PREPARATION AND CHARACTERISATION OF HEAT-TREATED OIL PALM-EMPTY FRUIT BUNCH/HIGH DENSITY POLYETHYLENE COMPOSITES

NUR AFIFAH BINTI NORDIN

FACULTY OF SCIENCE
UNIVERSITI MALAYA
KUALA LUMPUR

2020

PREPARATION AND CHARACTERISATION OF HEAT-TREATED OIL PALM-EMPTY FRUIT BUNCH/HIGH DENSITY POLYETHYLENE COMPOSITES

NUR AFIFAH BINTI NORDIN

DISSERTATION SUBMITTED IN FULFILMENT OF
THE REQUIREMENT FOR THE DEGREE OF MASTER OF
SCIENCE

DEPARTMENT OF CHEMISTRY
FACULTY OF SCIENCE
UNIVERSITI MALAYA
KUALA LUMPUR

2020

UNIVERSITI MALAYA

ORIGINAL LITERARY WORK DECLARATION

Name of Candidate: **NUR AFIFAH BINTI NORDIN**

Matric No: **SGR150050**

Name of Degree: **MASTER OF SCIENCE**

Title of Project Paper/Research Report/Dissertation/Thesis ("this Work")

PREPARATION AND CHARACTERISATION OF HEAT-TREATED OIL PALM-EMPTY FRUIT BUNCH/HIGH DENSITY POLYETHYLENE COMPOSITES

Field of Study: **CHEMISTRY (MATERIALS CHEMISTRY)**

I do solemnly and sincerely declare that:

(1) I am the sole author/writer of this Work;
(2) This Work is original;
(3) Any use of any work in which copyright exists was done by way of fair dealing and for permitted purposes and any excerpt or extract from, or reference to or reproduction of any copyright work has been disclosed expressly and sufficiently and the title of the Work and its authorship have been acknowledged in this Work;
(4) I do not have any actual knowledge nor do I ought reasonably to know that the making of this work constitutes an infringement of any copyright work;
(5) I hereby assign all and every rights in the copyright to this Work to the University of Malaya ("UM"), who henceforth shall be owner of the copyright in this Work and that any reproduction or use in any form or by any means whatsoever is prohibited without the written consent of UM having been first had and obtained;
(6) I am fully aware that if in the course of making this Work, I have infringed any copyright whether intentionally or otherwise, I may be subject to legal action or any other action as may be determined by UM.

Candidate's Signature Date:

Subscribed and solemnly declared before,

Witness's Signature Date:

Name:

Designation

PREPARATION AND CHARACTERISATION OF HEAT-TREATED OIL PALM-EMPTY FRUIT BUNCH /HIGH DENSITY POLYETHYLENE COMPOSITES

ABTRACTS

Natural fibre composites (NFCs) are composite materials, in which the reinforcing fibres are derived from renewable and carbon dioxide neutral resources such as wood or plants. NFC can be made by replacing synthetic fibres with various types of natural fibres. Oil palm empty fruit bunch (OPEFB) fibre and oil palm mesocarp fibre are two important types of fibrous materials left in the palm-oil industry. In this study, EFB will be used as a reinforcement to the high-density polyethylene (HDPE). EFB was heat-treated at 180°C using vacuum oven for one hour, extrusion compounded with HDPE at 10%, 20% and 30% weight fraction. The composites then were injection moulded into dumb-bell and bar shaped specimens using a Boy® 55M injection moulding machine. All of the composites were exposed to different environments and the effects of conditioning were discussed relative to the sample in dry as moulded condition as reference. The effects of composition and heat treatment on the thermal properties of composites were investigated using Thermogravimetric Analysis (TGA) and Differential Scanning Calorimetry (DSC). The tensile and flexural tests were also done using an Instron Universal Testing Machine. The impact test was run under Charpy mode using an Instron Dynatup 9210 instrumented falling weight impact tester with a V-shape impactor tup. TGA shows an increase in degradation peak temperature of the heat-treated composites. DSC results revealed that EFB had no significant effect on the melting behaviour of the composites. However, an increasing trend in the degree of crystallinity (X_c) of the composite is observed as the heat treated EFB was used in the system. An increment in the tensile modulus and tensile strain were observed for the treated fibre composites due to the improved wetting between fibre and matrix. Hence, leading to better interfacial adhesion between them. In addition, the tensile strength value was increased for treated fibre composites with lower fibre loading. In impact tests, the fracture energy (W) of composites increased with increasing of fibre content. However, the critical strain energy release rate decreases with increasing of fibre content. In addition, the peak load (P) and critical stress intensity factor (K_c) increased with fibre contents. Moreover, the value of P, W, K_c and G_c increased with fibre treatment. All composites absorb moisture to various extents with the untreated fibre composites show the highest value of absorption. Thermal and mechanical properties reduced after soil burial on the composites. For that condition, composites made from heat treated EFB shows better thermal stability due to the improved compatibility between fibres and matrices, thus lowering the moisture intake, hence improve better thermal stability. Mechanical properties reduced due to attack of microorganisms that promote further composite degradation rate. The treated fibre composites showed more resistance towards the environment. However, indoor exposure has no significant effect on thermal and mechanical properties of composites. The MFI values of untreated and treated fibre composites decreased with the increase of EFB fibre loading in the system

Keywords: Heat treatment, Empty fruit bunch; Natural fibre composites, Extrusion and injection moulding

PENYEDIAAN DAN PENCIRIAN KOMPOSIT SERAT TANDAN KELAPA SAWIT DIRAWAT DENGAN HABA / POLIETILENA KETUMPATAN TINGGI

ABSTRAK

Komposit serat semulajadi (NFCs) adalah bahan komposit, di mana gentian pengukuhan diperolehi daripada sumber neutral yang boleh diperbaharui seperti kayu atau tumbuh-tumbuhan. NFC boleh dibuat dengan menggantikan gentian sintetik dengan pelbagai jenis gentian semulajadi. Serat tandan buah kelapa sawit (OPEFB) dan serat mesokarp kelapa sawit adalah dua jenis gentian yang menjadi bahan buang di kilang sawit. Dalam kajian ini, EFB akan digunakan sebagai bahan penguat kepada polietilena ketumpatan tinggi (HDPE). EFB dirawat pada 180°C menggunakan ketuhar vakum selama satu jam, diekstrusi dan dikompaun dengan HDPE pada peratusan berat 10%, 20% dan 30%. Bahan komposit itu kemudiannya disuntik kepada bentuk *'dumb bell'* untuk diuji menggunakan mesin pengacuan suntikan Boy® 55M. Sampel komposit didedahkan kepada persekitaran yang berbeza dan kesannya dibincagkan dan dibandingkan dengan sampel kering sebagai rujukan. Kesan komposisi dan rawatan haba pada sifat-sifat terma komposit telah dikaji menggunakan Analisis Termogravimetri (TGA) dan ujian pengimbasan kalorimetri (DSC). Sifat tegangan dan lenturan juga diuji dengan menggunakan Mesin Ujian Universal Instron. Ujian impak dijalankan menggunakan Instron Dynatup 9210. TGA menunjukkan peningkatan suhu puncak degradasi bagi komposit yang mengandungi EFB yang dirawat berbanding komposit yang tidak dirawat. Keputusan ujian DSC menunjukkan bahawa EFB tidak mempunyai kesan yang ketara terhadap suhu lebur komposit. Walaubagaimanapun, nilai penghabluran didapati semakin meningkat apabila EFB yang telah dirawat digunakan didalam sistem komposit. Kenaikan dalam modulus tegangan dan pemanjangan tegangan diperolehi untuk komposit yang mengandungi gentian yang dirawat disebabkan oleh peningkatan perlekatan diantara gentian dan matrik. Hal ini menyebabkan peningkatan perlekatan antara muka yang lebih baik antara mereka. Selain itu, nilai kekuatan tegangan meningkat bagi komposit yang mengandungi gentian yang dirawat pada kuantiti yang rendah. Dalam ujian impak, nilai tenaga pematahan komposit meningkat dengan peningkatan kandungan gentian. Walau bagaimanapun, kadar lepas tenaga kritikal menurun dengan peningkatan kandungan gentian. Di samping itu, beban puncak (P) dan faktor keamatan tekanan kritikal (K_c) meningkat dengan kandungan gentian. Selain itu, nilai P, W, K_c dan G_c meningkat dengan rawatan gentian. Semua komposit menyerap kelembapan dengan komposit yang mengandungi gentian yang tidak dirawat menunjukkan nilai tertinggi penyerapan. Sifat terma dan mekanikal bahan komposit mengalami penurunan selepas bahan tersebut ditanam di dalam tanah. Dalam pendedahan tersebut, komposit yang mengandungi gentian dirawat menunjukkan kestabilan terma yang lebih baik kerana peningkatan keserasian antara gentian dan matriks yang menyebabkan penurunan pengambilan kelembapan dan seterusnya meningkatkan kestabilan haba. Sifat mekanikal menurun disebabkan oleh serangan mikroorganisma yang menggalakkan kadar pereputan bahan komposit. Komposit menggunakan gentian yang telah dirawat menunjukkan daya tahan terhadap alam sekitar yang lebih baik. Walau bagaimanapun, pendedahan tertutup tidak memberi kesan yang ketara terhadap sifat terma dan mekanikal komposit. Nilai-nilai MFI daripada komposit yang mengandungi gentian tidak dirawat dan dirawat menurun dengan peningkatan kuantiti EFB didalam sistem.

Kata kunci: Rawatan haba, serat tandan kelapa sawit, komposit gentian semulajadi, penyemperitan dan pengacuan suntikan

ACKNOWLEDGEMENTS

In the name of Allah, Most Gracious, Most Merciful

I would like to appreciate and thankful to my beloved supervisors, Prof. Dr. Aziz Hassan and Dr. Nor Mas Mira Abd Rahman, for their untiring patience and guidance during the whole journey of this research. I must thank to Prof. Dr. Rosiyah Yahya and Dr. Noordini Mohamad Salleh for indirectly involved and give assistance to me in this research journey.

I would like to convey my gratitude to the technical and support staffs, especially to Mr. Zulkifli Abu Hasan, Mr. Hafizie, Mr. Zaidi, for their continuous help and support to finish this research. I am also grateful to all the members of polymer and composite research group for contributed in many ways for the completion of this study.

I am thankful to the Government of Malaysia and the University of Malaya as the research done was funded by grant UMRG (RG343-15AFR), RU (RF025B-2018) and PG117-2016A. I am also thankful to the MyBrain15 (My Master) programme, which fund throughout this study.

Last but not least, I owe a special appreciation and gratitude to all my family members, especially my beloved parents for their continuous and endless support throughout this research journey. I must appreciate all my friends, who kept me in touch and support me indirectly with thoughts and always available when they were needed the most.

TABLE OF CONTENTS

CHAPTER 3: METHODOLOGY 49

LIST OF FIGURES

LIST OF TABLES

LIST OF SYMBOLS AND ABBREVIATIONS

a/D	Notch to depth ratio
ASTM	American Society for Testing and Materials
CFRP	Carbon fibres reinforced plastic
DSC	Differential scanning calorimetry
D_{Tp}	Derivative peak temperature
E	Young's modulus
EFB	Empty fruit bunch
FESEM	Field emission scanning electron microscopy
FTIR	Fourier transform infrared
G_c	Critical strain energy release rate
GFRP	Glass fibres reinforced plastic
GPa	Giga Pascal
HDPE	High density polyethylene
K_c	Critical stress intensity factor
LDPE	Low density polyethylene
MFI	Melt flow index
MPa	Mega Pascal
NFRC	Natural fibre reinforced composites
OPEFB	Oil palm empty fruit bunch
P	Peak load
PE	Polyethylene
PLA	Polylactic acid
PMC	Polymer matrix composites
PP	Polypropylene

PS	Polystyrene
PVC	Polyvinyl chloride
RPM	Revolutions per minute
S/D	Span to depth ration
SEM	Scanning electron microscopy
SEN	Single end notch
$T_{50\%}$	Temperature at 50% degradation
Tan δ	Tan delta
T_c	Crystallisation temperature
TGA	Thermogravimetric analysis
T_m	Melting temperature
Tonset	Onset temperature
UTM	Universal testing machine
UV	Ultra violet radiation
V	Volume
W	Energy to failure
W_f	Weight fraction
$wt_{\%}$	Weight percent
X_c	Degree of crystallinity
δ	Density
ΔH_c	Entalphy of crystallisation
ΔH_m	Entalphy of fusion

CHAPTER 1:INTRODUCTION

1.1 Polymer composites: An overview

A polymer matrix composite (PMC) is a composite material composed of a variety of short or continuous fibres bound together by an organic polymer matrix. PMCs are designed to transfer loads between fibres through the matrix. Some of the advantages with PMCs include their lightweight, high stiffness and their high strength along the direction of their reinforcements. Other advantages are good abrasion resistance and good corrosion resistance. During the sixties, the rise of composite materials began when glass fibres in combination with tough rigid resins could be produced on large scale. Throughout the world, more and more people understand that we need to sustainably manage our planet's resources and ecosystems. This awareness has been growing for about a century but has picked up momentum in the past decade. During the last decade, there has been a renewed interest in the natural fibre as a substitute for glass, motivated by potential advantages of weight saving, lower raw material price, and 'thermal recycling'. Renewed interest in the utilization of natural materials addresses ecological issues such as recyclability and environmental safety (Jiang et al., 2012). In these days, the natural fibres provide a fascinating substitute for the most widely applied reinforcing fibre in the composite technology, glass. On the other hand, natural fibres have their disadvantages, and these must be resolved to be comparable with glass. Low thermal stability, compatibility of natural fibres with polymer matrix, irregular in shape, poor resistance to moisture, weak interactions fibre-matrix adhesion is some of the drawbacks when using natural fibres in polymer matrix composites.

1.2 Applications of composite materials

The development of composite materials with properties stronger and lighter than the traditional materials such as aluminium, alloys and steel are making them an excellent choice in engineering industries. Moreover, composite materials possess many advantages such as lightweight, corrosion resistant and low cost. Applications of polymer composites can be divided into industries as follows.

1.2.1 Aerospace industry

The most common composites used are glass, carbon and aramid to manufacture the aerospace parts. Polymer composites are attractive because of its reduced airframe weight ranging from 20-50% enables fuel saving and therefore lowers its operating costs. Moreover, composites do not corrode like metals.

The most common composited of fibre glass was first used in the Boeing 707-passenger jet. In the jet, composites accounted for 2% of the structure. By contrast, composites on the 787 account for 50% of the aircraft's structural weight and composites make up about 25% of the total airframe of the Airbus A380 (Mrazova, 2013). Moreover, the reduction in weight of the aircraft makes them able to carry more weapons and increase its range, which makes more effective in combat missions (Bielawski, 2017). Initially, composite materials were used only in secondary structure, but as development of the materials has improved, their use in primary structure such as wings and fuselages has increased.

The leading usage of composite materials are in the military aircraft. Carbon and Glass Fibres Reinforced Plastic (CFRP and GFRP) are used commonly in modern military aviation especially in Lockheed Martin F-35 Lightning II. In this aircraft, composites used as vertical stabilizer, tail plane, flaps and wings. The typical mass reduction is around 25-30% by using composite materials as the components.

In addition, instead of usage in military aircraft, composite materials also been used in commercial aircrafts nowadays. There is a great potential to lower the weight and increase the payload of airplanes, thus reducing the fuel costs of the aircrafts, making the aircraft lighter. Thus, enable them to travel further and carry more passengers which is the important aspects and economically desirable for the airlines.

1.2.2 Oil and gas

Fibre-reinforced polymer composites have found extensive applications in the oil and gas industry for the last three decades. The global market of composites in oil & gas industry projected to reach USD 1.98 billion by 2021. The high cost in this industry when using steel piping in retrofit applications are driving the use of composites, which withstand the severe conditions experienced in offshore environment. In the offshore oil and gas industry, composites offer several potential advantages with the cost of manufacturing and erecting oil rigs could be significantly reduced if heavy metal pipelines be replaced with lighter ones which made of composites. The cost advantages of composite products are much greater when they replace expensive corrosion-resistant metals such as steel and aluminium. Due to their function as resistance to corrosion, they help in enhancing reliability and safety & leads to reduce life cycle costs. Composites used in this industry includes piping, ladder, gratings, flexible tubes, riser, high-pressure accumulator bottles and topside applications. Among these applications, pipes are the largest application of the composites in oil & gas industry. In addition, the use of composite pipes reduces the overall maintenance costs, thus reducing the maintenance costs of the overall installations.

The most common composites used is glass fibre due to its low cost of glass fibre and its superior properties such as strength, flexibility, durability, stability, lightweight, as well as resistance to heat, temperature, and moisture. However, epoxy resin-based oil and gas composites has the largest market share due to their resistance to corrosive liquids, excellent electrical insulation, good performance at elevated temperature, lower styrene emission in comparison to that of polyester resins, and good adhesion strength to different varieties of reinforcements.

1.2.3 Automotive industry

Similar in aerospace industry, polymer composites have high demand in automotive industry especially in Europe, whereby their special applications are in automotive industry. The global automotive polymer composites market size was valued at USD 6.40 billion in 2016 and is expected to reach USD 11.62 billion by 2025. In early 2000, the glass fibre reinforced composites were chosen because of their durability of automobile interior and exterior parts as well as cost effective compared to carbon fibres.

Moreover, glass fibre reinforced composites have good mechanical properties, light in weight and able to produce parts quickly with minimal manufacturing complexity. Therefore, they often be used in this industry to replace and lighten metallic components such as front-end bolsters, seat pans, bumper beams, battery trays, gears and engine. Increasing amount of component made from composites in automotive will provide vehicles with lighter weight, which will provide us energy efficiency, as well as fuel reduction. As commonly known, the lighter the vehicle, the less power required getting it moving and the less energy needed to maintain a constant speed. Therefore, it burns up less fuel by as much as 6-8 percent for every 10 percent reduction in poundage.

However, in the past few decades, car manufacturers and suppliers for door panels, seat backs, headliners, package trays, dashboards, and interior parts used the natural fibre composites. Natural fibres such as kenaf, hemp, flax, jute, and sisal offer such benefits as reductions in weight, cost, and CO_2, less reliance on foreign oil sources, and recyclability.

1.2.4 Construction and civil structure

The oldest engineering profession in the world is the civil engineering profession. Engineers have been involved over many centuries to build and maintain the basic infrastructure, from pyramids in Egypt to modern transportation systems. They design and supervise the construction of roads, airports, tunnels, bridges, water supply, sewage systems, and buildings. Therefore, it is called a sector that provides an essential service to society. Moreover, they are also concerned with the impact of their projects on the public and the environment, and they co-ordinate the needs of society with technical and economic feasibility. In addition, today, many challenges require building reinforced structures that can overcome natural disasters such as earthquakes and hurricanes. This requires the creative use of polymer composite materials in existing structures and structural systems. Composites are now successfully applied in making concrete structures more earthquake-resistant around the world. As a result, polymer composites can benefit most in construction and civil industry.

1.3 Problem statement

The increasing consumption of petroleum-based products and its negative impact on the environment has led to an increase in environmental consciousness when it comes to sustainable materials and products. As a renewable resource, natural fibres provide many benefits both to the environment and to those involved in the market that they create. Fibre reinforced composites have received much attention based on different applications because of the good properties and the advantages found over synthetic fibre. Moreover, natural fibre reinforced composites (NFRC) are well known as lightweight which results in a higher specific strength and stiffness than glass and low-cost material which makes the material an interesting product for low-wage countries. The construction industry has been one of the world's largest consumers of composite material in roofing, flooring, ceilings, beams and columns and even full hybrid construction systems.

This research is important due to high interest in using natural fibre as reinforcement materials in PMC. Empty fruit bunch (EFB) fibre was chosen in this research as reinforcement in the composites. EFB is the fibrous mass left behind after separating the fruits from sterilized (steam treatment at 294 kPa for 1 hour) fresh fruit bunches (EFB) (Shinoj et al., 2011). However, the wastes generated have created major disposal problem especially to the environment due to the open burning and it is being discouraged by the

Department of Environment in Malaysia few years back (Goh et al., 2010). Previously, the EFBs were burned and the ash is used as fertilizer, resulting in air pollution to the environment. To solve the issues, these oil palm wastes can be converted to a wide range of value-added products such as oil palm fronds, that can be processed and used as a roughage source for ruminants such as castles and goat (Shuit et al., 2009). Therefore, the need to optimize the use of EFB as to solve these problems are very important as feedstock for valuable products in the chemical, energy, and biomaterials sectors.

However, factors such as low thermal stability, compatibility of natural fib res with polymer matrix, irregular in shape, poor resistance to moisture, weak interactions fibre-matrix adhesion are some of the drawbacks when using natural fibres in polymer matrix composites. The main concern of using natural fibre in processing of composites are restricted maximum processing temperature. Natural fibres tend to degrade when exposed to high temperature. Therefore, it is suggested that the maximum processing temperature of most of the natural fibres are 150°C because natural fibres are thermally unstable and may degrade between 150-220°C. It is advised that temperatures above 150°C can lead to permanent modifications of the physical and chemical properties of the natural fibres (Santulli et al., 2017). Therefore, HDPE is chosen to be compounded with natural fibres due to the low melting point which is around 140°C.

In natural fibre composites, weak adhesion between fibres and matrix was found due to being different in nature, in which the natural fibres are hydrophilic while the polymer matrix is hydrophobic. This leads to poor compatibility between them thus, lowering the properties of final composites. Moreover, the stress transfer between polymer matrix is inefficient, resulting in reducing the tensile properties of composites. Thus, several treatments for the natural fibre are necessary to achieve good interface properties. In this study, heat treatment is a chosen to treat the fibres before compounding with matrix. However, heat treatment is expected to modify the polar nature of the natural fibre, consequently, improving the compatibility between fibres and matrix. Due to its cost effectiveness and no production of waste, the heat treatment was preferable among other treatments. Therefore, the objectives focus on the effect of heat treatment of EFB on mechanical and thermal properties on the composites.

1.4 Research Scope

This research covers the topic on raw materials characterisation, the effect of EFB fibre compositions and fibre treatment, moisture absorption of indoor and burial conditioning and the durability of composites towards environmental exposure at certain period of time.

Untreated EFB fibre was analysed for chemical compositions, Fourier transform infrared spectroscopy (FTIR), thermal, physical and tensile test to a get a general overview of the properties of raw materials used.

The optimum temperature for heat treatment of EFB fibre was investigated by varying the temperature of heat treatment. In this work, the temperature used were 140°C, 160°C, 180°C and 200°C for 1-hour treatment in vacuum oven. The effects of different treatment were analyses based on thermal, mechanical and the morphology of the fibre. Based on the results, the optimum heat treatment temperature was chosen to proceed with processing of the composites.

The effects of addition of 10%, 20% and 30% of untreated and treated EFB fibre were investigated on thermal, mechanical and the fractured surface specimens were observed by the field emission scanning electron microscopy (FESEM).

Samples were subjected to indoor and burial conditions to study the degradation and moisture absorption. The percentage of weight gain or loss were calculated. All the properties were discussed relative to the sample in dry condition as reference.

All composites were exposed to environmental exposures, which are indoor and soil burial to investigate the durability of composites after 6 months' period. The effects of conditioning on the thermal and mechanical properties were discussed relative to the sample in dry condition as reference.

1.5 Research Objectives

The objectives of this research are:

- **I.** Preparation of natural fibre reinforced polymer composites comprising of oil palm empty fruit bunch as the reinforced natural fibre and high density polyethylene as the polymer matrix.
- **II.** To investigate the optimum condition for the heat treatment of EFB fibre and the effect of heat treatment on the properties of composites.
- **III.** To evaluate the effects of fibre compositions and environmental exposure towards the properties of composites.
- **IV.** To elucidate the failure mechanisms through fracture surfaces of the composites.

1.6 Thesis outline

This thesis is divided into 5 chapters. Chapter 1 represents the introduction on the background, problem statement, justifications, scope of work, research objectives and the outline.

Chapter 2 explained about the literature review. All the subtopics discussed in this chapter are composites, polymer and its type, fibres, fibre reinforced composites, processing techniques, fibre treatments, fibre treatments, effects of fibre on thermal, mechanical, morphology and the durability of the composites.

Chapter 3 highlights the methodology of the research work. The materials used in this study, experimental works and characterisation techniques are such as thermal and mechanical properties are explained in detail.

Chapter 4 represent results and discussion. This chapter consists of two main topics. The first topic represents the results of properties of EFB fibres, while the second topic represents the characterisation of composite materials. The properties of EFB fibres includes the characterisation in terms of chemical properties, physical properties and also tensile properties of single continuous fibre. Moreover, this topic also discusses about the preliminary studies done on EFB fibre and optimum temperature chosen for the fibre pre-treatment.

The second topic discuss on the characterisation of composite materials in terms of morphology, thermal and mechanical. The effect of addition of EFB fibre, fibre treatment and conditioning (indoor and burial conditioning) on the properties of composite materials are discussed in this topic.

Chapter 5 concludes the thesis with some suggestions for further works.

CHAPTER 2:LITERATURE REVIEW

2.1 Composites

Composites are materials made from two or more constituent materials with significantly different physical or chemical properties that, when combined, produce a material with characteristics different from the individual components. Not all plastics are composites. In fact, most of plastics today are pure plastic, such as toys and soda bottles. When additional strength needed, many types of plastics can be reinforced (usually with reinforcing fibres). This combination of plastic and reinforcement can produce some of the strongest materials for their weight that technology has ever developed and most versatile. Fibre-reinforced polymeric is a material consisting of a polymer (resin) matrix combined with a fibrous reinforcing dispersed phase. They are comprised of short or continuous fibres bound together by an organic polymer matrix. Currently, plenty of research materials are being generated on the potential of cellulose based fibres as reinforcement for plastics. Moreover, natural fibre composites are environmentally superior in specific applications such as a side panel and insulation component for car as well as for transport pallets because of the lower environmental impacts and phase emissions due to the weight reduction and substitution of base polymer by higher volume percentage of natural fibre (Joshia et al., 2004). From the past three decades, researchers reported that plant-based natural fibre reinforced has been widely used in numerous applications especially in automotive industries due to their advantages (Onuaguluchi & Banthia, 2016; Shinoj et al., 2011). For example, coconut fibres bonded with natural rubber latex are being used in seats of the Mercedes Benz A-class model. Moreover, the automotive industry originally started considering the application of natural fibres, for interior part fillers, due to its lightweight characteristic.

2.2 Polymers

A polymer is a large compound consisting of long-chain molecules, each molecule made up of repeating units connected by covalent bonds. Most polymers are based on carbons and considered as organic chemicals. Polymers are composed of three types which are thermosets, thermoplastics and elastomers.

Thermoset are polymers that cross-link together during an irreversible process called curing to form chemical bond and become rigid (Fangueiro, 2011). As a result of their cross-linked structure, thermosets do not soften or flow when heated making thermosets ideal for high-heat applications such as electronics and appliances. Therefore, thermosets are often used in applications such as sealed products due to their resistance to deformation.

Thermoplastic polymers soften when heated and become more fluid as additional heat is applied. The molten polymer would then solidify as its temperature lowers. This process can be repeated numerous times for reprocessing, allowing them to be easily and economically shaped into products. Thermoplastic matrices offer high performance properties compared to thermosetting matrix resins. Thermoplastics are attractive due to their superior toughness and impact resistance, their post-process formability and improved reparability (Van de Velde & Kiekens, 2001). Thermoplastics such as poly (butylene terephthalate) (PBT), polypropylene (N. I. S. Anuar et al., 2018; Arutchelvi et al., 2008) and polyamides (PA) are generally use in the composite materials because of their excellent performance-process ability and profitability ratios (Hassan et al., 2012).

Elastomer is a polymer with both viscous and elastic properties is known as an elastomer. Polymers that exhibit ultimate elastic extensibility when subjected to relatively low mechanical stress also known as rubber. Elastomers have weak intermolecular forces; thus, some elastomers can be stretched by a factor of 10 and yet completely recover to their original shape. Although their properties are quite different from thermosets, they share a similar molecular structure that is different from the thermoplastics.

2.2.1 HDPE

Polyethylene (Polythene) is one of the world's most popular plastics. It is an enormously versatile polymer which is suited to a wide range of applications from heavy-duty damp proof membrane for new buildings to light, flexible bags and films. Polyethylene can de divide into two types which are low density polyethylene (LDPE) and high-density polyethylene (HDPE). One of widely used automotive plastics is high density polyethylene (HDPE) which often replaces heavier materials. It is an extremely flexible polymer which is suited to many applications from good electrical insulation, pipes, sheets and containers (Khanam et al., 2016). Moreover, the properties of plastics such as light, high thermal stability, impact strength, and resistance to abrasion make them suitable for use in automobiles. It also has higher hardness, tensile strength, and heat softening point temperature, as well as better processing properties, chemical and water resistance (Liang & Yang, 2007). Recently, the effects of some fillers, such fibres, clay, resin, organometallic compounds and mica on the properties of filled HDPE composites have been investigated. When considering natural fibres, the matrix preference is limited by the temperature at which natural fibres degrade (Pickering et al., 2016) The polymer used must be low in melting point due to the low degradation of natural fibres which is around 200°C. Thus, in this study, HDPE has been chosen to be compounded with EFB fibre.

2.3 Fibres

In stitching clothes, the threads sometimes split in two parts. The thin of the thread is referred to as fibre. Fibre is a part of hair-include material which are having continuity of filaments or are in a maximum elongated part, like a piece of thread (Mishra et al., 2016). They can be in forms of continuous and discontinuous forms. Some fibres are naturally made while some are synthetically made called as natural and synthetic fibre respectively. Fibres are classified by their chemical origin, divided to two families: natural fibres and manufactured fibres. Manufactured fibres are also referred to as manufactured or synthetic fibres. Figure 2.1 shows the classification of fibres. One of the important factors in favours of these type of fibres is because they are all biodegradable after an effective use (Rozman et al., 2001).

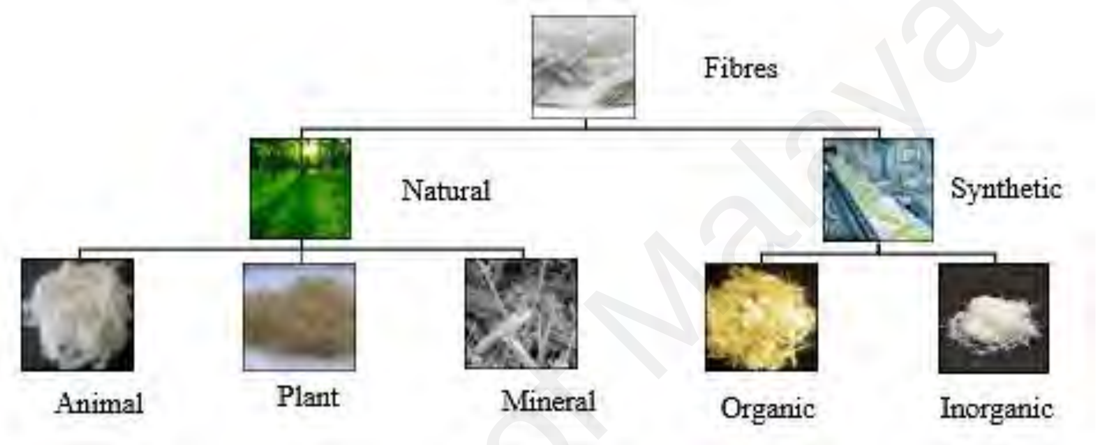


Figure 2.1: Classification of fibres (Kaboorani, 2009).

2.3.1 Synthetic fibres

Synthetic fibres are also called manufactured fibre. These fibres are those whose structural properties and chemical compositions are significantly modified during the formation and are manufactured artificially. The compounds that are used to make these fibres come from raw materials such as petroleum-based chemicals or petrochemicals. These materials are polymerized into a long, linear chemical that bond two adjacent carbon atoms. Different chemical compounds will be used to produce different types of fibres. There are several methods of manufacturing synthetic fibres, but the most common method is the melt spinning and extrusion process. Synthetic fibres can be divided into two groups, which are organic and inorganic. Examples of organic synthetic fibres are such as aramid, polyethylene, nylon, polyamide, polyester and acrylic while inorganic synthetic fibres are glass, carbon, boron and silica carbide. Most of all the synthetic fibres have quite similar properties.

2.3.2 Natural fibres

Natural fibres are hair-like threads obtained directly from plants, animals, and mineral sources. Botanically, a natural fibre is a collection of cells having long length and negligible diameter. They are obtained as continuous filaments or discrete elongated pieces like thread. They can be spun or twisted into yarn such as cloth and can be converted into nonwoven fabrics, such as paper or felt. Traditionally, natural fibre sources are broken down into plant, animal or mineral. All plant fibre contains cellulose as their major structural components whereas animal fibres contain protein and the mineral fibres are from asbestos group. Fibres from plant or vegetable sources are more properly referred to as cellulose-based and the annual production of plant fibres are more than that of animal or mineral fibres. Moreover, plant fibre can suitably be grown in many countries and can be harvested after short periods (Chandramohan & Marimuthu, 2011; Jawaid et al., 2011; Pickering et al., 2016).

Plant fibres

Based on the origin, natural fibres can be classified into bast, leaf, seed, fruit, stalk and other grass fibres based on their origin. Examples of bast fibres/ skin fibres are such as jute, flax, hemp, kenaf, banana fibre and rattan). These fibres usually have higher specific Young's modulus and tensile strength than other type of fibres due to higher cellulose content as well as the cellulose micro fibrils aligned more in the fibre direction. They are used for durable yarn, fabric, packaging and paper. Leaf fibres are that which collected from the leaves are pineapple, sisal, henequen and screw pine. Coir, cotton, kapok and oil palm empty fruit bunch are fibres collected from seeds and fruits. Coir contains a high lignin percentage thus makes the fibre very resistant to biodegradation while cotton has good strength and colour retention properties. Fibres from the stalk of the plant are straws of wheat, rice, barley, bamboo, grass and wood. The most common used plant fibres are cotton, flax and hemp. Hemp fibres are used as a seal within heating and sanitary industries while flax is used in thermal insulation and geotextiles (Chandramohan & Marimuthu, 2011; Dunne et al., 2016; Jawaid et al., 2011; Pickering et al., 2016).

Animal fibres

Fibres from animal sources are more properly known as protein-based fibres. They are harvested from an animal and are taken from either hair, silk, fur or feather. Hair fibres are obtained from hairy animals such as sheep's wool, goat hair; alpaca hair and horsehair while silk fibres are collected form silkworms, sea silk and spider silk. These silk fibres are obtained from dried saliva of bugs or insects during the preparations of cocoons. Avian fibre which fibre taken from the bird's feather. Fur fibre are taken from rabbit, mink, fox or beaver

Mineral fibres

Mineral fibres are naturally occurring fibres or those slightly modified fibre procured from minerals. These types of fibres can be divided into three groups, which are asbestos, ceramic and metal fibres. The only long thin naturally occurring mineral fibres is asbestos. Based on their crystalline structure, asbestos can be divided into two groups, which are serpentine and amphibole. There are six asbestos mineral types. Chrysotile is the only member of serpentine class. It exists in the form of a sheet or a layered structure. The remaining five asbestoses classified under amphibole are amosite, crocidolite, tremolite, anthophyllite and actinolite and these exists in exists in form of chain-like structure. Glass fibres (glass wool and quartz), aluminium oxide, silicon carbide and boron carbide are types of ceramic fibres while aluminium fibres fall under metal fibres (Chandramohan & Marimuthu, 2011). Table 2.1 below shows the mechanical properties of some synthetic and natural fibres.

Table 2.1: Mechanical properties of some synthetic and natural fibres (Anuar et al., 2017; Lilholt & Lawther, 2000; Mallick, 2010; Nair et al., 2016; Pickering et al., 2016).

Properties	Synthetic fibres (Glass fibres)	Natural fibres (Hemp fibres)
Density	High density as compared to polymers	Low density, high specific strength and stiffness
Mechanical properties	Good	Poor
Moisture uptake	Low, high compatibility with matrix	High, low compatibility with matrix
Cost	High	Low
Renewability	Poor recycling, Non-biodegradable	Renewable, biodegradable
Environmental effect	High (fossil fuels)	Low (solar energy)

Nowadays, natural fibres are offered to substitute synthetic fibres due to some advantages such as low cost, can be renewable and less hazards for the environment. Due to the changes in legislation and regulations, industry is forced to look for lighter and less environmentally harmful materials. This has caused a renewed interest in the use of natural fibres as alternatives that are more environmentally friendly in the industrial world. The use of natural fibres also is said to be the best efforts in reducing the environmental pollution. The natural fibre composites used especially in transportation (automobiles, railway coaches, aerospace) due to their light weight and low density make the automobiles more fuel efficient (Dunne et al., 2016; Ilyas et al., 2019). Natural fibres also used in military applications, building and construction industries (ceiling panelling, partition boards), packaging, and consumer products. Natural fibres are producible with low investment at low cost, which makes the material an interesting product for low wage countries. Moreover, it is renewable as the production requires little energy, and carbon dioxide is used while oxygen is given back to the environment (Ilyas et al., 2019). Most important thing is that natural fibres are not harmful to health as it reduced wear of tooling, no skin irritation and do not have health risk when inhaled. In terms of mechanical properties, natural fibres have a higher specific strength and stiffness compared to glass fibre due to its low specific weight (Chandramohan & Marimuthu, 2011; Wambua et al., 2003).

However, a part of their advantages, natural fibres suffer from disadvantages, natural fibres possess three disadvantages incompatibility of natural fibres with thermoplastic, moisture absorption and desorption characteristics and limited thermal stability (Kaboorani, 2009), such as poor compatibility with hydrophobic polymer matrix (poor interfacial adhesion) which leads to fibres to pull-out of the matrix (Dunne et al., 2016; Gupta et al., 2019; Suradi et al., 2011; Wambua et al., 2003) The formation of a weak interface, which results in poor mechanical properties, where the stress transfer at the interface between two different phases is determined by the degree of adhesion (Suradi et al., 2011). In contrast, natural fibres possess high moisture absorption natural due to large amount of cellulose, hemicelluloses, lignin and pectin thus; it tends to be active polar hydrophilic material (Gurunathan et al., 2015; Jawaid & Khalil, 2011; Kaboorani, 2009). When natural fibres are exposed to humid conditions, they will interact with water molecules by hydrogen bonding. This will also affect the interfacial adhesion thus lowering the mechanical properties. Moreover, natural fibres also have limited thermal stability. Due to the restriction, thermoplastics used with natural fibres should be melted below 200°C. As a result, polyethylene (D. U. Shah et al., 2012), polypropylene (Arutchelvi et al., 2008), polyvinyl chloride (PVC), and polystyrene (Yusoff et al., 2009) can use natural fibres as filler (Kaboorani, 2009). Table 2.2 shows the properties of different natural fibres.

Table 2.2: Properties of some natural fibres.

Fibre types	Origin	Density (kg/m^3)	Tensile strength (MPa)	Moisture content (%)	References
Hemp	Stem	1480-1500	277-1100	10	(Mwaikambo, 2006; Shahzad, 2011, 2013)
Jute	Stem	1300-1500	200-540	13.75-17	(Bos, 2004; Mwaikambo, 2006)
Coir	Fruit	1250	106-175	13	(Graupner & Müssig, 2010; Mwaikambo, 2006)
Sisal	Leaf	700-1500	80-855	14	(Bos, 2004; Mwaikambo, 2006)
Flax	Stem	1400-1500	400-1500	9.24-12	(Bos, 2004; Mwaikambo, 2006)
EFB	Fruit	700-1550	50-400	7.79	(Bakar et al., 2005; Kalam et al., 2005; Hanan et al., 2018; Sreekala et al., 2004; Sreekala & Thomas, 2003)

2.3.2.1 EFB fibres

Among the natural fibres available, Oil palm (Elaeis guineensis) is the highest yielding edible oil crop in the world. Crude Palm Oil (CPO) as the main product of oil palm fruit can be used in many different industries (Karina et al., 2008). Since Malaysia is one of the largest producers of palm oil and has over 2.5 million hectares of oil-palm plantations distributed extensively throughout the country (Suradi et al., 2011), abundance of oil palm cellulosic material can be readily obtained from the by-products which provide a new area for research development (Khalid et al., 2008). From oil palm tree, lignocellulosic fibres can be extracted from trunk, frond, fruit mesocarp and empty fruit bunch (EFB) (Shinoj et al., 2011). EFB is the fibrous mass left behind after separating the fruits from sterilised (steam treatment at 294 kPa for 1 h) fresh fruit bunches (FFB). Among the various fibre sources in an oil palm tree, EFB has potential to yield up to 73% fibres (Wirjosentono et al., 2004) and hence it is preferable in terms of reinforcement and cost reduction (Yusoff et al., 2009). Palm oil industries has to dispose about 1.1 ton of EFB per every ton of oil produced (Karina et al., 2008). At present, by-products of oil palm are not efficiently utilized, and the explosive expansion of oil palm plantation has generated enormous amounts of vegetable waste, creating problems in replanting operations and tremendous environmental concerns such as fouling and attraction of pests (Khalil et al., 2008; Law et al., 2007). Figure 2.2 shows the wastes of EFB piled up for disposal in a palm oil mill in India.

Figure 2.2: EFB left as waste in oil palm premise (Shinoj et al., 2011).

Oil palm fibre is firm and strong, similar to coir fibres (Hanan et al., 2018). These EFB consists of high cellulose content which is about 44.2% has the possibility to be an effective reinforcement in polymer. The amount of cellulose in fibres will affect the property and economic production of fibres for various uses. For example, for pulp and paper production, the strength of paper depends on the content of cellulose in the plant used (Khalil et al., 2008). Moreover, EFB also composed approximately 33.5% hemicellulose and 20.4% of the lignin (Astimar et al., 2002). The moisture content is about 10–15%. Table 2.3 shows the chemical composition of OPEFB fibres reported by different researchers within Malaysia, India and Colombia. This shows a variation in chemistry of the OPEFB fibres within the same region as well as outside the regional borders. These fibres consist of pores on the surface with average diameter of 0.07μ.m, which is useful for better mechanical interlocking with matrix resin in composite fabrication. All the nature fibres consist of basic chemical building block of cellulose, hemicellulose, and lignin with varying proportions. Other components such as pectin and waxes are also present in minor quantities in the fibrous materials.

Table 2.3: Chemical composition of OPEFB fibres reported by different researchers.

Location	Hemicellulose (%)	Cellulose (%)	Lignin (%)	References
Malaysia	34	44	20	(Astimar et al., 2002)
Malaysia	-	65	19	(Mahjoub et al., 2013; Piarpuzan et al., 2011)
Colombia	13	14	8	(Piarpuzan et al., 2011)
Malaysia	22	48	25	(Hassan et al., 2010)
Malaysia	84	50	21	(Khalil et al., 2010)
India	-	65	19	(Sreekala et al., 2001)

Cellulose

Cellulose is a very important polysaccharide because it is the most abundant organic compound on earth and closely associated with hemicellulose branches and lignin. Cellulose is a major component of tough cell walls that surround plant cells, and is what makes plant stems, leaves, and branches so strong. It is an organic compound with the formula $(C_6H_{10}O_5)_n$ and have positive influence on the mechanical strength of the fibre (N. I. S. Anuar et al., 2018). Typically, cellulose contributes the largest weight of the EFB which is about 24-65% (Chang, 2014). It is a polysaccharide consisting of a linear chain of several hundred to many thousands of β (1→4)- glycosidic bonds with each repeating unit containing three hydroxyl groups as shown in Figure 2.3. Due to the structure, cellulose possess the characteristics such as hydrophilicity, chirality, and broad chemical variability initiated by the reactivity oh hydroxyl groups. Moreover, the structural strength of EFB fibres is provided by the hydrogen bonds between different layers of cellulose chains, coupled with the crosslinking of lignin with both cellulose and hemicellulose (Dashtban et al., 2010).

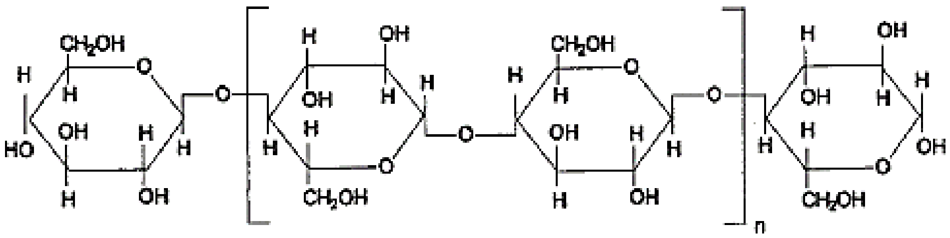


Figure 2.3: Structure of cellulose (Mohanty et al., 2002).

Hemicellulose

Hemicellulose is the second most abundant polymer. Like cellulose, hemicellulose function as supporting material in the cell wall. It can be easily hydrolysed by diluted acid, alkali or enzymes under mild conditions (Lee et al., 2014). The structure of hemicellulose in plant shown in Figure 2.4.

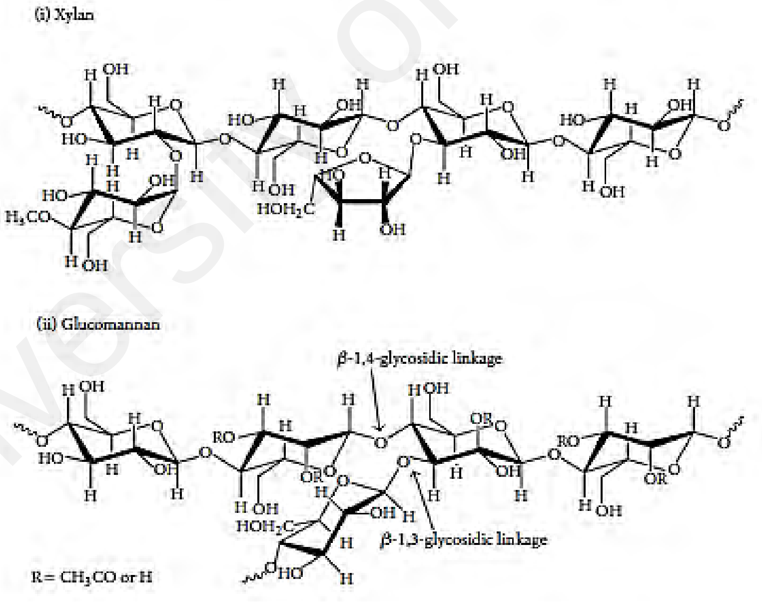


Figure 2.4: Chemical structure of hemicellulose compounds (Lee et al., 2014).

Lignin

Lignin is a three-dimensional polymer of phenylpropanoid units. It functions as the cellular glue, which provides compressive strength to the plant tissue and the individual fibres, resulting stiffness to the cell wall. According to the physical characteristics, lignin is hard, which increases the hardness of the cell wall. To prevent plant cell destruction, lignin acts as act as a protection layer for plant cell permeability and resistance against microbial attacks (Lee et al., 2014). Lignin is mostly observed as an integral part of the plant cell wall, embedded in a polymer matrix of cellulose and hemicellulose. Thus, lignin is the most non-biodegradable component of the plant cell wall.

Ash content

Ash content consists of calcium and magnesium, but certain plants contain high levels of silica that cause the knife to become blunt during the cutting process. This inorganic element will also create problems in a chemical sense, causing tearing during the pulping process, especially in the chemical recovery stage.

2.3.2.2 Modification of natural fibres

The incompatibility of natural fibres with the polymer matrix, the tendency to form aggregates during processing and poor resistance to moisture are some of the disadvantages of natural fibres (Bakar et al., 2005). This is due to the difference in nature between lignocellulosic filler and polymer matrix. Lignocellulosic are hydrophilic (contributed by hydroxyl groups in cellulose, lignin and hemicellulose), whereas, thermoplastics such as polypropylene (Arutchelvi et al.) and polyethylene (Shah et al., 2012) are hydrophobic (Rozman et al., 2003). To optimise effective interfacial bonding between fibre and matrix, the fibre surface needs to be modified with different treatments, addition of reactive additives and coupling agents.

The fibre treatment can be divided into chemical and physical methods (Gupta et al., 2019). In the chemical method, more reactive groups will be exposed on the fibre surface and consequently facilitate efficient coupling with the matrix. The ordinary chemical treatments are alkaline treatment, H_2SO_4 treatment and acetylated treatment (Ahad et al., 2009). Due to the reasons, composites with great mechanical and thermal properties will be produced (Dash et al., 2000).

Physical methods only involve stretching, calendaring and thermos treatments on the fibre surfaces, however no changes in the structural composition has been made (Kabir et al., 2011). The modification on the fibre surface will improve fibre-matrix interfacial adhesion thus enhance the composite properties (Abdelmouleh et al., 2007). Recently, heat treatment has been widely used to modify the fibre because it involves a simple method and is cheaper compared to chemical treatment. With this treatment, the fibre surface will be modified to make it have a good adhesion with thermoplastics and leads to better thermal stability (Kaboorani, 2009).

Coupling agents or compatibiliser in natural fibre reinforced plastic composites play an important role in improving the compatibility and adhesion between the polar natural fibre and the non-polar polymer matrices by introducing a chemical bond between the fibre and the matrix. So far, more than forty coupling agents have been used in the industrial production and research area. One of the examples of coupling agent that has been widely used is maleic anhydride polypropylene (MAPP) (Lu et al., 2007).

In addition, besides the treatment modification on the natural fibres, used of compatibiliser can promote interfacial adhesion polymer and fibres which having two different polarities. Based on study by Galav et al. (Galav et al., 2019), the tensile strength, tensile modulus, impact strength and flexural strength of observed to be enhanced by using compatibiliser due to better adhesion of the fibres and polymer matrix. This was also proved in SEM images where the fibres pull-out observed when use compatibiliser is less than the untreated composites because of the strong interfacial adhesion of polymer and matrix.

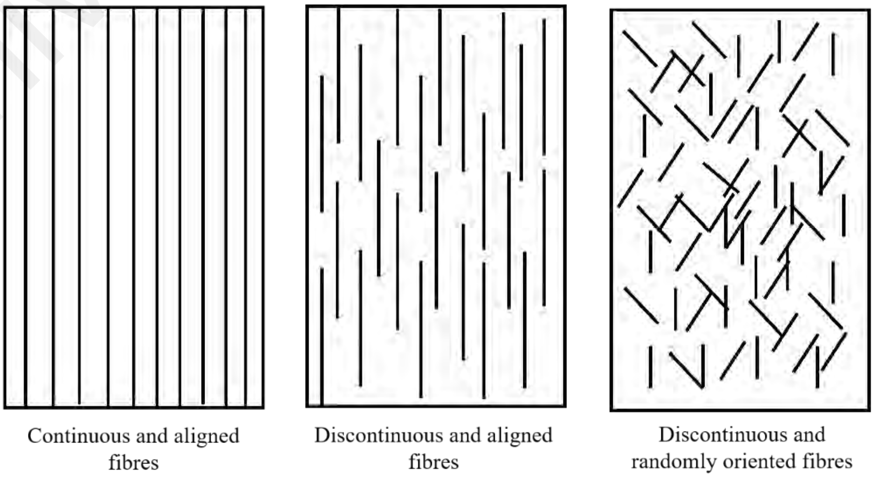
2.4 Fibre reinforced polymer composites

Fibre reinforced polymer composites (FRPC) are composite material composed of a polymer matrix that is reinforced with fibres. Natural and synthetic fibres can be used as reinforcement. The synthetic fibres are usually glass, carbon, or aramid, while natural fibres such as hemp, jute, kenaf and coir. The polymer that is widely used as matrix are polypropylene, high density polyethylene and epoxy.

FRPC are usually used in the aerospace, automotive, marine, and construction industries. The fibre reinforced polymer composites (FRPs) are increasingly being considered as a replacement for infrastructure components or systems mostly in car and aircrafts parts that are constructed of traditional civil engineering materials, namely concrete and steel.

FRPC can be formed by embedding fibres to the polymer matrix through several processes to form composites in several types: sheets, continuous mats and moulded specimen. However, most of the FRPC are created using moulds. The process begins with mixing fibres and matrix using extrusion technique, then moulded into parts needed. Figure 2.5 shows the illustration on dispersion of fibres in the composites.

(a)



(b)

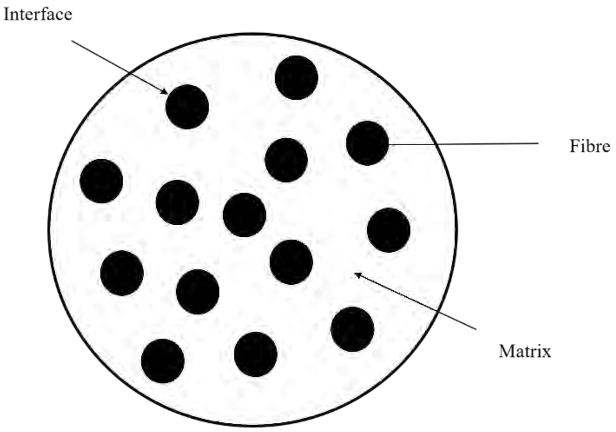


Figure 2.5: Illustration of fibre reinforced polymer composites: (a) Alignment of fibres in composite, (b) cross section of composite.

2.4.1 Processing

Good composites performances depend on the choice of matrix, fibres, bonding between fibre and matrix as well as good and suitable processing techniques. It is also known as conversion of composite to finish article. There is various technique of processing composites such as extrusion, injection moulding and compression moulding.

2.4.1.1 Compounding/Extrusion

Compounding is a process of melt blending plastics with other additives. This process changes the physical and thermal characteristics of the plastic. The final product is called a compound or composite. In this process, one or more screws rotating inside a heated barrel to generate pressure and melt the polymeric materials. Screws inside barrel has three sections which are feed, compression and metering as shown in the Figure 2.6 below Channel is deep in feed section to convey and compact the solids. Compression section progressively decreasing channel depth where melting occurs as heat supplied from the barrel and mechanical work from the screw rotation. Metering section with shallow/ constant depth to control output, generate pressure and mix the melt. The extrusion process begins by feeding raw materials (pellets/powder) into the feed hopper. The polymer granules are channel by rotating screw through the heated barrel. As the plastic passes through the screw channel, it is gradually heated and melted by the heat conducted from the heater clipped around the cylinder barrel together with the shear heat generated by the rotating screw. Along the barrel, the melt polymer is mixed with additives. Finally, it is pushed out of the die with properly regulated temperature and pressure to form the desired products. Then a puller through a cooling system, usually cooling water, pulls it. The final product is then cut into pellets using pelletizer (Jiang et al., 2012).

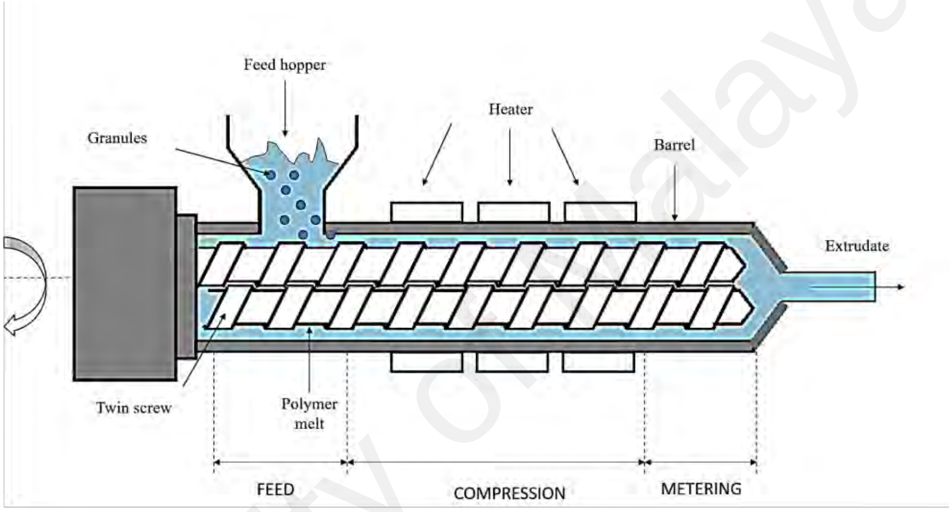


Figure 2.6: Illustration of the twin-screw extruder.

Studies done by Gang Sui et al. (Sui et al., 2007) used twin-screw extruder manufactured by Brabender Technologies Inc, Germany to compound sunflower seed hull sanding dust (SHSD) with polypropylene (Arutchelvi et al.) and clay. The temperature profile varies from hopper to die was 182°C, 188°C, 193°C, 199°C, 204°C, 210°C, 210°C, 210°C and the screw speed was fixed at 75 rpm. It was found that no obvious agglomerations of the fillers were found in the resulting composites indicating that mixing by the twin-screw extruder, which is an industry friendly method, is practical for dispersing the SHSD fillers uniformly in the PP matrix. By adding the SHSD fillers to PP, the mechanical and thermal properties of the composites were improved.

G. Gamon et al. (Gamon et al., 2013) used twin screw extruder for compounding poly (lactic acid) (Placet et al.) with Miscanthus fibres (MIS) and bamboo fibres, where the PLA was feed in the hopper while the fibres were introduced through a side feeder (Placet et al.). In this study, four different screw speed were tested (100, 150, 225 and 300 rpm) and temperature was set at 190°C in the melting zone while 165°C at the kneading zone. They found out that increasing the screw speed, will maintain the fibre bundle length, thus reduced blend viscosity in the polymer melts. Moreover, the composites have better thermal stability when compounded with lower screw speed due to less mechanical energy was spent. Optimum screw speed was found to be at 150 rpm screw for energy saving as 100 rpm was insufficient to have homogenous properties.

Different with the studies done by Andrzej K. Bledzki et al. (Bledzki & Faruk, 2004). Hard wood fibre with polypropylene (Arutchelvi et al., 2008) was mixed by high speed mixer (both with and without coupling agent). The temperature ranges from hopper to die was set at 160°-190°C and the screw speed was fixed at 55 rpm during the process. Based on the SEM image, it was observed the fibre pull out, de-bonding and fibrillation of the wood fibre-PP composites more visible in the extrusion process compared to the injection moulding. This is suggested due to the moisture present in the wood fibre which is released during the heating. Thus, the moisture retains in the melt until the composites being extrudate out of the die.

In addition, F.G. Torres and R.M. Diaz (Torres & Diaz, 2004) studies the morphological characterisation of natural fibre reinforced thermoplastics (NFRTP) processed by extrusion, compression and rotational moulding. The reinforcing fibres used were jute, sisal, cabuya, coconut, pecan and rice shell flour compounded with HDPE and PP as matrix. In extrusion, the reinforcing fibres were varied from 1 to 10% by volume. The composites extruded by using single screw extruder with temperature profile range from 160°C-230°C throughout the process. The fibre dispersion in composites were evaluated using stereomicroscopy. They found that several voids could be observed after the extrusion process, indications of the high incompatibility between the coconut fibres and PP compared to other natural fibres which are jute and sisal.

2.4.1.2 Moulding

Moulding of plastics can be defined as the process of shaping plastic using a rigid with huge design flexibility for both simple and highly complex designs. There are variety of moulding processes such as blow moulding, compression moulding and injection moulding.

Blow moulding is used in the production of large quantities of hollow plastic objects. The process follows the basic steps found in glass blowing. Blow moulding creates very uniform, thin walled containers. Plastic is melted and extruded into a hollow tube (a parison). This parison is then captured by closing it into a cooled metal mould. Air is then blown into the parison, inflating it into the shape of the hollow bottle, container, or part. In addition, (this method is very economical) it can do so very economically. Bottles, tubes and container are the examples from blow moulding process.

Blow moulding was used by A. Gracia-Rejon et al. (Rejon et al., 2002) to prepare long glass fibre (LGF) / HDPE composites. The LGF/HDPE composites were first compounded with extruder before proceeding to moulding. In this research, they studied the flow behaviour of LGF/HDPE composites during the extrusion and blowing stages of an intermittent blow moulding process. From this study, parison swell (diameter and thickness) decreased with increasing of fibre content. During the flow inside the die, the fibres are well oriented in the axial direction causing a reduction in the hoop stress at the die exit. This phenomenon leads to the formation of a tubular envelope that limits the radial expansion (diameter swell) of the parison while keeping its shape constant. No significant changes in fibre length distribution were observed when samples for different regions of the bottle were analysed separately. In conclusion, it appears that the blowing step does not change the orientation of the fibres.

Compression moulding is commonly used in manufacturing thermoset plastic parts. The raw materials for compression moulding are usually in the form of granules or pellets. In this type of moulding, the raw material is introduced into an open, heated mould cavity. The mould is closed, and certain amount of pressure is applied to force the material to contact all areas of the mould. Throughout the process heat and pressure are maintained until the polymer has cured. After a set of time, the pressure is released, and mould is open so the composite can easily be removed. Any resin flash around the edges is also removed. Compression moulding is a high-volume, high-pressure plastic moulding method that is suitable for moulding complex, high-strength objects. Because of short cycle time and high production rate, many organizations in the automotive industry have chosen compression moulding to produce parts.

Injection moulding process is commonly used in mass-production or prototyping of a product. Injection moulding machines were made in the 1930's. The polymer is introduced to the machine, soften and conveyed with screw along the barrel. The molten plastic that has been melted from pellet form in the barrel of the moulding machine is injected under pressure into the mould. The pressure is applied to ensure all cavities are filled. The mould is cooled, and plastic parts are then allowed to solidify in the mould. Injection mould cooling consumes about 85% of the cycle time for the entire process. After cooling, the mould separates and the newly formed plastic part is ejected from its mould. The part is cleaned of any extra plastic from the mould. The process can be repeated. Figure 2.7 below shows the illustration of injection moulding machine. There are advantages and disadvantages associated with plastic injection moulding. This process is more suitable to produce products in large amount and lower costs for labour as the bulk of the work is done by machine. Injection moulding also has the added benefit of lower scrap costs because the mould is so precisely made. The drawbacks are that the operating costs will be increased due to the equipment needed is expensive.

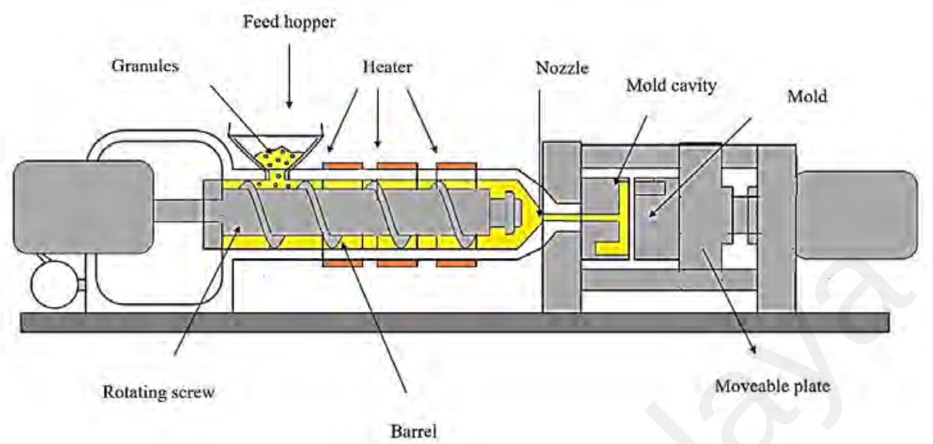


Figure 2.7: Illustration process of injection moulding machine.

Fibre length and orientation in injection moulded natural fibre/starch acetate composites studies was conducted by Heidi Peltola et al. (Peltola et al., 2011). In this research, flax and hemp fibre were used as the filler. The composites extruded using twin screw extruder were injection moulded with Engel injection moulding machine (ES 200/50 HL, Engel Austria GmbH, Schwartzberg, Austria) to tensile test specimen according to ISO 3167. It was found that the fibre length decreased after full process of the preparation of the composites. The full processing includes compounding, pelletising and injection moulding of the composite with 20 wt.% of hemp and 40 wt.% of flax fibres. The average of the unprocessed fibres is 11.7 mm while the fully processed fibres is 0.151mm compared. In conclusion, the reinforcement efficiency of hemp fibres was improved by the processing, but not with flax fibres.

Moreover, Amir Etaati et al. (Etaati et al., 2015) using injection moulding machine to prepare composites of 40 wt.% hemp fibre (untreated and treated), 5wt% maleic anhydride polypropylene (MAPP), and polypropylene. The tensile, flexural and impact properties were measured in this study. Based on the study, the tensile strength was reduced as the fibre length increased. It was suggested that using the longer fibres in compounding and injection moulding machines will not improve the tensile properties of the composites, instead it can degrade the properties, apparently due to the formation of fibre lump and thus making the dispersion of fibre heterogeneous. The same trend was observed with tensile modulus, flexural strength and flexural modulus. In addition, in impact tests, the peak load was observed to reduce as the fibre length increased. As mention before, the agglomeration of fibres is more likely to form with longer fibres. So, the crack can be initiated from fibre/matrix interface in the agglomeration sites. As a result, poor dispersion of fibres reduces composite impact resistance during compression, shear, and tensile by reduction of interface bonding between the fibre and matrix in agglomeration sites. In conclusion, increasing the fibre length reduced the mechanical properties of the composites.

On the other hand, B. Nyström et al. (Nyström et al., 2007) studied the microstructure and strength of injection moulded natural fibre composites. Wood powder (WP), chemitherm mechanical pulp and flax fibre were used as reinforcement with polypropylene and maleic anhydride polypropylene (MAPP) was used as the compatibiliser. The composites were prepared by extrusion and injection moulded to dog-bone shaped for characterisation. In order to study presence of agglomerates of fibres in the composites, 5 g pieces were cut out of injection moulded specimens after mechanical tests and heat pressed into thick sheets of 300 μm. It was found that the dispersion of WP particles is not good as it remains as bundles in the compound, however the compound with MAPP shows better dispersion of fibres in composites. Moreover, the fibre length decreases after processing using extrusion and injection moulding. In conclusion, processing has a large impact on composite strength. Properties of fibre length, fibre dispersion and processing parameters may influence the final orientation of fibres in the composite, which should already be taken into account during design of injection moulding instruments.

2.4.2 Properties of composite

2.4.2.1 Thermal properties

Thermal properties explain the reaction of a material to the application of heat. The major components of thermal properties are heat capacity, thermal expansion, heat stress and thermal conductivity. There are several techniques to measure the thermal properties of materials such as differential scanning calorimetry, thermogravimetric analysis and etc.

Differential scanning calorimetry

The differential scanning calorimetry (DSC) is one of the versatile thermal analysis techniques that measures the difference in the heat flow of samples based on the defined temperature program (various combinations of thermal scans heating/cooling, and isothermal cycles), a pressure (stable) and an atmosphere (inert or reactive) DSC is also known as a physical characterisation method used to study thermal behaviour of neat polymers, copolymers, polymer blends and composites.

Based on mechanism of operation DSC can be classified into two types, which are power compensated DSC and heat flux DSC. For power compensated DSC, sample and reference temperatures are kept equal by heating with separate heaters while external heat is increased or decreased linearly. In heat flux DSC, the sample and the reference are heated from the same source and the temperature difference is measured as the sample temperature is increased or decreased (Haines, 2012).

DSC have been widely used for identification of characteristics and performances of the materials. DSC measure the properties such as glass transition temperature, melting point, freezing point, boiling point, crystallisation temperature and time, percentage of crystallinity, specific heat capacity and oxidative stability.

For example, DSC was used to describe compost organic matter quality regarding input materials and humification. In this study, degradation of biogenic waste materials during composting leads to decreasing intensities of heat flow peaks. Humification is indicated by the increase of the second exothermic peak if heat flows are referring to organic matter. However, for humic acid determination time-consuming analyses are needed. Thus, thermal features provide more comprehensive information on compost organic matter quality (Smidt & Tintner, 2007).

DSC analysis was used by Mohammad Mazedul Kabir et al. (Kabir et al., 2013) to study the thermal properties of jute reinforced composites. Jute fabric reinforced polyester composite was prepared by hand lay-up process. The samples were kept at room temperature overnight and post cured at 80°C for 4 hours. The sample was released from the mould and kept in an oven at 80°C for 4 hours to post-cure. DSC analysis on treated fibres was performed by using a thermal analyser (DSC Instrument, Model No. Q100). The samples were heated at 10°C to 450°C at the rate of 10°C/min under nitrogen to remove volatiles from the samples. It was reported that alkali treatment removed hemicellulose, lignin and cellulose from the fibre surface, resulting to more thermally stable, improved hydrophobic nature and facilitate better adhesion possibilities with the matrix.

Thermal properties of high density polyethylene (HDPE) with natural fibres was studied by J.R. Araujo et al. (Araujo et al., 2008) using cuarau fibres as the reinforcement, maleic anhydride grafted polyethylene (PE-g-MA) and ethylene-vinyl acetate (Arrigo et al.) as the coupling agent. The DSC was conducted using TA instruments with temperature range of -50°C to 200°C under argon glow of 50 mL/ min at rate of 10°C/min. The composite made with EVA as a coupling agent shows higher ΔH_m than expected than the composite made with PE-g- MA. All composites show higher crystallinity degrees than expected, with is highest for the composite without coupling agent, while the composites with PE-g- MA show lower crystallinity due to increase of crystallinity provided by the fibres, that are acting as a nucleating agent, provided by the strong interaction between the curaua fibre and the matrix in the presence of PE-g-MA as coupling agent. In conclusion, the increase of the crystallinity for the composites occurred due to the trans crystallinity effect. The presence of the coupling agents decreases this effect due to its reactions with the OH groups at the fibre surface.

Thermogravimetric analysis

Thermogravimetric analysis (TGA) is the second most used thermal technique after DSC. It measures weight change of materials as function of temperature or time in atmosphere of inert or reactive gases such as nitrogen, helium or other gases. When the temperature increases, the materials degrade. As it degrades, the weight of the sample is reducing on the balance. The main use of TGA is to measure material thermal stability and their composition. Besides, inorganic materials, metals, polymers and plastics, ceramics, glasses, and composite materials can be analysed. Moreover, TGA also can be used to determine water content or the residual solvents in a material.

TGA used by Mehdi Tajvidi et al. (Tajvidi & Ebrahimi, 2003) to study the thermal degradation behaviour of natural fibre polypropylene composites. They reported that among different natural fibres, rice hulls proved to be the least thermally stable ones as seen in their lower thermal degradation. Moreover, the use of compatibiliser induced a slight reduction in thermal stability, which was attributed to the lower thermal stability of MAPP.

Aukasz Klapiszewski et al. (Klapiszewski et al., 2016) studies influence of processing conditions on the thermal stability and mechanical properties of PP/silica-lignin composites. The TGA was performed using a Jupiter STA449 F3 (Netzsch, Germany) at heat rate of 10°C/min from 30 to 1000°C in a nitrogen atmosphere, at a flowrate of 40 cm^3/min. the results revealed that as lignin content in hybrid filler increased, the number or larger particles increases, thus the thermal stability deceased. Moreover, the composites with higher screw speed during processing exhibit lower thermal stability.

Ain U. Md. Shah et al. (Shah et al., 2017) studies the thermal properties of bamboo fibre composites. Two types of composites were prepared which are bamboo fibre filled epoxy composites (EP-BFC), and bamboo powder filled glass/ epoxy composites (EP/G-BFC). The TGA was conducted at heating rate of 0°C/min from 30 to 900°C. the results confirmed that as the BF increased in both composites, the thermal stability decreased. However, the EP/G-BFC exhibit higher thermal stability, due to the existence of glass fibre.

2.4.2.2 Mechanical Properties

Tensile properties

Tensile test is the most frequently used to indicate how the material will react to forces being applied in tension mode. The specimen used is in dumb-bell shaped to prevent any fracture deformation of the material during testing. In this test, the sample is pull between two grips, and the force required to produce the deformation is measure using a load cell using universal testing machine (UTM). Accurate measurement need extensometer whether in contact or non-contact so the measurement of each sample. A variety of tests can be performed on each sample tested. For example, tensile strength, tensile strain, tensile modulus, elasticity, hardness and fatigue.

Properties that are directly measured via a tensile test are ultimate tensile strength, tensile strain and tensile modulus. The stress-strain behaviour represents the response of a material to loading. A curve will result showing how it reacted to the forces being applied. The point of failure is of much interest and is typically called its "Ultimate Strength" (UTS) on the chart as shown in Figure 2.8 below:

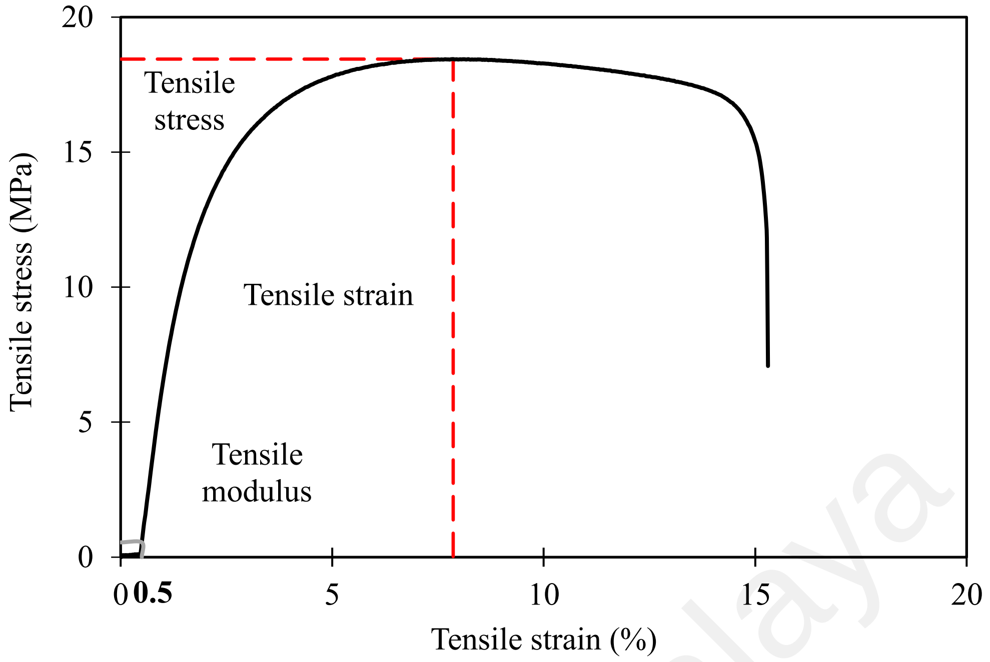


Figure 2.8: Example of stress-strain curve.

UTS is the maximum load the specimen can sustains during the test. The UTS may or may not equate to the strength at break. Due to the viscoelastic effect, the stress-strain relationship of a plastic composites is depending on the type of materials being tested such as brittle, ductile or a substance that exhibits both properties. Moreover, sometimes a material may be ductile when tested in a lab, which has high humidity, but when is placed in service and exposed to extreme cold and dry temperatures, it may transition to brittle behaviour. However, stiffer response may be achieved when materials is tested at higher cross-speed.

The modulus of elasticity is a measure of the stiffness of the material, but it only applies in the linear region of the curve, at early stage of tests. If a specimen is loaded within this linear region, the material will return to its exact same condition if the load is removed. Tensile modulus can be calculated from tensile test stress/strain graphs–derived from load/extension graphs. The slope of the graph is used to calculate tensile modulus, E when the material is obeying Hooke’s law. Unlike metals, plastics do not have a linear relationship between stress and strain. Hooke's Law no longer applies if some permanent deformation occurs in the specimen. From this point on in the tensile test, the material reacts plastically to any further increase in load or stress. It will not return to its original, unstressed condition if the load were removed. The tangent modulus is useful in describing the behaviour of materials that have been stressed beyond the elastic region. The tangent modulus quantifies the "softening" or "hardening" of material that generally occurs when it begins to yield. There is reason for each method and is chosen based on the application of the data. In order to ensure compatibility, standards are used to define criteria for modulus measurement.

The stretch or elongation the specimen undergoes during tensile testing can also be determined easily. This can be expressed as an absolute measurement in the change in length or as a relative measurement called "tensile strain". It is the ratio of the change in length to the original length, engineering strain formula. During this process, the elongation of the gauge section is recorded against the applied force. The data is manipulated so that it is not specific to the geometry of the test sample. The elongation measurement is calculate using the following equation:

$$\varepsilon = \frac{\Delta L}{L_0} = \frac{L-L_0}{L} \tag{2.2}$$

Where ΔL is the change in gauge length, L_0 is the initial gauge length, and L is the final length.

P.J. Herrera-Franco et. al (Herrera-Franco & Valadez-Gonzalez, 2004) studied mechanical behaviour high density polyethylene (HDPE) reinforced with continuous henequen fibres (Agave fourcroydes). In this study, the fibres were treated with NaOH solution for an hour at 25°C and treated with an amount of Silane coupling agent. The composites were compression moulded at 180°C at a pressure of 1 ton using a Carver laboratory press. Based on this study, they found that there is 10% increase in tensile strength of the treated composites compared to untreated. For treated composites, the improvement in the fibre-matrix interactions plays a more important role. The combined effect of the Silane treatment of the henequen fibre surface, (chemical interactions) and the pre-impregnation process (enhancing the mechanical interlocking) are considered key factors for the composite tensile transverse strength behaviour. This were proven by scanning electron microscope, where there are traces of matrix still coating the fibre. This is an indication that the adhesion between fibre and matrix has improved.

N I S Anuar et. al. (Anuar et al., 2017) studied the effect of kenaf and EFB fibre loading on polypropylene nonwoven composite at different mixture ratio. This study was conducted to identify the optimum fibre loading of nonwoven polypropylene composite and their effect on the mechanical strength. The study was designed at 40%, 50%, 60% and 70% of fibre content in nonwoven mat and composite. The researchers found that the highest tensile strength for nonwoven Kenaf/PP (KPNC) and EFB/PP nonwoven (EPNC) were 31.6 MPa and 20.1MPa at 50% and 60% fibre content respectively. Fibre tensile strength of Kenaf showed almost 3 times higher than EFB and this may result to higher tensile strength of KPNC compared to EPNC. Both KPNC and EPNC showed increment in tensile strength from 40% to 50% fibre loading. The tensile strength of KPNC dropped at 60% while EPNC dropped at 70% fibre content. This may be due to at high fibre loading, the amount of PP was not enough to wet and bond with fibres, leading to bad interfacial adhesion between fibre and matrix.

Flexural properties

Flexural analysis is the study of a materials resistance towards bending. Flexural or compressive strength is the mechanical measure of maximum load bearing capability of a material without undergoing any permanent deformation. It is important to study flexural in our daily life as it helps in designing elements like beams, cantilever and shafts. In addition, it will also provide parameter for development of stronger constructional materials and helps to judge quality of structures being used for construction. Moreover, it is important to know that, environmental conditions as if temperature and moisture affect the flexural strength of a material to a great extent. As example, lower temperatures and dry air tends to extract moisture that binds the particles of the object, and hence, make it more brittle leads to lower value of strength. However, higher temperatures and a humid climate helps the object retain moisture and thus, increases its flexural strength.

Flexural modulus is defined as the ratio of stress to strain in flexural deformation. Both three-point and four-point loading configurations are used. Three-point loading consists of a support point near each end of the beam and one load point at the mid-span. For four-point loading, there are two load points at equal distances from the support points. This distance is typically one-fourth of the span length (thus, the term quarter-point four-point loading), but a distance of one-third of the span length (third-point four-point loading) is sometimes used.

At the edge of the object on the inside of the bend (concave), the stress will be at its maximum compressive stress value. Whereas the outside of the band (convex), the stress will be at its maximum tensile value. Most materials fail under tensile stress before they fail under compressive stress, so the maximum tensile stress value that can be sustained before the beam or rod fails if it’s flexural strength.

Davindrabrabu Mathivanan et al. (Davindrabrabu et al., 2015) studied about the effect of fibre loading on the flexural properties of natural fibre reinforced polymer composites and they used pineapple leaf fibres (PALF) as reinforcement in plastics. They found that the result showed decrease in flexural modulus at higher weight fractions (20%) of fibre loading. The authors suggested at higher fibre content; a weak interface was created due to the increase in fibre-to-fibre interactions. Moreover, natural fibres are irregular in shaped, thus the ability to support stress transfer is poor, resulting in weaker composites.

Different with Shivnand H. et al. (Shivnand et al., 2010), from evaluation of tensile and flexural properties of hemp and polypropylene based natural fibre composites, the flexural strength of all the composites increased with increasing the hemp fibre/filler content, similar to the tensile strength. In addition, it was also found that addition of coupling agents like MAPP resulted in increase in the composite tensile and flexural strengths slightly, due to the improvements in the fibre-matrix interfacial adhesion.

Impact properties

Impact strength or impact resistance is the resistance of a material to fracture under a sudden impact, or shock force. It is a measure of the toughness of the material (Ciullo, 1996). The impact properties of a material represent a measure of the amount of energy that a material can absorb before fracturing under a high rate of deformation. Impact strength is used to determine whether a material will act in a brittle or ductile manner when subjected to sudden applied loads. In other way, it gives an idea of a material's "toughness". Usually it is used to test the toughness of metals, but similar tests are used for polymers, ceramics and composites. There are several factors that affect the impact strength of a material include its volume, modulus of elasticity, yield strength, and distribution of forces through the material section. As an example, brittle materials have low toughness as a result of the small amount of plastic deformation that they can endure.

Metal industry sectors include oil and gas, aerospace, power generation, automotive, and nuclear are the common industry use this method to measure the impact performances. For example, in forging industry, the impact test can be used to determine the malleability and ductility of a material that is being forged while in the rubber industry, the test can be used for determining the shock absorbing ability of a type of rubber so that its proper application could be decided. In the case of an aircraft, impact can take place during take-off and landing the aircraft may be struck by debris that is present on the runway, and as well as other causes. Moreover, in plastic or automotive industry, the test is used for analysing the breaking strength of a sample when it is subjected to a high impact from a pendulum. This helps in ensuring that the material is best for an application where it is subjected to such impacts. Standard impact test methods are available for metals and unreinforced polymer are (ASTM E23) and (ASTM D256) respectively. In addition, this standard is also being used for fibre reinforced composites.

Impact testing most commonly consists of Izod Impact and Charpy Impact (Ciullo, 1996). The Izod test is the more popular method for plastic materials whereas Charpy is very common for metals. For Izod test, a notched impact specimen is clamped and broken with a swinging weight or a “tup” attached to a swinging pendulum. The specimen breaks at its notched cross-section upon impact, and the upward swing of the pendulum is used to determine the amount of energy absorbed (notch toughness) in the process. Energy absorption is directly related to the brittleness of the material. The energy absorbed by the sample is calculated from the height the arm swings to after hitting the sample. Different with Charpy impact, the specimen is clamp in a flexural mode; specific weight load is drop from a specific height to put an impact on a notched sample. With the help of the Charpy Impact Test, one can easily analyses the strength of material. The quantitative results of the test give the understanding of the amount of force that a material can absorb when it reaches the point of fracture or failure. The qualitative results of the test procedure can provide information about the ductility of the materials being test. Figure 2.9 below shows different of the specimen for both Charpy and Izod tests.

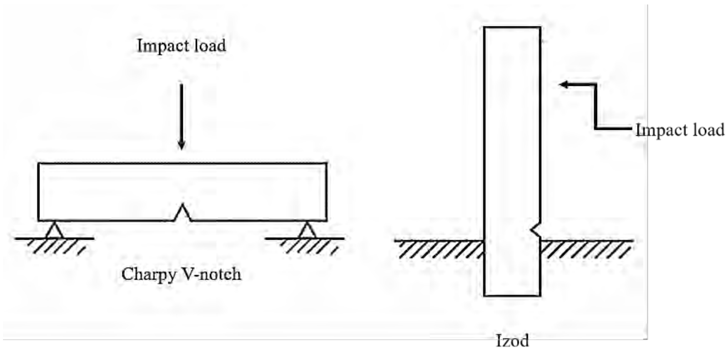


Figure 2.9: Differences between Charpy and Izod impact test set up.

From Hardik Modi et al. (Modi et al., 2017), studies on natural fibre composites were done by using cotton and bamboo fibres as reinforcement. The fibres act as main load carrying member in composites. The samples were prepared for longitudinal placed fibre and transverse placed fibre using Izod impact test method. They found that the impact strength of the composite gradually increases with increase in the weight fraction of the fibre. In addition, the impact strength of the composite material increases for both longitudinal placed fibre reinforced composites and transverse placed fibre reinforced composites. The transverse placed fibre reinforced composite has more impact strength compared to longitudinal placed fibre reinforced composites.

In addition, studies done on reinforced PLA composites by Nuthong et al., the impact strength of natural fibre reinforced PLA composites decreased with the increased of natural fibre content (Nuthong et al., 2013). This indicated that the addition of natural fibre was not effective to improve the brittleness and strength of PLA. This may be due to the weak interfacial adhesion strength between matrix and fibre thus affected the impact strength of composites. Impact energy dissipated by debonding, fibre and/or matrix fracture and causing fibre pull out. However, an increment of impact strength shows in the epoxy treated bamboo and coconut fibre reinforced PLA composites when compared with untreated composites proves that good adhesion between fibre and matrix after treatment.

Studies on impact properties also done by Mohamad Jawaid et al. (Jawaid et al., 2011) on jute and EFB fibre with epoxy resin. The composites were tested at ambient condition according to ASTM D 256 and average values was taken. It was found that the composites compounded with coupling agents of EFB/Jute/EFB exhibit higher impact properties compared to Jute/EFB/Jute and suggested may be due to the high interfacial bonding between fibre and matrix.

2.4.2.3 Interfacial adhesion and compatibility between fibre and matrix

The interfacial adhesion between fibre and matrix is the important part in composites especially during the stress transfer from the matrix to the fibre during loading. A good adhesion will lead in effective stress transfer producing a tougher and stiffer materials. Good interfacial adhesion requires a good wetting between the fibre and the matrix, to achieve an extensive and proper interfacial contact for this the surface energies of the two materials are important parameters. However, there is challenges when using natural fibres as the reinforcement as they are hydrophilic that are contributed by hydroxyl groups in cellulose, lignin and hemicellulose), while thermoplastics such as polypropylene and polyethylene are hydrophobic (Rozman et al., 2003). This hydrophilic nature causes ineffective reaction with the matrix. In addition to this, pectin and waxy substance covers the reactive functional groups of the fibre and act as a barrier for the fibre to make adhesion with matrix.

To optimize good interfacial bonding between fibre and matrix, the fibre surface needs to be modified with different methods such as chemical and physical treatments. The chemical treatments include alkaline treatment, silane coupling treatment and acetylation. In these methods of treatments, a third material is introduced to act as a compatibiliser or coupling agent between the fibre and the matrix (Shah et al., 2012). Alkaline treatment is one of the most used chemical treatment of natural fibres when used to reinforce thermoplastics and thermosets. The important of modification done by alkaline treatment is the interruption of hydrogen bonding in the structure, therefore increasing surface roughness, thus leads to better adhesion of fibre and matrix. The chemical process is as the equation below:

$$Fibre + -OH \rightarrow fibre - O - Na + H_2O \tag{2.3}$$

(Ravi et al., 2018)

Physical treatments do not change the chemical composition of the fibres, but they change the structural and surface properties of the fibre and thereby influence the mechanical bonding to polymers. There are several ways to treat fibres using physical such as plasma, heat, stretching and calendaring. Based on previous study by Juliana Cruz and Raul Fangueiro, plasma treatment can introduce several groups at the surface and can form strong bond between fibre and matrix. Moreover, the clean and surface roughness of the natural fibres improved, thus results in better interfacial adhesion between fibres and matrix (Cruz & Fangueiro, 2016; Tran et al., 2015). In addition, physical treatments were chosen among the other treatments because they minimise the environmental impact of natural fibre surface treatments (Lee et al., 2011). Compared with chemical treatments, they will produce large quantities of hazardous chemicals the process of hydrophobising the natural fibres and the chemical waste must be handled and disposed in appropriate ways to reduce effect to the environment. This adds extra cost to the production of natural fibre reinforced composites, making the chemical treatments less attractive as a solution to the problems encounter.

The researchers found that there is improvement in mechanical properties after these treatments to this fibre, indicating the improvement to the interfacial adhesion. As example from M Asim et al. (Asim et al., 2018) study, the fibre has rougher surface and free from impurities after alkali treatment, thus increasing interfacial adhesion. Moreover, the tensile strength of the treated composites is higher compared to untreated with optimum concentration and soaking time. Higher chemicals concentration at higher soaking time may remove impurity efficiently but may decrease tensile strength due to lignocelluloses degradation and rupture of fibre surface. For heat treatment, optimum and suitable temperature are needed to treat the fibre due to lower thermal stability of natural fibres. Based on Shahzad A. (Shahzad, 2011) studies, the optimum temperature to treat hemp fibres was at 100°C and 150°C as they show an improvement in tensile strength, but no significant improvement was observed for 200°C treatment. Heat treatment at high temperature also results in embrittlement and damage of fibre, which is the evidence in considerable reduction in strain to failure of composites.

2.4.2.4 Failure mechanisms through fracture surfaces of the composites

Fibre reinforced polymer composites consists of two main components which are fibres and matrix. Fibres is the component that carry most of the loads, while matrix carries nearly no loads, but they ensure the interaction between fibres are good so that the composite will have excellent mechanical properties. There are several types of failure mechanism in composites including breaking of fibre, single fibre pull-out, interfacial gaps around fibres and debonding, failure of the matrix (Amir et al., 2017; Le Moigne et al., 2018). The different types of failure of composites have been illustrated in Figure 2.10 below.

Scanning Electron Microscopy (Kartal et al.) can be used to analyse the failure mechanisms and behaviour of the composites which provide direct visualization of the morphology at the filler/matrix interface (Amir et al., 2017). However, SEM will only provide qualitative information on the microstructure of the interfacial breakage mechanisms (failure surface), and quantitative information can be obtained by image analysing with measurements of microstructural parameters such as distribution and orientation of reinforcements.

Most of fibres exhibit brittle failure. Strong interfacial adhesion between fibres and matrix will lead to distinct fibre breaking without pull-out, hence produced good mechanical properties of the composites. Moreover, if the fibre percentage in the composites is maximised, the fibre and composite will fail at the same load level. However, there will be situations where the fibres do not fail at a specific load level. Besides, fibres fail in a series of failure events, with the weakest fibre breaking first.

Moreover, brittle failure of fibres with single fibre pull-out: In this type of failure, the fibre not only exhibits brittle failure, but also pulls out of matrix due to debonding of fibre and matrix. This situation may be due to poor interfacial adhesion between fibres and matrix. Hence, the voids will make the loading transfer at the fibre-matrix interface is ineffective making the composites poor in mechanical properties.

In addition, the influence of the fibre bundles during a tensile experiment may also affect the failure mechanism of composites. There may be some occurrence of micro cracks through the transversely oriented fibre bundles, which induce a dislocation between the elementary fibres. This observation evidences that the weakness of natural fibres composites was mainly located at the bundles of fibres rather than the fibre matrix interfacial adhesion.

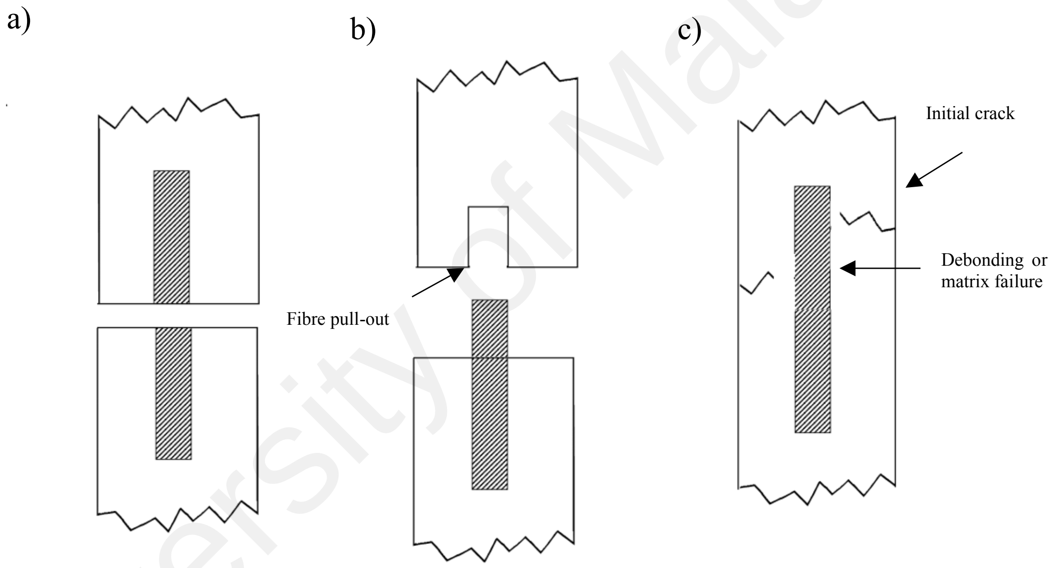


Figure 2.10: The different types of failure of composites
(a) Fibre breakage, (b) single fibre pull-out,(c) debonding and/or matrix cracking.

2.4.2.5 Resistance against environment

Durability of a material, in general is defined as the service life of a material under given environmental conditions (Ramakrishna et al., 2010). Natural fibre composites combine plant-derived lignocellulose fibres with a polymeric matrix. The natural fibre component may be wood, sisal, hemp, coconut, cotton, kenaf, flax, EFB, jute, abaca, banana leaf fibres, bamboo, wheat straw or other fibrous material, and the matrix can be a polymeric material such as PP, HDPE and PE. Advantages of natural fibre composites include lightweight, low-energy production and degradable which depends on the materials used. In order to study the durability of natural fibre composites, their exposure to various environments, degradation and weathering characteristics must be investigated.

Based on studies done by Ramakrishna G. et al. (Ramakrishna et al., 2010), the composites were exposed with NaOH in medium concentration. As a result, they reflect the changes in the strength due to the interaction between the matrix and any medium considered (NaOH in the present case) and hence can be used with confidence to evaluate the durability of natural fibre reinforced mortar composites and the relative performance of composites.

The application of natural fibres as reinforcement in composite materials is constantly in development especially in civil engineering field. Composites are used in permanent formworks, facades, tanks, pipes and long span roofing elements. Therefore, it is necessary to understand the nature and the compositions of natural fibres to gain a deep understanding on the degradation matters. Natural fibre composites have high opportunity of degradation when subjected to outdoor applications as compared to composites with synthetic fibres. This is attributed to the characteristics of natural fibres, which are susceptible to biodegradation (Azwa et al., 2013; Ramakrishna et al., 2010). Degradation of natural fibre/polymer composite in an outdoor environment is influenced by factors such as moisture absorption, temperature, ultraviolet radiation, and microorganism activities.

As reported, natural fibres absorb moisture due to content of hemicellulose and may be affect the performances of the composites. It has been stated that the water absorption for natural fibre composites was found to be typically 0.7–2% after 24 h, 1–5% after a week, and up to 18–22% after several months (Dittenber & GangaRao, 2012). This behaviour was attributed to the wood fibre chemical compositions in having high hydrophilic content. One of fibre component that responsible for this water absorption is the high content of hemicellulose. The water absorption of composites will lead to poor adhesion between fibre particles and polymer matrix generates void spaces around the fibre particles. Higher fibre volume composites immersed in water generally have lowering their strength in tensile and flexural properties compared to dry samples. This can be reduced through several types of fibre modifications as mention before.

Other than moisture absorption, these bio composites also facing problems where they have the potential to degrade when subjected to outdoor application prior to properties of natural fibres which is biodegradable (Azwa et al., 2013; Dittenber & GangaRao, 2012) of a composite may occur with the degradation of its individual constituents by losing interfacial strength between them such as hemicellulose and lignin.

CHAPTER 3:METHODOLOGY

3.1 Materials

A commercial semi-crystalline high-density polyethylene (HDPE), Titanzex HI 1100 with melt-flow index of 6.6 g/10 min (load of 2.16 kg at 190°C) and density of 0.96 g/cm^{3}manufactured by Titan Petchem (M) Sdn. Bhd, Malaysia was used as the matrix of the composites. Oil palm empty fruit bunch (EFB) fibre with average length of 3 mm (mesh 40) was used as reinforcement to the composites. Preliminary to the mixing process, EFB was dried for 24 h at 80°C using vacuum oven.

The physical properties of HDPE and EFB fibre was measured and data obtained are presented in Table 3.1.

Table 3.1: Physical properties of HDPE and EFB fibre.

Property	HDPE	EFB fibre (Untreated)	EFB fibre (Treated)	ASTM method
Melting temperature (°C)	135.14	-	-	-
Melt index (g/10 min)	6.60	-	-	D1238
Density (g/cm^3)	0.96	0.35	-	-
Tensile strength (MPa)	21.27	76.32	79.72	D 638, C 1557
Tensile modulus (GPa)	1.65	2.14	2.28	D 638, C 1557
Tensile strain (%)	9.39	17.32	14.88	D 638, C 1557

3.2 Preliminary studies on EFB fibres

EFB fibre was dried in a vacuum oven to remove moisture content at 80°C for about 24 hours. The dried fibre was subjected to four different temperature profiles which are: 140, 160, 180 and 200°C for 1 hour using vacuum oven. The thermal, mechanical behaviour and morphology of the treated fibre was determined. The optimum fibre temperature condition was chosen and used for the next stage.

3.3 Processing

Processing of Polymer Matrix Composites are the major manufacturing processes used for polymer matrix composites and describes process details, process parameters, analytical and experimental methods used for understanding process conditions. The preparation of polymer matrix composites involves many steps from raw materials until it become the sample for testing. The detailed process of the preparations was discussed in this chapter.

3.3.1 Compounding

According to desired weight fraction, fibre and matrix required were calculated and weighted. The EFB/HDPE composites were prepared for untreated EFB and the second set were prepared using treated EFB fibre as presented in Table 3.2. EFB was dried for 24 h at 80°C using vacuum oven before the mixing process. EFB fibre was heat treated at 180°C using vacuum oven for 1 hour in this study. The materials with different EFB content (10, 20 and 30 wt.%) were physically mixed in 500 g portions and compounded using the co-rotating twin screw extruder with the screw diameter and screw aspect ratio of 20 and 40 mm, respectively (Brabender KETSE 20/40, Germany). The screw speed of 80 rpm was used and the temperature profile was set between 140°C and 165°. The materials extruded from the formulations were pelletized into length of about 6 mm. the pelletized samples were stored in sealed plastic bag inside a desiccator after being oven dried at 80°C for 24 h.

Table 3.2: Formulations of composites.

Sample code	**High density polyethylene (w/w %)**	**EFB fibre (w/w %)**	**Fibre treatment**
HDPE	100	0	-
HDPE/EFB$_{U}$10	90	10	Untreated
HDPE/EFB$_{U}$20	80	20	Untreated
HDPE/EFB$_{U}$30	70	30	Untreated
HDPE/EFB$_{180}$10	90	10	Heat treated 180°C /1hour
HDPE/EFB$_{180}$20	80	20	Heat treated 180°C /1hour
HDPE/EFB$_{180}$30	70	30	Heat treated 180°C /1hour

3.3.2 Injection moulding

The dried pelletized composites were injection moulded into dumb-bell shaped and bar shaped specimens according to ASTM D638 and ASTM E23 (ASTM D638-10 in Standard Test Method for Tensile Properties of Plastics, 2010; ASTM E23-07, in Standard Test Methods for Notched Bar Impact Testing of Metallic Materials, 2007) respectively using a Boy 55M (Germany), with a 55-tonne clamping force injection moulding machine. The processing temperature were set at 170°C, injection pressure of 100-200 bar and mould temperature of 25°C.

3.4 Conditioning

Subjected to two different environments, the injection moulded tensile and impact were expected to be encountered in the service life of the composites material in order to assess their effects on the composite properties. Figure 3.1 shows the set up for both conditions.

3.4.1 Indoor conditioning

In the laboratory, all samples arranged on a rack and exposed to ambient conditions in the laboratory. Both untreated and treated composites were condition at 6 months' period time. The samples were removed and tested for thermal and mechanical properties at the end of the period.

3.4.2 Soil burial conditioning

Samples buried in garden soil to a depth of 30 cm in an open space free from shade for a period of 6 months from June 2016 until December 2016. Then, the samples were removed, washed, wiped clean and left to air dry at ambient temperature for 24 h prior to testing. The samples were tested for mechanical and thermal tests to assess the degree of degradation due to soil burial environment. Samples were buried at the University of Malaya main campus and the burial station location were geographically located at Petaling Jaya on the latitude of 03º 07` north of the Equator and longitude 101º 38` east of the Prime Meridian. The mean temperature and total rainfall in Petaling Jaya were compiled for each month according to the data obtained from Malaysian Meteorological Agency and presented in Table 3.3.

Table 3.3: Petaling Jaya weather data from June 1st to December 31st 2016.

Month	**Mean relative humidity (%)**	**Mean rainfall (mm)**	**Mean temperature (°C)**	
			Min	Max
June	72.2	9.6	25.7	33.4
July	74.4	8.1	25.4	33.3
August	69.4	4.0	26.2	33.4
September	71.4	9.0	25.3	33.1
October	73.0	5.8	25.5	32.7
November	81.0	15.8	24.9	32.0
December	78.3	10.3	24.9	32.0

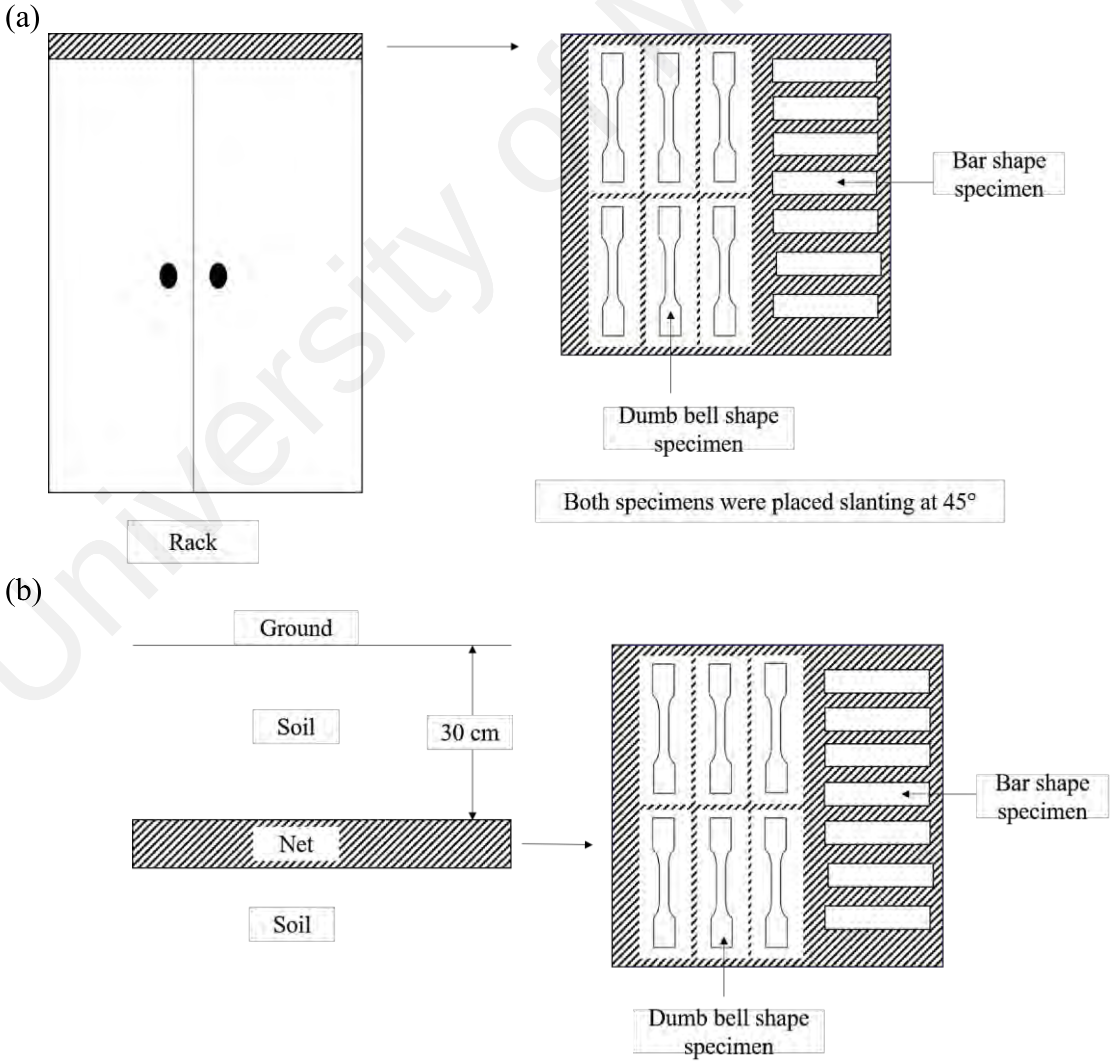


Figure 3.1: conditioning set up: (a) indoor, (b) soil burial.

3.5 Characterisation of EFB fibres

Before compounding with polymer matrix, the EFB fibre were characterized for both untreated and treated condition. They were characterized by chemical, physical, thermal and mechanical properties. For treatment, the fibres were tested at four different temperatures treatment to choose the optimum condition to compound with the polymer matrix. The detailed characterisation were discussed further.

3.5.1 Chemical properties

Chemical composition of EFB fibres

The chemical composition of untreated and treated EFB fibre were done in Malaysian Agricultural Research and Development Institute (MARDI). By using the van Soest et al. Van Soest method, the compositions of hemicellulose, cellulose and lignin in EFB were analysed. The method had successfully separated the plant cells by using two detergents known as neutral detergent fibre (NDF) and acid detergent fibre (ADF). NDF analysis measures the lignin, hemicellulose and cellulose components while ADF analysis measures the cellulose and lignin component in plant cell. Acid detergent lignin (ADL) analysis measures the lignin content of the fibre. According to the ASTM E1755 (ASTM E1755 – 01, Standard Test Method for Ash in Biomass, 2015), the ash content was determined.

Fourier transform infrared spectroscopic analysis (FTIR)

Infrared absorption spectra of raw materials and composites were recorded using a FTIR-ATR model spectrum 400 (Perkin Elmer, USA) at a resolution of 4 cm^{-1} with 64 sample scans for each spectrum in the wavelength of 4000 cm^{-1} to 500 cm^{-1}. Samples were pressed against the diamond crystal of ATR unit.

3.5.2 Physical properties

Determination of EFB fibre density

The density of EFB fibre was determined according to ASTM E212 with some modifications. A measuring cylinder with a known volume, V was placed on a weighing balance and tared to zero weight. The moisture-free EFB fibre was loaded into the cylinder until filled and overflowing. A spatula was used to level off the EFB fibre with the top of the measuring cylinder. To avoid spilling of EFB fibre during transfer, the side of measuring cylinder was taped gently. The filled cylinder was weighted to the nearest 0.1 g to determine mass, M of EFB fibre. The apparent density of the fibre was calculated using the following equation;

$$\rho = \frac{M}{V} \tag{3.1}$$

where, M is the mass of EFB fibre in grams and V is the volume of the measuring cylinder.

Fibre length distribution

Image Analyser, Leco model IA-32 was used to measure the fibre length. On the observation stage of a transmitted light microscope with resolution of 5Xm, a glass slide was placed. A video camera attached to the microscope transmitted live fibre image to the Image Analyser. Samples from the central portion of injection-moulded tensile test pieces were cut and the polymer matrix was removed by using the Soxhlet extraction method. Xylene were used as a solvent in the method. The extraction process was conducted in boiling xylene (~138°C) for 3 days. The extracted fibres were used for the measurement. Not less than 500 fibres lengths were measured for the determination of fibre length distribution (FLD).

Moisture content

EFB fibre was dried in vacuum oven at temperature of 80°C for 24 hours to a constant weight. The moisture content was expressed as a percentage loss in weight of the final oven-dried weight using Equation 3.2;

$$Moisture\ content\ (\%) = \frac{W_1 - W_0}{W_0}\ x\ 100\% \quad (3.2)$$

where, W_0 is weight of fibre before drying and W_1 is weight of fibre after drying.

Determination of EFB surface morphology

EFB fibre surface were examined using a Hitachi Field Emission Scanning Electron Microscopy model SU8220 to study the morphology of the fibres. The specimen surface was coated prior to testing to prevent charging.

3.5.3 Tensile test for single fibre

Universal Testing Machine, Instron 3345 (USA) was used to measure the tensile test of EFB fibre. Load cell of 0.1 kN was used, following ASTM C1557 E1755 (ASTM C1557-14, Standard Test Method for Tensile Strength and Young's Modulus of Fibers., 2015), with a cross-head speed of 2 mm/min at room temperature of about 25°C. The diameter of fibre was required to obtain its cross-sectional area before carrying out the tensile test. Natural fibres possessed non-uniform diameter unlike synthetic fibres. Therefore, natural fibres were assumed to be in a perfect cylindrical geometry. It was determined using Leco IA-32 Image Analyser an optical microscope, ten measurements were taken and average value for each specimen was calculated. At least 90 specimens were prepared for each untreated and heat-treated sample in this study. Single fibre strand was randomly selected from the fibre bundle and was glued to a paperboard with dimension presented by Figure 3.2.

Single fibre tensile specimen was mounted to the testing machine so that the fibres are positioned vertically and the side frame (shown in Figure 3.2) was cut before running the test. The test was carried out at a crosshead speed of 1 mm/min with a 0.1 kN load cell. Tensile modulus, tensile strain and tensile strength data points were taken to correlate with the diameter distribution of fibre. Meanwhile, the average value for tensile modulus, tensile and tensile strength were calculated.

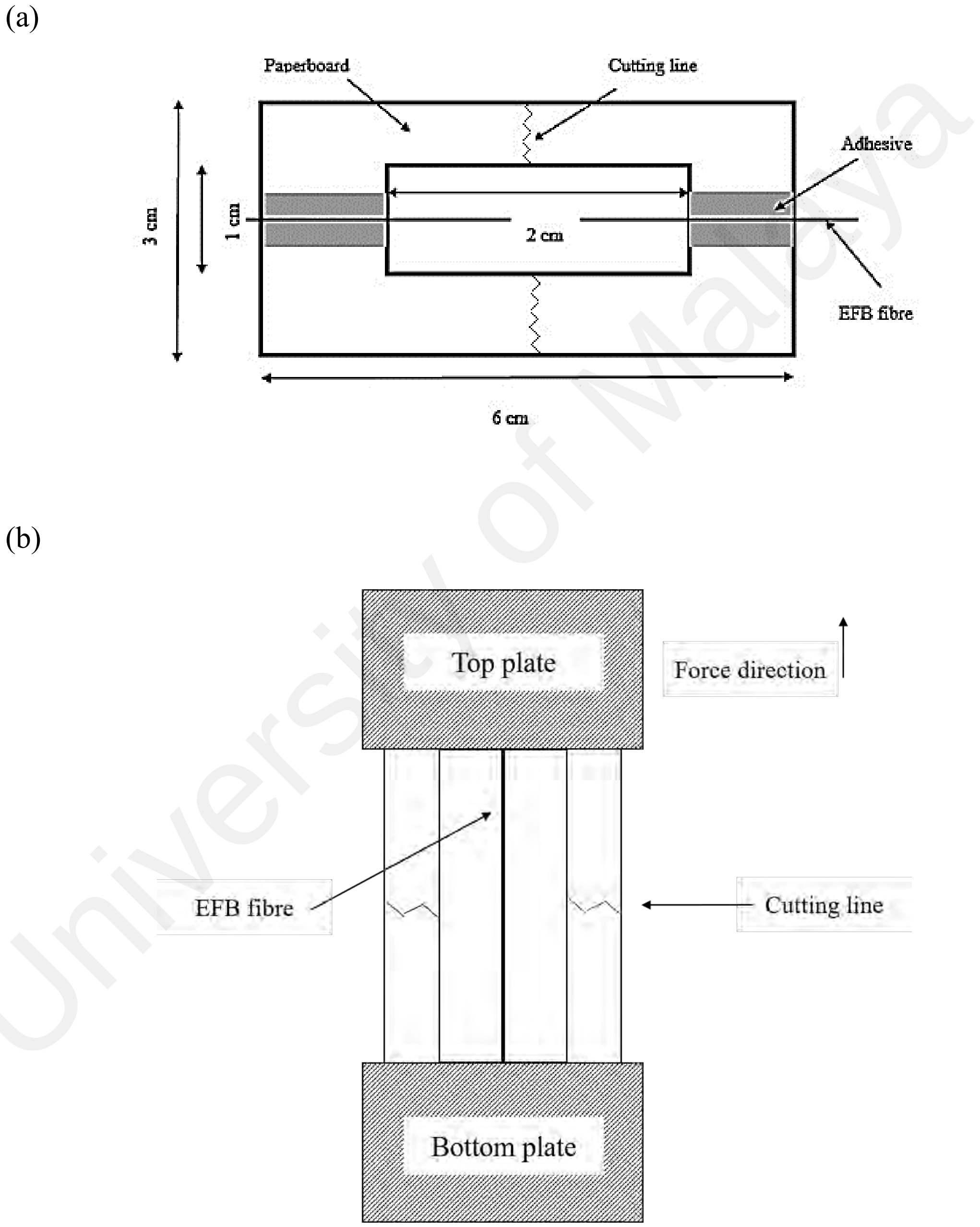


Figure 3.2: Tensile specimen for single continuous fibre: (a) mounting board dimension, (b) test set up.

3.6 Characterisation of composite materials

3.6.1 Morphology analysis

Selected fractured specimens from tensile and impact tests were evaluated using a Hitachi Field Emission Scanning Electron Microscopy (FESEM) model SU8220 to study the morphology and the fracture behaviour of the composite. The specimen surface was coated prior to testing to prevent charging.

3.6.2 Thermal characterisation

Thermogravimetric Analysis (TGA)

Thermogravimetric analysis (TGA) study was used to investigate the decomposition temperature, thermal stability and composition of the composite. TGA was done using a TGA 6 Thermogravimetric Analyser (Perkin Elmer) under nitrogen atmosphere with a flow rate of 20 mL/min and a scan rate of 10°C/ min over a temperature range of 30°C to 900°C. A test sample of 8-10 mg was used for each run. The curves obtained from the instrument were analysed for $T_{50\%}$ (temperature at 50% degradation) and T_{onset} (onset temperature)

Differential Scanning Calorimetry (DSC)

The melting temperature, crystallisation temperature and glass transition temperature could be obtained from Differential Scanning Calorimetry (DSC) analysis. The DSC experiments were performed with a Perkin Elmer Diamond DSC (USA). Each sample was subjected to heating, cooling and second heating cycles at a scanning rate of 10°C/ min under nitrogen atmosphere with the nitrogen flow rate of 20 mL/min, to prevent oxidation. The test sample of between 5 and 10 mg was crimped in an aluminium pan and tested over a temperature range of 30°C to 250°C.

Melt Flow Index (MFI)

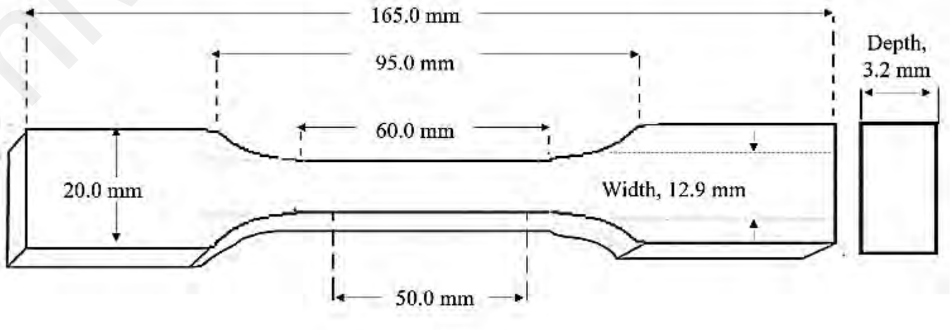
The melt flow indexes for the composites were tested using KARG Industri Technik Multiflow. The load weight used was 2.16kg and temperature at 190°C was in accordance to ASTM D1238 (ASTM D1238-13, Standard Test Method for Melt Flow Rates of Thermoplastics by Extrusion Plastometer, 2013). MFI measures the ease of flow of the melt of a thermoplastic polymer.

3.6.3 Mechanical Characterisation

Tensile test

Tensile tests for composites were carried out using a Universal Testing Machine, Instron 5569 equipped with a load cell of 50kN and a mechanical extensometer. Composites were tested using ASTM standard D638 (ASTM D638-10 in Standard Test Method for Tensile Properties of Plastics, 2010) with a constant cross-head speed of 10 mm/min at room temperature of about 25°C. Stress–strain curves were obtained from this test. Tensile modulus, tensile strength (maximum stress) and tensile strain were calculated from these curves. Figure 3.3 shows the tensile specimen dimensions and tensile testing set up.

(a)



(b)

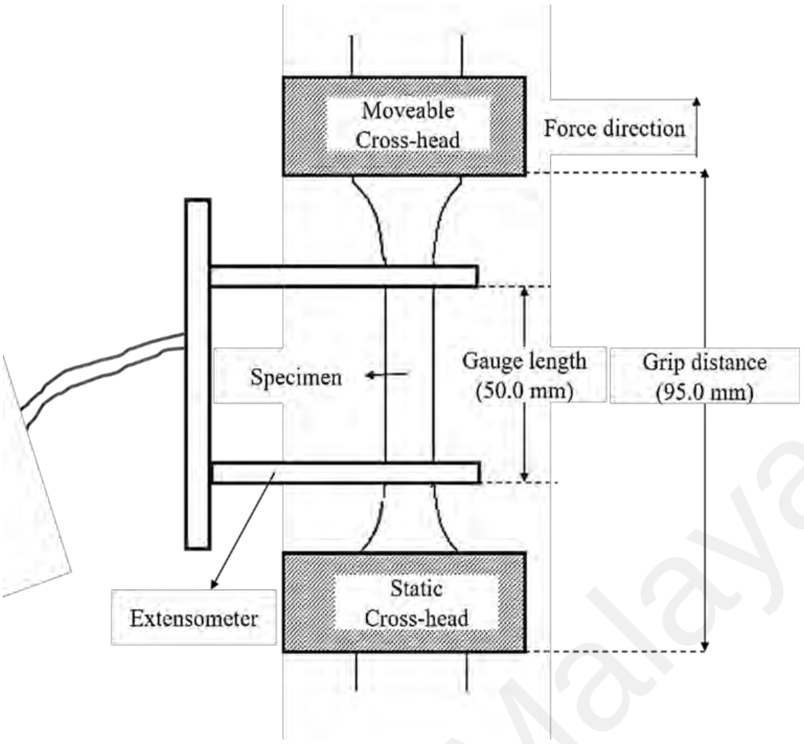


Figure 3.3: Tensile testing: (a) specimen dimensions, (b) set up.

Flexural test

Flexural tests were carried out using a Universal Testing Machine, under three-point bending mode, according to ASTM standard D790 D790 (ASTM D790-10 in Standard Test Methods for Flexural Properties of Unreinforced and Reinforced Plastics and Electrical Insulating Materials, 2010). The distance between the support spans (L) was set at 50 cm. All the specimens were tested at a constant cross-head speed of 1.27 mm/min at room temperature of about 25°C. The speed of cross-head motion (R) was calculated by using the equation:

$$R = \frac{ZL^2}{6d} \qquad (3.3)$$

Where *L* and *d* are the specimens support span and depth respectively. Z is the rate of straining equal to 0.01. Ten specimens per batch were conducted and the best six results were recorded. Flexural strength and modulus were calculated using Bluehill® software form the curve obtained. Figure 3.4 show the flexural testing set up.

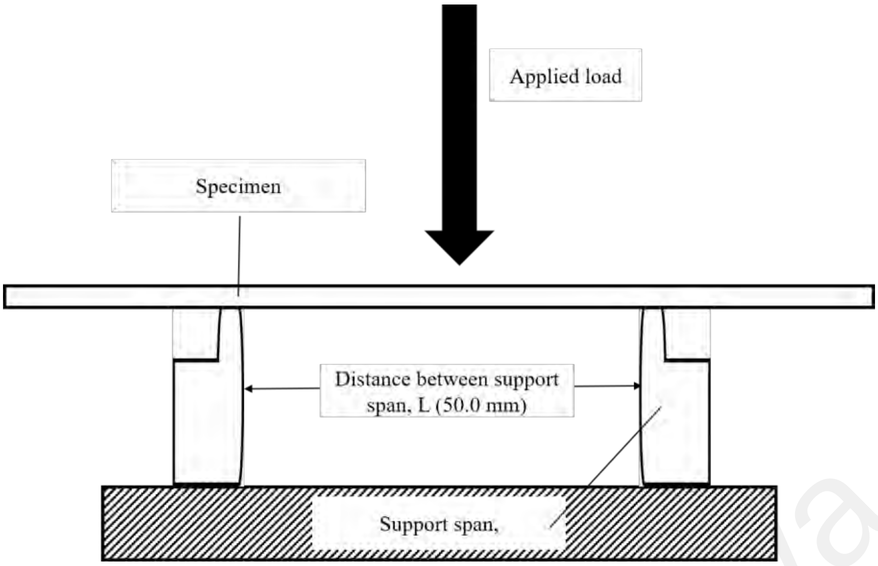


Figure 3.4: Flexural testing set up.

Impact test

The impact test bars were notched at the centre of one edge to produce single edge notch (SEN) impact test specimen. The impact test bars were tested at four different notch-to-depth (a/D) ratios of 0.1, 0.2, 0.3 and 0.4. The support span-to-depth ratio (S/D) was maintained at four throughout the experiment. The impact test is run under charpy mode using an Instron Dynatup 9210 (USA) instrumented falling weight impact tester with a V-shape impactor tup. The specimen dimension is accordance with ASTM E23-07 (ASTM E23-07, in Standard Test Methods for Notched Bar Impact Testing of Metallic Materials, 2007). Figure 3.5 show the impact specimen dimensions and testing set up.

3.6.4 Determination of water absorption behaviour

Samples which undergo soil burial and indoor conditioning for 6 months were measured. For every 2 days, the samples were removed, cleaned and wiped. Then the samples were weighted and put back in the conditions.

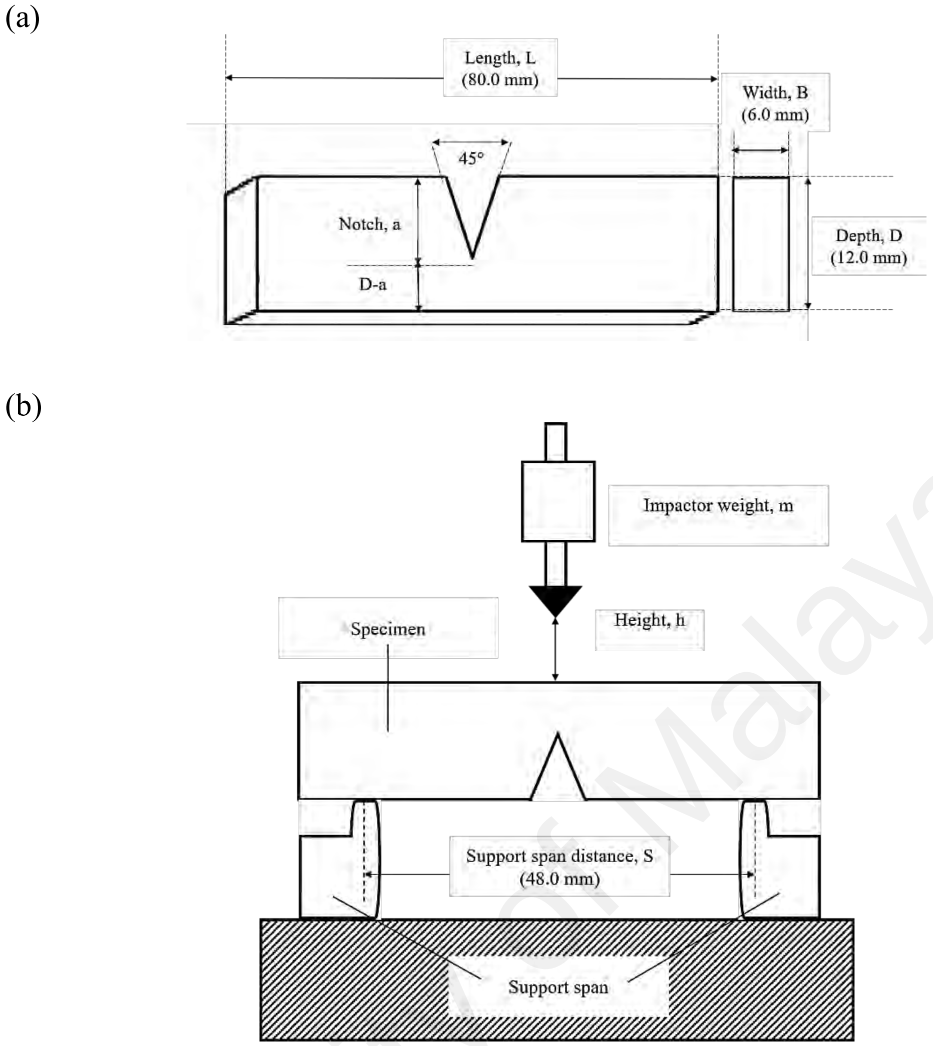


Figure 3.5: Impact testing: (a) Specimen dimensions, (b) set up.

CHAPTER 4: RESULTS & DISCUSSION

4.1 Properties of EFB fibres

4.1.1 Chemical properties

Chemical composition of EFB fibres

Cellulose, hemicellulose and lignin that forms main constituents of the natural fibres might differ depending on plant age and growth conditions, soil conditions, weather effect, and testing methods used. Natural fibre also consists of ash content and other extractives and impurities. As the chemistry of the natural fibre such as EFB is based on plant they grow, it changes during growth. The properties of the fibres such as tensile strength, flexural strengths, and rigidity depend on the alignment of cellulose fibrils, which are generally arranged along the fibre length. Table 4.1 below shows the chemical composition of untreated and treated raw EFB fibre, which the untreated EFB fibre consists of 45.3% cellulose, 23.1% of hemicellulose, 14.1% of lignin and about 17.4% of others content. Others can be part of ash content, impurities and inorganic element such as calcium, magnesium and silica (Anuar et al., 2018; Ibrahim et al., 2015). The silica content can be clearly seen embedded on the fibre surface when observed under SEM (Figure 4.7). As mention before, cellulose is the crystalline part in EFB and have a formula of $(C_6H_{10}O_5)_n$ consisting of hundreds to thousands of β (1→4) glucose unit, which connected to form a straight linear chain. Cellulose is tough, fibrous and water-insoluble polymers that support the structure framework in the cell walls. For treated EFB fibres, the composition of cellulose, hemicellulose and lignin have a slight increment compared to the untreated EFB. This indicates that heat treatment was not an effective way to remove the composition of cellulose, hemicellulose and lignin compare to chemical treatment (Hassan & Badri, 2016). However, heat treatment shows an observable reduction on other compositions after the treatment. The percentage of others composition in untreated EFB is 17.4% while for the treated EFB was 7.8%. This can be concluded that heat treatment may remove other compositions such as impurities and silica (Figure 4.8). High content of silica in plant can cause the decrease in mechanical properties of the composites as the fibre-matrix adhesion will be poor. All this information will be used as a guidance for the treatment temperature of EFB fibres, as well as the processing settings for the composites.

Table 4.1: Chemical composition of untreated and treated EFB fibre.

Chemical constituents	% Composition (Untreated)	% Composition (Treated)
Cellulose	45.3	51.4
Hemicellulose	23.1	24.0
Lignin	14.1	16.9
Others	17.4	7.8

Fourier-transform infra-red properties (FTIR)

FTIR is an important tool used to study and recognized the existence of functional groups in a molecule. The FTIR of both untreated and heat treated at different temperature is presented in Figure 4.1. A broad peak of cellulose hydroxyl groups (Wang et al.) appears at 3320 cm^{-1} and another peak at 1590 cm^{-1} belongs to C=C stretching, indicating that EFB fibres contains fatty acid from lignin. The bands at 2920 cm^{-1} is attributed to C-H stretching vibration of methyl or methylene group in cellulose and hemicellulose contain in EFB fibre (Alam et al., 2015; Sefadi & Luyt, 2012). A shoulder band at 1620 cm^{-1}observed belongs to C=O carbonyl stretching (Sefadi & Luyt, 2012). The peak appears around 1310cm^{-1} observed to be belong to the aromatic skeletal (=CH) vibrations in lignin. Two small bands appear at 850 cm^{-1} and 893 cm^{-1}, which were characteristic of the β-glycosidic linkages between sugar units in hemicellulose and cellulose (Alam et al., 2015). The spectra look similar even after the treatment as the entire samples exhibit the characteristic signals of lignocellulosic. Despite the similarities observed in all spectra, a decrease in intensity of the (Wang et al.) absorption band at 3320 cm^{-1} is observed in the heat-treated samples. For example, at 180°C fibre treatment, the OH intensity decrease from 806 cm^{-1} to 751 cm^{-1} when compare to the untreated EFB fibres. This suggests that O-H group content in the heat treated EFB is slightly reduced after heat treatment despite the temperature of treatment. This decrease occurs as a result of elimination of O-H groups in EFB, making the fibre change from hydrophilic to hydrophobic nature to bond with HDPE for the preparation of composites (Jayamani et al., 2014). However, the OH intensity of 200°C EFB fibre treatment seems to be increasing when comparing to the untreated fibres. This may be due to the fibre degradation as the heat fibre is being treated at high temperature.

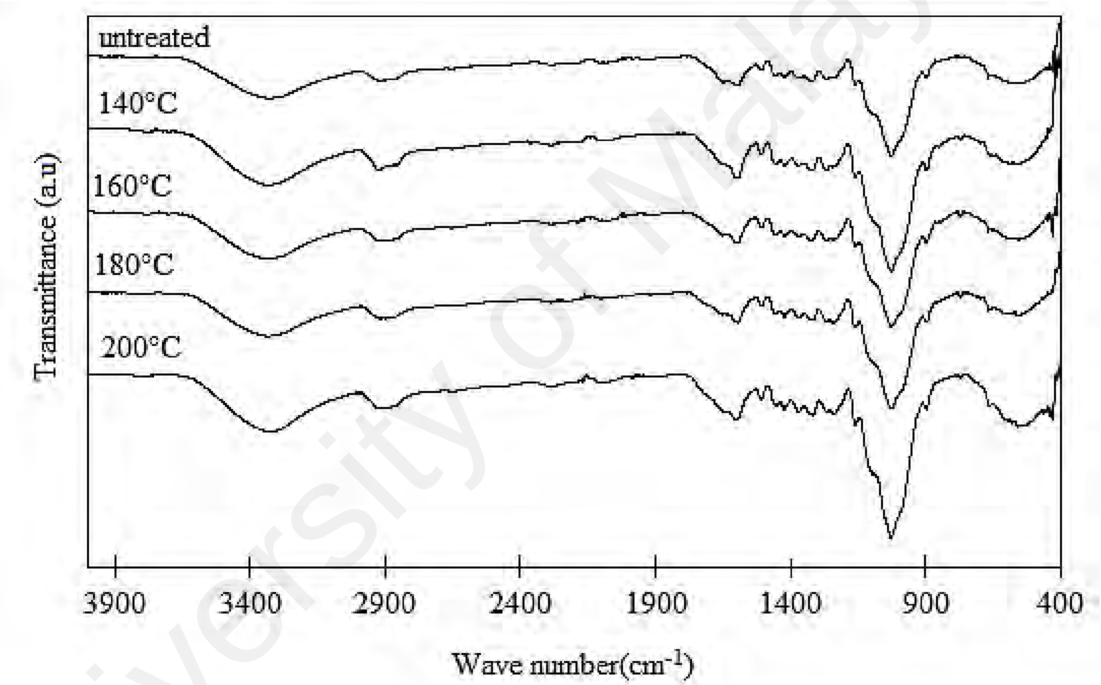


Figure 4.1: FTIR spectra of untreated and treated EFB fibre at different temperature.

4.1.2 Physical properties

Fibre length distribution (FLD)

Fibre length

Histogram of percentage of fibre counts against fibre length before compounding is shown in Figure 4.2. From the figure, the fibre length distribution of EFB fibre before compounding is quite normal with a bell shape. The highest of the fibre distribution (193 counts) was observed in the range of 1.50–1.99 mm followed by range 2.00-2.49 mm with 148 counts. The lowest fibre population was observed in the range of 4.50-4.99 mm with the fibre counts of two fibres. Number-average fibre length L_n and weight-average fibre length L_w were also calculated using the following equations:

$$L_n = \frac{\sum n_i \ L_i}{\sum n_i} \tag{4.1}$$

$$L_w = \frac{\sum n_i \ L_i2}{\sum n_i} \tag{4.2}$$

Where n_i is the number of fibres of length L_i and a mid-point of fibre length range was taken as an average value of fibre length, L_i. From calculation, L_n and L_w values of the fibre length were found to be 1.86 mm and 2.11 mm respectively.

Histograms of fibre length distribution (FLD) of raw EFB, untreated and treated EFB fibre after compounding are shown in Figures 4.2, 4.3 and 4.4. The EFB fibre were expected to break to a shorter length during processing. After being compounded with HDPE, the length of EFB fibre from untreated fibre composites reduced about 2mm. The highest fibre length observed after the processing is at range of 2.51-3.00 mm for 13 fibres count, whereas the lowest fibre length is at 0.00-0.50 mm for 37 fibre counts. From studies done by F. Willems and C. Bonten (Willems & Bonten, 2016), the fibre breakage is suggested to an increase in fibre-fibre interaction in the melt phase of the polymer. Moreover, the reduction of the fibres after processing may also be due to geometrical factors whereby the fibre is compound with polymer and extrude into a certain shape. The smaller the shape of the final products, the higher the fibre reduction. From calculation, L_n and L_w values of the fibre length were found to be 1.25 mm and 1.52 mm respectively.

The same trend was observed with treated fibre composites. The length of EFB fibre is reduced but to a lower extend where it was reduced to 1 mm compared to the untreated fibre composites. The treated fibre composites showed highest fibre length population at the range of 3.51-4.00 mm for 1 fibre count, whereas the lowest fibre length at range of

0-0.5 mm for 46 fibre counts. On the other hand, L_n and L_w values of the fibre length were found to be 1.60 mm and 1.88 mm respectively which are higher than that of measured for untreated fibre composites.

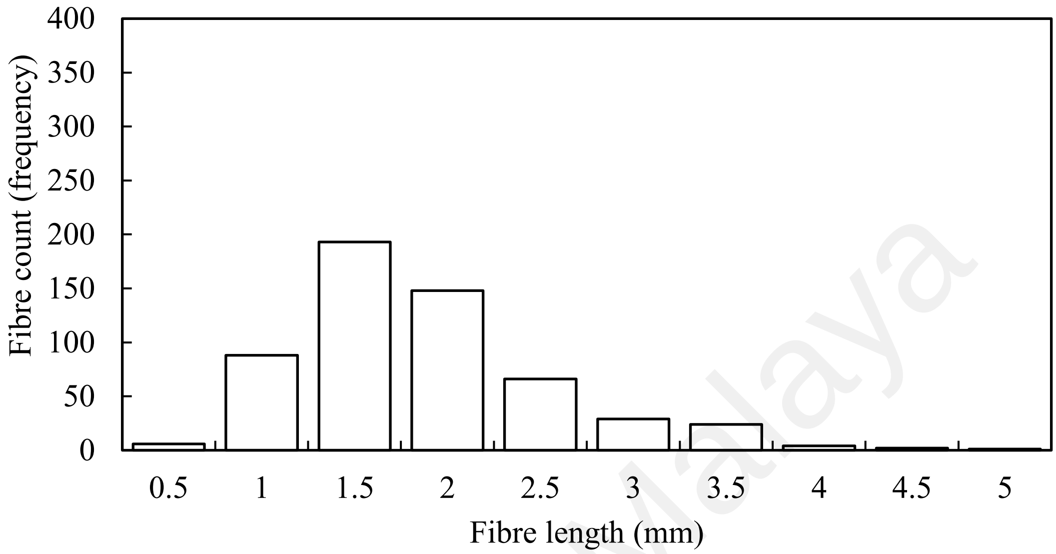


Figure 4.2: Fibre length distribution of raw EFB fibre before compounding.

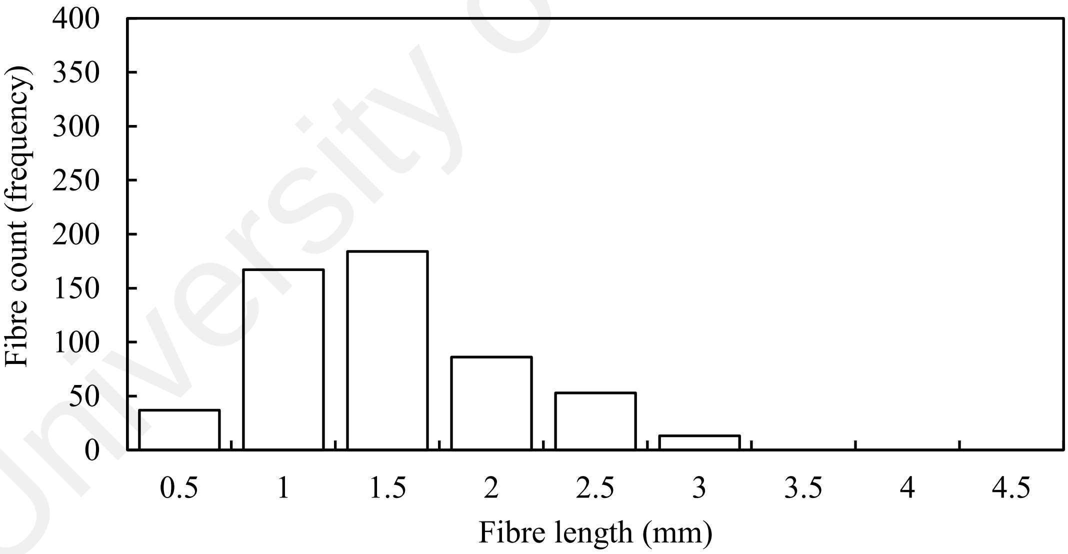


Figure 4.3: Fibre length distribution of EFB fibre from 30% untreated fibre composites after compounding.

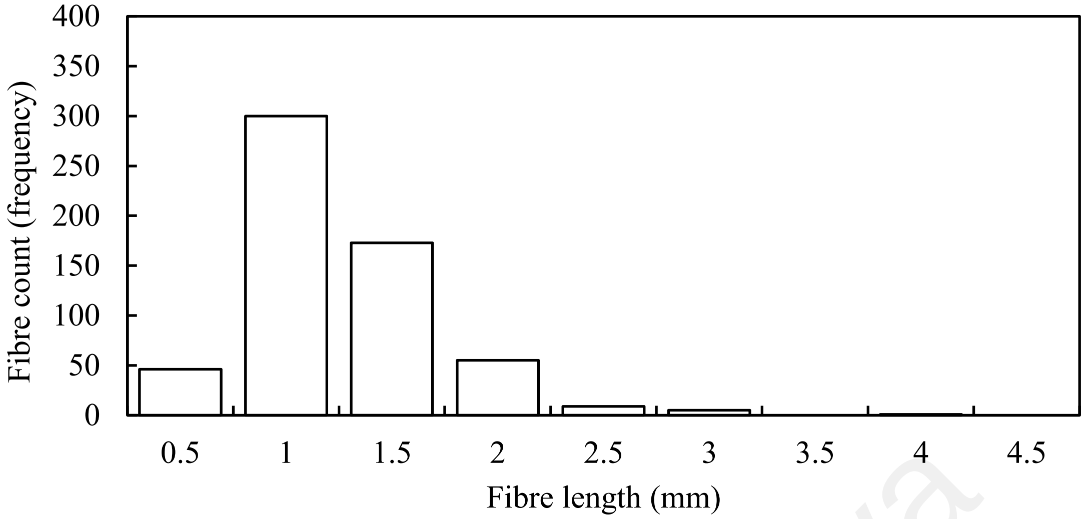


Figure 4.4: Fibre length distribution of EFB fibre from 30% treated fibre composites after compounding.

Fibre diameter

The histogram of fibre diameter before and after the treatment was shown in Figure 4.5 and 4.6. The histogram shows that the fibre diameter has a normal bell shape distribution in both figures. Before fibre treatment, the highest population was in the range of 401-450 µm with 30 fibre counts whereas the lowest fibre range was in the range of 601-650 µm with fibre count of two. Number-average fibre diameter, D_n and weight-average fibre diameter, D_w were calculated using the following equations:

$$D_n = \frac{\sum n_i \; D_i}{\sum n_i} \tag{4.3}$$

$$D_w = \frac{\sum n_i \; D_{i2}}{\sum n_i \; D_i} \tag{4.4}$$

From calculation, D_n and D_w values of the fibre length were found to be 392.31 µm and 408.21 µm respectively.

However, the fibre diameter is reduced after the treatment compared with the untreated fibres. The highest population was in range of 351-400 µm with 25 fibre counts whereas the lowest fibre range was in the range of 601-650 µm with fibre count of one. This reduction is expected because the heat treatment removes the waxy layer and some impurities such as silica, hemicellulose and lignin on the amorphous surface of EFB fibre (Asim et al., 2018). This can be seen in SEM image of EFB fibre in Figure 4.7 (b) where the surface of EFB fibre after treatment is clean and rough compared with the untreated fibres in Figure 4.7(a). From calculation, D_n and D_w values of the fibre diameter were found to be 364.3 μm and 394.5 μm respectively.

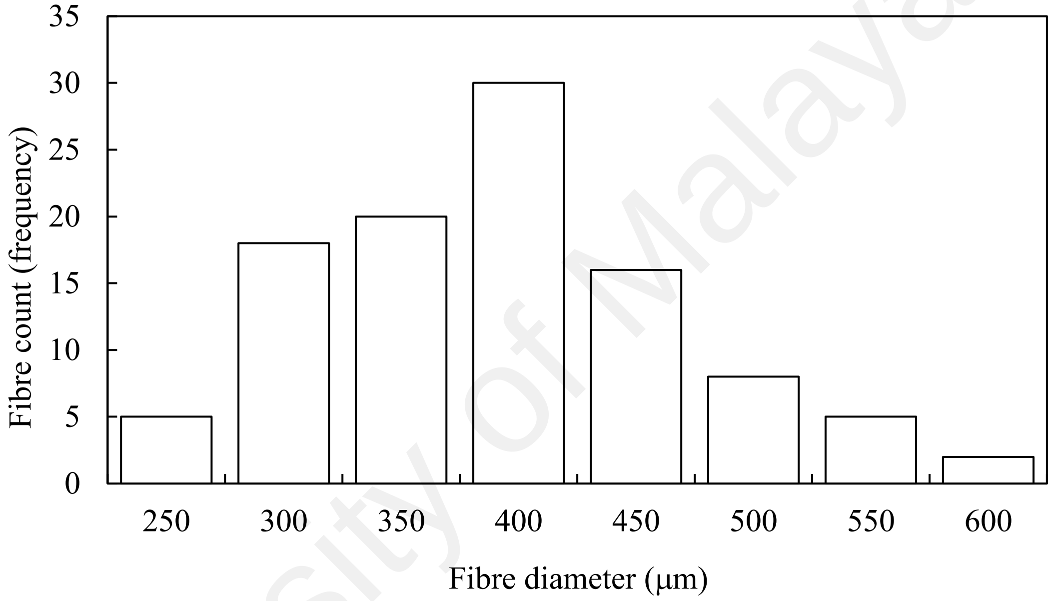


Figure 4.5: Fibre diameter before treatment.

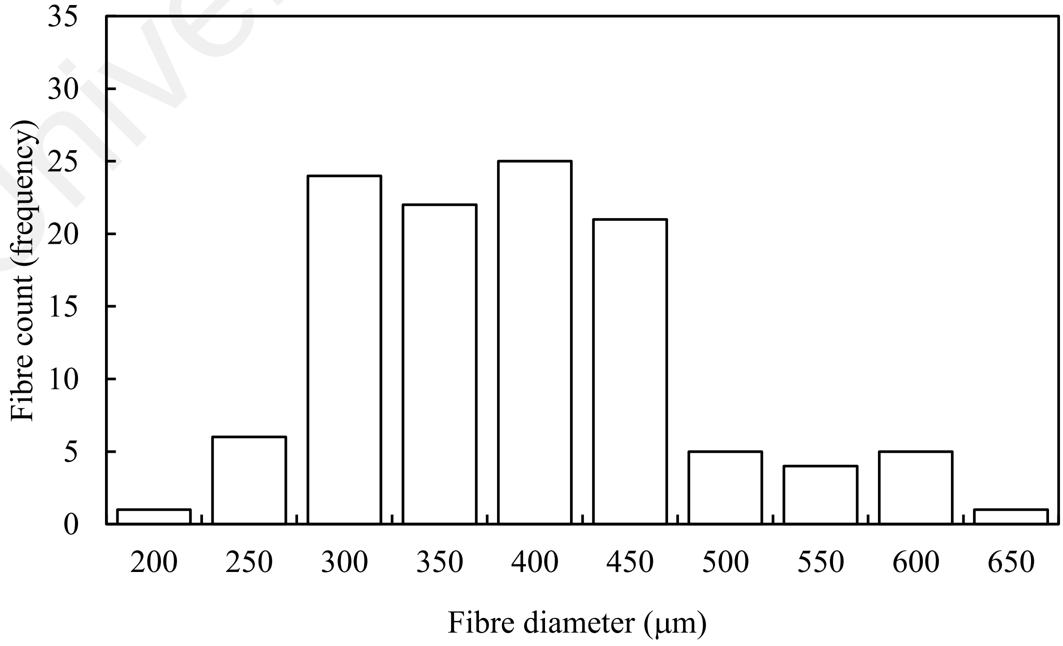


Figure 4.6: Fibre diameter after heat treatment 180°C.

EFB surface morphology

Figure 4.7(a) and (b) illustrate the SEM images for the surface of raw untreated EFB fibre with different magnification. The EFB fibre had a smooth surface with a layer of materials like lignin or waxes that covered the whole surface of the fibre. This layer may be acting as the protective layer present in most plants to avoid water loss (Shamsudin et al., 2012).

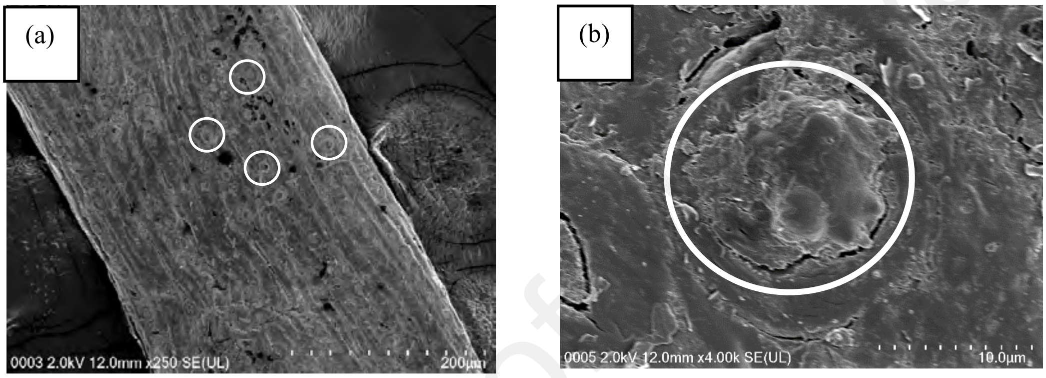


Figure 4.7: SEM images (a) and (b) of raw EFB fibre at different magnification (250x; 6000x).

EFB surface morphology

Moreover, there are mainly silica bodies found on the surface of the fibre strands as presented in the Figure 4.7 (a) and (b) but they were covered with the waxy layer and can’t be seen clearly. The silica bodies are connected to circular craters, which spread uniformly over the strand’s surface. Silica bodies are the most often mineral found on the surface of woody plants, formed by the invasion and hardening of soil minerals into sedimentary cavities between and within cell wall during plant growth (Yoon & Kim, 2008). Silica, which called silicon dioxide, compound of the two most abundant elements in Earth's crust, silicon and oxygen, SiO_2. The surface of EFB fibre acts as a main physical barrier for enzymatic hydrolysis process due to the difficulty in penetrating the surface to access the cellulose and hemicellulose for sugar production. Although the silica bodies are hard, it can be removed mechanically by several types of treatments, leaving behind empty crater, which would enhance penetration of matrix during processing of composites (Shinoj et al., 2011). This can enhance the interfacial adhesion between fibre and matrix resulting in better mechanical properties.

In the FESEM image of the untreated EFB surfaces (Figure 4.8(a)), it can be observed that the silica bodies were protected by the waxy layer and impurities resulting in smooth surface of the fibre. The removal of the impurities was expected to be happened during the treatment process, resulting in the appearance of a rough surface of the treated EFB (Figure 4.8(b)). The removal of the impurities also expected to reduce the diameter of the fibre. Theoretically, the smaller the diameter, the larger the surface area of a total fibre with the same loading of fibre and this will eventually increase the interaction between fibre and matrix (Sefadi & Luyt, 2012). In addition, the removal of the silica bodies during treatment also resulted in the presence of empty craters on the fibre surface, as can be seen in Figure 4.8(c). The open structure of the craters will benefit the adhesion between HDPE and EFB where HDPE can penetrate the structure of the fibre, thus adhesion is improved. The development of a rough surface and craters will result in a better mechanical interlocking between fibre and matrix thus improved the fibre–matrix interfacial adhesion in the composites (Gurunathan et al., 2015). Moreover, removal of the silica bodies makes the fibre surfaces rough, so the open structured of fibre due to the rough surface will increase the adhesion between HDPE and EFB.

Moisture content of EFB fibre

Moisture content of raw EFB was about 9.1-11.43% as shown in Table 4.2. Mechanical and physical properties of the composites may be affected by the moisture content of natural fibre. Higher moisture content in the composites leads to poor interfacial bonding between fibre and matrix thus lowering their mechanical properties.

Tensile strength may be with high moisture content due to the voids present in the composites (Sharba et al., 2015).

In addition, the high moisture content at 70 and 95% RH will cause part shrinkage and residual stress development during processing leads to deformation of finish parts and poor microstructural analysis. It was noticeable that plant fibres swell and shrink as they absorb and desorb moisture, respectively (Moudood et al., 2019). Moreover, composites final furnishing may have rough surface and thickness of laminate composites may not uniform, attributed to the poor adhesion of natural fibres with resin (Sharba et al., 2015; Sharba et al., 2016).

Table 4.2: Moisture content of EFB.

Sample	Weight of fibre before drying (g)	Weight of fibre after drying (g)	Moisture content (%)
1	245	217	11.43
2	450	409	9.10
3	423	376	11.11
4	460	408	11.30
5	202	179	11.39

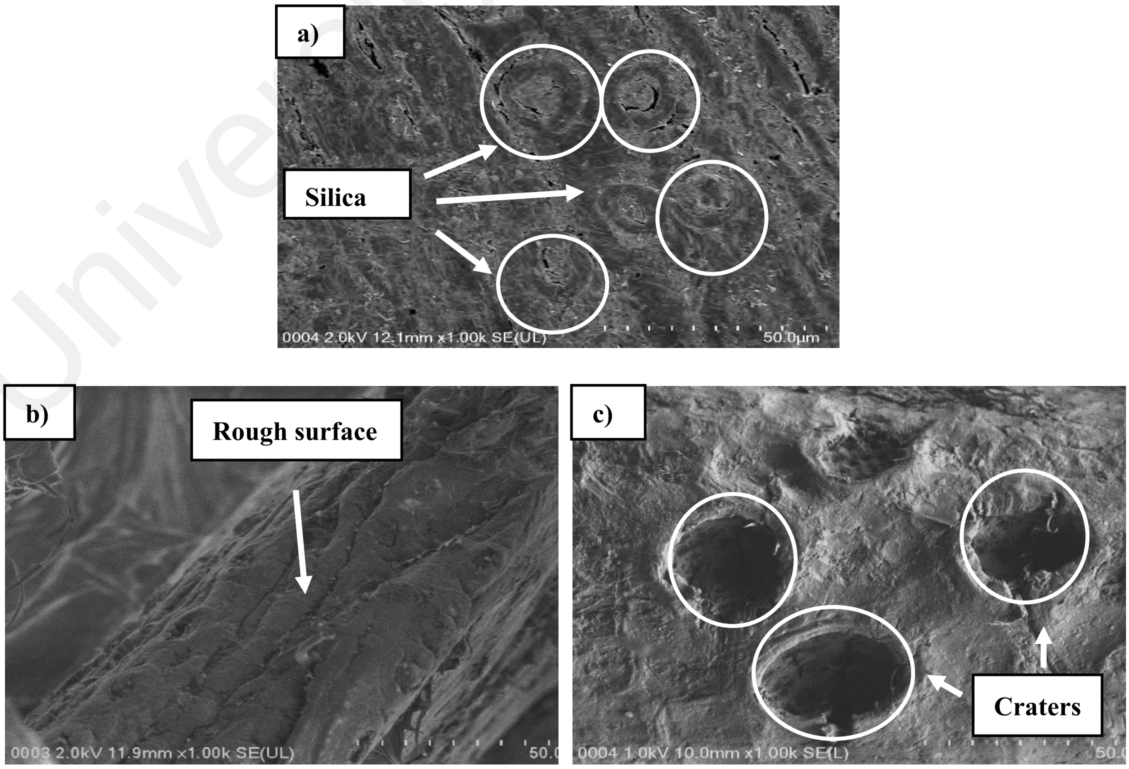


Figure 4.8: Surface morphology of (a) untreated EFB, (b) and (c) 180°C heat treatment at 1000x magnification.

Thermogravimetric analysis (TGA)

The thermal stability of the fibre was studied by the TGA technique. Understanding the thermal stability of materials is important as it allows us to determine a safe service temperature for a material itself. The samples were subjected to a temperature scan from 50 to 900°C with heating rate of 10°C/min in this study. Figure 4.9 shows the TGA scans in the form of weight change and derivative weight change versus temperature for untreated and treated EFB. There are 4 degradation transitions, A, B, C and D. The first transition, A, is related to the evaporation of water, which occurs below 100°C. The second transition B starts at about 250°C and corresponds to the polymerizations of hemicellulose, while the third transition, C, at about 350°C is due to the random cleavage of the glycosidic linkage of cellulose. The fourth transition is a slow mass loss between 400 and 600°C and is associated with the degradation of lignin (Rosa et al., 2009; Sefadi & Luyt, 2012). Table 4.3 shows the quantitative values of the onset, derivative peak temperature and the temperatures at 50% of weight loss, which are referred to as: T_{onset}, DT_P and $T_{50\%}$ respectively.

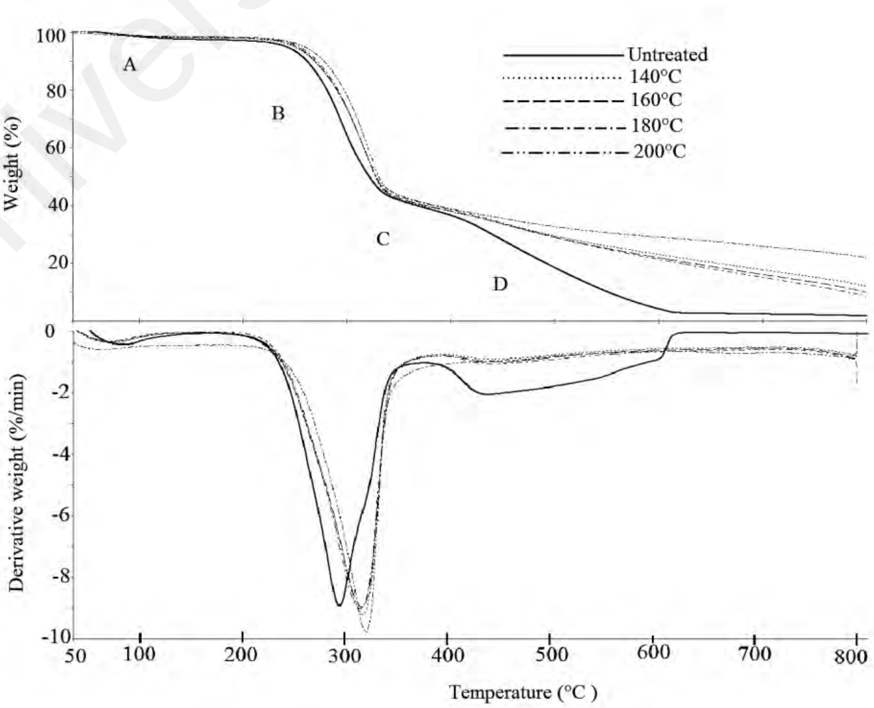


Figure 4.9: TGA thermograms of untreated and treated EFB fibre.

The information form TGA analyses of the raw materials was used to predict the possible maximum temperature range for processing the composites using twin-screw extruder. From the result, EFB degrades at about 184°C. For that reason, the maximum temperature for processing should be lower than the degradation temperature to make sure the fibre not degrade and effect the composites. This is because consideration needs to be given for the heat build-up from shear and friction during the processing. On the other hand, due to the incompatibility of fibre and matrix, EFB fibre is suggested to undergo a surface treatment. In this study, for the preliminary finding, the EFB was treated by subjecting the fibres to four different temperatures which are 140, 160, 180 and 200°C at 1 hour using vacuum oven. TGA thermograms of untreated and treated EFB were presented in Figure 4.9. The untreated and treated EFB fibres show a similar trend of decomposition.

From Table 4.3, the T_{onset} and T_{50} of EFB increased as the temperature of treatment increased. This observation shows that the thermal stability increased with the treatment of EFB. Untreated fibre degrades at lower temperature due to presence of thermally unstable fibre constituents such as hemicelluloses, holocellulose and ash, whereas the treated fibre is more stable due to the removal of these constituents during the treatment process (Khalid et al., 2008). As can be seen from the TGA curves in Figure 4.9, there are differences between the curves of untreated and heat-treated fibres, which clearly show that treated fibre had much higher thermal resistance than untreated fibres. Based on these results, the thermal stability of lignocellulose materials is suggested to be increased by the heat treatments.

Consequently, crystallinity of the EFB fibre increase after the treatment due to the removal of amorphous structure of hemicellulose and lignin (Solikhin et al., 2016). The greater crystalline structure led to a high resistance towards heat and increase in the maximum temperature for thermal degradation. From Figure 4.9 and Table 4.3, we can see that at 200°C treatment has the highest thermal stability when compared to others. In processing terms, we concerned about the relationship between treatment temperatures with the processing temperature as to determine the cause of failure. In case of thermal stability, heat treatment conducted in this study shows that 200°C treatment is possible for processing temperature, as the fibre is thermally stable. However, at 200°C, the fibres show a significantly different colour change than fibres another temperature. We suggest that may occur other degradation when fibres are treated with 200°C. Therefore, in this study we choose 180°C as the optimum temperature for fibre treatment.

Table 4.3: TGA data for untreated and treated EFB fibre.

Sample	**Decomposition temperature range (°C)**		**T_{onset} (°C)**	**DT_p (°C)**	**T_{10} (°C)**	**T_{50} (°C)**
Untreated	A	35.87 - 148.81	269.33	321.10	263.70	324.00
	B	186.55 - 399.55				
	C	399.55 - 625.98				
	D	> 625.98				
140°C	A	35.94 - 161.44	269.49	318.08	267.27	326.92
	B	174.62 - 400.47				
	C	400.47 - 699.40				
	D	> 699.40				
160°C	A	37.10-166.95	270.18	319.98	267.32	327.50
	B	178.27 - 399.40				
	C	399.40 - 699.00				
	D	> 699.00				
180°C	A	36.46 - 166.38	272.20	315.06	271.25	325.72
	B	179.52 - 396.07				
	C	396.07 - 699.40				
	D	> 699.40				
200°C	A	38.10-169.98	276.08	319.74	264.41	346.47
	B	185.26 - 403.62				
	C	403.62 - 712.86				
	D	> 712.86				

4.1.3 Tensile properties of single continuous fibre

The mechanical properties of EFB fibre in term of tensile modulus, tensile strength and tensile strain were discussed. EFB diameter was found to be in the range of 200-755 μm for both untreated and heat treated. The average tensile strength and tensile modulus of the EFB fibres with different diameter were presented in Table 4.4. The different results may be due to the pressure, which holds the fibre during the tensile test from the upper and lower gripper. The inconsistent diameter of fibre also affects the result where some of the samples did not break in the middle of fibre. The diameter of the fibre can affect the tensile properties where the tensile strength decreased when the diameter was increased as shown in Table 4.4. Based on the table below, the highest tensile strength (90.84 MPa) was found at the smallest diameter range which is 200-300 μm, while the lowest tensile strength (31.14 MPa) at fibre range of 600-700 μm. The same trend was observed with tensile modulus with tensile modulus value of 2.78 GPa at the diameter range of 200-300 μm. It was reported that smaller fibre diameters lead to better stress transfer from fibres to matrix than the similar larger fibres (Alcock et al., 2018).

Table 4.4: Average tensile strength, strain and modulus of different fibre diameter.

Diameter range (μm)	Fibre count	Tensile strength, σ (MPa)	Tensile strain, ε (%)	Tensile Modulus, E (GPa)
200-300	25	90.84	14.32	2.78
300-400	46	69.54	19.56	2.15
400-500	26	55.19	14.58	1.98
500-600	7	52.50	16.91	1.87
600-700	2	31.14	19.62	1.00
700-800	1	40.86	6.57	2.04

The fibre treatment shows slight increment in the value of tensile strength compared to the untreated fibres. This may be due to the increased in crystallinity of fibres by heat treatment. In addition, some materials i.e. lignin, wax and oils were removed. However, a reduction in tensile modulus and tensile strain were noticed as indicated in Table 3.1

previously. These were probably due to fibre surface damage which causes non-uniform micro fibrils diameter and micro-crack formation, then affects fibre deformation during tensile test (Izani et al., 2013). Consequently, the micro fibrils of the heat-treated fibre became visible which resulted in increment in the tensile strength of treated fibres.

4.2 Characterisation of composite materials

4.2.1 Morphology analysis

The fracture surface of the tensile test specimens was examined using a scanning electron microscope (Kartal et al.). Based on Figure 4.10(a) below, smooth surface was observed in the composite with 10% untreated EFB fibre while Figure 4.10 (b) shows rougher surface as 30% untreated EFB incorporated in the composites.

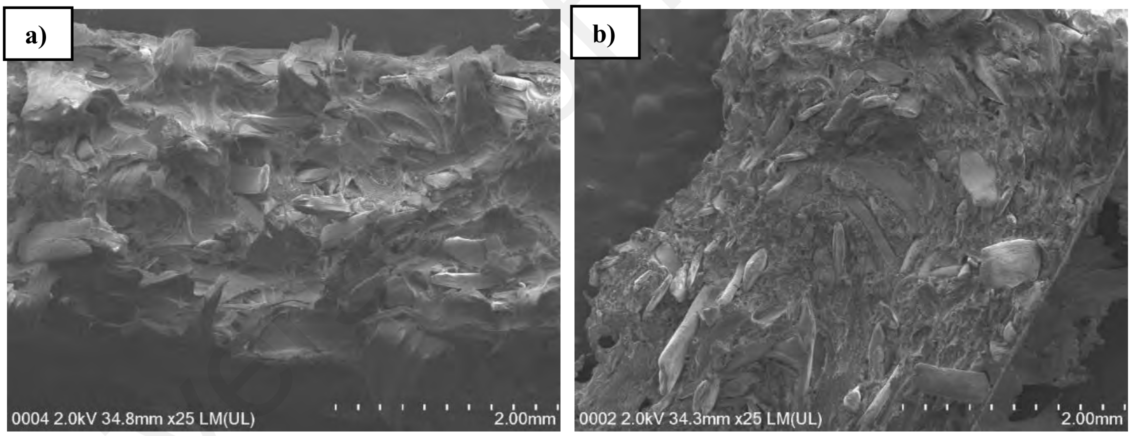


Figure 4.10: SEM micrograph of tensile fracture surface (a) composites with 10% untreated EFB, (b) composites with 30% untreated EFB.

4.2.2 Thermal characterisation

4.2.2.1 Thermogravimetric Analysis (TGA)

Effect of W_f

Figure 4.11 shows the TGA scans in the form of weight change and derivative weight change versus temperature for HDPE and untreated fibre composites. For HDPE, there is only one step of weight loss process. For HDPE/EFB composites, the curves exhibit two mass loss peaks. The first peak, at approximately 200°C to 400°C is due to the degradation of hemicellulose, cellulose, and lignin. Lignocellulosic materials, being chemically active, decompose thermo-chemically in the range of 150°C to 500°C. Hemicellulose degrades between 150°C and 350°C, cellulose decomposes between 240°C and 350°C, and lignin between 250°C and 500°C (Yang et al., 2005). The second peak, at approximately 400°C to 550°C is due to degradation of HDPE. As the filler loading increased, the thermal stability of the composites slightly decreased. These results show that the thermal stability of the composites decreased as the lignocellulosic filler content increased, which is a logical consequence of the lower thermal stability of the lignocellulosic filler compared to the pure HDPE.

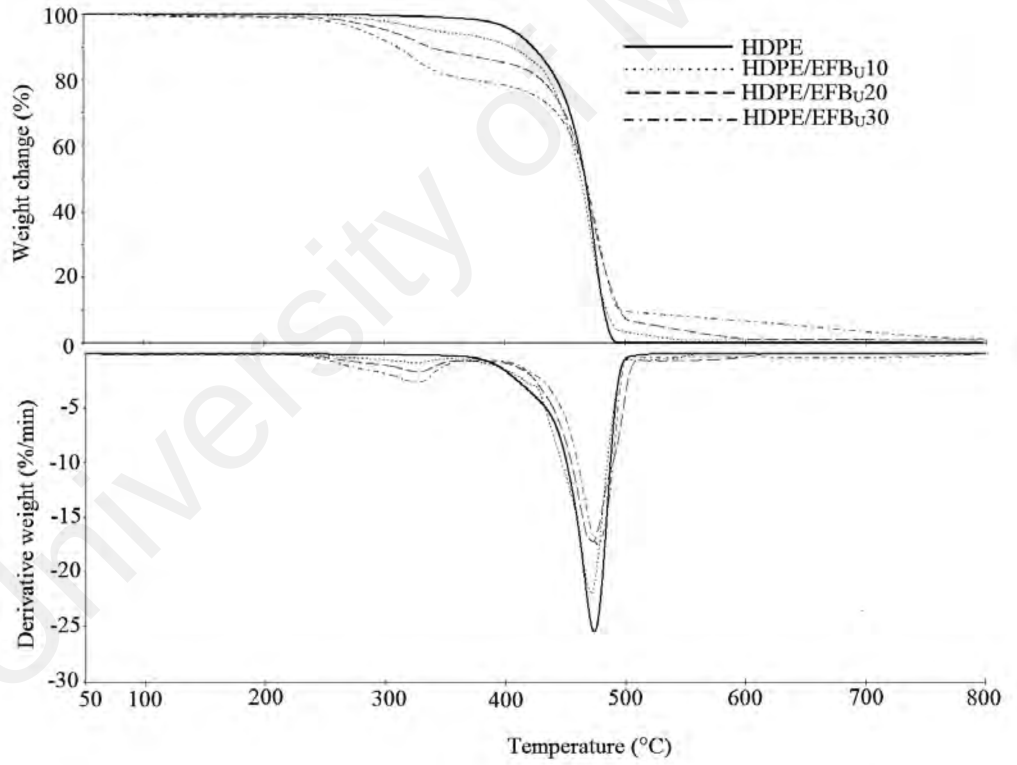


Figure 4.11: TGA thermograms of HDPE and untreated fibre composites.

Table 4.5: TGA thermogravimetric data for HDPE and all type of composites.

Sample	**Decomposition temperature range (°C)**		**T$_{onset}$ (°C)**	**T$_{50\%}$ (°C)**	**DTp (°C)**
HDPE	A	341.52 - 521.45	446.86	466.96	474.92
HDPE/EFB$_{U}$10	A	232.58 - 352.00	435.34	463.26	470.56
	B	352.00 – 567.94			
HDPE/EFB$_{U}$20	A	218.46 – 372.66	433.28	472.79	480.74
	B	372.66 – 624.17			
HDPE/EFB$_{U}$30	A	222.66 – 381.97	437.37	466.47	471.15
	B	381.97 – 764.75			
HDPE/EFB$_{180}$10	A	244.77 – 370.57	453.08	465.70	473.86
	B	370.57 – 603.08			
HDPE/EFB$_{180}$20	A	226.42 – 373.55	439.86	467.09	477.19
	B	373.55 – 603.89			
HDPE/EFB$_{180}$30	A	249.54 – 365.80	456.76	473.87	483.65
	B	365.80 – 601.49			

The thermal stability of HDPE and the various composites as measured by TGA shown in Table 4.5. The composites exhibit a slight decrease in weight in the temperature range of 100°C–150°C. This could be explained by the presence of moisture in EFB fibre released at above 100°C (Arrakhiz et al., 2013). It is also observed that the addition of fibres decreases the onset temperature of thermal degradation when compared to HDPE. This effect is suggested due to the presence of EFB fibre, which possesses lower thermal stability compared to HDPE. The TGA curves shows that as the fibre loading in the composites were increased, the overall thermal stability of composites was reduced. It can be seen from Figure 4.11 that the composites start to lose weight at lower temperatures when compared to HDPE. HDPE begins to degrade at 341.52°C while composites with 10 % EFB degrades 232.58°C. These results show that the thermal stability of the composites decreased as the EFB fibres content increased, which is a logical consequence of the lower thermal stability of the lignocellulosic filler compared to that of the polyolefin, whereas, the thermal degradation of the composite was observed above 500°C, because of the existence of ash content (Yang et al., 2005).

Moreover, the high shear and frictional forces that occur between the fibres and HDPE matrix during extrusion are the cause of breakage of the HDPE polymeric chains, resulting in a decrease in thermal stability of the composite (Beckermann & Pickering, 2008). From Table 4.5, degradation of HDPE started at a temperature greater than 300°C and degraded with much higher speed than EFB fibres. Initial degradation temperature of EFB fibre was much lower than HDPPE, but the speed was much slower. As the EFB fibre increased, composites degrade at lower temperature but at slower rate when compared to pure HDPE. This is may cause by the slow decomposition of hemicellulose, cellulose and lignin.

For treated composites, the same trend was observed where the incorporation of fibres decreases the onset temperature of thermal degradation when compared to HDPE as shown in Figure 4.12. This effect is said to be a consequence of the lower thermal stability of the lignocellulosic filler compared to the pure HDPE (Shah et al., 2017).

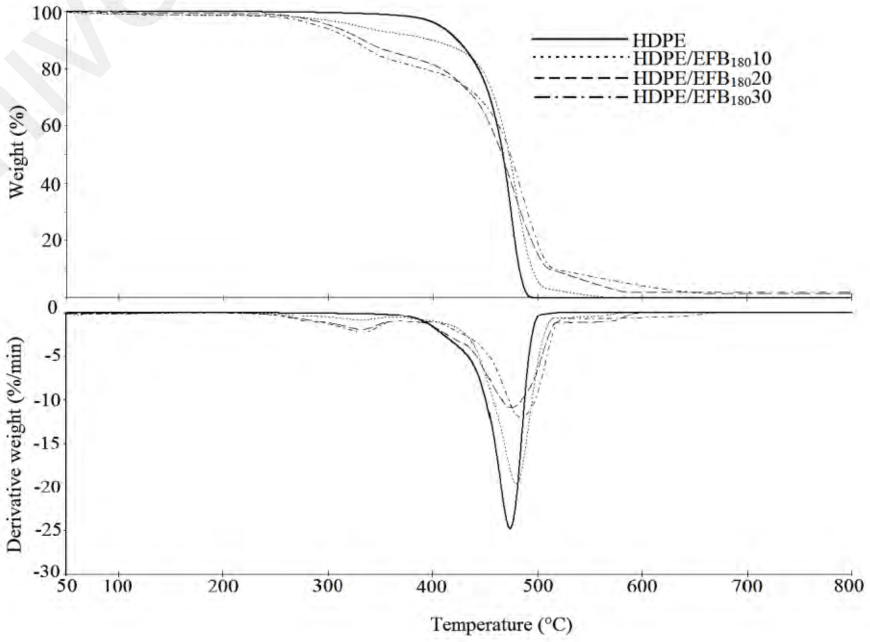


Figure 4.12: TGA thermograms of HDPE and treated fibre composites.

Effect of fibre treatment

The TGA curves in Figure 4.13 – 4.15 demonstrate that heat treatment of EFB fibre gives enhancement to the thermal stability of the material. It can be observed that the degradation takes place at higher temperature in the presence of treated fibre. As shown in Table 4.4, regardless the fibre content, the T_{onset} of treated fibre composite is higher than that of untreated fibre composite. The T_{onset} of HDPE/EFB$_{U}$10 increase from 435.34°C to 453.08°C for HDPE/EFB$_{180}$10. Similar trend was observed with DT$_{p}$ value, indicating that the most sensitive components of EFB were removed or their structures were changed as a result of heat treatment (Kaboorani, 2009). As it can be seen form Figure 4.13, the derivative curve for HDPE/EFB$_{180}$10 shifted towards higher temperature range, indicates that it is more thermally stable compared to untreated fibre composites.

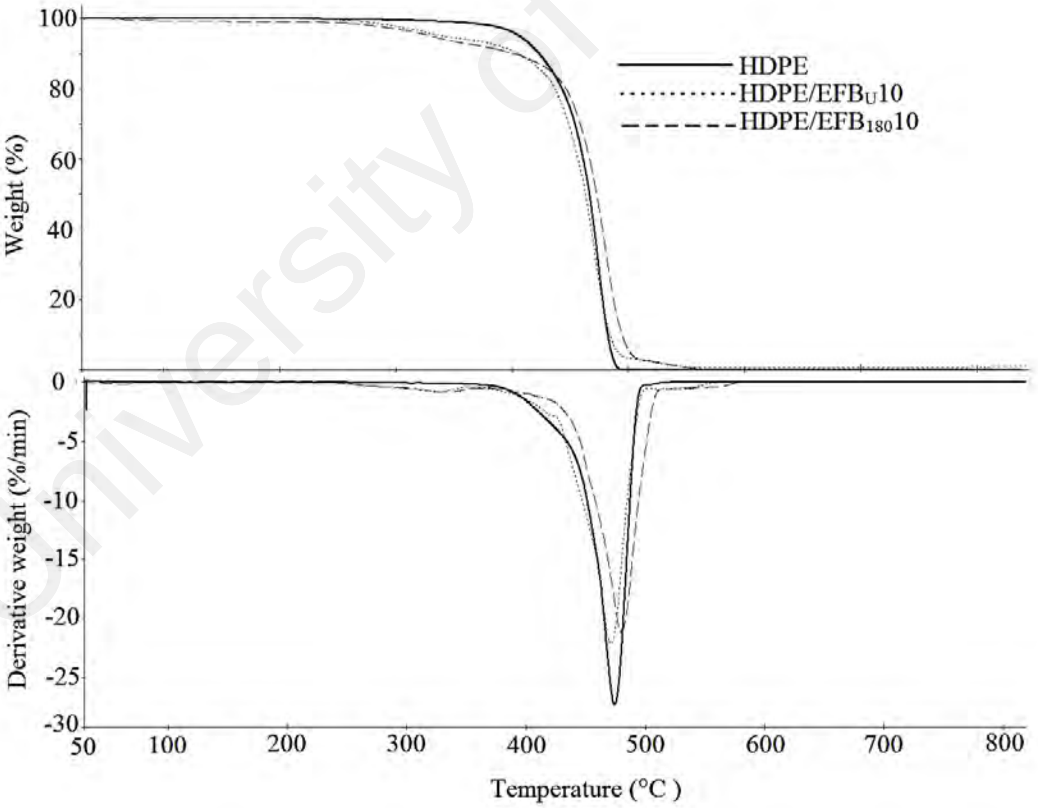


Figure 4.13: TGA thermograms of HDPE and 10% fibre composites.

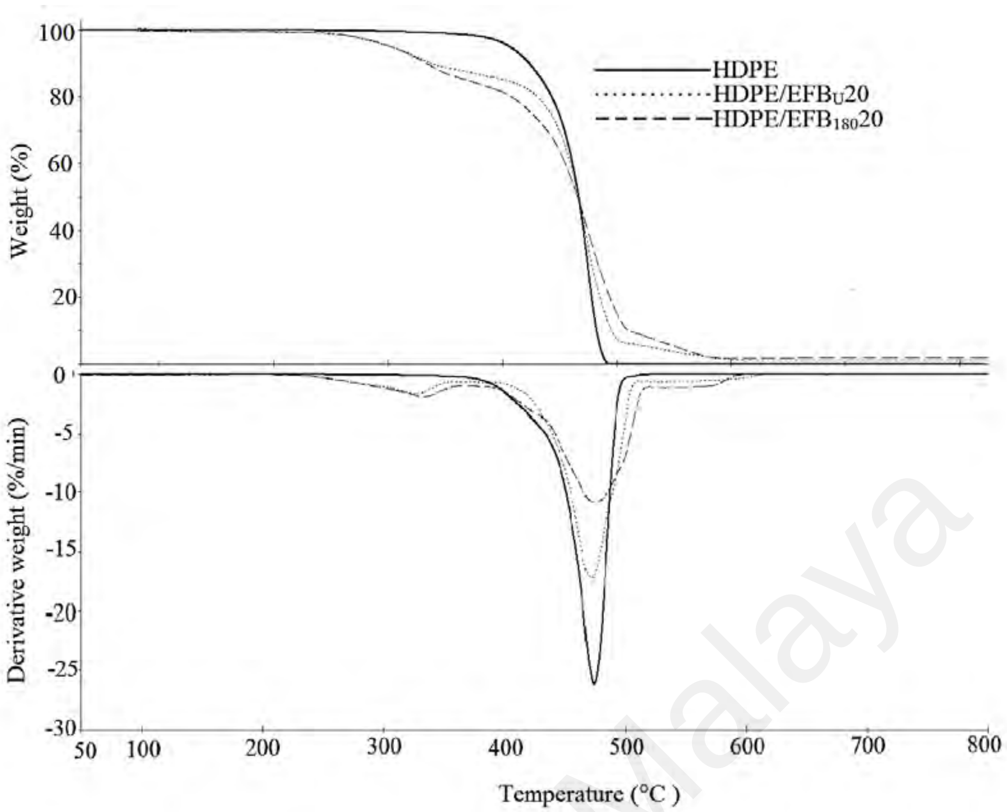


Figure 4.14: TGA thermograms of HDPE and 20% fibre composites.

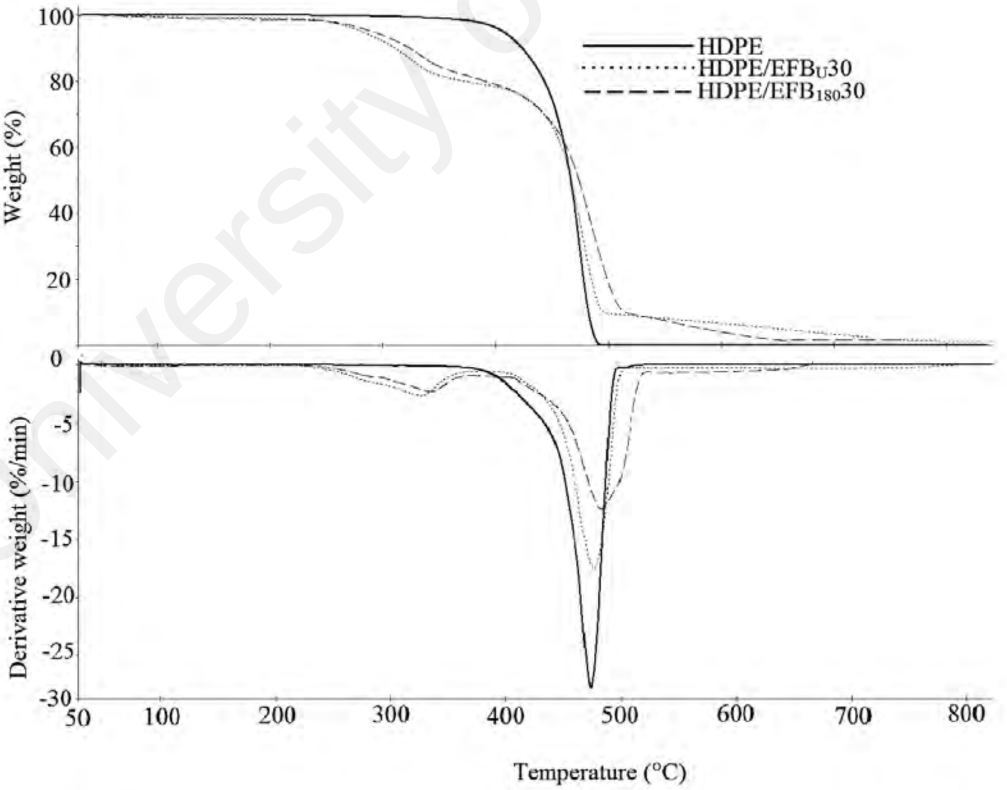


Figure 4.15: TGA thermograms of HDPE and 30% fibre composites.

Effect of conditioning

Indoor conditioning

The TGA curves of HDPE and composite conditioned under dry and indoor environment are presented in Figure 4.16 – 4.18. Table 4.6 shows the data extracted from those curves.

Table 4.6: TGA thermogravimetric data for composites under 6 months indoor exposure condition.

Sample	Decomposition temperature range (°C)	T_{onset} (°C)	$T_{50\%}$ (°C)	DT_p (°C)
HDPE	350.45 – 553.78	470.11	487.21	494.15
HDPE/EFB_U10	249.34 – 618.14	461.39	487.20	497.37
HDPE/EFB_U20	238.36 – 674.38	461.76	493.42	504.69
HDPE/EFB_U30	226.99 – 690.21	439.24	471.11	490.88
HDPE/EFB_{180}10	224.70 – 513.54	451.64	470.94	473.59
HDPE/EFB_{180}20	203.55 – 529.71	440.54	460.96	471.07
HDPE/EFB_{180}30	200.45 – 522.70	437.34	459.78	472.80

From Figure 4.16, the thermal stability of HDPE under indoor exposure shows slightly higher value of T_{onset}, at 470.11ºC, compared to 446.86ºC for dry HDPE. Same trend was observed for composites with the dry and indoor exposure. From Figure 4.17 and 4.18, it can see seen that a slight reduction of weight around 100°C for indoor exposure composites which is suggested due to water evaporation indicating that moisture from the surrounding environment have been absorbed by the composites. For untreated fibre composites, composites with highest fibre loading (30 wt.%) absorb the highest amount moisture, which is 0.963%. high fibre content will absorb moisture. With the increase of EFB fibre loading, the polar character of the fibre increases, resulting in higher water uptake (Feng et al., 2014; Kartal et al., 2013). In addition, the amount of hydrophilic –OH in the lignocellulosic fibres can also be the reason of the higher water uptake in the high fibre loading composites (Feng et al., 2014; Tajvidi & Ebrahimi, 2003).

Alternately, for treated fibre composites, the composites with lowest fibre loading absorb the highest amount of moisture, which is 2.12%. The value is however higher than untreated fibre composites, which can be concluded that at the time of exposure of the treated fibre composites, the humidity of the surrounding is may be higher than that time of exposure of the untreated fibre composites. The decomposition range of the sample under indoor exposure has no significant difference from the dry as moulded samples. From data extracted from Table 4.6, exposing the samples to indoor condition has no remarkable effects on the thermal behaviour of the composite material.

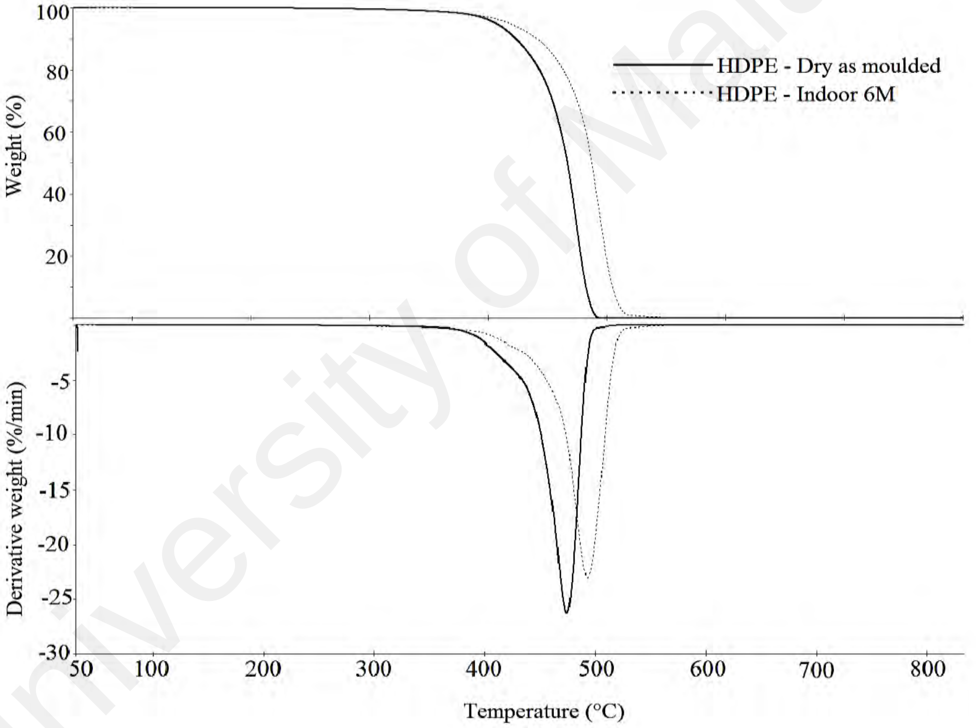


Figure 4.16: TGA thermograms of HDPE conditioned under dry and indoor environment.

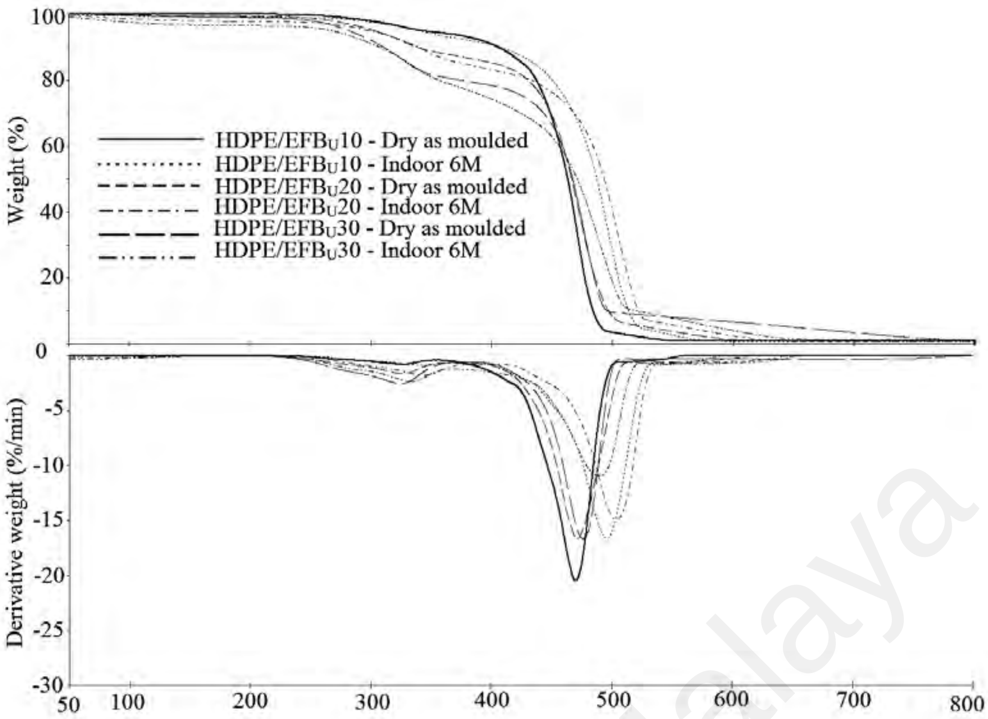


Figure 4.17: TGA thermograms of untreated fibre composites under dry and 6 months indoor exposure.

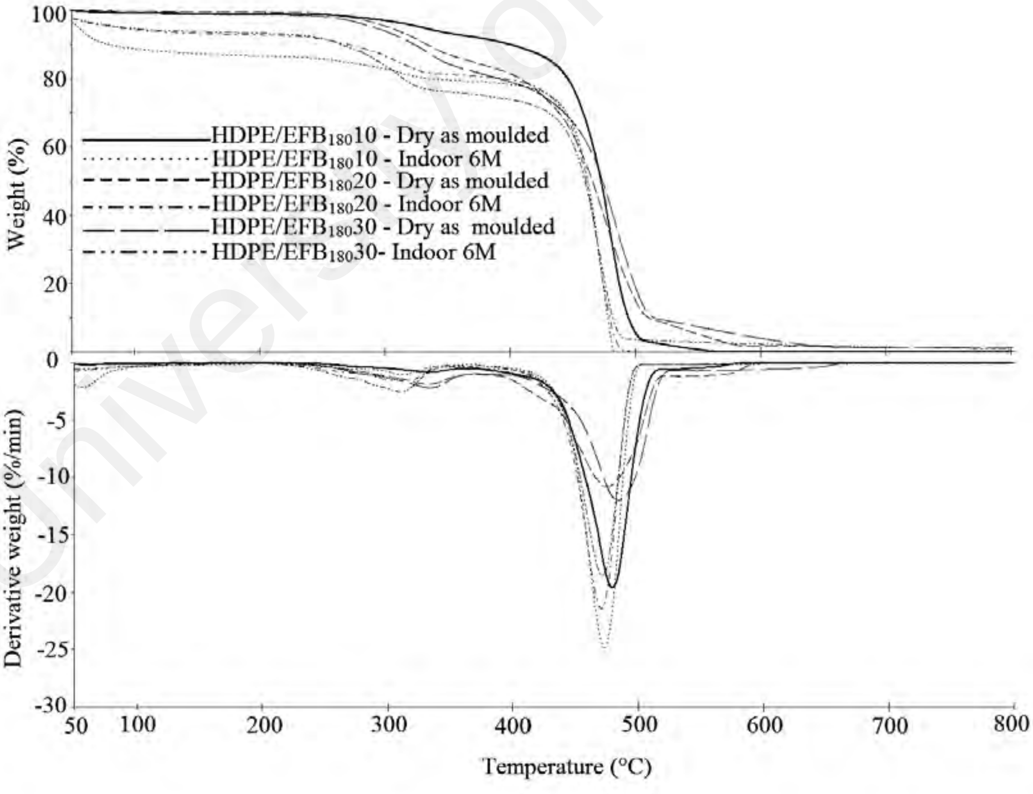


Figure 4.18: TGA thermograms of treated fibre composites under dry and 6 months indoor exposure.

Burial conditioning

Figure 4.19-4.21 presents the TGA curves of dry as moulded and soil burial HDPE and all type of composites. Data extracted from these curves are given in Table 4.7. From the table, the DT_p, T_{onset}, $T_{50\%}$ and decomposition range of the dry as moulded sample is lower that the soil burial sample for both 1 and 3 months of burial. Moreover, Figure 4.19 also shows that thermal stability of HDPE under 1 month soil burial is more stable than the dry as moulded. This result is contradicted with studies done by Mashael Alshabanat (Alshabanat, 2019), where in the studies, soil burial LDPE degrades faster than the dry as moulded samples due to the biodegradation effect. Moreover, studies done by Rida Tajau et al. (Tajau et al., 2016) also mentioned that the thermal properties of soil burial samples will decrease as they were exposed with soil microorganism in certain duration of time. However, for the composites the samples clearly degraded faster after the soil burial, probably due to deformation of the crystal structure after some microorganisms consume the composites (Alshabanat, 2019). From Figure 4.20, the soil burial composites show an increase in thermal stability relative to the dry as moulded one at all fibre loadings. For example, at 10% fibre loading the value of T_{onset} increase to 500.18°C for soil burial samples comparing to 435.34°C for the dry as moulded samples. Moreover, the DT_p and $T_{50\%}$ of the soil burial samples also shows an increment of about 29°C compared to dry as moulded. The same trend was observed with treated fibre composites where the thermal stability of the composites increases after the soil burial as shown in Figure 4.21. A peak about 100°C show the amount of water contained in the soil buried samples. In the untreated fibre composites, 0.30%, 0.455% and 0.597% water evaporated in the buried samples of 10%, 20% and 30% fibre loading. Same trend was observed with the treated fibre composites where the highest fibre loading composites absorb the highest moisture form the surrounding. As can be seen form Figure 4.21, most of the curves of buried and dry as moulded samples are overlapping each other but has a significant difference between them. The value of T_{onset}, $T_{50\%}$ and DT_p of 20% fibre loading shows a quite significant improvement where the values of T_{onset} increase from 439.86°C to 497.68°C, DT_p from 477.19°C to 503.31°C and $T_{50\%}$ from 467.09°C to 497.64°C when comparing from dry as moulded samples and soil burial respectively. No specific evidence could be given for this observation. However, the existence of fungal growth on the surface of the composites could be one of the reasons for this. In addition, the higher fibre loading should have more fungal on the composites, which might probably the reason why the thermal stability of soil burial composites is higher at higher fibre loading comparing to the dry as moulded samples.

Though heat treatment seems to affect the thermal stability of the soil buried composites, a clear trend could not be established with respect to the fibre treatment. It is expected that the treated fibre composite will have better compatibility between fibres and matrices, thus lowering the moisture intake to have better thermal stability.

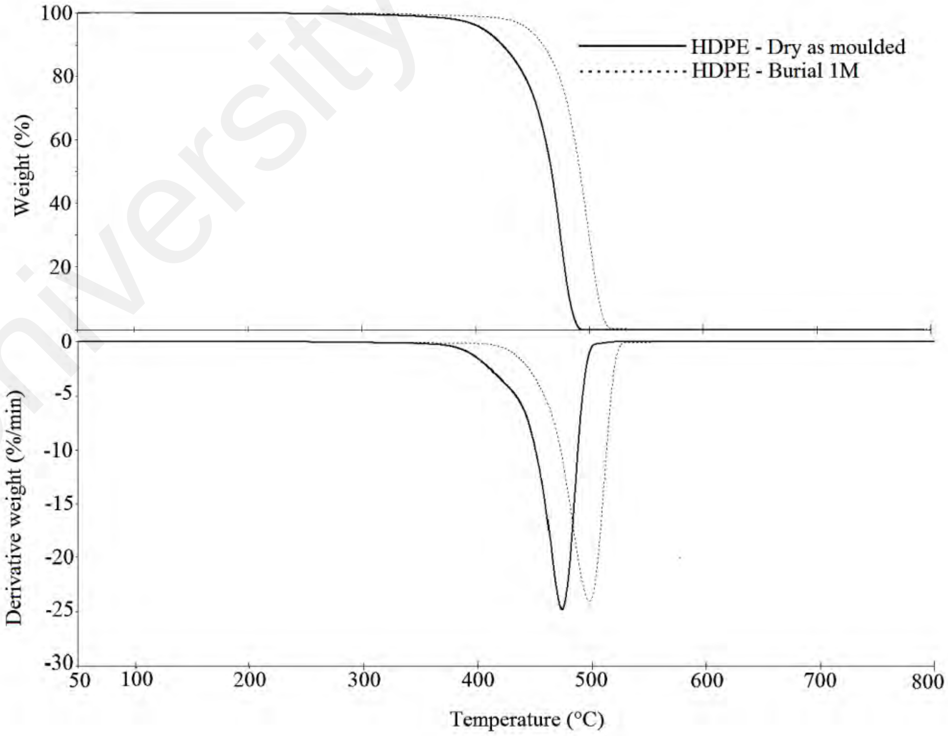


Figure 4.19: TGA thermograms of HDPE conditioned under dry and soil burial environment.

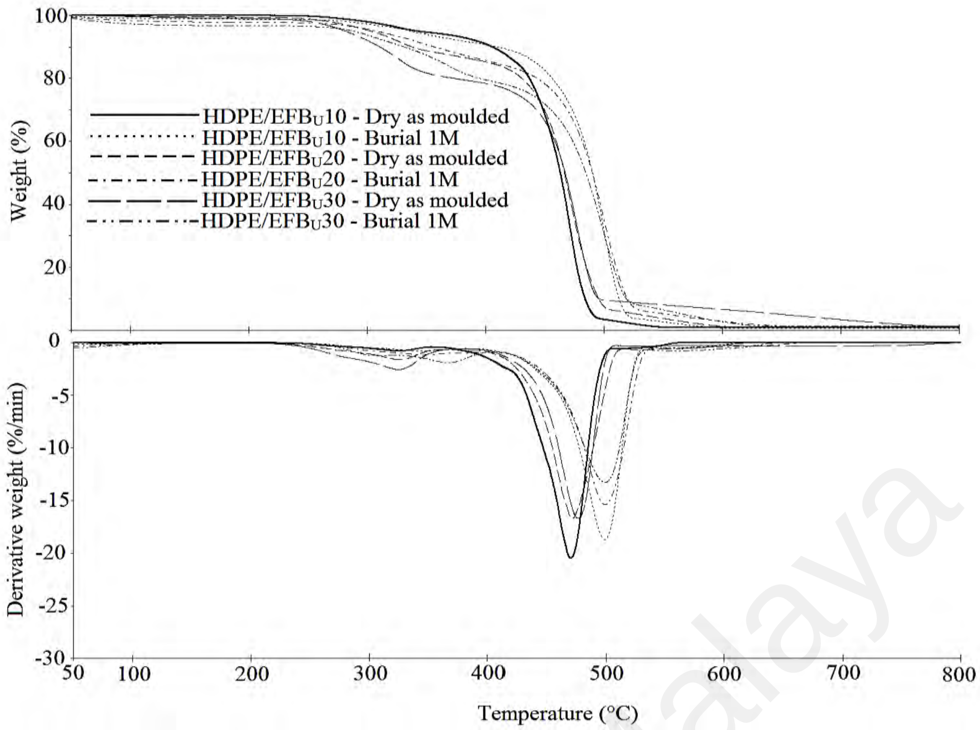


Figure 4.20: TGA thermograms of untreated fibre composites under dry and 1month soil burial exposure.

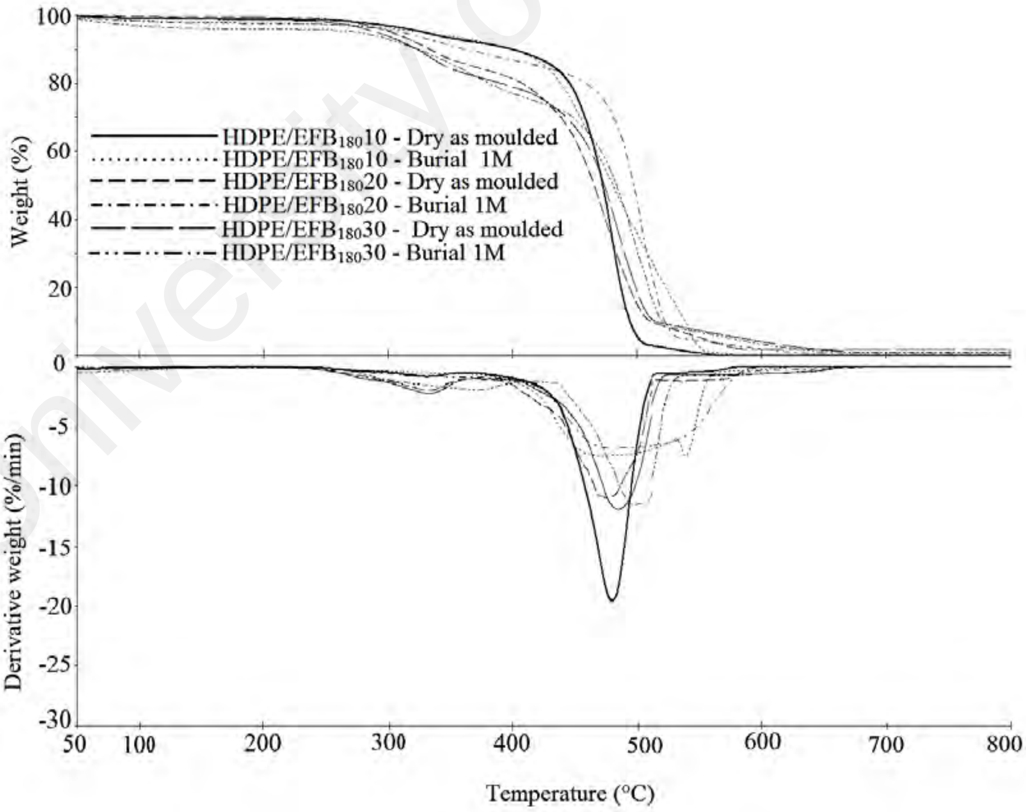


Figure 4.21: TGA thermograms of treated fibre composites under dry and 1month soil burial exposure.

Table 4.7: TGA thermogravimetric data for HDPE and composites under 1 and 3 months soil burial exposure.

Soil burial	Sample	Decomposition temperature range (°C)	T_{onset} (°C)	$T_{50\%}$ (°C)	DT_p (°C)
1 month	HDPE	382.88 – 551.81	470.38	491.49	498.12
	HDPE/EFB$_U$10	261.52 – 641.37	500.18	490.22	500.18
	HDPE/EFB$_U$20	242.50 – 677.48	458.72	490.23	500.77
	HDPE/EFB$_U$30	216.21 – 668.56	447.59	484.53	500.18
	HDPE/EFB$_{180}$10	238.86 – 649.39	467.50	487.45	491.66
	HDPE/EFB$_{180}$20	228.00 – 667.39	472.94	497.64	503.31
	HDPE/EFB$_{180}$30	211.45 – 688.67	444.44	483.38	501.50
3 months	HDPE	338.26 – 550.05	443.65	468.86	478.76
	HDPE/EFB$_U$10	238.60 – 590.62	427.96	462.56	476.91
	HDPE/EFB$_U$20	218.92 – 612.13	422.76	462.33	474.47
	HDPE/EFB$_U$30	215.12 – 615.57	424.17	462.42	471.26
	HDPE/EFB$_{180}$10	214.06 – 600.74	437.34	468.94	480.51
	HDPE/EFB$_{180}$20	208.98 – 601.19	424.11	460.43	468.35
	HDPE/EFB$_{180}$30	216.99 – 632.54	428.59	465.31	477.78

4.2.2.2 Differential Scanning Calorimetry (DSC)

DSC studies would allow greater understanding of the phase changes that the composite went through in the temperature range studied. The DSC measurement was done between the temperature ranges of 30°C to 250°C. The DSC thermograms is used to estimate the melting temperature (T_m), crystallization temperature (Arutchelvi et al., 2008) and enthalpy of melting (ΔH_m). DSC of all the samples were carried out in first heating, cooling and second heating cycle. However, only the results from the cooling and second heating are taken into consideration for discussion. This is due to the results obtained from the first heating may be influenced by the sample preparation and storage conditions. Therefore, it is more useful to study the second heating in order to obtain reproducible thermal behaviour of the samples (Özdilek et al., 2004).

The melting point of a material is defined as the temperature at which the solid phase exists in equilibrium with its liquid phase. When a material undergoes a melting phase transition, the high degree of molecular arrangement existing in the solid will be replaced by the disordered character of the liquid phase. In terms of kinetic molecular approach, the melting point represents the temperature at which the attractive forces holding the solid together are overcome by the disruptive forces of thermal motion.

Effect of total W_f

Table 4.8 presents the quantitative values of the melting temperature, crystallization temperature, enthalpy of melting, enthalpy of cooling and degree of crystallinity which are referred to as: T_m, T_c, ΔH_m, ΔH_c and X_c, respectively. Figures 4.22 and 4.23 shows the DSC thermograms for HDPE, untreated and treated composites. The degree of crystallinity (X_c) is calculated by using the following equation:

$$X_c = \frac{\Delta H_m}{\Delta H_m^*} x \, 100 \, (\%) \tag{4.1}$$

Where ΔH_m and ΔH_m^* are the enthalpies of composite specimen and fully crystalline HDPE matrix, respectively. The values in table have been normalised according to actual HDPE content in the composites. T_m and ΔH_m were obtained from the heating cycle while T_c and ΔH_c were obtained from cooling cycle.

Only one peak was observed at temperature 131°C – 135°C which is corresponds to the melting temperature of the HDPE matrix (Figure 4.22). In general, the addition of fibres did not significantly affect the melting temperature of the composite. However, the Tm of the treated composites was slightly higher of about 0.7°C to 1.9°C than the untreated composites. This is believed due to the surface modifications of fibre by heat treatment which causes an increment in the compatibility between the fibre and HDPE. The favouring interaction between the fibre and HDPE resulted in the increment of melting temperature value (Joseph et al., 2003). From Table 4.8, T_m of 10% treated fibre composites is observed at 134.7°C while for 10% untreated fibre composite, it was observed at 133.8°C. The same trend was observed with 20 and 30% fibre composites.

Table 4.8: DSC data for HDPE and all composites.

Sample	**T_m (°C)**	**T_c (°C)**	**ΔH_m (J/g)**	**ΔH_c (J/g)**	**X_c (%)**
HDPE	135.1	116.5	140.8	146.3	48.1
HDPE/EFB$_{U}$10	133.8	118.0	126.3	127.0	43.1
HDPE/EFB$_{U}$20	134.1	117.0	114.5	114.7	39.1
HDPE/EFB$_{U}$30	132.5	117.2	119.8	121.5	40.9
HDPE/EFB$_{180}$10	134.7	118.1	146.5	125.3	50.0
HDPE/EFB$_{180}$20	134.8	118.4	145.2	158.2	49.6
HDPE/EFB$_{180}$30	134.4	118.8	141.6	146.2	48.3

Figure 4.22 shows the DSC cooling curves for HDPE and all untreated composites. Based on the data tabulated in Table 4.8, there is only slight increase in the T_c with increasing of fibre content. The ΔH_c of pure HDPE obtained was -146.3 J/g. however, with the addition of EFB fibre; the magnitudes of ΔH_c of the composites became lower when compared to that of pure HDPE. The reduction of ΔH_c with addition of EFB fibre is related to the changes in the crystal structure of cooling process. However, at 20% fibre composites, the value is higher than HDPE.

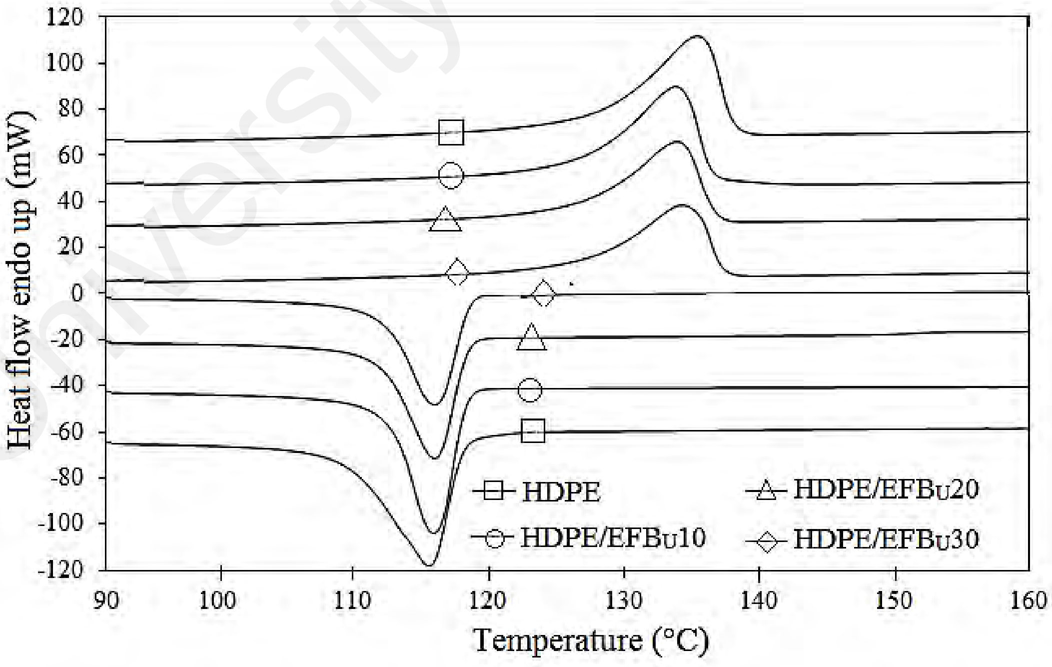


Figure 4.22: DSC curves of HDPE and untreated fibre composites (curves were shifted vertically for clarity).

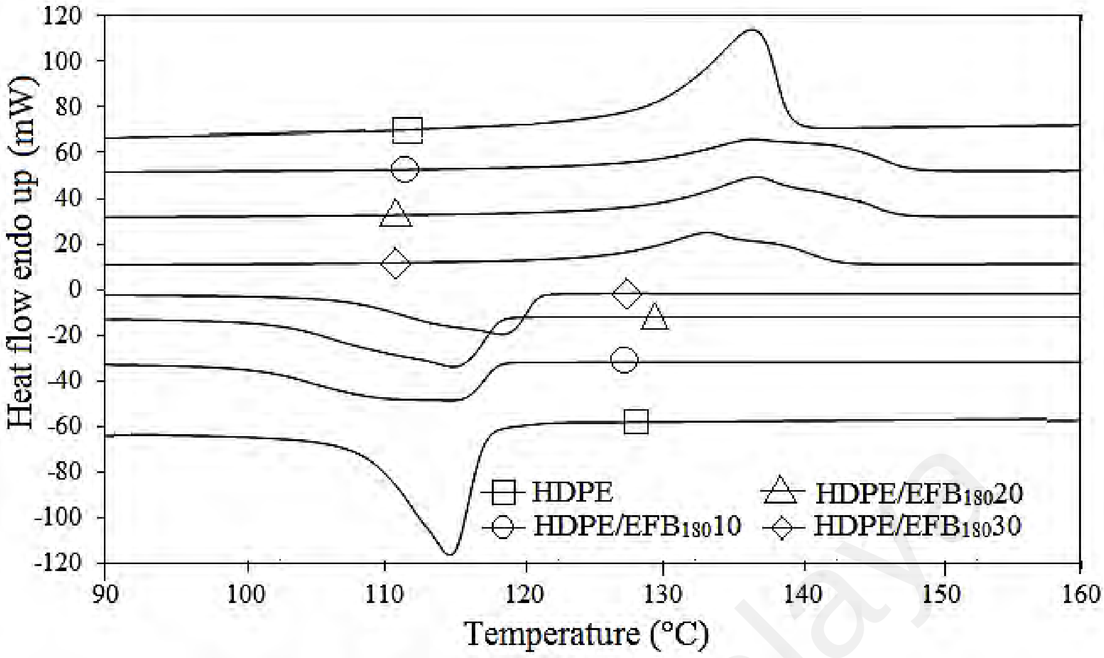


Figure 4.23: DSC curves of HDPE and treated fibre composites (curves were shifted vertically for clarity).

Incorporation of untreated EFB fibres into HDPE matrix results in a remarkable decrease in the X_c value compared to HDPE. As tabulated in Table 4.9, X_c of the untreated fibre composites decrease from 48.1% for HDPE to 43.1%, 39.1% and 40.1% for 10%, 20% and 30% EFB content, respectively. In the composites, the growth of crystallites was slow down due to compact alignment of matrix molecules network caused by natural fibres, this leads to a low mobility of polymer segments in the regions of lamellae surfaces during polymer melting process (Arrakhiz et al., 2013). The same trend observed in treated fibre composites, with slightly higher increment than untreated fibre composites. In Table 4.8, the value of 10% fibre composites is reported higher than that of 30% fibre composites. In comparison to HDPE, there were more crystallites formed in HDPE with 10% EFB fibre due to the role of natural fibres as nucleating agent, which initiate the germination and spherolites growth. Beyond this percentage, fibres became barriers which prevent the spherolites growth (Sui et al., 2009). However, there is a slight reduction at 20% and 30% compared to 10% treated fibre composites. It has been reported the crystallisation process has been slowing down due to the particles act as diluents to the matrix and restrict crystal growth at higher filler loading.

Effect of fibre treatment

Heat treatment on EFB fibre observed to have slight effect on the T_m of composites. The T_m of the treated composites was slightly higher than the untreated fibre composites. This was believed due to the surface modifications of fibre by heat treatment, which causes an increment in the compatibility between the fibre and HDPE. The favouring interaction between the treated fibre and HDPE resulted in the increment of melting temperature value (Joseph et al., 2003) compare to untreated fibre composites. Same trend was observed with T_c, where the treated fibre composites show slight increment comparing to the untreated fibre composites. On the other hand, X_c of treated fibre composites shows improvement whereby the value of $X_{c.}$ increases form 43.1% for untreated fibre composites to 50% for treated fibre composites at 10% fibre loading. Moreover, 20% treated fibre composites have value of 49.6% comparing with untreated fibre composites, which has value of 39.1% at same fibre loading. The increase in crystallinity on composites with the heat is may be due to the removal of hemicellulose and partial removal of the non-cellulosic components of EFB fibres (Sosiati & Harsojo, 2014). Moreover, based on studies by the results suggest that usage of the treated fibre blends the adhesion between the polymer and fibre is improved, hence increasing the crystallinity of composites (Bajsić et al., 2012).

Effect of conditioning

Indoor conditioning

To determine the extent of degradation, the effects of indoor environment on the properties of the composites are investigated. The cooling and second heating DSC curves of HDPE under 6 months' indoor exposure condition is presented in Figure 4.24. The data from these thermograms together with samples under 3 and 6 months' indoor condition was extracted and given in Table 4.9. The T_c values for samples under 3 and 6 months' indoor condition does not significantly change compared to dry as moulded samples. For

T_m, the value shows slight increment for 3- and 6-months indoor condition compared to dry as moulded. Besides, **Δ**H_m and **Δ**H_c show a similar trend for all fibre loadings compared to the dry as moulded specimen. On the other hand, the value of X_c for 3 and 6 months of exposure were higher than the dry as moulded specimen despite the EFB fibre loading. For example, at 30% EFB fibre loading the value of X_c increased from 41.9% to 49.9%, for dry as moulded and 3 months exposure respectively. The value has further increment to 51.2% for sample under 6 months exposure.

Table 4.9: DSC data of untreated fibre composites indoor exposed for 3 and 6 months.

Indoor Exposure	Sample	T_m (°C)	T_c (°C)	ΔH_m (J/g)	ΔH_c (J/g)	X_c (%)
3 months	HDPE	137.6	116.3	150.4	164.5	51.3
	HDPE/EFB$_U$10	135.3	118.0	149.0	137.8	50.9
	HDPE/EFB$_U$20	135.0	118.6	140.7	116.4	48.0
	HDPE/EFB$_U$30	134.6	118.8	146.2	102.8	49.9
6 months	HDPE	136.0	116.5	158.0	160.3	53.9
	HDPE/EFB$_U$10	135.3	117.8	150.8	136.4	51.5
	HDPE/EFB$_U$20	135.1	118.5	163.1	125.8	55.7
	HDPE/EFB$_U$30	135.3	118.4	150.1	105.2	51.2

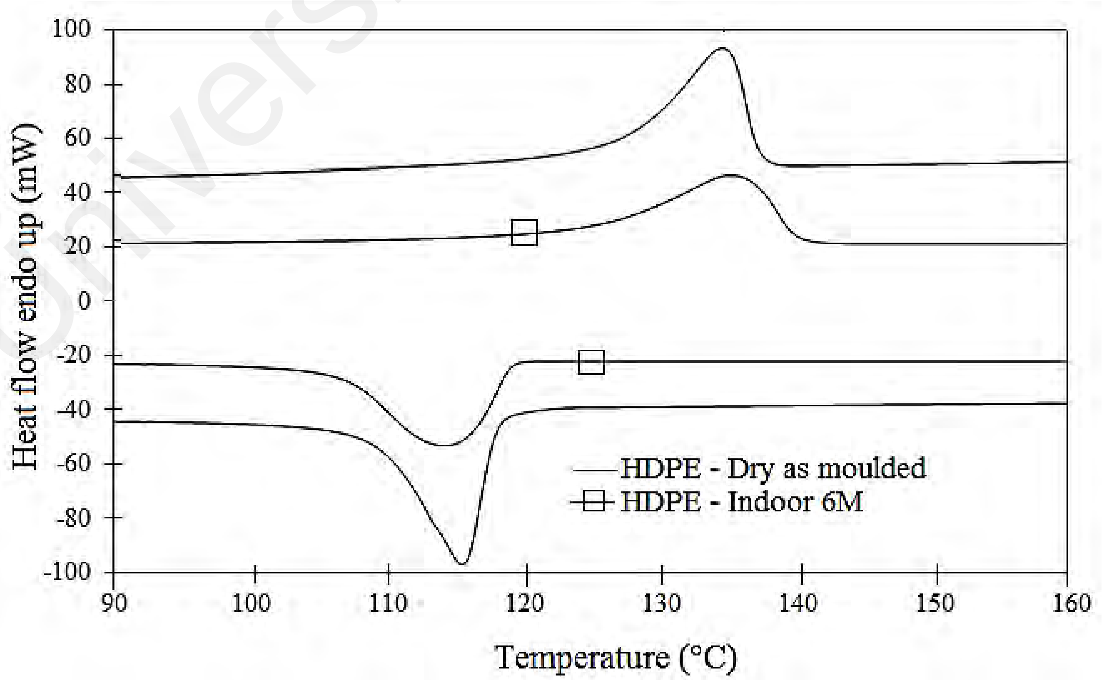


Figure 4.24: DSC curves of HDPE conditioned under dry and 6 months indoor environment (curves were shifted vertically for clarity).

Soil burial conditioning

The effects of soil burial on the properties of HDPE, untreated and treated fibre composites were investigated to understand the resistance of the composites after being buried under the soil for a period (1 and 6 months).

DSC curves of HDPE under dry and 6 months soil burial conditions are presented in Figure 4.25. For treated fibre composites, the curves are presented in Figure 4.26. The data extracted from these curves are also presented in Table 4.10. Based on the table, there was a notable change in T_m and T_c of the HDPE for both 1 month and 6 months soil burial, implying that soil burial has no observable effects on HDPE. However, the crystallinity of both untreated and treated fibre composites increases after soil burial exposure at 1 month and 6 months compared to the dry as moulded samples corresponds to a degradation of the amorphous phase in the polymer blends. This is may be due to the presence of microbes in soil will cause the amorphous phase to be consumed first, thus resulting in an increase in the portion of the crystalline phase in the remaining blends after the soil burial (Sam et al., 2011). On the other hand, the value of ΔH_m and $-\Delta H_c$ slightly increased. In addition, the T_m and T_c of all type of composites also show the same trends with HDPE, concluded that natural soil burial has no significant effect on melting and crystallisation temperature.

Table 4.10: DSC data or buried samples for 1 month and 6 months soil burial.

Soil burial	Sample	T_m (°C)	T_c (°C)	ΔH_m (J/g)	ΔH_c (J/g)	X_c (%)
1 month	HDPE	136.5	116.6	167.0	165.2	57.0
	HDPE/EFB$_U$10	135.5	117.6	120.5	117.5	41.1
	HDPE/EFB$_U$20	135.9	117.9	162.9	131.9	55.6
	HDPE/EFB$_U$30	135.5	118.1	162.5	110.9	55.5
	HDPE/EFB$_{180}$10	135.0	141.0	131.2	137.6	44.8
	HDPE/EFB$_{180}$20	135.6	117.9	166.3	133.9	56.8
	HDPE/EFB$_{180}$30	135.0	118.2	150.4	109.6	51.3
6 months	HDPE	137.8	115.8	161.5	172.3	55.1
	HDPE/EFB$_U$10	135.1	118.1	159.6	146.3	54.5
	HDPE/EFB$_U$20	135.2	118.2	149.6	120.6	51.1
	HDPE/EFB$_U$30	135.4	118.0	169.6	123.2	57.9
	HDPE/EFB$_{180}$10	135.2	118.1	154.0	139.8	52.6
	HDPE/EFB$_{180}$20	135.2	118.2	152.9	124.8	52.2
	HDPE/EFB$_{180}$30	135.3	118.1	154.1	111.0	52.6

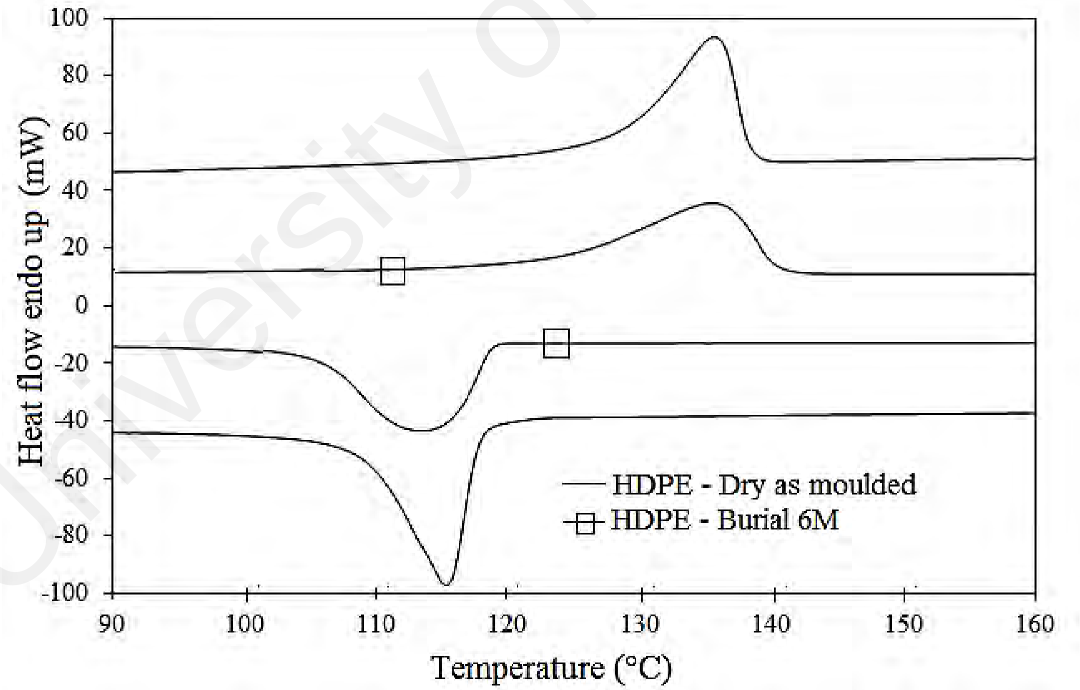


Figure 4.25: DSC curves of dry as moulded and soil burial for 6 month HDPE (curves were shifted vertically for clarity).

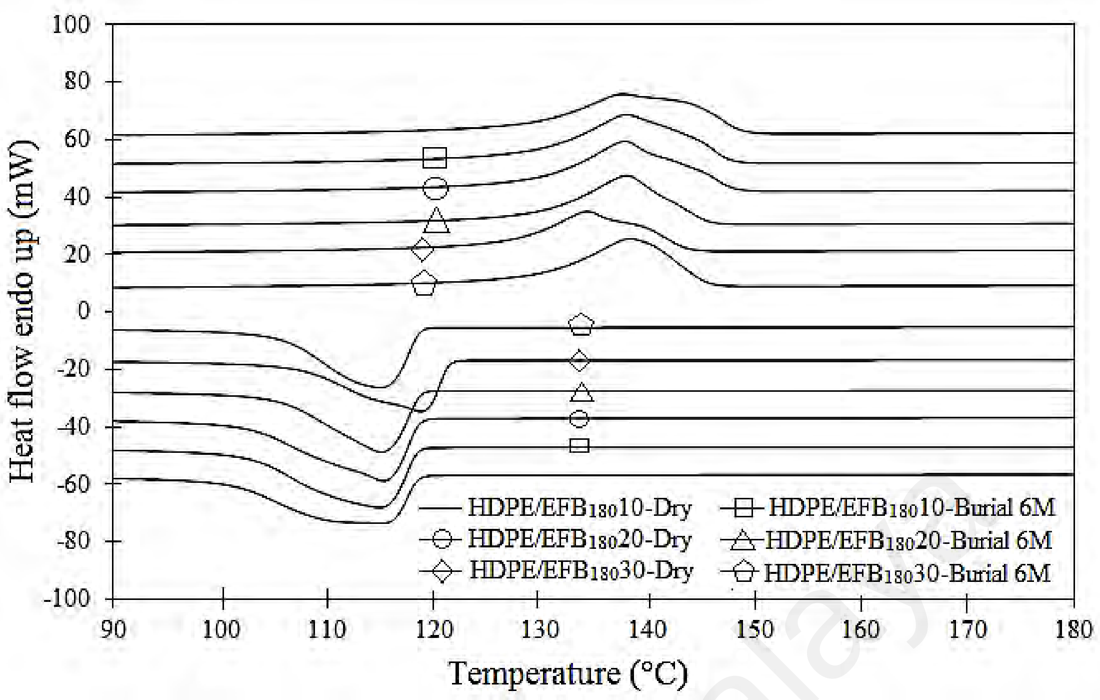


Figure 4.26: DSC curves of dry as moulded and soil burial for 6 month treated fibre composites (curves were shifted vertically for clarity).

4.2.2.3 Melt flow behaviour

The melt flow index (MFI) is a measure of the ease of flow of the melt of a thermoplastic polymer. The melt flow rate is inversely proportional to viscosity of the melt at the conditions of the test.

Table 4.11 below show the melt flow rate of HDPE and the composites. From the data extracted, the MFI of all composites reduced with the addition of fillers in the system. This trend is agreeable with findings by Eroglu (Eroglu, 2007) where the MFI values were decreased to lower value when fillers added in the PP composites. The same trend observed with treated fibre composites. This result can be related to the deformity of the EFB fibre and its lack contribution to the flow. Moreover, the presence of fibre will disturb the normal flow of the composites melt and hinder the mobility of chain segment of the flow especially at higher fibre loading. At higher filler loading, the flow of the composites was disturbed and resisted (Salmah et al., 2013). On the other hand, the MFI of treated fibre composites exhibit higher MFI compared to the untreated fibre composites. It is indicated that the heat treatment enhanced the compatibility between

EFB fibre and HDPE by having better dispersion of treated fibre in the matrix, thus, the composites flow smoothly compared to the untreated fibre (Viksne et al., 2004).

Table 4.11: Melt flow index of HDPE and the composites.

Sample	Untreated (g/10min)	Treated (g/10min)
HDPE	6.376	-
HDPE/EFB10	5.818	5.814
HDPE/EFB20	4.518	4.584
HDPE/EFB30	3.183	3.257

4.2.3 Mechanical Characterisation

4.2.3.1 Tensile properties

Mechanical properties of a composite material depend on the individual tensile properties of reinforcement fibres and the matrix. In this study, the mechanical properties of the EFB fibre and composites were determined to evaluate the average tensile strength, strain and modulus of the materials. Data are provided in Table 4.12.

Effect of total W_f

Reinforcing fibres are introduced to thermoplastics to improve their mechanical properties depends on type of applications. Thermoplastic composite has the advantage in terms of lower weight value, which is suitable for many different applications, due to high strength and relatively low processing costs when compared to traditional materials such as steel. In this study, the effect of fibre weight fraction (W_f) on tensile properties was studied. Tensile properties for all HDPE/EFB composites with different total W_f are shown in Table 4.12. Figure 4.27 – 4.29 show histogram of tensile properties of composites studied.

Table 4.12: Tensile properties for all composites.

Sample	Tensile modulus, E (GPa)	Tensile strength, σ (Singh et al.)	Tensile strain, ε (%)
Untreated EFB	2.14	76.32	17.32
Treated EFB	2.28	79.72	14.88
HDPE	1.65	21.27	9.39
HDPE/EFB$_{U}$10	0.88	19.51	11.18
HDPE/EFB$_{180}$10	0.93	19.73	11.25
HDPE/EFB$_{U}$20	1.26	19.46	7.15
HDPE/EFB$_{180}$20	1.32	19.07	8.65
HDPE/EFB$_{U}$30	1.56	19.38	5.37
HDPE/EFB$_{180}$30	1.61	18.39	5.94

Tensile modulus, E

From the Figure 4.27, tensile modulus increases steadily with increasing in fibre loading. Tensile modulus for untreated composites increases from 0.88 GPa to 1.26 GPa as the fibre loading is increased from 10% to 20%. Tensile modulus was then further increased to 1.56 GPa for composite with the fibre loading of 30%. For treated composites, tensile modulus increased from 0.93 GPa to 1.32 GPa as the fibre loading is increased from 10% to 20%. At 30% fibre loading, tensile modulus further increased to 1.61 GPa which exhibit the highest modulus among the composites. Same trend observed for both untreated and treated fibre composites.

The increment in tensile modulus values with increase in fibre loading is suggested due to higher fibre content could withstand more load and the fabricated composites became stiffer as can be seen in Figure 4.27. As can be seen in Table 4.12, the tensile modulus for untreated EFB fibre was 2.18 GPa while for HDPE was measured at 0.88 GPa. In addition, EFB fibre has higher stiffness compared to pure HDPE (El-Shekeil et al., 2014). Moreover, the incorporation of EFB fibre improved the tensile modulus of HDPE, indicating that the transferring of stress from the polymer matrix to the stiffer fibre has occurred.

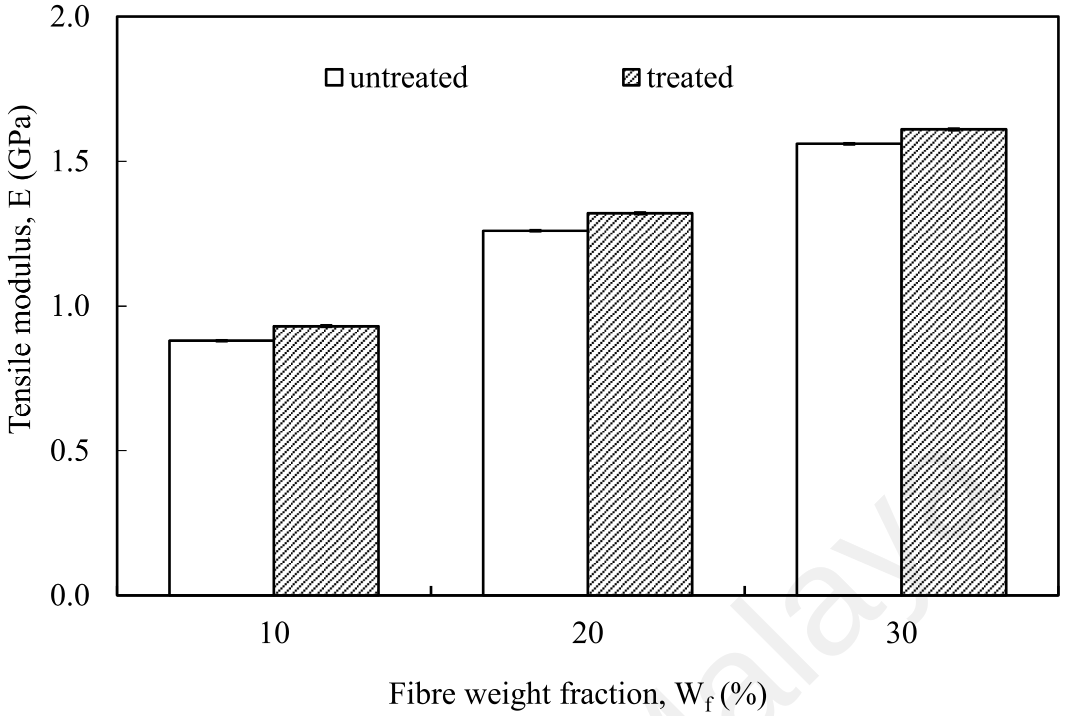


Figure 4.27: Tensile modulus of composites at different total W_f.

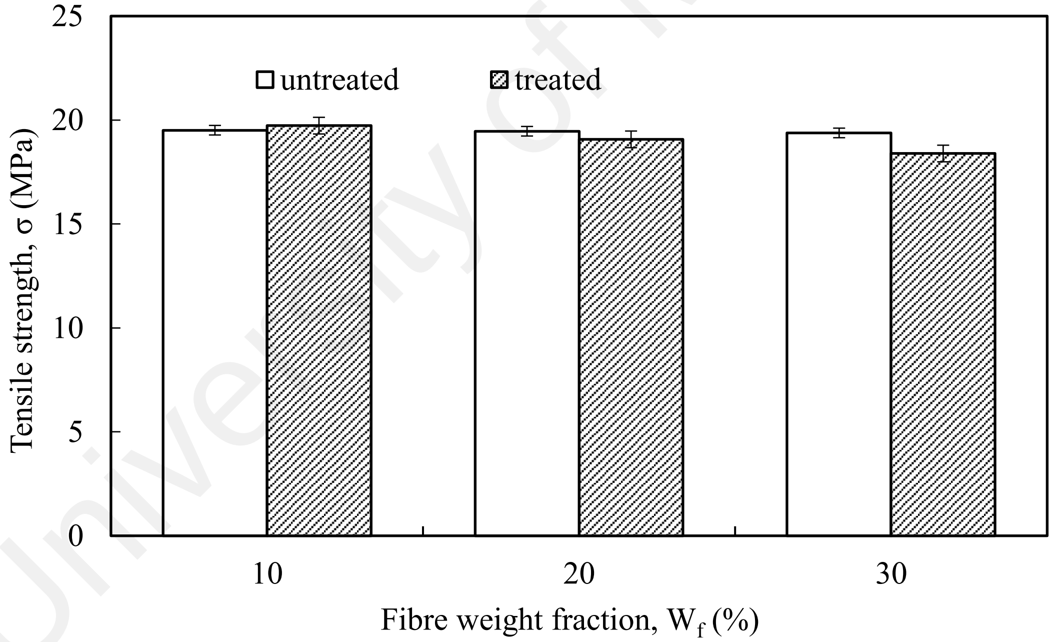


Figure 4.28: Tensile strength of composites at different total W_f.

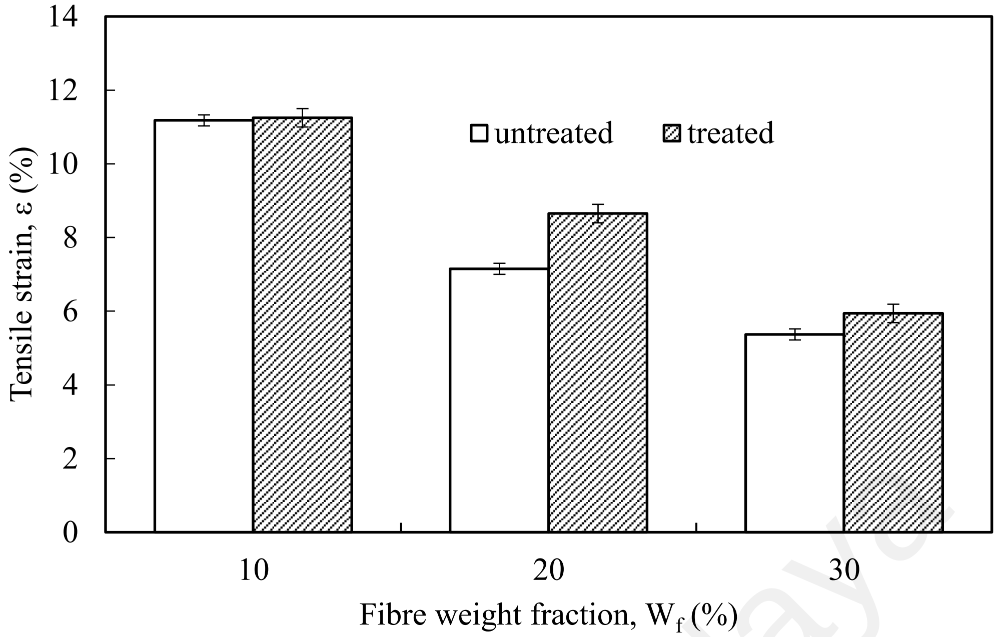


Figure 4.29: Tensile strain of composites at different W_f.

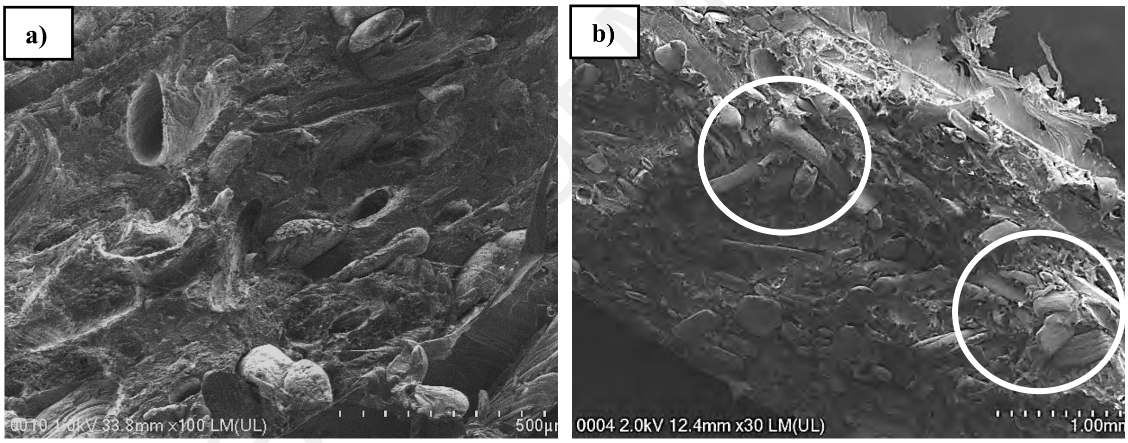


Figure 4.30: SEM micrograph of tensile fractured surface of composites containing (a) 30% untreated EFB fibres, (b) 30% treated EFB fibres

Tensile strength, σ

Generally, the tensile strength of untreated and treated fibre composites shows a slight reduction as the EFB fibre loading are increased (Figure 4.28) The tensile strength reduced from 21.27 MPa for HDPE to 19.51 MPa and 19.46 MPa for untreated composites with 10% and 20% fibre loading respectively. The composites showed further reduction to 19.38 MPa at 30% fibre loading. Same trend was observed for treated composites. Tensile strength has a slight reduction from 19.37 MPa to 19.07 MPa as EFB loading increased from 10% to 20%. The composites exhibit further decrement of tensile strength to 18.39 MPa at 30% fibre loading. This reduction in strength may indicate the possibility of the creation of more stress points in the composite as more EFB fibres are incorporated in the system. Due to the introduction of fibres, the composites become less ductile as the molecular rearrangement does not have time to take place. The notching effect of the fibres is also important in which considerable stress concentration is induced in the matrix at the fibre end and matrix flow constrained by adjacent fibres (Hassan et al., 2011). Further addition of the EFB to 20% and 30% resulted in slightly reduction of the tensile strength value, compared to the composite with 10% fibre loading. It is suggested that the higher the content of EFB fibres, the possibility of crack propagation at lower stress is higher, giving rise to a reduction in tensile strength (Sefadi & Luyt, 2012).This may be brought about by the increased incompatibility in the interfacial region between the matrix and the fibres (poor fibre-matrix adhesion) (Rozman et al., 2001). In addition, based on studies by C. Tezara et al, they found that several factors such as fibre content, water absorption in fibre, fibres forms or during fabrication may affect the tensile properties of the composites (Tezara et al., 2016). If the composites exhibit beyond the optimal weight fraction of fibres, the mechanical properties for both composites were experiencing steep decline because of poor interfacial bonding between fibres and matrix. This is because maximum fibre content will allow the fibres to fully moisten thus effect the bonding (Ishak et al., 2010).

In contrast with tensile modulus, the values taken at very low strain, which is below 0.5% compared to that fracture for tensile strength. At the lower point of strain, the void space and the interface adhesion between the fibre and matrix are not yet significant. Therefore, at this stage, the EFB fibre give their full strength to the composites before deformation starts (Hassan et al., 2011).

It is assumed that at high deformations, when there is a poor adhesion at the interface, the presence of fillers or fibres in a polymer matrix gives rise to defects at the interface, which are responsible for the strength reduction (Ishak et al., 2001). The other believable reason for the decreasing in tensile strength of composites maybe due to fibre orientation during the preparation of composite whereby it was impossible to align the fibres in a straight manner (Singh et al., 2014a).

Tensile strain, ε

Generally, tensile strain would decrease with increasing fibre content. Figure 4.29 shows the tensile strain of HDPE and its composites. Overall, as the fibre loading increase in the composites, the value of tensile strain decreases. For untreated composites, the tensile strain is reduced from 11.18% to 7.15% and 5.37% as the EFB content increased from 10% to 20% and 30%, respectively. Same trend observed in treated composites in which the tensile strain reduced from 11.25% to 8.65% and 5.94% as the fibre content increase from 10%, 20% and 30% respectively. The reduction in the tensile strain is expected as it indicates the increment in the stiffness of composites is high due to incorporation of stiffer materials, which is EFB fibre, that has lower elongation compared to HDPE. Stiffer materials would resist any deformation to its shape. In composites where content of fibre is low, failure of the fibres does not result in failure of the composite, as the matrix alone is capable of supporting the imposed stress. For composites with higher fibre content, the fibre content would cause insufficient wetting of fibre by matrix, increase the restriction on matrix by matrix thus limiting the chain mobility (Ismail & Ishak, 2018).

Effect of fibre treatment

The effect of heat treatment on tensile modulus, tensile strength and tensile strain for all type of composites were shown in Figure 4.27-4.29.

Tensile modulus, E

From Figure 4.27, it can be seen that at all fibre loading, tensile modulus value was increase after the fibre treatment. Tensile modulus increases from 0.88 GPa to 0.93 GPa for 10% EFB loading, 1.26 GPa to 1.32 GPa and 1.56 GPa to 1.61 GPa for 20% and 30% fibre loading respectively. This could be attributed the removal of moisture, oil, impurities and wax on fibre surface thus enhancing the interfacial adhesion between EFB and HDPE (Shahzad, 2011). However, since tensile modulus were calculated at 0.5%, the effect of rearrangement may not yet take place.

Tensile strength, σ

There is a slight increase for tensile strength at 10% EFB loading after treatment (Figure 4.28). Tensile strength increases from 19.51 MPa to 19.73 MPa. However, for 20% and 30% fibre loading, tensile strength decreased from 19.46 MPa to 19.07 MPa and 19.38 MPa to 18.39 MPa, respectively. The same trend was observed with the untreated fibres, which the value of tensile strength decreases with increasing of the fibre loading. This may be due to wetting problems, in which at high fibre loading as reported earlier with the untreated composites as can be seen in Figure 4.30.

Tensile strain, ε

Tensile strain increased slightly at all fibre loading after treatment. At 10% fibre loading, tensile strain increased from 11.18% to 11.25%. This trend followed by 20% and 30%, in which the tensile strain increased from 7.15% to 8.85% and 5.37% to 5.94% respectively. This possibly due to the improved interfacial adhesion between fibre and matrix.

Effect of conditioning

Indoor conditioning

The mechanical properties of the composites exposed under indoor condition were tested to investigate the response of both untreated and treated composites to ambient conditions. The tensile modulus, tensile strength and tensile strain of the composites are provided in Figure 4.31 - Figure 4.36.

Tensile modulus, E

The tensile modulus of HDPE under dry and indoor conditions were recorded at 1.65 GPa and 1.72 GPa, respectively. There is only a slight change observed in the tensile modulus of these two samples. (Figure 4.31). This result suggested that indoor exposure has no detrimental effect on the tensile modulus of the HDPE matrix. However, the value of tensile modulus of untreated fibre composites observed the highest at 1 month of indoor exposure despite the fibre loading (Figure 4.31). This result may be due to the high moisture content in the specimen after being exposed for the first 1 month (30 days) of absorption of moisture of the composites. This was suggested the absorption of moisture may filled up the gaps between fibre and matrix during manufacturing process because of shrinkage of the matrix, thus leads in the better adhesion and eventually increase the tensile properties (Munoz & García-Manrique, 2015). For the treated fibre composites, the tensile modulus has slight decrement with the time of exposure.

Tensile strength, σ

The tensile strength of composites under dry as moulded and indoor condition are shown. In Figure 4.33, pure HDPE shows insignificant change in the tensile strength with respect of different indoor condition exposure. Values of 21.27 MPa and 21.8 MPa are obtained for sample of dry as moulded and indoor condition, respectively. Similar trend has been observed for the untreated and treated fibre composites (Figure 4.33 and 4.34). This can be concluded that indoor environment is not destructive to the composites.

Tensile strain, ε

Figure 4.35 and 4.36 presents the tensile strain values for dry as moulded and indoor condition sample for untreated and treated fibre composites, respectively. The HDPE under dry condition has a tensile strain of 9.23% while the values has a slight increment to 10.32% for sample undergo 1 month of indoor exposure. It can be due to the water absorption causing the plasticization of HDPE (Munoz & García-Manrique, 2015). However, the value of tensile strain decreases to 7.15% and 5.37% for 3 and 6 months of exposure. This may be due to the further degradation of HDPE consequence of large drop in the molecular weight after certain exposure time. (Sharkh & Hamid, 2004). The value of tensile strain in untreated fibre composites generally observed to be increased with increasing time of exposure (Figure 4.35). In addition, the treated fibre composites also show decreasing value of tensile strain as the exposure time is increased but with higher value compared to untreated fibre composites (Figure 4.36). It is suggested that, the different break mechanism and the possible enhanced adhesion can be the main cause of the near constant strain at break of the composite samples, thus reducing the strain value (Sharkh & Hamid, 2004).

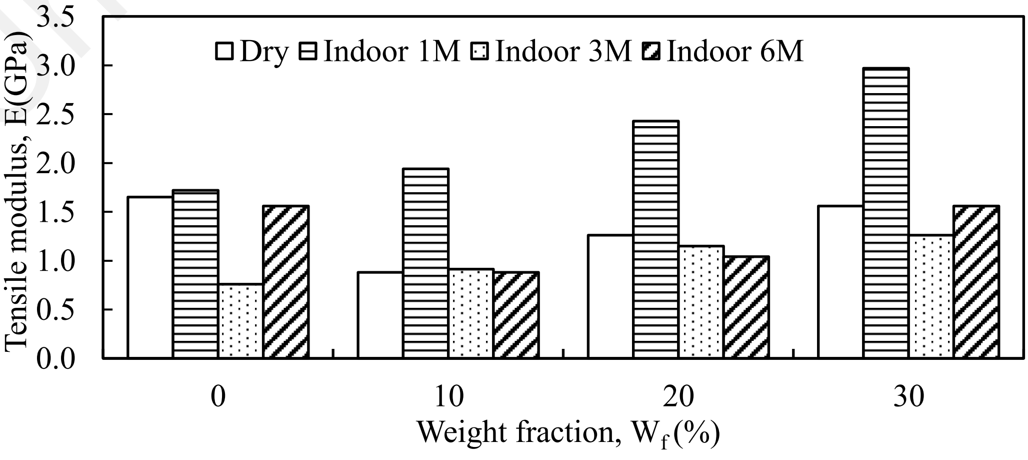


Figure 4.31: Tensile modulus of untreated fibre composite under dry and indoor exposure at different time.

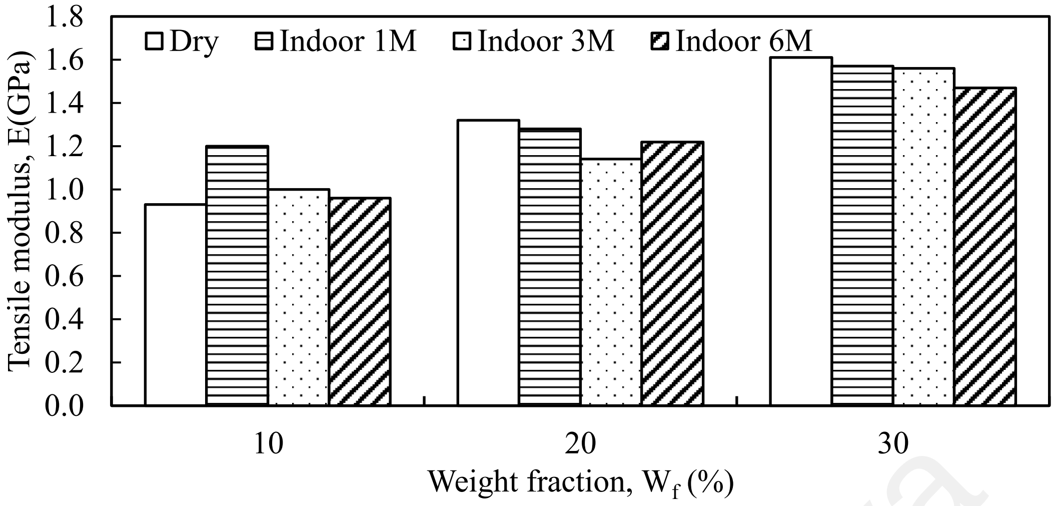


Figure 4.32: Tensile modulus of treated fibre composite under dry and indoor exposure at different time.

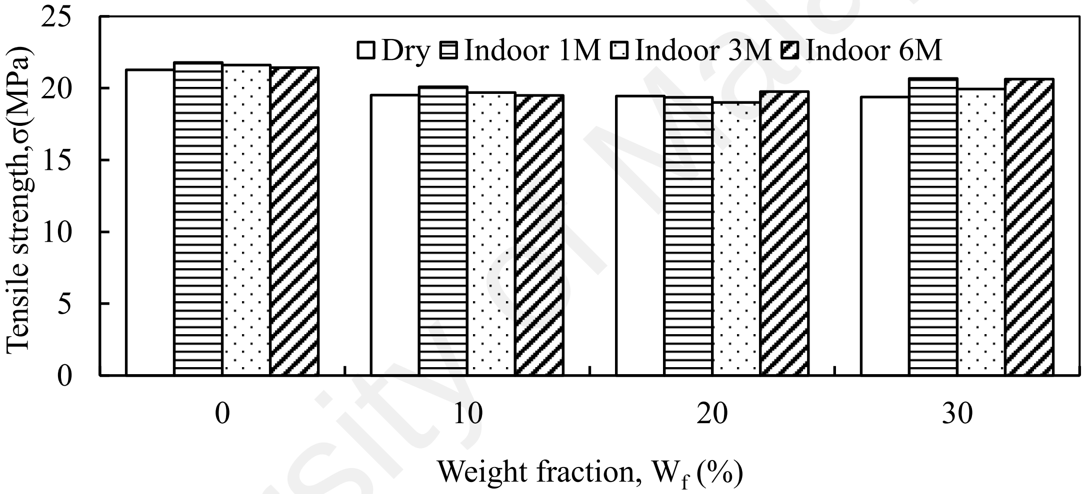


Figure 4.33: Tensile strength of untreated fibre composite under dry and indoor exposure at different time.

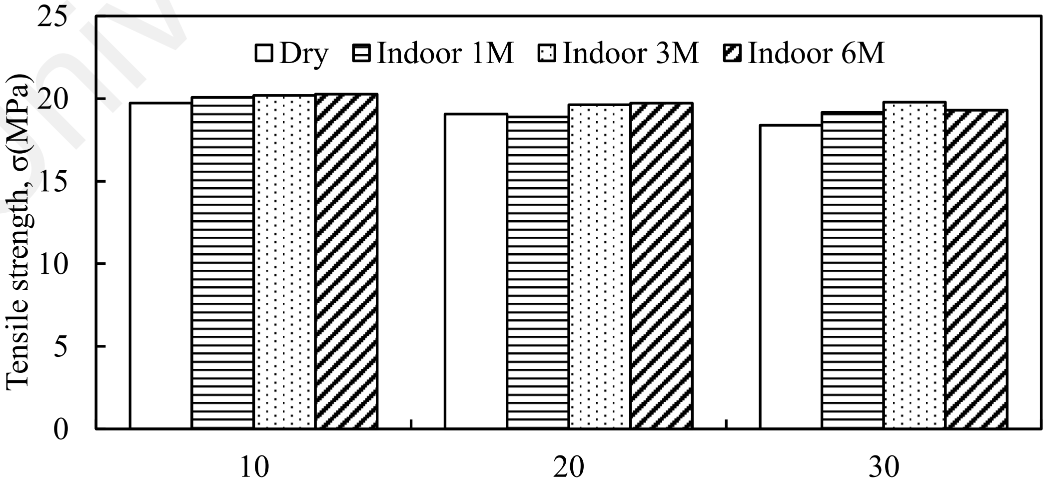


Figure 4.34: Tensile strength of treated fibre composite under dry and indoor exposure at different time.

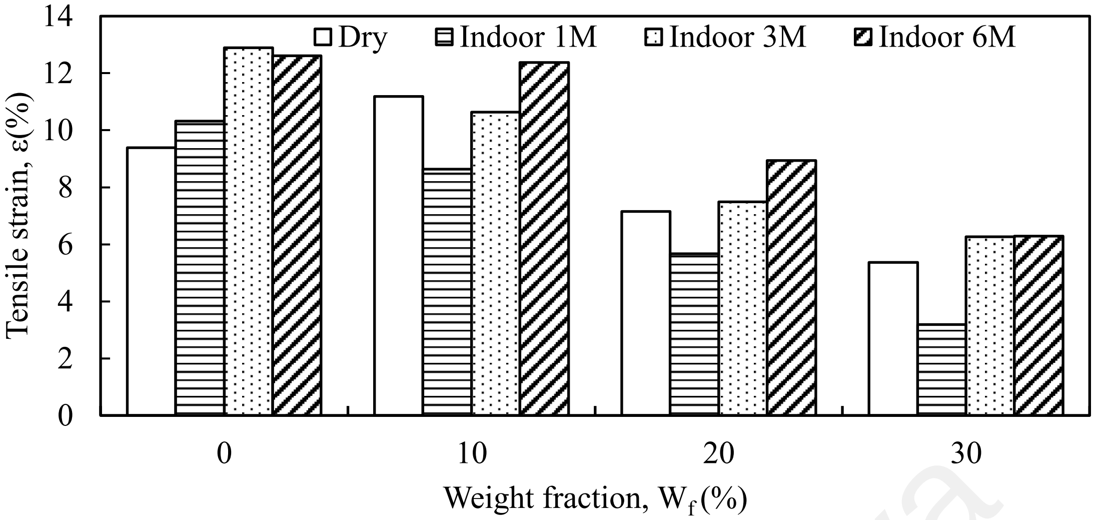


Figure 4.35: Tensile strain of untreated fibre composite under dry and indoor exposure at different time.

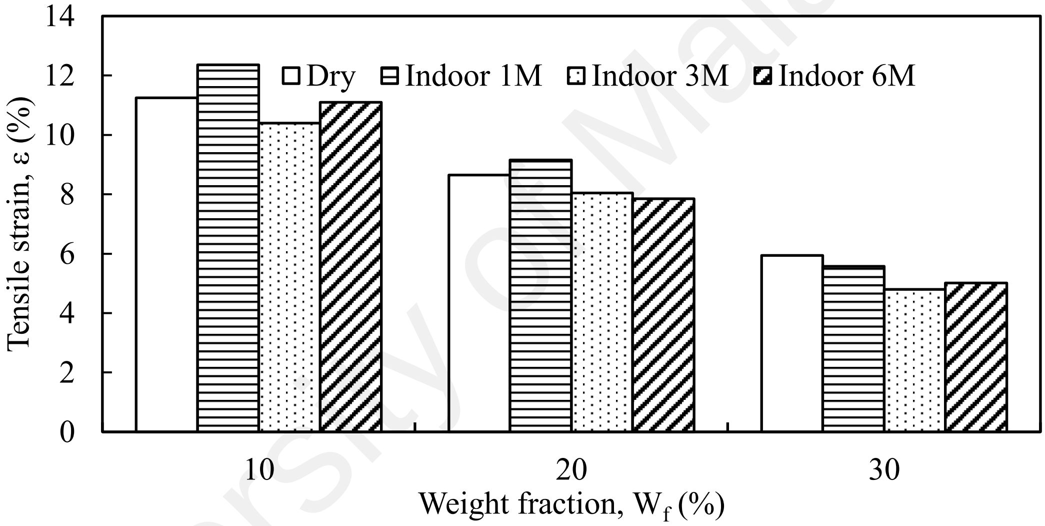


Figure 4.36: Tensile strain of treated fibre composite under dry and indoor exposure at different time.

Soil burial conditioning

The mechanical properties of the composites subjected to soil burial condition were tested to investigate the response of both untreated and treated composites when in contact with soil in the open environment. This study is also to observe the possible deterioration of the materials if any. The tensile modulus, tensile strength and tensile strain of the composites are plotted in Figure 4.38 – 4.43.

Tensile modulus, E

The tensile modulus of untreated fibre composites for dry and buried samples are shown in Figure 4.38. Tensile modulus of HDPE was dramatically dropped at 1 month of burial compared to dry samples. This result may be due to the high moisture content in the specimen after being buried for the first 1 month (30 days) influenced the modulus of HDPE (Rashdi et al., 2010). In addition, the tensile modulus of HDPE keeps reducing at 3 and 6 months. This are may be due to biodegradation of HDPE due to exposure of bacteria and fungi in soil (Arutchelvi et al., 2008). There is no specific trend observed in the untreated fibre composites but overall, the tensile modulus decreased with the increasing duration of soil burial. For example, at 30% fibre loading, the value of tensile modulus decreased from 1.56 GPa for dry to 1.19 GPa after 1 month of burial. The value has further reduction to 1.03 GPa and 0.77 GPa at 3 and 6 months soil burial, respectively. This can be explained by the fungal activities as we can see the fungal growth on the surface of the composites in Figure 4.37. The microorganisms may attack the composites, increased the porosity, thus facilitating the access of free oxygen, enzymes and free radicals, leading to promote further degradation. This will reduce the stiffness of composites, leading to lower tensile modulus (Ołdak et al., 2005). However, at 10% fibre content, the tensile modulus increases after the exposure to soil burial. This is may be due to good adhesion between HDPE and EFB at 10 % fibre loading, thus acts could act as a protection covering to the soil microorganisms attack (Obasi et al., 2013).

The same trend observed with treated fibre composites whereby the tensile modulus decreased as the time of soil burial increased (Figure 4.39) However, the value of treated fibre composites slightly higher compared to untreated fibre composites. This is due to the removal of impurities during the fibre treatment, lowering the possibilities of high moisture uptake and fungal growth.

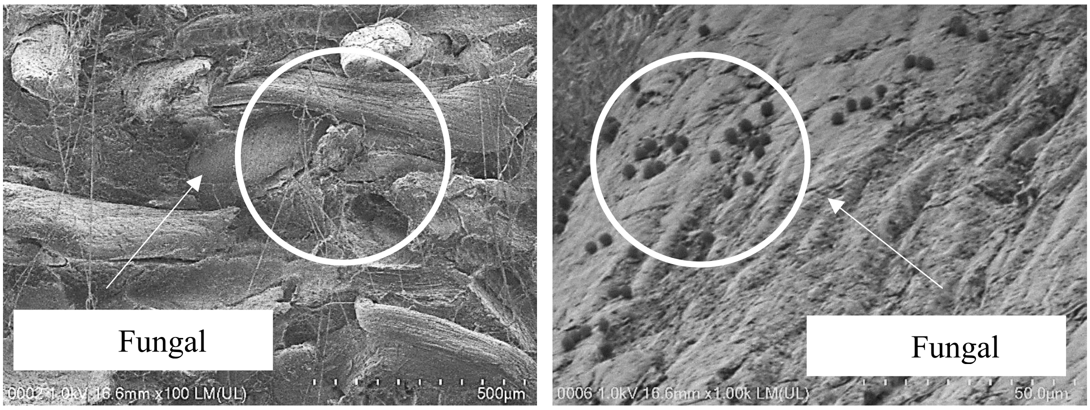


Figure 4.37: SEM micrographs for tensile fracture of soil buried 30% untreated EFB composites (6 months).

Tensile strength, σ

Generally, the tensile strength for the composites after soil burial exposure was decreased with increasing time burial. However, the tensile strength for pure HDPE has a slight increment but it is said to be insignificant. The tensile strength for untreated fibre composites has a decrement when compared to dry samples due to moisture intake (Rashdi et al., 2010) (Figure 4.40). Moreover, the decrease in value of tensile strength is due to the presence of EFB fibre, which is a lignocellulosic material, thus absorb more water easily and tends to degrade faster (Abdul Khalil et al., 2010). The flow of water molecules into the interfaces between fibre and matrix also initiates the debonding process of the samples. In addition, high fibre content in the composites may leads to more water penetration into the interface through the micro cracks induced by swelling of fibres creating swelling stresses thus making the composites to fail (Alomayri et al., 2014). As can be seen in 30% untreated fibre composites, the value of tensile strength is almost the same for 3 and 6 months which is 14.11 MPa and 14.01 MPa respectively.

Therefore, it is possible that the duration of exposure (6 months) of this specimen is not sufficient to cause more decline in the value of tensile strength. The trend observed with the treated fibre composites but with higher value comparing to the untreated fibre composites which is 14.74 MPa and 14.23 MPa for 3 and 6 months soil burial respectively

(Figure 4.41). The same reason explained before, this may be due to removal of impurities during the fibre treatment, lowering the possibilities of high moisture uptake.

Tensile strain, ε

The tensile strain of the buried samples is generally higher than the dry as moulded for both untreated and treated fibre composites (Figure 4.42 and 4.43). This may be a result of the exposure to the degradative elements in the soil. It can be due to the water absorption during the soil exposure causing the plasticization of the composite samples (Munoz & García-Manrique, 2015).

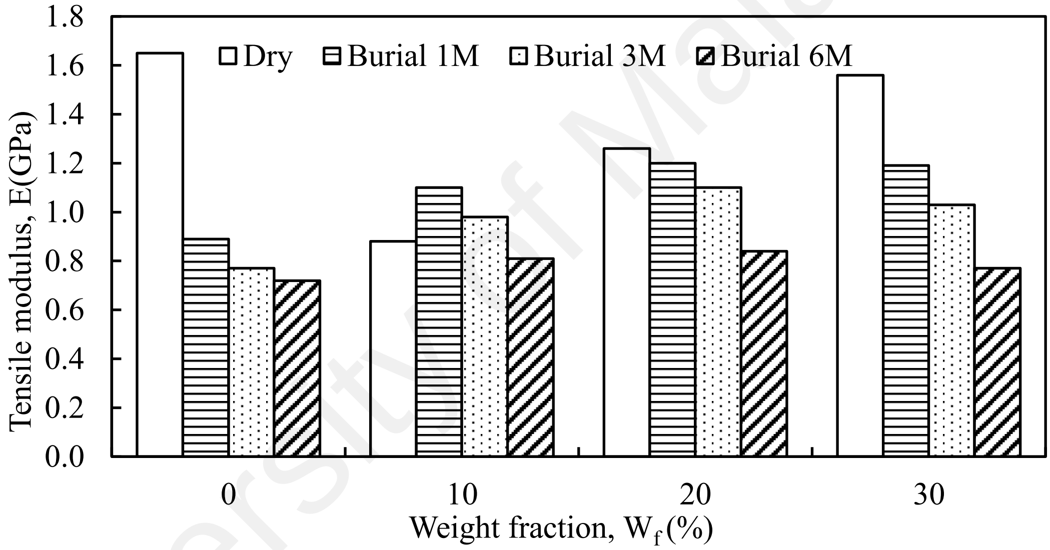


Figure 4.38: Tensile modulus of untreated fibre composite under dry and soil burial exposure at different time.

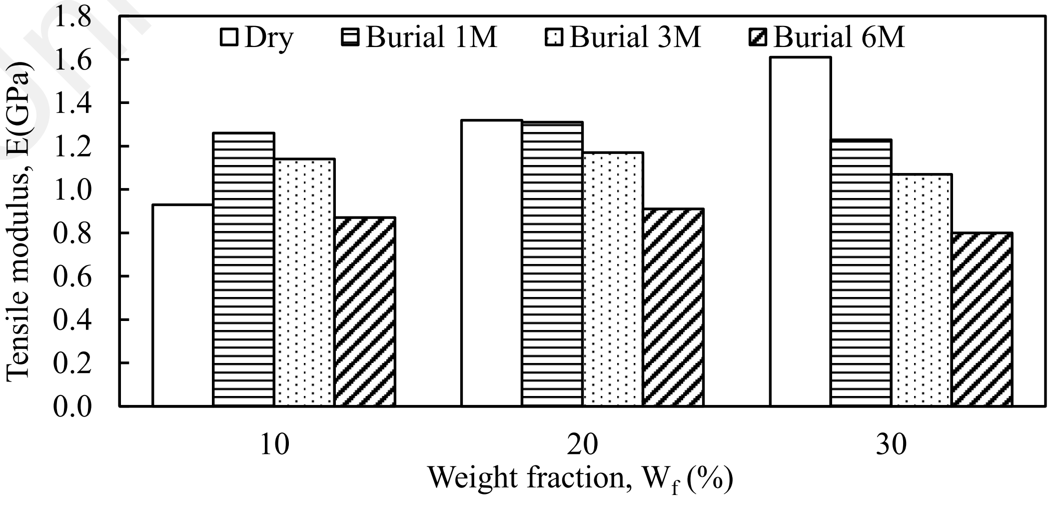


Figure 4.39: Tensile modulus of treated fibre composite under dry and soil burial exposure at different time.

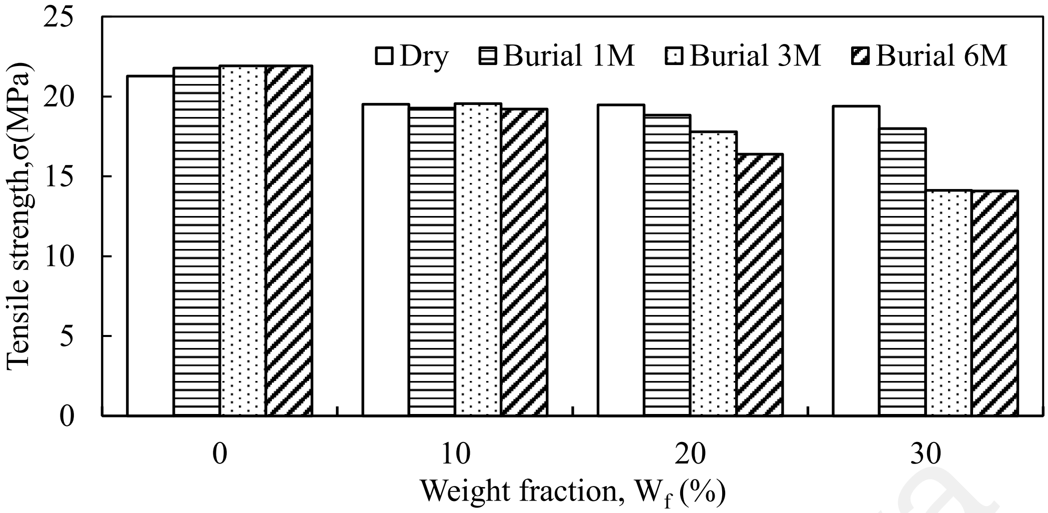


Figure 4.40: Tensile strength of untreated fibre composite under dry and soil burial exposure at different time.

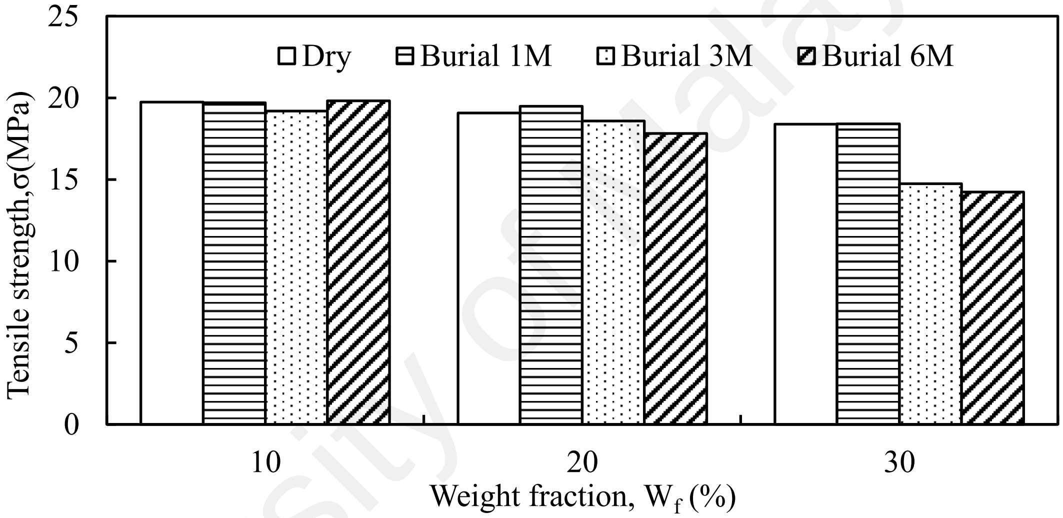


Figure 4.41: Tensile strength of treated fibre composite under dry and soil burial exposure at different time.

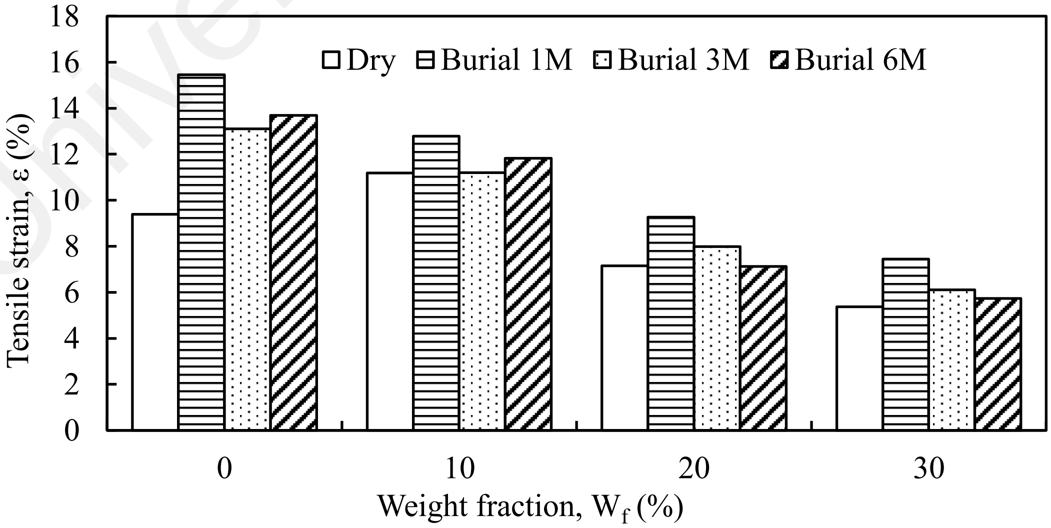


Figure 4.42: Tensile strain of untreated fibre composite under dry and soil burial exposure at different time.

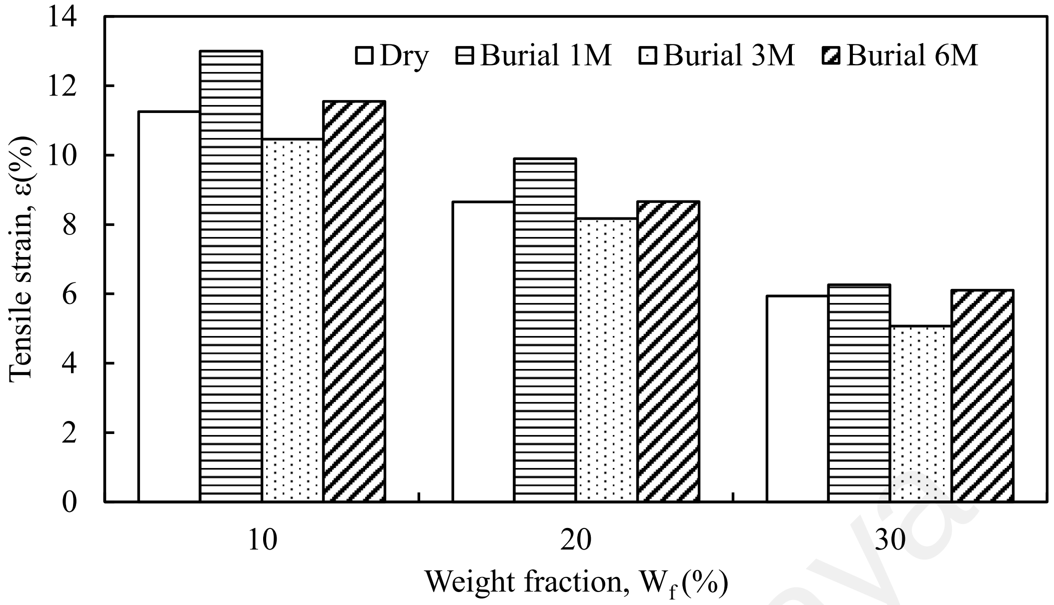


Figure 4.43: Tensile strain of treated fibre composite under dry and soil burial exposure at different time.

4.2.3.2 Flexural properties

Flexural testing studies the bending properties of a material. The flexural test measures the force required to bend a beam under three-point loading conditions. The data used to select materials for parts that will support loads without flexing such as polymers, wood composites. Specimens tested undergo a range of stresses across its depth. At the edge of the object on the inside of the bend (concave face), the stress will be at its maximum compressive stress value. At the outside of the bend (convex face), the stress will be at its maximum tensile value. At the outside of the bend (convex face), the stress will be at its maximum tensile value. (Ramakrishna & Huang, 2003). The flexural strength of any material or object defined as the maximum stress in the outermost fibres measured at 5% deformation/ strain. The test beam is under compressive stress at the concave surface. For flexural modulus, the value calculated from the slope of the stress vs. deflection curve. The value for flexural modulus, flexural strength and flexural displacement of composites are illustrated in Table 4.13 for all composites.

Table 4.13: Flexural properties for composite materials.

Sample	Flexural modulus, E (GPa)	Flexural strength, σ (Singh et al.)	Flexural displacement (mm)
HDPE	0.52	21.56	0.08
HDPE/EFB$_{U}$10	0.54	21.73	0.07
HDPE/EFB$_{180}$10	0.59	23.60	0.08
HDPE/EFB$_{U}$20	0.71	24.98	0.07
HDPE/EFB$_{180}$20	0.74	26.24	0.07
HDPE/EFB$_{U}$30	1.10	31.74	0.06
HDPE/EFB$_{180}$30	1.05	29.85	0.06

Effect of total W$_f$

Flexural modulus, E

The results show that flexural modulus increased steadily with increasing of fibre content for both untreated and treated fibre composites. The present of fibre causes the energy absorbed to break the test pieces to be increased. This is due to the reinforcing effect of the EFB fibre on the HDPE, which resulted in a good stress transfer from the fibres to the matrix. For untreated fibre composites, the flexural modulus has a slight increase form 0.52 MPa to 0.54 MPa for HDPE to 10% EFB fibre loading. Moreover, the modulus increases steadily from 0.54 MPa from 10% fibre loading to 0.71 MPa and 1.10 MPa at 20 and 30% fibre loading, respectively. The same trend observed for the treated fibre composites, where the flexural modulus values increase with the increasing in fibre loading. The values increased from 0.59 MPa for 10% fibre loading to 0.74 MPa and 1.05 MPa for composites with 20% and 30% fibre loading, respectively.

Flexural strength, σ

The flexural strength of untreated fibre composites increases from 21.56 MPa for pure HDPE to 21.73 MPa, 24.98 MPa and 37.74 MPa for 10%, 20% EFB and 30% EFB content respectively. Increment in the fibre loading causes the energy absorbed to break the specimen to be increased. This is due to the reinforcing effect of the EFB in which the EFB makes flexible HDPE become more rigid, thus increasing the flexural strength. Same trend was observed for treated fibre composites, in which the flexural strength also increases with increasing in EFB content. The flexural strength increased steadily from 23.60 MPa for 10% fibre loading to 26.245 MPa and 29.85 MPa for 20% to 30% fibre content, respectively.

Flexural Displacement

The flexural displacement of composites decreased with increasing in fibre content for both untreated and treated fibre composites. This result indicates that at higher fibre loading, the fibre dominates the fracture process of failure between fibre HDPE and EFB fibre. The decreased in the displacement explained by the fact that natural fibres have less tensile than the matrix. Therefore, addition of brittle fibres makes the matrix more rigid thus decreasing the mobility of polymer matrix (Joseph et al., 2002).

Effect of fibre treatment

Flexural modulus

The treated fibre composites have higher value of flexural modulus compared to untreated fibre composites at 10% and 20% EFB loading. This result is in line with earlier reports where heat treated fibre composites were found to improve the mechanical properties compared to the untreated fibre composites (Kaboorani et al., 2008). However, at 30% EFB loading, the values of flexural modulus have a slight decrease from 1.10 GPa for untreated fibre composites to 1.05 GPa for treated fibre composites.

Flexural strength

The treated fibre composites have higher value of strength compared to the untreated composites at 10% and 20% fibre loading. This improvement explained by an enhancement of fibres-matrix interaction under the compressive stresses during bending, developed in the transverse section of the flexural specimens. Moreover, increment in the fibre loading causes the energy absorbed to break the specimen to be increased. This is due to the reinforcing effect of the EFB fibre on the HDPE which resulted in good stress transfer from the fillers to the matrix. However, at 30% fibre loading, the values of flexural strength have a slight decrease when comparing to untreated composites. This is may be due to the deterioration of fibres as the treatment causes over extraction of extractives of fibres (Sood & Dwivedi, 2017).

Flexural Displacement

The treatment of fibre shows almost similar value of the flexural displacement compared with the untreated fibre composites. The treatment of fibre shows a slight increment in flexural displacement when compared to untreated fibre composites from 0.07 mm to 0.08 mm at 10% fibre loading.

Effect of conditioning

Indoor conditioning

Flexural modulus, E

The flexural modulus of untreated composites shown in Figure 4.44 below. From the figure, flexural modulus of HDPE under dry condition shows minimal changes after the exposure to indoor conditions. The value increase from 0.52 GPa for dry specimen to 0.53 GPa for sample under 1 month indoor exposure. However, the flexural modulus was reduced to 0.49 GPa and 0.50 GPa for 3 and 6 months indoor exposure, respectively. Same trend observed in all types of composites in which there is minimal changes in flexural modulus of the conditioned and dry as moulded for indoor 3 months exposure but in no trend. This can be concluded that indoor environmental exposure does not change flexural modulus of composites in any noticeable way. However, the flexural modulus increased at 10% and 20% untreated fibre composites.

For the treated composites, flexural modulus increases slightly after being exposed to indoor environment (1 to 6 months) (Figure 4.45). However, at higher fibre loading 30% fibre loading, there is a decrease in value from 1.05 GPa for dry sample to 0.96 GPa for samples exposed to indoor environment in 1 month. The same trend was observed with untreated fibre composites where the value decrease at 30% fibre loading. This is may be due to the higher water absorption during the exposure especially at the first month of exposure at the highest fibre loading (Munoz & García-Manrique, 2015).

Flexural strength, σ

Similar with flexural modulus, flexural strength of composites shows insignificant changes after the time of exposure to indoor environment. Values of flexural strength of dry moulded HDPE (21.56) MPa in almost the same as the indoor exposure (21.67 MPa). As for the untreated composites, the trend observed was similar to HDPE in which the values of flexural strength did not differ much from the dry as moulded samples except for 30% fibre loading composites. The changes at 30% fibre loading quite significant but with no specific trend. Similarly, the flexural displacement of the dry as moulded and indoor exposed composites is almost the same (Figure 4.46). Therefore, it can be concluding that indoor exposure is favourable to the flexural properties of composites.

The trend observed similar of the treated fibre composites whereby the indoor environment does not change the flexural strength of the composites (Figure 4.47). At 10% fibre loading composites, values of the flexural strength at dry as moulded (23.60 MPa) is similar with the indoor 1-month exposure (23.81 MPa). However, there is a slight change in composites exposed to indoor exposure for 3 and 6 months, but in no particular trend.

Flexural Displacement

Flexural displacement of composites shows insignificant changes after the time of exposure to indoor environment. Values of flexural strength of dry moulded HDPE (21.56) MPa in almost the same as the indoor exposure (21.67 MPa). As for untreated fibre composites, the trend observed was similar to HDPE in which the values of flexural strength did not differ much from the dry as moulded samples except for 10% fibre loading composites (Figure 4.48). The changes at 10% fibre loading quite significant but with no specific trend. Similarly, the flexural displacement of the dry as moulded and indoor exposed treated fibre composites is almost the same except decrease at 20% fibre loading during 3 months of exposure (Figure 4.49). Therefore, it can be concluding that indoor exposure is favourable to the flexural properties of composites.

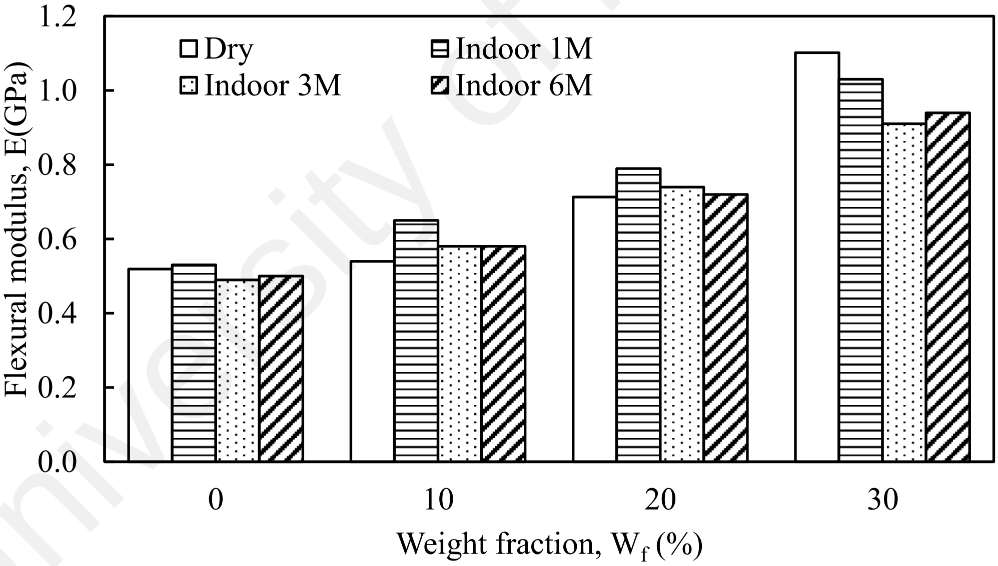


Figure 4.44: Flexural modulus of untreated fibre composite under dry and indoor exposure at different time.

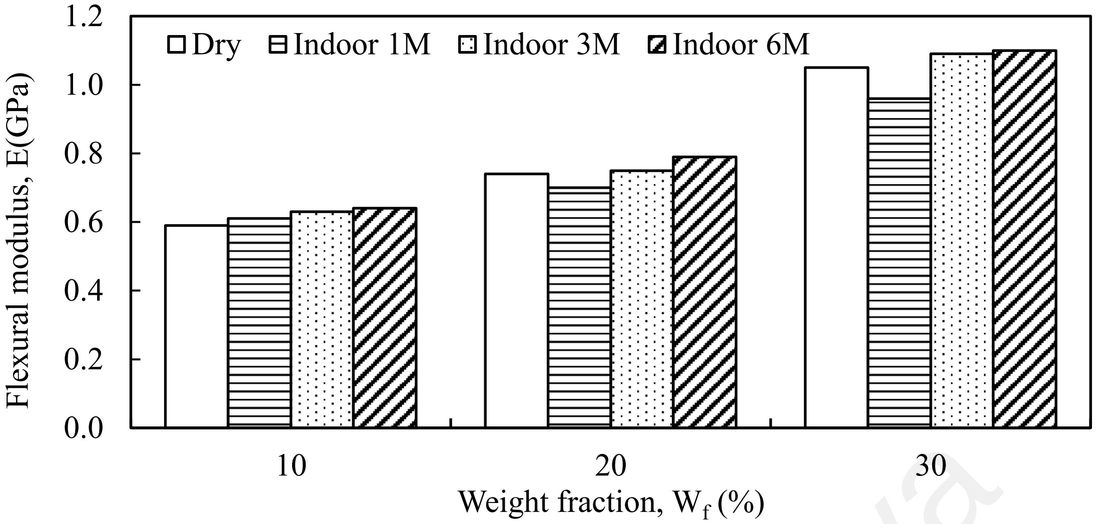


Figure 4.45: Flexural modulus of treated fibre composite under dry and indoor exposure at different time.

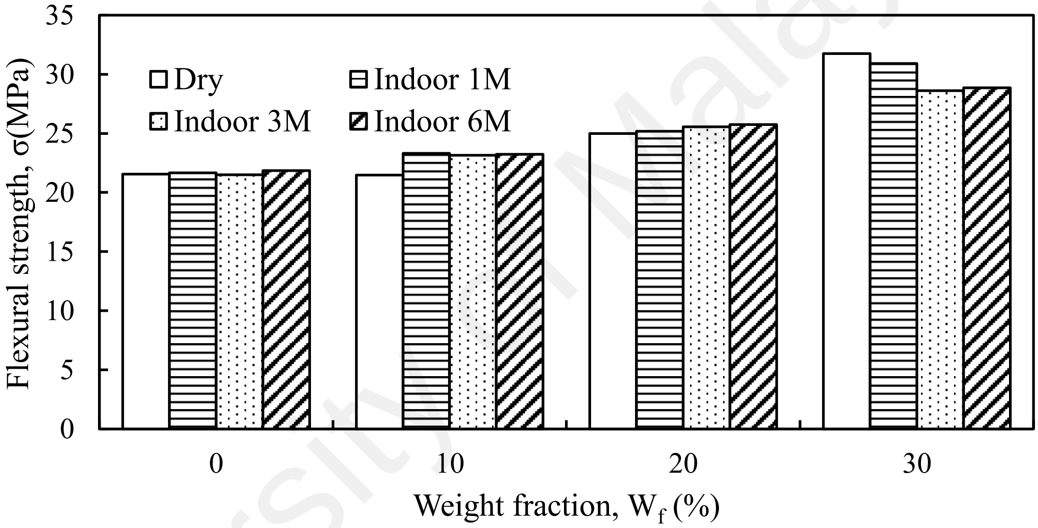


Figure 4.46: Flexural strength of untreated fibre composite under dry and indoor exposure at different time.

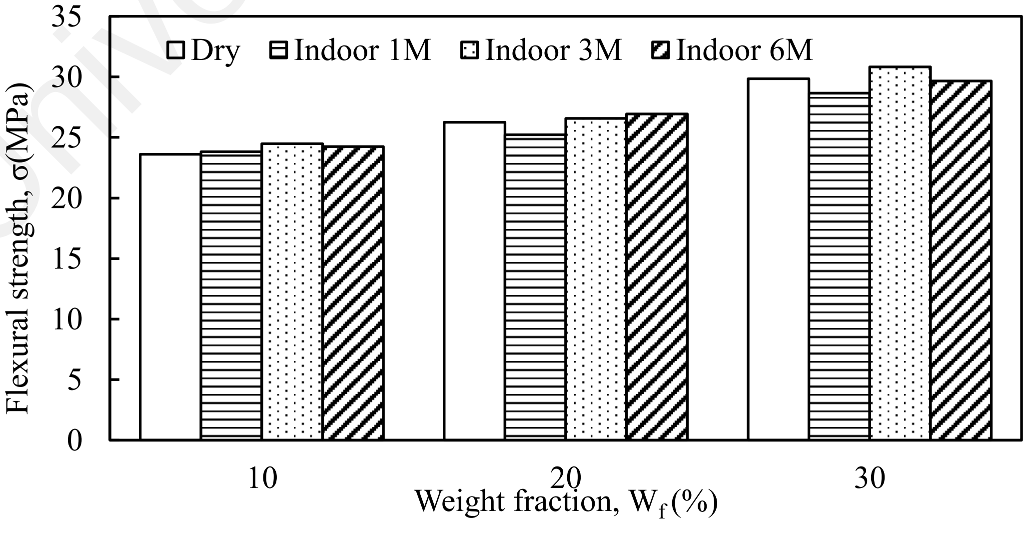


Figure 4.47: Flexural strength of treated fibre composite under dry and indoor exposure at different time.

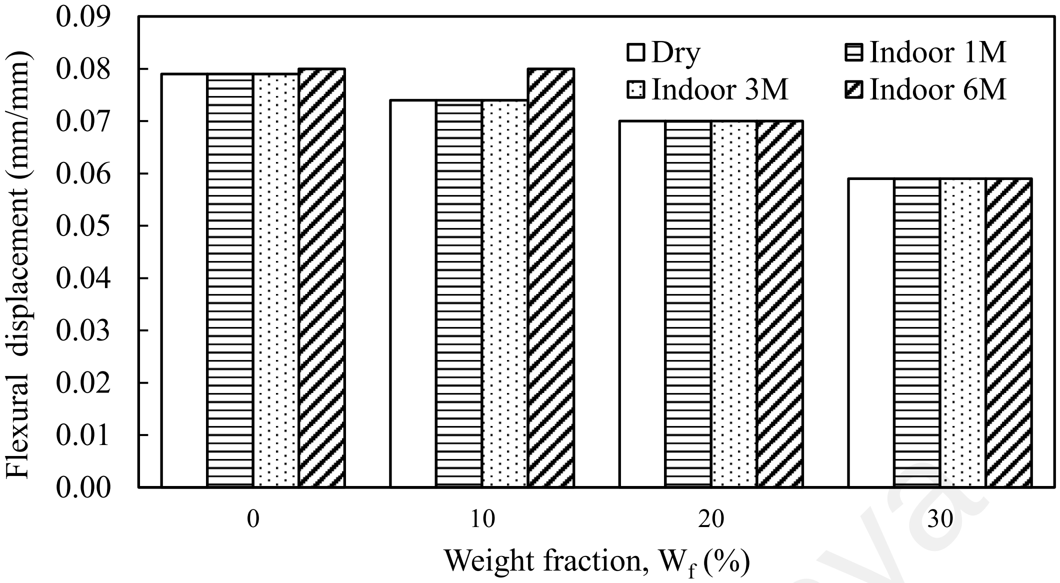


Figure 4.48: Flexural displacement of untreated fibre composite under dry and indoor exposure at different time.

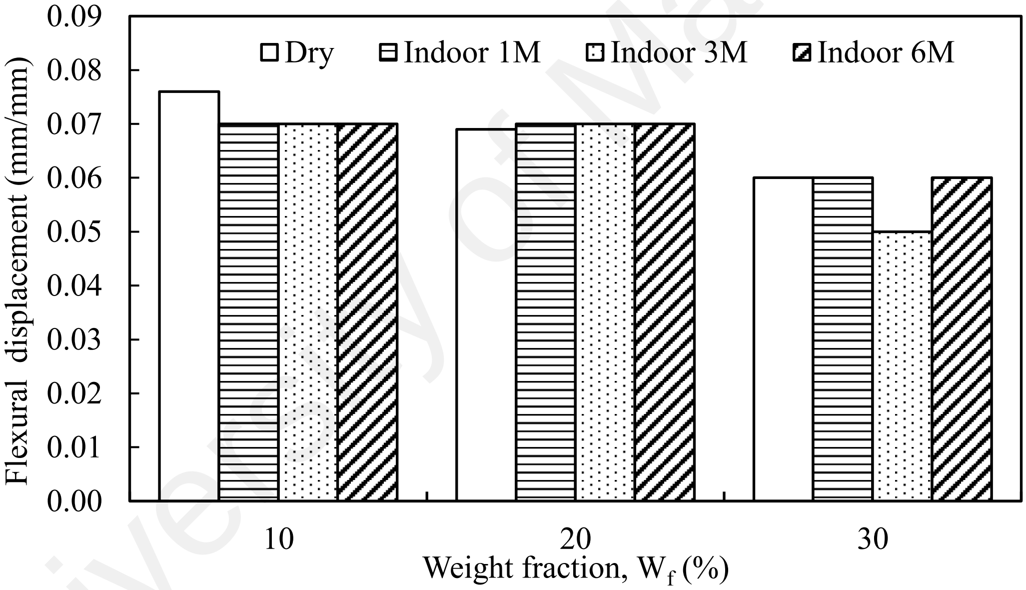


Figure 4.49: Flexural displacement of treated fibre composite under dry and indoor exposure at different time.

Soil burial conditioning

Flexural modulus, E

The flexural modulus of untreated fibre composites was shown in Figure 4.50 below. From the figure, there is no much change in the flexural modulus for HDPE upon soil burial conditions. Values of 0.52 GPa is obtained for the dry as moulded HDPE, burial for 1 month and 3 months, while 0.54 Gpa is recorded for the sample after 6 months soil burial exposure. As for the untreated composites (Figure 4.50), there is a general decrease in value of tensile modulus of the buried sample compared to the dry as moulded sample. In composites with 20% fibre loading, the flexural modulus decreases from 0.71 Gpa for the dry as moulded sample to 0.66 Gpa for sample under 1 month of soil burial condition. The flexural modulus has further decrease to 0.65 Gpa and 0.57 Gpa,for 3 and 6 months burial samples, respectively. The trend is similar to a study done by Abdul Khalil H.P.S (Khalil et al., 2010), where the flexural modulus decreases in the increasing time of soil burial. As the moisture being absorbed during the soil burial condition, it will penetrate the EFB structure and causing the adhesion between EFB and HDPE to be distracted, hence resulted in a swelling behaviour. Due to the swelling, poor adhesion between fibre and matrix is expected thus initiates crack formation, and composites expected to fail easily.

For treated fibre composites (Figure 4.51), at 10% fibre loading, there is no significant changes in the values of flexural modulus after buried in soil (1 to 6 months). However, at 20% and 30% fibre loading, there is a decrease in value from dry samples until 6 months of burial. At 20% fibre loading, the values of flexural modulus have a slight decrease from 0.74 Gpa for dry sample to 0.68 Gpa at 1 month soil burial. The values have further reduction to 0.64 Gpa and 0.60 Gpa for 3 and 6 months of soil burial respectively. At 30% fibre loading, the flexural modulus decreases from 1.05 Gpa for dry samples to 0.74 Gpa at 1 month of soil burial. The values have further reduction to 0.61 Gpa and 0.60 Gpa at 3 and 6 months of soil burial respectively. The values however higher than the untreated fibre composites. This may be due to the removal of impurities of fibres during the treatment, leading to a better interaction of fibre and matrix.

Flexural strength, σ

Similar with flexural modulus, flexural strength values for HDPE did not change upon soil burial. Values of 21.56 MPa for dry as moulded is obtained for the dry as moulded samples while 22.19 MPa for soil burial 1 month, 22.68 MPa and 23.36 MPa for burial 3 months and 6 months respectively. Same trend was observed for untreated fibre composites with 10% and 20% fibre loading (Figure 4.52). However, at 30% fibre loading, the flexural strength has a significant decrease where values of 31.74 MPa is recorded for dry as moulded decrease to 25.11 MPa for 1-month soil burial and further decrease to 23.38 MPa and 20.81 MPa for 3 months and 6 months soil burial respectively. This was probably due to high amount of absorption of water during soil burial with consequent fibre swelling that may lead to micro cracking of the HDPE matrix surrounding the EFB in addition to relatively poor interaction between EFB and HDPE matrix (Khalil et al., 2010).

The flexural strength value of treated fibre composites shown in Figure 4.53. The same trend observed which the value of tensile strength decreases as the time of buried increase but with higher value compared to untreated fibre composites. Not much changes were observed with 10% and 20% treated fibre composites. At 30% treated fibre composites, the value decrease form 29.85 MPa for dry as moulded to 26.28 MPa at 1-month soil burial. The tensile strength further decreased to 22.53 MPa and 21.49 MPa for 3 and 6 months soil burial respectively. From this observation, the effect of heat treated EFB in resisting the effect of fungal activities is more pronounced as the flexural strength has a higher value than untreated fibre composites.

Flexural displacement

The variation of flexural displacement of the dry as moulded and soil buried composites is shown in Figure 4.54 and Figure 4.55. Flexural displacement shows insignificant changes after the soil burial at all types of composites. Generally, the value of flexural displacement of all composites have same or higher value than the dry as moulded samples. This trend is expected because interfacial degradation as a result of fungal activities should increase displacement at yield of the composites.

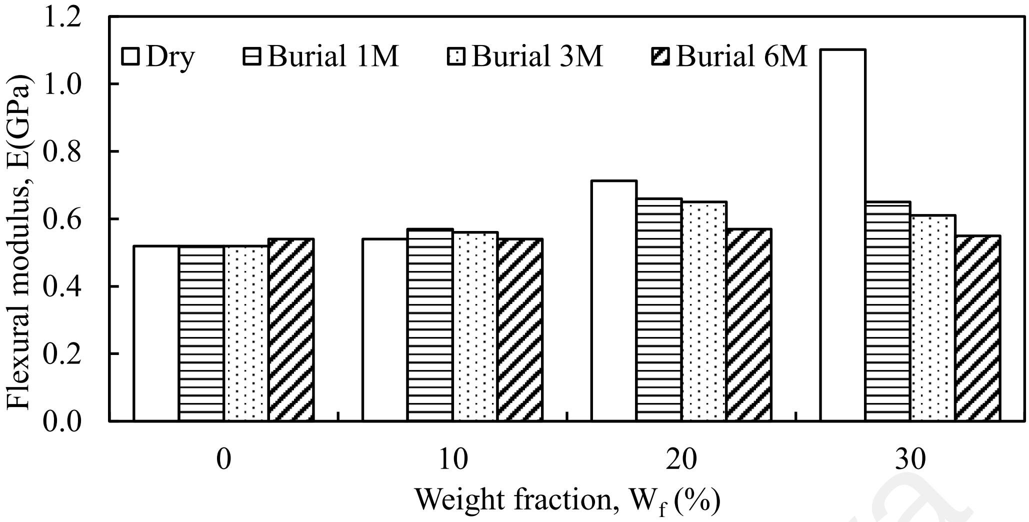


Figure 4.50: Flexural modulus of untreated fibre composite under dry and soil burial exposure at different time.

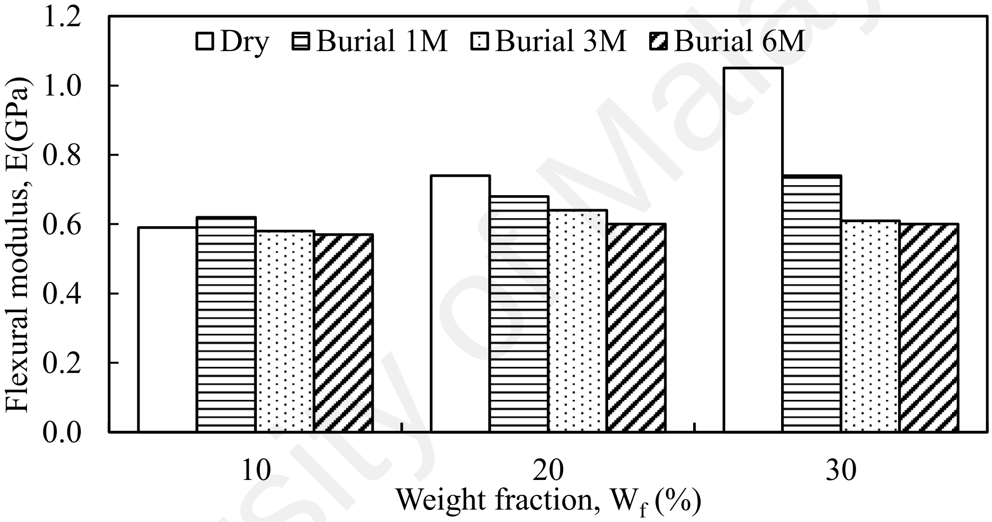


Figure 4.51: Flexural modulus of treated fibre composite under dry and soil burial exposure at different time.

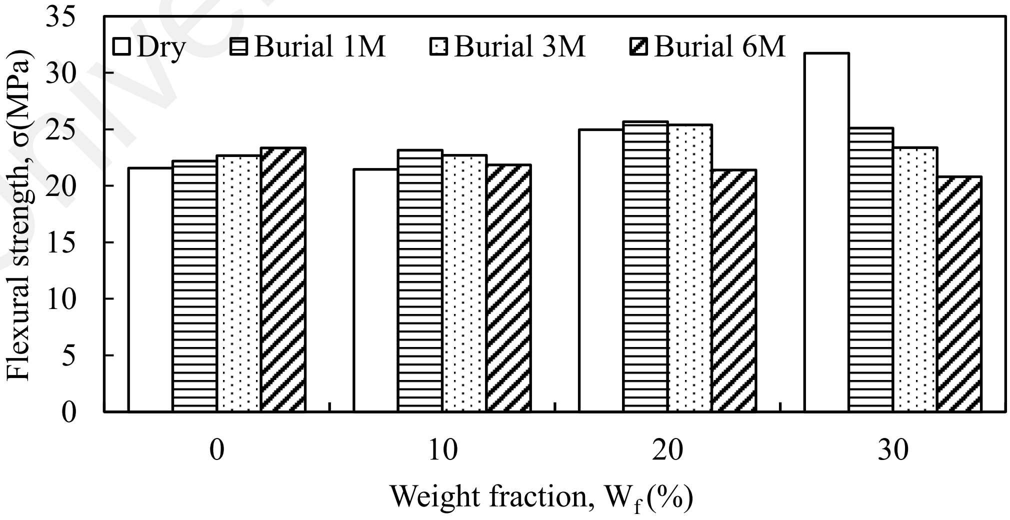


Figure 4.52: Flexural strength of untreated fibre composite under dry and soil burial exposure at different time.

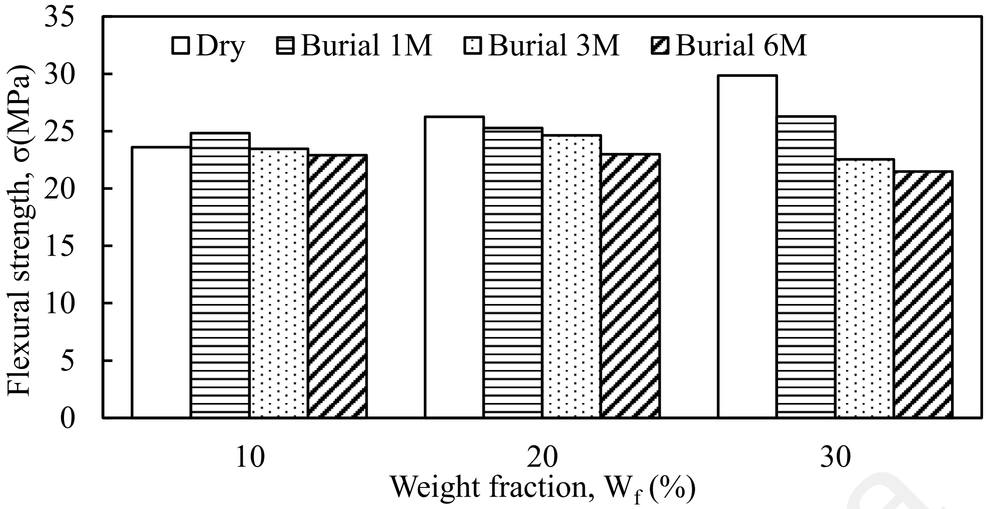


Figure 4.53: Flexural strength of treated fibre composite under dry and soil burial exposure at different time.

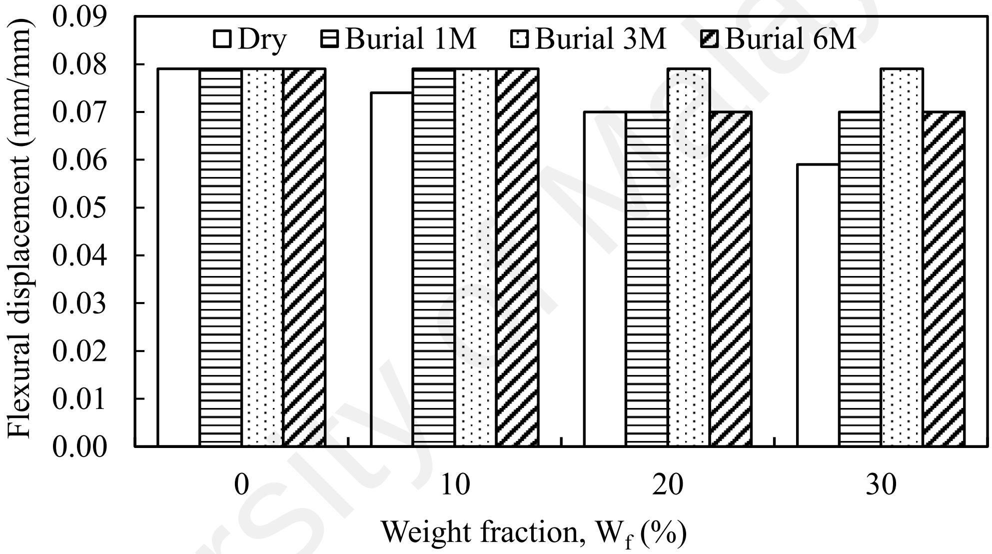


Figure 4.54: Flexural displacement of untreated fibre composite under dry and soil burial exposure at different time.

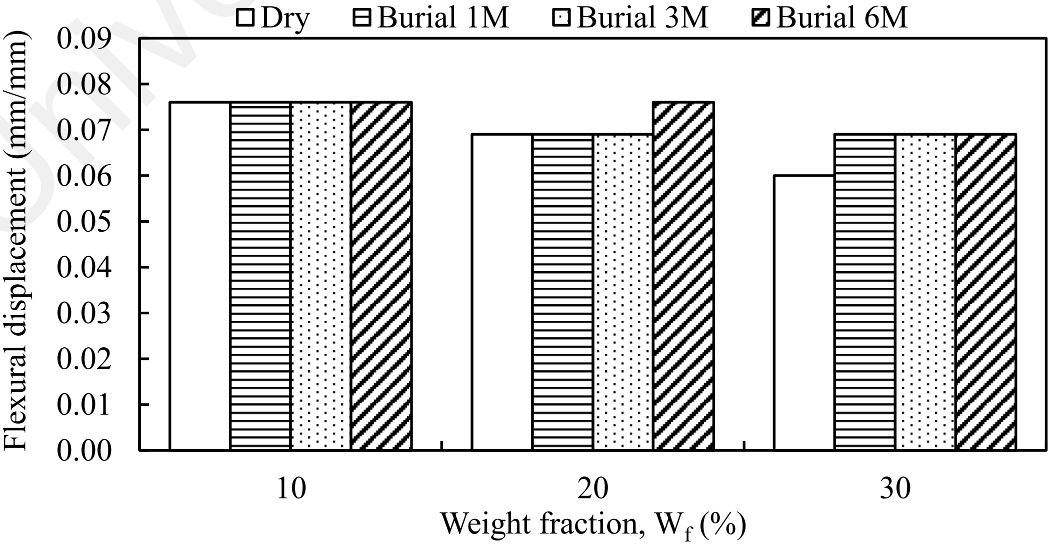


Figure 4.55: Flexural displacement of treated fibre composite under dry and soil burial exposure at different time.

4.2.3.3 Impact properties

Impact behaviour is one of the important aspects to characterise materials. Impact resistance is a material's ability to withstand intense force or shock applied to it over a short period. Impacts force can come in the form of natural elements, such as wind or earthquakes, explosions, collisions, and rapid loading. In application, it is important to consider impact behaviour if the materials have tendency to experience high risk of impact. A material with high impact resistance is able to absorb more energy and disperse the energy efficiently. In the lab, impact properties are often measured through a common Charpy test, where a weighted pendulum strikes a sample located opposite of the notch with a V-shape impactor tup.

Impact energy of a material is the amount of energy required to fracture in a given volume of material. Brittle materials have low toughness as a result of the small amount of plastic deformation that they can endure. The impact value of a material can also changes with temperature. Generally, at lower temperatures, the impact energy of a material decreases.

Impact test discovers the amount of energy absorbed by a material during fracture. This absorbed energy is a measure of a given material's toughness and acts as a tool to study temperature-dependent brittle-ductile transition. It is to decide whether the material is brittle or ductile in nature (François et al., 2013).

From impact tests, energy vs. time graphs were produced for each tested specimen. From these graphs, there are two important parameters were extracted which are fracture energy (W) and peak load (P). Fracture energy is described as the energy required to open unit area of crack surface or the total amount of energy dissipated during crack growth (Mueller & Knauss, 1971). It is a material property and does not depend on size of structure. Fracture energy is measured by calculating the area under the graph of the sample fracture. The peak load was obtained from the highest point in the graph.

The relationship between W and the critical strain energy release rate (Ngo et al., 2014) and specimen geometry function (BDΦ) is given by,

$$W = G_c BD\,\Phi \qquad (4.4)$$

where B and D are the width and depth of the specimen, respectively. A correction factor, Φ is given by,

$$\phi = \frac{1}{2}\left(\frac{a}{D}\right) + \frac{1}{18\pi}\left(\frac{S}{D}\right)\frac{1}{\left(\frac{a}{D}\right)} \qquad (4.5)$$

where a and S are notch depth (crack length) and the specimens support span respectively.

A plot of W against BDΦ produced a straight line, where its slope is equal to the G_c of the materials. Strain energy release rate (G) is defines as the energy dissipated during fracture per unit of newly formed surface. When the G value exceeded the critical value, G_c, the crack would grow, and failure occur. The trend observed for G_c value is the same as observed for W. G_c increases as the fibre loading increased. Increment in G_c increase is due to more energy needed to form new surface as failure occurs caused by increase in fibre content. The fibres would hinder crack propagation by forcing it around the fibre ends.

The G_c qualities of the composite can be further enhanced by improving the interfacial interaction of the fibres and the matrix. High density polyethylene is non-polar while EFB fibre is polar. Therefore, it is highly unlikely that there is any significantly strong interaction between these two materials. Enhancement in the fibre-matrix can be done via surface treatment of the EFB fibres.

The relationship between the critical stress intensity factor (K_c) with nominal fracture stress (σ), geometry correction factor (Y) and notch or crack length (a) is given by the following equation,

$$\sigma Y = \frac{K_c}{\sqrt{a}} \tag{4.7}$$

In three-point bend test, σ is given by simple bending theory as,

$$\sigma = \frac{6PS}{4BD^2} \tag{4.8}$$

For the three-point bend test specimen, where S/D is equal to 4, Y is given by,

$$Y = 1.93 - 3.07\left(\frac{a}{D}\right) + 14.53\left(\frac{a}{D}\right)^2 - 25.11\left(\frac{a}{D}\right)^3 + 25.80\left(\frac{a}{D}\right)^4 \tag{4.9}$$

A plot of σY against $a^{-0.5}$ produced a straight line, where its slope is equal to the K_c of the materials. K_c values for fibres composites increases with increase in fibre loading. K_c is more sensitive towards the rigidity of the material and describes the initial part of failure (crack initiation).

Effect of W_f

Fracture energy, W

Figure 4.56 and Figure 4.57 show the fracture energy (W) against fibre loading and at all values of notch to depth ratio (a/D) for both untreated and treated composites. From the figures, it is noticed that increasing the a/D ratio would cause the fracture energy to decrease. The fracture energy of 10% untreated fibre composites at 0.1 a/D is 260.12 mJ decrease to 182.38 mJ, followed by 143.26 mJ and 133.5 mJ as the a/D increases to 0.2, 0.3 and 0.4 respectively as shown in Figure 4.60. Similar trend was observed for 10% treated fibre composites (Figure 4.57), where the fracture energy decrease from 263.29 mJ at 0.1 a/D to 198.13 mJ followed by 172.43 mJ and 153.9 mJ as the a/D increases to 0.2, 0.3 and 0.4 respectively. As stated before, the fracture energy is related to the distance where the crack needs to travel during failure. Higher a/D explained that the crack initiated is longer, thus it will shorten the distance for crack to propagate (small fracture area) resulting in lower fracture value (Hassan, et al., 2011).

The effect of fibre loading on the fracture energy of the composites were also investigated. For untreated composites, facture energy at a/D 0.2 increases from HDPE to 10% and 20 fibres loading then slightly decreased at 30% fibre loading. The increase in fracture energy is due to more fibres present in the composite, thus more energy needed to break the specimens. The fibres also hinder the crack growth by causing the crack to move along its interface resulting in higher W. However, when the fibres added beyond 20% fibre loading, the property of the material seems to be reduced. This may be due to wetting problem with such high EFB fibre content. This statement is supported by the fracture surface field emission scanning electron microscopy (FESEM) analysis. Figure 4.58 shows the FESEM image of fracture surface of untreated fibre composites with 30% fibre loading. From the image, it can be seen that EFB fibres does not dispersed well in the composites and tend to cling together in bundle, causing wetting problem, thus reducing the mechanical properties of the fibres (Bakar et al., 2005a).

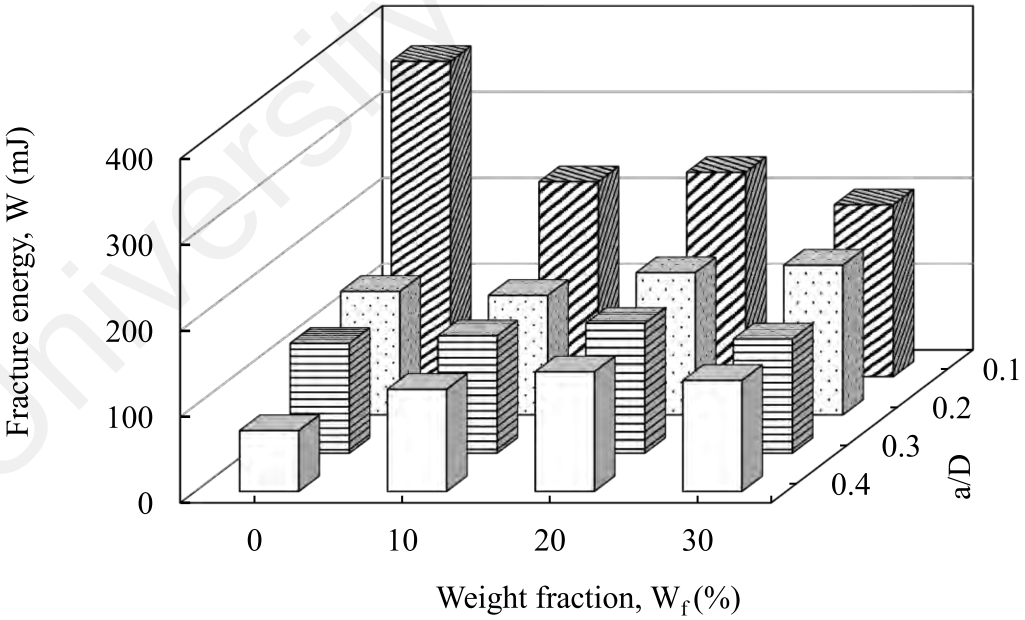


Figure 4.56: Fracture energy for untreated composites at different fibre loading.

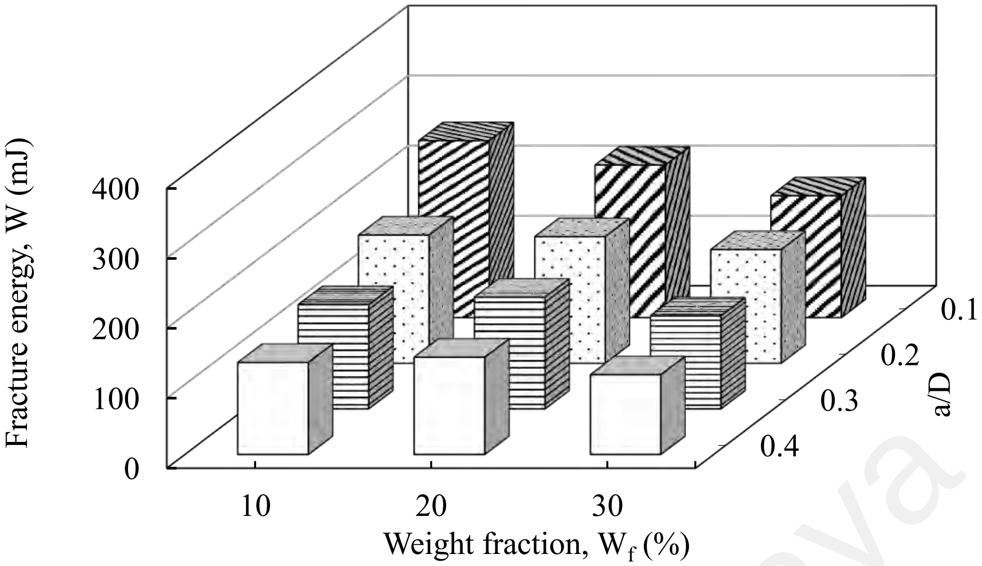


Figure 4.57: Fracture energy for treated composites at different fibre loading.

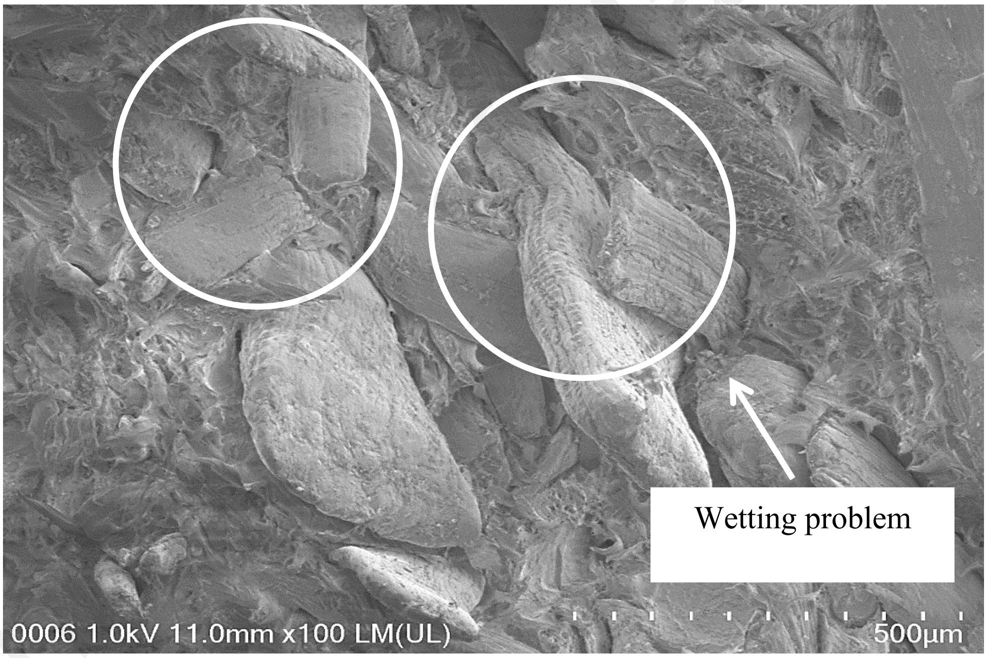


Figure 4.58: Impact fracture surface of 30% untreated fibre composites.

When the fibre content exceeds the optimum level, the improvement in the mechanical properties would start to level off and even decrease as the content is further increased. (Drzal & Madhukar, 1993). This observation caused by the wetting problem faced by the composite. Interfacial interaction between fibre and matrix is one of the most important components in a composite. Good interfacial interaction would allow stress to transfer from the matrix to the fibre in an efficient manner, producing superior composite.

However, when fibre content is too high, the fibres and matrix to be poorly mix. As a result, various defects such as voids and undispersed fibre bundles are present in the mix. These flaws act as a stress concentrator and weaken the composite (Bakar et al., 2005a).

Critical energy release rate, G_c

Strain energy release rate (G) defines as the energy dissipated during fracture per unit of newly formed surface and only when G exceed the critical value, G_c, the crack grows, and failure occurs. Generally, the trend observed regarding G_c is in agreement with the trends earlier observed in W. An example of the W against BDΦ is given in Figure 4.59. Similar graphs were plotted for all types of composites and their G_c value were tabulated in Table 4.14. A plot of W against BDΦ produced a straight line, where its slope is equal to the G_c of the materials.

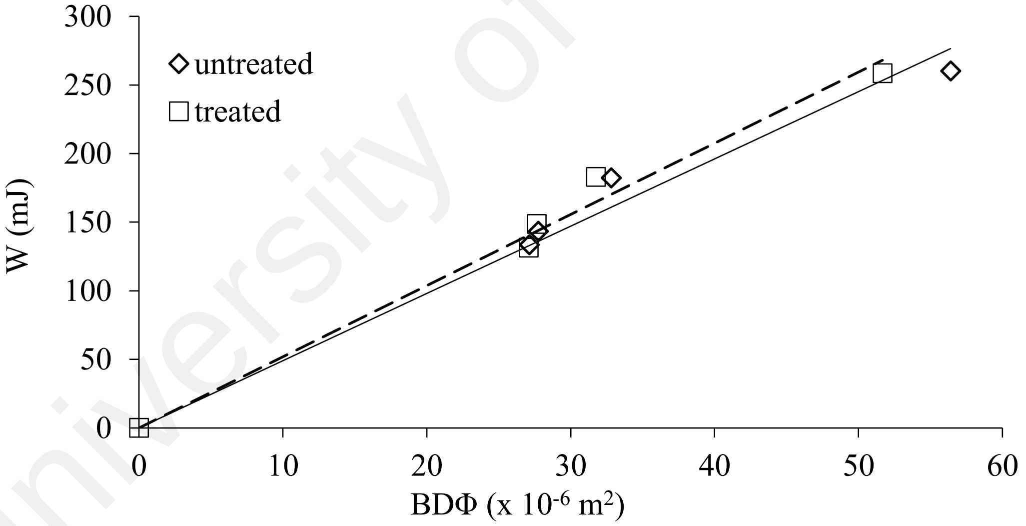


Figure 4.59: W against BDΦ plot for 10% untreated and treated fibre composites.

Value of G_c for HDPE and all types of composites presented in Table 4.14. Pure HDPE exhibit the highest value of G_c which is 5.86 kJ m^{-2}. The high value of HDPE is due to slow crack propagation of the ductile behaviour of pure HDPE. For untreated composites, the values of G_c decreased with addition of fibre loading. The highest value obtained for 10% EFB fibre loading is 5.56 kJ m^{-2}, whereas for 30% EFB fibre loading G_c value obtained is 4.88 kJ m^{-2}, The decreased observed in value of G_c with higher fibre loading is may be due to the presence of more EFB fibre ends within the HDPE which tends to allows more crack propagation. Moreover, at higher fibre content, the possibility of the EFB fibres to agglomerate is high. This agglomeration may act as stress concentration area, enhancing the crack growth. For treated composites, the value of G_c, the same trend was observed as the untreated fibre composites, however the value is higher. This may be due to improve adhesion between fibre and matrix, so that energy dissipative processes involved in the toughening of the composites is well to improve the mechanical properties of composites (Hughes et al., 2002).

Table 4.14: G_c and K_c for all type of composites.

Sample	**G_c (kJm^{-2})**	**K_c (MPam $^{1/2}$)**
HDPE	4.77	1.91
HDPE/EFB$_U$10	5.56	1.88
HDPE/EFB$_U$20	5.54	2.01
HDPE/EFB$_U$30	4.88	2.11
HDPE/EFB$_{180}$10	6.24	2.05
HDPE/EFB$_{180}$20	6.05	2.00
HDPE/EFB$_{180}$30	5.21	2.12

Peak load, P

Peak load is defined as the maximum force needed to cause the fracture of the sample. It depicts the highest point on the load-deflection curve and is a function of the damage resistance of a material. The peak load values of composites are illustrated in Figure 4.60 and Figure 4.61. Based on the graph, *P* decreases with notch to depth ratio, a/D. When a/D increase, the available length for crack propagation reduced thus decreases the maximum load required for facture. The neat HDPE however has the highest value of *P* comparing to the composites. When introducing EFB fibre, the values of P slightly reduced at all a/D values suggested. This can be said due to the brittle behaviour of the EFB fibre. In addition, the incorporation of fibres also restrict the movement in the flexible polymer matrix (Bakar et al., 2005). Lower values of *P* were observed at low

EFB content (10 wt. %), indicates that the samples cannot sustain the maximum load. However, at 20 and 30% EFB fibre loading, the increment in *P* values compared to 10 wt. % was observed. The increase is due to an increase in the rigidity of the material. The presence of fibres restricts the movement of the polymer chains resulting in increased stiffness. Stiffer materials tend to have higher value of peak load when tested the initial part of failure (crack initiation).

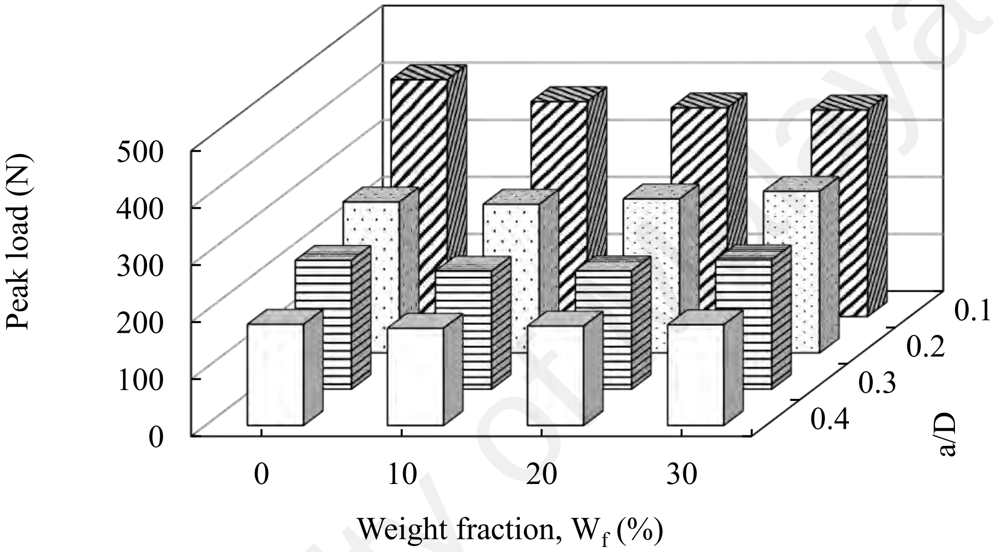


Figure 4.60: Peak load for untreated composites at different fibre loading, W_f (%).

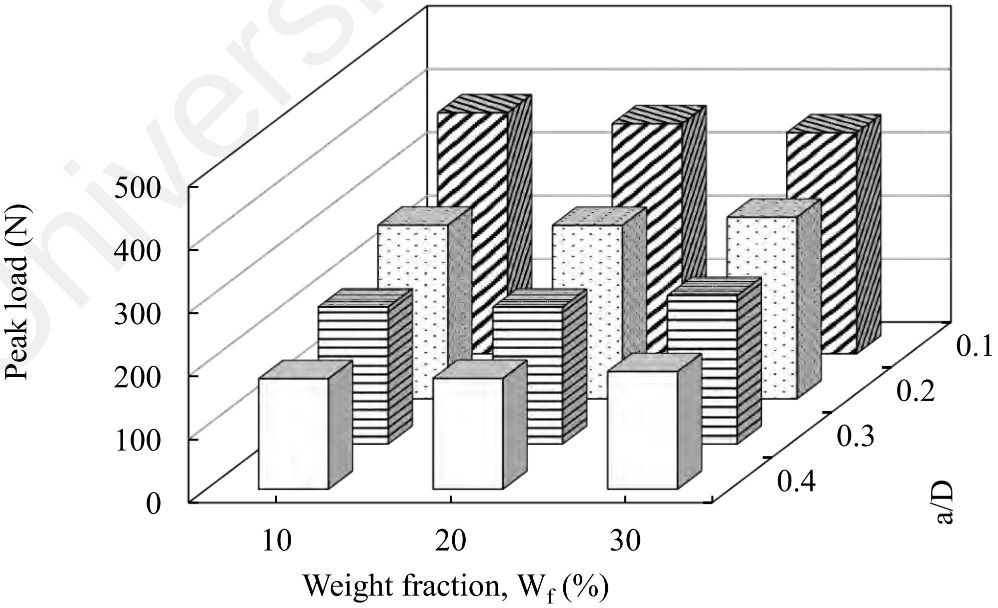


Figure 4.61: Peak load for treated composites at different fibre loading, W_f (%).

Critical stress intensity factor, K_c

An example of σY against $a^{-0.5}$ plot is illustrated in Figure 4.62 below. Kc values for all the composites are extracted from their respective curves of σY against $a^{-0.5}$ and tabulated in Table 4.14. A plot of σY against $a^{-0.5}$ produced a straight line, where its slope is equal to the K_c of the materials.

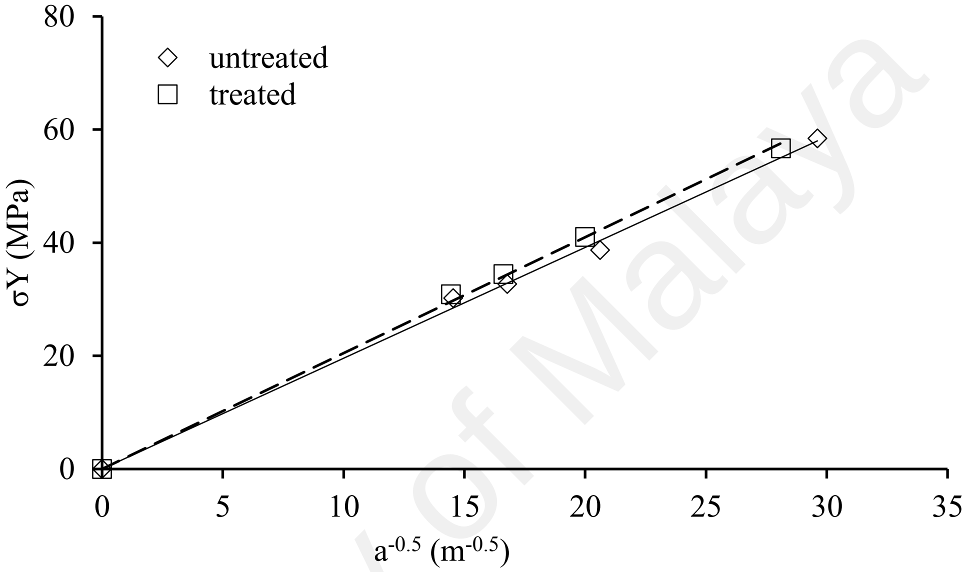


Figure 4.62: of σY against $a^{-0.5}$ plot for 10% untreated and treated fibre composites.

Effect of fibre treatment

Fracture energy, W

The effect of heat treatment on W of the composites can be seen in Figure 4.57. Generally, the value of fracture energy in treated fibre composites is higher than the untreated fibre composites despite the a/D ratio and fibre compositions. The value of fracture energy of 10% untreated fibre composites at 0.2 a/D increase from 182.38 mJ to 197.14 mJ after the heat treatment. The increase in value of W is expected, indicating that treating the EFB fibre has modified the EFB fibre at certain degree, which then enhances the compatibility between the non-polar HDPE matrix and the polar EFB fibre relative to the untreated fibre composites.

Critical energy release rate, G_c

The effect of heat treatment on G_c of the composites is also shown in Table 4.15. From the data extracted, it was observed that the value of G_c increased linearly with heat treatment at all fibre loadings, with HDPE/EFB$_{180}$10 has the highest value of 6.24 kJ m^{-2}. At 30% untreated fibre composites, the value observed was 4.88 kJ m^{-2} and it is increased to 5.22 kJ m^{-2} after the treatment.

Peak load, N

The effect of heat treatment on *P* values of the composites is also presented in Figure 4.65. Based on the data extracted in the graph, the values of P in treated fibre composites is higher that untreated fibre composites. This result is expected and could be attributed to the reduced hydrophilicity of the fibre after treatment, thus increase the compatibility between EFB fibre and HDPE. This can be proven by the SEM image in Figure 4.63. EFB fibre is pull out from the matrix, indicating the bonding between EFB and HDPE is poor whereas in Figure 4.64, some portion of HDPE is still adhering at the EFB particles, indicating better interfacial adhesion after the treatment.

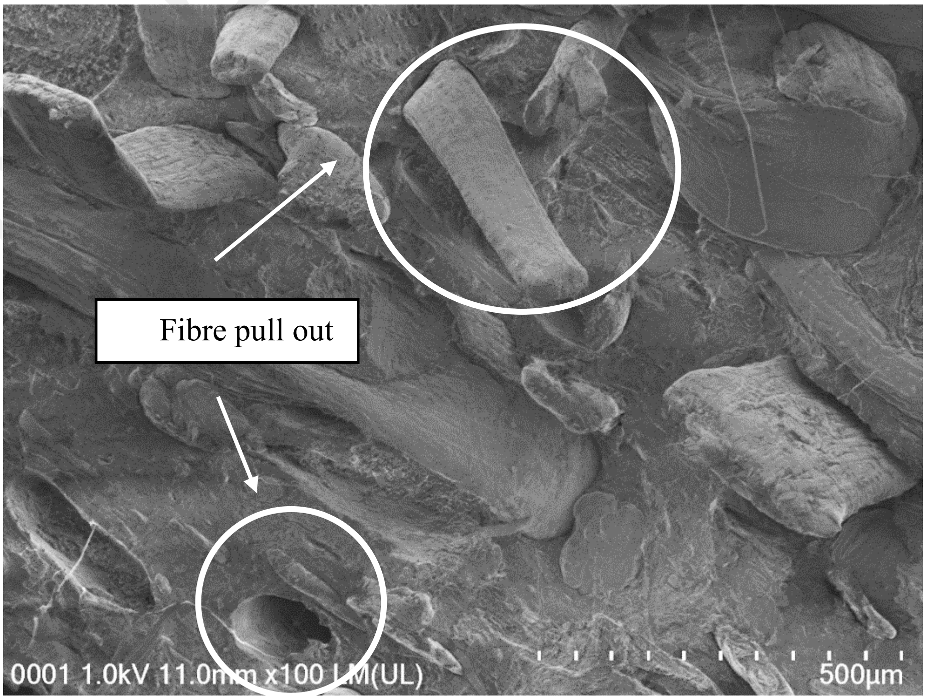


Figure 4.63: Impact fractured surface of 30% untreated fibre composites.

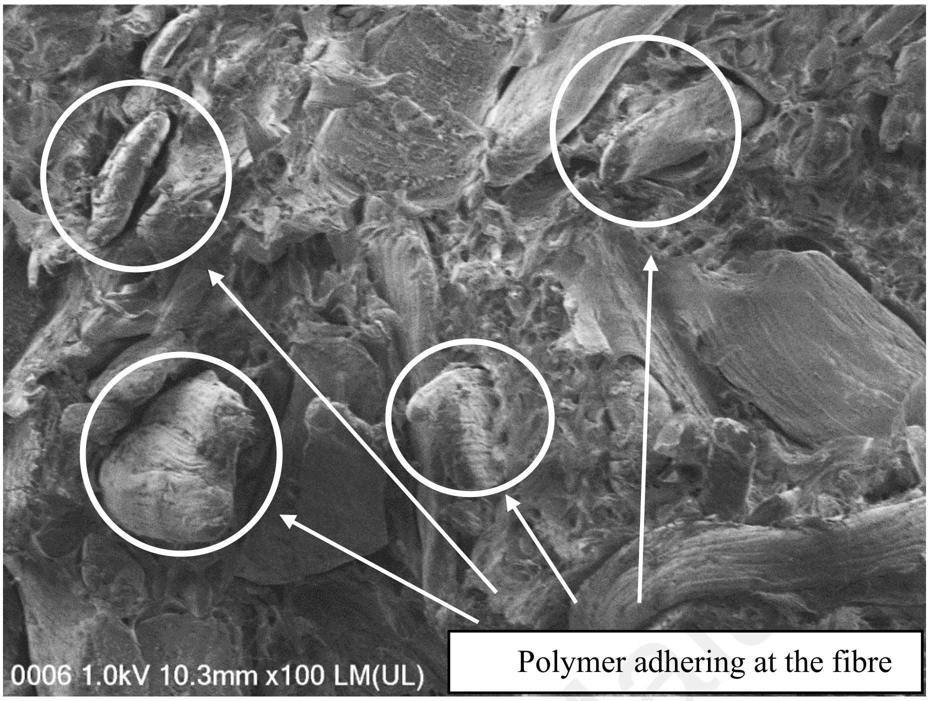


Figure 4.64: Impact fractured surface of 30% treated fibre composites.

Critical stress intensity factor, K_c

The effect of K_c in fibre treatment are presented in Table 4.15. Values of K_c increases with heat treatment relative to the untreated fibre composites for all fibre compositions. The value of K_c increase from 1.88 MPa $m^{0.5}$ for HDPE/EFB$_U$10 to 2.05 MPa $m^{0.5}$ for HDPE/EFB$_{180}$10. Moreover, the highest K_c value of 2.12 MPa $m^{0.5}$ is shown by the HDPE/EFB$_{180}$30 composites. Increase in K_c values may be due to the improved compatibility between EFB and HDPE, leading to better dispersion of the fillers within matrix and resulted in increased stiffness of the composites.

Effect of conditioning

Impact testing was done for composite with a/D 0.2 that undergo indoor exposure and soil burial conditioning.

Indoor conditioning

Fracture energy, W and critical energy release rate, G_c

From Figure 4.65 and Figure 4.66, generally the values of W decrease with the exposure despite the fibre loading. Pure HDPE presents slight decrement value of fracture energy of 150.51 mJ at 1 month indoor exposure while the dry as moulded has value of 179.98 mJ. The value however has a slight increment to 189.64 mJ at 3 months of indoor exposure and decreased to 167.17 mJ at 6 months of exposure. The values of *W* of untreated fibre composites at lower fibre loading (10%) show the same trend with pure HDPE. At 20% fibre loading, the values decreased at 1 month indoor exposure, then increased slightly at 3 and 6 months of exposure. Different with highest fibre loading composites (30%), the value of *W* decreased as the time of indoor exposure increasing linearly and can be said insignificant due to small changes. Treated fibre composites show the same trend of W whereby at lower fibre loading, the trend is inconsistent but with 30% fibre loading, the value of W decreasing linearly with increasing time of exposure. G_c value shows slight changes with same trend for both untreated and treated fibre composites (Figure 4.67 and Figure 4.68). The values of G_c following the trend of W in which the values decreasing with exposure at all fibre loading. This can be concluded that indoor environment causes the W and G_c of the composites to have a slight reduction.

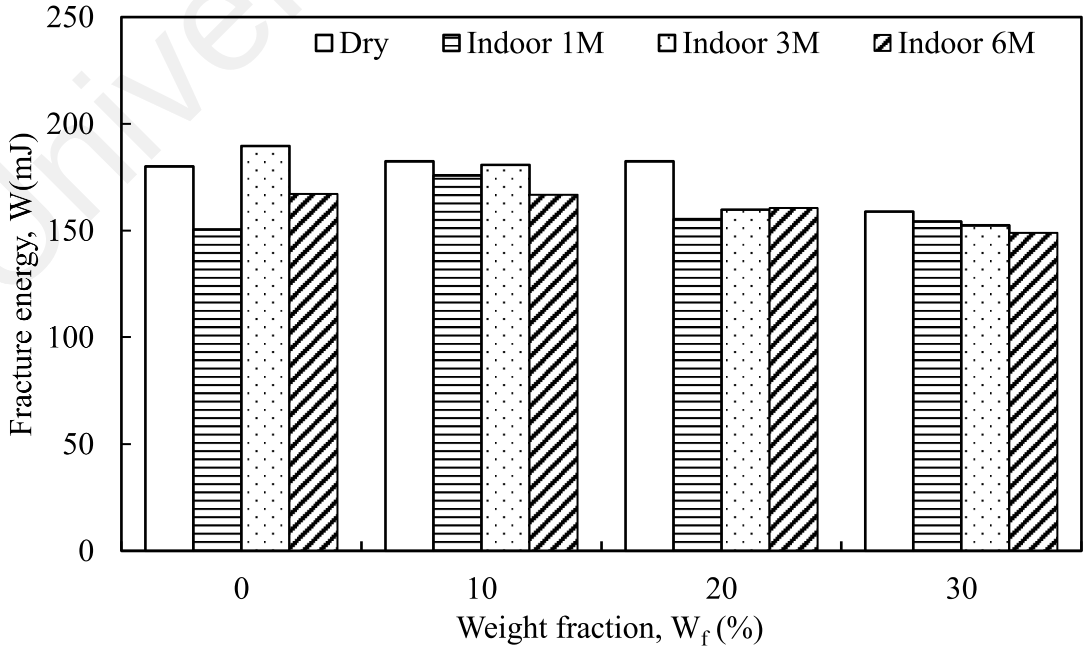


Figure 4.65: Fracture energy of untreated fibre composite under dry and indoor exposure at different time.

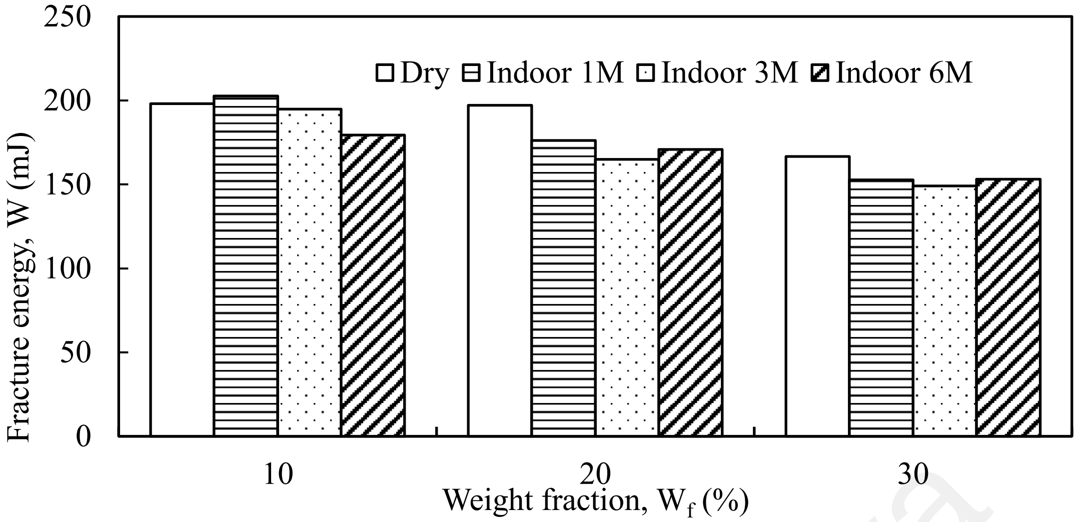


Figure 4.66: Fracture energy of treated fibre composite under dry and indoor exposure at different time.

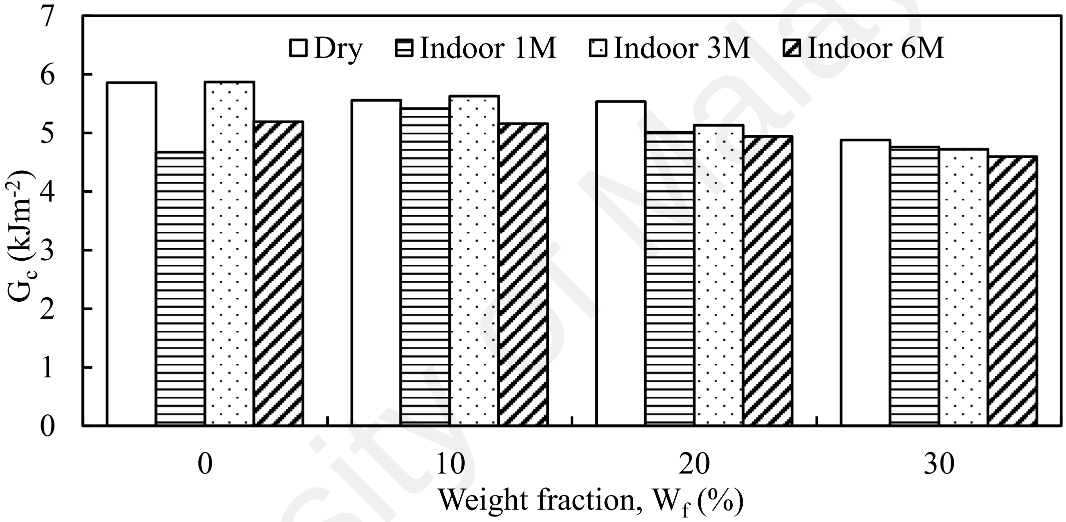


Figure 4.67: G_c of untreated fibre composite under dry and indoor exposure at different time.

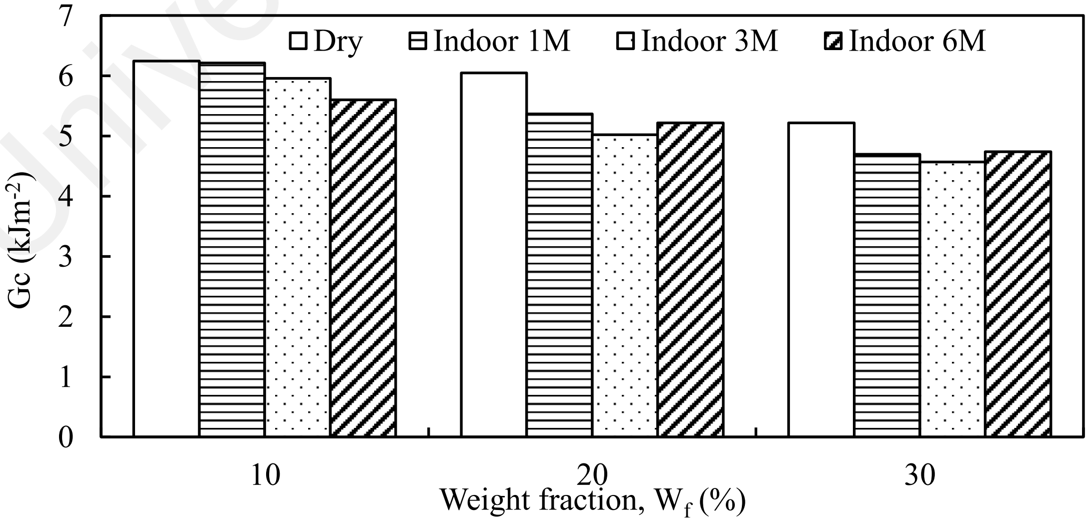


Figure 4.68: G_c of treated fibre composite under dry and indoor exposure at different time.

Peak load, N and critical stress intensity factor, K_c

Figure 4.69 shows the peak load of dry as moulded and indoor exposure of untreated fibre composites at different time. In general, values of P show slight increment with the indoor exposure at all fibre loading. The dry as moulded HDPE presents the peak load values of 264.16 N while for indoor exposed HDPE show values of 270.01 N, 299.34 N and 290.87 N for 1, 3 and 6 months of indoor exposure respectively. This result shows a slight increase in the P of indoor exposed samples relative to the dry as moulded ones. It is not clear what is responsible for this trend because the indoor exposed samples show no visible signs of degradation. On the other hand, treated fibre composites show slight increment without a clear trend in the indoor exposed samples despite the fibre loading and the exposure time compared to the dry as moulded (Figure 4.70). This can be concluded, that there is no variation is observed regard to the heat treatment on EFB fibre. The same trend was observed with the K_c as can be seen in Figure 4.71 and Figure 4.72 for untreated and treated fibre composites respectively. However, treated fibre composites exhibit higher value of K_c compared to untreated fibre composites.

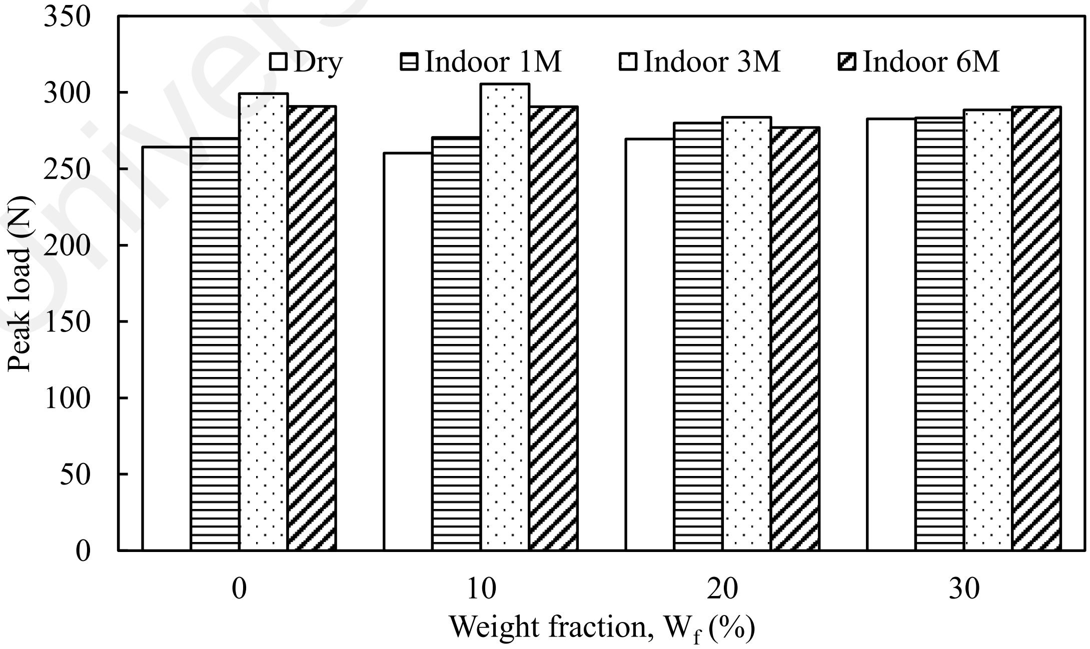


Figure 4.69: Peak load of untreated fibre composite under dry and indoor exposure at different time.

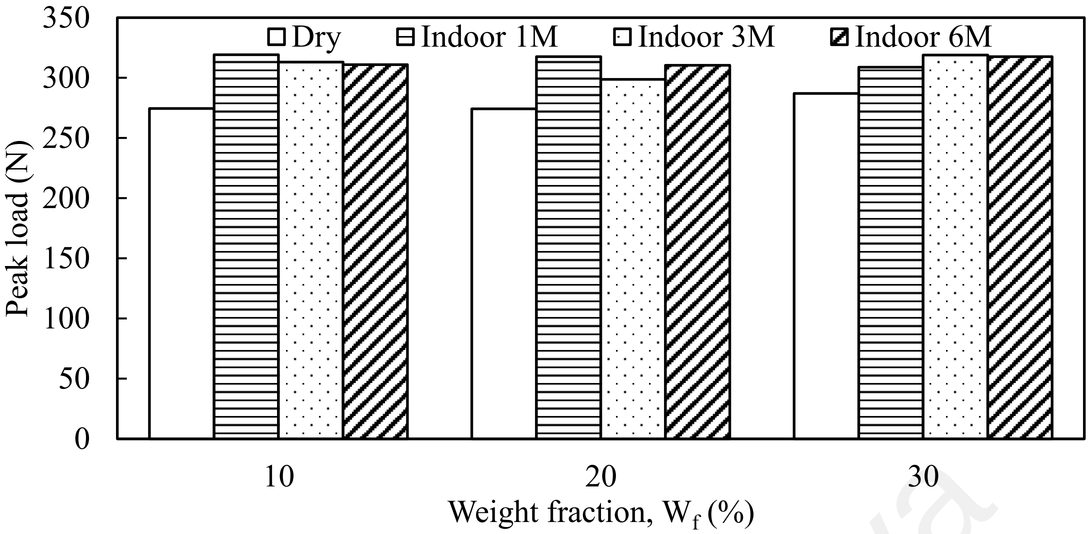


Figure 4.70: Peak load of treated fibre composite under dry and indoor exposure at different time.

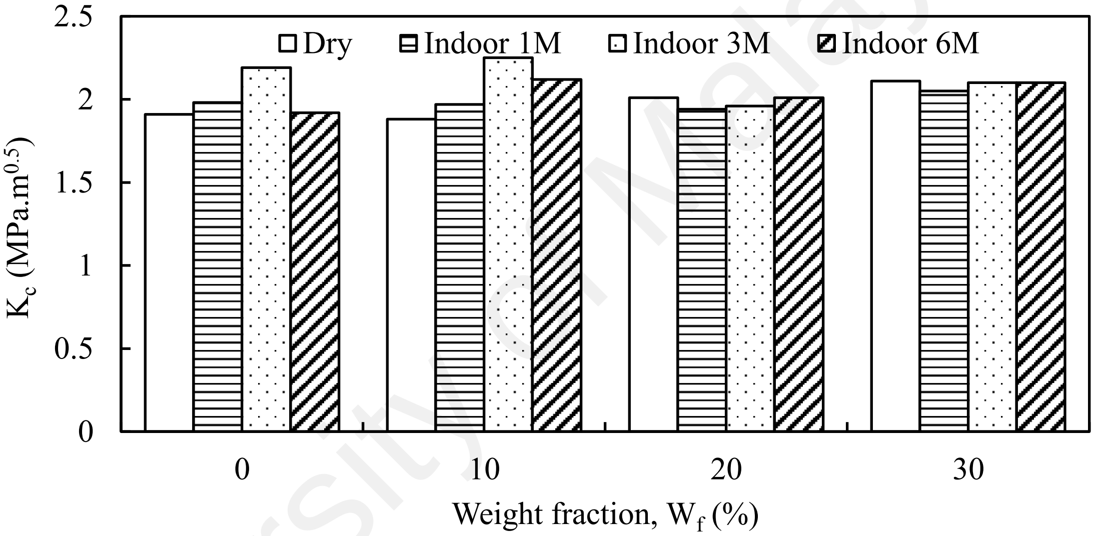


Figure 4.71: K_c of untreated fibre composite under dry and indoor exposure at different time.

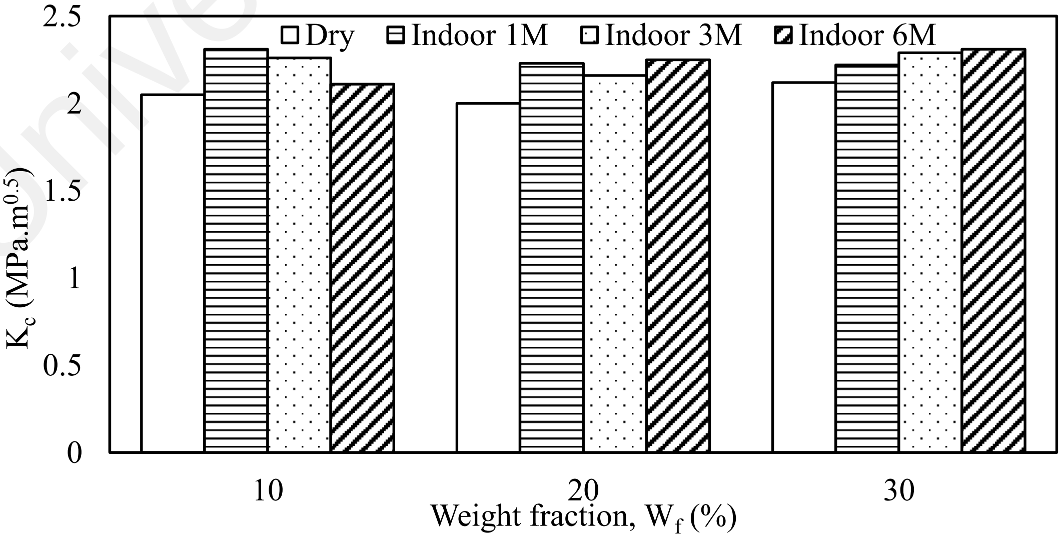


Figure 4.72: K_c of treated fibre composite under dry and indoor exposure at different time.

Soil burial conditioning

Fracture energy, W and critical energy release rate, G_c

Figure 4.73 shows fracture energy of dry as moulded soil burial of HDPE and untreated fibre composites at different time. There is significant trend observed in HDPE and both type of composites after soil burial. The fracture energy of untreated fibre composites decreases after 1 month of soil burial, but increase after 3 and 6 months after the condition at all fibre loading. The dry as moulded HDPE presents the values of 179.98 mJ, followed by 125.70 mJ for 1 month soil burial, and 206.91 mJ and 194.67 mJ for soil burial 3 and 6 months respectively. The decrease in *W* after 1 month of burial may be said that the HDPE is not severely degraded with soil burial. However, at 3 months and 6 months soil burial, the HDPE starts to degrade and show higher values of W following the trend of untreated fibre composites. In addition, untreated fibre composites show a slight increase in W with soil burial despite the fibre loading, especially at lower fibre loading (Figure 4.73). A possible reason for this trend is that even the EFB is degradable, when it is compounded with polymer matrix, the degradability may be lowered. This is because, at lower fibre content, especially at 10% fibre loading, the EFB are covered by the HDPE, thus it is protected from attack by microorganisms. However, as the EFB fibre increases, at 30% fibre loading, wetting effects occurs and the surface of the fibre may be exposed clearly on the surface of the composites and makes the organisms easier to attack. Thus, the response of composites with higher EFB loading to failure in impact properties is expectedly higher than with lower EFB fibre loading. In addition, the same trend was observed with treated fibre composites where the value of *W* is higher with lower EFB loading (Figure 4.74).

The value of G_c is in sequence with the trend observed in the fracture energy. Both untreated and treated fibre composites show the same trend in which the G_c increases after soil burial (Figure 4.75 and Figure 4.76). The results are not expected as the value of Gc expected to decrease with time of exposure to soil burial, same trend with *W* explained above.

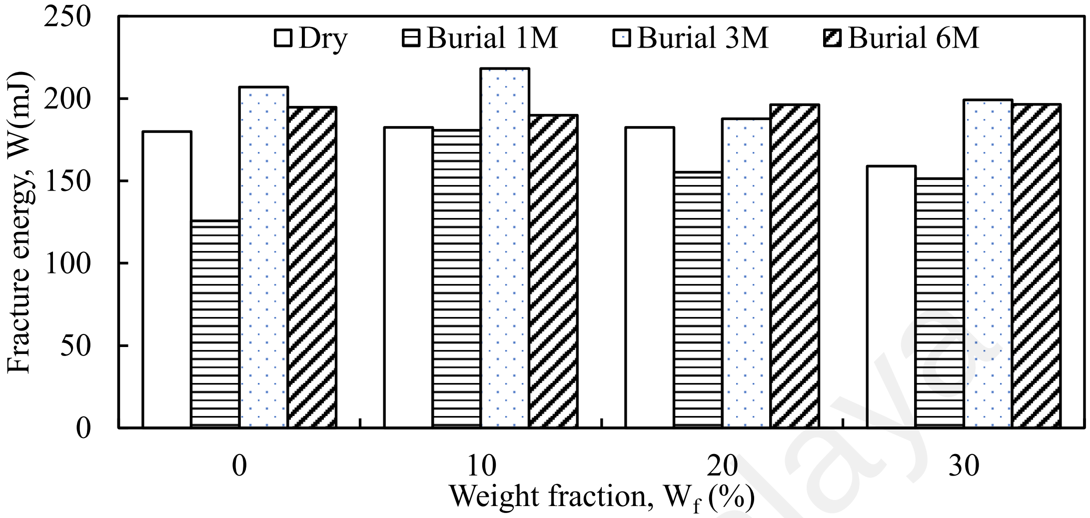


Figure 4.73: Fracture energy of untreated fibre composite under dry and soil burial exposure at different time.

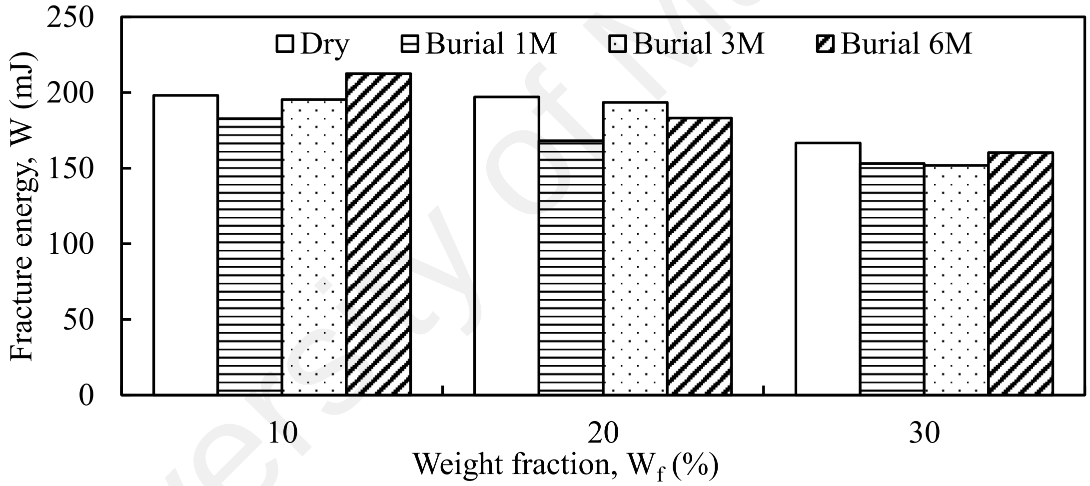


Figure 4.74: Fracture energy of treated fibre composite under dry and soil burial exposure at different time.

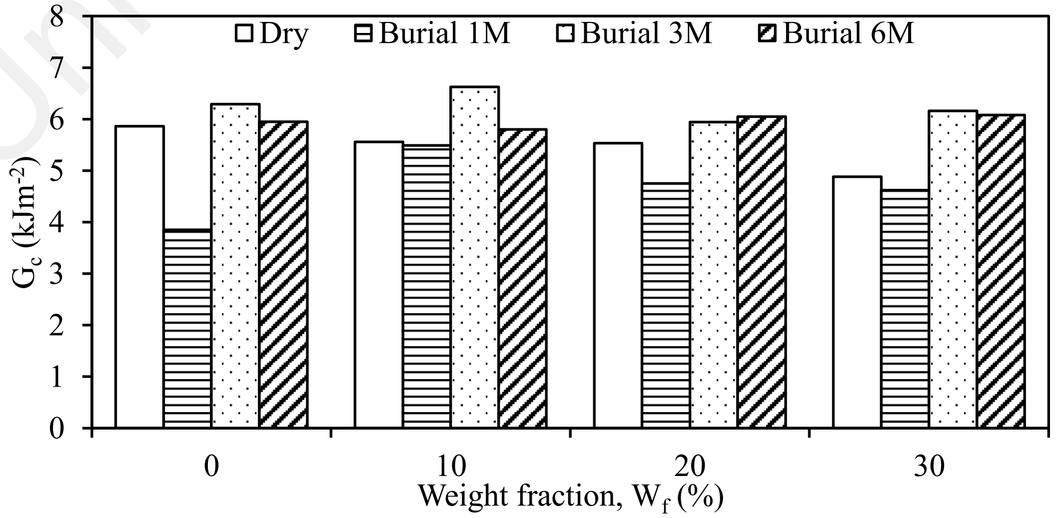


Figure 4.75: G_c of untreated fibre composite under dry and soil burial exposure at different time.

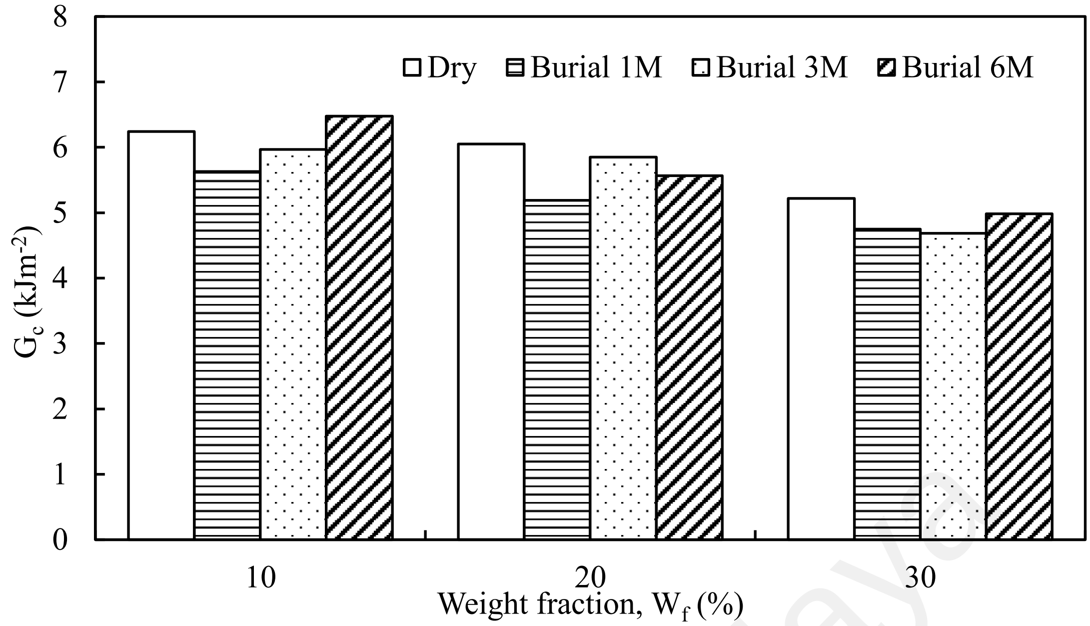


Figure 4.76: Gc treated fibre composite under dry and soil burial exposure at different time.

Peak load, N and critical stress intensity factor, K_c

Based on Figure 4.77, the value of peak load increase for HDPE, 10% and 20% untreated fibre composites at 1 month and 3 months of soil burial. The values however have a reduction after 6 months of soil burial. Unlike the 30% untreated fibre composites, the value of P decreasing with increasing of time of soil burial. This is expected as the composites may undergo ageing and degradation process after exposed to microorganisms in the soil. In treated fibre composites, values of P exhibit same trend as the untreated fibre composites (Figure 4.78). This concluded that the treatment done on the fibre surface did not influence the values of P appreciably in the buried samples compared to untreated fibre composites.

The trend observed for K_c are in agreement with the trend observed in P. K_c values of HDPE slightly decreased after 1 month of soil burial but increased at 3 and 6 months of soil burial. The values ok Kc decrease with increasing of EFB fibre loading for both untreated and treated fibre composites, with treated fibre composites exhibit higher value than untreated fibre composites (Figure 4.79 and Figure 4.80). Both composites (10% and

20%) fibre loading show increment in K_c values while at 30% fibre loading, the values of Kc are decreasing. At 10% and 20% fibre loading, the composites may absorb a lot of water during 1 and 3 months of soil burial. It is possible that, as moisture saturated in the samples, the composite system becomes pliable, thus increasing the K_c (Hobbs, 1998). At 30% fibre composites, the K_c reduced with increasing time of soil burial for both untreated and treated fibre composites. This result is quite relevant as the composites with higher loading expected to degrade faster with the attack of microorganisms towards the fibres.

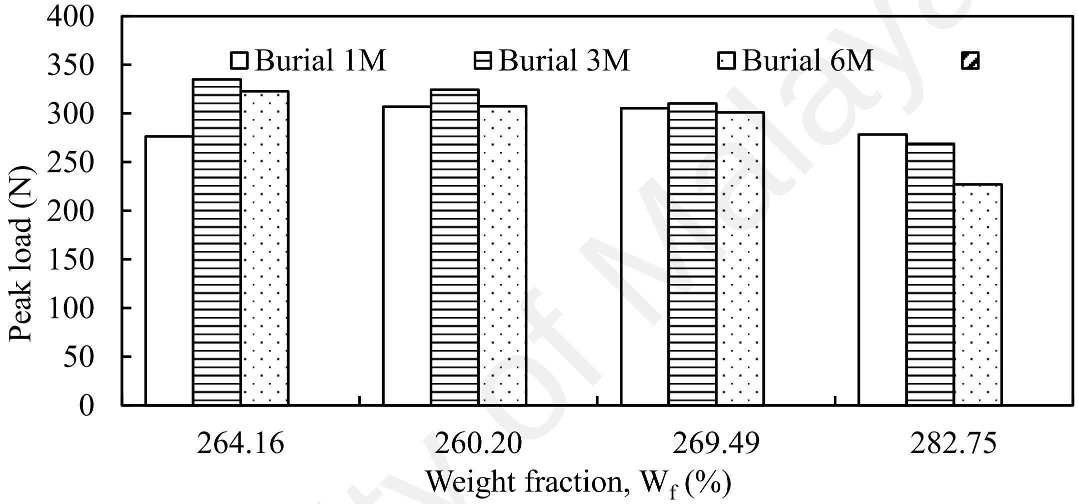


Figure 4.77: Peak load of untreated fibre composite under dry and soil burial exposure at different time.

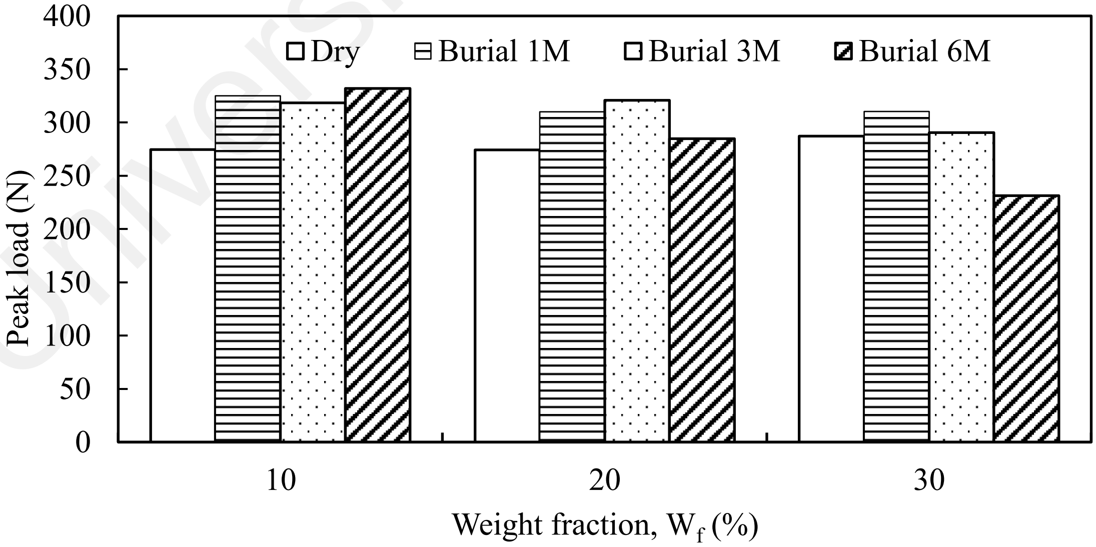


Figure 4.78: Peak load treated fibre composite under dry and soil burial exposure at different time.

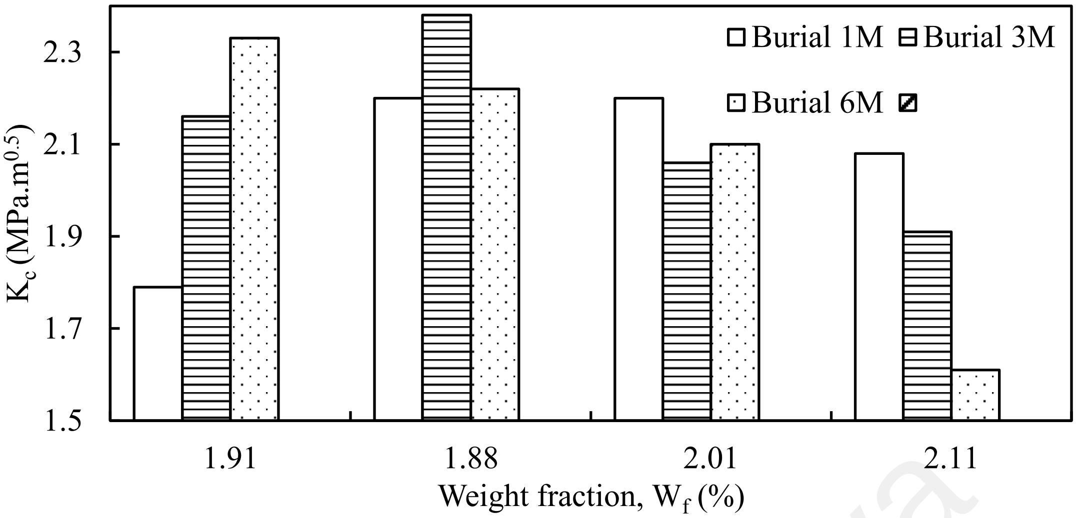


Figure 4.79: K_c of untreated fibre composite under dry and soil burial exposure at different time.

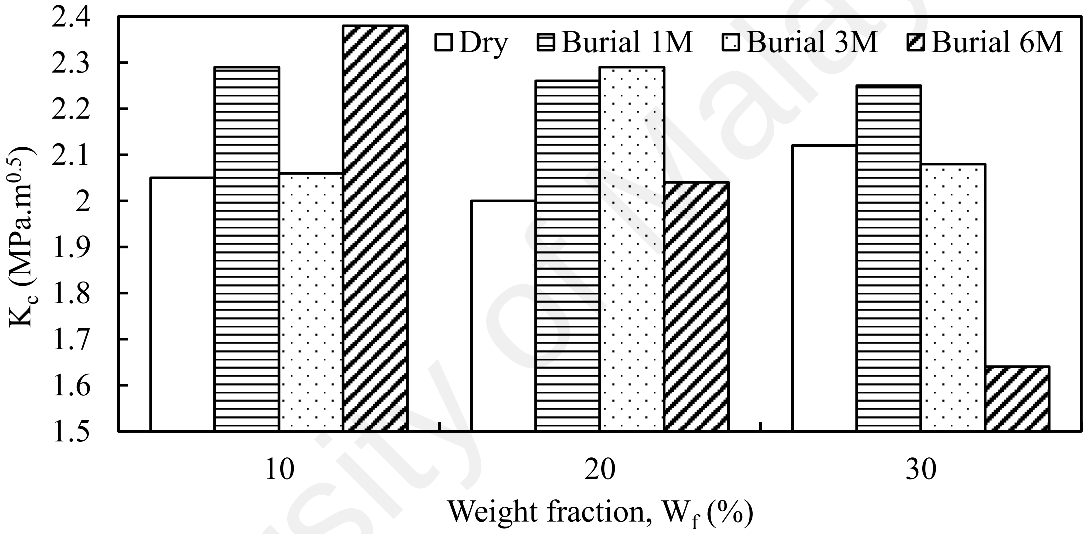


Figure 4.80: K_c treated fibre composite under dry and soil burial exposure at different time.

4.2.4 Effect of water absorption on composites

EFB fibre is a lignocellulosic fibre and the nature of this fibre is hydrophilic and are permeable to water. EFB can absorb water up to 21.39% based on studies done by M. Jawaid et al. (Jawaid et al., 2011). The addition of EFB fibre to the polymeric materials, HDPE increases the water absorption of the composites. The high hydrophilicity of the EFB fibres is due to the availability of free –OH groups in cellulose, hemicellulose and lignin in EFB fibres will form hydrogen bonds with water molecules (Mu et al., 2018). Water absorption will affect the properties of composites through degradation of fibre-matrix interface and structure, resulting in changes of properties of composites, thus creating poor stress transfer efficiencies. The effect of indoor exposure and soil burial conditioning on the absorption of water were studied at both untreated and treated fibre composites.

4.2.4.1 Water absorption behaviour

The hydrophilic nature of EFB fibre remains as disadvantage for its composites when it used in outdoor environment or other related moisture applications. The incorporation of EFB in composites increases the amount of water absorbed. This is well understood and agreed by most researchers is due to the hydrophilic nature of EFB fibre or other natural fibres. Water absorption increase with increasing of fibre loading and this is also expected due to the pore structures of EFB fibre itself, hence has higher ability to retain water (Razak & Kalam, 2012). In this section, the effect of fibre loading and heat treatment were evaluated on the water absorption behaviour in different environmental exposure.

Water uptake, W_t, was calculated using this formula,

$$W_t = \frac{W_2 - W_1}{W_1} X\ 100\ (\%) \qquad (5.0)$$

where W_1 and W_2 are the dry weight and weight after time t of exposure to the different environment respectively. A plot of moisture content against time of exposure for the different EFB fibre loadings of untreated and treated fibre composites is presented in Figure 4.85 – 4.86 for indoor exposure and soil burial respectively.

Indoor conditioning

Based on the Figure 4.81 below, for the pure HDPE show least water absorption over the period of study. This is because of the nature of HDPE, which is hydrophobic, and low water absorption (Mu et al., 2018). For the whole period exposed, the HDPE absorb water up to 0.04% at days 24 of exposure to the condition. However, both untreated and treated fibre composites absorb water to various extend with composite having 30%

untreated fibre show the highest water absorption up to 1.11% after 108 days of exposure. As the HDPE is hydrophobic, the water uptake of both types of composites may be attributed to the availability of free –OH groups in cellulose, hemicellulose and lignin in EFB fibres that will form hydrogen bonds with water molecules (Azeh et al., 2012; Mu et al., 2018). From the graph, water absorption of the composites increased gradually with the progression time, until they approached a saturation level, where the amount of water absorbed by the composites is almost the same. As noticed from the graph, composites with higher EFB content shows more water absorption. One of the reasons to explain this situation was the compatibility between fibre and matrix, which can affect the water absorption. With the increase of EFB fibre loading, the polar character of the fibre increases, resulting in higher water uptake (Feng et al., 2014; Kartal et al., 2013). In addition, the amount of hydrophilic –OH in the lignocellulosic fibres can also be the reason of the higher water uptake in the high fibre loading composites (Feng et al., 2014; Tajvidi & Ebrahimi, 2003). The same trend was observed with treated fibre composites where 30% treated fibre composites exhibit highest value of 1.01% after exposure to indoor conditioning for 108 days. The formation of lump increases as the EFB fibre increases due to the difficulties of having a homogeneous dispersion of filler at a high filler content. The agglomeration of the filler in composites increases the water absorption of the composites (Khalil et al., 2010). The decreased percentage in the water absorption in treated fibre composites may be due to the improvement of adhesion between fibre and matrix by providing strong bonding, and hence further hindering the water diffusion through the composites (Ismail & Ishak, 2018).

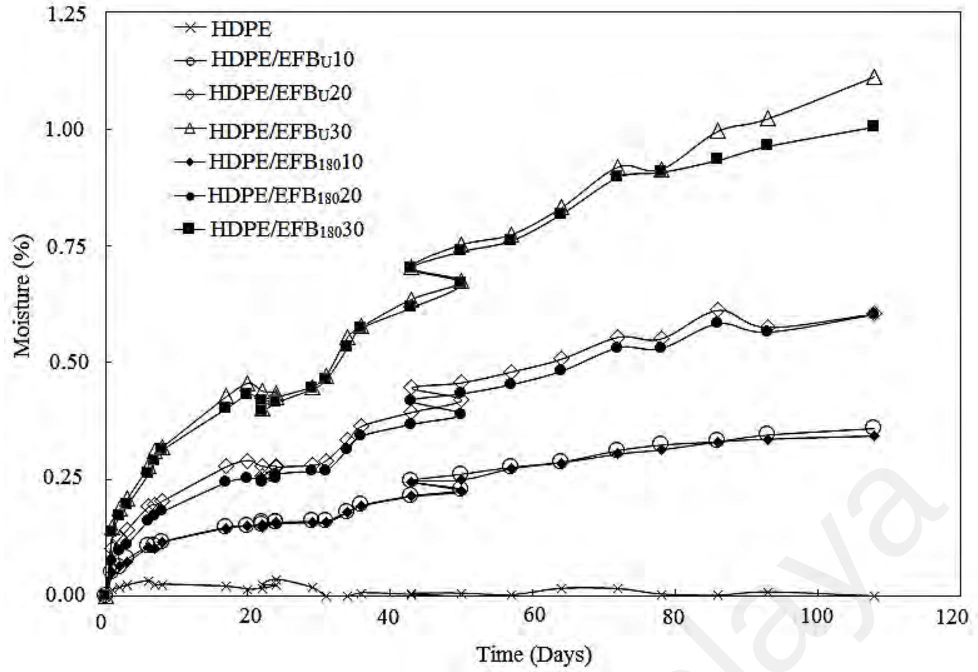


Figure 4.81: Moisture absorption of composites as a function of EFB fibre content and heat treatment of indoor exposure.

Soil burial conditioning

The effect of HDPE and its composites on the water absorption for soil burial environment over the period of study is shown in Figure 4.82 below. From the figures below, it was observed that HDPE and the composites showed a higher degree of water absorption compared to the indoor exposure samples. It can be observed that the water uptake increases as the time of soil burial increasing due to high amount of water allowed to be absorbed by the materials (Jumaidin et al., 2017). Comparing with the indoor exposure samples, the soil burial samples absorb more water due to environment as reported in the weathering data before, whereas the indoor exposure samples only exposed to the moisture. The trend however is the same as the indoor exposure samples where the highest fibre loading absorbs more water than the lower fibre loading composites. As stated before, the treated fibre composites exhibit lower absorption of water due to the strong bonding between fibre and matrix. This could also be attributed to the removal of impurities, water sensitive components, hemicellulose, in EFB fibre by the heat treatment, thereby rendering the composites more water resistant. Moreover, free hydroxyl groups of the cellulose are reduced with heat treatment which could result into a decrease in water absorption of the EFB fibre in the composites.

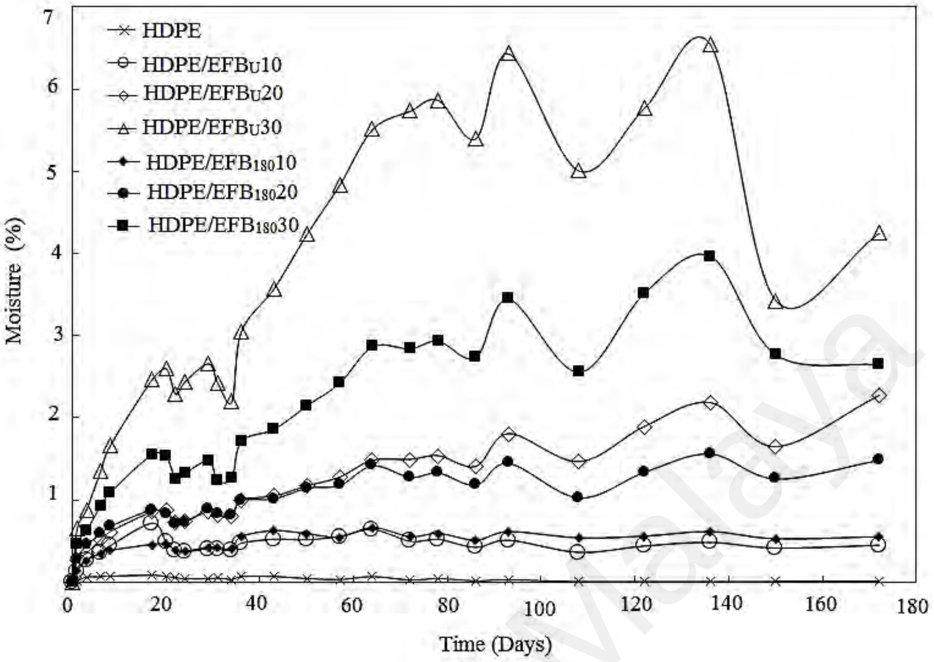


Figure 4.82: Moisture absorption of composites as a function of EFB fibre content and heat treatment of soil burial.

CHAPTER 5:CONCLUSIONS

5.1 Conclusions

Heat treated oil palm empty fruit bunch/high density polyethylene composites were successfully prepared and characterised on several characterisations mentioned in this study. From this research, it can be concluded that the heat treatment on EFB fibre has given positive effect, which reduced the hydrophilicity, and it is more compatible with a non-polar matrix, HDPE, resulting in better interfacial adhesion between HDPE and EFB fibre, leading to improvement in their properties. The effect of heat treatment on the thermal and mechanical properties of composites has been studied and several conclusions have been made:

i. From the SEM image of surface of EFB, the silica is removed, and rougher surface of the fibre is observed after the fibre treatment at 180°C, leading to better adhesion of HDPE and EFB. The optimum condition for the heat treatment of EFB was chosen at 180°C, 1 hour.

ii. As the fibre loading increased, the thermal stability of the composites decreases due to the lower thermal stability of the lignocellulosic filler comparing to pure HDPE.

iii. Based on the information from TGA and SEM the thermal stability of the EFB fibre increased after the heat treatment due to the removal of the hemicellulose, lignin, ash and other impurities during the heat treatment.

iv. The melting temperature was not significantly affected by the heat treatment. However, the degree of crystallinity increases slightly after heat treatment on EFB.

v. The EFB heat treatment shows improvement in tensile strength compared to the untreated fibre composites at lower fibre loading. Moreover, the value of tensile modulus increases linearly with increase of lignocellulosic fibre indicating that there is a good HDPE-EFB adhesion.

vi. Flexural strength and modulus increasing with fibre treatment and EFB loading, regardless the fibre proportions. Flexural displacement however did not show any significant effect to the composites.

vii. Impact properties of heat treated composites show improvement in which the value of G_c and K_c increase relatively.

viii. Soil burial conditioning shows that the composites have undergo some degradation due to reduction on the mechanical properties which can be concluded from the attack of microorganisms during the conditioning period. However, the treated fibre composites showed more resistance towards the environment.

ix. Indoor exposure has no detrimental effect on thermal and mechanical properties of composites; thus, it can be used to produce products in domestic applications.

x. Treated fibre composites absorb less moisture compared to untreated fibre composites, regardless the exposure condition, due to the improvement of adhesion between fibre and matrix by providing strong bonding, and hence further hindering the water diffusion through the composites.

xi. Treated fibre composites exhibit higher value of MFI compared to the untreated fibre composites, indicated that treatment improve the compatibility between EFB fibre and HDPE, thus decreased the viscosity of composites and enhanced the flow ability.

5.2 Suggestions for future studies

Based on the findings in the studies, there are several suggestions can be made for future studies. First, the interfacial adhesion between fibre and matrix can be improved by introducing coupling agents or compatibiliser such as Maleic anhydride polyethylene (MAPE) or through double treatment of the fibre surface.

In addition, hybridisation of fibres can be made to prepare the composites. For examples, EFB fibres can be hybrid with other natural fibres such jute, hemp, bast and kenaf fibre to improve the properties of the composites.

Lastly, the conditioning time may need to be extended to have better understanding of the degree of the material's response to environmental degradation. For example, in this research, the period was set up to six months exposure but in future can be set up to 12 months. In addition, other exposure which is outdoor conditioning may be added to understand the composite's behaviour towards it as well. For conditioning exposure, colour spectrometry test may need to be analysed to study the effect of colouration towards the degradation due to the exposure.

REFERENCES

Abdelmouleh, M., Boufi, S., Belgacem, M. N., & Dufresne, A. (2007). Short natural-fibre reinforced polyethylene and natural rubber composites: Effect of silane coupling agents and fibres loading. *Composites Science and Technology, 67*(7-8), 1627-1639.

Abdul Khalil, H., Poh, B., Jawaid, M., Ridzuan, R., Suriana, R., Said, M. R. & Nik Fuad, N. A. (2010). The effect of soil burial degradation of oil palm trunk fiber-filled recycled polypropylene composites. *Journal of Reinforced Plastics and Composites, 29*(11), 1653-1663.

Abdul Khalil, H., Siti Alwani, M., Ridzuan, R., Kamarudin, H., & Khairul, A. (2008). Chemical composition, morphological characteristics, and cell wall structure of Malaysian oil palm fibers. *Polymer-Plastics Technology and Engineering, 47*(3), 273-280.

Abu-Sharkh, B., & Hamid, H. (2004). Degradation study of date palm fibre/polypropylene composites in natural and artificial weathering: Mechanical and thermal analysis. *Polymer Degradation and Stability, 85*(3), 967-973.

Ahad, N. A., Parimin, N., Mahmed, N., Ibrahim, S. S., Nizzam, K., & Ho, Y. M. (2009). Effect of chemical treatment on the surface of natural fiber. *Journal of Nuclear and Related Technologies, 6*(1), 155-158.

Alam, M., Beg, M., Mina, M., Mamun, A., & Bledzki, A. (2015). Degradation and stability of green composites fabricated from oil palm empty fruit bunch fiber and polylactic acid: Effect of fiber length. *Journal of Composite Materials, 49*(25), 3103-3114.

Alcock, M., Ahmed, S., DuCharme, S., & Ulven, C. (2018). Influence of stem diameter on fiber diameter and the mechanical properties of technical flax fibers from linseed flax. *Fibers, 6*(1), 1-16.

Alomayri, T., Assaedi, H., Shaikh, F., & Low, I. M. (2014). Effect of water absorption on the mechanical properties of cotton fabric-reinforced geopolymer composites. *Journal of Asian Ceramic Societies, 2*(3), 223-230.

Alshabanat, M. (2019). Morphological, thermal, and biodegradation properties of LLDPE/treated date palm waste composite buried in a soil environment. *Journal of Saudi Chemical Society, 23*(3), 355-364.

Amir, N., Abidin, K. A. Z., & Shiri, F. B. M. (2017). Effects of fibre configuration on mechanical properties of banana fibre/PP/MAPP natural fibre reinforced polymer composite. *Procedia Engineering, 184*, 573-580.

Anuar, N., Zakaria, S., Harun, J., & Wang, C. (2017). Kenaf/PP and EFB/PP: Effect of fibre loading on the mechanical properties of polypropylene composites.*Paper presented at the IOP Conference Series: Materials Science and Engineering*, *217*(1), 1-10.

Anuar, N. I. S., Zakaria, S., Kaco, H., Hua, C. C., Chunhong, W., & Abdullah, H. S. (2018). Physico-mechanical, chemical composition, thermal degradation and crystallinity of oil palm empty fruit bunch, kenaf and polypropylene fibres: A comparatives study. *Sains Malaysiana, 47*(4), 839-851.

Araujo, J., Waldman, W., & De Paoli, M. (2008). Thermal properties of high density polyethylene composites with natural fibres: Coupling agent effect. *Polymer Degradation and Stability, 93*(10), 1770-1775.

Arrakhiz, F., El Achaby, M., Malha, M., Bensalah, M., Fassi-Fehri, O., & Bouhfid, R. (2013). Mechanical and thermal properties of natural fibers reinforced polymer composites: Doum/low density polyethylene. *Materials & Design, 43*, 200-205.

Arrigo, R., Morici, E., & Dintcheva, N. T. (2017). High-performance thermoplastic elastomers/carbon nanotubes nanocomposites: Mechanical behavior, rheology, and durability. *Polymer Composite, 38*, 381-391.

Arutchelvi, J., Muniyasamy, S., Arkatkar, A., Doble, M., Bhaduri, S., & Veera Uppara, P. (2008). Biodegradation of polyethylene and polypropylene. *Indian Journal of Biotechnology, 7*, 9-22.

Asim, M., Jawaid, M., Abdan, K., & Nasir, M. (2018). Effect of Alkali treatments on physical and Mechanical strength of Pineapple leaf fibres.*Paper presented at the IOP Conference Series: Materials Science and Engineering, 290*(1),1-6.

Astimar, A. A., Anis, M., & Mohamad, H. (2002). Preparation of cellulose from oil palm empty fruit bunches via ethanol digestion: Effect of acid and alkali catalysts. *Journal of Oil Palm Research, 14(1*), 9-14.

ASTM, ASTM C1557-14, *Standard Test Method for Tensile Strength and Young's Modulus of Fibers.* (2014). ASTM International. West Conshohocken, PA.

ASTM, ASTM D638-*10 in Standard Test Method for Tensile Properties of Plastics.* (2010). ASTM International West Conshohocken, PA.

ASTM, ASTM D790-10 *in Standard Test Methods for Flexural Properties of Unreinforced and Reinforced Plastics and Electrical Insulating Materials.* (2010). ASTM International. West Conshohocken, PA.

ASTM, ASTM D1238-13, *Standard Test Method for Melt Flow Rates of Thermoplastics by Extrusion Plastometer.* (2013). ASTM International. West Conshohocken, PA.

ASTM, ASTM E23-07, *in Standard Test Methods for Notched Bar Impact Testing of Metallic Materials.* (2007). ASTM international. West Conshohocken, PA.

ASTM, ASTM E1755-01, *Standard Test Method for Ash in Biomass.* (2015). ASTM International. West Conshohocken, PA.

Azeh, Y., Olatunji, G. A., & Mamza, P. A. (2012). Scanning electron microscopy and kinetic studies of ketene-acetylated wood/cellulose high-density polyethylene blends. *International Journal of Carbohydrate Chemistry, 2012*, 1-7.

Azwa, Z., Yousif, B., Manalo, A., & Karunasena, W. (2013). A review on the degradability of polymeric composites based on natural fibres. *Materials & Design, 47*, 424-442.

Bajsić, E. G., Rek, V., Bulatović, V. O., & Pavić, B. O. (2012). *Effect of filler treatment on the thermal properties of composites and polymer blends.* Paper presented at the ECCM 15-15th European conference on composite materials, 24th – 28th June 2012, Venice, Italy.

Bakar, A. A., Hassan, A., & Yusof, A. M. (2005a). Effect of oil palm empty fruit bunch and acrylic impact modifier on mechanical properties and processability of unplasticized poly (vinyl chloride) composites. *Polymer-Plastic Technology and Engineering, 44*(6), 1125-1137.

Bakar, A. A., Hassan, A., & Yusof, A. M. (2005b). Mechanical and thermal properties of oil palm empty fruit bunch-filled unplasticized poly (vinyl chloride) composites. *Polymers & Polymer Composites, 13*(6), 607-617.

Beckermann, G., & Pickering, K. L. (2008). Engineering and evaluation of hemp fibre reinforced polypropylene composites: fibre treatment and matrix modification. *Composites Part A: Applied Science and Manufacturing, 39*(6), 979-988.

Bielawski, R. (2017). Composite materials in military aviation and selected problems with implementation. *Review of the Air Force Academy, 1*(33), 11-16.

Bledzki, A. K., & Faruk, O. (2004). Extrusion and injection moulded microcellular wood fibre reinforced polypropylene composites. *Cellular Polymers, 23*(4), 211-227.

Bos, H. L. (2004). *The potential of flax fibres as reinforcement for composite materials*. Netherlands, Technische Universiteit Eindhoven Eindhoven.

Chandramohan, D., & Marimuthu, K. (2011). A review on natural fibers. *International Journal of Research and Reviews in Applied Sciences, 8*(2), 194-206.

Chang, S. H. (2014). An overview of empty fruit bunch from oil palm as feedstock for bio-oil production. *Biomass and Bioenergy, 62*, 174-181.

Ciullo, P. A., & Hewitt, N. (1996). Industrial minerals and their uses: A handbook and formulary, Peter A. Ciullo (Ed.), Noyes Publications, 1996. In P.A. Ciullo & N. Hewitt (Eds.), The Rubber Formulary (pp. 55-72). Norwich, New York: William Andrew Publishing.

Cruz, J., & Fangueiro, R. (2016). Surface modification of natural fibers: A review. *Procedia Engineering, 155*, 285-288.

Dash, B., Rana, A., Mishra, S., Mishra, H., Nayak, S., & Tripathy, S. (2000). Novel low-cost jute–polyester composite. II. SEM observation of the fractured surfaces. *Polymer-Plastics Technology and Engineering, 39*(2), 333-350.

Dashtban, M., Schraft, H., Syed, T. A., & Qin, W. (2010). Fungal biodegradation and enzymatic modification of lignin. *International Journal of Biochemistry and Molecular Biology, 1*(1), 36-50.

Davindrabrabu, M., Januar, P. S., Dandi, B., Ruzaimi, M., Rejab, M., & Cionita, T. (2015). Effect of fibre loading on the flexural properties of natural fibre reinforced polymer composites. *Paper presented at the Applied Mechanics and Materials, 695,* 85-88.

Dittenber, D. B., & GangaRao, H. V. S. (2012). Critical review of recent publications on use of natural composites in infrastructure. *Composites Part A: Applied Science and Manufacturing, 43*(8), 1419-1429.

Drzal, L., & Madhukar, M. (1993). Fibre-matrix adhesion and its relationship to composite mechanical properties. *Journal of Materials Science, 28*(3), 569-610.

Dunne, R., Desai, D., Sadiku, R., & Jayaramudu, J. (2016). A review of natural fibres, their sustainability and automotive applications. *Journal of Reinforced Plastics and Composites, 35*(13), 1041-1050.

El-Shekeil, Y., Sapuan, S., Jawaid, M., & Al-Shuja'a, O. (2014). Influence of fiber content on mechanical, morphological and thermal properties of kenaf fibers reinforced poly (vinyl chloride)/thermoplastic polyurethane poly-blend composites. *Materials & Design, 58*, 130-135.

Eroglu, M. (2007). Effect of talc and heat treatment on the properties of polypropylene/EVA composite. *International Journal of Science & Technology, 2*(1), 63-73.

Etaati, A., Pather, S., Rahman, M., & Wang, H. (2015). Ground hemp fibers as filler/reinforcement for thermoplastic biocomposites. *Advances in Materials Science and Engineering, 2015*, 1-11.

Fangueiro, R. (2011). *Fibrous and composite materials for civil engineering applications*. United Kingdom: Elsevier.

Feng, J., Shi, Q., Chen, Y., & Huang, X. (2014). Mold resistance and water absorption of Wood/HDPE and Bamboo/HDPE composites. *Journal of Applied Sciences, 14*(8), 776-783.

François, D., Pineau, A., & Zaoui, A. (2013). *Ductile-Brittle Transition. In Mechanical Behaviour of Materials*. Dordrecht: Springer.

Galav, L., Mukhopadhyay, S., & Deopura, B. (2019). Effect of compatibilizer on mechanical properties of chemically treated coir/polypropylene composite. *Indian Journal of Fibre & Textile Research, 44,* 155-162.

Gamon, G., Evon, P., & Rigal, L. (2013). Twin-screw extrusion impact on natural fibre morphology and material properties in poly (lactic acid) based biocomposites. *Industrial Crops and Products, 46*, 173-185.

Garcia-Rejon, A., Meddad, A., Turcott, E., & Carmel, M. (2002). Extrusion blow molding of long fiber reinforced polyolefins. *Polymer Engineering & Science, 42*(2), 346-364.

Goh, C. S., Tan, K. T., Lee, K. T., & Bhatia, S. (2010). Bio-ethanol from lignocellulose: Status, perspectives and challenges in Malaysia. *Bioresource Technology, 101*(13), 4834-4841.

Graupner, N., & Müssig, J. (2010). *Technical applications of natural fibres: An overview. Industrial applications of natural fibres: Structure, properties and technical applications*. Germany: Wiley.

Gupta, H., Kanaujia, K. K., Rizvi, S. M. A., & Shukla, R. (2019). A Review on the mechanical properties of natural fibre reinforced polypropylene composites. *International Research Journal of Engineering and Technology (IRJET), 6*(6), 339-342.

Gurunathan, T., Mohanty, S., & Nayak, S. K. (2015). A review of the recent developments in biocomposites based on natural fibres and their application perspectives. *Composites Part A: Applied Science and Manufacturing, 77*, 1-25.

Haines, P. J. (2012*). Thermal methods of analysis: principles, applications and problems.* Scotland: Springer.

Hanan, F., Jawaid, M., & Paridah, M. (2018). Oil palm EFB/kenaf fibre reinforced epoxy hybrid composites: dimension stability behaviours. *Paper presented at the IOP Conference Series: Materials Science and Engineering, 368*(1),1-8.

Hassan, A., Hassan, A. A., & Rafiq, M. (2011). Impact properties of injection molded glass fiber/polyamide-6 composites: Effect of testing parameters. *Journal of Reinforced Plastics and Composites, 30*(10), 889-898.

Hassan, A., Rahman, N. A., & Yahya, R. (2011). Extrusion and injection-molding of glass fiber/MAPP/polypropylene: Effect of coupling agent on DSC, DMA, and mechanical properties. *Journal of Reinforced Plastics and Composites, 30*(14), 1223-1232.

Hassan, A., Rahman, N. A., & Yahya, R. (2012). Moisture absorption effect on thermal, dynamic mechanical and mechanical properties of injection-molded short glass-fiber/polyamide 6, 6 composites. *Fibers and Polymers, 13*(7), 899-906.

Hassan, A., Salema, A. A., Ani, F. N., & Bakar, A. A. (2010). A review on oil palm empty fruit bunch fiber-reinforced polymer composite materials. *Polymer Composites, 31*(12), 2079-2101.

Hassan, A., Yahya, R., Rafiq, M., Hussin, A., Sheikh, M., & Hornsby, P. (2011). Interfacial shear strength and tensile properties of injection-molded, short-and long-glass fiber-reinforced polyamide 6, 6 composites. *Journal of Reinforced Plastics and Composites, 30*(14), 1233-1242.

Hassan, N. S., & Badri, H. (2016). Thermal behaviors of oil palm empty fruit bunch fiber upon exposure to acid-base aqueous solutions. *Malaysian Journal of Analytical Sciences, 20*(5), 1095-1103.

Herrera-Franco, P., & Valadez-Gonzalez, A. (2004). Mechanical properties of continuous natural fibre-reinforced polymer composites. *Composites Part A: Applied Science and Manufacturing, 35*(3), 339-345.

Hobbs, J. (1998). The fracture of poly (hydroxybutyrate) Part I Fracture mechanics study during ageing. *Journal of Materials Science, 33*(10), 2509-2514.

Hughes, M., Hill, C., & Hague, J. (2002). The fracture toughness of bast fibre reinforced polyester composites Part 1 Evaluation and analysis. *Journal of Materials Science, 37*(21), 4669-4676.

Ibrahim, Z., Abdul Aziz, A., Ramli, R., Jusoff, K., Ahmad, M., & Jamaludin, M. A. (2015). Effect of treatment on the oil content and surface morphology of oil palm (Elaeis guineensis) empty fruit bunches (EFB) fibres. *Wood Research, 60*(1), 157-166.

Ilyas, R., Sapuan, S., Norizan, M. N., Atikah, M., Huzaifah, M., & Radzi, A. (2019). *Potential of natural fibre composites for transport industry: A review Paper.* Paper presented at Prosiding Seminar Enau Kebangsaan, 1 April 2019, Negeri Sembilan, Malaysia.

Ishak, M., Leman, Z., Sapuan, S. a., Edeerozey, A., & Othman, I. (2010). Mechanical properties of kenaf bast and core fibre reinforced unsaturated polyester composites. *Paper presented at the IOP Conference Series: Materials Science and Engineering, 11*(1), 1-7.

Ishak, Z. M., Ariffin, A., & Senawi, R. (2001). Effects of hygrothermal aging and a silane coupling agent on the tensile properties of injection molded short glass fiber reinforced poly (butylene terephthalate) composites. *European Polymer Journal, 37*(8), 1635-1647.

Ismail, N., & Ishak, Z. (2018*).* Effect of fiber loading on mechanical and water absorption capacity of Polylactic acid/Polyhydroxybutyrate-co-hydroxyhexanoate/Kenaf composite. *Paper presented at the IOP Conference Series: Materials Science and Engineering, 368*(1), 1-12.

Izani, M. N., Paridah, M., Anwar, U., Nor, M. M., & H'ng, P. (2013). Effects of fiber treatment on morphology, tensile and thermogravimetric analysis of oil palm empty fruit bunches fibers. *Composites Part B: Engineering, 45*(1), 1251-1257.

Jawaid, M., Abdul Khalil, H., Bhat, A., & Abu Baker, A. (2011). Impact properties of natural fiber hybrid reinforced epoxy composites. *Paper presented at the Advanced Materials Research, 264,* 688-693.

Jawaid, M., & Khalil, H. A. (2011). Cellulosic/synthetic fibre reinforced polymer hybrid composites: A review. *Carbohydrate Polymers, 86*(1), 1-18.

Jawaid, M., Khalil, H. A., Khanam, P. N., & Bakar, A. A. (2011). Hybrid composites made from oil palm empty fruit bunches/jute fibres: Water absorption, thickness swelling and density behaviours. *Journal of Polymers and the Environment, 19*(1), 106-109.

Jayamani, E., Hamdan, S., Rahman, M. R., & Bakri, M. K. B. (2014). Investigation of fiber surface treatment on mechanical, acoustical and thermal properties of betelnut fiber polyester composites. *Procedia Engineering, 97*, 545-554.

Jiang, Z., Yang, Y., Mo, S., Yao, K., & Gao, F. (2012). Polymer extrusion: From control system design to product quality. *Industrial & Engineering Chemistry Research, 51*(45), 14759-14770.

Joseph, P., Joseph, K., Thomas, S., Pillai, C., Prasad, V., & Groeninckx, G. (2003). The thermal and crystallisation studies of short sisal fibre reinforced polypropylene composites. *Composites Part A: Applied Science and Manufacturing, 34*(3), 253-266.

Joseph, P., Rabello, M. S., Mattoso, L., Joseph, K., & Thomas, S. (2002). Environmental effects on the degradation behaviour of sisal fibre reinforced polypropylene composites. *Composites Science and Technology, 62*(10-11), 1357-1372.

Joshia, S. V., Drzalb, L. T., Mohantyb, A. K., & Arorac, S. (2004). Are natural fiber composites environmentally superior to glass fiber reinforced composites?. *Composites Part A : Applied Science and Manufacturing, 35*, 371-376.

Jumaidin, R., Sapuan, S. M., Jawaid, M., Ishak, M. R., & Sahari, J. (2017). Thermal, mechanical, and physical properties of seaweed/sugar palm fibre reinforced thermoplastic sugar palm starch/agar hybrid composites. *International Journal of Biological Macromolecules, 97*, 606-615.

Kabir, M., Wang, H., Aravinthan, T., Cardona, F., & Lau, K.-T. (2011). Effects of natural fibre surface on composite properties: A review. *Paper presented at the Proceedings of the 1st international postgraduate conference on engineering, designing and developing the built environment for sustainable wellbeing (eddBE2011),* 94-99.

Kabir, M. M., Islam, M. M., & Wang, H. (2013). Mechanical and thermal properties of jute fibre reinforced composites. *Journal of Multifunctional Composites, 1*(1), 71-77.

Kaboorani, A. (2009). Thermal properties of composites made of heat-treated wood and polypropylene. *Journal of Composite Materials, 43*(22), 2599-2607.

Kaboorani, A., Faezipour, M., & Ebrahim, G. (2008). Feasibility of using heat treated wood in wood/thermoplastic composites. *Journal of Reinforced Plastics and Composites, 27*(16), 1689-1699.

Kalam, A., Sahari, B., Khalid, Y., & Wong, S. (2005). Fatigue behaviour of oil palm fruit bunch fibre/epoxy and carbon fibre/epoxy composites. *Composite Structures, 71*(1), 34-44.

Karina, M., Onggo, H., Abdullah, A. D., & Syampurwadi, A. (2008). Effect of oil palm empty fruit bunch fiber on the physical and mechanical properties of fiber glass reinforced polyester resin. *Journal of Biological Sciences, 8*(1), 101-106.

Kartal, S. N., Aysal, S., Terzi, E., Yılgör, N., Yoshimura, T., & Tsunoda, K. (2013). Wood and bamboo-PP composites: Fungal and termite resistance, water absorption, and FT-IR analyses. *Bioresources, 8*(1), 1222-1244.

Khalid, M., Ratnam, C., Chuah, T., Ali, S., & Choong, T. S. (2008). Comparative study of polypropylene composites reinforced with oil palm empty fruit bunch fiber and oil palm derived cellulose. *Materials & Design, 29*(1), 173-178.

Khanam, P. N., AlMaadeed, M., AlMaadeed, S., Kunhoth, S., Ouederni, M., Sun, D & Mayoral, B. (2016). Optimization and prediction of mechanical and thermal properties of graphene/LLDPE nanocomposites by using artificial neural networks. *International Journal of Polymer Science, 2015.* 1-15.

Klapiszewski, Ł., Bula, K., Sobczak, M., & Jesionowski, T. (2016). Influence of processing conditions on the thermal stability and mechanical properties of PP/silica-lignin composites. *International Journal of Polymer Science, 2016*, 1-9.

Law, K.-N., Daud, W. R. W., & Ghazali, A. (2007). Morphological and chemical nature of fiber strands of oil palm empty-fruit-bunch (OPEFB). *Bioresources, 2*(3), 351-362.

Lee, H., Hamid, S. B. A., & Zain, S. (2014). Conversion of lignocellulosic biomass to nanocellulose: Structure and chemical process. *The Scientific World Journal, 2014,* 1-20.

Lee, K.-Y., Delille, A., & Bismarck, A. (2011). Greener surface treatments of natural fibres for the production of renewable composite materials. In Cellulose fibers: bio-and nano-polymer composites (pp. 155-178). Springer, Berlin, Heidelberg.

Liang, J.-Z., & Yang, Q.-Q. (2007). Mechanical, thermal, and flow properties of HDPE–mica composites. *Journal of Thermoplastic Composite Materials, 20*(2), 225-236.

Lilholt, H., & Lawther, J. M. (2000). Natural organic fibers. In A. Kelly & C. Zweben (Eds.), Comprehensive Composite Materials (pp. 303-325). Oxford: Pergamon.

Lu, J. Z., Wu, Q., & McNabb, H. S. (2007). Chemical coupling in wood fiber and polymer composites: A review of coupling agents and treatments. *Wood and Fiber Science, 32*(1), 88-104.

Mahjoub, R., Bin Mohamad Yatim, J., Sam, M., & Rahman, A. (2013). A review of structural performance of oil palm empty fruit bunch fiber in polymer composites. *Advances in Materials Science and Engineering, 2013*, 1-9.

Mallick, P. (2010). Thermoplastics and thermoplastic–matrix composites for lightweight automotive structures. In P. K. Mallick (Ed.), Materials, Design and Manufacturing for Lightweight Vehicles (pp. 174-207): Woodhead Publishing.

Mishra, A., Pandey, A., & Dwivedi, M. (2016). Natural fibre reinforcement polymer composites-A review. *International Journal of Advance Research in Science and Engineering, 5*(5), 453-458.

Modi, H., Chokshi, S., & Patel, D. (2017). Impact strength prediction of natural fiber reinforced polyester composites. *International Journal of Engineering Technology Science and Research, 4*(8), 595-604.

Mohanty, A. K., Misra, M., & Drzal, L. (2002). Sustainable bio-composites from renewable resources: Opportunities and challenges in the green materials world. *Journal of Polymers and the Environment, 10*(1-2), 19-26.

Moigne, L. N., Otazaghine, B., Corn, S., Angellier-Coussy, H., & Bergeret, A. (2018). Characterization of the Interface/Interphase in Natural Fibre Based Composites. In *Surfaces and Interfaces in Natural Fibre Reinforced Composites* (pp. 101-133). France: Springer.

Moudood, A., Hall, W., Öchsner, A., Li, H., Rahman, A., & Francucci, G. (2019). Effect of moisture in flax fibres on the quality of their composites. *Journal of Natural Fibers, 16*(2), 209-224.

Mrazova, M. (2013). Advanced composite materials of the future in aerospace industry. *Incas bulletin, 5*(3), 139-150.

Mu, B., Wang, H., Hao, X., & Wang, Q. (2018). Morphology, mechanical properties and dimensional stability of biomass particles/high density polyethylene composites: Effect of species and composition. *Polymers, 10*(3), 1-15.

Mueller, H., & Knauss, W. (1971). The fracture energy and some mechanical properties of a polyurethane elastomer. *Transactions of The Society of Rheology, 15*(2), 217-233.

Munoz, E., & García-Manrique, J. A. (2015). Water absorption behaviour and its effect on the mechanical properties of flax fibre reinforced bioepoxy composites. *International Journal of Polymer Science, 2015,* 1-10.

Mwaikambo, L. (2006). Review of the history, properties and application of plant fibres. *African Journal of Science and Technology (AJST) Science and Engineering Series, 7*(2), 120-133.

Nair, V., Khosla, P., & Ramachandran, M. (2016). Review on mechanical properties of various natural fibers reinforced composites. *Research Journal of Pharmaceutical Biological and Chemical Sciences, 7*(1), 2001-2004.

Ngo, W., Pang, M., Yong, L., & Tshai, K. (2014). Mechanical properties of natural fibre (kenaf, oil palm empty fruit bunch) reinforced polymer composites. *Advances in Environmental Biology*, *8*(8) 2742-2748.

Nuthong, W., Uawongsuwan, P., Pivsa-Art, W., & Hamada, H. (2013). Impact property of flexible epoxy treated natural fiber reinforced PLA composites. *Energy Procedia, 34*, 839-847.

Nyström, B., Joffe, R., & Långström, R. (2007). Microstructure and strength of injection molded natural fiber composites. *Journal of Reinforced Plastics and Composites, 26*(6), 579-599.

Obasi, H. C., Igwe, I. O., & Madufor, I. C. (2013). Effect of soil burial on tensile properties of polypropylene/plasticized cassava starch blends. *Advances in Materials Science and Engineering, 2013,* 1-5.

Ołdak, D., Kaczmarek, H., Buffeteau, T., & Sourisseau, C. (2005). Photo-and bio-degradation processes in polyethylene, cellulose and their blends studied by ATR-FTIR and Raman spectroscopies. *Journal of Materials Science, 40*(16), 4189-4198.

Onuaguluchi, O., & Banthia, N. (2016). Plant-based natural fibre reinforced cement composites: A review. *Cement and Concrete Composites, 68*, 96-108.

Özdilek, C., Kazimierczak, K., van der Beek, D., & Picken, S. J. (2004). Preparation and properties of polyamide-6-boehmite nanocomposites. *Polymer, 45*(15), 5207-5214.

Peltola, H., Madsen, B., Joffe, R., & Nättinen, K. (2011). Experimental study of fiber length and orientation in injection molded natural fiber/starch acetate composites. *Advances in Materials Science and Engineering, 2011,* 1-7.

Piarpuzan, D., Quintero, J. A., & Cardona, C. A. (2011). Empty fruit bunches from oil palm as a potential raw material for fuel ethanol production. *Biomass and Bioenergy, 35*(3), 1130-1137.

Pickering, K. L., Efendy, M. A., & Le, T. M. (2016). A review of recent developments in natural fibre composites and their mechanical performance. *Composites Part A: Applied Science and Manufacturing, 83*, 98-112.

Placet, V., Trivaudey, F., Cisse, O., Guicheret-Retel, V., & Boubakar, L. (2012). Diameter dependence of the apparent tensile modulus of hemp fibres: A morphological, structural or ultrastructural effect?.*Composites Part A: Applied Science and Manufacturing*, *43*(2), 275-287.

Ramakrishna, G., Sundararajan, T., & Kothandaraman, S. (2010). Evaluation of durability of natural fibre reinforced cement mortar composite-a new approach. *ARPN Journal of Engineering and Applied Sciences, 5*(6), 44-51.

Ramakrishna, S., & Huang, Z. M. (2003). 9.06 - Biocomposites. In I. Milne, R. O. Ritchie, & B. Karihaloo (Eds.), *Comprehensive Structural Integrity* (pp. 215-296). Oxford: Pergamon.

Rashdi, A., Sapuan, S., Ahmad, M., & Khalina, A. (2010). Combined effects of water absorption due to water immersion, soil buried and natural weather on mechanical properties of kenaf fibre unsaturated polyester composites (KFUPC). *International Journal of Mechanical and Materials Engineering, 5*(1), 11-17.

Ravi, M., Rajnish, R. D., Alok, S., Saikat, G., & Kumar, C. A. (2018). Effect of surface treatment on Natural fibers composite. *IOP Conference Series: Materials Science and Engineering, 376*(1), 1-5.

Razak, N., & Kalam, A. (2012). Effect of OPEFB size on the mechanical properties and water absorption behaviour of OPEFB/PPnanoclay/PP hybrid composites. *Procedia Engineering, 41*, 1593-1599.

Rosa, M. F., Chiou, B.-s., Medeiros, E. S., Wood, D. F., Williams, T. G., Mattoso, L. H. & Imam, S. H. (2009). Effect of fiber treatments on tensile and thermal properties of starch/ethylene vinyl alcohol copolymers/coir biocomposites. *Bioresource Technology, 100*(21), 5196-5202.

Rozman, H., Saad, M., & Ishak, Z. M. (2003). Flexural and impact properties of oil palm empty fruit bunch (EFB)–polypropylene composites—the effect of maleic anhydride chemical modification of EFB. *Polymer Testing, 22*(3), 335-341.

Rozman, H., Tay, G., Kumar, R., Abusamah, A., Ismail, H., & Ishak, Z. M. (2001). Polypropylene–oil palm empty fruit bunch–glass fibre hybrid composites: A preliminary study on the flexural and tensile properties. *European Polymer Journal, 37*(6), 1283-1291.

Salmah, H., Lim, B., & Teh, P. (2013). Rheological and thermal properties of palm kernel shell–filled low-density polyethylene composites with acrylic acid. *Journal of Thermoplastic Composite Materials, 26*(9), 1155-1167.

Sam, S., Ismail, H., & Ahmad, Z. (2011). Soil burial of polyethylene-g-(maleic anhydride) compatibilised LLDPE/soya powder blends. *Polymer-Plastics Technology and Engineering, 50*(8), 851-861.

Santulli, C., Sarasini, F., Puglia, D., & Kenny, J. M. (2017). 8 Injection moulding of plant fibre composites. In *Advanced Composite Materials: Properties and Applications* (pp. 420-439): Sciendo Migration.

Sefadi, J. S., & Luyt, A. (2012). Morphology and properties of EVA/empty fruit bunch composites. *Journal of Thermoplastic Composite Materials, 25*(7), 895-914.

Shah, A. U. M., Sultan, M. T. H., Cardona, F., Jawaid, M., Talib, A. R. A., & Yidris, N. (2017). Thermal analysis of bamboo fibre and its composites. *Bioresources, 12*(2), 2394-2406.

Shah, D. U., Schubel, P. J., Licence, P., & Clifford, M. J. (2012). Hydroxyethylcellulose surface treatment of natural fibres: The new 'twist'in yarn preparation and optimization for composites applicability. *Journal of Materials Science, 47*(6), 2700-2711.

Shahzad, A. (2011). Effects of fibre surface treatments on mechanical properties of hemp fibre composites. *Composite Interfaces 18*, 737-754.

Shahzad, A. (2013). A study in physical and mechanical properties of hemp fibres. *Advances in Materials Science and Engineering, 2013*, 1-9.

Shamsudin, S., Shah, U. K. M., Zainudin, H., Abd-Aziz, S., Kamal, S. M. M., Shirai, Y., & Hassan, M. A. (2012). Effect of steam pretreatment on oil palm empty fruit bunch for the production of sugars. *Biomass and Bioenergy, 36*, 280-288.

Sharba, M. J., Salman, S. D., Leman, Z., Sultan, M. T., Ishak, M. R., & Hanim, M. A. A. (2015). Effects of processing method, moisture content, and resin system on physical and mechanical properties of woven kenaf plant fiber composites. *Bioresources, 11*(1), 1466-1476.

Shinoj, S., Visvanathan, R., Panigrahi, S., & Kochubabu, M. (2011). Oil palm fiber (OPF) and its composites: A review. *Industrial Crops and Products, 33*(1), 7-22.

Shivnand, H., Inamdar, P. S., & Sapthagiri, G. (2010). *Evaluation of tensile and flexural properties of hemp and polypropylene based natural fiber composites*. Paper presented at the 2010 2nd International Conference on Chemical, Biological and Environmental Engineering (ICBEE 2010), 2nd to 4th November 2010, Cairo, Egypt.

Shuit, S. H., Tan, K. T., Lee, K. T., & Kamaruddin, A. (2009). Oil palm biomass as a sustainable energy source: A Malaysian case study. *Energy, 34*(9), 1225-1235.

Singh, S., Deepak, D., Aggarwal, L., & Gupta, V. (2014a). Tensile and flexural behavior Singh, S., Deepak, D., Aggarwal, L., & Gupta, V. (2014). Tensile and flexural behavior of hemp fiber reinforced virgin-recycled HDPE matrix composites. *Procedia Materials Science, 6*, 1696-1702.

Smidt, E., & Tintner, J. (2007). Application of differential scanning calorimetry (DSC) to evaluate the quality of compost organic matter. *Thermochimica Acta, 459*(1-2), 87-93.

Solikhin, A., Hadi, Y. S., Massijaya, M. Y., & Nikmatin, S. (2016). Basic Properties of oven–heat treated oil palm empty fruit bunch stalk fibers. *Bioresources, 11*(1), 2224-2237.

Sood, M., & Dwivedi, G. (2017). Effect of fiber treatment on flexural properties of natural fiber reinforced composites: A review. *Egyptian Journal of Petroleum, 27*(4), 775-783.

Sosiati, H., & Harsojo. (2014). Effect of combined treatment methods on the crystallinity and surface morphology of kenaf bast fibers. *Cellulose Chemistry and Technology, 48*(1-2), 33-43.

Sreekala, M., Kumaran, M., Geethakumariamma, M., & Thomas, S. (2004). Environmental effects in oil palm fiber reinforced phenol formaldehyde composites: Studies on thermal, biological, moisture and high energy radiation effects. *Advanced Composite Materials, 13*(3-4), 171-197.

Sreekala, M., Kumaran, M., Joseph, R., & Thomas, S. (2001). Stress-relaxation behaviour in composites based on short oil-palm fibres and phenol formaldehyde resin. *Composites Science and Technology, 61*(9), 1175-1188.

Sreekala, M., & Thomas, S. (2003). Effect of fibre surface modification on water-sorption characteristics of oil palm fibres. *Composites Science and Technology, 63*(6), 861-869.

Sui, G., Fuqua, M., Ulven, C., & Zhong, W. (2009). A plant fiber reinforced polymer composite prepared by a twin-screw extruder. *Bioresource Technology, 100*(3), 1246-1251.

Sui, G., Zhong, W., Fuqua, M., & Ulven, C. (2007). *Properties of polypropylene composites fabricated using twin-screw extrusion*. Paper presented at the Proceedings of the 16th International Conference on Composite Materials, 8th – 13th July 2007, Kyoto, Japan.

Suradi, S., Yunus, R., & Beg, M. (2011). Oil palm bio-fiber-reinforced polypropylene composites: Effects of alkali fiber treatment and coupling agents. *Journal of Composite Materials, 45*(18), 1853-1861.

Tajau, R., Salleh, M. Z., Salleh, N. G. N., Abdurahman, M. N., Salih, A. M., Fathy, S. F., . . . Hamidi, N. A. (2016). *Soil burial biodegradation studies of palm oil-based UV-curable films.* Paper presented at the AIP Conference Proceedings.

Tajvidi, M., & Ebrahimi, G. (2003). Water uptake and mechanical characteristics of natural filler–polypropylene composites. *Journal of Applied Polymer Science, 88*(4), 941-946.

Tezara, C., Siregar, J. P., Lim, H., Fauzi, F., Yazdi, M. H., & Moey, L. (2016). Factors that affect the mechanical properties of kenaf fiber reinforced polymer: A review. *Journal of Mechanical Engineering and Sciences, 10*(2), 2159-2175.

Torres, F., & Diaz, R. (2004). Morphological characterisation of natural fibre reinforced thermoplastics (NFRTP) processed by extrusion, compression and rotational moulding. *Polymers and Polymer Composites, 12*(8), 705-718.

Tran, L., Yuan, X., Bhattacharyya, D., Fuentes, C., Van Vuure, A., & Verpoest, I. (2015). Fiber-matrix interfacial adhesion in natural fiber composites. *International Journal of Modern Physics B, 29*(10n11), 1540018.

Van de Velde, K., & Kiekens, P. (2001). Thermoplastic pultrusion of natural fibre reinforced composites. *Composite Structures, 54*(2-3), 355-360.

Viksne, A., Rence, L., Kalnins, M., & Bledzki, A. K. (2004). The effect of paraffin on fiber dispersion and mechanical properties of polyolefin–sawdust composites. *Journal of Applied Polymer Science, 93*(5), 2385-2393.

Wambua, P., Ivens, J., & Verpoest, I. (2003). Natural fibres: Can they replace glass in fibre reinforced plastics? *Composites Science and Technology, 63*(9), 1259-1264.

Wang, P.-H., Ghoshal, S., Gulgunje, P., Verghese, N., & Kumar, S. (2016). Polypropylene nanocomposites with polymer coated multiwall carbon nanotubes. *Polymer, 100*, 244-258.

Willems, F., & Bonten, C. (2016). *Influence of processing on the fiber length degradation in fiber reinforced plastic parts.* Paper presented at the AIP Conference Proceedings, 1799 (1), 1-5.

Wirjosentono, B., Guritno, P., & Ismail, H. (2004). Oil palm empty fruit bunch filled polypropylene composites. *International Journal of Polymeric Materials, 53*(4), 295-306.

Yang, H.-S., Wolcott, M., Kim, H.-S., & Kim, H.-J. (2005). Thermal properties of lignocellulosic filler-thermoplastic polymer bio-composites. *Journal of Thermal Analysis and Calorimetry, 82*(1), 157-160.

Yoon, C. J., & Kim, K. W. (2008). Anatomical descriptions of silicified woods from Madagascar and Indonesia by scanning electron microscopy. *Micron, 39*(7), 825-831.

Yusoff, M. Z. M., Salit, M. S., & Ismail, N. (2009). Tensile properties of single oil palm empty fruit bunch (OPEFB) fibre. *Sains Malaysiana, 38*(4), 525-529.

LIST OF PUBLICATIONS AND PAPER PRESENTED

PUBLICATIONS

1. **Nordin, N. A.,** Abd Rahman, N. M. M., & Hassan, A. (2019). Thermal and mechanical properties of injection moulded heat-treated oil palm empty fruit bunch fibre-reinforced high-density polyethylene composites. *Plastics, Rubber and Composites,* 1-12.

2. **Nordin, N. A.,** Abd Rahman, N. M. M., & Hassan, A. (2020). Conditioning effect on the mechanical and thermal properties of heat-treated oil palm empty fruit bunch/high-density polyethylene composite. *High Performance Polymers, 32*(2), 158-167.

PAPERS PRESENTED

1. **Nordin, N. A.,** Abd Rahman, N. M. M., & Hassan, A. (2019). *Conditioning effect on the mechanical and thermal properties of heat-treated oil palm empty fruit bunch/high density polyethylene composite.* Paper presented at the 3rd International Conference on Science & Engineering of Materials (ICSEM-2019), 19th – 21st July 2019, Greater Noida, India.

2. **Nordin, N. A.,** Abd Rahman, N. M. M., & Hassan, A. (2016). *Thermal and mechanical properties of injection moulded oil palm empty fruit bunch fibre reinforced high density polyethylene composites.* Paper presented at the 1st Asian Researcher Symposium (ARS) 2016, 25-26th April 2016, Jakarta, Indonesia.