS	v
LIST OF FIGURES	vii
LIST OF TABLES	viii
LIST OF ABBREVIATIONS	ix
CHAPTER 1: INTRODUCTION	1
1.1 Background of Study	1
1.1.1 Oil Palm Empty Fruit Bunch.	2
1.1.2 Sawdust	2
1.2 Problem Statement	3
1.3 Objective and Scope of Study.	4
1.3.1 Objective	4
1.3.2 Scope of Study	4
CHAPTER 2: THEORY / LITERATURE REVIEW	5
2.1 Composite	5
2.2 Chemical Composition of Natural Fibre	5
2.2.1 Cellulose	6
2.2.2 Hemicellulose	7
2.2.3 Lignin	7
2.2.4 Pectin	7
2.2.5 Wax	7
2.3 Mechanical Properties of Natural Fibre	8
2.4 Literature Review	8
2.4.1 Oil Palm Empty Fruit Bunch (EFB)	9
2.4.2 Sawdust	10
2.4.3 Other natural fillers	10
CHAPTER 3: METHODOLOGY	13
3.1 Materials	13
3.1.1 Polymer Matrix	13
3.1.2 Natural Fibres	13
3.2 Fibre Surface Treatment	13
3.3 Composite Preparation	14
3.4 Mechanical Testing	14
3.5 Morphology Study	14

3.6	Project Activities	. . .	14
CHAPTER 4:	RESULT AND DISCUSSION	. . .	15
4.1	Morphological Characterization of Natural Fibres	. . .	15
4.1.1	Oil Palm Empty Fruit Bunch	. . .	15
4.1.2	Sawdust	. . .	17
4.2	Mechanical Properties of Natural Fibre-Filled HDPE Composites	. . .	18
4.2.1	Tensile Strength	. . .	18
4.2.2	Flexural Strength	. . .	20
4.3	Morphology Study of the Composites Fractured Surface	. . .	21
4.3.1	Tensile-Fractured Surface	. . .	21
4.3.2	Flexural-Fractured Surface	. . .	23
CHAPTER 5:	CONCLUSIONS & RECOMMENDATIONS		25
REFERENCES		27
APPENDICES		31

LIST OF FIGURES

Figure 1-1 Biomass produced in Malaysia [5]	3
Figure 4-1 Scanning electron micrographs of oil palm empty fruit bunch surface (500X) (a) before dry, (b) after dry, (c) after 3 %wt silane treatment.	16
Figure 4-2 Scanning electron micrographs of sawdust surface (500X) (a) before dry, (b) after dry, (c) after 3 %wt silane treatment.	17
Figure 4-3 Tensile strength of EFB and sawdust-filled HDPE composite at different filler loading.	19
Figure 4-4 The sawdust-filled HDPE composite specimen.	20
Figure 4-5 Flexural strength of EFB and sawdust-filled HDPE composite at different filler loading.	21
Figure 4-6 Scanning electron micrograph of tensile-fractured surface of 30 %wt EFB-filled composite (30X).	22
Figure 4-7 Scanning electron micrograph of tensile-fractured surface of 30 %wt sawdust-filled composite (150X).	23
Figure 4-8 Scanning electron micrograph of flexural-fractured surface of 30 %wt EFB-filled composite (30X).	24
Figure 4-9 Scanning electron micrograph of flexural-fractured surface of 30 %wt sawdust-filled composite (30X).	24

LIST OF TABLES

Table 2-1 Chemical composition of natural fibres [10,11]	6
Table 2-2 Mechanical properties of natural and synthetic fibres [14]	8

LIST OF ABBREVIATIONS

CF	Coir fibre
EFB	Empty fruit bunch
HDPE	High density polyethylene
OPF	Oil palm frond
PP	Polypropylene
SEM	Scanning Electron Microscope
SHSD	Sunflower hull sanding dust
STD	Standard deviation
TWF	Teak wood flour
VTS	Vinyltriethoxysilane

CHAPTER 1

INTRODUCTION

1.1 BACKGROUND OF STUDY

Composites consist of one or more discontinuous phases embedded in a continuous phase known as reinforcement and matrix respectively [1]. The resultant composite materials possess better and enhanced properties compared to that of the individual components. Composite materials have been widely used in various applications such as in aircraft structures, automobile industries and construction field.

Fibres are the most important part of a composite material. They are categorized into two categories, namely organic (carbon, graphite, aramid, polymer) and inorganic fibres (glass) [1]. Apart from these synthetic fibres, plant-based fibres or also known as natural fibres are also used as filler in polymers [1].

The use of natural fibres as filler in composite material to replace the widely used synthetic fibres is gaining considerable attention among researchers [2-4, 10-11, 14-25, 29-30]. Replacing synthetic fibres with natural fibres is likely to bring both environmental and economic advantages. The advantages of plant fibres over synthetic fibres are they are abundantly available at a low cost and easily obtained from renewable resources [2]. Furthermore, these natural fibres also display properties such as low density, good thermal insulation, and demonstrate high specific strength. These features make natural fibres suitable as fillers in plastics, be it thermosets or thermoplastics. Apart from that, natural fibres impose no health hazards, they reduced amount of machine wear during processing and they are recyclable [3].

Being derived from ligno-cellulose, which contain strongly polarized hydroxyl groups makes natural fibres hydrophilic in nature [3]. The hydrophilic nature of the natural fibre affects adhesion to a hydrophobic polymer matrix. Various treatments, ranging from graft copolymerization of monomers onto the fibre surface to the use of coupling agents (silanes, titanates, zirconates, triazine compounds) can be used to improve fibre – matrix adhesion [4].

1.1.1 Oil Palm Empty Fruit Bunch (EFB)

Oil palm (*Elais guineensis*) which originated from West Africa is an important species of *Elais* genus which belongs to the Palmae family [5]. Nowadays, oil palm is planted in most countries including Malaysia. The oil palm fruit grows in large bunches which weighs between 10 to 40 kg [5]. The fruit has a promising market because it is widely used in industries. Each part of the oil palm fruit has their own benefit. For example, oil can be extracted from the pulp and the fruit itself while the kernel is used in soap production [5]. Another important part of the oil palm fruit is the empty fruit bunch. This empty fruit bunch is indeed the waste obtained upon completion of oil extraction process, when the fruits and nuts have been stripped off from the bunch [6]. This waste is harmful to the environment because it can cause pollution and attracts pests. However, the empty fruit bunch is rich in fibre. The fibre is beneficial in such a way that it can be used in compounding composite material to improve the mechanical properties of the resultant products. Hence, the utilization of fibre from oil palm empty fruit bunch (EFB) in composite will eventually help to increase the usability of this residual waste and at the same time reducing the environmental problems caused by the waste.

1.1.2 Sawdust

Wood, which originated from shrubs and trees is a natural composite composed of 40-50% cellulose and 15-25% hemicelluloses bind together by 15-30% lignin [1]. Wood can be classified into softwood and hardwood. The distinct difference between softwood and hardwood is the existence of vessels (pores) in their structure where hardwood structure is filled with vessels while there are no such vessels in softwood

structure [1]. Woods are mainly used in industries such as furniture and paper production. The processing of woods to produce products, contributes to generation of high number of waste in the form of sawdust. These unused sawdust have found its application in the production of plywood. Despite their usage in plywood production, there are still a large portion of these residues available. Another option to fully utilize the sawdust is by introducing them into polymers, to produce composite with enhanced properties.

1.2 PROBLEM STATEMENT

Each year, the agricultural field in Malaysia produces more than 70 million tonnes of agricultural by-products [7]. These agricultural by-products which are the most abundant biomass resources include oil palm fibre, oil palm empty fruit bunch (EFB) and coconut husk. Other types of biomass produced in Malaysia are as shown in Figure 1-1 [5]. The tropical weather in Malaysia, where there is adequate amount of rain and sunlight throughout the year contributes to the high production rate of biomass [7]. Unfortunately, the arising number of the produced by-products creates waste management problem where the disposal of the by-products in large quantity is difficult and costly [7]. Taking into account the environmental effects caused by these bio- waste, numbers of research and study have considered converting these biomass into high value products that will serve as raw materials to support other industries [7].

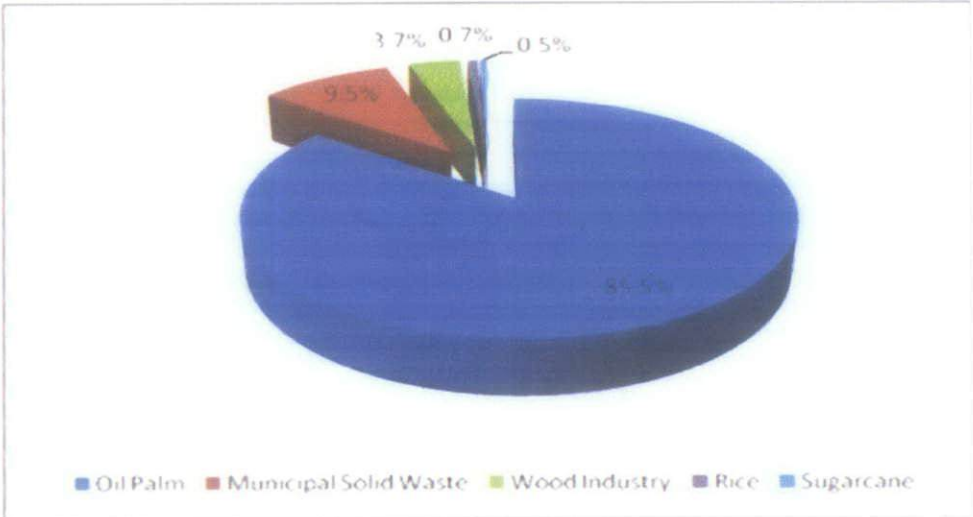


Figure 1-1 Biomass produced in Malaysia [5].

Therefore, extensive research has been carried out regarding the use of natural fibres extracted from these agricultural by-products. Natural fibres are much cheaper if compared to traditional glass and carbon fibres used in the production of advanced composites. These natural fibres are used as filler in polymers such as polyethylene to produce new class of materials for various applications.

However, before these fibre-filled composites can be used for products development, the characteristics of the fibre-filled composite materials have to be studied thoroughly to ensure its reliability and suitability. These include the fibre/matrix compatibility and the mechanical performance such as tensile and flexural properties of the resultant composites.

1.3 OBJECTIVE AND SCOPE OF STUDY

1.3.1 Objective

To investigate the influence of various types of natural fillers and their loading on tensile and flexural properties of EFB and sawdust filled high density polyethylene (HDPE) composites.

1.3.2 Scope of Study

Composite material consists of matrix material and the reinforcing material. For the purpose of this study, high-density polyethylene (HDPE) is chosen as the matrix material while oil palm empty fruit bunch (EFB) and sawdust with fibre loading ranging from 0 to 30 %wt are used as the fillers. The hydrophilic nature of the natural fibres contradicts the hydrophobic nature of the matrix. Therefore, all the natural fibres used in this study undergo surface treatment where 3% silane coupling agent is used to enhance their compatibility with the matrix. The manufacturing and processing of the natural fibre composites involve injection molding process. The produced composites are then tested for their tensile and flexural properties.

CHAPTER 2

THEORY / LITERATURE REVIEW

2.1 COMPOSITE

Composite is defined as a mixture of two or more distinct constituents or phases [8]. The constituents consist of continuous and discontinuous phases. The continuous phase is known as the matrix while the discontinuous phase is known as the reinforcing phase or reinforcement. The reinforcing component or filler acts to improve the properties of the matrix. Therefore, the filler used is usually harder, stiffer and stronger than the matrix. There are many ways to manufacture composite materials which include compression molding, injection molding, pultrusion, extrusion and filament winding [9]. Normally, the constituent used in composite production are present at a reasonable proportion and exhibit different properties. The combinations of these different properties will later produce a composite material with enhanced properties compared to the properties of the individual constituent [8].

2.2 CHEMICAL COMPOSITION OF NATURAL FIBRE

Natural fibres can be obtained from various sources, be it from animal, minerals or plants [1]. Animal fibres are mainly composed of proteins. Examples of animal fibres include silk (derived from dried saliva of silkworms) and wool (sheep) [1]. As for mineral fibre, one of the examples is asbestos. Despite of its excellent thermal resistance, asbestos impose health hazards to humans [1].

Plant fibres on the other hand are composed of cellulose. Examples of plant fibres are cotton, jute, flax, sisal and hemp [1]. Plant fibres are categorized into seed fibre,

leaf fibre, bast or skin fibre, fruit fibre and stalk fibre. These classifications of plant fibres are based on the part from which the fibres are extracted. Natural fibres have complex structures. According to Wool [2], these natural fibres are generally lignocellulosic and consist of helically wound cellulose microfibrils in an amorphous matrix of lignin and hemicelluloses. The chemical composition of selected natural fibres is given in Table 2-1.

Table 2-1 Chemical composition of natural fibres [10,11].

Fibre	Cellulose (% wt)	Hemicelluloses (% wt)	Lignin (% wt)	Pectin (% wt)
Flax	71	18.6-20.6	2.2	2.3
Hemp	70-74	17.9-22.4	3.7-5.7	0.9
Jute	61-71.5	13.6-20.4	12-13	0.2
Kenaf	45-57	21.5	8-13	3-5
Ramie	68.6-76.2	13.1-16.7	0.6-0.7	1.9
Nettle	86			
Sisal	66-78	10-14	10-14	10
Henequen	77.6	4-8	13.1	
Banana	63-64	10	5	
Abaca	56-63		12-13	1
Oil palm empty fruit bunch	49.6-65	18	19-21.2	
Oil palm mesocarp	60		11	
Cotton	85-90	5.7		0-1
Coir	32-43	0.15-0.25	40-45	3-4
Cereal straw	38-45	15-31	12-20	8

2.2.1 Cellulose

Cellulose is an organic compound produced by plant and it is the basic structural component of all plant fibres. Cellulose molecules consist of glucose units linked together in long chains [12]. These long chains are linked together in bundles known as microfibrils [12]. The cellulose microfibrils have a very high tensile strength, theoretically estimated to be 7.5 GPa [12]. These cellulose microfibrils are highly concentrated at the second layer of the secondary wall of the fibre wall structure, which made up almost 50% of the cell wall structure, therefore giving the fibres a very high tensile strength.

2.2.2 Hemicellulose

Hemicellulose is a group of polysaccharides that is bonded together in relatively short, branching chains [12]. It is highly related to the cellulose microfibrils, embedding them together in a matrix [12]. Hemicellulose is hydrophilic in nature where it contains many sites at which water can readily be bonded [12].

2.2.3 Lignin

Lignin is a three-dimensional polymer having an amorphous structure with high molecular weight [12]. Lignin is the compound which provides rigidity to the plant [12]. In the absence of lignin, plants such as trees will not be able to grow and reach great heights. Compared to cellulose and hemicelluloses, lignin has low affinity for water and it is thermoplastic, where it softens and starts to flow at temperatures around 90°C and 170°C respectively [12].

2.2.4 Pectins

Pectins are the major constituent of the primary cell walls. They are the most complex polysaccharides in plant cell walls with regards to the structure and functions [13]. Pectins play a great role in determining the growth of a plant, the morphology and the plant development [13]. Pectins also act as the defense mechanism against wounding, stress and infections caused by plant pathogens [13].

2.2.5 Wax

Wax is the material which makes up the part of fibre that can only be extracted using organic solution [1]. It is a fatty substance which is usually found in the cell walls of cells on the outside surface of the plant. It acts to provide water proof surface to the plant in order to prevent evaporation and dehydration.

2.3 MECHANICAL PROPERTIES OF NATURAL FIBRE

In general, the cellulose content and the microfibrils angle of the fibres play a major role in affecting the strength and stiffness of the fibres [1]. The cellulose content determines the crystallinity index and the microfibrils angle of the fibre [1]. Fibres with high crystallinity index and/or cellulose content generally exhibit greater mechanical properties compared to fibres with low cellulose content [1]. For example, hemp with 70-74 %wt cellulose content has tensile strength of 690 MPa, while sisal which has 66-78 %wt of cellulose possesses lower tensile strength of 511-635 MPa. The mechanical properties of natural fibres as compared to synthetic fibres are shown in Table 2-2.

Table 2-2 Mechanical properties of natural and synthetic fibres [14].

Fibre	Density (g/cm³)	Elongation (%)	Tensile strength (MPa)	Young's modulus (GPa)
Natural:				
Cotton	1.5-1.6	7.0-8.0	287-597	5.5-12.6
Jute	1.3	1.5-1.8	393-773	26.5
Flax	1.5	2.7-3.2	345-1035	27.6
Hemp	-	1.6	690	-
Ramie	-	3.6-3.8	400-938	61.4-128
Sisal	1.5	2.0-2.5	511-635	9.4-22.0
Coir	1.2	30.0	175	4.0-6.0
Viscose (cord)	-	11.4	593	11.0
Soft wood kraft	1.5	-	1000	40.0
Synthetic:				
E-glass	2.5	2.5	2000-3500	70.0
S-glass	2.5	2.8	4570	86.0
Aramide (normal)	1.4	3.3-3.7	3000-3150	63.0-67.0
Carbon (standard)	1.4	1.4-1.8	4000	230.0-240.0

2.4 LITERATURE REVIEW

The use of natural fibres as reinforcing agent or filler in thermoplastic matrices, replacing the utilization of synthetic fibres has become the subject of interest among researchers. A number of studies have reported successful incorporation of these natural fibres into elastomers and thermoplastics [2-4, 10-11, 14-26, 29].

Abundantly available natural fibres such as oil palm empty fruit bunch and sawdust can be used as filler in producing bio composite material. Different types of natural fibre differ in their chemical composition. Thus, they exert different effects on the resultant composite. However, other parameters such as fibre loading, particle size, surface treatment and manufacturing process also affect the physical and mechanical properties of the natural fibre-filled composite. Therefore, extensive studies have been conducted by researchers with regards to the effects of different type of natural fibre on the mechanical properties of the resultant composite.

2.4.1 Oil Palm Empty Fruit Bunch (EFB)

M.Zuhri et al. [15] conducted a study on the mechanical properties of short random oil palm fibre reinforced epoxy composites. The composites were fabricated by hand lay-up techniques and the volume fractions of oil palm fibre used was varied at 5 vol%, 10 vol%, 15 vol% and 20 vol%. Results obtained showed that as the fibre loading was increased, the tensile and flexural properties decreased. It was observed that the highest tensile properties was achieved at 5 vol% of fibre loading and the addition of more than 5 vol% of fibre did not have any significant effect to the flexural properties of the composite.

Another researcher, M. Khalid et al. [16] studied the mechanical properties of the polypropylene (PP)-cellulose (derived from oil palm empty fruit bunch) and PP-oil palm empty fruit bunch (OPEFB) bio composites, taking into consideration the effect of trimethylolpropane triacrylate (TMPTA) as the coupling agent. The fibre loading was fixed at 30% while the coupling agent concentration is varied from 2.0 to 7.0 wt%. Results obtained showed that the mechanical properties of PP-cellulose bio composite was improved at 2.0 wt% of TMPTA concentration whereby there was no significant improvements in the mechanical properties of PP- OPEFB bio composites.

2.4.2 Sawdust

Another commonly used natural fibre that has been incorporated in the production of bio composite is the sawdust. Jutarat Prachayawarakorn and Kanita Anggulalat [17] studied the influence of meranti sawdust aspect ratios and amount of loadings on mechanical and morphological properties of composites from polypropylene and meranti sawdust. Results obtained showed increased in Young's modulus and flexural modulus in line with the increment of sawdust aspect ratio and the percentage of sawdust content. Jutarat Prachayawarakorn and Kanita Anggulalat [17] also studied the effect of maleic anhydride-grafted-polypropylene on meranti sawdust reinforced polypropylene and it was reported that there was improvement in adhesion between the fibre-matrix interfaces.

In another work [18], the mechanical properties of teak wood flour-reinforced HDPE composites were studied. The tensile and impact strength behavior of teak wood flour (TWF)-filled HDPE composites were evaluated at 0 – 0.32 volume fraction of TWF [18]. Results obtained showed that the tensile modulus and strength initially increased up to 0.09 volume fraction of TWF, whereas a decrease is observed with further increase in the volume fraction of TWF [18]. Kamini Sewda et al. [18] also reported increased in the tensile modulus and strength of teak wood flour-reinforced HDPE, in the presence of maleic anhydride-grafted HDPE (HDPE-g-MAH).

2.4.3 Other natural fillers

Other natural fibres that have also captured the researchers' interest include the wheat straw fibres, sunflower hull sanding dust (SHSD), coir fibre, bagasse, henequen fibre, bamboo and rice husk. The introduction of these natural fibres into the polymer matrix have shown enhancement in the mechanical properties of the resultant composite. Panthapulakkal et al. [19] reported that composites of polypropylene filled with 30% wheat straw fibres extracted by both mechanical and chemical processes have shown enhanced mechanical properties compared to virgin polypropylene. On the other hand, W.H. Zhong et al. [20] conducted a study on plant fibre reinforced polymer composite prepared by a twin-screw extruder where polypropylene (PP) was reinforced with sunflower hull sanding dust (SHSD).

Results obtained showed that the addition of SHSD into the PP demonstrated improvement in the flexural strength and flexural modulus of the composites.

Research on coconut fibre or also known as coir has been carried out by quite a number of researchers. Huang Gu and Wang Wei [21] in their research on characterization and utilization of natural coconut fibres composites reported that 60% volume fraction of coir fibre is the best amount that should be used for reinforcement in rubber matrix. Volume fraction higher or lower than 60% will eventually caused reduction in the tensile strength of the composites. They also studied the effects of composite manufacturing process to the strength of the produced composite. Huang Gu and Wang Wei [21] reported that coir fibre-reinforced rubber composite fabricated using the heat press technique with temperature range of 130-160 °C did not effect the tensile strength of the composites.

Meanwhile, M. Brahmakumar et al. [3] studied the effect of natural waxy surface layer of coconut fibre on fibre-matrix interfacial bonding and strength of the composite. Results showed that the coconut fibre waxy layer provided good fibre-matrix bond such that removal of the layer caused decreased in tensile strength and modulus of the composite.

Sugar cane waste or also known as bagasse has also found its application in the composite fabrication. Researchers have carried out experiments to study the characteristics of this bagasse fibre when used as reinforcement in composite. M. Mariatti et al. [22] studied the effect of fibre loading on the properties of bagasse fibre-reinforced unsaturated polyester composites. Results obtained showed that as the fibre loading increased, the tensile and flexural properties of the bagasse fibre-reinforced polyester composites also increased. It was also reported that the mechanical properties of acrylic acid treated bagasse fibre was better compared to sodium hydroxide treated bagasse fibre. Daniella Regina Mulinari et al. [23] in another study examined the effects of fibre modification using zirconium oxychloride on the mechanical properties of the bagasse reinforced high density polyethylene composites. Compared to the non-modified bagasse fibre composite, the modified fibre composites showed better tensile strength. The modified bagasse

composite also reduced the composite elongation by 26% and increased the tensile modulus by 50%.

P.J Herrera Franco et al. [4] conducted a study on short henequen fibres reinforced HDPE composite. The study focused on the effect of fibre-matrix adhesion to the mechanical properties of the composite. The study involved surface treatments which consisted of alkali treatment, silane coupling agent and the pre-impregnation process of the HDPE/xylene solution. The presence of Si-O-cellulose and Si-O-Si bonds on the lignocellulosic surface confirmed that the silane coupling agent was efficiently held on the fibres surface through both condensation with cellulose hydroxyl groups and self-condensation between silanol groups. Results obtained showed that silane treatment produced significant increase in tensile and flexural strength of the HDPE-henequen fibre composite, while the tensile and flexural modulus remained unaffected. Similar trend in mechanical properties enhancement was also observed by other researchers. Ismail et al. [24] reported that the presence of bonding agent in bamboo-fibre-reinforced natural rubber composites resulted in improved mechanical properties of the composites. Mohd Shahril Ezuan Mustapa et al. [25] studied the mechanical properties of polypropylene rice husk composite. Improvement of 35% in flexural strength was observed with the addition of 4 wt% of maleic anhydride grafted polypropylene into the composites with 30 wt% rice husk.

CHAPTER 3

METHODOLOGY

3.1 MATERIALS

3.1.1 Polymer Matrix

High-density polyethylene (Titanzex® HDPE) was supplied by Titan Petchem (M) Sdn. Bhd., Pasir Gudang, Johor, Malaysia. The HDPE has a mass flow index of 7 g/10 min with a density of 0.961 g/cm³.

3.1.2 Natural Fibres

Oil palm empty fruit bunch (EFB) and sawdusts were obtained locally. All the fibres were ground and then sieved into 425 µm using ELE International Lab Test Sieve. Finally, all the fibres were dried in a hot air oven at 80°C for 24 hours to remove the moisture content.

3.2 FIBRE SURFACE TREATMENT

The fibres were treated with 3 %wt of silane coupling agent in an ethanol/water (60/40) solution for 1 h under agitation. The pH of the solution was adjusted to 3.5 – 4 with acetic acid. Then, the fibres were dried in an oven at 80°C for 24 h. For the purpose of this study, vinyltriethoxysilane (VTS) was used. The silane was supplied by Dow Corning Corp.

3.3 COMPOSITE PREPARATION

The specimens were prepared in two steps, firstly compounding followed by injection molding. The compounding was done manually where HDPE were mixed together with the natural fibre at different loading (0, 10, 20 and 30 %wt) by hand. The compounds were then molded using Tat Ming ME20 III injection molding machine in order to produce dog-bone shaped specimens for mechanical testing. Injection pressure of 80 bar and temperatures set at 120°C (zone 1) and 180°C (zone 2 and 3) were used for the molding process.

3.4 MECHANICAL TESTING

Both tensile and flexural tests were performed according to ISO 527-2 and ISO 178 respectively, using Lloyd Instrument Universal Testing Machine Model LR5K, equipped with a 5 kN load cell. The cross-head speed of 5 mm/min was used for the tensile test. For the flexural test (three point bending), a cross-head speed of 2 mm/min was used. Five identical samples of each natural fibre composite were prepared for the purpose of the tensile and flexural testing.

3.5 MORPHOLOGY STUDY

The tensile and flexural fractured surfaces of the specimens were used to illustrate the morphology of the composite with different types of fibres. This analysis was performed using Model 7353 LEO 1430VP Scanning Electron Microscope (SEM) with the resolution of 137 eV at 5.9 KeV.

3.6 PROJECT ACTIVITIES

The project activities were executed based on the Gantt chart attached in Appendix A.

CHAPTER 4

RESULT AND DISCUSSION

4.1 MORPHOLOGICAL CHARACTERIZATION OF NATURAL FIBRES

The morphology and structural characteristics of the surface of each natural fibres used in this study was investigated using the scanning electron microscopy (SEM). The surface of the natural fibres was examined for any morphological changes at three different conditions which include:

- a) before drying (initial condition)
- b) after drying (fibres were oven dried at 80°C for 24 hours)
- c) after treatment with silane

4.1.1 Oil Palm Empty Fruit Bunch (EFB)

The scanning electron micrographs of the EFB surfaces before and after drying revealed some changes on the fibre surface structure. Figure 4-1 (a) shows the OPEFB fibre surface at magnifications of 500X before being dried. The image shows the distribution of stomata and microscopic pores on the fibre surface. The porous surface morphology is useful because it provides better mechanical interlocking between the fibre and the matrix for composite fabrication [26]. However, there were changes in the surface condition of the dried EFB fibres. The pores became smaller in size and less visible as shown in Figure 4-1 (b). The micrograph of silane treated fibre surface is shown in Figure 4-1 (c). It was observed that the pores have become more prominent after the fibre treatment [26]. The surface of the fibre also turned out to be rougher and more textured compared to untreated fibres, providing better surface for fibre-matrix adhesion.

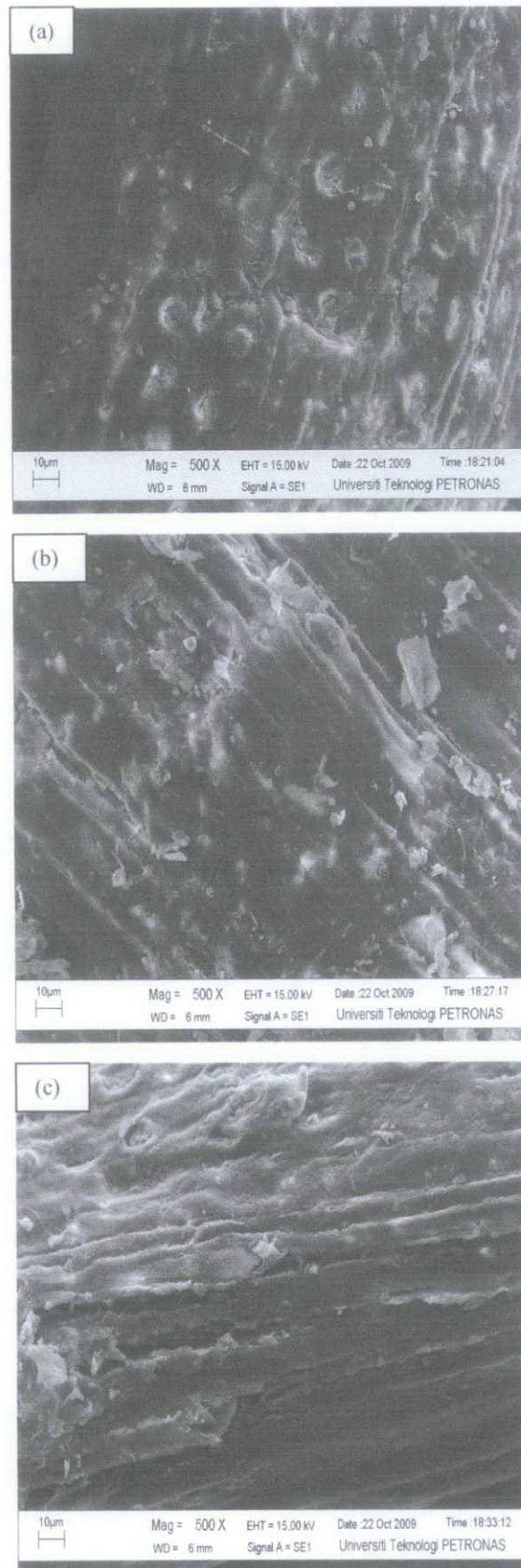


Figure 4-1 Scanning electron micrographs of oil palm empty fruit bunch surface (500X) (a) before drying, (b) after drying, (c) after 3 %wt silane treatment.

4.1.2 Sawdust

The scanning electron micrographs of sawdust were obtained for three different conditions. Changes in the structure of the sawdust are shown in Figure 4-2 (a), (b) and (c). Before being dried, the sawdust surfaces show extensive deformation of tracheids where irregular torn ends was observed [27]. According to a study conducted by L. Donaldson et al. [27] on three-dimensional imaging of a sawn surface using scanning electron microscopy, it was reported that the tracheids of earlywood surfaces were highly compressed. The compressed surface formed flat sheets of cell wall that were either bent over or partially erected as shown in Figure 4-2 (a). After undergoing drying process, where removal of water or moisture from the sawdust took place, the sawdust surface became more oriented and properly arranged (Figure 4-2 (b)). Surface modifications of the fibres by silane treatment produced image as shown in Figure 4-2 (c). The sawdust surface was rougher with deep grooves, promising a better interlocking surface between the fibre and matrix.

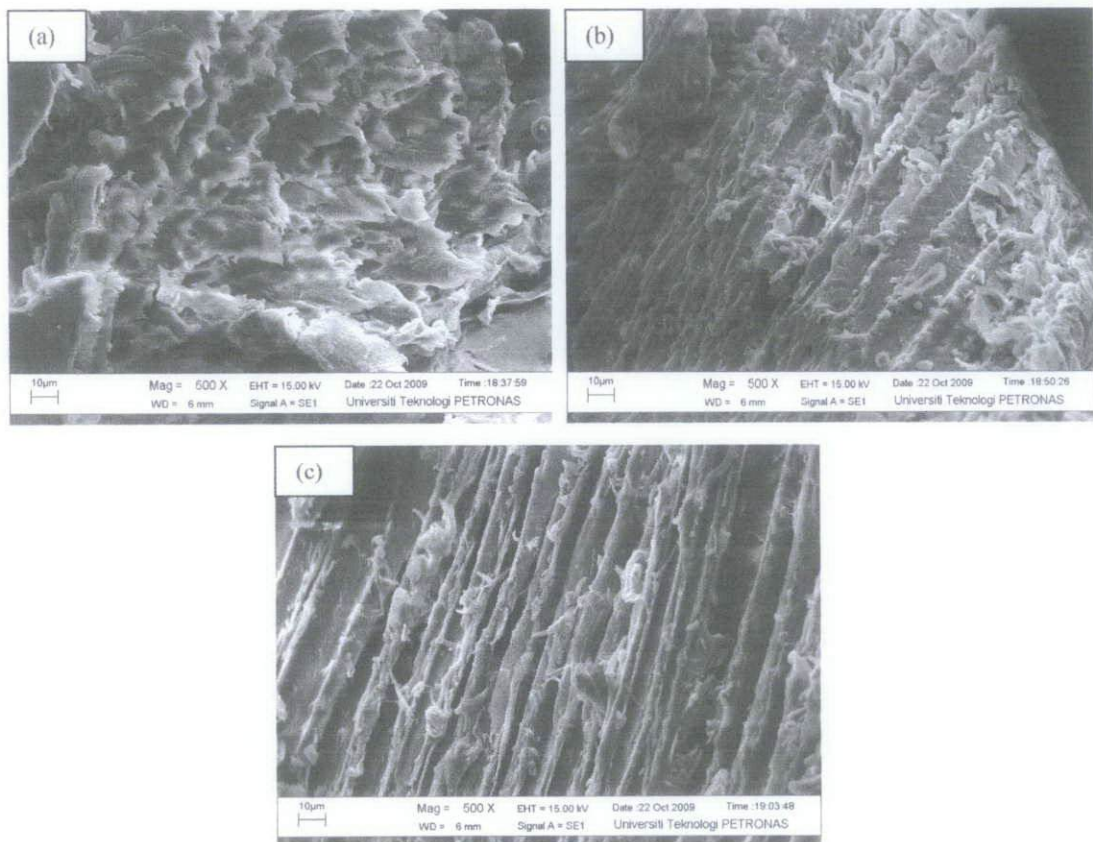


Figure 4-2 Scanning electron micrographs of sawdust surface (500X)
(a) before drying, (b) after drying, (c) after 3 %wt silane treatment.

4.2 MECHANICAL PROPERTIES OF NATURAL FIBRE-FILLED HIGH DENSITY POLYETHYLENE (HDPE) COMPOSITE

The influence of different types of natural fillers on the mechanical properties of the resultant composite was investigated. The results presented in this section are based on the tensile test performed using the Lloyd Instrument Universal Tensile Machine equipped with a 5 kN load cell with cross-head speed of 5 mm/min. Five identical dog bone specimens of each natural fibre-filled composites were tested and the average values were reported. The detailed results of the test are attached in Appendix B.

4.2.1 Tensile Strength

The tensile strength of the EFB and sawdust-filled composite as a function of filler loading (%wt) is presented in Figure 4-3. In general, the introduction of filler was found to decrease the tensile strength of EFB-filled composite by 25% whereby 24% reduction was observed in sawdust-filled composite. The EFB and sawdust loading were varied from 10 to 30 %wt. Results obtained showed that varying the filler loading does not impart any significant effect on the tensile strength of both EFB and sawdust-filled composites.

A study [15] was conducted on the mechanical properties of short random oil palm fibre reinforced epoxy composites. Based on the study, it is observed that the tensile strength of pure epoxy is much higher than the reinforced composite. The result [15] supported the result of the current study where pure HDPE have higher tensile strength compared to the EFB and sawdust-filled composites.

This could be due to the incompatibility between hydrophilic fibres and the hydrophobic HDPE matrix. Both EFB and sawdust are lignocellulosic materials in which their surfaces are covered by cellulose, hemicelluloses and lignin which comes from polar hydroxyl group [28]. The incorporation of the polar lignocellulosic fibres into the non-polar HDPE resulted in increasing number of incompatible interfacial regions [28]. The presence of these weak interfacial regions resulted in

low stress transfer from the matrix to the filler materials thus weakening the composites [15].

The decrease in tensile strength value could also be contributed by the non-homogeneity of the composite specimens [21]. An example of the sawdust-filled HDPE composite specimen is as shown in Figure 4-4. From the figure, it can be seen that the coarse and fine sawdust fibres are not evenly distributed throughout the specimens. It is believed that this non-homogeneity condition has adverse effect on the tensile strength of the composite [21].

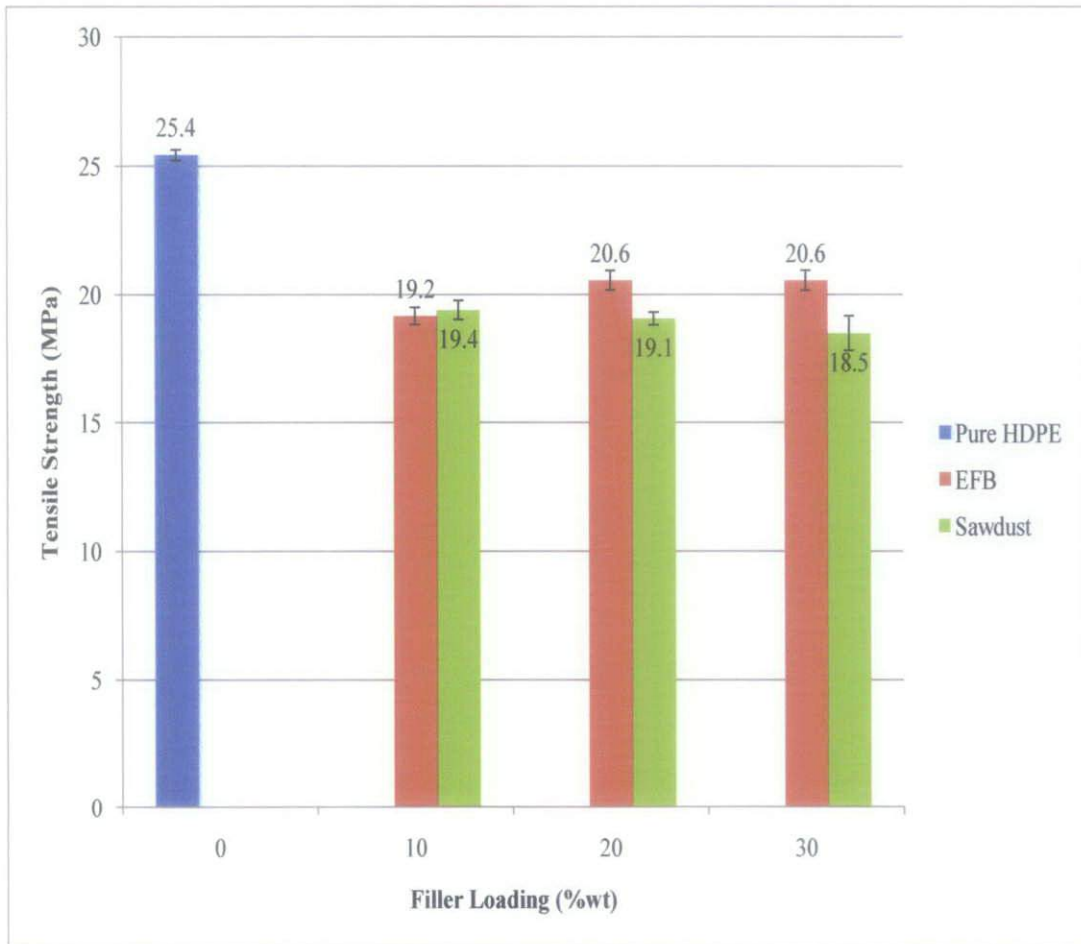


Figure 4-3 Tensile strength of EFB and sawdust-filled HDPE composite at different filler loading.



Figure 4-4 The sawdust-filled HDPE composite specimen.

4.2.2 Flexural Strength

Flexural strength can be defined as the ability of a material to resist deformation when subjected to load [29]. Based on Figure 4-5, the incorporation of filler was found to increase the flexural strength of EFB and sawdust-filled composites up to 11% and 14% respectively. Varying the filler loading from 10 to 30 %wt was found to increase the flexural strength of EFB-filled composite up to 10%. The result is vice versa for sawdust-filled composite where no significant improvements were observed in the flexural strength of the composite as the filler loading was varied.

As shown by various studies [15, 29], the increase in flexural strength is very unlikely to occur when fillers were incorporated into the matrix material. In contrast, results obtained from this current study showed that the introduction of filler caused the flexural strength of both EFB and sawdust-filled composites to increase. This may be due to increase in stress transfer from the matrix to the filler, thus increasing the stress at failure and eventually resulted in high value for flexural strength.

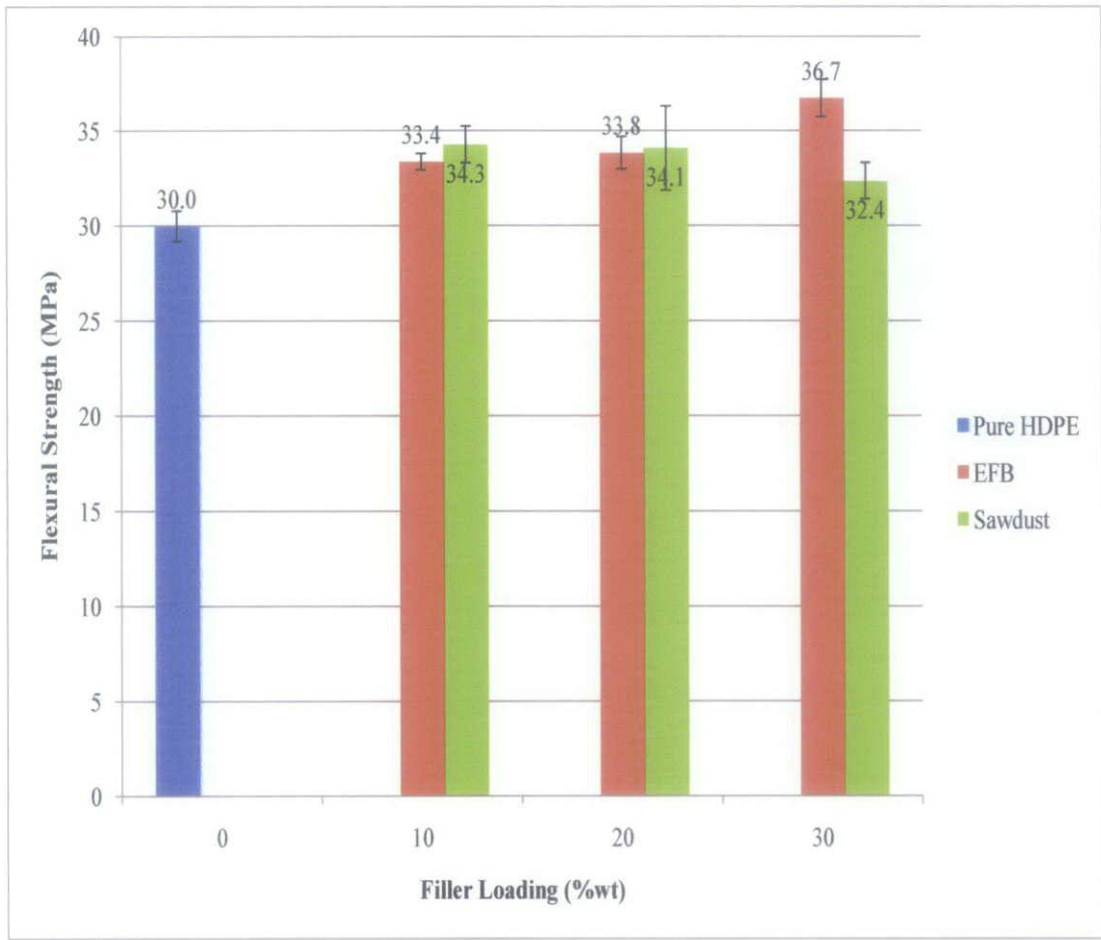


Figure 4-5 Flexural strength of EFB and sawdust-filled HDPE composite at different filler loading.

4.3 MORPHOLOGY STUDY OF THE COMPOSITES FRACTURED SURFACE

The tensile and flexural-fractured surface of both EFB and sawdust-filled composites were studied using the scanning electron microscope (SEM). The micrographs obtained reveal the interaction between the fillers and the polymer matrix.

4.3.1 Tensile-Fractured Surface

The micrographs of 30 %wt EFB-filled composite as shown in Figure 4-6 revealed the presence of fibre pull out. Fibre pull out occurrence is attributed by poor adhesion and dispersion between the matrix and fibre [15]. The micrographs in Figure 4-6 also show the presence of a small gap between the fibre and matrix. The

formation of the small gap was most probably due to incomplete wettability or bonding between the matrix and fibre during the compounding stage of the composite [15].

On the other hand, the tensile-fractured surface of 30 %wt sawdust-filled composite showed excessive amount of matrix tearing as shown in Figure 4-7. This indicated that the composite material possess ductile properties. During mechanical testing, the energy absorbed by the material is mainly transferred in matrix tearing formation and only limited amount of stress or energy is transferred to the sawdust [30]. As a consequence, the sawdust filler did not impart any significant reinforcing effect to the composite [30].

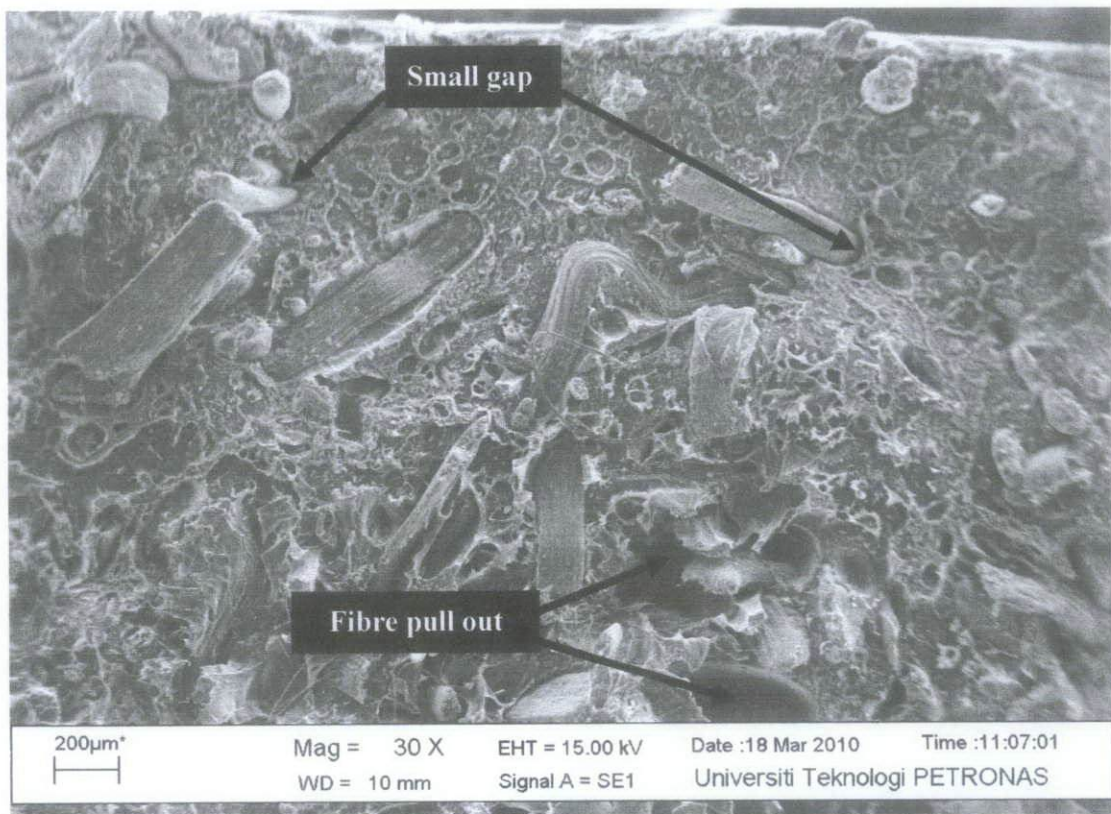


Figure 4-6 Scanning electron micrograph of tensile-fractured surface of 30 %wt EFB-filled composite (30X).

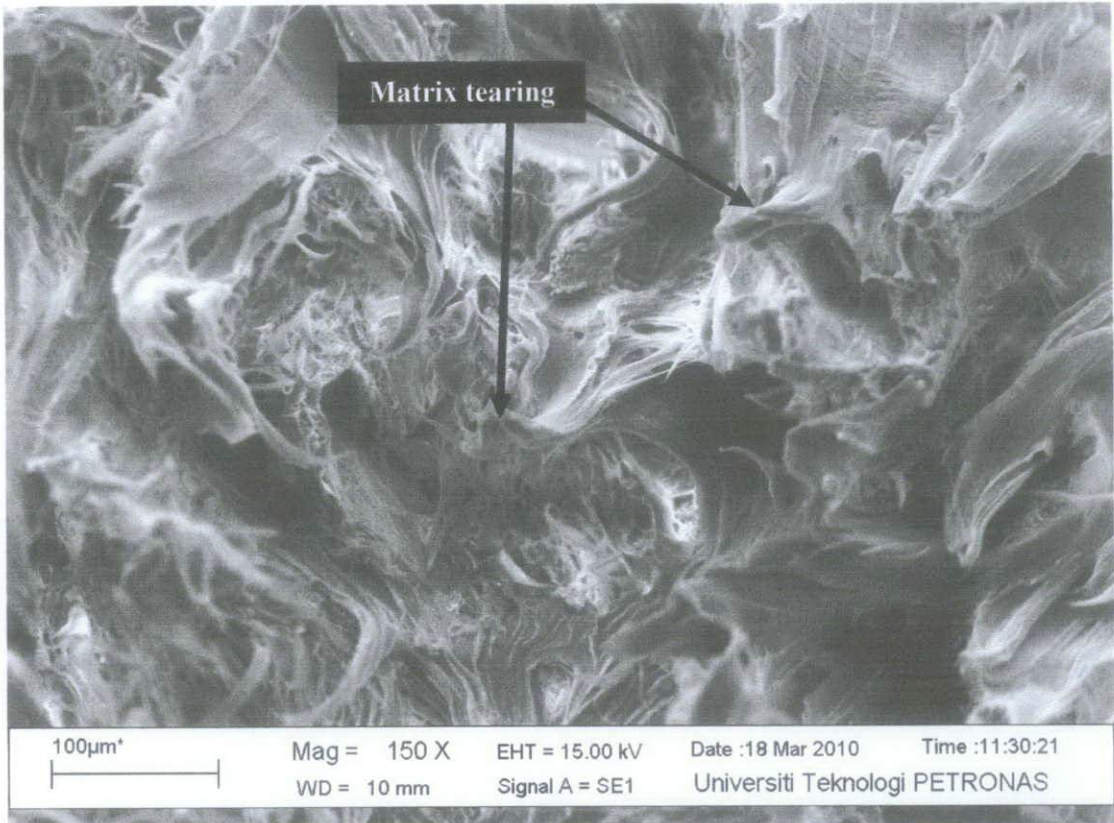


Figure 4-7 Scanning electron micrograph of tensile-fractured surface of 30 %wt sawdust-filled composite (150X).

4.3.2 Flexural-fractured Surface

The micrograph of 30 %wt EFB-filled composite as shown in Figure 4-8 revealed brittle behavior of the composite, characterized by the smooth fracture surface [29].

In contrast, Figure 4-9 showed the rough fracture surface of 30 %wt sawdust-filled composite. At high sawdust loading, poor particle wetting by the matrix resulted in weak interfacial adhesion between the filler and matrix material. Therefore, when subjected to load, crack propagates at a high rate [29]. The fast crack propagation path was distorted by the presence of sawdust particles thus producing the rough fracture surface [29].

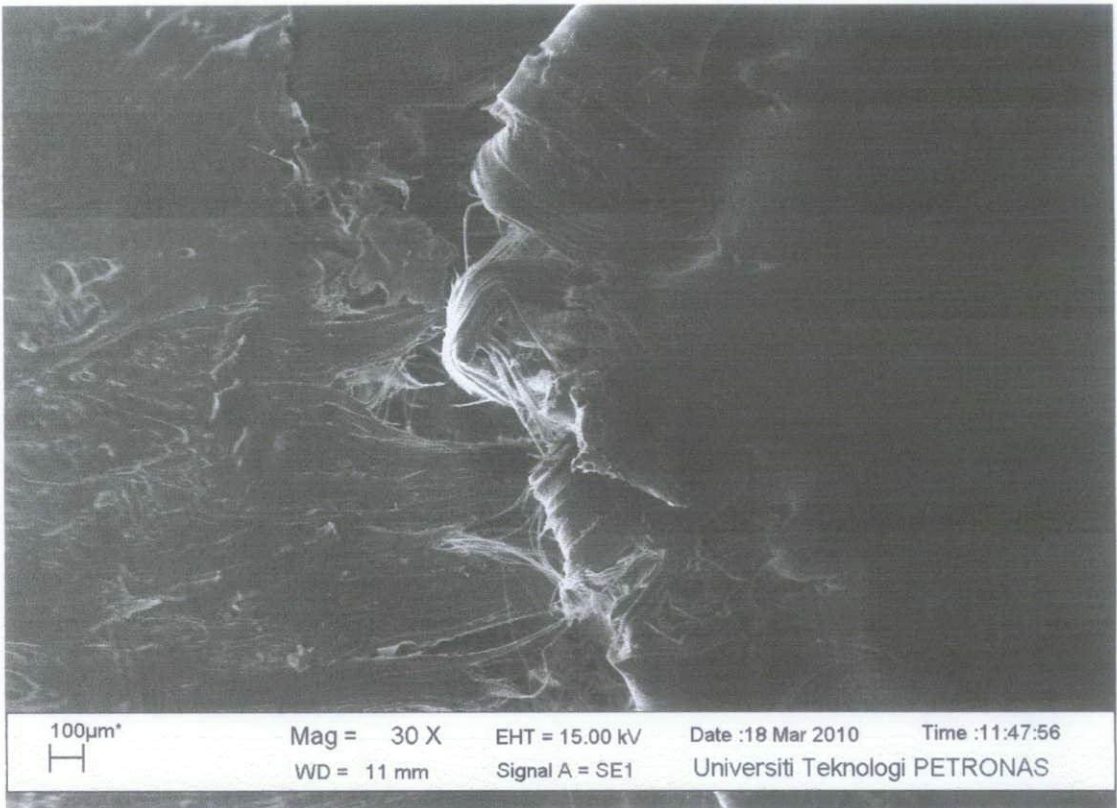


Figure 4-8 Scanning electron micrograph of flexural-fractured surface of 30 %wt EFB-filled composite (30X).

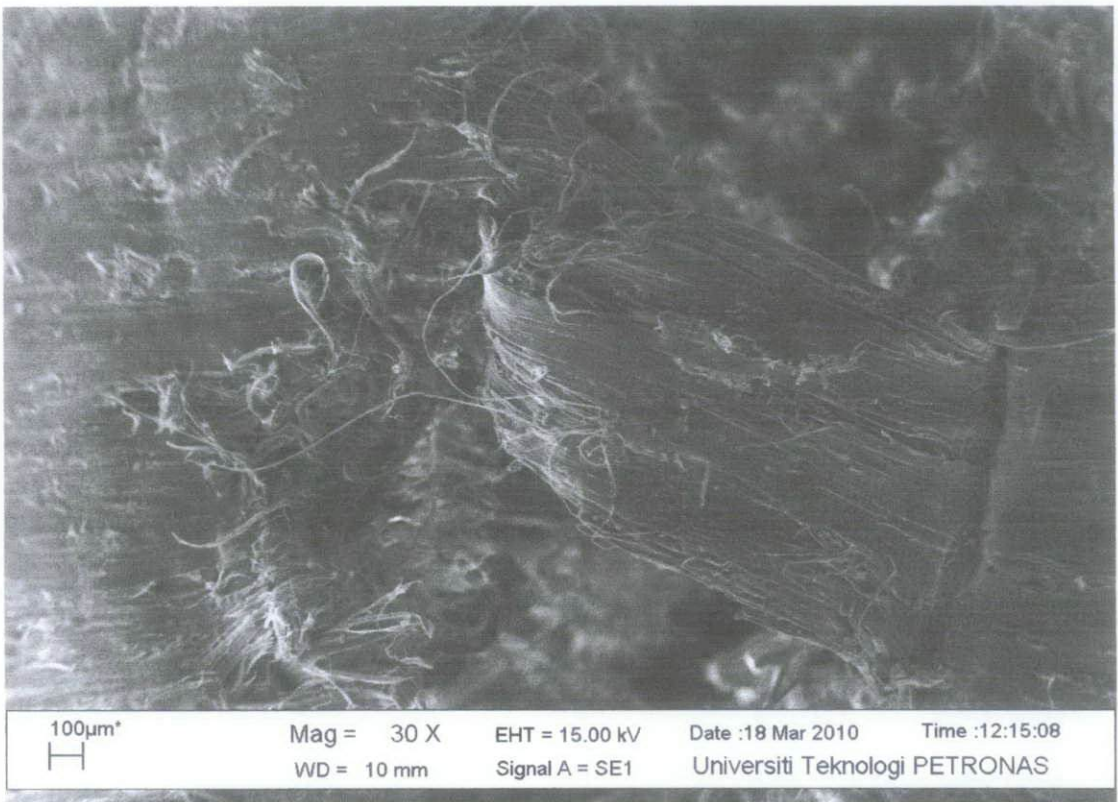


Figure 4-9 Scanning electron micrograph of flexural-fractured surface of 30 %wt sawdust-filled composite (30X).

CHAPTER 5

CONCLUSIONS AND RECOMMENDATION

5.1 Conclusions

The influence of different types of natural fibres and their loading on the tensile and flexural properties of natural fibre-filled HDPE composites were investigated. It was found that the incorporation of EFB and sawdust fillers with HDPE matrix resulted in decreased of the tensile strength of both types of biocomposites. No significant change was observed in the tensile strength of both EFB and sawdust biocomposites as the filler loading was varied from 10 - 30 %wt. However, EFB biocomposites was found to have higher tensile strength value compared to that of the sawdust biocomposites.

Results revealed that the incorporation of 10 %wt EFB and sawdust filler increased the flexural strength of both EFB and sawdust biocomposites up to 11 - 14% compared to that of pure HDPE. However, increasing the filler loading from 10 - 30 %wt did not influence the flexural strength of the sawdust biocomposites whereas the flexural strength of EFB biocomposites showed a marked increase of 22% compared to the flexural strength of pure HDPE as 30 %wt of filler was incorporated.

5.2 Recommendations

For the purpose of future work, extrusion process should be introduced as a mean of compounding the filler and matrix material in order to ensure good filler dispersion in the composite material.

On the other hand, in order to determine the suitability of the composite material to be used for future products development, it is feasible to produce the prototype of the product to enable further investigation on its mechanical properties.

REFERENCES

- [1] Navin, C., & Mohammed, F. 2008, *Tribology of natural fiber polymer composites*, England, Woodhead Publishing Limited. p. 2-10, 39-41, 180-182, 199.
- [2] George I. W., & Richard P. W., 2000, "Composites from natural fibers and soy oil resins," *Applied Composite Materials* **7**: p. 421-432.
- [3] M. Brahmakumar, C. Pavithran, & R.M. Pillai., 2005, "Coconut fibre reinforced polyethylene composites: effect of natural waxy surface layer of the fibre on fibre/matrix interfacial bonding and strength of composites," *Composites Science and Technology* **65**: p. 563-569.
- [4] Herrera-Franco, P.J. and A. Valadez-Gonzalez, 2005, "A study of the mechanical properties of short natural-fiber reinforced composites," *Composites: Part B* **36**: p. 597-608.
- [5] K.T. Lee, S.H. Shuit et al., 2009, "Oil palm biomass as a sustainable energy source: A Malaysian case study," *Energy* **34**: p. 1225-1235.
- [6] Huzaiifa bin Hashim. 2008, *The effect of palm oil fiber on concrete properties*, BSc. Thesis, Universiti Teknologi Malaysia. p. 8.
- [7] Ivy Tan Ai Wei. 2008, *Preparation, characterization and evaluation of mesoporous activated carbons derived from agricultural by-products for adsorption of methylene blue and 2,4,6-trichlorophenol*, Ph.D. Thesis, Universiti Sains Malaysia. p. 10-11.
- [8] F. L. Matthews and R. D. Rawlings. 1999, *Composite materials: Engineering and science*, England, Woodhead Publishing Limited. p. 1-5.
- [9] P.K. Mallick. 2007, *Fiber-reinforced composites: Materials, manufacturing*

and design, CRC Press. p. 377.

- [10] Bismarck, A., S. Mishra, and T. Lampke, 2005, "Plant Fibers as Reinforcement for Green Composites, in natural Fibers, Biopolymers, and Biocomposites," *A.K. Mohanty, M. Misra, and L.T. Drzal, Editors*, CRC Press. p. 37.
- [11] Khalil, H.P.S.A., et al., 2007, "Agro-hybrid Composite: The effects on Mechanical and Physical Properties of Oil Palm Fiber (EFB)/Glass Hybrid Reinforced Polyester Composites," *Journal of Reinforced Plastics and Composites* **26**(2): p. 203-217.
- [12] Sabu Thomas & Maya Jacob, 2008, "Biofibres and biocomposites," *Carbohydrate Polymers* **71**: p. 343-364.
- [13] Mohnen D., 2008, "Pectin structure and biosynthesis," *Current Opinion In Plant Biology* **11** (3): p. 266-77
- [14] J. Gassan & A.K. Bledzki, 1999, "Composites reinforced with cellulose based fibres," *Prog. Polymer Science* **24**: p. 221-274.
- [15] Mohd Zuhri et al., 2010, "Mechanical properties of short random oil palm fibre reinforced epoxy composites," *Sains Malaysiana* **39** (1): p. 87-92.
- [16] M. Khalid et al., 2008, "Effect of trimethylolpropane triacrylate (TMPTA) on the mechanical properties of palm fiber empty fruit bunch and cellulose fiber biocomposite," *Journal of Engineering Science and Technology* **3**: p. 153-162
- [17] Prachayawarakorn J. and Anggulalat K., 2003, "Influence of Meranti sawdust aspect ratios and amount of loadings on mechanical and morphological properties of composites from polypropylene and Meranti sawdust. *Songklanakarin Junior Science Technology* **25**(5): p. 595-606.

- [18] Kamini Sewda, S. N. Maiti, 2009, "Mechanical properties of teak wood flour-reinforced HDPE composites," *Journal of Applied Polymer Science* **112(3)**: p. 1826-1834.
- [19] Panthapulakkal S., Zereshkian A., Sain M., 2006, "Preparation and characterization of wheat straw for reinforcing application in injection molded thermoplastic composites," *Bioresource Technology* **97**: p. 265-272.
- [20] W.H. Zhong et al., 2009, "A plant fiber reinforced composite prepared by a twin-screw extruder," *Bioresource Technology* **100**: p. 1246-1251.
- [21] Huang Gu & Wang Wei, 2009, "Characterisation and utilization of natural coconut fibres composites," *Materials and Design* **30**: p.2741-2744.
- [22] M. Mariatti et al., 2008, "Effect of fiber surface treatment and fiber loading on the properties of bagasse fiber-reinforced unsaturated polyester composites," *Composite Science and Technology* **68**: p. 631-638.
- [23] Daniella Regina Mulinari et al., 2009, "Sugarcane bagasse cellulose/HDPE composites obtained by extrusion," *Composites Science and Technology* **69**: p. 214-219.
- [24] Ismail H., Edyham MR, Wirjosentono B., 2002, "Bamboo fibre filled natural rubber composites: the effects of filler loading and bonding agent," *Polymer Test* **21**: p. 139-144.
- [25] M. Shahril E. M., Azman H., and A. R. Rahmat. 2005, "Preliminary study on the mechanical properties of polypropelene rice husk composites" in Symposium Polimer Kebangsaan Ke-V. Retrieved 18 August 2009 from http://eprints.utm.my/3267/1/L14_spk05_fp_Mohd_Shahril.pdf.
- [26] S. Thomas et al., 1997, "Oil Palm Fibers: Morphology, Chemical Composition, Surface Modification, and Mechanical Properties," *Journal of*

Applied Polymer Science **66**: p. 821-835.

- [27] L.Donaldson et al., 2007, "Three-dimensional imaging of a sawn surface: a comparison of confocal microscopy, scanning electron microscopy, and light microscopy combined with serial sectioning," *Wood Science Technology* **41**: p. 551-564.

- [28] A.K. Mohanty, M. Misra and L.T. Drzal. 2005, *Natural fibers, biopolymers, and biocomposites*, CRC Press. p. 414.

- [29] Hazizan Md Akil et al., 2006, "The flexural and morphological properties of α -alumina filled epoxy composites," *Malaysian Polymer Journal (MPJ)*, Vol 1, No. 1, p. 25-38.

- [30] Muhamad Fatikul Arif., 2008, *Investigation of Oil Palm Empty Fruit Bunch Composites Based on High Density Polyethylene Matrix*, M.Sc. Thesis, Universiti Teknologi PETRONAS. p. 40.

APPENDICES

Gantt Chart

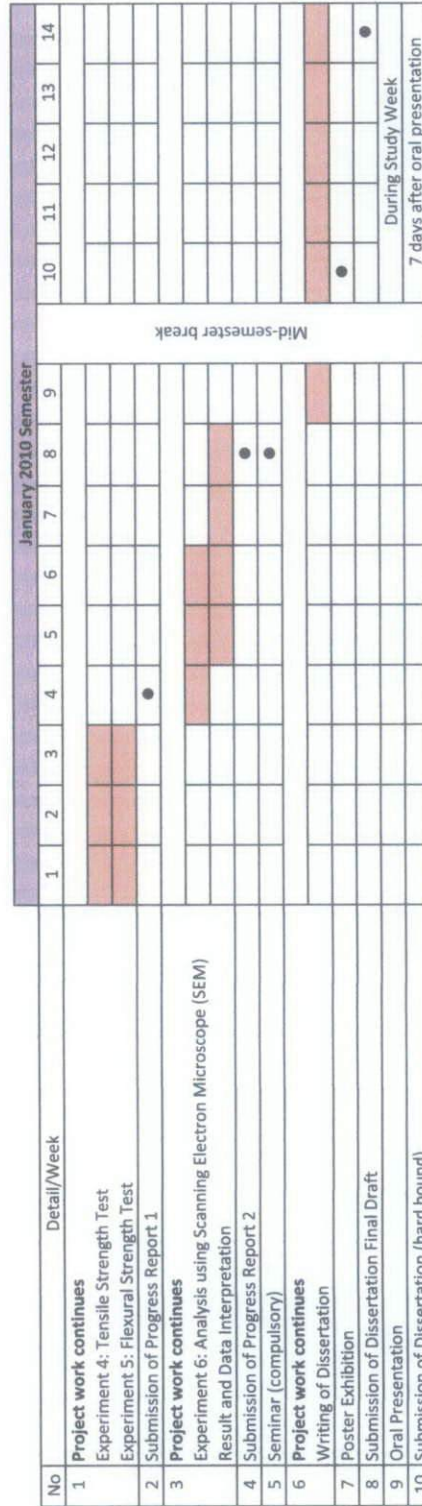
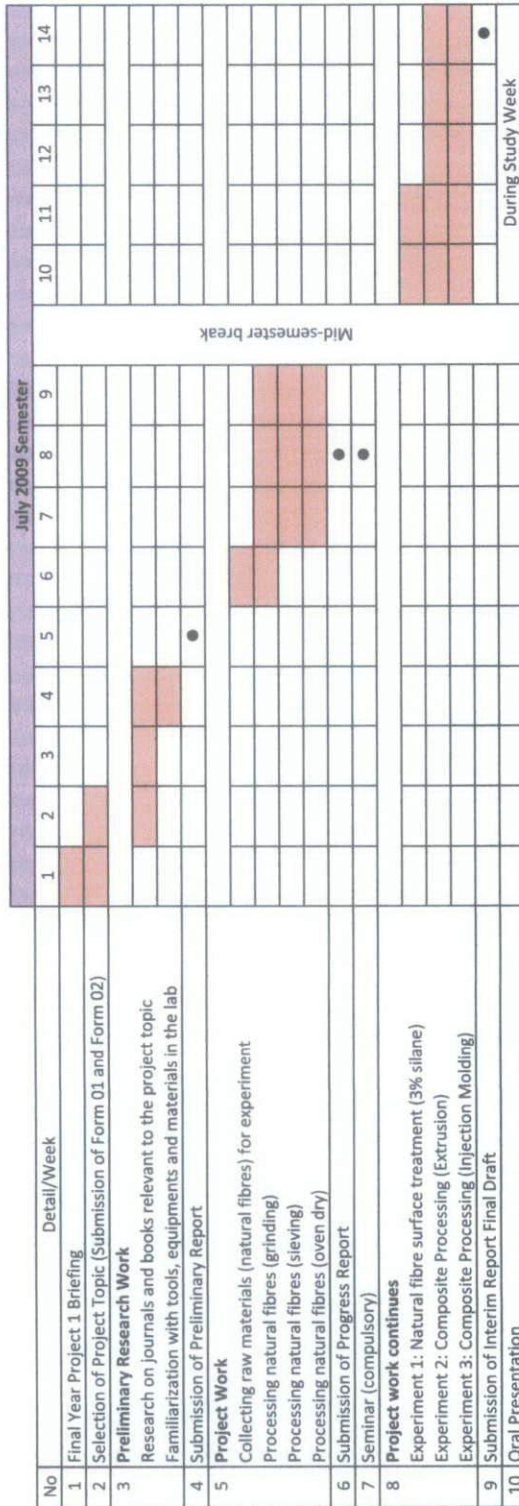


Table B-1 The mechanical properties of EFB-Filled HDPE composites with varying EFB loading.

EFB Filler Loading (%wt)	Mechanical Properties		
	Tensile Strength (MPa)	Flexural Strength (MPa)	
0	25.20796	29.16689	
	25.71603	29.72272	
	25.37685	30.74131	
	25.29377	29.40285	
	25.5998	30.90719	
	Mean	25.43888	29.98819
	STD	0.212684	0.790468
10	19.24405	33.93095	
	19.31417	32.97953	
	18.78578	33.28667	
	19.59775	33.6754	
	18.88687	32.96316	
	Mean	19.16572	33.36714
	STD	0.330506	0.428024
20	20.40711	33.57267	
	21.03625	33.20155	
	20.83691	33.2366	
	20.07519	33.88273	
	20.41932	35.27068	
	Mean	20.55495	33.83285
	STD	0.381305	0.850235
30	20.66131	36.64347	
	20.0959	36.54508	
	20.39493	35.23096	
	20.45589	37.29793	
	21.15304	37.90508	
	Mean	20.55221	36.7245
	STD	0.392154	0.999

Table B-2 The mechanical properties of sawdust-filled HDPE composites with varying sawdust loading.

Sawdust Filler Loading (%wt)	Mechanical Properties		
	Tensile Strength (MPa)	Flexural Strength (MPa)	
0		25.20796	29.16689
		25.71603	29.72272
		25.37685	30.74131
		25.29377	29.40285
		25.5998	30.90719
	Mean	25.43888	29.98819
	STD	0.212684	0.790468
10		19.09153	34.65033
		19.82278	35.71831
		19.59956	33.17257
		18.94372	34.03147
		19.52105	33.84811
	Mean	19.39573	34.28416
	STD	0.366221	0.959229
20		18.95229	35.83705
		18.85601	34.93063
		18.82931	35.23401
		19.29811	34.16886
		19.34891	30.25548
	Mean	19.05693	34.08521
	STD	0.248267	2.22358
30		17.43417	33.22835
		19.13318	33.14783
		18.34414	30.90351
		19.03512	32.49991
		18.44978	32.0076
	Mean	18.47928	32.35744
	STD	0.679694	0.953912