CHARACTERIZATION OF POLY (3-HYDROXYBUTYRATE-CO-3-HYDROXYVALERATE) (PHBV) REINFORCED WITH PINEAPPLE LEAF FIBER COMPOSITE FOR POTENTIAL APPLICATION AS PACKAGING MATERIAL

This report is submitted in accordance with requirement of the University Teknikal Malaysia Melaka (UTeM) for Bachelor Degree of Manufacturing Engineering (Hons.)

by

MUHAMMAD NAZMI BIN MOHD NASSIR

B052110133

971023-04-5351

FACULTY OF INDUSTRIAL AND MANUFACTURING TECHNOLOGY AND ENGINEERING

2024

UNIVERSITI TEKNIKAL MALAYSIA MELAKA

BORANG PENGESAHAN STATUS LAPORAN PROJEK SARJANA MUDA

Tajuk: **CHARACTERIZATION OF POLY (3-HYDROXYBUTYRATE-CO-3-HYDROXYVALERATE) (PHBV) REINFORCED PINEAPPLE LEAF FIBER COMPOSITE FOR POTENTIAL APPLICATION AS PACKAGING MATERIAL**

Sesi Pengajian: **2023/2024 Semester 2**

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Disahkan oleh:

Muhammad Nazmi.

Alamat Tetap:
110, Pengkalan Rama Pantai,
75100, Melaka.

Tarikh: 12/7/2024.

Cop Rasmi:
ASSOC. PROF. DR. ZALEHA BINTI MUSTAFA
Associate Professor
Faculty of Manufacturing Engineering
Universiti Teknikal Malaysia Melaka
Hang Tuah Jaya
76100 Durian Tunggal, Melaka

Tarikh: 12/7/2024

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I hereby, declared this report entitled "Characterization of Poly (3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) Reinforced Pineapple Leaf Fiber Composite for Potential Application as Packaging Material" is the result of my own research except as cited in references.

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Author's Name : MUHAMMAD NAZMI BIN MOHD NASSIR

Date : 12/7/2024.

APPROVAL

This report is submitted to the Faculty of Manufacturing Engineering of Universiti Teknikal Malaysia Melaka as a partial fulfilment of the requirement for Degree of Manufacturing Engineering (Hons). The member of the supervisory committee is as follow:

.....

(Prof Madya Dr Zaleha Binti Mustafa)

ABSTRAK

Polimer biodegradasi, seperti Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV), mendapat perhatian sebagai alternatif mesra alam berbanding polimer berasaskan petroleum. Walaupun sesuai untuk bahan pembungkusan, ia mempunyai kerapuhan tinggi dan kekuatan mekanikal terhad, memerlukan pengukuhan serat semulajadi. Serat Daun Nanas (PALF), yang kaya dengan selulosa, dipilih untuk mengukuhkan PHBV, bagi meningkatkan sifat komposit dalam industri pembungkusan, Kajian ini bertujuan untuk membandingkan kesan pengisian PALF terhadap sifat mekanikal tegangan, menyiasat kesan kelembapan terhadap sifat mekanikal tegangan, dan menilai permukaan patah komposit. PALF dirawat alkali dimasukkan ke dalam PHBV dengan pemuatan berbeza. Ujian tegangan dijalankan untuk menilai Modulus Young, kekuatan, dan regangan patah. Analisis morfologi menggunakan Mikroskop Elektron Imbasan (SEM) dilakukan untuk menganalisis permukaan patah. Komposit diuji penyerapan air selama 14 hari untuk mensimulasikan kelembapan, diikuti ujian fizikal, ujian tegangan, dan analisis morfologi. Keputusan menunjukkan peningkatan PALF meningkatkan tegangan dari 29.57 MPa (PHBV tulen) kepada 46.74 MPa (30 wt.%), manakala modulus menurun dari 0.0193 GPa kepada 0.0083 GPa. Regangan patah meningkat dari 1.73% kepada 6.19%, menunjukkan peningkatan keanjalan. Kelembapan memberi kesan pada parameter mekanikal; untuk 20 wt.% PALF, tegangan menurun dari 29.81 MPa kepada 19.87 MPa, modulus meningkat dari 0.0075 GPa kepada 0.0111 GPa, dan regangan patah menurun dari 3.38% kepada 2.03% akibat pembengkakan serat dan retakan mikro. SEM mendedahkan bahawa PHBV tulen menunjukkan kerapuhan, manakala 20 wt.% PALF menunjukkan ikatan kuat pada sampel tidak berumur tetapi penarikan serat dan kerosakan yang ketara pada sampel berumur akibat kelembapan. Penyelidikan ini menunjukkan potensi komposituntuk meningkatkan sifat mekanikal dan ketahanan, menawarkan alternatif lestari untuk bahan pembungkusan.

ABSTRACT

Biodegradable polymer, such as Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV), is gaining popularity as an eco- friendly alternative to petroleum-based polymers. Despite its suitability in packaging material, it has high brittleness and limited mechanical strength, requiring reinforcement from natural fiber. Pineapple Leaf Fiber (PALF), containing high cellulose, was chosen to reinforce with PHBV, for improving composite properties in packaging industry. Thus, the objectives of this study are to compare the effect of PALF loading on the tensile mechanical properties, investigate the effect of moisture on the tensile mechanical properties, and evaluate the surface fracture behavior of the PHBV/PALF composite. Alkaline-treated PALF was filled into PHBV at varying loadings (0-30 wt.%). Then, tensile tests were conducted to evaluate Young's Modulus, strength, and strain at break. Next, morphological analysis of composites was conducted using a Scanning Electron Microscope (SEM) to analyze fracture surfaces. Composites underwent a 14-day water absorption test to simulate moisture exposure, followed by physical tests, tensile tests, and morphological analysis. Results showed that increasing PALF loading improved tensile strength from 29.57 MPa (neat PHBV) to 46.74 MPa (30 wt.%), whereas modulus decreased from 0.0193 GPa to 0.0083 GPa. Strain at break increased from 1.73% to 6.19%, indicating enhanced ductility. Moisture exposure significantly impacted mechanical properties; for 20 wt.% PALF, tensile strength decreased from 29.81 MPa to 19.87 MPa, modulus increased from 0.0075 GPa to 0.0111 GPa, and strain at break decreased from 3.38% to 2.03% due to fiber swelling and microcracks. SEM analysis showed neat PHBV exhibited brittle failure, while 20 wt.% PALF showed strong bonding in unaged samples but significant fiber pull-out and breakage in aged samples due to moisture. This research demonstrates the potential of PHBV/PALF composites to improve the mechanical properties and durability, offering sustainable alternatives for packaging materials.

DEDICATION

My beloved father, Mohd Nassir Bin Abdul Aziz

My beloved mother, Noraini Binti Tamat

My adored sister and brother, Nazrul, Haziq, Hazarif and Nurhazimah

for giving me moral support, money, cooperation, encouragement and also understandings

Thank You So Much & Love You All Forever

ACKNOWLEDGEMENT

In the name of ALLAH, the most gracious, the most merciful, with the highest praise to Allah that I manage to complete this final year project successfully without difficulty.

My respected supervisor, Prof Madya Dr Zaleha Binti Mustafa. Her kindness, unwavering patience and mentorship guided me through the process, her easily understood explanations and open mind allowed me to grow and learn in such a way that I am now a better researcher.

Last but not least, I would like to give a special thanks to my family for their motivation and moral support in completing this report. Next, I would like to thank my classmates for lend me their hand along the journey of preparing this report. Thanks for the great family and friendship.

Finally, I would like to thank everybody who was important to this FYP report, as well as expressing my apology that I could not mention personally each one of you.

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LIST OF ABBREVIATIONS

ASTM	-	American Society for Testing and Materials
BF	-	Basalt Fiber
DSC	-	Differential Scanning Calorimetry
ENR	-	Epoxidized Natural Rubber
ISO	-	International Organization for Standardization
PALF	-	Pineapple Leaf Fiber
PHA	-	Polyhydroxyalkanoate
PHBV	-	Poly (3-hydroxybutyrate-co-3-hydroxyvalerate)
PLA	-	Polylactic Acid
PP	-	Polypropylene
SEM	-	Scanning Electron Microscope
TGA	-	Thermogravimetric Analysis
UV	-	Ultraviolet
WF	-	Wood Fiber
XRD	-	X-Ray Diffraction

LIST OF SYMBOLS

σ	-	Stress
MPa	-	Mega Pascal
GPa	-	Giga Pascal
°C	-	Degree Celcius
Mt	-	Metrics Ton
CO^2	-	Carbon Dioxide
CH_4	-	Methane
%	-	Percent
wt%	-	Weight Percent
°	-	Degree
Tm	-	Melting Point
g/cm^3	-	Gram per Centimetre Cubic
mm	-	Millimetre
mmHg	-	Millimetre of Mercury
mm/min	-	Millimetre per Minute
ε	-	Strain
E	-	Young's Modulus

CHAPTER 1

INTRODUCTION

1.1 Background of Study

In this new era, engineering material manufacturers are looking for new and more efficient ways to counteract the adverse impacts such as water pollutions, toxic waste generation, and high energy consumption associated with functional materials. To mitigate these negative consequences, more effective and sustainable approaches are needed as technology develops and the need for innovative solutions increases. One of the ways to cope with these problems is by development and utilization polymer composites. These composite materials have developed as a desirable alternative in a variety of industries, including packaging, automotive, aerospace, and various others. Their appeal lies in their exceptional versatility, as they offer a good alternative to many traditional materials like metals, ceramics, glass, and concrete.

In the context of packaging materials, there has been a noticeable and robust growth in the utilization of bio-based materials within the sustainable packaging industry. This surge in popularity can be attributed to recent consumer trends that prioritize environmentally friendly packaging options and a reduction of waste (Reichert et al., 2020). As a response to the increasing demand for environmentally friendly packaging solutions, bio-composites have been introduced as a notable innovation. These bio-composites are engineered using biodegradable polymeric matrices and natural fibers, combining the benefits of biodegradability and ecological friendly.

An essential aspect of these bio-composites is their capacity to mitigate the environmental impact of packaging materials. Once their intended use is fulfilled, these "green" composites can be easily composted, contributing to a reduction in waste and posing no significant threat to the environment. Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) is one of the candidates stands out due to its biodegradable nature and compatibility with biological systems. However, the drawbacks of this biopolymer are their high brittleness and limited mechanical strength, which can hinder it broader applications in various industries. Thus, they are often used with natural fiber to improve their mechanical properties and at the same time maintaining it degradability.

Pineapple Leaf Fiber (PALF) has low microfibrillar angle, which is the key factor in its enhanced strength. These characteristics make PALF a highly suitable candidate for reinforcing PHBV, as it promises to significantly improve the overall mechanical performance of the biopolymer, while simultaneously offering cost-efficiency. This strategic application of PALF combined with PHBV demonstrates a well-informed technique to addressing the limitations of biodegradable polymers and shows the appropriate technique to producing more sustainable and ecologically friendly packaging materials.

1.2 Problem Statement

In the context of environmental sustainability and efficient waste management, it is critical to acknowledge the significant drawbacks of conventional packaging materials, many of which are derived from petroleum-based plastics. These materials are non-biodegradable, meaning they do not naturally decompose, and they can cause serious environmental hazards. The relentless of such non-biodegradable materials in the environment has raised concerns about pollution and waste accumulation.

In contrast, packaging materials made of biodegradable polymers provide a more environmentally friendly option. Biodegradable polymers can break down naturally over time, reducing the long-term environmental impact. In this project, Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) stands out as a remarkable example. It is regarded as an alternative for petrochemical polymers because of its high tensile strength (22–34 MPa) and Young's modulus (3–6 GPa), which are comparable to those of commonly used

Polypropylene (PP). Moreover, due to its good oxygen and water barrier properties, it is one of the best options in packaging applications. However, this polymer has high brittleness with relatively low impact strength, high cost to produce and may degrade during storage, making commercial use challenging (Conceição et al., 2023; K. E. Mazur et al., 2022).

To address these limitations and enhance the ability of PHBV as a packaging material, effort has been done by exploring the incorporation of natural fibers as reinforcements. By adding natural fibers as reinforcements, it is an effective and economical way to enhance the strength of biodegradable polymers while providing desired properties. Commonly used natural fibers have cellulose, lignin, pectin, and hemicellulose. Composites reinforced with lignocellulosic fibers offer high stiffness, biodegradability, and lightweight characteristics, with a favorable Young's modulus/density ratio (K. E. Mazur et al., 2022). By blending natural fibers with biodegradable polymers, it becomes possible to create composite materials that combine the benefits of both components.

In the context of this project, the choice to introduce Pineapple Leaf Fiber (PALF) as the reinforcement for PHBV is noteworthy. Todkar and Patil (2019) research highlighted that PALF is a suitable option due to its specific properties. PALF has the highest cellulose content of any natural fiber derived from plant leaves, contributing to its good strength and durability.

However, another property of PALF is Hydrophilic, which can attract water, while PHBV is Hydrophobic, these two opposites properties will be a challenge in forming strong bonds in composites. Hydrophilic nature of PALF may limit effective bonding with PHBV, potentially impacting composite strength. Consequently, this composite will be sensitive to moisture, potentially affecting its properties and performance since material for packaging will be used commonly in the environment that have moisture in air. Moisture presence may alter the physical and mechanical characteristics of this composites, requiring strategic design and precautions to cater these effects and ensure the desired performance and longevity of the composite material.

1.3 Objectives

1.3.1 To compare the effect of fiber loading on the tensile properties of PHBV/PALF composites.

1.3.2 To analyze the effect of moisture on the tensile properties of the PHBV/PALF composites.

1.3.3 To evaluate the surface fracture of the PHBV/PALF composite.

1.4 Scope

This project involves fabricating composites using Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) and short Pineapple Leaf Fiber (PALF) with a length below 6mm. Tensile tests will be conducted with varying fiber loadings (0%, 10%, 20%, 30%) to evaluate the composites strength. The water absorption of the composites along 14 days at room's temperature (23ºC) will be examined, and the fracture surfaces will be analyzed using a Scanning Electron Microscope (SEM) machine.

CHAPTER 2

LITERATURE REVIEW

This chapter discusses research papers that have been made by researchers related to the title of this research. This chapter will be discussing about the biodegradable polymers, natural fibers as reinforcements in composites, surface treatment, fiber architectures, the examples of PHBV composites, and their applications.

2.1 Introduction

The increasing use of plastics in packaging has become an important factor of environmental impact, posing various issues in terms of disposal and management at the end of their life cycle. As a result, 22 Mt of plastic materials leaking into the environment in 2019, contributing to persistent plastic pollution (Jeswani et al., 2023; OECD, 2022). Based on Figure 2.1 below, huge percentages of plastics that leaked to the environment which is 82% (macroplastics) are come from mismanaged waste, littering, and marine activities. Other factors that contributing to microplastics are come from transport related microplastics, microplastics dust, wastewater sludge and other.

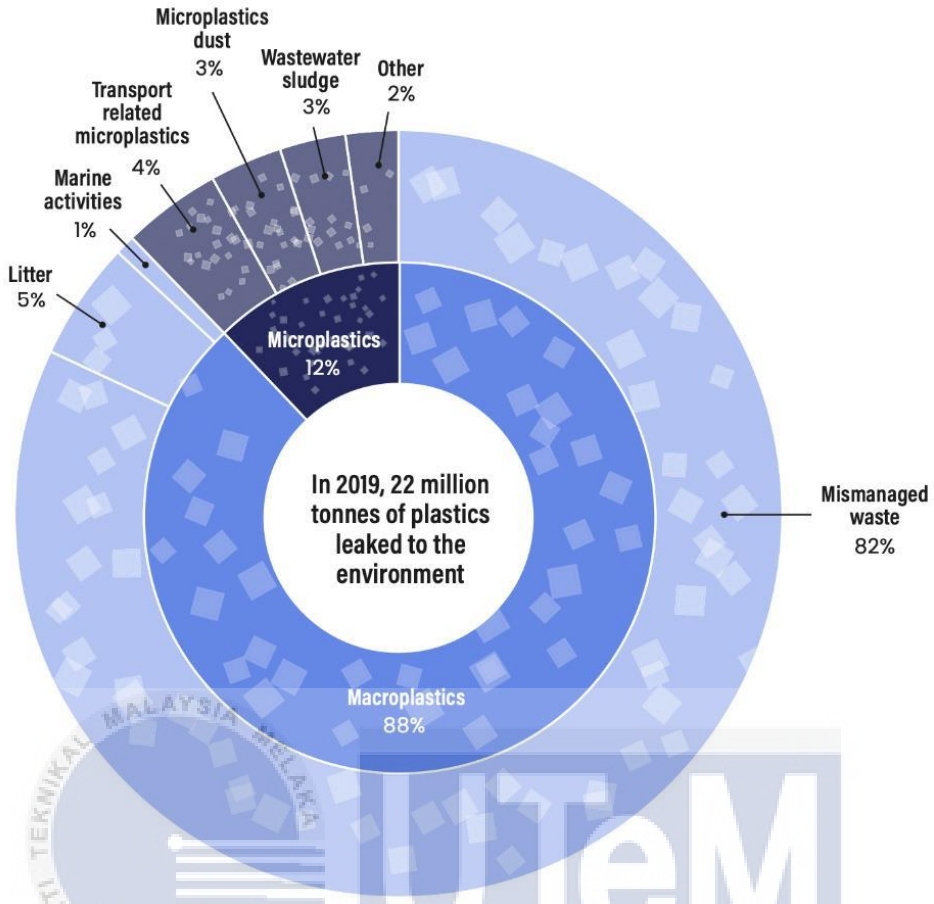

Figure 2.1: Global leakage of macro-and microplastics to the environment in 2019. (OECD, 2022)

To minimize the consequences related to the use of fossil fuel plastics in packaging usage, there is an effective way to develop materials that may easily degrade in waste disposal systems, such as landfilling, anaerobic digestion, and industrial composting. Polyester-based biodegradable plastics emerge as a good option to replace current single-use plastics, offering a potential solution to the plastic waste problem. Various aliphatic polyesters, including poly (lactic acid) (PLA), polyhydroxyalkanoates (PHA), and polyhydroxybutyrate (PHB), have garnered attention due to their production from renewable feedstocks and their capacity for biodegradation (Jeswani et al., 2023).

Biobased polymers can be made from a variety of renewable resources, including first-generation feedstock (sugar, sugarcane, beet, potato, corn, and hemp); second generation feedstock (food crops, wood residues, and other non-food crops) and lastly third generation feedstock (biomass derived from algae or methane (CH_4) (Sikorska et al., 2021).

2.2 Biodegradable Polymers

Biodegradable polymers are a class of materials that have gained significant attention due to their potential to address environmental concerns and reduce the carbon footprint comparing to petroleum-based polymers. These polymers are designed to degrade through the action of naturally occurring microorganisms such as bacteria, fungi, and algae. During biodegradation, they will show a reduction in specific properties like mass, tensile strength, elasticity, or opacity. For a polymer to be considered mineralized, it must completely transform into carbon dioxide, water, biomass, and minerals, with no byproducts remaining (Albright & Chai, 2021).

Based on Figure 2.2 below, biodegradable polymers can be produced through diverse techniques, including ring opening polymerization (ROP), chemoenzymatic methods, photo-initiated radical polymerization, enzymatic polymerization, and various types of ring opening polymerization such as cationic, anionic, enzymatic, coordinative, and radical methods (Meghana et al., 2023).

Figure 2.2: Strategies involved in the synthesis of biodegradable polymers (Meghana et al., 2023)

Due to its beneficial characteristics, which are non-toxicity, environmentally friendly nature, and high mechanical strength, they have been explored in variety of applications. Some of them are drug delivery system, aquaculture tools, and electronics. The applications of biodegradable polymers are shown in Figure 2.3 below.

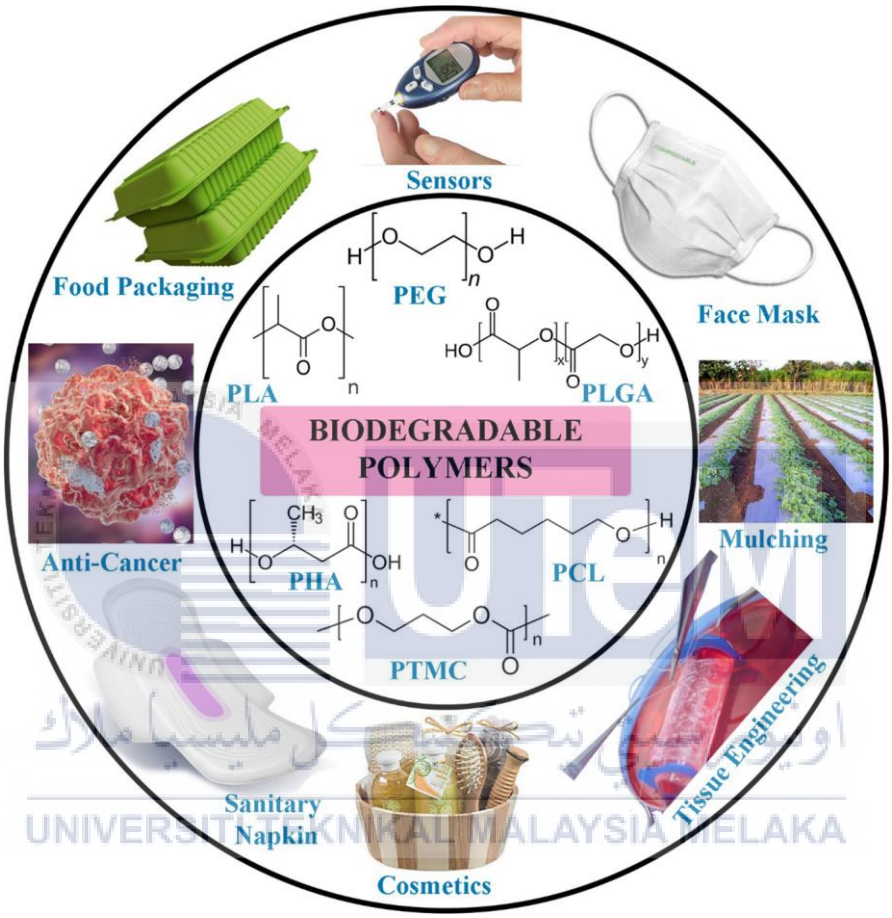

Figure 2.3: Examples of application of biodegradable polymers. (Meghana et al., 2023)

2.2.1 Poly (3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV)

Polyhydroxyalkanoates (PHAs) are a family of biodegradable polymers that are synthesized by microorganisms as energy and carbon storage compounds. PHAs are biopolymers that are biosynthesized by various microorganisms as granules under stressed environmental conditions, typically in the presence of excess carbon and limited essential

growth nutrients such as phosphate and nitrogen. According to Figure 2.4 below, the biosynthesis of PHAs is part of the closed carbon cycle of nature, and their degradation does not contribute to an increase in atmospheric CO^2 levels, making them environmentally friendly (Ibrahim et al., 2021).

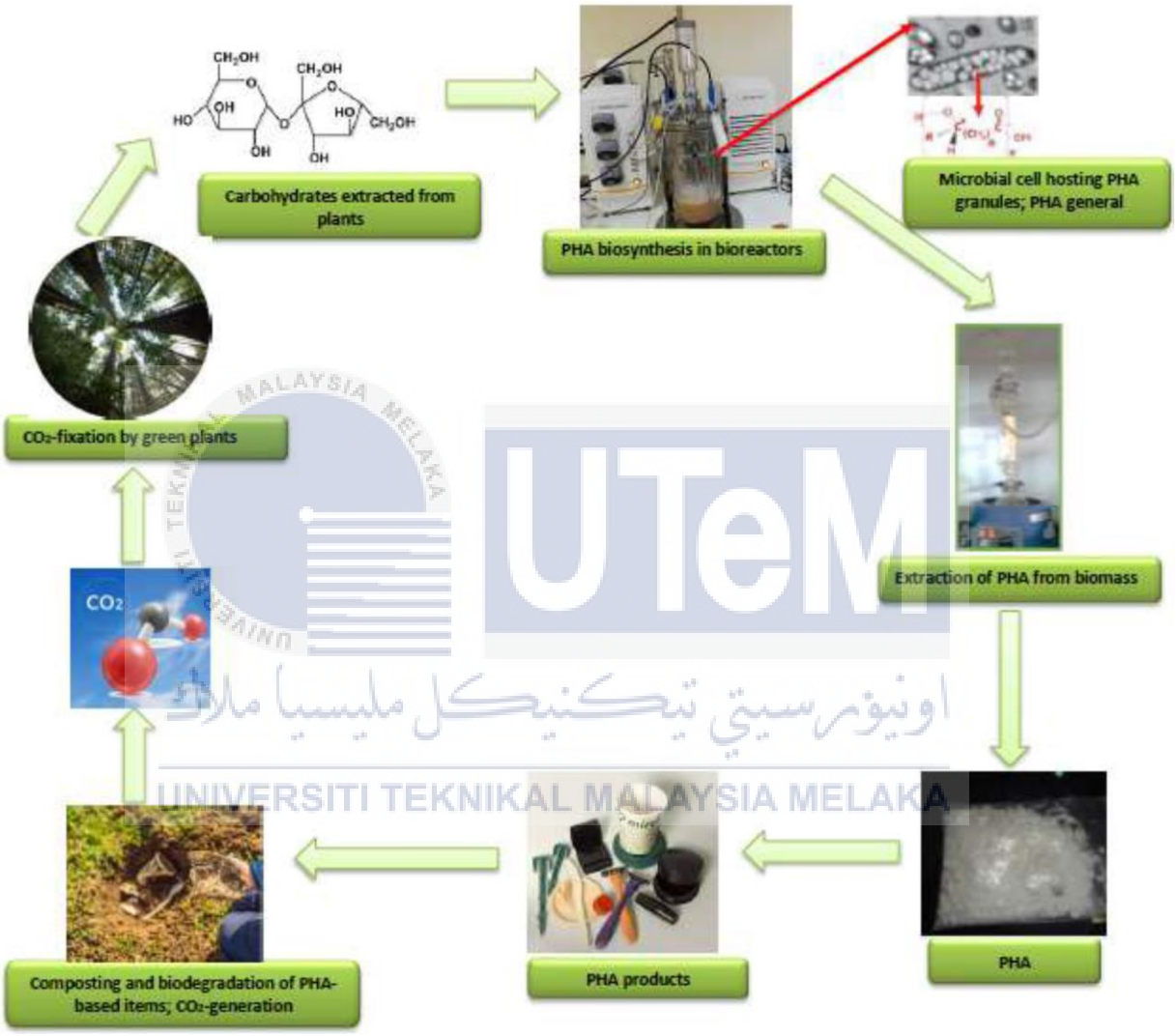

Figure 2.4: Cyclic biological process of PHA (Ibrahim et al., 2021).

Poly (3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV), is one of the materials comes from polyhydroxyalkanoate (PHA) polymer family, have been the subject of recent advances in biodegradability research. It is a copolymer composed of 3-hydroxybutyrate (3HB) and 3-hydroxyvalerate (3HV) monomeric units. The incorporation of HV into the polymer chain alters its properties, making it more flexible and less crystalline compared to

the homopolymer poly(3-hydroxybutyrate) (PHB) (Meereboer et al., 2020). Figure 2.5 shows the molecular structure of PHBV.

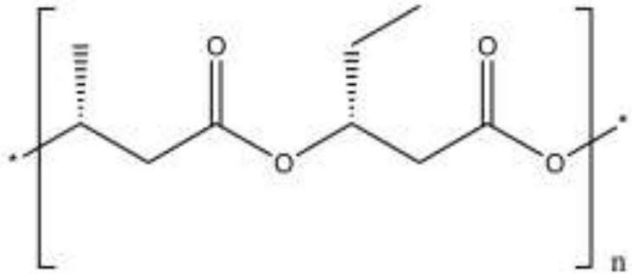

Figure 2.5: Molecular structure of PHBV. Adapted from (Ibrahim et al., 2021).

PHBV exhibits several advantageous properties, including biodegradability, biocompatibility, and being a biobased polymer derived from renewable resources . These features make PHBV a promising candidate for various applications, particularly in the biomedical and pharmaceutical fields, such as drug delivery systems, bone scaffolds, implant coatings, and tissue engineering (Ibrahim et al., 2021). With the reinforcement with natural fiber, PHBV biocomposites have high demands in markets like packaging materials, disposable items, automotive parts, and medical devices due to their fully biodegradable nature and high mechanical properties (Ten et al., 2015).

The biodegradability of PHBV in the microbial perspective highlight PHBV as an energy source. In microbially active environments, enzymes secreted by microorganisms break down PHBV into segments used as a carbon source for microbial growth. Biodegradation rates depend on factors like surface area, microbial activity, pH, temperature, and moisture. PHBV degrades rapidly in anaerobic sewage and slowly in seawater, with end products being carbon dioxide, water, and humus in aerobic conditions. Specific environments like simulated landfills and anaerobic sewage demonstrate the biodegradability of PHBV, making it an environmentally friendly polymer (Luzier, 1992). Figure 2.6 below shows the PHBV cycle from the process of synthesis, until the process of biodegradation.

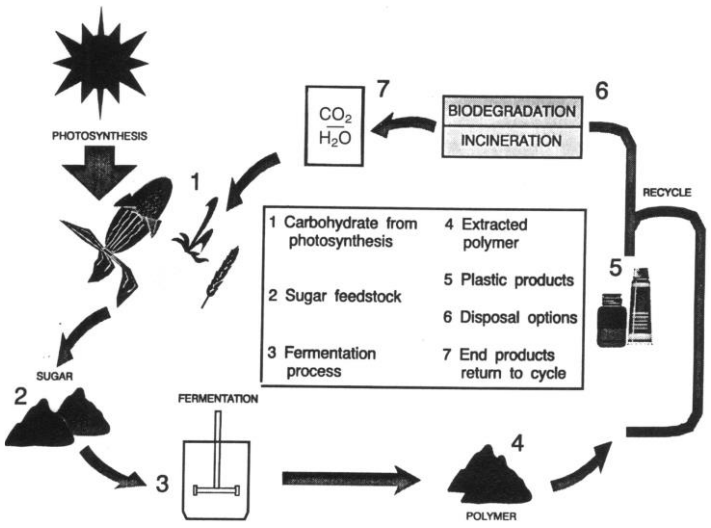

Figure 2.6: Life cycle of PHBV. (Luzier, 1992)

However, when compared to other biodegradable or commercial polymers, commercial PHBV have limitations such as low impact strength, high brittleness, low thermal stability, considerable hydrophobicity, and poor flexibility, making it unsuitable for certain industrial and commercial applications (Ibrahim et al., 2021). According to Technical Data Sheet of PHBV (ENMAT Y1000), the mechanical properties of PHBV was shown in the Table 2.1 below:

Table 2.1: PHBV mechanical properties. Adapted from (Technical Data Sheet PHBV ENMAT Y1000).

Mechanical Properties	Values
Tensile Strength (MPa)	36-39
Young's Modulus (MPa)	1600-2100
Elongation at Break (%)	1-3

To address the drawbacks of PHBV, the forming of bio-composites where the incorporation of natural fiber can improve mechanical properties compared to virgin polymer itself. The properties are dependent on the aspect ratio, fiber modulus, morphology, and interfacial adhesion of the fiber to the polymer. These bio-composites have been explored for applications in packaging, reinforced materials, and biomedical fields. (Ibrahim et al., 2021; Meereboer et al., 2020).

2.3 Natural Fiber as Reinforcement

The growing attention to natural fiber as a reinforcement in polymer composite is significantly increasing in industrial applications. Natural fibers, derived from sources such as plants and wood, have gained popularity due to their advantage including renewability, widespread availability, biodegradability, cost-effectiveness, recyclability, and low density. With these advantages, they were picked as the most ecologically sustainable and appealing substitute with traditional materials like carbon, synthetic fibers, and glass in the preparation of composites. The classifications of natural fiber are shown in Figure 2.7. Plant fibers such as cotton, flax, hemp, sisal, jute, kenaf, ramie, pineapple, banana, bamboo, and wood, are frequently used as reinforcement materials in composite fabrication (Lakshmi Narayana & Bhaskara Rao, 2021).

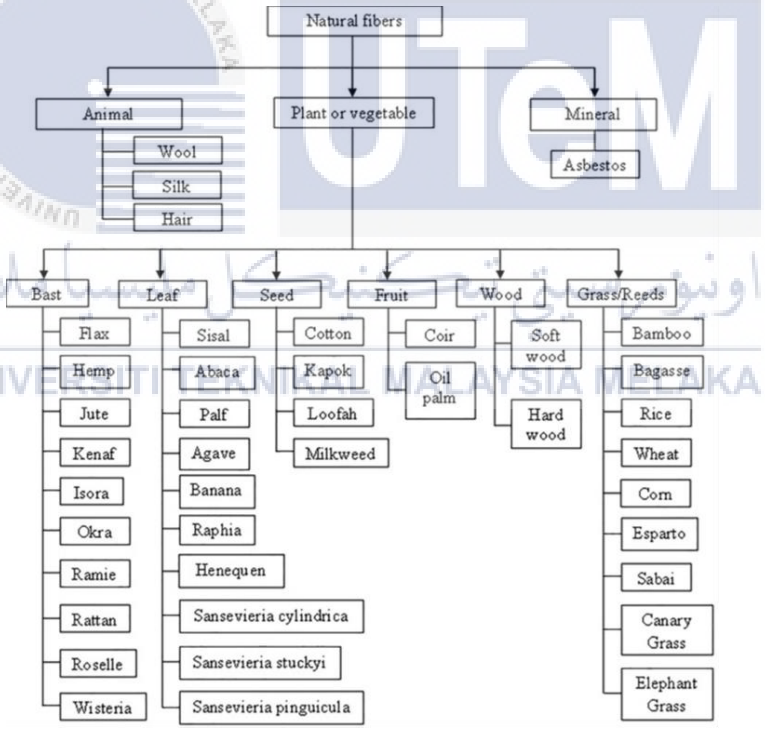

Figure 2.7: Classification of Natural Fibers. (Lakshmi Narayana & Bhaskara Rao, 2021)

Natural fibers from plant consists of cellulose, hemicellulose, lignin, pectin, and other smaller impurities. The structure of natural fiber is shown in Figure 2.8:

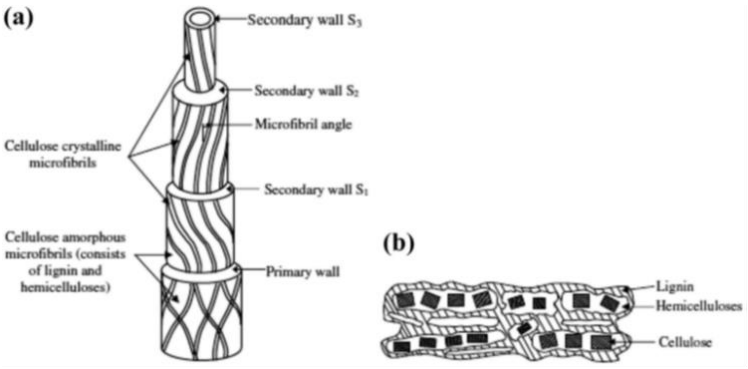

Figure 2.8: (a) Structure of natural fiber. (b) Constituents of natural fiber. (Samanth & Subrahmanya Bhat, 2023)

In natural fibers, there are several main ingredients such as cellulose, hemicellulose, lignin, pectin, waxes, and some water-soluble compounds. The complicated structure of lignin is linked by an aromatic ring attached to it, functioned to protect the cellulose and hemicellulose matrices and to improve their elasticity (Samanth & Subrahmanya Bhat, 2023). Table 2.2 below shows the chemical composition of selected natural fibers

Table 2.2: Chemical composition of natural fibers. Adapted from (Mohamed et al., 2021).

Fiber	Cellulose, (wt%)	Hemicellulose, (wt%)	Lignin, (wt%)	Waxes, (wt%)
Alfa	45	39	7-9	2
Bamboo	26-43	30	21-31	-
Banana	63-64	19	5	-
Cotton	85-90	6	-	0.6
Flax	71	19-21	2	1.5
Hemp	68	15	10	0.8
Jute	61-71	14-20	12-13	0.5
Kenaf	72	20	9	-
Nettle	86	10	-	4
Oil Palm	65	-	29	-
Pineapple Leaf Fiber	70-82	12.3	3.5	-
Sisal	65	12	10	2
Straw (wheat)	38-45	15-31	12-20	-

Due to the presence of these components, natural fibers have high moisture absorption, poor compatibility, and thermal degradation. These properties make them undesirable in several composite applications (Lakshmi Narayana & Bhaskara Rao, 2021).

2.3.1 Pineapple Leaf Fiber

One of the most major tropical crops in Malaysia is the pineapple, and PALF is a byproduct of pineapple harvesting. Consequently, PALF can be extracted from pineapple leaves by scraping, removing, or peeling them, and it can be utilized for necessary duties for no further cost. Figure 2.9 shows the fibers that obtained from pineapple leaf.

Figure 2.9: Fibers from pineapple leaf. (Mohamed et al., 2021)

As a natural fiber, PALF has special mechanical qualities that should be thoroughly studied for use in a variety of fields of study, such as materials science and engineering. PALF has a cellulose content of 70-82% and a fiber structure that is roughly 83% like cotton. The composition of hemicellulose (12.3%) and lignin (3.5%) are both low in PALF. Moreover, the low microfibrillar angle (8−15°) and high amount of alpha-cellulose in PALFs contribute to their outstanding mechanical properties. Due to these unique qualities, PALF

is an excellent substitute raw material that may be used by industries to reinforce composite matrixes (Mohamed et al., 2021). Table 2.2 below shows the values of mechanical properties of PALF.

Table 2.3: PALF mechanical properties. Adapted from (Ho et al., 2012; Mohamed et al., 2021).

Mechanical Properties	Values
Tensile Strength (MPa)	170-1627
Young's Modulus (GPa)	60-82
Elongation at break (%)	2.4

2.4 Surface Treatment

Natural fibers are incompatible with thermoplastics due to their hydrophilic nature, which is the main drawback of employing them as reinforcement with hydrophobic material. This feature causes poor interfacial contact between the fibers and the matrix, which in turn causes the mechanical properties of the composite to be less than ideal. To mitigate this problem and improve the composites' overall performance, natural fibers must be altered to decrease their hydrophilicity (Thyavihalli Girijappa et al., 2019).

Surface treatment is employed on natural fibers with the aim of improving their characteristics. This process includes clean the surfaces and geometry of the fiber, eliminating impurities, strengthening the fibers, and reduce the moisture content. These modifications can improve the interaction between the fibril and the matrix. The purpose is to optimize the physical properties of natural fibers, making them more suitable for a variety of applications such as automotive, construction and packaging sectors (Mohammed et al., 2022; Samanth & Subrahmanya Bhat, 2023).

2.4.1 Alkaline Treatment

The outside layer of the natural fiber cell wall is covered with natural oils, lignin, pectin, and waxy components. Sodium hydroxide (NaOH) is one of the chemical reagents used in the chemical treatment process, which modifies the structure of natural fibers. By alkalizing the surface and removing the cellulose in the plant fibers, the alkaline reagent modifies the structure of the cellulose (Thyavihalli Girijappa et al., 2019).

To improve the hydrophobicity and adhesion properties of natural fiber to the matrix, the alkaline treatment approach is one of the simplest, most affordable, and effective options. Natural fibers that undergoing an alkaline treatment will breaks down fiber bundles into smaller fibers and fibrillation occur. As a result, a rough surface topography develops, enhancing mechanical properties and fiber matrix adhesion (Mohammed et al., 2022). Figure 2.10 below shows the interfacial adhesion mechanisms between fiber and matrix after alkaline treatment.

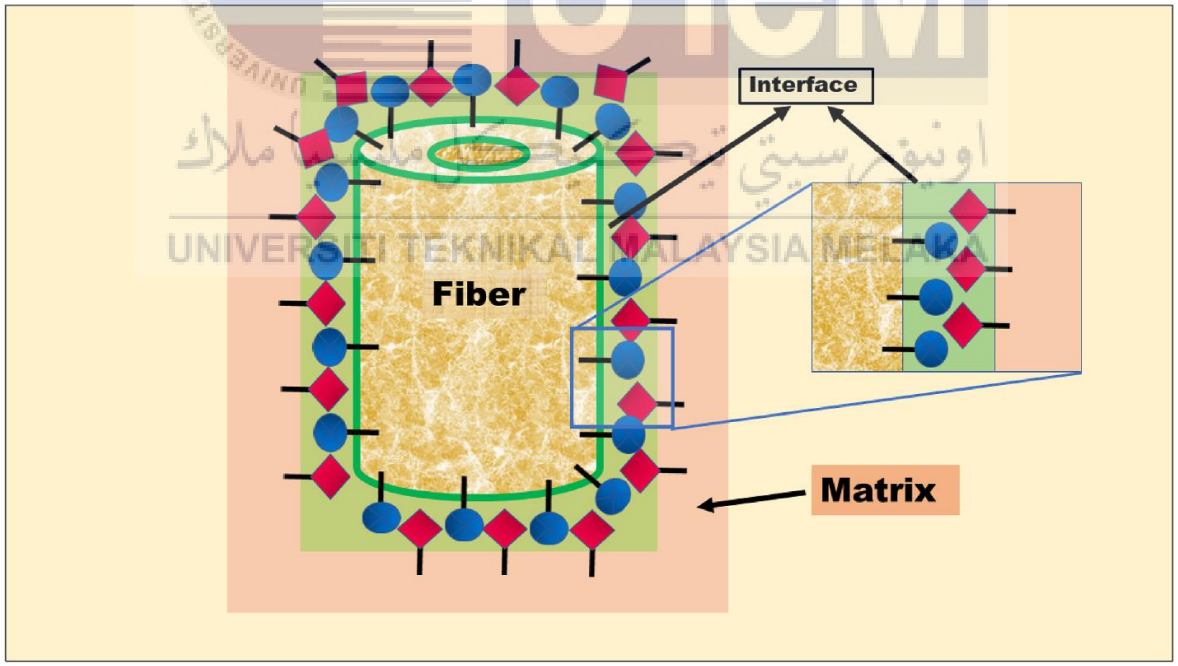

Figure 2.10: Interfacial chemical bonding mechanisms between fiber and matrix. (Mohammed et al., 2022)

Related to the Equation 1 below, the hydroxyl groups (OH) that came from hemicellulose and lignin will react preferentially with alkali during alkali treatment, resulting in the natural fiber's disintegration. The natural fiber will have an ability to withstand moisture and hydrophilic hydroxyl groups were reduced. Moreover, the tensile strength, impact strength, flexural strength, and modulus of elasticity will be improved after the alkaline treatment process (Samanth & Subrahmanya Bhat, 2023).

$$\text{Fiber-OH} + \text{NaOH} \rightarrow \text{Fiber-O-Na} + \text{H2O} \qquad \text{(Equation 1)}$$

2.5 Fiber Architecture

Several factors influence composite mechanical properties, with natural fiber architecture plays an important role. This includes factors such as cellulose concentration in the filler, fiber length, fiber loading or volume fraction, fiber orientation, and the interfacial adhesion between polymer matrix and natural fiber (Mohamed et al., 2021).

2.5.1 Fiber Loading

The influence of fiber loading on the tensile mechanical properties and moisture absorption of biodegradable composites has been extensively studied when the tensile strength of flax fiber-reinforced polylactic acid composites was affected by fiber volume fraction (25%, 30%, 35%, 40%, 45% and 50%) highlighting the significance of fiber loading in determining mechanical properties. Moreover, it was noted that in this research fiber content should not exceed 35%, as water intake increases exponentially until 50% (Singh et al., 2021).

Next, the mechanical properties and water absorption of kenaf/pineapple leaf fiber-reinforced polypropylene (PP) hybrid composites, emphasizing the importance of fiber loading in influencing tensile properties, flexural properties, impact properties, and water absorption properties. The results demonstrated that the proportion of fibers significantly

influenced the mechanical properties of composites. Introducing PALF (pineapple leaf fiber) in hybrid composites had a positive impact, leading to increased mechanical strength as the PALF content increase. This improvement was attributed to PALF's higher cellulose content, enhancing its inherent strength. While PALF has high moisture absorption due to its cellulose content, incorporating kenaf fiber in hybrid composites improved moisture resistance because of kenaf's lower cellulose and higher lignin content. Combining kenaf and PALF in hybrid composites not only enhanced mechanical properties but also improved water resistance in the resulting composite materials (Feng et al., 2020).

2.5.2 Length of Fiber

In addition to fiber loading, the length of fibers also is another crucial factor that influences the mechanical properties of the composites. In the making of composites, the directions of long fibers are much easier to control comparing to short fibers. Due to the random orientation of short fibers, composite samples would have a brittle property comparing to long fibers. However, flexural modulus of the short PALF sample was 25 times higher than long PALF composite. It was observed that as the fiber length decreases with increasing fiber content, the elongation at break will be reduced (Todkar & Patil, 2019).

The effect of short fiber length on composites was discovered by Capela et al. (2017) when the influence of the fiber length (2, 4 and 6 mm) on mechanical properties of highly reinforced epoxy resin was observed. Tensile strength and stiffness of the composites improved, but only marginally. As fiber length went from 2 to 4 mm, stiffness increases by 25%, and when fiber length reached 6 mm, it tended to decrease. Regarding the tensile strength, a similar trend was noted. Poor fibers dispersion and disorder were achieved for high fibers content composites, particularly for 6 mm long fibers. When the fiber length went from 2 to 4 mm, the storage modulus increased and continued to grow marginally until it reached 6 mm.

2.6 Water Absorption Mechanism in Natural Fiber Based Composites

Biodegradable composites, particularly those involving biodegradable polymer and natural fibers, can absorb moisture in humid conditions or when immersed in water, leading to poor stress transfer efficiency within the natural fiber-polymer matrix interface. The moisture absorption by natural fibers will affects their physical, mechanical, and thermal properties. Biocomposites typically absorb 0.7–2% of water after 24 hours, 1–5% after a week, and up to 18–22% for over several months.(Dittenber & Gangarao, 2012).

The capacity of natural fibers to absorb moisture is dependent on their inherent hydrophilic nature. Natural fibers are composed of numerous hydroxyl groups (-OH), primarily found in hemicellulose and cellulose. While these components contribute to moisture absorption, lignin, another component in natural fibers, exhibits hydrophobic characteristics with a low -OH-to-C ratio. Cellulose and hemicellulose, in contrast, possess high -OH-to-C ratio, making them accessible to water absorption when their hydroxyl groups are exposed (Mokhothu & John, 2015).

Water absorption process by natural fibers starts with the fibers swelling, a result of water molecules fills the spaces between microfibrils, then creating a temporary microcapillary network. This absorption happens in two ways: (1) water forms hydrogen bonds with hydroxyl groups (-OH), binding to cell walls and middle lamella, and (2) free water fills micro- and macropores in cell walls, retained by capillary forces. Based on Figure 2.11, as water vapor increases, it can penetrate the cell walls and middle lamella through hydrogen bonds, causing significant cell swelling. Bound water saturates the cell until the water saturation point is reached. The cellular cavities like lumens and porosities become fully saturated as they are filled by free water (Nurazzi et al., 2021).

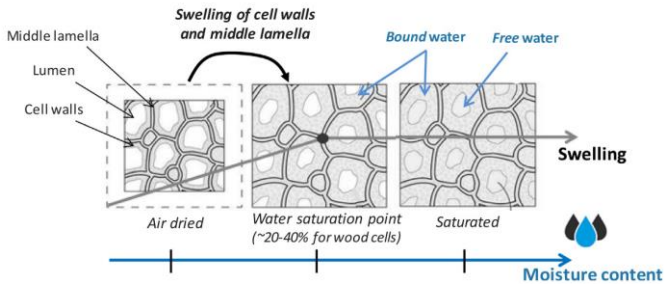

Figure 2.11: Water absorption in natural fibers. (Nurazzi et al., 2021)

Fickian diffusion is the process of water spreading from regions of higher to lower concentration, driven by a concentration gradient. Figure 2.12 shows graphs of Fickian and non-Fickian diffusion. Water absorption and desorption patterns at room temperature follows Fickian behaviour, characterized by linear water uptake in the beginning, slowing, and approaching saturation over time. While at higher temperatures of immersion, non-Fickian behaviour is observed, attributed to differences in sorption behaviour and water molecule states in the composites. The diffusion coefficient, indicating solvent molecule movement among polymer segments, increases with rising temperature, causing microcracks and an increase in the permeability coefficient (Azwa et al., 2013).

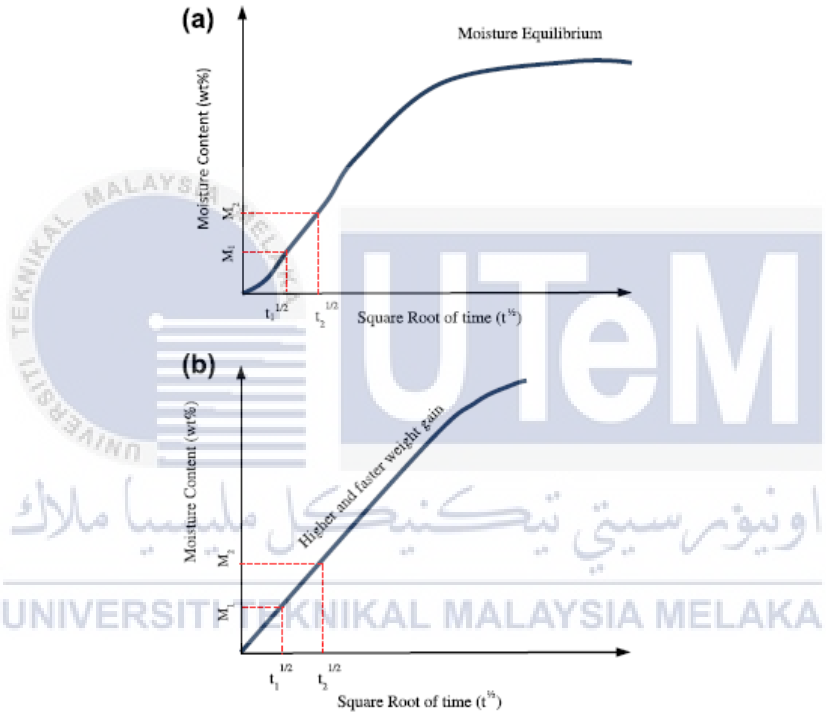

Figure 2.12: (a) Fickian diffusion (room temperature). (b) Non-Fickian diffusion (elevated temperature) (Azwa et al., 2013)

The process of debonding between the fiber and matrix begins with the formation of osmotic pressure pockets at the fiber surface, resulting from the leaching of water-soluble substances. This process is illustrated in Figure 2.13. Over an extended period, biological activities like fungi growth contribute to the degradation of natural fibers. The characteristics of natural fiber composites when immersed in water are influenced by various factors, including the properties of the fiber and matrix materials, relative humidity, and manufacturing techniques. These factors determine features such as porosity and the volume

of natural fiber content. The way in which composite materials absorb water is depends on many factors such as temperature, volume of fiber content, the orientation of natural fiber, permeability of the fiber, the area of exposed surfaces, diffusivity, the reaction between water and the matrix, and surface protection (Azwa et al., 2013).

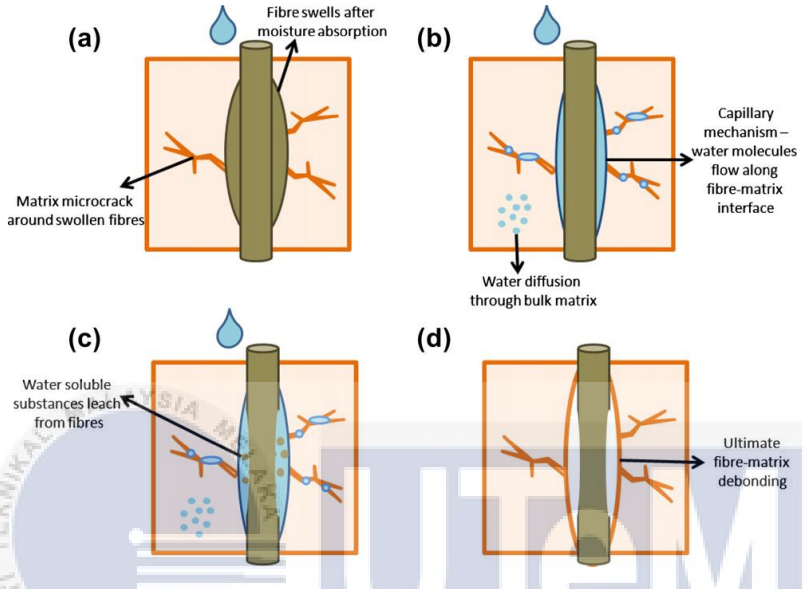

Figure 2.13: Effect of water on fiber-matrix interface. (Azwa et al., 2013)

2.7 PHBV and Natural Fibers Composites

Based on recent studies, some efforts have been made by reinforcing PHBV with natural fiber to improve the mechanical properties. A variety of techniques have been recommended, including crosslinking, reinforcing with natural and synthetic fibers, and the use of nanomaterials. Among these, the use of natural fibers to plastics have gained significance since it lowers production costs while simultaneously improving mechanical properties. The current market for biodegradable, environmentally friendly materials has resulted in the development of biopolymer composites, which are biopolymer matrix-embedded natural fibers (Rivera-Briso & Serrano-Aroca, 2018; K. Mazur & Kuciel, 2019; Varghese et al., 2020). Table 2.4 summarizes recent researches have been made related to PHBV/natural fibers composites.

Table 2.4: Summary of recent researches related to PHBV/Natural Fiber Composites.

Author/Title	Summary	Testing Methods	Findings
(K. E. Mazur et al., 2022) Mechanical, thermal and hydrodegradation behavior of poly (3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) composites with agricultural fibers as reinforcing fillers	The study explored how agricultural fibers like nettle fibers, pine cones flour, and walnut shells flour, affect the mechanical, thermal, and hydrodegradation properties of PHBV composites.	- Tensile Test. - Flexural Test. - Charpy impact test. - TGA. - DSC. - Water Uptake Test. - Mechanical Testing after Immersion	- Stiffness at elevated temperature were improved, as well as specific modulus and specific strength. - All the composites have favourable thermal properties, and degradation process between 200-300 °C. - Fast degradation in water, indicating their potential for reduced composting time and improved biodegradability. - Nettle fibers and walnut shells flour showing the most promising results in mechanical and thermal properties.
(Kuciel et al., 2019) Novel Biorenewable Composites Based on Poly (3-hydroxybutyrate-co-3-hydroxyvalerate) with Natural Fillers	The study combining PHBV with natural fillers like nanocellulose, walnut shell flour, eggshell flour, and tuff. It aimed to assess their mechanical and thermal properties for potential applications in industries such as automotive and medicine.	- Tensile Test (ISO 527-1:2012) - Flexural Test (PN-EN ISO 178) - Charpy impact test. - DSC. - TGA. - SEM. - Density	- Those containing nanocellulose and eggshell flour shows increment on tensile strength and modulus of elasticity. In contrast, composites with walnut shell flour and tuff enhanced flexural modulus and flexural stress at 3.5% strain. - Biocomposites had higher crystallinity compared to unmodified PHBV. - Natural fillers affected the melting temperature (T_m) and melting enthalpy of the biocomposites.Thermal stability was also enhanced in the biocomposites by using TGA. - The composites absorbed more water over time, leading to a decrease in tensile strength and modulus of elasticity after exposure to water. - Composites with natural fillers degraded faster than unmodified PHBV.
(K. Mazur & Kuciel, 2019) Mechanical and hydrothermal aging behaviour of polyhydroxybutyrate-co-valerate (PHBV) composites reinforced by natural fibers	The research focuses on the mechanical and hydrothermal aging behavior PHBV composites reinforced by natural fibers. The study investigates the effects of incorporating Wood Fibers (WF) and Basalt	- SEM. - Thermal expansion measurements. - Tensile test - Bending test - Impact test	- WF in composite resulted in a decrease in flexural strength and an increase in flexural modulus across all temperature. - BF enhanced all mechanical properties (flexural strength and flexural modulus) at every temperature.

	Fibers (BF) on the properties of the composites.		- Weight gain rate percentage changed over time for PHBV composites with WF and BF in saline solution at 40°C. - The composites containing lignocellulosic fillers showed the highest water absorption (WF and BF) - Neat PHBV had the lowest water absorption. - The presence of fibers influenced the rate of water absorption, with composites having 15% by weight of WF showing the highest absorption rate. - Mechanical properties slightly decreased by around 10% after both the first and second weeks.
(Conceição et al., 2023) A Biodegradable Composite of Poly (3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) with Short Cellulose Fiber for Packaging	This research focuses on the development of a biodegradable composite material for packaging using Poly (3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) and short cellulose fibers. The study addresses the problem of improper disposal of polymeric waste by providing a sustainable alternative for packaging materials.	- FTIR. - TGA. - DSC. - Contact angle measurements using the force-based method and the sessile drop method to determine the wettability of the samples. - XRD. - Impact resistance tests (ASTM D256). - Vicat softening temperature test (ASTM D-1525)	- Increased cellulose content led to reduced molecular mobility, heightened crystallinity, decreased impact strength, and increased agglomerate formation, acting as stress concentrators. - FTIR analysis indicated that cellulose fibers served as nucleating agents, evidenced by the reduced intensity of specific peaks.
(Varghese et al., 2020) Novel biodegradable polymer films based on poly(3-hydroxybutyrate-co-3-hydroxyvalerate) and Ceiba pentandra natural fibers for packaging applications	The research focuses on the development of biodegradable polymer films for packaging applications using PHBV polymer composites with Ceiba pentandra natural fibers. The study investigates the effects of these fibers on various properties of the polymer composites.	- Tensile Test (ASTM D638-14) - TGA. - DSC. - FTIR - Biodegradability Test (15,30, and 45 days) - UV visible spectrophotometer - Food Packaging application	- Adding sisal and coconut fibers made the films stronger, based on tensile strength tests. The films showed good flexibility and elongation properties. - The films had antimicrobial properties, preventing the growth of Escherichia coli (E. coli) and Staphylococcus aureus (S. aureus). - After 7 days of storage, researcher compared the quality of the strawberries in the films with those left unpacked. The films successfully preserved the freshness and quality of the strawberries, suggesting their suitability for food packaging. - The films demonstrated strong thermal stability, experiencing minimal weight loss until reaching a temperature of 700 °C.

			- Sisal and coconut fibers enhanced the films' crystallinity, suggesting better structural properties. - The findings indicated that the films are biodegradable, as they showed a decrease in weight over time. This suggests their potential as environmentally friendly materials."
(Zaidi & Crosky, 2019) Unidirectional rubber-toughened green composites based on PHBV	The study aims to improve the mechanical characteristics of the bio-based polymer poly(hydroxybutyrate-co-valerate) (PHBV). This is achieved by strengthening it with unidirectional flax and adding toughness through the use of either poly(butylene adipate-co-terephthalate) (PBAT) or epoxidized natural rubber (ENR)	- TGA. - Tensile Testing. - Impact Testing. - SEM. - DSC.	- Adding unidirectional flax to PHBV composites led to considerable improvements in tensile, flexural, and impact properties but minimal changes in thermal properties. - Impact resistance of the composites increased, where the original impact resistance of neat PHBV was 1.4 kJ/m2, which was increased 24 to 37 – fold in the composites. - The microstructure characterization revealed that the rubber particles were well bonded to the PHBV matrix, while the fibers were only weakly bonded to the matrix in all the composites. - Different behavior observed in the PHBV/ENR/flax composites, and this behavior was linked to the larger size of the ENR particles.

2.8 PHBV Composites Applications

In the field of engineering materials, PHBV demonstrated comparable performance to traditional plastics like polypropylene, indicating their suitability as eco-friendly alternatives for various industrial purposes, including applications in semirigid packaging and the construction of lightweight automobile body panels (Zhao et al., 2021). By incorporating magnetostrictive cobalt ferrites (CFO) into the PHBV matrix, magnetically responsive composites are produced, proving effective in cell and tissue differentiation for tissue engineering applications (Amaro et al., 2020)

Related to the natural fiber in composites, PHBV and Natural Rubber (NR) has been developed to address the limitations of pure PHBV, making it a potential substitute for petroleum-based plastics in food packaging. Blending PHBV with 10-15% NR achieves the best balance of processing and performance properties, like traditional plastics. These blends have the potential to be used in the production of compostable freezer-to-microwave food packaging (Zhao et al., 2019).

Moreover, lignocellulosic fibers obtained by dry grinding of three different agricultural residues (wheat straw, brewing spent grains, and olive mills) were compared for their potential use as fillers PHBV for food packaging. The results suggest that PHBV composites with wheat straw fibers hold promise for packaging respiring food products, while PHBV composites with olive mills fibers may be better suited for products sensitive to water (Berthet et al., 2015).

In conclusion, PHBV composites have shown great potential for a wide range of applications, including tissue engineering, automobile, and food packaging. This ongoing research aims to further evaluate the mechanical, biocompability, and moisture resistance of the PHBV and PALF composites focusing in packaging applications.

CHAPTER 3

METHODOLOGY

3.1 Introduction

This chapter plays a crucial part in this research, explaining the methods used to achieve the research objectives. The process flowchart (Figure 3.1) function as an organizational tool, outlining methodology of the research. The methodology starts with the surface treatment of Pineapple Leaf Fiber (PALF) to enhance adhesion with Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV). FTIR analysis is performed to determine the impacts of the surface treatment of PALF. Then, the compounding process of the samples focused on project scope by using PALF with random fiber orientation with length less than 6mm and fiber loadings of 0%, 10%, 20% and 30%. Next, a series of tensile tests with varying fiber loadings are performed. After that, morphological analysis is performed, which involves analysing the fracture surfaces of the composites using techniques such as Scanning Electron Microscopy (SEM). For another series of samples, another test was performed which is Water Absorption Test, for determining the composite performance over a specific time which is 14 days and under specific temperature. Finally, tensile test was performed to analyse the effect of moisture on the tensile mechanical properties. This methodical approach seeks to provide complete understanding into the effects of fiber loading on the tensile mechanical properties, effect of moisture on tensile mechanical properties, and morphological properties of PHBV/PALF composite.

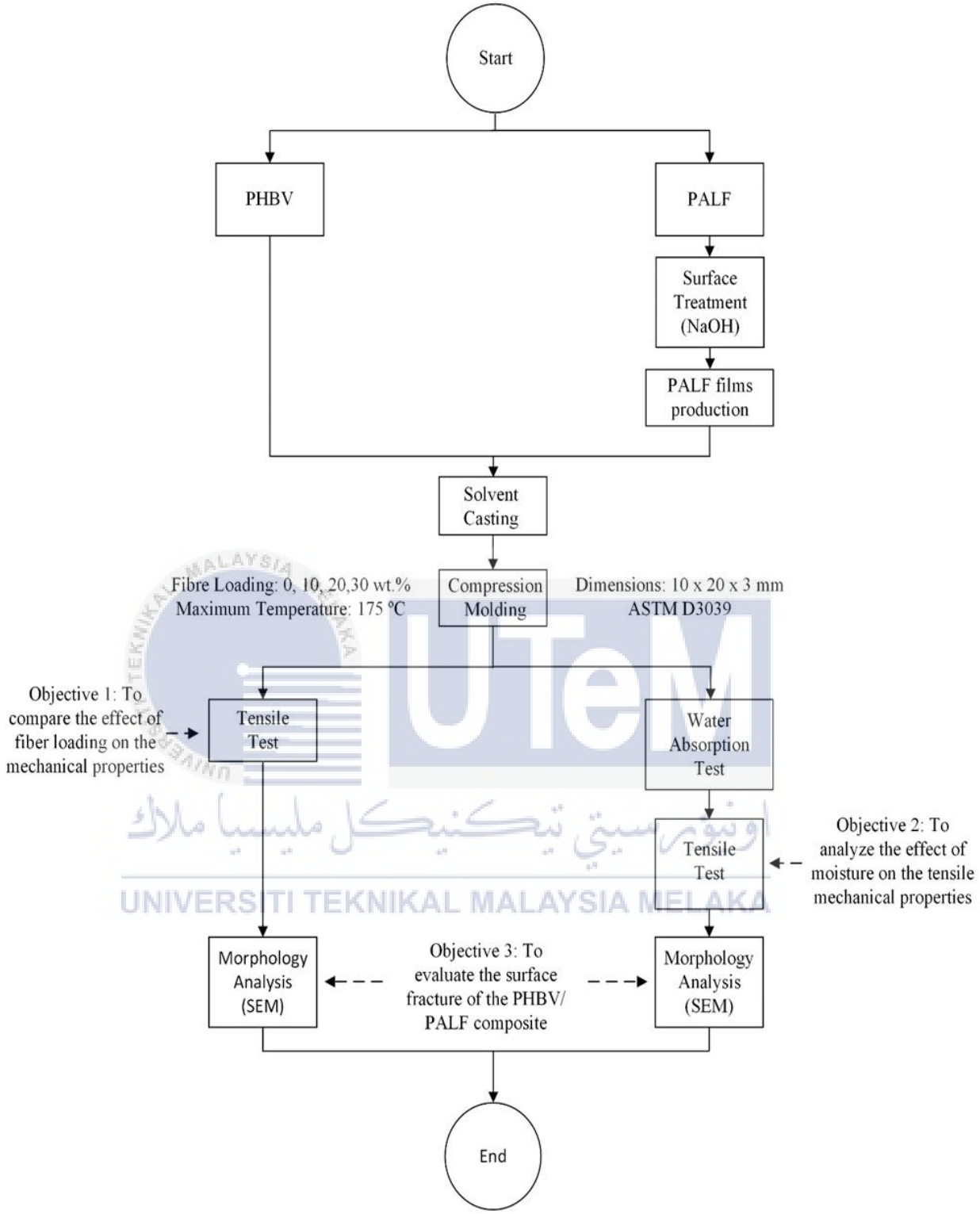

Figure 3.1: Methodology Flow Chart.

3.2 Materials

In methodology, materials that were used are Poly (3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV), Pineapple Leaf Fiber (PALF), and Sodium Hydroxide (NaOH). PHBV functions as matrix, while PALF is utilized as reinforcement element, and NaOH is used in surface treatment process.

3.2.1 PHBV

PHBV (ENMAT Y1000) (Figure 3.2) was purchased from Tianan Biologic Material Company LTD (China) was used as a matrix. It is in the form of powder and it is 100% biodegradable polymer with the following properties: specific gravity – 1.25, melt flow index – 1-5, yield stress – 31-36 MPa, Young's Modulus – 1600-2100 Mpa, and flexural modulus – 800-1100 MPa.

Figure 3.2: Poly (3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV).

3.2.2 PALF

Pineapple Leaf Fiber (PALF) (Figure 3.3) was obtained from a local supplier as a byproduct of pineapple cultivation. The PALF is in long fiber with a density of 1.526 g/cm^3. Then, continuous PALF were cut with length below 6 mm for surface treatment.

Figure 3.3: Pineapple Leaf Fiber (PALF).

3.2.3 NaOH

Sodium Hydroxide (NaOH) solution was obtained from Sigma Aldrich (Figure 3.4) with the following properties: Density – 1.320-1.340 g/cm3, Vapor Pressure – 3 mmHg at 37 °C and pH Value – 14.

Figure 3.4: Sodium Hydroxide (NaOH).

3.3 Surface Treatment

In surface treatment process, Untreated PALF were soaked with 5% Sodium Hydroxide (NaOH) solution for surface treatment process at room temperature for 1 hours. Then, PALF were washed several times for removing alkali until the colour of litmus paper indicate pH 7. Finally, fibers were dried in drying oven at 80 °C for 48 hours to removes moisture.

3.4 PALF films production

Short PALF were pulverized using a blender (Figure 3.5) to ensure the short PALF (below 6mm) are evenly distributed in distilled water. Then, the slurry is poured into a paper mould, which shapes it into a thin film of short PALF with dimensions of 100 x 200 mm and a weight of 1 gram per film. The thickness of the film can be adjusted by changing the amount of slurry in the mold (Figure 3.6). The films are dried after the process is done.

Figure 3. 5: Blender (Panasonic, Malaysia)

Figure 3.6: The procedure of PALF film production.

3.5 Sample Preparation

3.5.1 Solvent Casting

PALF and PHBV composite films were made using a solvent casting method. In the vacuum hood, chloroform was stirred with a magnetic stirrer until reach 50 ºC and PHBV powder was poured to make it completely dissolved. All apparatus of solvent casting technique is shown in Figure 3.7.

Figure 3.7: Apparatus of Solvent Casting.

After PHBV powder completely dissolved in Chloroform, the solution then was poured into the glass container for dipping PALF films until the solution finished. Each PALF and PHBV was spread on a clean glass plate and left to dry in the vacuum hood. The next day, all dried films was removed from the glass plate. The composite films then were dried in the vacuum oven (Figure 3.8) at 60 ºC for 24 hours to remove chloroform from the films.

Figure 3.8: Vacuum Oven Machine (Memmert, Germany).

3.5.2 Compression Moulding

Finally, samples (ASTM D3039) were produced by using compression moulding machine (Figure 3.9). The samples were fabricated with 10, 20, 30 wt% PALF in the PHBV matrix. The remaining mass of PHBV and PALF/PHBV films were layered in sandwich form in mould of 200 x 150 mm and compressed at 175 ºC from room temperature. The mould was left for soaking about 23 minutes at maximum temperature before cooled down to room temperature, using water quenching. The samples were then cut according to dimensions of ASTM D3039. The mass of PHBV matrix and PALF were calculated using Equation 2 and Equation 3 below, and the calculations for 0, 10,20 and 30 wt.% PALF fiber loading are shown in Table 3.1 below:

$$\text{Density of Composite, } \rho_c = V_{PALF}\rho_{PALF} + V_{PHBV}\rho_{PHBV} \qquad \text{(Equation 2)}$$

where V_{PALF} is the volume of PALF; ρ_{PALF} is the density of PALF; V_{PHBV} is the volume of PHBV; ρ_{PHBV} is the density of PHBV.

Density, $\rho = \frac{m}{V}$ (Equation 3)

where m is mass; V is volume.

Table 3.1: Calculations of PHBV and PALF

PALF Fiber Loading (wt.%)	PHBV Matrix Loading (wt.%)	Density of Composite (g/cm^3)	Mass of Composite (g)	Mass of PALF (g)	Mass of PHBV (g)
0	100	1.25	75	0	75
10	90	1.28	76.62	7.66	69.96
20	80	1.30	78.24	15.65	62.59
30	70	1.33	79.86	23.96	55.90

Figure 3.9: Compression Moulding Machine (Carver, USA).

3.6 Tensile Testing

Tensile test (ASTM D3039) for unaged samples (normal sample) and aged samples (water aging) were conducted by using the Shimadzu AGS-X Universal Testing Machine (Figure 3.10) and the crosshead speed for tensile tests was 2mm/min.

Figure 3.10: Universal Testing Machine (Shimadzu, Japan).

The temperature for performing tensile test were at ambient temperature which is at 23°C. At least five separate tests were conducted to produced average values of reading. The dimensions of samples for tensile testing are illustrated at Figure 3.11 and Table 3.2 below:

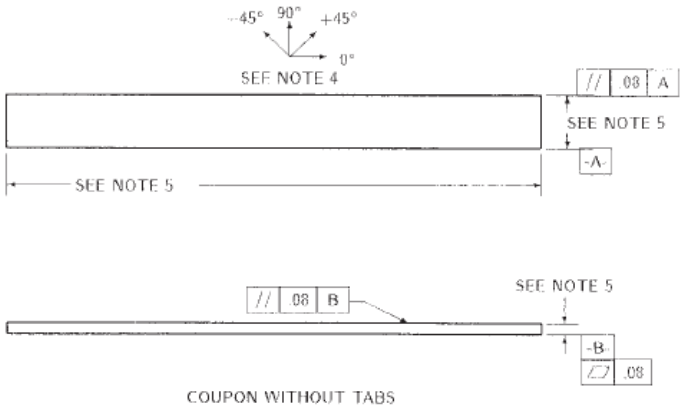

Figure 3.11: Sample dimensions. Adapted from (ASTM D3039).

Table 3.2: Sample dimensions. Adapted from (ASTM D3039).

Description	Length (mm)
Length	120
Width	20
Thickness	3
Clamping length	20 x 2

Data reading of Force (N) and Displacement (mm) were recorded for every samples. Then, the value of stress (σ), strain (ε) and Young's Modulus (E) was calculated by using Equation 4, 5 and 6 below:

$$\sigma = \frac{F}{A} \qquad \text{(Equation 4)}$$

where σ is the stress; F is the applied force; A is cross-sectional area of sample.

$$\varepsilon = \frac{\Delta L}{Lo} \qquad \text{(Equation 5)}$$

where ε is the strain; ΔL is the change in length; L_o is the original length.

$$E = \frac{\sigma}{\varepsilon} \qquad \text{(Equation 6)}$$

where E is the Young's Modulus; σ is the stress; ε is the strain.

3.7 Water Absorption Test

Water absorption tests were conducted over 14 days to simulate conditions under which the composite may be exposed to moisture (Figure 3.12). Distilled water was used, and the temperature was maintained at a constant 23°C. Samples were removed from the water daily and dabbed with tissue paper to remove excess water from the surface of the samples. Data such as initial thickness, final thickness, initial mass, and final mass were recorded. Thickness swelling and percentage of water content (Mt) for each day were calculated using Equation 7 and 8. After recording the data, all the samples were re-immersed to continue the water absorption test. At the end of the test, all the samples were dried in an oven at temperature until constant weight was achieved. Then, tensile tests were conducted to evaluate the effect of moisture on the tensile properties of the composites. The samples that underwent water absorption were labelled as aged samples, while the samples that did not undergo water absorption were labelled as unaged samples.

$$\text{Thickness Swelling} = \left(\frac{Ti - To}{To}\right) \times 100 \qquad \text{(Equation 7)}$$

where T_0 is the initial thickness of the sample before water exposure; T_i is the thickness of the specimen after water exposure.

$$Mt = \left[\frac{Wt - Wo}{Wo}\right] \times 100 \qquad \text{(Equation 8)}$$

where M_t is the percentage of water content; W_t is the instantaneous weight of the sample, and W_0 is the initial weight of the sample.

Figure 3.12: Water absorption apparatus.

3.8 Morphology Analysis

3.8.1 Scanning Electron Microscope (SEM)

Fracture surfaces of PHBV/PALF composite is analysed using Carl Zeiss Evo 50 Scanning Electron Microscope (SEM) Machine (Figure 3.13). To minimize the electrical charging while improve the quality during imaging, gold coating is applied on the fractured surfaces.

Figure 3.13: SEM Machine (Carl Zeiss, Germany).

CHAPTER 4

RESULTS AND DISCUSSION

4.1 Introduction

This chapter presents the findings of the research and will be analysed in detail. The results from thoroughly investigation, data collection, and analysis were carried out over the course of the research timeline. The characterization of Poly (3-Hydroxybutyrate-co-3-Hydroxyvalerate) (PHBV) reinforced with 0 to 30 wt % of Pineapple Leaf Fiber (PALF) were analysed using mechanical analysis. The composite's durability when exposed to moisture is tested by using water absorption test. Additionally, the surface fracture of composites was analyzed using Scanning Electron Microscopy (SEM) to examine the morphology and understand the failure mechanisms.

4.2 Effect of different fiber loading on the tensile properties of PALF/PHBV composites

The tensile properties of PHBV/PALF composites were evaluated to understand the effect of varying fiber loadings on the mechanical performance of the composite material. Tensile tests were conducted on composites with 0%, 10%, 20%, and 30% wt. PALF to determine how the increase of fiber content affected the tensile strength, modulus, and elongation at break of the composites.

4.2.1 Tensile properties

The impact of different fiber loadings on the tensile properties of PHBV/PALF composites was examined using fiber reinforcements of 0, 10, 20, and 30 wt.%. Figures 4.1, Figure 4.2, and Figure 4.3 shows the effect of fiber loading on the tensile strength, modulus, and strain at break of PHBV/PALF composites.

Based on Figure 4.1, starting with 0 wt.% PALF, or neat PHBV, the tensile strength is 29.57 MPa. At 10 wt.% fiber loading, the tensile strength slightly decreases to 28.71 MPa, and at 20 wt.% fiber loading, it increases slightly to 29.81 MPa. A similar trend of tensile strength was recorded by Ismail & Ishak (2018) when kenaf fiber incorporated into PLA. They observed that at lower fiber loadings (10 wt.%), the tensile strength did not significantly improve due to fiber-matrix adhesion and inadequate stress transfer. However, as the fiber loading increased to an optimal range, which is at 20 wt.%, the dispersion and distribution of the fibers improved, resulting in enhanced tensile strength. The minimal improvement of tensile strength value in Figure 4.1 (20 wt.%) suggests poor dispersion and fiber distribution, as well as the presence of pores and cracks in the specimens, as shown on SEM images (Figure 4.6 – Figure 4.9), which may lead to weak adhesion between the fibers and the matrix. This poor adhesion limits effective stress transfer through the matrix, resulting in reduced composite strength.

However, at 30 wt.% fiber loading, the tensile strength significantly improves to 46.74 MPa, representing an increase of approximately 58% compared to neat PHBV. This significant improvement indicates better stress transfer and effective reinforcement due to improved fiber distribution and dispersion within the composite. The SEM images (Figures 4.10 and 4.11) support this observation, revealing good fiber dispersion and enhanced adhesion between the fibers and the matrix at higher fiber loading. This effective dispersion helps to evenly distribute stress, minimize localized stress concentrations, and reduce weak points in the composite, resulting in superior tensile properties. As suggests by researcher, adding fibers to the polymer matrix generally enhances the mechanical properties of composites. Consequently, many researchers find the influence of fiber content on

the strength properties of fiber reinforced composites to be particularly significant and noteworthy (Frącz et al., 2021).

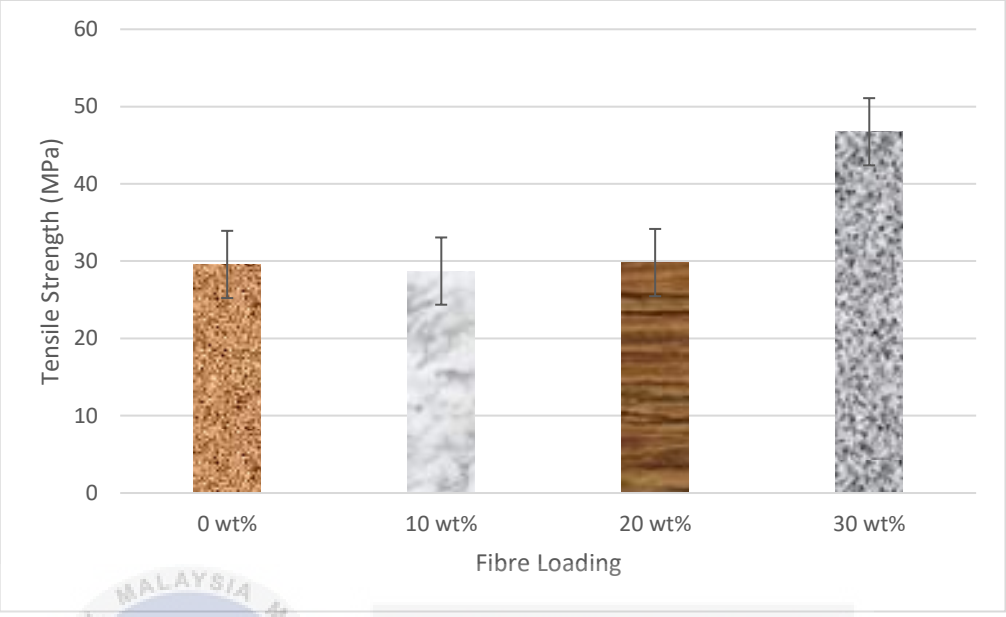

Figure 4. 1: Tensile Strength of PHBV/PALF composites at different fiber loadings.

For the modulus of the composites (Figure 4.2), 0 wt.% PALF shows the value at 0.0193 GPa, which is the highest among the samples, indicating the brittleness of neat PHBV. The SEM image (Figure 4.4) supports the higher value of modulus when there are smooth surfaces of fracture indicating brittle failure after tensile test. As fiber loading increases to 10 wt.% and 20 wt.%, the modulus decreases to 0.0087 GPa and 0.0075 GPa, respectively. This decline can be attributed to the porosity within the composites. Higher fiber loadings introduce more pores, as evidenced by the SEM images (Figure 4.7), which create points of weakness and reduce the overall stiffness of the composites. This porosity limits stress transfer and weakens the composite structure, resulting in a lower modulus.

However, at 30 wt.% fiber loading, a slight increase in modulus is observed, which is at 0.0083 GPa. This represents an improvement of approximately 10.7% compared to the 20 wt.% fiber loading. The SEM images (Figure 4.10 and 4.11) at this fiber loading demonstrate better fiber dispersion and less voids, contributing to the increasing value. This is likely because the fibers restrict the mobility of the

polymer matrix, resulting in a stiffer composite and an increase in modulus (Ismail & Ishak, 2018).

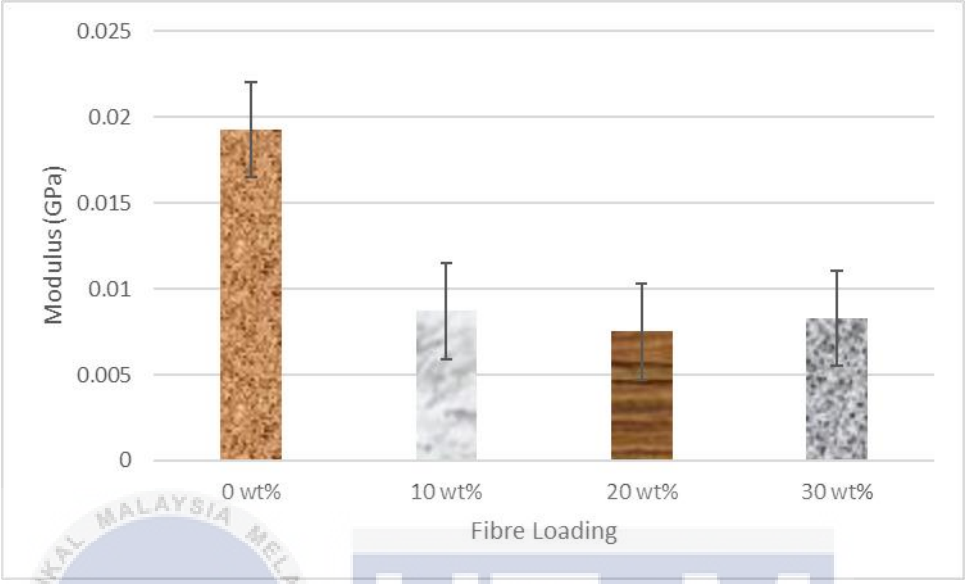

Figure 4. 2: Modulus of PHBV/PALF composites at different fiber loadings.

The elongation at break of the composites (Figure 4.3) increased to 3.57% when 10 wt.% PALF fiber was incorporated, compared to neat PHBV which had an elongation at break of 1.73%. This improvement indicates that the addition of fibers enhances the flexibility of the composite. However, when the fiber content reached 20 wt.%, the elongation at break reduced slightly to 3.38%. This reduction can be attributed to the less uniform stress distribution during deformation in bio composites with increasing fiber content, as suggested by Ismail and Ishak (2018). The non-uniform stress distribution can lead to localized failure, reducing the overall elongation at break.

Interestingly, at 30 wt.% fiber loading, the strain at break significantly improved to 6.19%, representing an increase of approximately 83% compared to the 20 wt.% fiber loading. This increase is likely due to better fiber dispersion and improved fiber-matrix adhesion, as indicated by the SEM images (Figure 4.10 and Figure 4.11). The improved distribution and adhesion reduce the stress

concentrations and enhance the composite's ability to deform uniformly, resulting in good flexibility and high elongation at break.

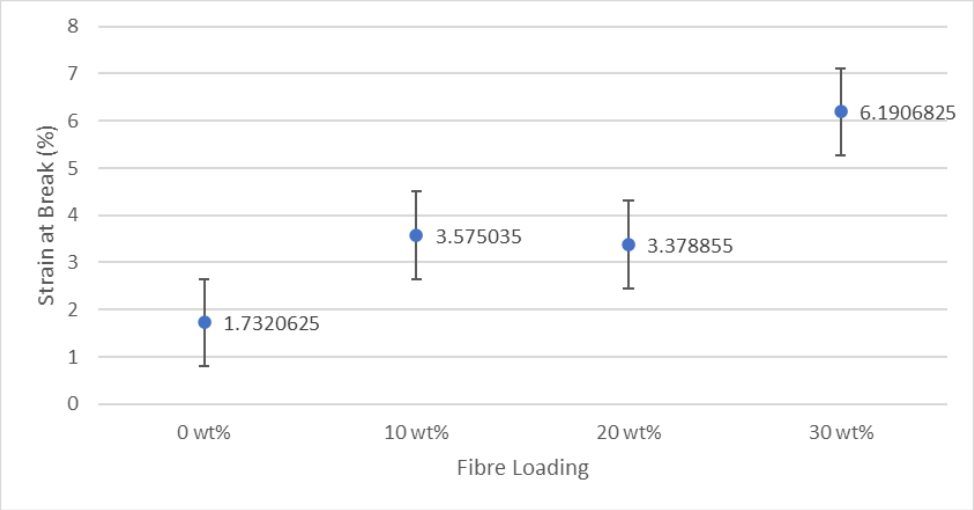

Figure 4. 3: Strain at break of PHBV/PALF at different fiber loadings.

4.2.2 Morphology Analysis

To assess the fracture surfaces of the composites after tensile testing, SEM images of 0, 10, 20, and 30 wt.% fiber loadings are shown in Figures 4.3, 4.4, 4.5, and 4.6, respectively. Morphology analysis was conducted using SEM to examine these fracture surfaces. The failure mechanisms and the interactions between the fibers and the matrix at different fiber loadings were analyzed to understand how these interactions affect the overall mechanical properties of the composites.

SEM images of neat PHBV (Figure 4.4 and Figure 4.5) show a relatively smooth fracture surface, demonstrating brittle failure without fiber reinforcement. This finding is also reported by Frącz et al. (2021) when the pure PHBV shows smooth surfaces comparing with fiber-reinforced PHBV. The higher modulus value in Figure 4.2 further supports the brittleness of the neat PHBV. This indicates that the neat PHBV lacks enough reinforcement to withstand higher stress, resulting in brittle failure, as it cannot effectively distribute the load. This results in localized stress concentrations around microcracks, as shown in Figure 4.4, which further weaken the material. When subjected to an external load, stress is concentrated around these microcracks rather than being uniformly distributed, causing premature failure in neat PHBV.

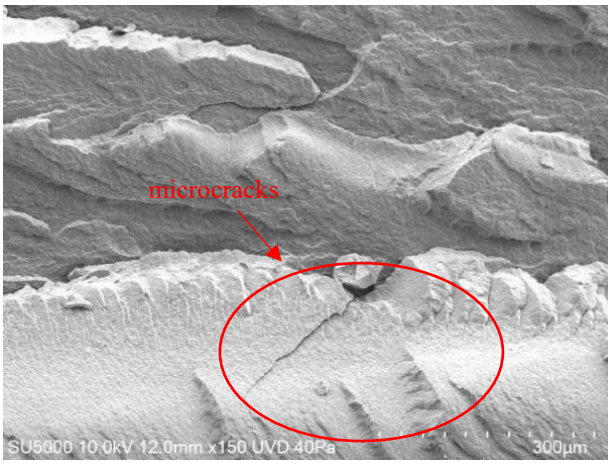

Figure 4. 4: SEM image of the fracture surfaces (Neat PHBV).

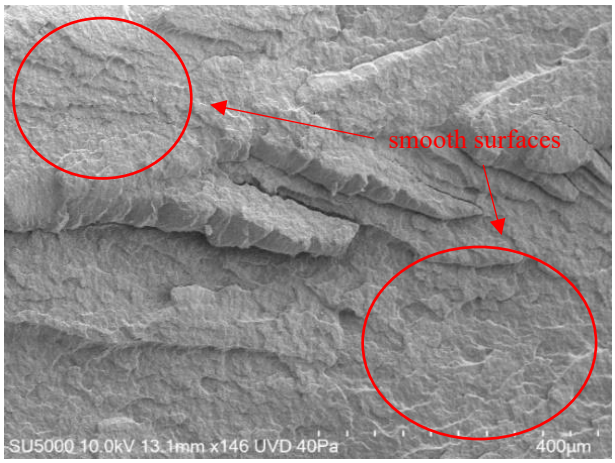

Figure 4. 5: SEM image of the fracture surfaces (Neat PHBV).

At 10 wt.% PALF loading, SEM images (Figure 4.6 and Figure 4.7) display fiber pull-out and macro-cracks in the PHBV matrix. This suggests some interaction between the fibers and the matrix but also significant matrix cracking. The presence of fiber pull-out indicates that the fibers are starting to contribute to the capacity of the composite to withstand load. Additionally, pores are observed in the composite matrix, which may result from incomplete fiber wetting or PHBV consolidation during the production process. The result of SEM images corresponds with the tensile test results (Figure 4.2), showing a decrease in value compared to neat PHBV. This finding is supported by Ismail & Ishak (2018), who reported that the tensile strength of the materials decreased due to insufficient wetting of fibers by the matrix.

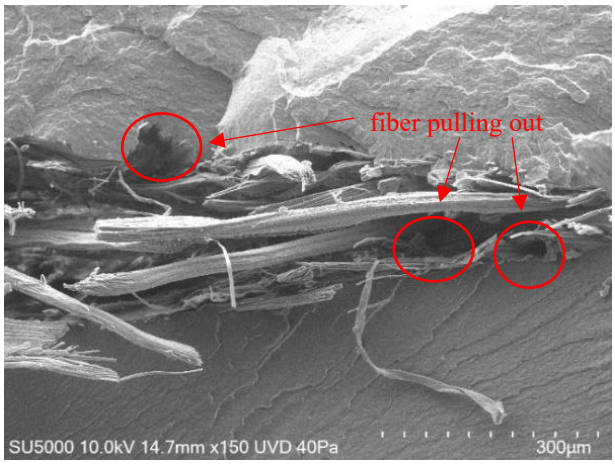

Figure 4. 6: SEM image of the fracture surfaces (10 wt.% PALF fiber loading).

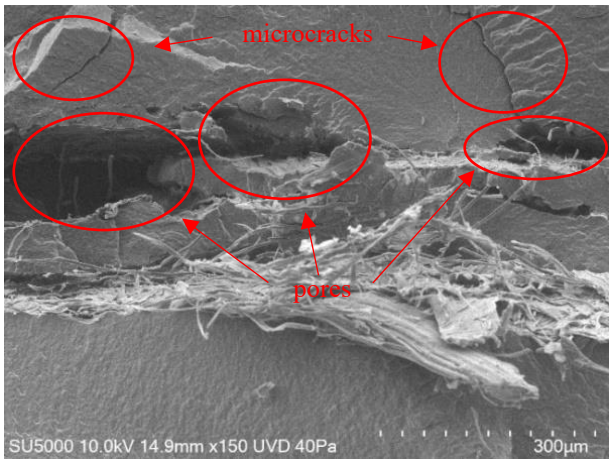

Figure 4. 7: SEM image of the fracture surfaces (10 wt.% PALF fiber loading).

SEM images (Figure 4.8 and Figure 4.9) demonstrate fiber pull-outs and breakages with a PALF loading of 20wt.%. This pull-out effect is associated with insufficient adhesion between the fibers and the matrix, impacting the mechanical properties of the composites (K. E. Mazur et al., 2022). Additionally, SEM images show areas of poor fiber dispersion, in which fibers are not evenly distributed throughout the matrix. This inadequate dispersion can cause localized stress concentrations and weak areas in the composite, further weakening its tensile properties as shown in Figure 4.2, resulting in inconsistent performance.

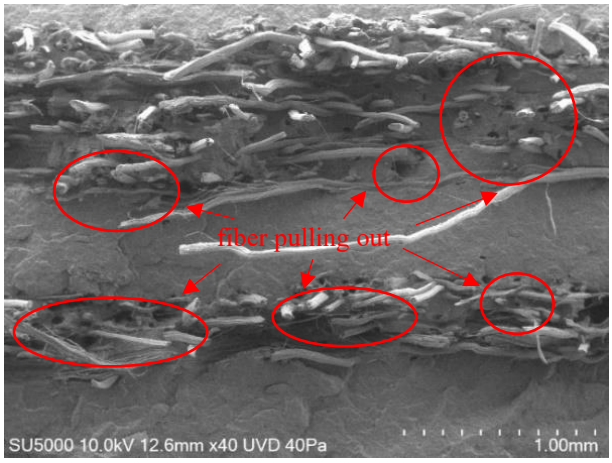

Figure 4. 8: SEM image of the fracture surfaces (20 wt.% PALF fiber loading).

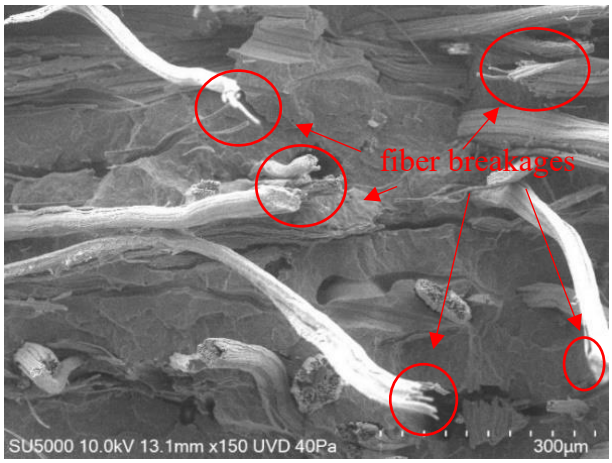

Figure 4. 9: SEM image of the fracture surfaces (20 wt.% PALF fiber loading)

At 30 wt.% PALF loading, SEM images (Figure 4.10 and Figure 4.11) reveal significant fiber breakage, indicating that the fibers within the composites are failing under tensile testing. This suggests that the adhesion between the fibers and the matrix are improved at this fiber loading. Furthermore, the SEM images demonstrate good fiber dispersion, with fibers evenly scattered throughout the matrix. This finding is also reported by Ismail & Ishak (2018), that observed higher fiber loading improves fiber distribution and dispersion, resulting in greater strength compared to lower fiber loading. This is consistent with Figure 4.2, which shows the highest tensile test values among other composites.

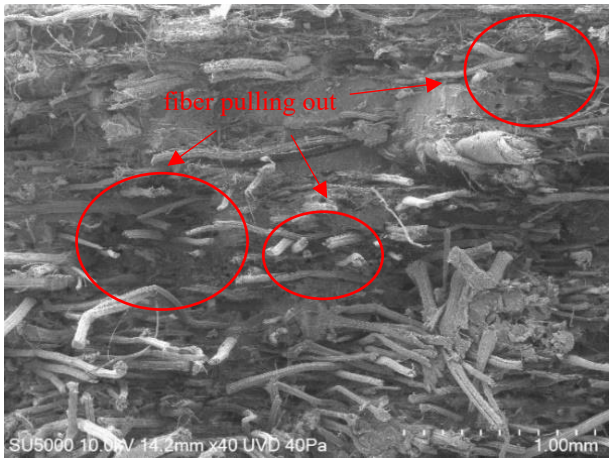

Figure 4. 10: SEM image of the fracture surfaces (30 wt% PALF fiber loading)

Figure 4. 11: SEM image of the fracture surfaces (30 wt% PALF fiber loading)

4.3 Effect of moisture on the mechanical properties of the PHBV/PALF composites

The effect of moisture on the mechanical properties of PHBV/PALF composites was investigated using water absorption tests within the period of 14 days. Percentage of water content and thickness swelling along 14 days of immersion were observed. Tensile tests were performed on aged samples or water absorption samples to evaluate changes in tensile strength, modulus, and strain at break due to moisture exposure. Additionally, the mechanical properties of aged samples were compared with unaged samples (not subjected to water absorption) with the same fiber loading, specifically at 0 and 20 wt.% PALF, to determine the impact of moisture exposure.

4.3.1 Water Absorption

The water absorption tests result (Figure 4.12) revealed that the PHBV/PALF composites exhibited varying degrees of water uptake depending on the fiber content. As the fiber loading increased, the water absorption also increased significantly over time. This is most probably due to the hydrophilic nature of the fibers and the lack

of proper fiber/matrix adhesion. The ability of natural fibers to absorb moisture is largely due to their hydrophilic properties. These fibers contain of hydroxyl groups (-OH), which are mainly present in hemicellulose and cellulose (Mokhothu & John, 2015). Additionally, the spaces between fibers and matrix form microchannels that allow water to easily penetrate the material (K. E. Mazur et al., 2022). Conversely, the neat PHBV or 0wt.% PALF fiber loading have smaller value of water absorption, due to the hydrophobic nature of the PHBV itself.

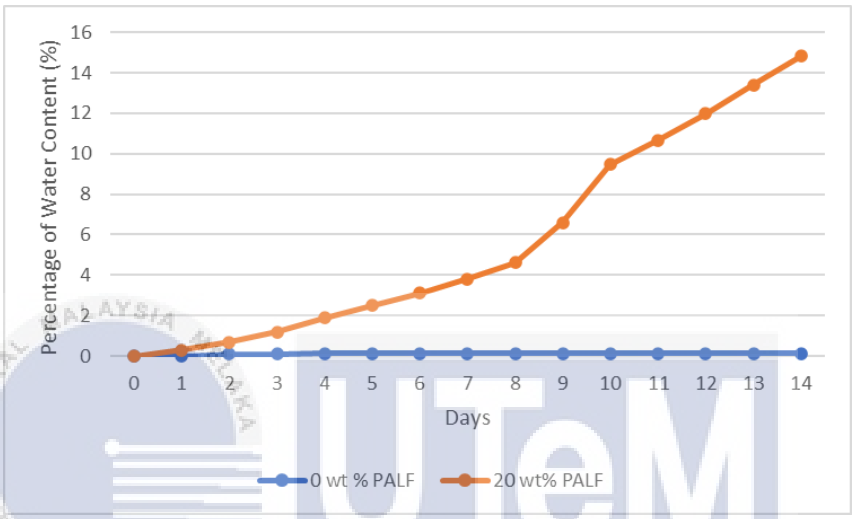

Figure 4. 12: Water absorption of the composites.

4.3.2 Thickness Swelling

Natural fibers absorb water by initially swelling as water molecules occupy the spaces between their microfibrils, forming a transient microcapillary network (Nurazzi et al., 2021). Thickness swelling data (Figure 4.13) showed a similar trend to water absorption, with higher fiber content composites showing more swelling. Neat PHBV samples displayed minimal thickness swelling, whereas composites with 20 wt.% PALF showed significant value. The increased thickness swelling with higher fiber content is primarily due to the swelling of the natural fibers when they absorb water. Furthermore, the presence of more fibers in the composite means a greater volume of fiber that can swell, leading to more pronounced thickness changes.

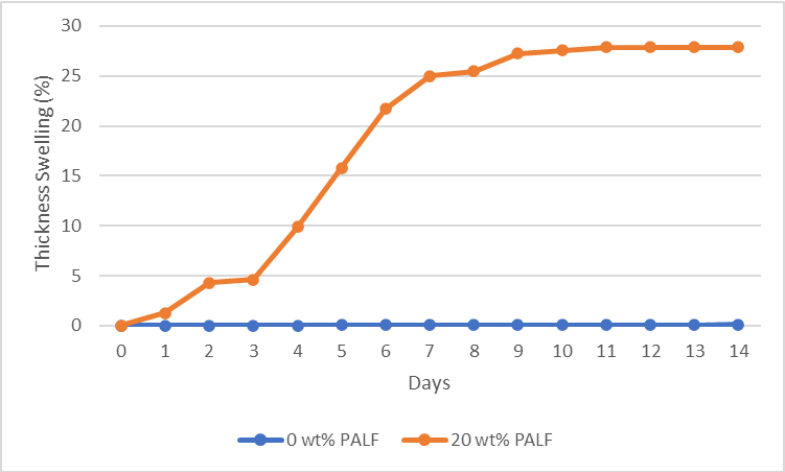

Figure 4. 13: Thickness swelling of the composites.

4.3.3 Tensile Properties

The mechanical properties of composites under moisture conditions were investigated to assess their performance in response to water aging. Figure 4.14 shows the tensile strength of neat PHBV increased slightly after water aging, from 29.57 MPa to 30.61 MPa, indicating a potential plasticizing effect in which water molecules improve the polymer's resistance to deformation under stress. This occurrence suggests improved mechanical performance because of greater chain mobility caused by water interactions.

Conversely, at 20wt.% fiber loading, the tensile strength of aged samples reduced from 29.81 MPa to 19.87 MPa. This reduction is attributed to water penetration within the composites, leading to swelling that can reduce the ability to tranfer stress at the fiber/matrix interface (Mazur et al., 2022).

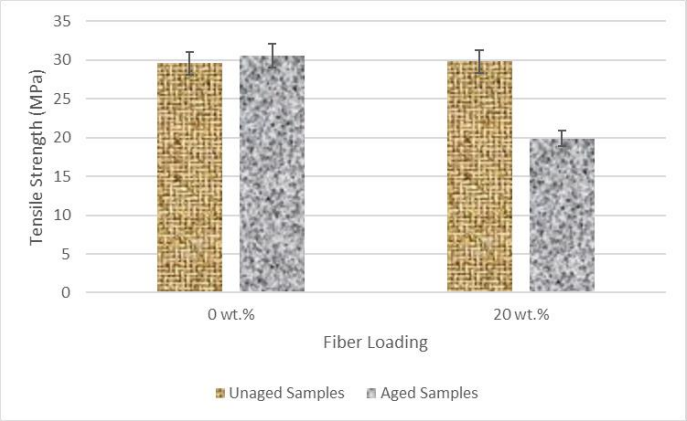

Figure 4. 14: Tensile strength of PHBV/PALF composites.

From Figure 4.15, the modulus of neat PHBV decrease significantly after water aging, from 0.0193 GPa to 0.0134 GPa, indicating lower stiffness and increased flexibility of the polymer. This is supported by SEM images (Figure 4.17), which show minimal signs of damage and indicate good resistance to moisture. However, for the 20 wt.% fiber loading, the modulus of elasticity increased from 0.0075 GPa to 0.0111 GPa after water ageing. This increase is likely due to plasticizing effect of water molecules penetrating the composites, which enhances the modulus. This supports by the SEM images (Figures 19 and Figure 20) that shows fiber pull-out and breakage, which are consistent with the improved modulus as fibers still contribute to the structural integrity of the composites even after moisture exposure.

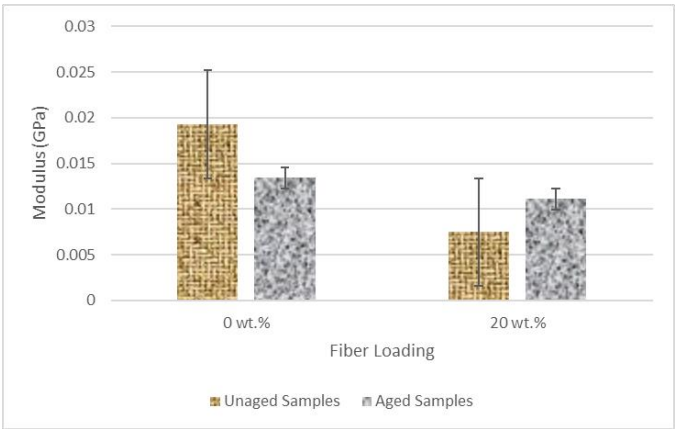

Figure 4. 15: Modulus of PHBV/PALF composites.

From Figure 4.16, for unaged samples, the strain at break for unfilled PHBV is 1.73%. Upon water aging, the strain increased to 2.53%, likely due to the plasticizing effect of water molecules. The SEM images of aged neat PHBV (Figure 4.17 and Figure 4.18) show a smooth fracture surface with minimal damage, indicating that water molecules may increase chain mobility and thus elongation at break.

For the unaged 20 wt.% PALF/PHBV composite, the strain at break is 3.38%. After water aging, the strain at break decreased to 2.03%, probably due to the presence of microcracks caused by the swelling of the fibers, which subsequently induces internal stresses and leads to the failure of the composite (Ismail & Ishak, 2018). The SEM images (Figure 4.19) display fiber pull-out and fiber breakage, suggesting that moisture weakens the interfacial bonds, causing microcracking and reducing the overall elongation at break. For unaged samples, the strain at break for unfilled PHBV is 1.73 %. Upon water ageing, the strain is increase to 2.53 %. This is due to the plasticizing effect due to the present of water molecule

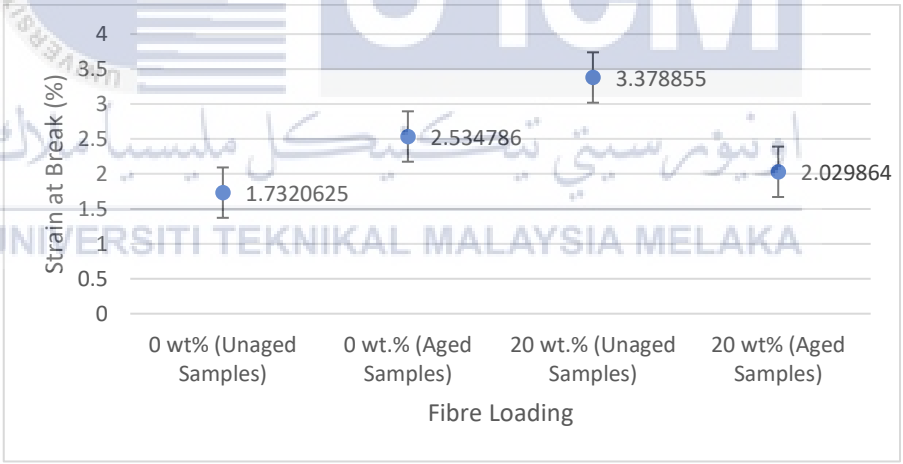

Figure 4. 16: Strain at break of PHBV/PALF composites.

4.3.4 Morphology Analysis

Morphology analysis was performed using SEM to study the fracture surfaces of the composites after tensile testing on water absorption samples. Fracture surfaces

provide information about the failure mechanisms and how moisture exposure impacts the structural integrity of the composite.

SEM images of neat PHBV (Figure 4.17 and Figure 4.18) show a relatively smooth fracture surface with less signs of damage. This shows that the absence of fibers reduces water absorption, resulting in reduced matrix failure. The value of tensile strength (Figure 4.14) supports the SEM images that shows brittle failure is occur, which is smooth surfaces. The hydrophobic characteristics of PHBV contributes to its resistance to moisture, which explains the minimal impact on its structural integrity.

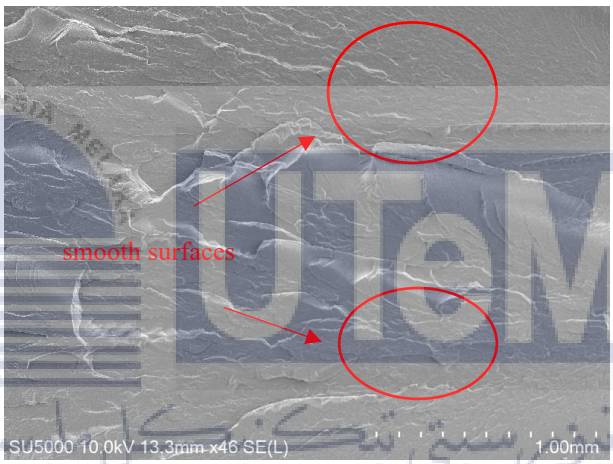

Figure 4. 17: SEM image of the fracture surfaces (Aged Neat PHBV).

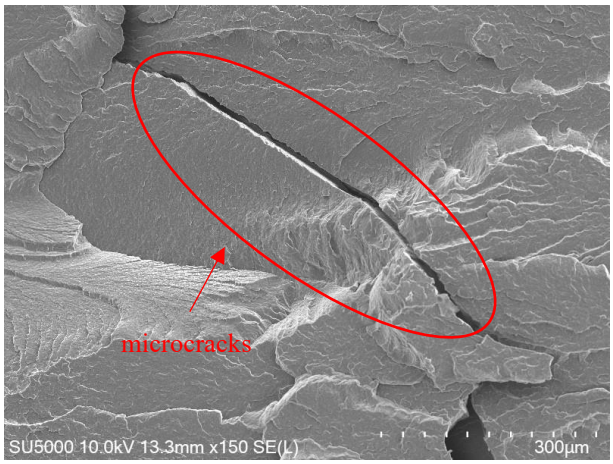

Figure 4. 18: SEM image of the fracture surfaces (Aged Neat PHBV).

At 20 wt% PALF loading, SEM images (Figure 4.19 and Figure 4.20) display fiber pull-out and fiber breakage. Fiber pull-out implies that moisture weakens the interfacial bond between fibers and matrix. However, fiber breakage indicates that some load is still being transferred to the fibers before failure.

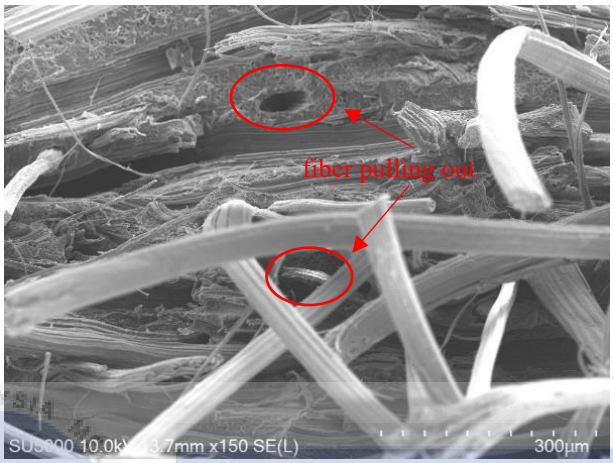

Figure 4. 19: SEM image of the fracture surfaces (Aged 20wt.% PALF).

Figure 4. 20: SEM image of the fracture surfaces (Aged 20wt.% PALF).

CHAPTER 5

CONCLUSION AND RECOMMENDATION

5.1 Conclusion

The study found that increasing PALF loading improves the tensile properties of PHBV/PALF composites. The tensile strength increases from 29.57 MPa for neat PHB to 46.74 MPa at 30 wt.% PALF, indicating improved stress transfer due to better fiber-matrix interaction. However, the modulus decreased from 0.0193 GPa for neat PHBV to 0.0075 GPa at 20 wt.% PALF fiber loading, likely due to porosity and voids. The strain at break increased from 1.73% to 6.19%, reflecting enhanced ductility and flexibility as fiber content increased.

Moisture exposure significantly affected the mechanical properties of the composites. After water aging, the tensile strength of the 20 wt.% PALF composites decreased from 29.81 MPa to 19.87 MPa. However, the modulus increased from 0.0075 GPa to 0.0111 GPa, suggesting plasticization effects due to water absorption. The strain at break decreased from 3.38% to 2.03%, attributed to microcracking caused by fiber swelling, which led to internal stresses and weakened the composite structure.

Morphology analysis revealed that neat PHBV, both unaged and aged, exhibited smooth fracture surfaces, indicating brittle failure. For 20 wt.% PALF loading, unaged samples showed strong fiber-matrix bonding with minimal pull-out, while aged samples displayed significant fiber pull-out and breakage, indicating weakened adhesion due to moisture exposure.

5.2 Recommendation for future research

Further research into the characterization of PHBV reinforced with PALF is recommended, with the following suggestions: -

i. To investigate the soil biodegradation performance of PHBV/PALF.
ii. To study the influence of long PALF on the mechanical performance of the composites.
iii. To enhance the mechanical performance of the composite by using nano-filler such as nano-cellulose, nano clay, and others.

5.3 Sustainable Design and Development

This research demonstrates sustainable design by developing eco-friendly composites using renewable pineapple leaf fiber (PALF) and biodegradable Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV). PALF, an agricultural byproduct, minimizes dependability on synthetic fibers and carbon footprints, while the biodegradability of PHBV addresses plastic pollution by breaking down into harmless byproducts. The combination of these elements produces high-performance, environmentally friendly composites, demonstrating a comprehensive to sustainability and encouraging the use of renewable and biodegradable materials in a variety of applications.

5.4 Complexity

The complexity of this research comes from numerous of issues, including the time-consuming and labor-intensive production of PALF films, as well as the fabrication process of composites, which uses sandwich technique that demands precision and consistency. Moreover, the use of chloroform in solvent casting has considerable health risks, demanding extensive safety standards. Furthermore, the manual operation of the hot press causes fatigue, which affects productivity and quality control.

5.5 Life Long Learning

This research practices lifelong learning by developing essential skills such as meticulous planning and adherence to schedules, engaging in critical discussions, and working independently with minimal supervision. It enhances the ability to evaluate information critically, particularly in analyzing data, promoting good decision making and problem solving. Additionally, the research promotes knowledge sustainability through suggestions for future improvements, ensuring continuous advancement in the material science field. The entrepreneurship component is also present, as the findings have the potential to be marketed and implemented in engineering projects, driving innovation, and providing market opportunities. These skills are essential for managing engineering projects, driving innovation, and making good engineering decisions.

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APPENDICES

Gantt Chart of PSM I

Appendix A

ACTIVITY	WEEK														
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
PSM Title selection								Mid Semester Break							
Collection for Literature Review															
Journal's Reaadings															
Introduction writing															
Literature Review writing															
Methodology writing															
Preparation for poster presentation															
Presentation week															
Logbook submission															
Report submission															

Gantt Chart of PSM II

Appendix B

ACTIVITY	WEEK														
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
Material Preparation								Mid Semester Break							
Composite Fabrication															
Sample Testing															
Data Collection															
Results and Discussion Writing															
Preparation for Oral Presentation															
Oral Presentation Week															
Logbook submission															
Report submission															
Technical Paper Submission															