UNIVERSITI TEKNOLOGI MARA

PROPERTIES OF POLY (BUTYLENE ADIPATE-CO-TEREPHTHALATE) BIOCOMPOSITE REINFORCED WITH KENAF, SEAWEED AND CHITOSAN AS BIO-FILLER

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MSc

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MUHAMAD HAIK AL BIN HAMDAN

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CONFIRMATION BY PANEL OF EXAMINERS

I certify that a Panel of Examiners has met on 12 January 2026 to conduct the final examination of Muhamad Haikal Bin Hamdan on his master of Science thesis entitled "Properties of Poly (Butylene Adipate-Co-Terephthalate) Reinforced with Kenaf, Seaweed and Chitosan as Bio-Filler" in accordance with Universiti Teknologi MARA Act 1976 (Akta 173). The Panel of Examiners recommends that the student be awarded the relevant degree. The Panel of Examiners was as follows:

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I declare that the work in this thesis was carried out in accordance with the regulations of Universiti Teknologi MARA. It is original and is the results of my own work, unless otherwise indicated or acknowledged as referenced work. This thesis has not been submitted to any other academic institution or non-academic institution for any degree or qualification.

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ABSTRACT

The increase in accumulated plastic waste into the environment is a significant contributor to plastic pollution. Biodegradable polymers, such as polybutylene adipate-co-terephthalate (PBAT), have the potential to replace conventional plastics in order to effectively reducing microplastic contamination. This study reports on the influence of kenaf fibre (KF), kenaf core (KC), seaweed (SW), and chitosan (C) on various aspects of PBAT bio-composites, including their chemical structure, crystallisation behaviour, thermal properties, microscopic morphology, and tensile properties. Bio-composites were synthesized through a melt mixing process involving PBAT combined with 10, 20, and 30 wt.% of KF, KC, SW, and C. The tensile characteristics reveal that the maximum tensile modulus is KF30 at 652 MPa, while the minimum tensile modulus is SW10 at 89.32 MPa. Dynamic Mechanical Analysis (DMA), Thermogravimetric Analysis (TGA), Differential Scanning Calorimetry (DSC), and Fourier Transform Infrared (FTIR) Spectroscopy were utilised for characterisation. The integration of bio-fillers led to an improvement in the stiffness of PBAT, accompanied by significant increases in storage modulus (E') across all bio-composite with the storage modulus of KF30 recorded is 620 MPa at 20 °C. It shows that it can be ascribed to the pronounced interactions between the carbonyl groups of the PBAT matrix and the hydroxyl groups of the bio-fillers. Furthermore, the use of bio-filler, characterised by its small surface area and diverse composition, affected the crystallization of PBAT. There are new peaks found in FTIR analysis ranging within 3900-3600 cm^{-1} which is ascribes as alcohol O-H stretching. In conclusion, this study presents an innovative method for the preparation of PBAT bio-composites, integrating environmentally friendly bio-fillers. These bio-composites present opportunities for the application of biodegradable materials, providing enhancements in functionality.

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LIST OF SYMBOLS

Symbols

cfu/g	Colony forming unit per gram
cnT[1]	Wavelength
E*	Complex Modulus
g/cm[3]	Gram per centimeter cubic
Hz	Hertz
J/g	Joules per gram
kN	Kilo Newton
kg/m3	Kilogram per centimeter cubic
m	Mass of the material
mg	Miligram
ml/min	Mililiter per minute
MPa	Mega Pascal
mm	Milimeter
mm/min	Milimeter per minute
pH	Potential of hydrogen
rpm	Revolutions per minutes
wt%	Weight percentage
℃	Degree Celsius
°C/min	Degree Celsius per minute
£	Strain
a	Stress
0	Heat added during melting
T	Temperature
t	Time
>	Bigger than
%	Percentage

LIST OF ABBREVIATIONS

Abbreviations

BA	Butanediol Adipic
BT	Butanediol Terephthalate
DTG	Derivative Thermogravimetric
H NMR	Proton Nuclear Magnetic Resonance
UV	Ultra Violet

CHAPTER 1
INTRODUCTION

1.1 Research Background

The increment of accumulated plastic waste to the environment is one of the major factors of plastic pollution. These increases of plastic pollution in a particular area of the environment are deemed "poorly reversible" as natural degrading processes occur slowly and technological-advanced remediation alternatives are improbable. Plastic contamination typically occurs in tropical landfills, deserts, and deep oceans. (MacLeod, 2021). Plastic manufacture is an integral aspect of the global economy and has grown tremendously, with an estimated 9.2 billion tons produced between 1950 and 2017. Every year, around 11 million tons of garbage emphasis of plastic-based leak into the seas. Polymer or plastic consumption varies, but 47 % of the total account come from packaging industry. Recycling and the circular economy are viewed as crucial to addressing the plastic problem, such as the Extended Producer Responsibility plan and the Deposit Return plan (Williams, 2022). From that circumstance, the plastic pollution effect really impacts the world environmentally and economically.

Therefore, the awareness of plastic pollution grown in our society in which leads to the expanding of demand for polymeric materials in which environmentally friendly among the consumers. Moreover, majority of manufacturers especially polymers industries were experimenting the development of low cost and environmentally friendly polymeric materials to cope the rising demand of the polymeric products. The need for environmentally friendly polymeric materials, which seeks to contribute to the green economy and address significant concerns regarding conventional thermoplastic polymers is increasing. The potential economic and ecological sustainability of bio-compostable composites is driving their emergence as novel environmentally benign materials in the fields of engineering and commerce (Awad, 2023a). These materials are typically made by blending bio-degradable polymers with organic particles. An extensive array of bio-compostable polymers, including those derived from starch and polyhydroxyalkanoates and polylactic acid, have been suggested as viable substances

for the fabrication of bio-compostable composites (AL-Oqla, 2022a). Within this framework, poly (butylene adipate-co-terephthalate) (PBAT) as an entirely biodegradable aliphatic and aromatic copolyester suitable for variety of uses, including food, medical, agriculture and pharmaceutical industries.

PBAT has high mechanical properties because of its aromatic structure in the molecule chain and high biodegradability because of its aliphatic structure in the molecular chain. Compared to low-density polyethylene (LDPE), PBAT's mechanical properties are more flexible than those biodegradable polyesters, including poly (lactic acid) (PLA) and poly (butylene-succinate) (PBS). Because of these mechanical qualities, PBAT is a very promising biodegradable material with many possible uses. However, the high melt viscosity (2.7 - 4.9 g/lOmin), low crystallization rate (53 °C) and low tensile strength around (35-44 N/mm^2) limit its process ability and industrial applications. PBAT is frequently combined with several bio polymers to create high-performance composite materials (Ferreira FV, 2019a).

To enhance the ultimate characteristics of this polymer, various natural particulates, including lignin (Xiong et al., 2020a), residual microalgae biomass (Gallo-Garcia et al., 2022), torrefied coffee grounds (Kim et al., 2024), and reed (Xie, 2023a) has been evaluated as fillers for PBAT. These materials utilized for fabrication of high-performance materials that applicable to a variety of applications while also enabling the valorisation of detritus from the marine, industrial, or agricultural sectors. Some recent years of studies have explored reinforcing biodegradable polymers with natural fibres (AL-Oqla, 2022b; Ilyas, 2022a), cellulose nanofibrils (EM, 2023) and biomass (Ferreira FV, 2019b; Karimah, 2021a) as an alternative method in order to increase mechanical and thermal properties while maintaining its biodegradability (Hong SH, 2022a).

Seaweed possesses significant potential as a sustainable source of biopolymers. Seaweed is a highly adaptable organism that generates a range of polysaccharides, such as agar, carrageenan, and alginate, which are widely utilized in the creation of biopolymers (Sarmin, 2023). Seaweed polysaccharide-based biopolymers have favourable characteristics due to their renewability, biodegradability, biocompatibility, and eco- friendliness (El-Sheekh et al., 2022; Sangeetha et al., 2023). Other than seaweed, chitosan also being utilized in this project. Chitosan is a chitin derivative generated by deacetylating a specific type of polymers obtained from chitin

(Sunilkumar, 2012). Chitosan-based composites have many appropriateness and potential especially for biomedical, packaging, and water purification fields because polysaccharides in which are renewable resources (Oladzadabbasabadi, 2022a). Chitosan can be advantageous due to its highly compatible with other biopolymers, so blending it with cellulose or incorporating cellulose nanofibre. Due to the presence of functional groups such as hydroxyl, acetamido, and amine, these distinctive polymers have emerged as a new type of physiological materials with a favourable solubility profile, less crystallinity, and chemical reactivity. Kenaf, a natural plant, is increasingly being recognized as a viable material for the manufacturing of bio-composites. Kenaf is chosen as a supplementary alternative material for manufacturing bio-composites due to its rapid growth characteristics, enabling it to provide a substantial amount of raw material within a short timeframe (Asim et al., 2019; Hassan et al., 2020).

The primary objective of this research is to enhance the sustainability and manufacturing of bio-composites from PBAT by employing seaweed, chitosan, and kenaf as fillers, which remain minimally investigated. The study aims to develop an approach for characterising the mechanical, viscoelastic, thermal, and molecular interactions of the bio-composites.

1.2 Problem Statement

Plastic pollution really impacted the environment where it leads to innumerable negative consequences such as natural disaster from the piles of plastic that have been thrown into the ocean, the soil, and air. Moreover, the disposal of the polymeric materials such as plastic really high cost consuming where the dispose plastic could not be reused again and the cost of the plastic manufacture could not be reversible. From that particular reason, the government, non-government agencies and the polymeric materials consumer emphasis these problems and raise an awareness towards a sustainability of environment to diminish the rate of plastic pollution that caused by polymeric materials consumptions.

Thus, there are several alternative growing interests in environmentally friendly materials generated from natural sources in order to satisfy the rising demand for products that combine sustainable qualities with improved mechanical properties. In

this context, a number of lignocellulosic fibres have been evaluated as reinforcing agents for biodegradable matrices with a dual objective of cost reduction and enhanced bio plasticity. On the other hand, the addition of organic fillers generally results in notable increase in stiffness, at the expense of the breaking properties of these polymers, leading to stiffer materials, resulting in a limitation of the application area. This compromises the toughness of the materials, which subsequently restricts their applicability. Therefore, in order to simplify the production process and reduce the number of phases affected, a large research effort has been devoted to discovering lignocellulosic additives which can increase bioplastics with no need for coupling agents, chemical treatment or plasticizer. Currently, there has been limited assessment of the practicality of integrating seaweed, chitosan, and kenaf into biodegradable polymers.

This study examined the viability of using seaweed, chitosan and kenaf as a reinforcing component in PBAT blends. The ultimate objective was to produce high-performance natural filler base that were reinforced with PBAT to improve the functional properties of the bio- composite so that they could be used in industrial application. Furthermore, this work aims to examine the interfacial compatibility of seaweed, chitosan, and kenaf filler with the PBAT matrix. This compatibility is crucial in determining the overall mechanical performance of the bio-composite. The utilization of natural resources material like seaweed, chitosan and kenaf as a natural filler have a high possibility to create a high-performance biopolymer. Due to the circumstances, the utilization of these natural resources has a potential to improve the mechanical and the physical properties of the PBAT matrix. This bio-composite is thought to be a highly promising renewable resource for the development of biocompatible and environmentally friendly materials.

1.3 Research Objectives

The investigation's principal objectives are as follows:

1 To evaluate the compatibility of PBAT with seaweed, chitosan, and kenaf as a bio-fillers material through viscoelasticity and mechanical properties of the bio-composites.

2 To determine the thermal properties and the molecular interaction of each bio-composite produced from PBAT/seaweed, PBAT/chitosan and PBAT/kenaf blends.

1.4 Significance of Study

The purpose of this research was to evaluate the compatibility of PBAT in combination with natural resources such are seaweed, chitosan, and kenaf to produce an enhanced bio-composite material. Seaweed, chitosan and kenaf are extremely promising sources of renewable biopolymers and also a promising material that can be utilized as natural filler to nourish the compatibility of PBAT in combination with seaweed, chitosan, and kenaf to produce a bio-composite material. The utilization of these additives' natural filler in the PBAT matrix has the potential to enhance the physical and mechanical properties of the PBAT polymer matrix.

This research also provides a collection of information regarding the molecular interaction and properties of bio-composite produced from PBAT/seaweed, PBAT/chitosan and PBAT/kenaf blends through viscoelasticity, mechanical and thermal evaluation. In addition, every matrix or mixture-based PBAT will undergo several characterization analyses such are mechanical strength analysis, dynamic mechanical analysis, morphological analysis through scanning electron micrograph (SEM), and thermogravimetric analysis in order to obtain the first-hand results of each analysis regarding the characterization of each matrix.

CHAPTER 2
LITERATURE REVIEW

2.1 Biopolymer

There is rising awareness on the adverse effects of environmental pollution arising from fossil fuels and waste from petrochemical industries. Extensive research has been conducted to investigate alternative options to petroleum-based goods that are both renewable and biodegradable, hence reducing environmental risks. Biopolymers are a leading class of functional materials ideal for high-value applications, and they are of significant interest to academics and experts from many fields. It offers a potential solution to the problem due to their characteristic of being biodegradable materials derived from renewable resources. The categorization of biopolymers is illustrated in Figure 2.1.

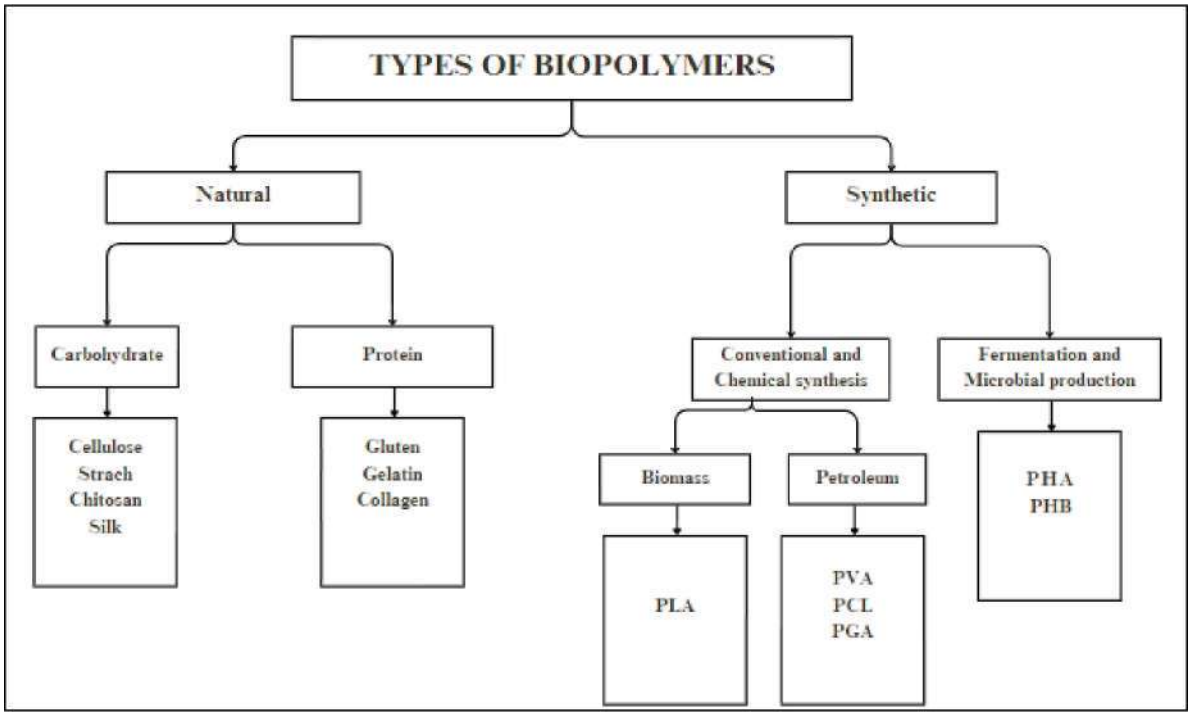

Figure 2. 1 Streamlined Categorization of Biopolymers

Biopolymers are polymers that consists of organic substances. They are complex biological molecules made up of repeating units (monomers). This encompasses a wide range of plant species, including corn and soybeans, as well as several tree species and even certain microbes (Phiri et al., 2023). Biopolymers are derived from sustainable sources and exhibit high biodegradability due to the presence of oxygen and nitrogen atoms in their structural framework. Biodegradation transforms them into carbon dioxide, water, biomass, organic matter, and other natural compounds. Their biocompatibility and biodegradability render them valuable in various sectors, including the pharmaceutical industry as dressing materials and medical implants such as organs, wound healing, and medical implants, as well as packaging materials and edible films and emulsions (Sivakanthan, 2020).

A notable differentiation between polymers and biopolymers is in their structural composition. Biopolymers, like all polymers, consist of monomers, which are strings or sequences of monomeric units. While these strings are frequently linear, they can also be circular, branched, closed, or cross-linked (George, 2020a). The fundamental structure of biopolymers pertains to the exact chemical composition and organization of the units comprising its structure. Biopolymers, such as nucleic acids, proteins, carbohydrates, lipids, and large non-polymeric molecules like lipids and macrocycles, are the most common types of macromolecules. Plastics, synthetic fibres, and experimental substances like carbon nanotubes are instances of synthetic macromolecules. Aside from the repetitive units of nucleic acids, saccharides, or amino acids, the molecular structures of these compounds can also include various chemical side chains that play a role in their functions. Poly (lactic acid) (PLA) and polyhydroxyalkanoates (PHAs) are biopolymers that can be generated from microorganisms or genetically engineered organisms using conventional chemical techniques (Baranwal, 2022).

2.1.1 Biopolymer Composites

Biopolymer composites are generated by blending biopolymers with suitable infill reinforcements to augment their functions. The fillers used depend on their

intended function and can include natural fibres, metals, and nanofillers based on metal oxides. The properties of biopolymer composites can be obtained and altered by combining an appropriate biopolymer with appropriate fillers or additives, which facilitates the interaction between the polymer and the filler. The chemical composition, degradation kinetics, and mechanical properties of biopolymer composites can be altered to meet the specific requirements of the application. Therefore, research into the replacement of petroleum derived commodity plastics with biodegradable polymers derived from biological and renewable sources has been given much attention (Alshammari, 2019). Due to their remarkable properties, such are better barrier performance, biodegradability and lighter nature, biopolymers derived from naturally occurring sources either via chemical processes or the biosynthesis of live organisms provide efficient solutions for these challenges (Ilyas, 2022b; Karimah, 2021b; Kumar, 2022a). However, they are typically characterized by poor mechanical properties, fatigue limited life expectancy, low chemical resistance, insufficient long-term durability, and weak processing capabilities (Chauhan, 2022; Dehury, 2022a). To address these limitations and create sophisticated materials suitable for many uses, biopolymers can be strengthened by incorporating fillers or nanofillers, resulting in the formation of bio-composites or bio-nanocomposites. The structure, environmental tolerance, and longevity of a biopolymer composite are primarily determined by the biopolymer matrix, whereas the stiffness and strength of the composite are determined by the reinforcement fibre. The utilization of innovative biopolymer composites in unique applications guarantees the possibility of expansion in global markets. The ongoing attempts to create environmentally friendly composite products with enhanced performance have resulted in significant global applications and utilization.

To procure biopolymer composites, there are numerous processing methods to acquire the biopolymer composites. Biopolymer composite preparations are commonly processed via compression molding, injection molding, and extrusion. Furthermore, biopolymer composites are typically obtained through solvent casting and melting methods. The solvent casting method is a process that related to the of a polymer and filler solubilisation, the distribution of the solution on a substrate, and solvent removal, which improves the polymer chain molecular orientation and intercalation of filler molecules. Therefore, solvent casting method is a low-cost processing, and it could

improve the physicochemical properties of the biopolymer composites (Borbolla-Jiménez et al., 2023). Next, the most used process is melting processing method where the biopolymer will be inserted into the screw like extruder along with the filler in a particular temperature and speed depending to types of material used (Velghe et al., 2023).

Currently, polyhydroxyalkanoates (PHAs), starch-based plastics, polybutylene adipate-co-terephthalate (PBAT), and poly (lactic acid) (PLA) are the most used biopolymers in the development of bio-composites (Awad, 2023b; Dehury, 2022b). Poly (lactic acid) (PLA) is an entirely biodegradable polymer that undergoes decomposition both within the human body and in composting facilities. Its biocidal properties are because of its propensity to endure hydrolysis at the surface and produce lactic acid. Recent studies on poly (lactic acid) (PLA) reinforced with fish scale which resulted in enhancing the flexural strength of the matrix as per the 20 % fish scale content were introduced with the poly (lactic acid) (PLA) (Arockiam et al., 2023). As a result, it is widely recognized as an exceptionally efficient alternative to polymers derived from petroleum in numerous sectors, including packaging, agriculture, personal care, cosmetics, biomedicine, and tissue engineering (Karimah, 2021c; Oladzadabbasabadi, 2022b). Biopolymers with PHAs are extremely intriguing. Produced by microorganisms in the absence of vital nutrients and in the presence of surplus carbon, they constitute a group of polyesters composed of hydroxyalkanoic acids. In different ecosystems, PHAs have thermoplastic properties like polypropylene and good mechanical characteristics as well as excellent biodegradability (Nayak, 2023a; Xie, 2023b). Phosphopropyrate-hydroxybutyrate homopolymer (PHB) and poly(3-hydroxybutyrate-co-3-hydroxyvalerate) copolyester (PHBV) with hydroxyvaleric acid are the most prevalent PHAs and are specially designed for use in food packaging (Ferreira FV, 2019c). They are expensive in comparison to other biopolymers, which has restricted their application in the pharmaceutical and medicinal industries despite their advantageous properties and biodegradability. For the production of diverse products with a wide range of structural and functional changes, renewable polymeric materials derived from vegetables or plant oils may be a reliable base material (Arumugam, 2020a; Xie, 2023c). Due to their widespread availability and very affordable price, they are highly appealing for use in the plastics sector. Various

vegetable oils, including soybean oil, castor oil, and linseed oil, have undergone polymerization in the presence of different fillers and fibres, such as clays, inorganic nanoparticles, hemp, flax, jute, or kenaf fibres (Kumar, 2022a; Oladzadabbasabadi, 2022c). This process has resulted in the development of bio-composites that exhibit notable enhancements in their mechanical properties and thermal stabilities.

2.1.2 Characterization of Biopolymer Composites

The properties of each biopolymer composite are reliant upon the variety of biopolymer composite (Gurupranes et al., 2023). It is in direct competition with commodity plastics because they offer a wide range of advantages when used in environments that are crucial for their performance. These advantages include low cost, sustainability, biocompatibility, lightweight, carbon dioxide sequestration, biodegradability, superior electrical and mechanical properties, recyclability, soil fertility, and efficient waste management. Biopolymer composite can be degraded by environmental factors, including air, light, heat, or bacteria (Díez-Pascual, 2022a). The structure, environmental resilience, and longevity of the biopolymer composite are primarily regulated by the biopolymer matrix, while the rigidity and strength of the composite are determined by the reinforcing. Potential advancements in global markets are guaranteed by the introduction of innovative applications of biopolymer composites (Elfaleh et al., 2023).

The integration of reinforcement materials originating from biological sources into biopolymers has greatly enhanced the mechanical characteristics of biopolymer composites, including tensile and impact strengths (Basheer et al., 2021). The resulting biopolymers demonstrate improved characteristics in terms of stiffness, resistance to heat-induced deformation, capacity to allow gases to pass through, decomposition, and, notably, the tensile strength of the component when combined with nanoparticles and nanofibres produced from biological sources (Das A., 2023). Moreover, the integration of reinforcement material with biopolymer, the electrical properties of the biopolymer composite (Dalwadi et al., 2023), and its resistance to insects or pests (Friuli et al., 2023) also improved.

Several previous studies recorded that the reinforcement of biopolymer with bio-filler increases the properties of the biopolymer composites. A study shows that a PBAT reinforced with coffee grounds (CG), ranging from 10% to 50% which resulting the elongation at break of the sample of PBAT/CG50 biopolymer composite has the highest reading compared to the other biopolymer samples (Boey et al., 2022).

The assimilation of bio-derived reinforcing materials into biopolymers improves the mechanical properties of composites, including impact and tensile strength. Incorporating bio-derived nanoparticles and nanofibres improves biopolymer attributes such as moduli, heat deformation temperature, gas permeability, decomposition, and tensile strength. Moreover, biopolymer composites are environmentally benign and cost-effective, inspiring researchers to build innovative products suitable for various applications. Research has focused on bio-composites made from cellulose, starch, poly-lactic acid (PLA), poly-vinyl alcohol (PVOH), poly-butylene succinate (PBS), poly-hydroxy butyrate (PBH), and other materials (George, 2020c).

The polymer component in bio-composites is responsible for the material's outstanding enhancement mechanical properties, including molecular weight, chemical composition, shape, and processing technique, which effectively impart desired functions. Therefore, it has been proven that bio-derived reinforcing materials into biopolymers improved the mechanical properties of the biopolymer composites. In addition, researcher is focusing on developing lightweight structural materials from natural resources with superior mechanical properties to address environmental concerns such as lignocellulosic-based composite materials are widely used in construction, building, and automotive components. For these applications, the developed product must be durable enough to sustain external forces within the material's maximum limit under cyclic loading.

PBAT is one of the excellent examples of biopolymer composite that exhibits lower strength, but significantly higher ductility (Dmitruk, 2023). The decrease in composite flexibility is explained by the lower mobility caused by the presence of rigid bio-composites (Oyeoka et al., 2021). Therefore, there are numerous biopolymer composites that have a lower mobility to ductile. Polylactide (PLA) is one of the biopolymer composites that is widely utilized in numerous applications due to its

brittleness (Xu, 2020). Circumstances, ductile biopolymer composites able to absorb or withstand a greater load (Hazwani & Todo, 2021).

Tribology is concerned with friction, wear, and lubrication between contacting surfaces. Interfacial molecular films are absorbed, self-assembled, or functionally grafted molecule systems formed near or on a surface via physical and chemical processes (Jabbarzadeh, 2018). Modifying the wear and frictional behaviour of composites is crucial as tribological conditions account for a significant portion of mechanical part failure (Sadasivuni, 2020c). Several researchers have tried new combinations of both reinforcement as well as the matrix and studied their tribological behaviour. However, tribology is one of the key limitations that must be addressed in the case of biopolymer composite materials; but this subject has received little attention (Thankachan et al., 2022).

2.1.3 Application of Biopolymer Composites

Recent growth in biopolymers industries have sparked interest in a variety of industries to improve the biopolymer composites properties and ease commercialization. Biopolymers such as collagen, keratin, chitosan, silk, and elastin were combined with synthetic polymers to create composites with improved properties. Biodegradable natural polymers are in high demand as packaging film materials for pharmaceuticals and food packaging applications due to their low toxicity content in the biopolymers (Udayakumar, 2021). Furthermore, the utilization of bast also gives a significant observation when it utilized as biopolymer composite. According to the researchers, bast fibres exhibit outstanding mechanical capabilities, making them a viable alternative to glass fibres as reinforcing components in polymer composites and suitable for a variety of applications. Environmentally friendly, bast fibre composites are used in autos, military applications, construction, and packaging industries (Prakash, 2022). It shows that biopolymer is used to improve certain properties of the composites in many applications including packaging, agriculture, personal care, cosmetics, biomedicine, and tissue engineering as shown in Table 1.

Table 1

Applications of Biopolymer Composites

Biopolymer composites	Applications	References
Chitosan **PVA/ chitosan (CS)**	Biomaterials applications	
Polyurethane **PU/hydroxyapatite (HA)**	Coatings, adhesives, rigid and flexible foams, films, fibres, thermal and electrical insulation, biomedical applications, construction, high-performance adhesives, footwear, and furniture packaging	(Díez-Pascual, 2022b) (Mumtaz et al., 2023)
Polyvinyl alcohol (PVA) **PVA/tannin acid (TA)** **PVA/ cellulose nanocrystal (CNCs)**	Medical applications: contact lenses, eye drop, embolization particles, tissues adhesion barrier, drug delivery, artificial cartilage and meniscus, solid polymer electrolyte, food packaging	(Gautam et al., 2022) (Tan et al., 2021)
Polyvinyl chloride (PVC)	Scientific applications: Gas separation, water and wastewater treatment, ion exchange gas	(Lieberzeit et al., 2022)
Poly (butylene adipate-co-terephthalate) (PBAT) **PBAT/ polylactic acid (PLA)**	Packaging applications: wrap and plastic bag	(Pietrosanto et al., 2020)

2.2 Poly (Butylene Adipate-Co-Terephthalate) (PBAT)

PBAT is commonly used in various applications due to its advantageous characteristics, such as lightweight and durability. PBAT is a polyester composed of terephthalic acid (TPA), 1,4-butanediol, and adipic acid. It is widely used in the manufacturing of plastics. PBAT is widely used for food packaging, agricultural, textile,

and various other industries. PBAT mulch film is utilized in agriculture to optimize soil conditions and augment crop yield (Burford et al., 2023). Unlike polymers formed by carbon-carbon linkages, PBAT was initially recognized as a compostable biopolymer due to the higher susceptibility of polyesters to enzymatic breakdown, particularly by esterase (Hong SH, 2022a). The utilization of PBAT is increasing in many industries due to its properties and capability to reduce white pollution. Therefore, several developed countries such are China, Europe, and North America became the major producer of PBAT. It reflects that PBAT global demand is growing (Xu, 2023). Moreover, numerous research has been conducted on PBAT with many types of materials such are zinc oxide (ZnO), lignin bio-composites and cellulose nano crystal (CNC), to assess its properties and its biocompatibility (de Souza et al., 2022; Vatansever et al., 2020; Xiong et al., 2020b). It shows that the demand of PBAT will keep increasing for industrial application and research purposes.

2.2.1 Synthesis of PBAT

The synthetization of PBAT can be divided into several stages including pre-mixing, pre-polymerization and final-polymerization process (Jian et al., 2020a). It is a substance formed by poly-condensing butanediol (BDO), adipic acid (AA), and terephthalic acid (PTA). It has been demonstrated to be an ideal composition in terms of outstanding characteristics and biodegradability. The preparation of PBAT necessitates a long reaction time, a high vacuum, and temperatures that are typically higher than 190°C (Hong SH, 2022b). These conditions must be met to promote condensation reactions and remove the lighter molecules (water) as a byproduct. It is a versatile aliphatic- aromatic co-polyester, it also has excellent chemical and hydrophobic properties. Moreover, the material is biodegradable and able to completely decompose in a matter of weeks with the help of natural enzyme sources found in soil (Xie, 2023d).

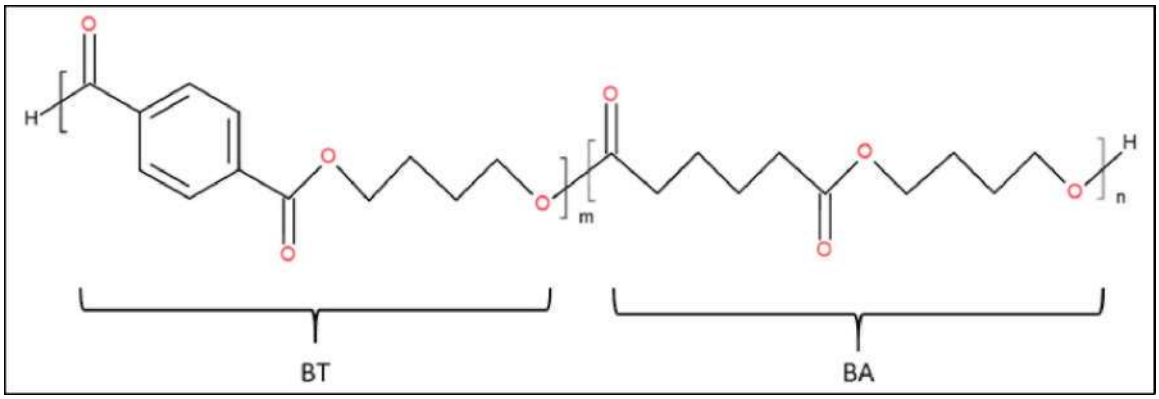

Figure 2. 2 Chemical Structure of PBAT (Su, 2021a)

Figure 2.2 shows the chemical structure of PBAT. It shows that the chemical structure of the PBAT is formed with butanediol terephthalate (BT) and butanediol adipic (BA) types of dimers. The first dimer was structured by 1,4 -butanediol terephthalic acid monomers in which a rigid section. Another dimer, BA unit formed by 1,4-butanediol and adipic acid monomers which in which forming a flexible section (Su, 2021b). Nevertheless, PBAT exhibits good mechanical and thermo-mechanical properties. Consequently, these limitations hinder its application in several industrial sectors. To mitigate these challenges, it is necessary to incorporate a strengthening agent that will enhance the characteristics of the polymer to fulfil specific application requirements, while also controlling the overall cost of the product. However, the quality of reinforcement is often determined by the precise arrangement of the microstructure and the stability of the link between the polymer matrix and the filler. The schematic diagram of synthesis of PBAT shown as below:

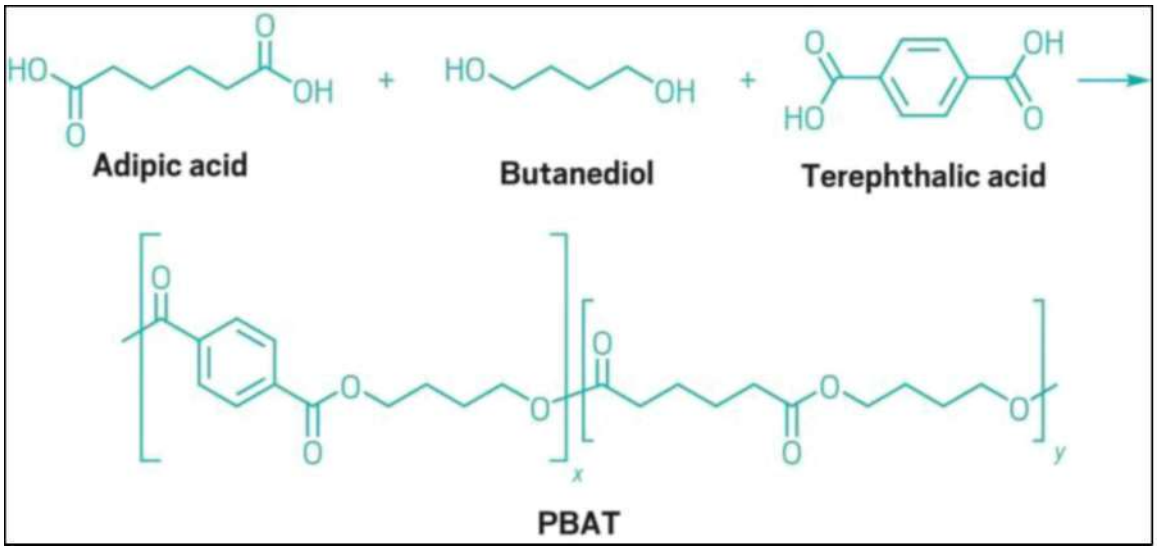

Figure 2. 3 The Schematic Diagram of Synthesis of PBAT (Tullo, 2021).

2.2.2 PBAT Based Bio-composites

The mechanical properties of PBAT have led to its frequent use in combination with other bio-composites materials to create high-performance or high-stability composites. Despite its high value and environmental friendliness, the widespread usage of biodegradable PBAT is constrained by its thermal stability compared to other polymers. As a result, there have been notable attempts to tackle these shortcomings by using additional biodegradable polymers like poly (lactic acid) (Balla et al., 2021), or inexpensive natural organic additions such as cellulose (Etale et al., 2023), starch (Hamaker, 2021), and plant protein (Gomes & Sobral, 2021). In addition, certain researchers have made efforts to merge PBAT with certain agricultural byproducts, such as peanut shell (Paczkowski et al., 2021) and coffee grounds (Leow et al., 2021). The integration of PBAT with an organic agricultural waste in the production of composites is expected to reduce the strain on landfills and mitigate associated environmental concerns. It is crucial to convert agricultural waste into more valuable goods by using them as functional additives in bioplastics.

Over the past decade, composite materials based on PBAT have been transformed into commercially available products. PBAT is one of the polymers that is frequently used and being studied to improve and enhanced its properties. PBAT-based

products have successfully achieved worldwide composability standards and have obtained compost certificates (Biernat, 2023). These products can be processed using typical plastic equipment, making them ideal for creating the same kind of products as traditional plastics. PBAT-based products have obtained extensive applications several industries, including packaging (Alpaslan Güler & Deniz, 2023), mulch film (Qiao et al., 2022), and cutlery (Mousa et al., 2022), due to their high quality, satisfactory performance, and cheap cost. Applications for PBAT as shown in Table 2.

Table 2

Application of PBAT

Application	**References**
Agricultural applications: decomposable bag mulch film	(Jian et al., 2020b)
Packaging application: food packaging utensils	(Zhao et al., 2024a)
3D printing application: 3D printing material	(Zhang et al., 2021)
Pharmaceutical applications: disposable medicine equipment	(Zhao et al., 2024b)

There are several commercially available compostable materials based on PBAT that can be used to make a packaging for certain purposes. BASF, Novamont, BIOTECH, and KINGFA are prominent businesses engaged in the development of materials based on PBAT. KINGFA, a prominent player in the field of biodegradable polymers, has developed a diverse range of compostable substances made from PBAT, starch, and PLA. These materials have been used in various packaging applications, such as compost bags and supermarket bags. KINGFA has overseen the extensive adoption of shopping bags composed of Starch-PBAT blends in upscale supermarkets across China, setting an example for the introduction of biodegradable plastics in the nation. Biodegradable mulch films, when inserted into the soil, are susceptible to degradation and decomposition as time passes. Consequently, they must exhibit specific

predetermined characteristics. KINGFA has manufactured mulch films using PBAT to meet agricultural requirements. When PBAT-based mulch films are used in soil, they show little vulnerability to water, elevated temperatures, and UV radiation. Moreover, these films have the capacity to fully decompose through biological processes once they have served their purpose. KINGFA has successfully employed agricultural films made from PBAT/PLA/Nano-particles composites in different regions and crops in China. These films have established a strong foundation for future progress in the country.

2.3 Natural Fibres

Back in the old days, natural fibres utilized as daily life equipment such are rope, fabric, and twine. The natural fibres do not gave bad impact towards the environment because of its sustainable characteristics, biocompatibility and biodegradability as well as its origin from renewable resources (Azammi et al., 2020). Due to characteristics, natural fibres is one of the alternatives to reduce or diminish the utilization of the synthetic fibres in composite industry (Getme & Patel, 2020). Due to its distinct characteristics, the properties of natural fibres are completely contradicted with the synthetic fibres. Natural fibres, which derived from renewable sources offers great properties such are easy production, cost effective, low density and lightweight (Hosseini, 2020).

Furthermore, numerous natural fibres are being utilized to reinforce with polymer matrix, which results in the polymer functioning as a binding material to hold the fibres. Due to the particular circumstances, it gave the fibres a dimensional stability (Aaliya, 2021a). By time with the rapid development of research regarding the utilization of natural fibres, natural fibre composites have enormously been utilized in numerous application in industrial applications, in automotive purposes, furniture and packaging where it is to increase the environmentally sustainability through the production of sustainable materials where it could be as an alternative for synthetic fibres (Mohamad et al., 2020). Since the natural fibre are made or can be obtain either harvesting or from processing method of natural resources, it shows that the chemical

properties of the natural fibres consists of protein, cellulose, hemicellulose, wax and lignin depends on the classification of the natural fibres (Ilyas et al., 2022a).

Natural fibres are a promising material that has been utilized in many industrial applications due to their excellent properties. Despite that, there are still some drawbacks that need to be emphasized when dealing with natural fibres. The limitations of using natural fibres are minimal impact strength, delamination, fracture toughness behaviour and brittleness. To overcome this limitations, treatment or reinforcement process of these natural fibres with polymer materials needed which resulting better composites (Verma et al., 2022). Moreover, the properties of natural fibres is not constant where it depend on harvesting season or any factor that could affect the properties of the natural fibres (Al-Azad et al., 2021). Example of natural fibres drawback is flax. It is common in clothing and textile application where flax is sewn able, inexpensive, and offer strong materials but its production process has significant environmental concerns, particularly due to its high-water usage and reliance on pesticides. Next, the natural fibres hemp, is quite famous to be studied due to its strength with compatibility to reinforce with polymer materials. Hemp fibres begin to degrade at a temperature of 150°C, making them an important subject of study in material science (Ahmed et al., 2022).

2.3.1 Classification of Natural Fibre

Fibres are hair-like materials that can exist in the form of continuous filaments or discrete elongated pieces, resembling threads. These materials possess the capacity to undergo a transformation into elongated and slender structures. They can function as a component of composite materials (Syduzzaman et al., 2020a). Natural fibres can be categorized into two distinct groups depending on their inherent qualities: natural fibres and synthetic fibres, which are generally referred to as man-made fibres (Kozlowski & Mackiewicz-Talarczyk, 2020). Figure 2.4 illustrates the classification of natural fibres.

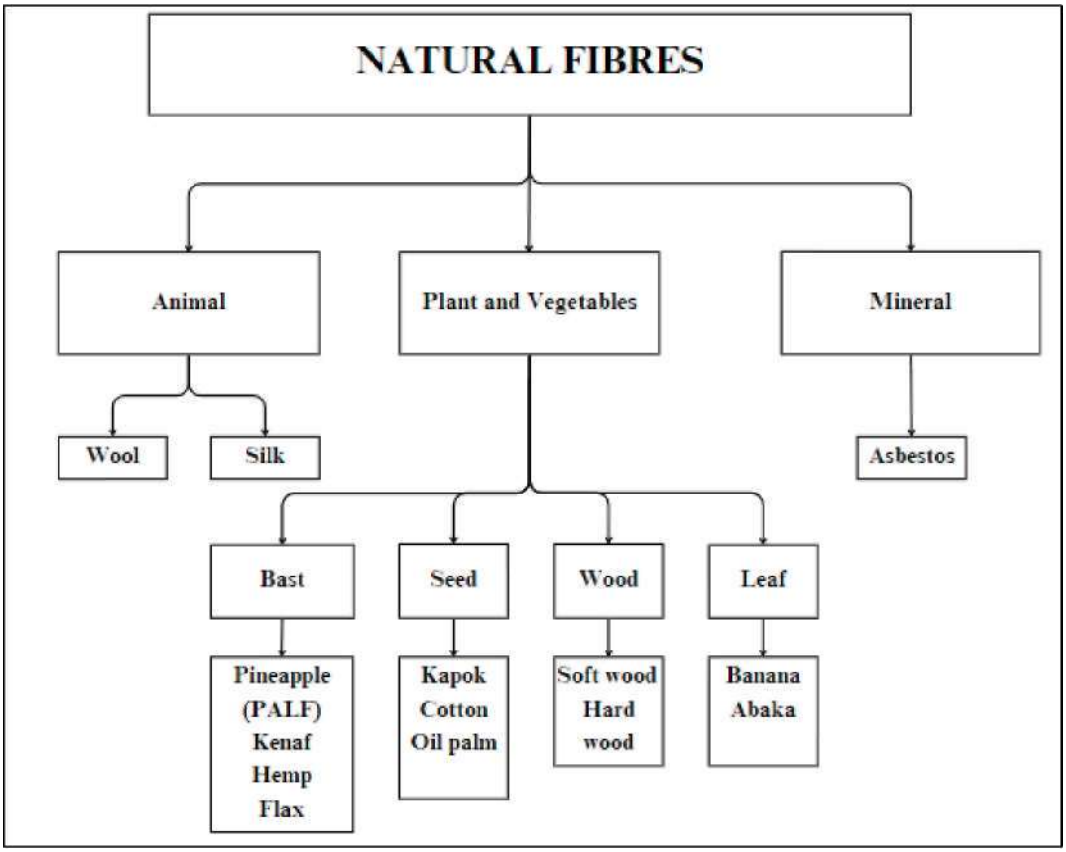

Figure 2. 4 Classification of Natural Fibres

Animal fibres can be harvested from animal sources such are feather, fur, and wool. Animal fibres are composed of protein from the animal. Animal fibres have flexibility, surface toughness and low hydrophilic compared to other fibres (Mhatre et al., 2019). The chemical, physical and mechanical properties make animal fibres an outstanding source that could be utilized as a proxy to reinforced with polymer composites (Aaliya, 2021b). In addition, the utilization of natural fibres like wool and silk is active in textile industry due to its flexibility properties. For instance, wool, from hairy or furry animals such as sheep, goat, and alpaca, is normally utilized for winter jackets due to its rate of heat release and hydrophilic. Wool is primarily composed of a line protein which is keratin. It contained sulphur, oxygen, hydrogen, and amino acid as its composition. Research has been done to investigate the chemical composition of high sulphur content which resulted in wool exhibiting good resistance to chemical impacts (Allafi et al., 2022). Due to the circumstances, it shows that the chemical composition of natural fibres affects the mechanical properties of the animal fibres

(Kerni et al., 2020). Recent studies show that the utilization of cow hair fibre reinforced with polyethylene (PP) is able to enhance the flexural characteristic of bio-composites (Ali et al., 2021). Another study shows that wool reinforced with poly (lactic acid) (PLA) accelerates the degradation cycle of the bio-composites (Szczepanik et al., 2024).

While for the plant fibres, it can be procured from plant sources like wood, plant bast such as kenaf, ramie and hemp. The usage of plant fibres is common for textile industry, automotive industry, clothing and for partition board production (Aaliya, 2021c). Figure 2.4 illustrates that plant fibres can be categorized based on their position within the plant. For instance, bast fibres such as hemp, jute or flax are obtained from the plant's stem, whereas other fibres can be obtained from seeds (cotton), fruit (coconut, pineapple), or even the plant's leaves (sisal). Plant fibres primarily a sugar-based polymer, including cellulose and hemicelluloses, along with pectin and lignin. Additional constituents, such as wax or oil, may also coexist with structural water. Numerous factors, including environmental circumstances, age, and degradation processes, influence the chemical composition of plants. This composition can vary not just between different plants but also among different portions of the same plant. Research from (Akter & Hossain, 2021a) performed a comprehensive literature review in which they recorded the mean chemical content of several plant species. It shows that the natural fibres from plants are high in several compositions as shown in Table 3.

Table 3

Average Chemical Composition of Plant Vegetables Fibre

Chemical composition	Percentages (%)
Cellulose	40-60
Semi-cellulose	20 - 40
Lignin	10 - 25
Ash, additives, and impurities	Low percentage

Composites enhanced with natural fibres have made considerable advancements in recent times due to their capacity to decompose naturally, their affordability, their low weight in comparison to other materials, their exceptional mechanical capabilities, and their renewable characteristics (Aravinth et al., 2022). These composites are

destined to have an increasing number of uses in the near future due to extensive research aimed at understanding and enhancing their properties. An essential aspect to consider when utilising these materials in various weathering circumstances is a comprehensive comprehension of their hygroscopic behaviour (Ilyas et al., 2022b). Plant fibres possess a hierarchical structure and can be utilized at various levels to enhance the strength of composite materials. Indeed, fibres can be processed into fabric yarn, a bundle of fibre, or even individual fibres. The layer with the greatest thickness is the S2 layer. It accounts for 70-80% of the thickness of the secondary wall. Therefore, the characteristics of the fibre are mostly determined by the properties of this layer (de Azevedo et al., 2021). Plant fibre yarns are composed of numerous relatively short plant fibres that are twisted at an angle to the yarn axis, enhancing the yarn's axial strength. The individual fibres possess a structure consisting of several cell walls. The section is polygonal; however, it is typically approximated as round for calculating mechanical parameters. The outer wall is referred to as the major wall. It has a relatively tiny thickness in comparison to the overall thickness of the fibre. The composition of this wall primarily consists of pectin, hemicellulose, low crystalline cellulose and a smaller quantity of waxes (Alhijazi et al., 2020). The secondary wall, comprising approximately 90% of the overall section, is segmented into three layers. The primary constituents of this substance are cellulose microfibrils, which are arranged in a parallel manner and surrounded by an amorphous matrix consisting of hemicellulose, pectin, and lignin. The three layers exhibit distinct characteristics due to variations in their thickness and structural arrangement, including microfibril angle and chemical composition. The mechanical properties of plant fibres are affected by numerous factors, such as the cellulose content, fibre diameter, temperature, microfibrillar angle, presence of defects, and water content inside the fibres.

A variety of plant fibres, including kenaf, cotton, sisal, banana, jute, pineapple, okra, oil palm, coir, and others, are extensively utilised in composite manufacturing. The utilisation of natural fibres in composites fabrication offers several benefits. These include a lower specific weight, resulting in a higher specific stiffness compared to glass. Natural fibres also possess low mechanical properties, particularly in terms of impact resistance. Additionally, they are a renewable resource and exhibit moisture sensitivity. The production of composites using natural fibres requires low investment

and offers low thermal stability and abrasion, resulting in less tool wear. However, natural fibres have low durability and poor fire resistance. On the positive side, they are abundantly available and biodegradable. In addition, plant fibres have been used in many applications, as indicated in Table 4.

Table 4

Application of Plant Fibres

Plant Fibre	**Application**	**References**
Pineapple leaf fibres (PALF)	textile materials (yarn) machineries components conveyer belts cords carpet manufacturing	(Pandit et al., 2020)
Biduri plants (Calotropis Gigantea.)	heat insulation materials textile application (wool)	(Sana et al., 2020)
Date palm tree fibre	thermoplastic, thermosetting biopolymer	(Awad et al., 2021)
Abaca, jute, sisal	automotive industry (car seat, backrest, backdoor liners)	(Syduzzaman et al., 2020b)
Hemp (Cannabis sativa)	textile application paper production	(Tulaphol et al., 2021)
Kenaf fibre	roofing, walling, and ceiling fibrous concrete board panel	(Abbas et al., 2022a)
Bamboo, banana fibre	bio-filler polymeric bio-composites	(Lohar et al., 2020)

An inherent drawback associated with utilising natural fibres as reinforcement in composites is the inadequate adhesion between the fibre and matrix. Within the context of composites, the matrix serves as a binding agent that facilitates the transmission of the fibres' stiffness throughout the material. If the adhesion between the fibres is insufficient, the composite will not possess the desired qualities (Fatriansyah et al., 2022). Furthermore, it will be susceptible to the surrounding conditions in which

it will be utilised, hence reducing its lifespan. Extensive research is being performed to intensify the adhesion between fibres and a polymeric matrix through surface modification of the fibres. The authors offer two approaches: physical therapies such as plasma or corona treatment, and chemical alteration using substances like sodium hydroxide, maleic anhydride, isocyanates, organosilanes, permanganate, and peroxide. Another drawback of these fibres is their susceptibility to variations in characteristics based on the batch, variety, and even the specific location inside the plant. For instance, when examining the mechanical characteristics of flax fibres situated at various points within the stem, it was observed that the flax fibres found in the centre exhibited superior mechanical properties compared to those in other locations (Hariprasad et al., 2020a).

2.3.2 Chitosan

Chitosan (CTS) is an aminoglucopyran that contains glucosamine (GlcN) and N- acetylglucosamine (GlcNAc) (Wang, 2020a). It derived from chitin that is composed of glucosamine and N-acetyl glucosamine (Pellis et al., 2022a). Moreover, chitosan is one of the natural biopolymers that have a modifiable structure and abundant functional group that made it able to process in many sizes and shapes (Wang & Zhuang, 2022). Chitosan can be procured from the exoskeleton of animals such are crabs, eggshells shells, and cells wall of fungi (Azmana et al., 2021a; Kou et al., 2021; Pellis et al., 2022b). Moreover, chitosan has an antibacterial behaviour, decent mechanical, thermal properties, biocompatibility, easy process ability and low viscosity (Aranaz et al., 2021; Azmana et al., 2021b). It has been proved that the reinforcement of chitosan with polymer does not affect the polymer properties, which signifies that chitosan is efficient to be utilized as bio-filler (Fila et al., 2024). To obtain chitosan, various processes are necessary. The first step involves washing and drying the materials that contain chitin, such as prawn shells and crab shells. Next, the shell waste will undergo the demineralization process where it will be diluted in hydrochloric acid (HCl, 1.0 M) or other acids. Then, the demineralized waste will go through to deproteinization process where the waste will be diluted in sodium hydroxide (NaOH, 1.0 M) in a certain period

time, before chitin will be procured. Chitin will undergo a deacetylation process in which it will be dissolved in a solution of sodium hydroxide (NaOH, 12.5 M). Subsequently, a substance called chitosan will be present (Souza et al., 2020).

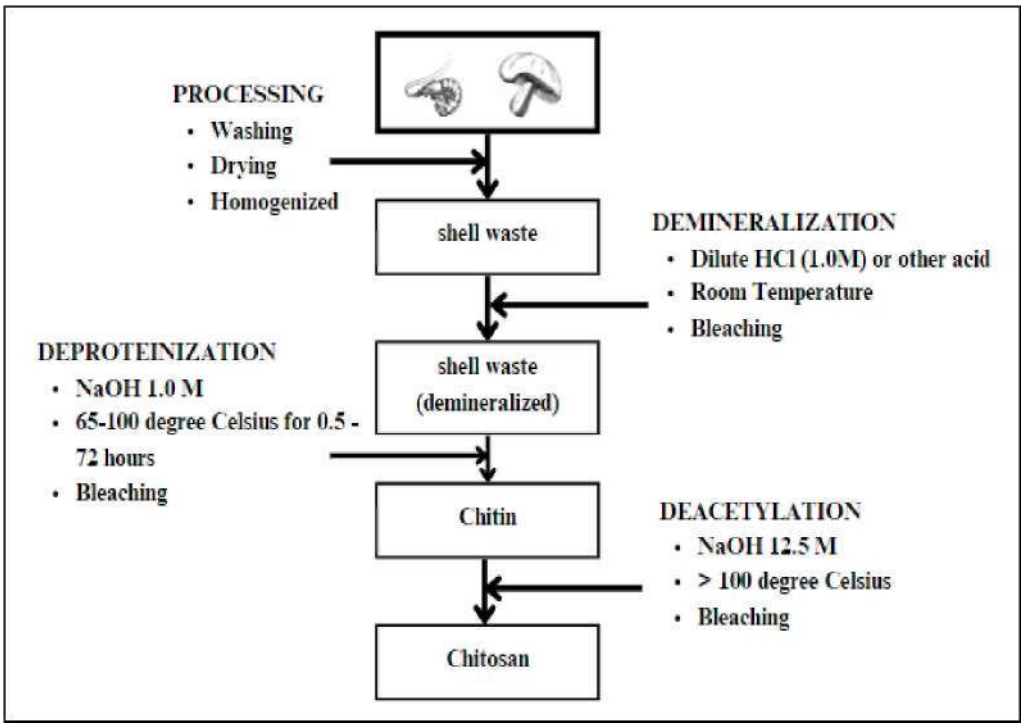

Figure 2. 5 Process of Chitosan Chemical Production Process

These unique polymers have evolved into a new class of physiological materials with good solubility, less crystallinity and are able to respond to changes in chemistry due to the existence of functions groups such as hydroxyl, acetamido or amoxicillin (Oladzadabbasabadi, 2022e; Qin, 2018a). Even though chitosan contain several advantages, such as nontoxicity, biodegradability and proliferation in nature, chitosan-based materials have poor water blocking capacity due to their superior hydrophilic character, which mainly affects their mechanical, gas penetrability, and thermal properties (Arumugam, 2020a; Wang, 2020a). Due to the particular circumstances, these polysaccharides have garnered significant attention as renewable resources. Among them, chitosan-based composites are relevance and are widely used in the sectors of packaging, biomedical, and water purification (Arumugam, 2020b; Gupta, 2019; Oladzadabbasabadi, 2022d).

Due to the high compatibility of chitosan with other biopolymers, mixing chitosan with cellulose and incorporating nanofibers isolated from cellulose can be beneficial (Liu, 2020). Consequently, mixing biopolymers to produce composites is one method for lowering chitosan's hydrophilicity. Combining chitosan with additional polysaccharides resulted in increased obstructive, mechanical, and aesthetic composite characteristics (Cazón, 2018; Tang, 2018). In addition, recent studies stated that the reinforcement of chitosan with the polyvinyl alcohol (PVA) able to improve the performance of the chitosan film via formation of intermolecular of the chitosan with the polymer (Zhao et al., 2023).

2.3.3 Seaweed

Seaweed is an important source of natural biopolymers, which are widely used around the world. Seaweed is classified into three groups which are *phaeophyta* (brown algae), *rhodophyta* (red algae) and *chlorophyte* (green algae). Japan, China and Malaysia have been using seaweed in a long period as for cooking materials and as thickening agent (Das D., 2023). Malaysia in one of the nations that producing red seaweeds (Bhuyar et al., 2020). *Kappaphycus alvarezii* is a red seaweed related to marine algae. These macro algae are primarily grown to produce its hydrocolloids, specifically kappa- carrageenan (k-carrageenan) (Rupert et al., 2022), which are utilized as a thickening and gelling agent in food and non-food applications. Due to its high organic content and impressive phycocolloids and biodegradability, the main seaweed derivatives, such as carrageenan and alginate, have been widely used in the agricultural industry, packaging, cosmetics, and food. It is also known as an effective gas barrier with mechanical properties (Shafie et al., 2022). There are several methods to extract the kappa-carrageenan (k-carrageenan) such are solid-liquid extraction (SLE), microwave assisted extraction method, supercritical fluid extraction method, and ultrasonic assisted extraction method. As for solid-liquid extraction (SLE) method, the red seaweed will be dried and pulverized to break the cell wall. Then, the red seaweed compound will be dissolved in the solvent such are water, methanol, ethanol or either mixed solvent to penetrate the pulverized cell wall to extract from red seaweed (Carpena

et al., 2022). However, the k-carrageenan content varies from 25 to 35% of the total seaweed weight, leaving a large amount of solid waste that has yet to be exploited (Ary Mauliva Hada Putri, 2023).

There has been a lot of study done on natural filler-reinforced polymer composites (Nayak, 2023b; Oladzadabbasabadi, 2022f; Qin, 2018b) however research on seaweed filler-based polymer composites is quite uncommon. Most recent investigations combine seaweed from various species into synthetic polymer matrixes such as poly(lactic) acid, polypropylene, poly (caprolactone) (PCL), high density polyethylene (HDPE), and poly(hydroxide-butyrate) (PHB), resulting in a non-compatible phase of the composite, increasing the water absorption of the matrix as the seaweed content increases. The figure of red seaweed as shown in Figure 2.6.

Figure 2. 6 Red Seaweed (*Kappaphycus alvarezii*).

2.3.4 Kenaf

Kenaf (*Hibiscus cannabinus L.*), a perennial species, is widely planted because it is easy to grow and produces more fibre than flax or ramie. In recent times, the inadequate production of ramie and flax fibres to meet the natural fibres in composites demand. It also has led to an increased reliance on plant bast fibres. Kenaf also has been

a possible substitute for a plant fibres such as jute (Xu et al., 2020). Kenaf is a plant that can be presented for economic and ecological opportunities due to its rapid growth rate where it achieved its maturity within three to four months, and it also requires minimal water. It also has been utilized in many industrial applications (Austin et al., 2024). Kenaf is a versatile plant that already cultivated in many countries especially in Asian countries like China, India, and Malaysia. The kenaf global market is projected to cross USD 854 million by 2025 where it shows that the demand of kenaf is keep increasing and growing (Vayabari et al., 2023). Figure 2.7 shows a harvesting process kenaf.

Figure 2. 7 Harvesting Kenaf (*Hibiscus cannabinus L.)*

Kenaf produces an assortment of edible parts, including seed, leaves, stalk bark, and stalk core. The stem bark comprises robust and lengthy fibres (bast fibres), whereas the stem core comprises short, dense fibres (core fibres). The bark contains bast fibres, which originated from extensive fibre strands that have traditionally been utilized to manufacture ropes and package sacks. Kenaf also known due to its cellulose content in its fibres and core which are 56% and 46% respectively (Khan et al., 2023). Consequently, kenaf has emerged as a globally recognized fibre crop (Abbas et al.,

2022a). To procure kenaf bast, the kenaf bark is separated from the stem and retted after harvesting, which is a fibre-forming technique. The fibre retting process removes the pectin-rich middle lamella that connects adjacent fibre cells, releasing bast fibres. Kenaf fibres have been used in combination with polymers to increase mechanical strength. It has been proven that kenaf exhibit an excellent mechanical properties such are flexural strength and tensile strength compared to other plant fibres such are banana, jute, bamboo and sisal (Krishnamurthy, 2020). Moreover, kenaf also is more adaptable to climatic conditions compared to the certain commercial grown fibres (Kujoana et al., 2023). Moreover, kenaf also known to its high yield (Salih & Qader, 2020) where kenaf yield more compared to other plant fibres such as hardwood where the high yield makes it a more efficient use of land and resource. Kenaf core and kenaf fibre as shown in Figure 2.8.

Figure 2. 8 Dry Separated Kenaf Fibre and Core

A study of kenaf in various polymer matrices has been conducted. The mechanical properties of polyolefins, including polyethylene and polypropylene, were found to be enhanced through research involving kenaf modification (Chauhan, 2022b). Kenaf fibres are commonly used to reinforce polymer matrices, resulting in the formation of fibre- reinforced polymeric composites (Ahmad Saffian et al., 2020). This process significantly enhances the properties of the polymers. Kenaf fibres have

mechanical properties similar to contemporary materials, making them appropriate for a variety of structural and non-structural industrial products using a polymer matrix (Sreenivas et al., 2020). Recent investigations and research efforts have revealed the numerous qualities of kenaf fibres in both original and reprocessed plastics. These features make kenaf fibres appropriate for use in construction materials, such as boards with varying densities, widths, and resistance to fire and insects.

2.4 Recent Work on PBAT Reinforced Natural Sources

The integration of natural fibres into a PBAT matrix significantly impacts the physical properties of the resultant composite material, influencing its mechanical, thermal, decomposition, and crystallinity performance. Continuous advancements in the comprehension of constituent materials provide improved management of various design factors, resulting in increased qualities of composite materials. These materials exhibit improved efficiency in several applications. Natural fibres are often used as reinforcement in polymer composites; however, their insufficient compatibility with the polymer matrix poses a considerable challenge. The adhesion between natural fibres and the matrix in composites is a crucial factor influenced by various elements. Factors include fibre surface, chemical compatibility, roughness, humidity, surface treatments, and intermolecular interactions. The matrix enables the transmission of applied stresses to the fibres within the composite material, protects them from external damage, and maintains the material's shape. Moreover, it must exhibit adequate deformability and compatibility with the reinforcement to impart critical mechanical qualities to the composite while facilitating fibre movement. To achieve this, it must attach efficiently to the fibres, enabling optimal stress transfer and reducing mobility.

The recent works on PBAT reinforced natural sources section delves into a variety of studies on PBAT bio-composites in order to gain a more comprehensive understanding of the cooperation between natural sources and PBAT matrix. Researchers, engineers, and industries able to promote sustainable and environmentally friendly solutions by making informed decisions regarding material selection and design by comprehending the characteristics and properties of PBAT bio-composites.

2.4.1 PBAT Reinforced Plant Fibres

Despite their ecological advantages, PBAT face limitations regarding thermal and mechanical properties. PBAT is an aliphatic-aromatic co-polyester known for its biodegradability, flexibility, and appropriateness for packaging applications. Nonetheless, its comparatively low stiffness and strength constrain its applicability across a wider spectrum of uses. An effective strategy to overcome these limitations is the use of functional additives that improve the mechanical strength, stiffness, and processing characteristics of the material while maintaining its biodegradability and minimal environmental impact. Agro-industrial residues have increasingly attracted attention as multifunctional reinforcements in biodegradable composites. The use of materials like rice husks, cotton straw, and sugarcane bagasse as functional additives in polymer matrices offers a cost-effective and plentiful source of lignocellulosic material that enhances barrier performance, strength, and stiffness. This method aligns with circular economy principles by diminishing dependence on fossil-based resources and transforming agricultural waste into value-added materials.

Leite-Barbosa et al. (2025) conduct an investigation into the reinforcement of PBAT using *açaí* seed residues (ASR), which is a plentiful agro-industrial byproduct originating from Brazil. The residues were analysed through FTIR, XRD, TGA, and SEM approaches, while the bio-composites were subjected to assessments of their mechanical, morphological, and thermal properties. In comparison to pure PBAT, the addition of ASR led to a decrease in tensile strength, while the elastic modulus and flexural strength saw an increase. Thermal analysis indicated that the filler had a nucleating effect, raising the crystallisation temperature from 65.55 °C to 79.90 °C. The thermal resistance exhibited by all bio-composites displayed a comparable weight loss curve profile in relation to temperature, accompanied by a minor decrease in the initial degradation temperature (T_{onset}) when contrasted with neat PBAT.

Moreover, another study from Yun et al. (2025) by reinforcing PBAT with cellulose-rich fibre (CRF) in which extracted from kimchi cabbage (KCB) in which an agro-industrial byproductthat most commonly well known in Korea. The KCB is quite known in country Korea in which it became a national food for locals. Therefore, the

study was done to fulfil the knowledge gap. The kimchi cabbage (KCB) residue was analysed through several analyses such as FTIR, DSC, SEM and XRD approaches. The bio-composites were subjected to assessments of their mechanical, morphological, and thermal properties. The PB AT/CRF bio-composites show the addition of high content of CRF within the PBAT matrix leads to increase brittleness compared to the neat PBAT. The reinforcement of cellulose-rich fibre (CRF) within the PBAT bio-composites depict that the crystallization temperature of the PBAT/CRF as the cellulose-rich fibre above 2.5% within the bio-composites. The results show that the addition of cellulose-rich fibre within the PBAT matrix acted as nucleating agent in which reflected the enhanced thermal stability of the crystalline regions of the PBAT/CRF bio-composites.

Moreover, another recent study from Xu et al. (2024) in which the investigation regarding the reinforcement of PBAT and reed fibre (RF) as bio-filler. The researcher recorded that reed *(Phragmites australis)* is a perennial herbaceous plant that has a rapid growth, high yield and ability to thrive without the need of fertilizers. Reed is well known in China due to its low density and excellent mechanical properties. The researcher believes the reed fibre has a high specific strength and modulus. The PBAT/RF bio-composites were subjected to several analyses such as degradation analysis, Fourier transform infrared spectroscopy (FTIR), scanning electron microscopy (SEM), and differential scanning calorimetry (DSC). The thermal analysis of PBAT/RF bio-composites recorded that the reed fibre (RF) functioned as nucleating agent in which increasing the crystallization peak temperature of the bio-composites. Moreover, the morphological analysis depicts that the PBAT/RF bio-composites becoming more hydrophilic compared to the neat PBAT due to the hydrophilic that reed fibre has, the hydrophilicity of the PBAT/RF bio-composites fasten the degradation process compared to the neat PBAT. It shows that the reinforcement of reed fibre (RF) increases the hydrophilic traits of PBAT/RF bio-composites.

In addition, Ye et al. (2024) conduct an investigation regarding the reinforcement of bamboo powder (BP) with PBAT in which this study emphasis on tensile modulus and creep resistance of the PBAT/BP bio-composites. Bamboo is one of the plants that has a short growth life cycle. The researcher recorded that the BP was surface modified with y-(2,3-epoxypropoxy) propyltrimethoxysilane (KH560). The

PBAT/BP bio-composites asses to mechanical properties analysis. The researcher recorded that the reinforcement of BP within the PBAT matrix increase the tensile modulus and flexural modulus of treated K-BP/PBAT composites by 225% and 608% as the BP concentration content is 25 wt% compared to the neat PBAT while the creep resistance of K-BP25 was increased by 3.25 compared to the neat PBAT. The researcher also recorded that the K-BP content within PBAT matrix enhanced the nucleation ability of PBAT.

A recent study by (Biswal, 2024) in which investigating the reinforcing effect of jute fibre (JF) in a poly (lactic acid) (PLA)/ poly (butylene adipate-co-terephthalate) (PBAT) matrix in advanced engineering application, as the reinforcement done through melt blending process. The researcher stated that the jute fibre (JF) is renewable and biodegradable in which suitable for reinforcing with PLA/PBAT. The PLA/PBAT/JF bio-composites were assessing its mechanical properties, thermal stability, and morphological features. Thus, the researcher compared the properties of PLA/PBAT/JF 15% bio-composites with the commercial car dash box. The researcher recorded by the reinforcement of JF within the PLA/PBAT matrix enhanced the impact toughness and mechanical compatibility comparable to the commercial engineered plastics, Acrylonitrile Butadiene Styrene (ABS). The SEM analysis also determines that the PLA/PBAT/JF has a good interfacial bonding.

Techawinyutham et al. (2024) conduct a study regarding the reinforcement between PBAT with durian and mangosteen peeled waste as natural pigment. The mangosteen and durian peel wastes were collected around local market in Thailand. The durian (D) and mangosteen (M) peeled skin were turned into powder to be reinforced with PBAT in which resulting three types of matrices in which M/PBAT, D/PBAT, and M/D/PBAT composites. The fabrication of brown mangosteen powder and light brown durian powder within the PBAT composite resulted in PBAT colour changes from white to black. These analyses were assessed by CIELAB colour system. The M/PBAT, D/PBAT and M/D/PBAT composites were subjected to morphological, thermal, mechanical, chemical composition and rheological properties analyses. The thermal analysis indicates that the M/PBAT is more superior compared to the D/PBAT. Therefore, the researcher stated that the addition of durian and mangosteen powder within the PBAT matrix able to add value to this waste and the bio-composites is

become rougher in which resulting that the bio-composites has promising potential for food packaging and medicine zipper packaging applications.

Ren et al. (2024) conduct an investigation regarding the utilization of soybean protein isolate (SPI) as filler to be reinforce with PBAT. The researcher stated that the utilization of SPI as filler is to reduce the cost of the PBAT due to its availability and inexpensive natural polymer. Therefore, the utilization of series of polycaprolactone-based polyurethane pre-polymers (PCLPUs) with different soft segment molecular weights is to improve the compatibility between the SPI/PBAT. The SPI/PBAT bio-composites were assessed through several analyses which are structural morphological and property analyses. The result recorded with the addition of 5% PCLPU-500 content to SPI/PBAT bio-composites indicate that the bio-composite tensile elongation, impact strength and the tensile strength was increased by 867%, 264%, and 150% compared to the SPI/PBAT bio-composites that contain none PCLPU-500 content within the SPI/PBAT bio-composite.

A study regarding improvement of mechanical and thermal expansion properties of pectinase treatment plant fibre, coconut coir and bamboo fibre reinforcing with starch/PBAT bio-composites by Yang et al. (2024). The study material is subjected to the several assessments such as evaluating thermal properties, mechanical properties, and microstructure analysis. Therefore, the result researcher recorded was the presence of 15 wt% of pectinase treated bamboo fibres within the starch/PBAT bio-composites indicates the highest improvement of bending strength by 65% compared to the starch/PBAT bio-composites without added fibres. Therefore, the researcher stated that the reinforcement of fibres is significantly enhanced the starch/PBAT bio-composites.

Another researcher Zeng et al. (2022) conduct a study regarding mechanical properties and tensile model between the reinforcement of hemp fibre (HF) and PBAT. The utilization of hemp fibre is widely known across China due to it traits where it has a good excellent mechanical property (tensile modulus and tensile strength), lightweight, and also able to give a good impact on the environment. Hemp fibre also is a promising potential to add value to the PBAT. However, due to the HF traits where it is hydrophilic, the HF was subjected to the alkylation treatment to procure silane (Si-HF), while the PBAT/HF bio-composites is assessed to morphology, density, mechanical properties and thermal stability analysis. The density of the treated HF (Si-

HF)/PBAT bio-composites were lower compared to the neat PBAT. Moreover, the tensile strength and tensile modulus of both Si-HF/PBAT and untreated HF/PBAT increased, However, the tensile strength and the tensile modulus of Si-HF/PBAT is higher compared to the Hi/PBAT bio-composite. As for the thermal stability, the thermal decomposition of HF/PBAT is lower compared to the pure PBAT while the thermal stability of Si-HF/PBAT was higher compared to HF/PBAT. The morphological studies indicate that the Si-HF has better interfacial interaction with PBAT compared to the HF. Thus, the researcher also stated that both bio-composites (Si-HF/PBAT and HF/PBAT) is suitable for lightweight composite application due to the reduction in bio-composites density.

A study from Meraj et al. (2025) in evaluating the properties of reinforcement between PBAT and microcrystalline cellulose (MCC) that procured from kenaf fibre by using eutectic green solvent in order for packaging applications. Both MCC functioned as fillers and PBAT were subjected for structural, morphological, tensile and thermal properties. The thermal stability analysis indicates that the PBAT/MCC bio-composites film exhibit enhanced thermal stability compared to the neat PBAT sample. In addition, 1.5 wt% PBAT/MCC films have higher peak temperature compared to the neat PBAT in which 408.56°C. It indicates that the PBAT/MCC films is suitable for typical food packaging in which able to withstand with moderate heat during handling and storage. The addition of MCC content within the PBAT matrix indicate that the surface of the MCC/PBAT films becomes rougher due to the presence of cavities and holes under the SEM. The tensile strength of the MCC/PBAT film decreased compared to the neat PBAT in which due to the addition of MCC content with the PBAT matrix that made the film become stiffer while the tensile modulus of PBAT/MCC higher compared to the neat PBAT. The researcher stated that the PBAT/MCC film is has promising potential to be utilized as compostable film for food packaging appliances.

Referring to the recent studies, the utilization of plant fibres as filler to be reinforce with PBAT is highly promising. There are many research and published study that have been recorded regarding the reinforcement between PBAT and plant fibres as additives or filler in the matrix in which signifies the good compatibility of both materials (PBAT and plant fibres). Therefore, the PBAT is a biodegradable polymer that should be highlighted for packaging applications due to its characterization that

similar to the low density polyethylene (LDPE). However, the utilization of plant fibres as filler able to altered the properties of PBAT such are increasing the tensile modulus, increasing the peak crystallization temperature, and even can be tough as commercialized plastic. The plant fibres also are cost-effective material that can be utilize as filler that able to be reinforce with PBAT. Therefore, most of the objective of the utilization of plant fibres as filler is to fully utilized the industrial residue or waste and in the same way it can reduce the cost of PBAT utilization while producing a promising bio-composites that suitable for lightweight composite and packaging applications.

2.4.2 PBAT Reinforced Animal Fibres

Despite of the limitation that occur while utilizing PBAT as matrix such as low modulus and low constrain strength. This aliphatic aromatic biodegradable polymer has extensively promising polymer due to its flexibility and appropriateness that suitable for packaging applications. Therefore, to cope with the low stiffness and strength constrain difficulties while utilizing PBAT, an effective alternative to overcome these difficulties is the utilization of functional additives that able to improve PBAT mechanical properties, stiffness and processing while maintaining PBAT biodegradability and minimal environmental impact.

Therefore, the utilization of animal fibres is one of the effective strategies. By utilizing the animal fibre as additives in PBAT blends it could affect several factors like sustainability and waste valorisation. It is due animal fibre like wool, cashmere, mohair, feather, and silk are most likely agricultural or industrial by-products. The animal fibres like wool and keratin is high elasticity, heat insulation, warmth retention and soft in which most well-known to applied in textile and clothing applications. Hence, the animal fibres often disposed as residue or waste. By utilizing the animal fibre as additive in reinforcement materials with PBAT, it could add the PBAT value while contributing to the circular bio economy goals. As an example, the feather of poultry was produced in millions tons annually, but it is underutilized. Thus, by utilizing the animal fibres, it

could lead to the sustainability and it can valorise the animal fibres that originated as industrial residue or waste.

Furthermore, the utilization of animal fibres like wool, hair, feathers silk is rich in fibroin and keratin. Keratin is well known as functional biopolymer that consists high tensile strength, good thermal stability and it contains of amino groups (-NH2), sulfyhydryl group or thiol group (-SH) and carboxyl group (-COOH) in which it is a promising for better bonding or chemical modification for PBAT. In addition, these properties able to alter and enhance the PBAT bio-composite performance in flexibility, rigidity, toughness and PBAT biodegradability. Unfortunately, there are limitation and difficulties to assess the knowledge regarding the reinforcement between PBAT and animal fibres through recent studies and research. One of the reasons that can be a hypothesis is due to research of animal fibre reinforcement with PBAT is extremely niche compared to the reinforcement of PBAT and plant fibre research. Therefore, keratin-based bio-filler (e.g., poultry feathers, wool) are most likely explored in other polymers while as for PBAT, the utilization of plant fibres (e.g., jute and reed) as bio-fillers are commonly found in research.

However, there are limited research that have been recorded in recent years regarding the reinforcement of animal fibres and PBAT in order to assess to fulfil the knowledge gap regarding the lack of research for reinforcement between animal fibres and PBAT. A study from Vadillo et al. (2025) regarding the utilization of steam-exploded feather as additives within the PBAT matrix. Chicken feather are predominantly composed of keratin in which it is light weight, thermal insulation properties and high strength. Therefore, keratin is well known as with its rich in nitrogen in which it is very valuable functioned as key nutrient of plants. The utilization of feather additive undergoes through steam explosion treatment in order to procure steam exploded feather (SEF). The PBAT/SEF bio-composite sample were subjected to the several assessments which are ecotoxity test in soil, disintegration in soil, biodegradation in soil, mechanical testing, field-emission SEM, TGA, and density analysis. The tensile modulus of PBAT/SEF bio-composites shows an improvement in which the tensile modulus of PBAT/SEF are recorded higher compared to the pure PBAT with 200 MPa and 119 MPa respectively. As for the thermal analysis (TGA), the thermal stability of PBAT/SEF bio-composite is more higher compared to the neat

PBAT. This study also recorded that the addition of SEF within the PBAT matrix indeed reducing the density of the bio-composites. Therefore, the SEF within the PBAT tends to degrade at lower temperature approximately around 200 °C.

In addition, Fodorean et al. (2024) conduct a research regarding the possibilities of utilizing natural fibre and wools for fabrication of bio-composite materials. This research reinforcing a sheep wool as a bio-fillers to be reinforced with starch/PBAT composite and the other PBAT matrix with miscanthus fibre. The ratio of the mixture recorded was 60 % starch (S), 30 % PBAT, and 10 % sheep wool (SW) while the other PBAT matrix is 60 % starch (S), 30 % PBAT, and 10 % miscanthus fibre (MF). Both of the PBAT matrix is subjected to tensile test in which consists of young's modulus, tensile strength, and elongation at break. The research recorded that the tensile strength of S/PBAT/SW bio-composites is higher compared to the S/PBAT/MF bio-composites. It indicates that the S/PBAT/SW bio-composites is more flexible compared to the other bio-composites in which shows for more suitability for packaging film applications while the S/PBAT/MF bio-composites are prone for structural packaging applications.

The limitation of research regarding reinforcement with animal fibre and PBAT indicates to limiting the assess of knowledge whereas the reinforcement between PBAT and animal fibres has a promising result for PBAT reinforce with animal fibres. The reinforcement between PBAT and animal fibres indicates that the matrix tend to be more flexible and ductile compared to the reinforcement between PBAT and plant fibres in which resulting stiffer and rigid matrix. Moreover, by reinforcing the PBAT with animal fibres has a potential to improved its toughness, impact resistance, and also the elongation at break of the matrix in which it is beneficial for foams and packaging films where the flexibility of the matrix is needed.

The reinforcement of PBAT with animal fibres is promising and also underexplored strategy to create or discover a strong biodegradable composite using waste or residue for packaging applications and this is also aligned with the sustainability goals. However, the limited published studies of reinforcement between PBAT and animal fibres can lead to lack of standard methods or benchmarking data. In addition, research or published study of reinforcement animal fibres with PBAT is highly niche compared to the published study regarding the reinforcement of plant fibres and PBAT where wool or keratin or other animal fibres has been commonly

investigated with other polymers such as PLA, poly hydroxylbutyrate (PHB) but less research with PBAT.

2.4.3 PBAT Reinforced Mineral Fillers

In order to enhance and improve the aliphatic aromatic biodegradable polymer, PBAT, countless of extensive research has been done by engineers and researchers to extent the performance of PBAT due to its lower modulus and lower constraint compared to the other commercialized polymer. Therefore, researcher and engineers has done an extensive discovery to cope with the disadvantages while utilizing PBAT. Moreover, the utilization of mineral fibres (e.g., fibreglass, slag wool, nanoclay and etc.) is one of the strategies to improve and enhance the capability of PBAT. PBAT is a ductile and flexible polymer, by utilizing mineral fibres like basalt, kaolinite and nanoclay, it has a potential to enhance the rigidity of the ductile PBAT in which it able to make the PBAT is suitable for extensive utilization and applications. Thus, the mineral fibres often to produce waste from during production and post-use disposal, Mineral fibre waste is hazardous for human, environment and also living things due to its structure that it is very fine that could harm living things. The utilization of mineral fibres like asbestos also has been banned in many countries due to its microscopic features that could lead to lung cancer and asbestosis. Mineral fibres also are not prone to biodegradable but it takes a long period to be decay. Therefore, by utilizing the mineral fibres as additives for PBAT it could give positive impact not only to human but also environment. Therefore, there is extensive study that has been done regarding the reinforcement of mineral fibres and PBAT.

Zhang et al. (2024) conduct an investigation regarding properties of glass fibre (GF)/PBAT shape memory composites in which the purpose of this study is to enhance the mechanical properties of PBAT while also utilizing modifiers, chain extender ADR4370 and 3-aminopropyltriethoxysilane (KH550). The utilization of this additives also to reducing the cost of PBAT. The composites were subjected to mechanical properties analysis. The findings recorded show that the higher GF content within the PBAT matrix, resulted in reduction in fracture strength and elongation at break while it

increasing the tensile modulus of the GF/PB AT. The researcher stated the modifiers, KH550 functioned to controlled the mechanical properties while the GF concentration is low (5 wt% and 10 wt%) while other modifiers, ADR 3470 controlled the mechanical properties of higher GF concentration within the PBAT matrix (20 wt% and 25 wt%). Therefore, the researcher believes that the GF/PBAT composites exhibit a good mechanical property and able to low the production cost.

Silva et al. (2025) conduct an investigation regarding the reinforcement of PBAT with titanium dioxide ($Ti0_2$) and organoclay (B8 OMMT) to assess the properties of bio-composites for food packaging applications. To assess the properties, the bio-composites were subjected to X-ray diffraction (XRD), differential scanning calorimetry (DSC), thermogravimetric analysis (TGA), field emission gun-scanning electron microscopy (FEG-SEM) and tensile testing analysis. The thermal properties analysis findings recorded that the nanoparticle did not significantly alter the PBAT crystallinity or thermal stability (Tonset) in which 375 °C in which favourable for packaging stability. The presence of Ti02 within the PBAT did not significantly affect the young's modulus of $Ti0_2$/PBAT composites (67.5 MPa) while $Ti0_2$/B8/PBAT exhibit a higher Young's modulus compared to the $Ti0_2$/PBAT composites with the $Ti0_2$ concentration of 0.3 wt% and 0.5 wt% (77.6 MPa) and (97,0 MPa). It shows that the reinforcement of $Ti0_2$ within the PBAT matrix increase the stiffness of the composites. The researcher stated that the composites is favourable to be a candidate for biodegradable packaging applications.

Togliatti et al. (2022) conduct an investigation regarding the PBAT-based bio-composite reinforced with bioresorbable phosphate glass (PG) microparticles. The PBAT is reinforced with PG at 2, 10, and 40 wt%. The PBAT/PG bio-composites was subjected to SEM, dynamic mechanical analysis (DMA), and tensile properties analysis. The recorded findings show that the young's modulus of PBAT/PG bio-composites increases along with the addition of the PG concentration (2, 10, and 40 wt%) (123 MPa) within the PBAT matrix in which resulting the young's modulus of PBAT/PG is higher compared to the neat PBAT (102 MPa). It indicates that the PBAT/PG bio-composites become stiffer compared to the neat PBAT. The modulus storage, (E') also shows significant increases as the PG content within the PBAT matrix (252 MPa) compared to the neat PBAT (178 MPa). The morphological study indicates that the PG

particles shows an irregular geometry in which there is no voids at the interface recorded resulting in good compatibility of PG particles with the PBAT matrix.

Venkatesan et al. (2023) conduct an investigation regarding reinforcement between kaolin clay (KO) with PBAT in order to assess the characterization of the PBAT/KO bio-composites. Kaolin clay is commonly known as China clay in which very known in industrial application like ceramics, paper, and plastics. Kaolin is chemically formed by two layers which is octahedral $AlO_2(OH)_4$ and tetrahedral SiO_4. This research is assessed through several characterization analyses like FTIR, XRD, SEM and mechanical test with the KO different concentration within the PBAT matrix. The morphological study indicates that the presence of KO within the PBAT matrix made the interface of PBAT/KO was rougher compared to the smooth surface of PBAT. The thermal analysis indicates the higher KO content within the PBAT matrix, the higher residual weight. The addition of KO within the PBAT matrix increased the tensile strength of PBAT/KO bio-composites where 1.0 wt% PBAT/KO (19.20 MPa) while the neat PBAT tensile strength is 18.34 MPa. The elongation at break of PBAT/KO decreasing as the KO concentration is increases within the PBAT matrix compared to the neat PBAT. The researcher stated that the PBAT/KO offered potential as materials for food packaging that able to extend its shelf life and able to prevent bacterial growth.

A study regarding reinforcement between PBAT and $CaCO_3$ by Liu et al. (2023) in which it is investigating the properties of PBAT/ $CaCO_3$ composites modified with the tittanate coupling agent (TC). $CaCO_3$ is normally found in limestone or chalk. The properties of unmodified and modified $CaCO_3$ were subjected to tensile analysis while the unmodified PBAT/$CaCO_3$ (TC-1) and modified PBAT/$CaCO_3$ (TC-2) composites were subjected to several characterization analyses including tensile testing, SEM, DSC, and TGA. The tensile findings indicate that the both tensile strength and elongation at break of PBAT/ $CaCO_3$ composites (TC-1) is lower compared to the PBAT/T-$CaCO_3$ (TC-2) composites. As the morphological analysis, the surfaced of PBAT/ $CaCO_3$ composites (TC-1) is more rough compared to the PBAT/T-$CaCO_3$ (TC-2) composites. It is due TC-2 uniformly distributed compared to the TC-1 composites. The thermal analysis indicates that the treated $CaCO_3$ functioned as nucleating agent in which increases the crystallization temperature of the composites from 97.51 °C to

99.67 °C. The TC-2 also indicates the increases of decomposition temperature in which increasing the thermal stability of the composites. The researcher believes that PBAT/modified CaCCb composites has a good performance and has a promising for broad application prospect in plastic packaging field due to its high-performance and low production cost.

Post et al. (2021) conduct a study regarding the reinforcement of PBAT with mineral additives, talc. Talc is mineral with chemical compound of magnesium silicate hydroxide (Mg3Si40io(OH)2). Talc also commonly known in talcum powder, ceramics, paints and as filler for plastics and elastomer. This study determines the mechanical properties changes between several polymers (PBAT, polybutylene succinate (PBS), polybutylene succinate adipate (PBSA)) and several mineral fillers (talc, CaCCb, chalk powder and kaolin) with different loadings. The PBAT and talc mechanical properties changes evaluated by mechanical characterization and physical characterization analysis. The tensile modulus of the PBAT/talc increases as the talc filler concentration (%) increases within the PBAT matrix while the elongation at break of PBAT/talc-50 wt% composites indicate higher reading compared to the neat PBAT. Thus, the strength at break of PBAT/talc also shows increases as the talc concentration increases within the PBAT matrix. The impact resistance of PBAT/talc-10 wt% outperformed all the other polymers that reinforced with talc The researcher believes that PBAT/talc still applicable for plastic products (e.g., home appliances, toys and rigid food packaging).

Referring to the recent and previous research and study, it shows that the reinforcement of PBAT with mineral fillers emphasis on improvement or enhancement the properties of the PBAT itself in also to reduce the cost, and extent the PBAT application range in which focusing on sustainability and biodegradable plastics. Thus, by referring to the previous research mineral fillers (talc, kaolin clay, and glass fibre) able to improve the mechanical properties (impact resistance and tensile modulus) of the PBAT matrix. The reinforcement between PBAT and mineral fillers also able to enhance the thermal stability in which it able to improve the thermal stability of the PBAT bio-composites. Since the mineral fillers is the material that has high potential to be waste, especially after the end of the product life and yet the mineral fillers do not degradable, but it will decay in a long period and the residue remains. The mineral residue able to become a sloid waste is it is not decay. Therefore, the utilization of

minerals as fillers really helps to sustain and reducing the waste that from minerals sources. The reinforcement of PBAT with minerals has a promising prospect to produce a stronger bio-composites, cheaper and applicable in real world especially in biodegradable films, packaging and compostable products.

CHAPTER 3
RESEARCH METHODOLOGY

3.1 Raw Material Preparation

Poly (butylene adipate-co-terephthalate) (PBAT), ecoflex® F Blend C1200 was purchased from BASF SE, 67056 Ludwigshafen, Germany. It is in natural colour, and it has faint specific odour which is non-hazardous to consumer. The form of PBAT supplied in granules form and in a solid-state matter. It is semi crystalline, and its melting range ranges within 100 - 120 °C. It is not highly flammable. The density of PBAT is around 0.8 - 1.4 g/cm^3 at 20 °C, while the bulk density of delivered PBAT is approximately 500 - 1,000 kg/m^3 at the same temperature. Additionally, PBAT does not cause corrosion on metal surfaces. Supplied PBAT is incompatible with strong oxidizing agents. As a precaution step, the PBAT was avoided to all sources of ignition such are heat, sparks, and open flame. The PBAT shown in Figure 3.1.

Figure 3. 1 PBAT

Chitosan 38906 (molecule weight 345,500 g mol-1 with degree of deacetylation 84.5 %) was purchased from Chemiz (M) Sdn Bhd, Selangor Malaysia. The chitosan

substance is from 2-Amino-2-deoxy-(l—>4)-P-D-glucopyranan, poly-(l,4-P-D-glucopyranosamine), poly-(l—>4)-P-D-glucosamine. Chitosan supplied in powder state from crab shell with no odour and with melting point of 102.5 °C. The chitosan density is 1 g/cm^3. Chitosan is chemically stable under standard ambient condition (room temperature), stored in temperature below 30 °C. Chitosan is incompatible with strong oxidizing agents. The specification of chitosan 38906 as shown in Table 5. The Figure 3.2 shows chitosan powder that provided by Chemiz (M) Sdn Bhd.

Table 5

Specifications of Chitosan (MSDS, Sigma Aldrich)

Specifications	
De-Acetylation (DAC)	min 90 %
Viscosity (mPas)	53 - 800
Moisture	Max. 10.0%
Ash	Max. 1.5%
pH	6.5-7.8
Particle size (mesh)	100 mesh
Arsenic (As)	max. 0.00005%
Lead (Pb)	max. 0.00005%
Mercury (Hg)	max. 0.00005%
Cadmium (Cd)	max. 0.00005%
Heavy metals (as Pb)	max. 0.001%
Total plate count (cfu/g)	max. 1000
Yeast & mold (cfu/g)	max 100
Micro organism	negative

Figure 3. 2 Chitosan Powder is Provided by Chemiz (M) Sdn Bhd.

The raw red seaweed (*Kappaphycus alvarezii*) was supplied from Green Leaf Synergy Sdn. Bhd. Tawau, Sabah, Malaysia. The red seaweed subsequently rinsed to eliminate superfluous impurities and then subjected to a 24-hour drying process in an oven set at a temperature of 80 °C. Subsequently, the dried seaweed was reduced in size and pulverised using a grinder machine (THOMAS-Willey, Laboratory Mill Model 4, USA) to produce seaweed particles. The dried seaweed was crushed into a fine powder, sieved using a vibrating sieve shaker (Endecotts, EFL2000), and then stored in zip-lock containers. The dried seaweed has an average particle size ranging from 100 to 150 micron and its moisture content varies between 3% to 5%. An analysis of the chemical composition of seaweed was undertaken by the Malaysian Agricultural Research and Development Institute (MARDI) in Selangor, Malaysia. The results are presented in Table 6. The Figure 3.3 shows the raw dried seaweed to a dimension of 100 to 150 microns. The process of preparation of seaweed as shown in Figure 3.4.

Table 6

Chemical Composition of Red Seaweed (*K.alvarezii)*

Chemical compounds	Composition (wt%)
Carbohydrate	58.70
Crude protein	4.20
Cellulose	6.95
Hemicellulose	3.33
Lignin	4.83

Ash	34.49

Figure 3. 3 Dried Red Seaweed from its Raw State to a Dimension of 100 to 150 Microns.

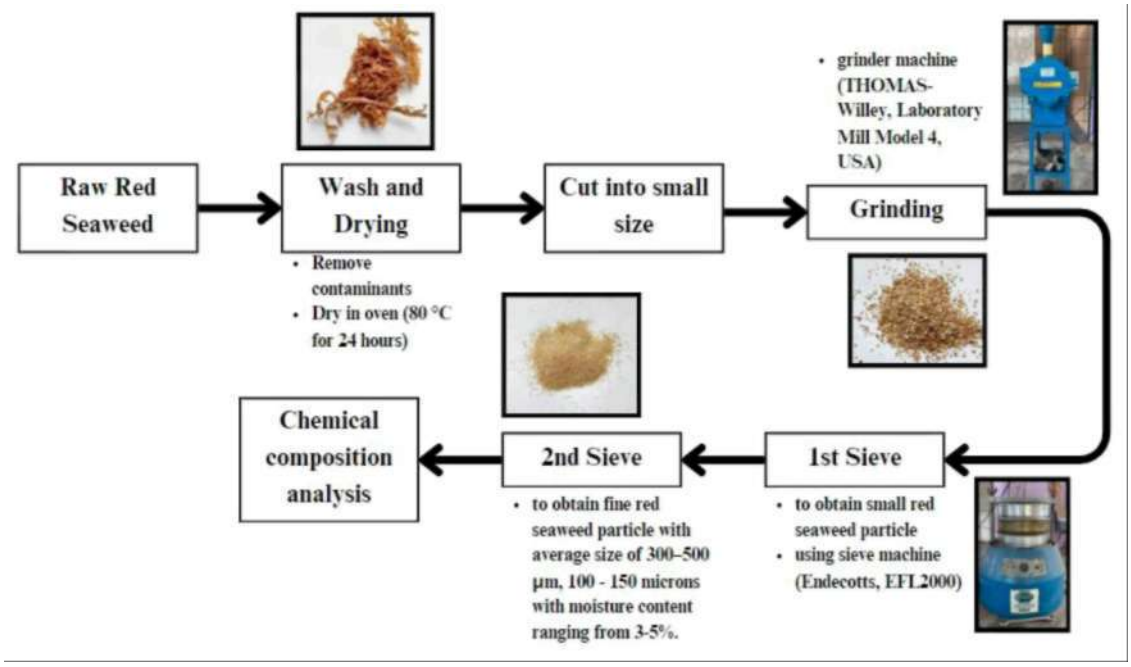

Figure 3. 4 Flowchart of Red Seaweed Preparation Process

The kenaf stems, illustrated in Figure 3.5, were provided by the National Kenaf & Tobacco Board (NKTB), situated in Kuantan, Pahang, Malaysia. The fully grown kenaf plants have reached an age of 4 to 6 months. The kenaf stems were randomly selected and the kenaf bast and cores were manually separated as shown in Figure 3.6. They were then chipped using a laboratory Pallmann Mini Chipper and crushed using a Pallmann Ring Knives Flaker to obtain tiny particles of kenaf. The particles were further dried to a moisture content (MC) of 5% using an industrial oven and then pulverized using a grinder machine (THOMAS-Willey, Laboratory Mill Model 4, USA). The particles went through further screening using a vibrating screener, with only particles that passed through the 100-mesh sieve size being used to produce bio-composite. Both the core and kenaf particle were utilised separately as fillers. Subsequently, the kenaf core and bast were sent to the Malaysian Agricultural Research and Development Institute (MARDI) for chemical composition analysis, as presented in Table 7. The process for kenaf preparation is illustrated in Figure 3.7.

Table 7 Chemical Composition of Kenaf (Core and Fibre)

Chemical compounds	Kenaf (fibre + core)	Kenaf fibre	Kenaf core
Extractive (%)	6.55	5.63	4.6
Holocellulose (%)	86.74	85.62	87.3
a-Cellulose (%)	52	54	47
Lignin (%)	20.19	14.56	19.32
Ash (%)	3.52	5.4	2.1

Figure 3. 5 Harvesting of Kenaf in Kuantan, Pahang, Malaysia

Figure 3. 6 Dried Kenaf Bast (Left) and Kenaf Core (Right)

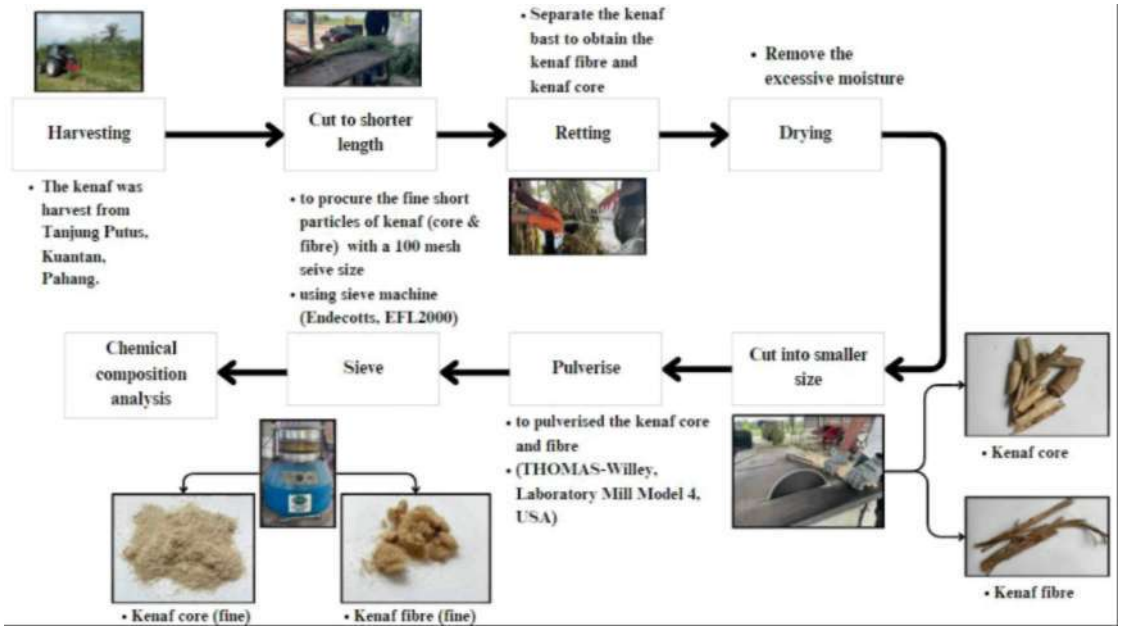

Figure 3. 7 Flowchart of Kenaf (Core and Fibre) Preparation Process

3.2 Fabrication of Bio-Composite Sample

The fabrication of the bio-composite specimen consisted of two main steps: mix preparation and hot pressing in a steel cast mould. The procedure for preparing the blend is outlined as follows: The PBAT polymers were initially melted using a melt-blending, which involved employing a co-rotating Rheomixer batch mixer (HAAKETM Rheomix, Germany) set at a temperature of 120°C and a speed of 60 rpm for a duration of three minutes. The filler materials: kenaf fibre, kenaf core, seaweed, and chitosan, were introduced into the Rheomixer in various weight ratios (as indicated

in Table 8) and further blended for an additional five minutes. A neat PBAT sample was prepared as a control. Following the melt blending process, each blend pelletised using a crusher machine (model:1506 s, MIKASA Sangyo Co., Ltd., Japan).

Table 8

Weight Ratio of Bio-composite Made of PBAT/KF, PBAT/KC, PBAT/SW, PBAT/C Blends with 10, 20 and 30 wt% Ratio

Sample code	**PBAT wt%**	**10 wt%**	**20 wt%**	**30 wt%**	**Total weight blend**
PBAT	80g	-	-	-	80g
KF10	72g	8g	-	-	80g
KF20	64g	-	16g	-	80g
KF30	56g	-	-	24g	80g
KCIO	72g	8g	-	-	80g
KC20	64g	-	16g	-	80g
KC30	56g	-	-	24g	80g
SWIO	72g	8g	-	-	80g
SW20	64g	-	16g	-	80g
SW30	56g	-	-	24g	80g
CIO	72g	8g	-	-	80g
C20	64g	-	16g	-	80g
C30	56g	-	-	24g	80g

The subsequent step in the process included using a steel cast mould to press the panel at a high temperature. The pellets were weighed and placed into a mould with approximate dimensions of 300 mm × 300 mm × 3 mm, with the aim of achieving a density of 1000kg/m^3. Afterwards, they were relocated to the hot press (HEI 100 H). The samples were subjected to hot pressing for a duration of seven minutes at a temperature of 110°C, under a pressure of 5 MPa. The samples encountered post-curing at room temperature for a duration of 24 hours prior to analysis. The procedure for preparing the samples depicted in Figure 3.8.

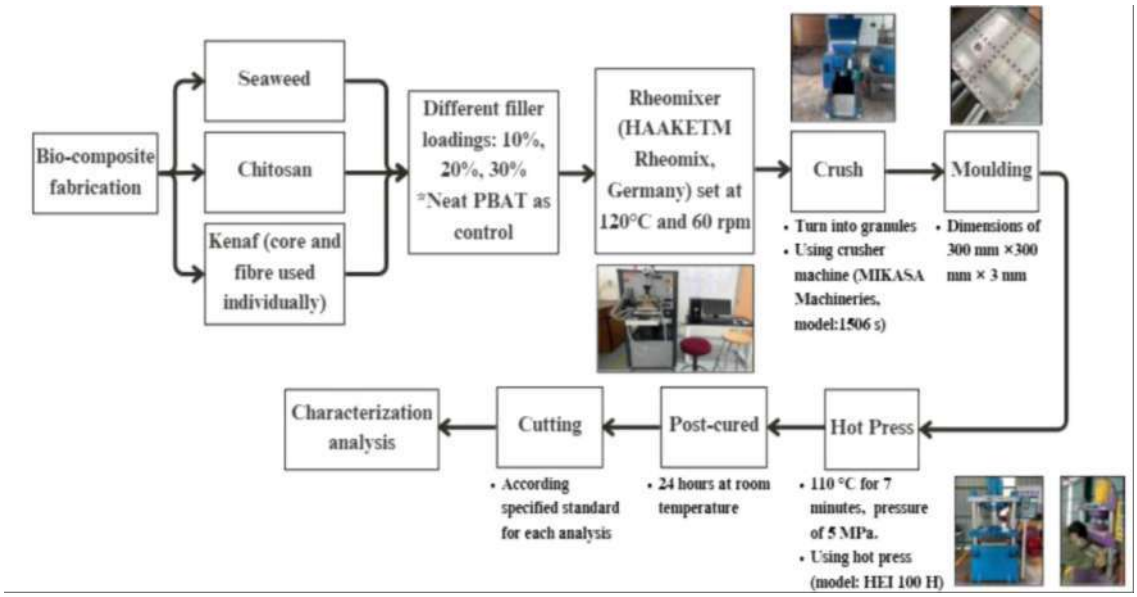

Figure 3. 8 Flowchart of Fabrication of Bio-composite Sample Process

3.3 Characterization Analysis

The characterization of bio-composites was conducted through several analyses following the production of the bio-composites, as depicted in Figure 3.9.

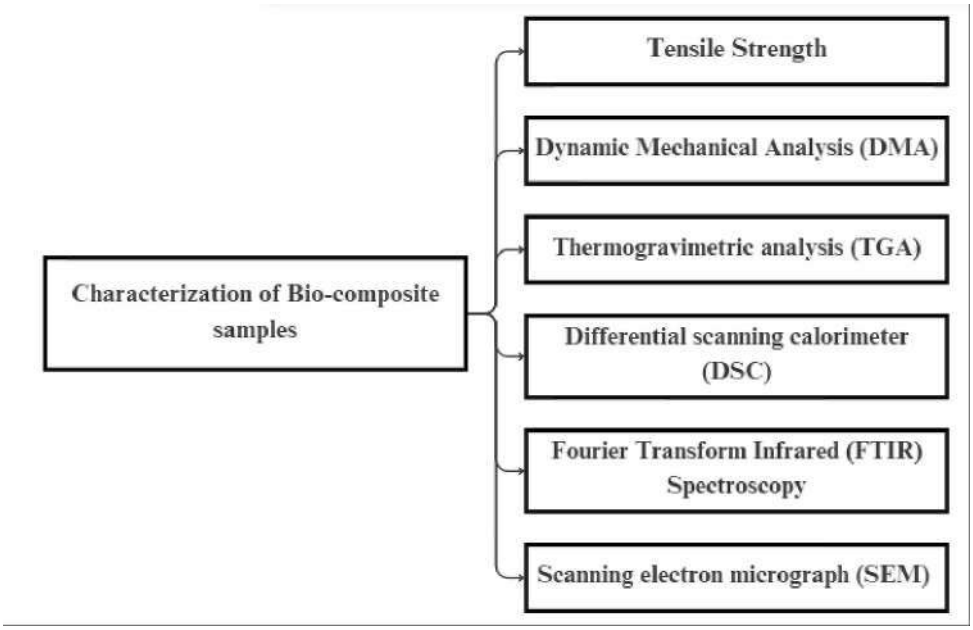

Figure 3. 9 Characterization Analysis of Bio-Composite Samples

3.4 Tensile Properties Test

The tensile properties were determined in accordance with the test standard stated in ASTM D 638 type VI. The test was conducted using a Universal Testing Machine (50 kN Blue Hill INSTRON Universal Testing Machine, INSTRON Corp., United States) equipped with a 50 kN load-cell that was computer-controlled. The crosshead speed was set at 1 mm/min for a duration of 3 minutes until the specimen failed. The dog-bone specimens were obtained by cutting a compressed specimen with dimensions of 115 x 19 x 3 mm in length, width, and thickness. Five specimens of each sample were tested and the mean values of the tensile strength, strain at break and modulus of elasticity of all the specimens tested were calculated. Figure 3.10 illustrates the tensile strength sample and Figure 3.11 illustrates the tensile strength test.

Figure 3. 10 Tensile Strength Sample

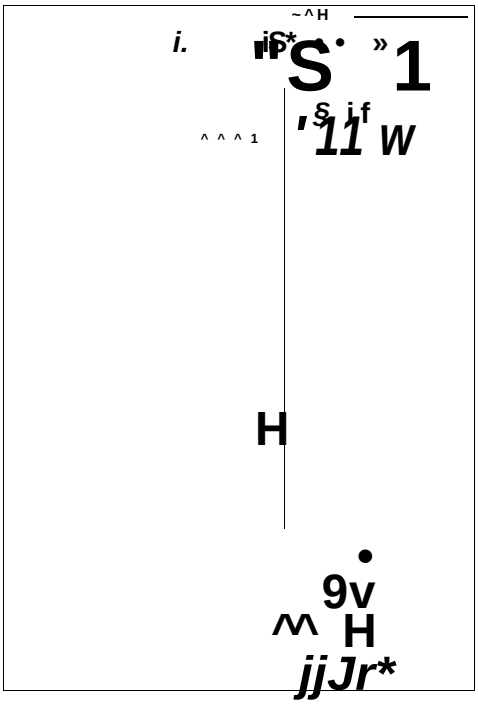

Figure 3.11 Tensile Strength Test Using Universal Testing Machine (50 Kn Blue Hill INSTRON Universal Testing Machine, INSTRON Corp., United States.

3.5 Dynamic Mechanical Analysis (DMA)

The Dynamic Mechanical Analysis (DMA) was performed using the TA Q800 instrument. Rectangular specimens measuring 60 mm x 12.5 mm x 3 mm were employed for performing dynamic mechanical experiments. A temperature scan from -50 to +100 °C was conducted in a nitrogen atmosphere using a three-point bending mode at a heating rate of 3 °C/min and a frequency of 1 Hz. The mechanical loss factor (tan 8), loss modulus (E''), storage modulus (E^1) were measured in relation to temperature during the measurement.

The DMA analysis shows that the complex modulus is a major distinguishing feature of any material sequence which changes over time, as shown by Equation 1.

$$E^* = \frac{\mathbf{q(t)}}{\mathbf{£(t)}} = \frac{a_0 e^{i(a)t+8)}}{e_0 e^{IOJt}} = e^{lS} = [\cos S + i \sin S] \qquad (1)$$

As demonstrated in Equation 2, the storage and loss modulus, two essential components of this time-dependent distinguishing feature, will likewise be related to temperature.

$$E^* = E' + iE'' \tag{2}$$

The product of these two quantities, $E' = —[\cos 8]$ and $E'' = —[\sin S]$, yields an estimate of elastic retrieval of distortion energy and heat dissipation. Alternatively, measurements of the material's stiffness and damping capability was performed. The loss factor was calculated as a function of temperature to investigate energy dissipation and deformation energy storage per cycle, as shown in Equation 3.

$$\tan 5 = \frac{E''}{E'} \tag{3}$$

Figure 3. 12 Dual Cantilever Mode Used for Dynamic Mechanical Analysis (DMA)

3.6 Thermogravimetric Analysis (TGA)

The thermal stability of the composite was examined using thermogravimetric analysis (TGA) in line with ASTM El 131-0. TGA was performed, which typically involved measuring polymer mass as a temperature function or time by providing the sample to a regulated temperature programmed in a controlled environment. In the oxidative environment, polymers start to lose mass, although mass growth can be observed prior to breakdown at a gradual rate of heat. All measurements were done at the INTROP, UPM Laboratory of Bio-composites using a Mettler TGA Q 500 TA (Schwerzenbach, Switzerland). To avoid oxidation, the TGA was measured in a nitrogen atmosphere at a flow rate of 50 ml/min. The weighed composite samples (40 mg to 45 mg) placed in an alumina and warmed to different temperatures ranging from 0 °C to 600 °C at a rate of 10 °C/min.

3.7 Differential Scanning Calorimeter (DSC)

Thermal characterisations were conducted using the Q20 differential scanning calorimeter from TA Instruments. The nitrogen purge was conducted on all samples, ranges in weight from 6 to 9 milligrams. To measure non-isothermal crystallisation, all samples initially scanned to determine the material's thermal history. Before the second heating scan to 180 °C at a rate of 10 °C/min, each sample was heated to 180 °C and subsequently chilled to -100 °C. The chilling scan was used to determine the crystallisation peak temperature (Tc). The second heating readings were used to ascertain the degree of crystallinity (Xc), melting temperature (Tm), and glass transition temperature (Tg). The Xc of the sample was determined by comparing the heat of fusion, AH_m with the theoretical 100% crystalline heat of fusion (A//^) using the Equation 4:

$$*c(\%) = \frac{AHm}{A < x (1 - a)} X\ 10o \qquad (4)$$

K)

where A//^ is the heat of fusion for PBAT and an is the weight fraction of the KLPs, which is included to normalise the enthalpy based on the polymer fraction present in the sample. The enthalpy of entirely crystalline PBAT determined by accounting for the contributions of aromatic (0.487) and aliphatic (0.513) groups, as previously determined by 1H NMR. The melting enthalpy of fully crystalline PBAT is 144.5 J/g x 0.487 = 70.37 J/g, where 144.5 J/g is the enthalpy of fusion of fully crystalline poly (butylene terephthalate), (PBT). This is because only the aromatic part can crystallise in PBAT. The Thomson-Gibbs (Equation 5) was employed to determine the crystal structure's thickness:

where L^a is the crystal thickness, <7eis the surface energy of the PBT basal crystal plane (75 x 10-7) J x cnT^2, 7^ is the equilibrium melting temperature of PBT crystals (244 °C). The DSC scans of the samples were used to obtain the T_m. To investigate isothermal crystallisation, the specimens were cooled at a rate of 30 °C/min from 180 °C to a select temperature (T_c), 115 °C, and then crystallised. After crystallisation, the melting behaviour of the samples was examined by heating/melting them at a rate of 10 °C/min until they reached 180 °C.

3.8 Fourier Transform Infrared (FTIR) Spectroscopy

The molecular interaction between PBAT and the filler particle was confirmed using an FTIR Perkin-Elmer 1720X spectrometer in transmission mode. The FTIR spectra were captured with a resolution of 4 cnT^1 and 32 samples in the spectral range of 4000 cnT^1 to 700 cm^{-1}. The OC and EOC specimens were produced by pressing 100 mg of desiccated KBr with two mg of powder to produce a thin disc. The KBr disc was employed as a reference.

3.9 Scanning Electron Micrograph (SEM)

The SEM analysis was performed using the EM-30AX scanning electron microscope (COXEM, Daejeon, Korea) as shown in Figure 3.13 at an accelerating voltage of 2 kV to evaluate the morphological properties of the filler and the matrix. Samples were gold-coated by an electro deposition technique to impart electrical conduction.

Figure 3. 13 Scanning Electron Microscope EM-30AX (COXEM, Daejeon, Korea).

CHAPTER 4
RESULTS & DISCUSSION

4.1 Tensile Strength Properties

The bio-composites test specimens underwent tensile testing, and the resulting data are illustrated in Figures 4.1 until 4.3. The addition of bio-filler; KF, KC, SW, and C resulted in a decrease in ultimate tensile strength and elongation at break. Nevertheless, the tensile moduli exhibited significant enhancement due to the incorporation of bio-filler. Furthermore, an increase in the quantity of bio-filler within the bio-composite corresponded with an increase in tensile moduli.

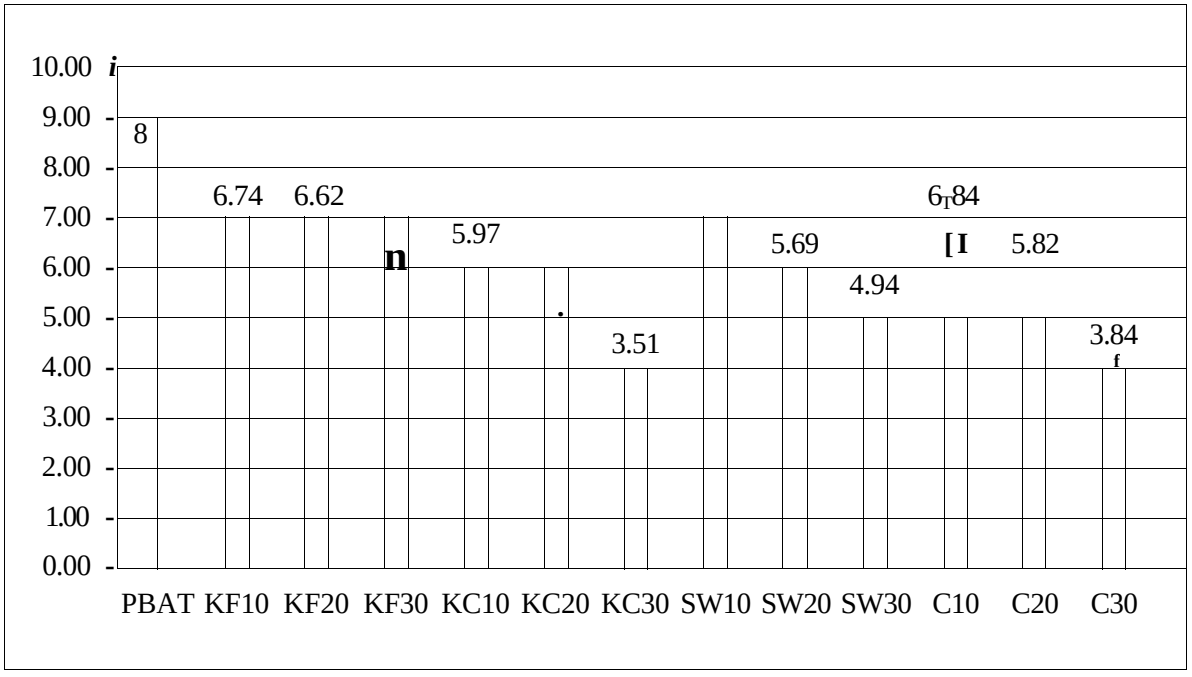

Figure 4. 1 Tensile Strength Properties of Bio-composite

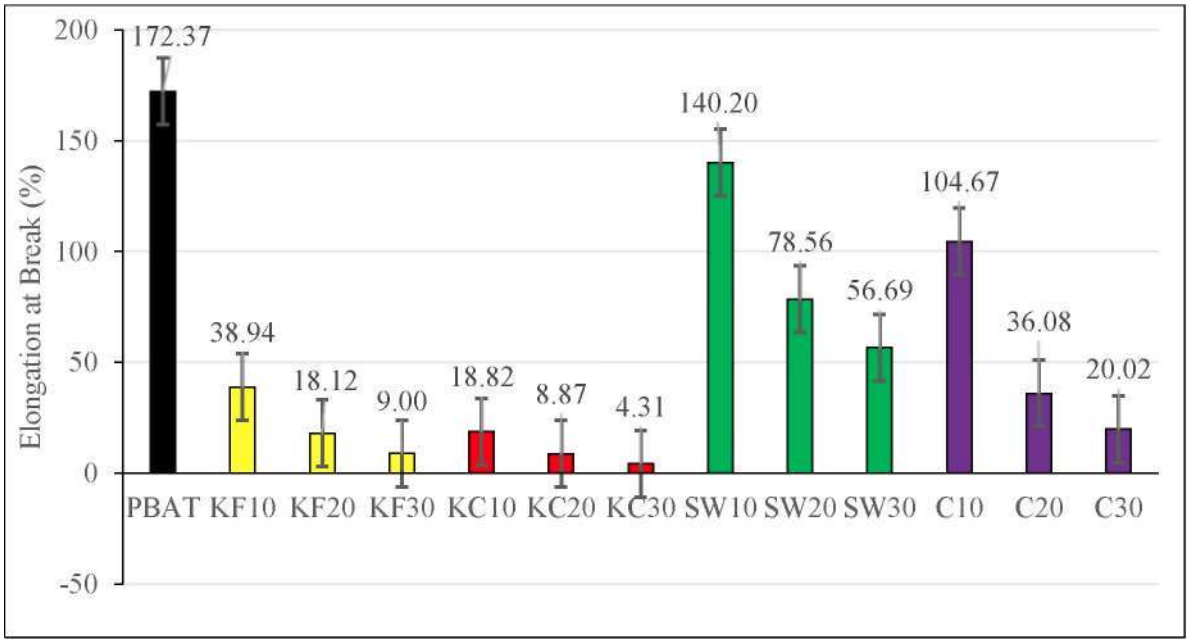

Figure 4. 2 Elongation at Break of Bio-composite

The tensile strength has significantly decreased to 6.74 MPa (KF10), 5.97 MPa (KC10), 6.28 MPa (SW10), and 6.84 MPa (C10) from an initial value of 8.26 MPa (PBAT). The observed trend for elongation at break percentage demonstrated a notable decline, decreasing to 38.94% (KF10), 18.82% (KC10), 140.20% (SW10), and 104.67% (C10) from an initial value of 172.37% (PBAT) as shown in Figure 4.2. This decrease indicates a deterioration in the interfacial adhesion between the bio-filler and PBAT chains. The elongation at break of the bio-composite decreased as the concentration of bio-fillers within the PBAT bio-composites increased. Marques et al. (2015a), Pinheiro et al. (2019) and Da Silva et al. (2017) have documented similar behaviour in their studies of PBAT-based composites including *curauá* (5 wt.%), munguba (10 wt.%), and peach palm tree fibres (20 wt.%), respectively. The highest tensile strength recorded is C10 at 6.84 MPa, and the lowest is KC30 at 3.51 MPa. The decrease in tensile strength of the bio-composite is likely due to the initial variability of the specimens, a phenomenon expected when rigid particles are included into the polymer matrix. This is because the interface between the bio-filler and matrix, inclusive of the presence of voids, might serve as flaws in the composites, despite the strong adhesion between the bio-filler and matrix. Moreover, the incorporation of bio-fillers may have altered the configuration of PBAT polymer chains inside the crystal

lattice, resulting in reduced crystallinity of the composite, thus diminishing tensile strength.

With the steady rise in bio-fillers concentration content of the bio-composite, both tensile strength and elongation at break demonstrate a decreasing trend, as depicted in Figures 4.1 and 4.2. The elongation at break of the bio-composite decreased as the concentration of bio-fillers within the PBAT bio-composites increased. The highest elongation at break (%) recorded is SW10 (140.20%) and the lowest recorded is KC30 (4.31%). This phenomenon can be ascribing to the existence of embrittlement effect that cause by the presence of bio-fillers within the PBAT matrix in which resulting to restrict the chain mobility of the PBAT bio-composites. A similar findings found by Mäder et al. (2025) in which resulting that the bio-composites samples exhibits the same embrittlement effect as the concentration content of bio-fillers utilised increased within the bio-composites sample while utilising PBS/PBAT reinforced with spent coffee ground (SCG). The specific surface area of the particles infill will affect the number of molecular chains that can be adsorbed, thereby enhancing the material's tendency to agglomerate. The diminished strength was due to the lack of shear yielding during the mixing and stretching process. The findings demonstrate that the incorporation of bio-fillers into the blends leads to its dispersion with PBAT, hence enhancing steric hindrance. This constrains the tensile properties of PBAT (Dammak et al., 2020a; Nunes et al., 2020).

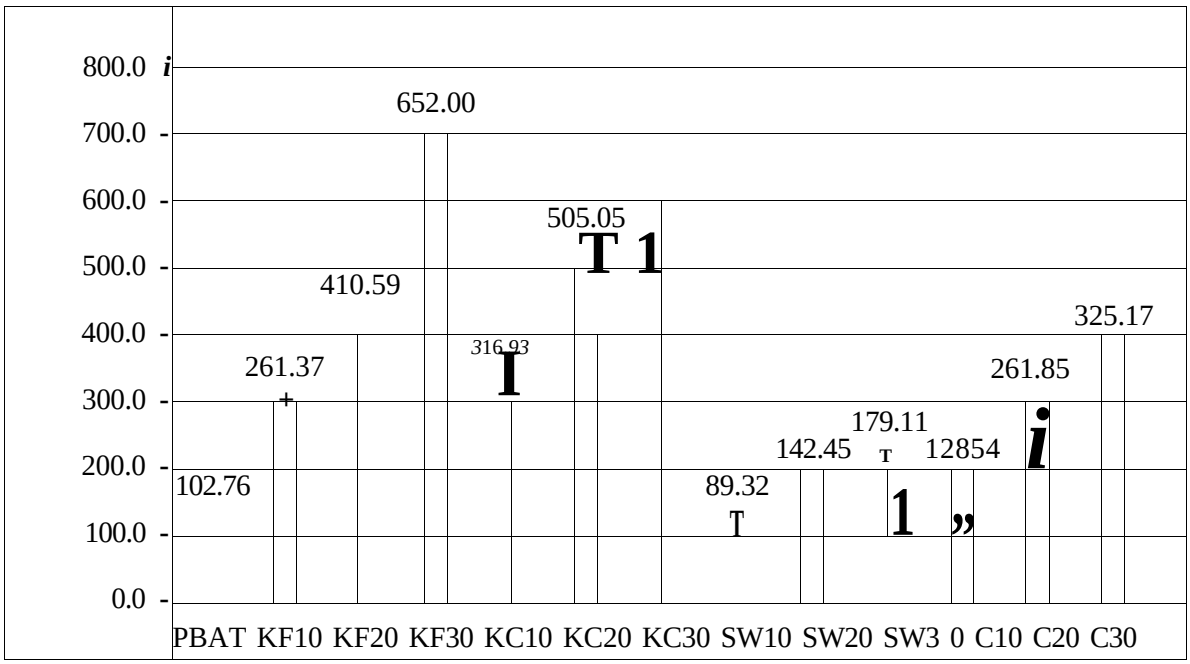

Figure 4. 3 Tensile Modulus of Bio-composite

The bio-composites flexural modulus increases proportionally as the content of bio-fillers, as anticipated for bio-filler reinforced materials. Ahankari et al. (2011) have observed same findings for PHBV-based blends using various forms of biomass, ranging from 30 to 40 wt.%. The elevating of increase in flexural modulus can mostly be attributed to mechanical interlocking among bio-fillers.

The incorporation of kenaf bio-filler significantly enhanced the tensile moduli, which increased proportionally with the content of kenaf bio-filler within the bio-composite when compared to seaweed and chitosan. A 250% increase in modulus rates was observed with the integration of 10 wt.% KF and KC, respectively, while preserving sufficient toughness, as indicated by the notably high elongation at break values. The highest tensile modulus recorded is KF30 (652.00 MPa) while the lowest modulus recorded is SW10 (89.32MPa). The carboxyl groups on the surface of the functionalized kenaf fibres interact with the hydroxyl groups of the PBAT matrix, leading to improved stiffness, as supported by data from other research groups employing PBAT as the matrix (Botta, Titone, et al., 2021; da Costa et al., 2023a; Dammak et al., 2020b). The observed increase in tensile moduli may result from the transfer of certain stresses to the dispersed phase. Furthermore, the stress concentration zones around bio-fillers increase as their proximity intensifies at elevated concentrations.

4.2 Dynamic Mechanical Analysis (DMA)

Dynamic Mechanical Analysis (DMA) serves as a method to investigate the correlation among time, temperature, and frequency in relation to the viscoelastic properties of polymers. The impact of temperature on the load-bearing capacity of specimens was assessed, as demonstrated in Figure 4.4 (PBAT/KF), Figure 4.5 (PBAT/KC), Figure 4.6 (PBAT/SW), and Figure 4.7 (PBAT/C). All plots present a comparison of the curves for the blends against the pristine PBAT polymer, detailing the storage modulus, loss modulus, and tan 8.

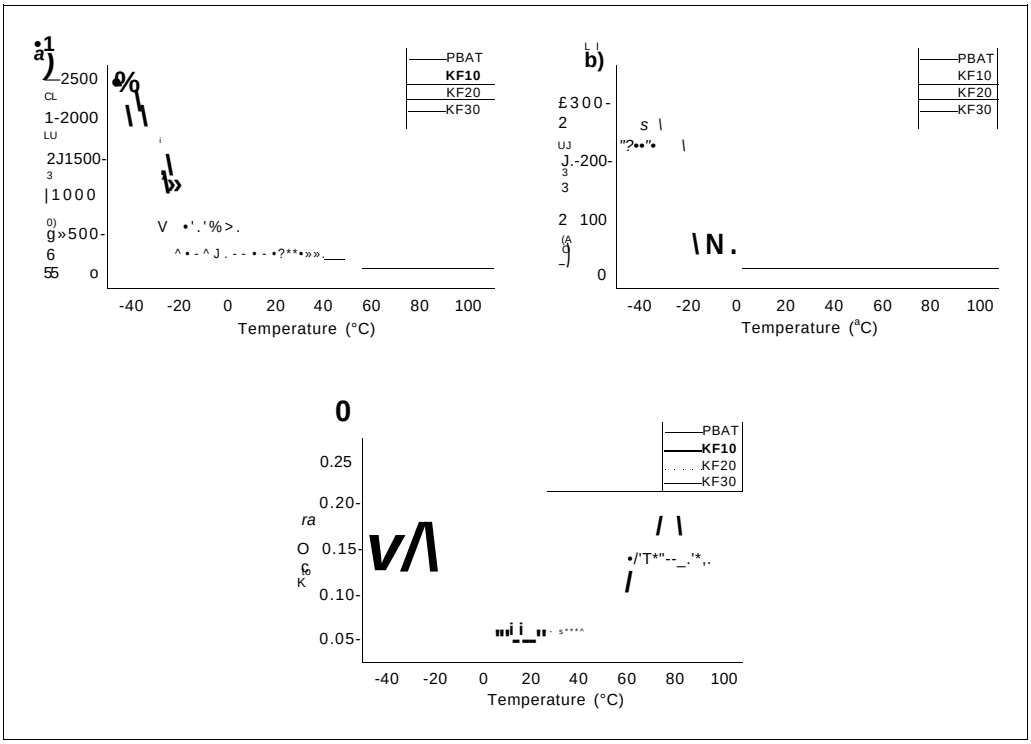

Figure 4. 4 DMA of PBAT/KF Bio-composite: (a) Storage Modulus (E') (b) Loss Modulus (E") and (c) Damping Factor (Tans)

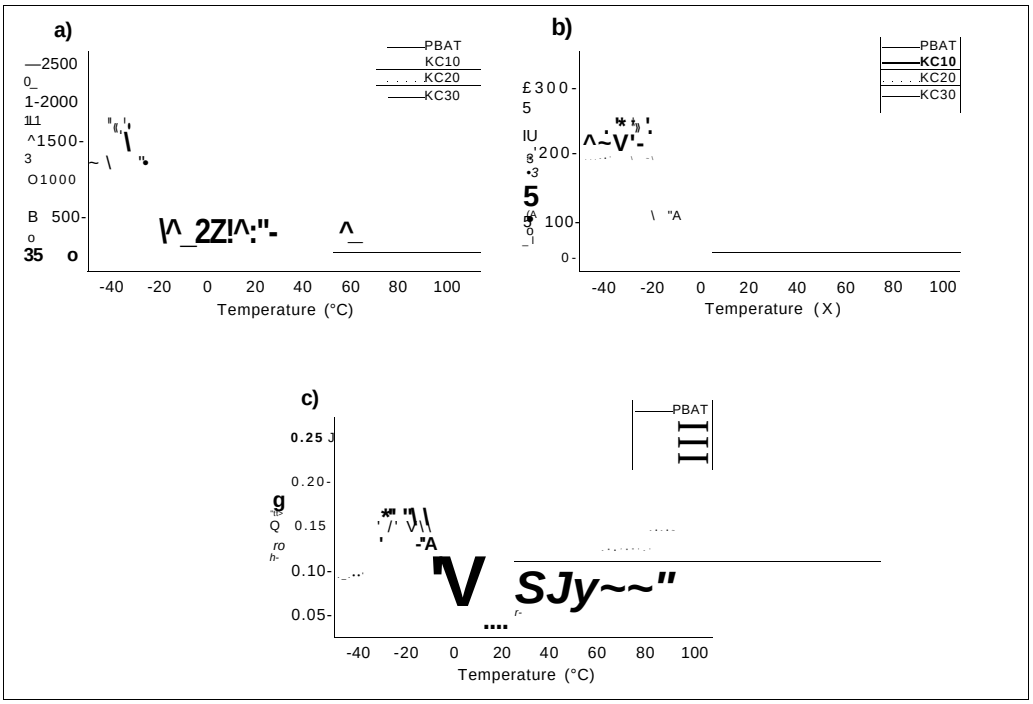

Figure 4. 5 DMA of PBAT/KC Bio-composite: (a) Storage Modulus (E') (b) Loss Modulus (E") and (c) Damping Factor (Tans)

The incorporation of KF and KC fibre resulted a notable enhancement of the storage modulus across the evaluated temperature range. At 20 °C, the pure PBAT exhibited a storage modulus of 225 MPa. Bio-composites containing 30 wt.% KF (KF30) demonstrated a modulus of 620 MPa, while those with 30 wt.% KC (KC30) showed a modulus of 510 MPa. The glass transition temperature, indicated by the peak of the loss modulus, exhibited a slight increase with the addition of KF and KC. This indicates a reduction in the polymer chains mobility due to the presence of the fibres. The Tans value, which indicates the glass transition in polymeric bio-composites, is measured between -30 °C and -20 °C. The height of the Tans peak diminished with increasing amounts of KF and KC, consistent with the reduced proportion of PBAT in the bio-composite.

The modulus in the glassy region was significantly greater for the bio-composites compared to neat PBAT, especially for those with 30 wt.% of KF and KC. The existence of the fibres significantly influenced the modulus in the rubbery region, with this effect becoming more pronounced as the amounts of KF and KC increased. The presence of fibres leads to an increased modulus. The increase in modulus with fibres inclusion corresponds with the results of tensile testing, indicating the reinforcing

effect of the fibres. Recent studies demonstrated similar performance when incorporating bamboo powder in PBAT compositions (Ye et al., 2024a) and when blending rice straw microparticles with the PLA/PBAT matrix (Jubinville et al., 2024a). The manifestation of KC observed as a minor reduction in the storage modulus, loss modulus, and Tans values in comparison to KF. This observation may be associated with the chemical composition of KC compared to KF since the chemical composition of bio-fillers influenced the properties of bio-composites. A study by Ahmetli et al. (2025) indicates that the chemical composition of hazelnut waste shell (cellulose and lignin) able to alter the properties of bio-composite.

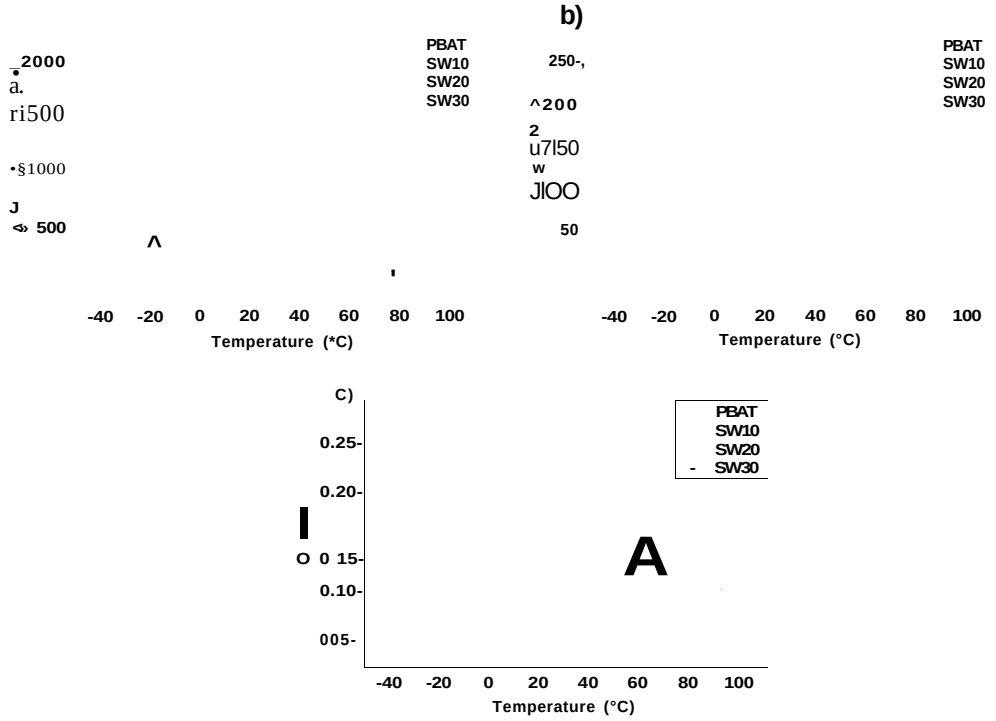

Figure 4. 6 DMA of PBAT/SW Bio-composite: (a) Storage Modulus (E') (b) Loss Modulus (E") and (c) Damping Factor (Tans)

Figure 4.6 (a) depicts the correlation between the storage modulus E' and temperature for neat PBAT and PBAT/SW bio-composites, assessed at a frequency of 1 Hz. The observed initial temperature range is -40 to -30 °C, indicating the matrix's stiffness under extremely flat conditions (glassy state). The sample with 20wt% SW demonstrates a higher E' value of 1937 MPa, in contrast to the 10wt% sample at 1690

MPa and the 30wt% sample at 1156 MPa during the initial phase. Many polymers exhibit a distinct plateau below the glass transition temperature (T_g) (Ferreira et al., 2019; Ratshoshi et al., 2024a). The presence of SW on this plateau partially obscures the reinforcing effect. The filler effect is consistently observed in the PBAT matrix, irrespective of the type of filler or processing methods employed (da Costa et al., 2023b; Olonisakin et al., 2024a).

At elevated temperatures, both PBAT and SW exhibit a decrease in stiffness, resulting in a decline in the storage modulus. Bio-composites reinforced with 10wt%, 20wt%, and pure PBAT exhibit a more pronounced decrease in E values with increasing temperature compared to those containing 30 wt% SW. The second and third segments of the curves, occurring at temperatures between -20 and 10°C, are associated with polymer relaxation. This leads to a significant reduction in the material's storage modulus. The degradation of the polymeric matrix in both areas lead to an enhanced reinforcing effect of the SW. The storage modulus of bio-composites with 20% by weight is approximately 28.7 MPa at a temperature of about 100°C. The figure significantly exceeds the storage moduli of the other bio-composites. The inclusion of SW has a minimal effect on the E' value of the bio-composites in the plastic region. This trend demonstrates that a concentration of 20 wt% enhances the compatibility of the PBAT polymer utilized as the matrix.

Figure 4.6 (b) depicts the variations in E" for PBAT and PBAT/SW bio-composites as a function of temperature. The data indicates that the loss modulus of all materials increases in the plastic zone (-40 to -30 °C) and subsequently decreases in the elastic region (-20 to 10 °C) with rising temperature. Observations indicate that the presence of SW influences the value of E", both below and above the glass transition temperatures, which are the temperatures at which E" reaches its maximum values. The loss modulus of PBAT is lower than that of the reinforced composites in both the plastic and rubbery regions. The incorporation of SW enhances the loss modulus of composites at elevated temperatures.

The damping curves of the PBAT/SW in Figure 4.6 (c) showed in a reduction in the height of the Tans peak. The lack of constraints on chain movement within the pure PBAT matrix accounts for this phenomenon. The incorporation of SW particles limits the mobility of the chains, resulting in a reduction in both the sharpness and height

of the Tans peak. The obtained values are consistent with those documented in the literature (Kargarzadeh et al., 2020a; Olonisakin et al., 2024b; Ratshoshi et al., 2024b) and indicate a correlation with the SW content. The presence of two thermal transitions is linked to the aliphatic and aromatic constituents of PBAT, respectively.

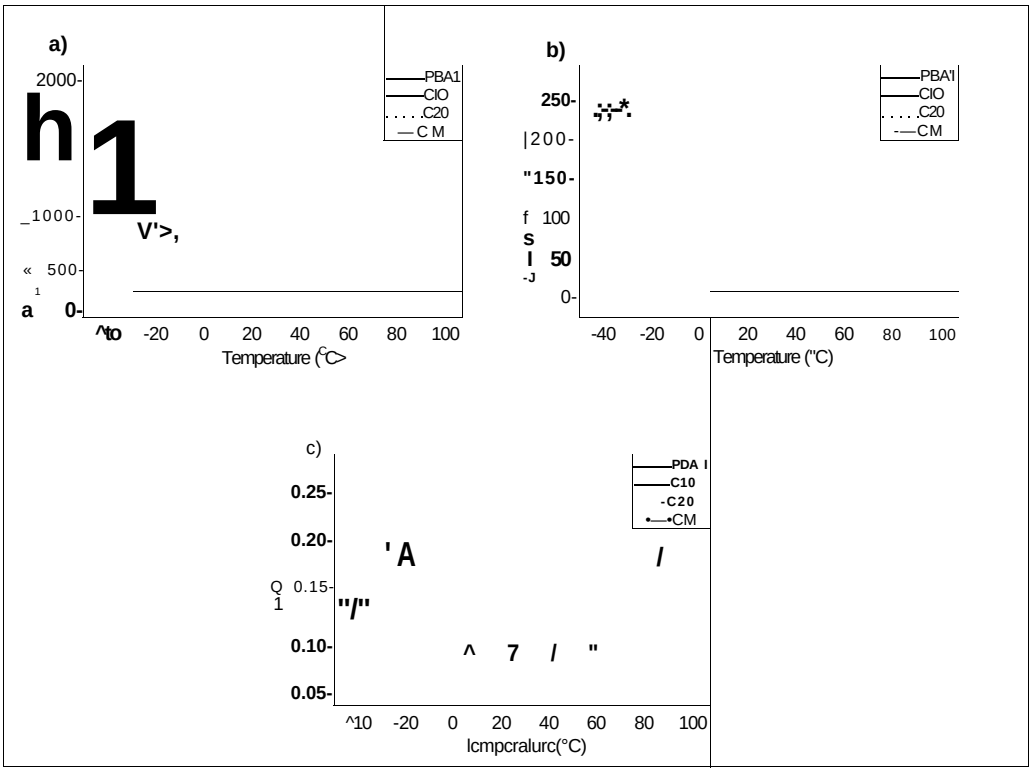

Figure 4. 7 DMA of PBAT/C Bio-composite: (a) Storage Modulus (E') (b) Loss Modulus (E'') and (c) Damping Factor (Tans)

Figure 4.7 (a) demonstrates the correlation between the storage modulus E' and temperature for neat PBAT and PBAT/C bio-composites, assessed at a frequency of 1 Hz. The observed initial temperature is -40 to -30 °C, which indicates that the PBAT/C is in a glassy state. The incorporation of chitosan content within the PBAT matrix resulted in an enhancement of modulus storage of PBAT/C in which signifying the higher concentration of C content within the PBAT matrix, the higher the storage modulus. It has been proven that C30 has the highest storage modulus, E compared to neat PBAT. Meanwhile, Figure 4.7 (b) indicates that the peak of loss modulus of PBAT/C bio-composites slightly increases as the C content concentration increases within the PBAT matrix. The observed initial temperature is -40 to -30 °C. It can be

related to the tensile modulus of PBAT/C bio-composite that increases as the C content increases within the matrix which leads the PBAT/C to become stiffer compared to neat PBAT. It leads to reduction and restriction of polymer chain in matrix. The presence of the C content influenced the modulus in the rubbery region, with this effect becoming more distinct as the amounts of C increased. The damping curves of PBAT/C in Figure 4.7 (c) demonstrated that the presence of chitosan content within the PBAT matrix constrain the chain interaction of PBAT and chitosan. The C content limited and hindered the chain mobility of the PBAT/C bio-composites.

All bio-composite samples exhibit a notable alteration when the bio-fillers were reinforced with the PBAT matrix, while a notable result recorded in which resulting increase in modulus storage, E' and loss modulus, E''. The highest peak of modulus storage, E' recorded is KF30 while the lowest peak of storage modulus, E' recorded is SW30. The highest peak of loss modulus recorded, E'' recorded is KC30 while the lowest peak of loss modulus, E'' recorded is SW30. This is due to the response of chemical composition that influenced the behaviour of bio-fillers when reinforced with PBAT.

4.3 Thermal Analysis

4.3.1 Thermogravimetric Analysis (TGA)

TGA curve yielded substantial insights into the material's behaviour under elevated temperatures. Figures 4.8 and 4.9 present the TGA and DTG data at varying heating rates for PBAT/KF and PBAT/KC, respectively. Investigations into thermal stability were conducted to determine the effects of KF and KC on the PBAT matrix. The onset degradation temperature (Ton) for the bio-composite with both fillers, KF and KC, ranged from approximately 250°C to 280°C, significantly lower than the Ton of the neat PBAT, which was recorded at around 370°C.

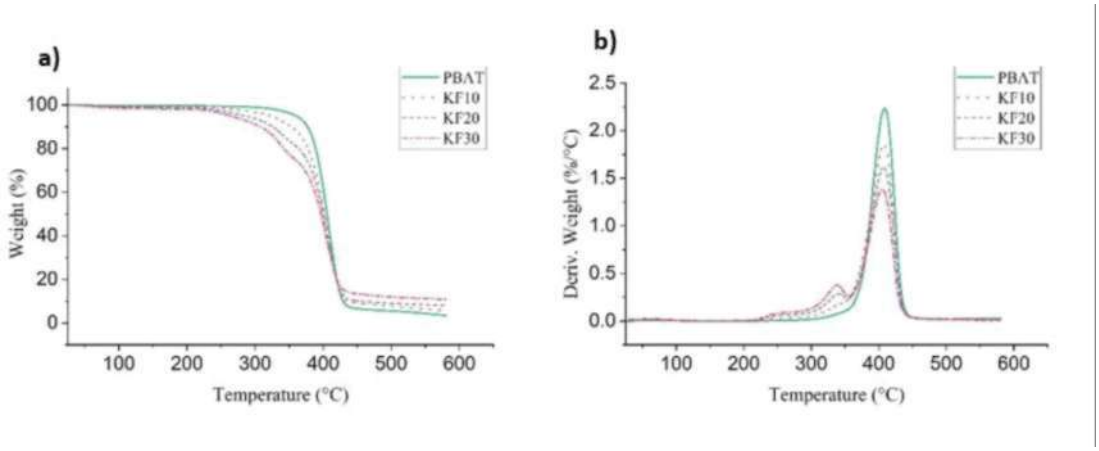

Figure 4. 8 Thermogravimetric Curves of PBAT/KF Bio-composite: (a) TGA and (b) DTG

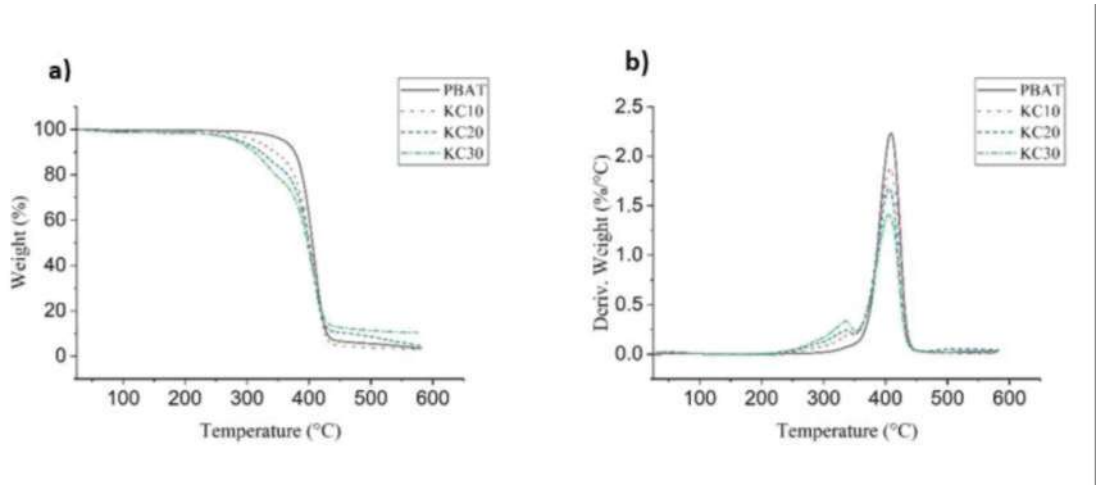

Figure 4. 9 Thermogravimetric Curves of PBAT/KC Bio-composite: (a) TGA and (b) DTG

The thermal decomposition of PBAT/KF and PBAT/KC bio-composite occurs in three stages: from 50°C to approximately 230°C, the sample mass gradually decreases due to dehydration and the breakdown of holocellulose, lignin, and small fragments; from 300°C to around 400°C, the mass decreases rapidly as cellulose and PBAT decompose; above 400°C to nearly 600°C, the decomposition of additional substances in the bio-composite material occurs. The char formation noted at the final stage of bio-composites degradation remained consistent across the varying loading levels of KF and KC, as outlined in Table 9.

Table 9

TGA and DSC Data of PBAT Bio-composites

Samples	T at 5 %	T at 25 %	T at 50 %	T at 75 %	Residue at 600 °C (%)
PBAT	**360.43**	**392.61**	**405.67**	**417.16**	**3.46**
KF10	**322.72**	**383.75**	**401.86**	**415.67**	**5.71**
KF20	**285.04**	**376.04**	**400.05**	**415.77**	**8.08**
KF30	**264.13**	**359.21**	**396.62**	**415.18**	**10.83**
KC 10	**315.08**	**382.49**	**401.01**	**414.56**	**3.06**
KC 20	**288.95**	**376.52**	**399.52**	**414.76**	**4.56**
KC 30	**281.64**	**364.93**	**397.25**	**415.40**	**10.24**
SW 10	**182.38**	**376.78**	**404.30**	**463.05**	**19.91**
SW 20	**180.77**	**374.54**	**403.72**	**434.12**	**19.99**
SW 30	**186.96**	**378.54**	**403.90**	**430.97**	**16.91**
C10	**271.06**	**360.19**	**396.76**	**414.58**	**7.97**
C20	**281.37**	**364.00**	**396.02**	**412.51**	**1.14**
C30	**283.53**	**354.86**	**398.67**	**415.50**	**7.81**

Note: Temperatures are given based on total weight lost at 5%,25%,50% and 75%

The TGA/DTG graph (Figures 4.8 and 4.9) indicates that the degradation temperature for all PBAT/KF and PBAT/KC bio-composites ranges from 300°C to 400°C. The degradation temperature of bio-composites increases as the content of KF and KC fibres rises from 10 wt.% to 30 wt.%, leading to a higher main degradation peak temperature and a substantial decrease in the rate of degradation of the produced bio-composites. Thermal stability is enhanced with 30 wt.% KF and KC fibres loadings in comparison to 10 wt.% and 20 wt.% loadings. At the total 75 % of weight loss (%) of T 75%, KF samples exhibits slight decrease which is KF10 (415.77°C), KF20 (415.77°C) and KF30 (415.18°C) which higher compared to the KC bio-composites sample, KC10 (414.56°C), KC20 (414.76°C), and KC30 (415.40°C). The thermal decomposition temperature of KF and KC bio-composites decreased compared to neat PBAT, due to the inclusion of kenaf fibres, which demonstrates low thermal stability (Marques et al., 2015b; Sudha et al., 2024).

The incorporation of both KF and KC significantly decreases the thermal stability of the PBAT matrix. The observed phenomenon can be attributed to the

interfacial interactions within the PBAT, influenced by the addition of KF and KC fibres. This results in a structure that shows reduced robustness but can withstand elevated temperatures prior degradation occurs. The integration of KF and KC fibres creates a boundary within the PBAT matrix, hindering the transfer of heat and mass during thermal degradation. The boundary effect hinders the degradation process, thereby reducing the thermal stability of PBAT. Comparable assertions have been presented concerning the incorporation of jute fibre as reinforcement in PLA/PBAT blends (Tang et al., 2024) and coir fibre reinforced TPS/PBAT composites (Kargarzadeh et al., 2020b).

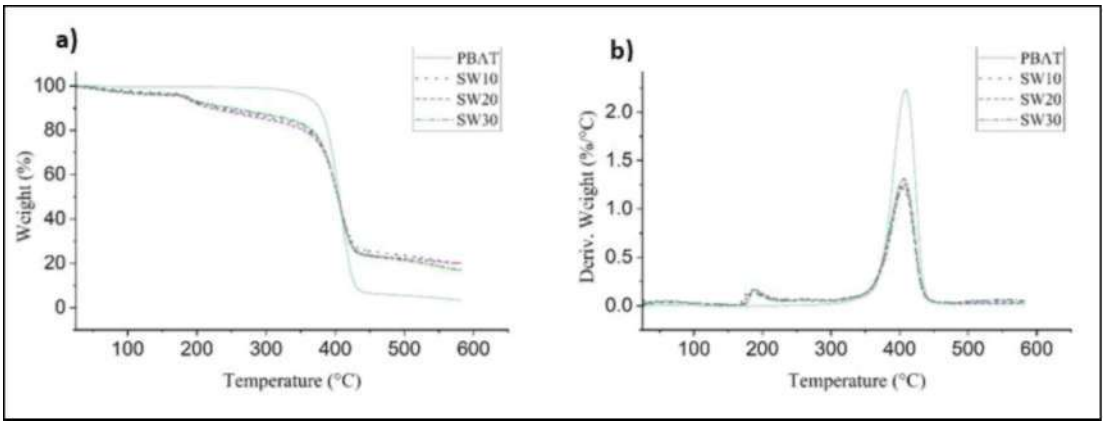

Figure 4. 10 Thermogravimetric Curves of PBAT/SW Bio-composite: (a) TGA and (b) DTG

Table 9 and Figure 4.10 present the effects of SW incorporation on the thermal stability of PBAT. Table 9 presents the temperatures associated with deterioration resulting in weight losses of 5%, 25%, 50%, and 75%. The TGA curves for pure PBAT, illustrated in Figure 4.10 (a), indicate a singular primary weight loss stage of the PBAT matrix occurring at a temperature of 383°C. The PBAT/SW composites demonstrate two separate degradation phases resulting from the breakdown of SW particles. With increasing SW concentration, the decomposition temperature of the composites fluctuates toward that of the SW. To enable precise observation, various temperature ranges of deterioration are delineated in the DTG plot, as illustrated in Figure 4.10 (b). The early phases of weight loss transpired within a temperature range of 100 to 380 °C, primarily linked to the presence of SW, were likely due to the degradation of galactose

units. The sample shows a gradual weight decrease, attributed to the further fragmentation of the composition products from the second phase. At the 75 % of total weight loss (%) of T 75%, SW bio-composites exhibit slight increases compared to the neat PBAT at (417.16 °C) in which at SW10 (463.05 °C), SW20 (434.12 °C), and SW30 (430.97 °C). The ash concentration of the residual material at 600 °C is approximately 19% by weight. The final weight loss observed between 350 and 450 °C is directly associated with the thermal degradation of PBAT. The intermediate degradation temperature readings suggest a consistent interaction between the SW and PBAT matrix. PBAT demonstrates superior thermal stability. The thermal stability of PBAT/SW composites diminishes with an increase in SW content, primarily due to the low thermal degradation of the SW. A similar finding found by (Hamdan et al., 2025) where the thermal stability of PBAT/SW bio-composites is decreases compared to the neat PBAT due to the low thermal stability of SW content within the PBAT matrix. The thermal stability of the PBAT/SW bio-composite exhibits the following order: SW30 > SW10 > SW20. An increase in SW concentration correlates with a rise in the residue char of the composites.

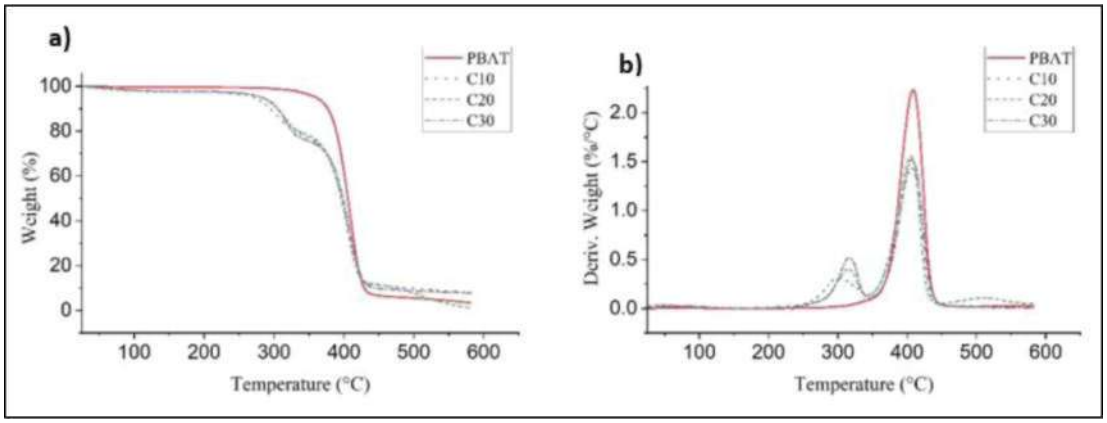

Figure 4. 11 Thermogravimetric Curves of PBAT/C Bio-composite: (a) TGA and (b) DTG

Table 9 and Figure 4.11 present the influences of chitosan integration on the thermal stability of PBAT. The TGA curve for neat PBAT, illustrated in Figure 4.11 (a), indicates a singular primary weight loss stage of the PBAT matrix occurring at a temperature of 360 °C. The TGA curves of PBAT/C also indicated there were two

separate degradations occurred, included the degradation of chitosan. The early weight loss occurs at 100 °C to 360 °C, in which the degradation of chitosan particles (alkaline polysaccharide) within the PBAT matrix. Therefore, the TGA curves indicate that the decomposition of PBAT/C fluctuated as the C content concentration increases. At the 75 % of total weight loss (%) of T 75%, C bio-composites exhibit slight decreases compared to the neat PBAT at (417.16 °C) in which C10 (414.58 °C), C20 (412.51 °C), and C30 (415.50 °C). The ash concentration of the residual material at 600 °C is approximately 7% by weight. Therefore, the least residue produced was from C20. The final weight loss observed between 350 and 450 °C is directly associated with the thermal degradation of PBAT.

The incorporation of C decreases the thermal stability of the PBAT matrix. The observed phenomenon can be attributed to the interfacial interactions within the PBAT, influenced by the addition of C particles within the PBAT matrix. A similar finding found by Salazar et al. (2022) in which the utilization of chitosan in polylactide-polthylene glycol (PL-PG/C) bio-composites in which resulting decreases of thermal stability. It indicates that the thermal stability of neat PBAT is superior compared to the PBAT/C bio-composites. It is due to the reduction of heat transfer that has been created by the presence of chitosan particles within the PBAT matrix. The presence of C content within the matrix hinders the degradation process in which it reduces the thermal stability of PBAT matrix. The thermal stability of PBAT/C diminished as the chitosan content increases.

Therefore, all PBAT bio-composites sample exhibits an alteration in thermal stability. The thermal stability of PBAT bio-composites influenced by the chemical composition of each bio-fillers utilized. The following order of PBAT bio-composite stability is PBAT/KF > PBAT/KC > PBAT/C > PBAT/SW.

4.3.2 Differential Scanning Calorimeter (DSC)

Figures 4.12 to 4.15 present the differential scanning calorimeter (DSC) thermograms for the PBAT/KF, PBAT/KC, PBAT/SW, and PBAT/C bio-composites. A peak is observed above the glass transition temperature for all bio-composite blends. This phenomenon is referred to as enthalpic relaxation or thermal history. The enthalpic

relaxation refers to a phenomenon characterized by a minor peak occurring above the glass transition temperature, which arises when a heated material is gradually cooled through Tg or maintained below Tg for a duration (Bevis et al., 2008). The degradation temperature of biocomposites increases when the bio-filler content rises from 10 wt.% to 30 wt.%, resulting in a higher primary degradation peak temperature and a marked reduction in the degradation rate of the resulting biocomposites. Thermal stability is improved with 30 wt.% bio-filler loadings compared to 10 wt.% and 20 wt.% loadings. The thermal degradation temperature of biocomposites was diminished relative to unmodified PBAT due to the incorporation of bio-filler, which exhibits low thermal stability.

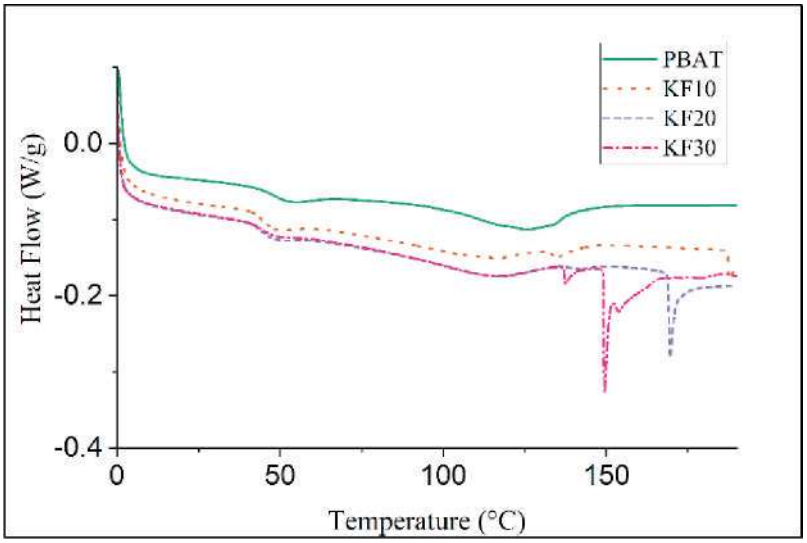

Figure 4. 12 DSC Heating Curves of PBAT/KF Bio-composite

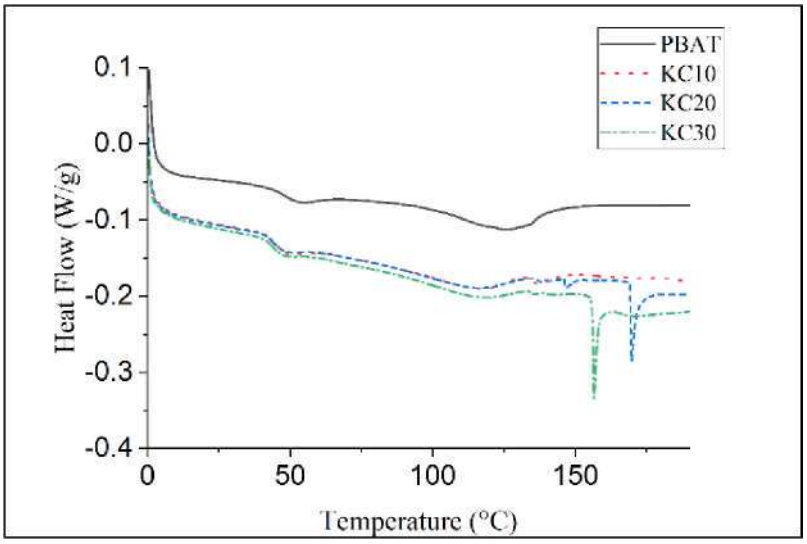

Figure 4. 13 DSC Heating Curves of PBAT/KC Bio-composite

The incorporation of KF and KC into PBAT influences thermal and kinetic properties, indicating that the melting temperatures of the bio-composites were largely unaffected, whereas the degree of crystallinity decreased, leading to the absence of PBAT endotherms in the DSC scans. Kargarzadeh et al. (Kargarzadeh et al., 2020c) noted a comparable situation when kraft lignin particles were integrated into a PBAT.

The current scenario indicates that the reduction of PBAT crystallinity is likely responsible for the improvement of the mechanical response, particularly at low temperatures. The melting rate of the bio-composites increased as the integration of PBAT with KC and KF compared to the neat PBAT (see Table 9). The melting rates of PBAT/KF and PBAT/KC bio-composites, as illustrated in Figures 4.12 and 4.13, indicate that all compositions display comparable behaviour. KF and KC accelerated the melting rate of the biocomposites compared to the unmodified PBAT (see Table 9). The melting rates of PBAT/KF and PBAT/KC biocomposites, illustrated in Figure 6, indicate that all compositions display comparable behaviour. Nevertheless, the KF30 and KC30 biocomposites demonstrated a σ_{max} that surpassed that of other biocomposites.

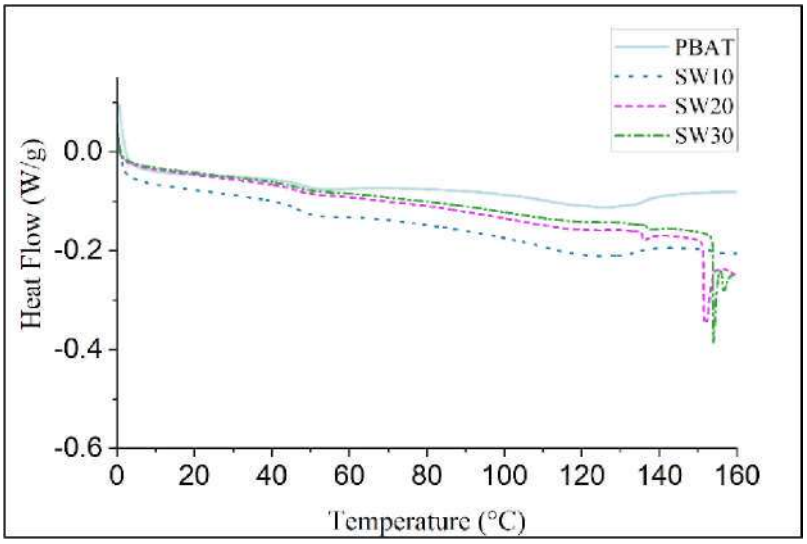

Figure 4. 14 DSC Heating Curves of PBAT/SW Bio-composite

Figure 4.14 presents the DSC thermograms of the PBAT/SW bio-composite, illustrating a singular fusion endotherm that signifies the fusion of the PBAT matrix. This peak exhibits a lower elevation and a broader profile relative to the other thermograms. The melting temperature shows a slight increase with rising SW concentration. This phenomenon may result from intermolecular interactions between SW particles and PBAT chains, leading to the formation of thinner lamellar crystals. The anticipated interaction involves hydrogen bonding between the carbonyl units of PBAT and the sulfate, hydroxyl and glycosidic groups of SW. Interactions occur in the amorphous state, exhibiting a diminished effect on the degree of crystallization relative to PBAT domains. Figure 4.14 illustrates that the addition of SW results in an elevated crystallization temperature (Tc) of the composites in comparison to plain PBAT. This behavior suggests that SW acts as a nucleation agent, facilitating the crystallization of PBAT. The nucleation outcome becomes more pronounced with increasing SW quantities, as a larger amount of SW leads to an increased number of heterogeneous nuclei for crystallization.

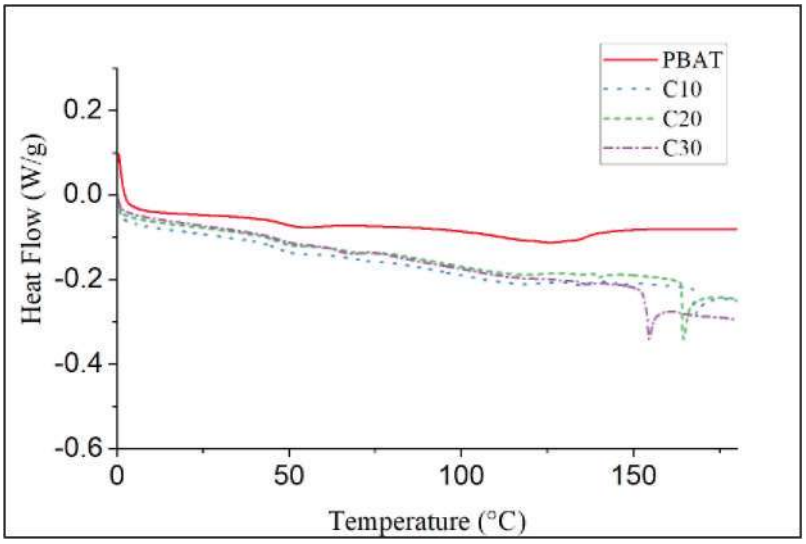

Figure 4. 15 DSC Heating Curves of PBAT/C Bio-composite

Figure 4.15 presents the DSC thermograms illustrating the cooling and heating curves of PBAT/C at concentrations of 10 wt.%, 20 wt.%, and 30 wt.%, respectively. Each composite exhibited a singular broad peak indicating of the melting temperature of the blends, reflecting enhanced dispersion among the blend components. Compared to pure PBAT, the melting peaks of the bio-composites containing chitosan exhibited slight decreases in the melting curves. The kinetics and extent of PBAT crystallisation during cooling may be slightly hindered, potentially leading to the formation of predominantly imperfect crystals that melt at lower temperatures compared to PBAT crystals. Furthermore, the inclusion of rigid aromatic structures restricted chain mobility. The observed phenomena were likely due to an increase in polymer viscosity following the addition of chitosan, which diminished the mobility of macromolecular chains at the crystal growth front; reminiscent observations were noted by Yu et al. (Yu et al., 2025), in which the integration of wood flour within PBAT matrix increase the crystallization rate of the bio-composites. The integration of chitosan in which has a composition of acetyl group does increase the melting temperature of PBAT/C composites and its enthalpy of fusional, resulted the PBAT/C require more energy to melt the PBAT/C bio-composites compared to neat PBAT. Therefore, further application of heat is required to facilitate the melting of the PBAT/Chitosan composite. Thus, their solid state is preserved at excessive temperatures, which may be noteworthy

for their mechanical properties, particularly in the design of items meant to endure high temperatures.

4.4 Fourier Transform Infrared (FTIR) Spectroscopy

Infrared spectroscopy was applied to determine the principal functional groups of PBAT and PBAT bio-composites, as well as determine the probable interactions occurring between them during the melt mixing process. The most notable spectra are presented in the spectra within the wavenumber range of 700 to 4000 cm^{-1}, highlighting the presence of the principal diagnostic peaks. The FTIR spectra of PBAT and their blends are shown in Figure 4.16, Figure 4.17, Figure 4.18 and Figure 4.19.

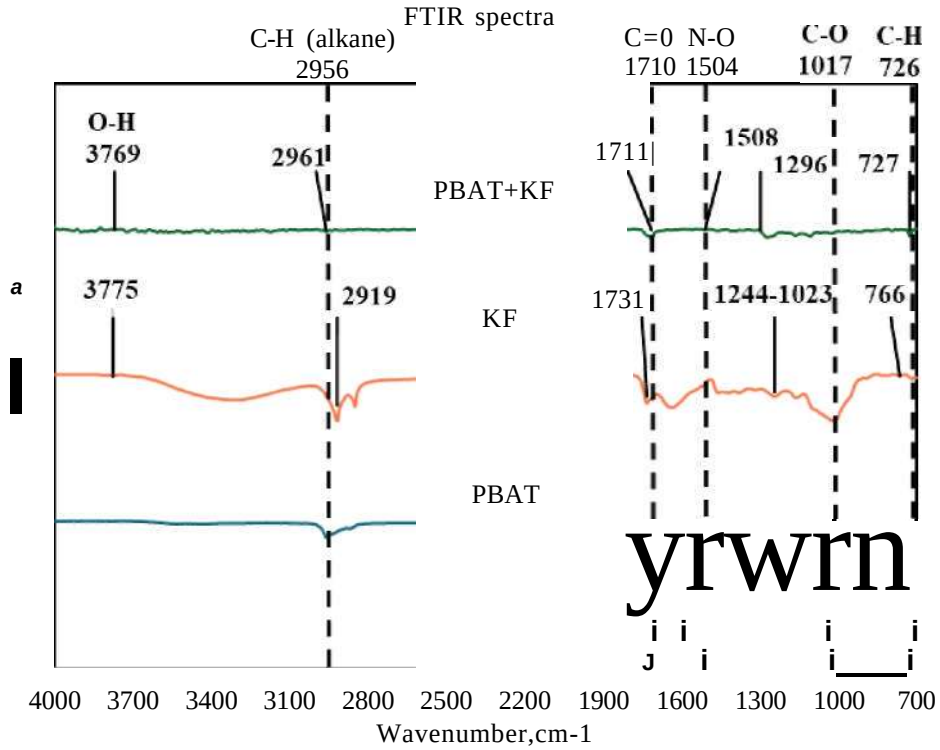

Figure 4. 16 FTIR Spectra Wavenumber (cm^{-1}) Graph of PBAT + KF

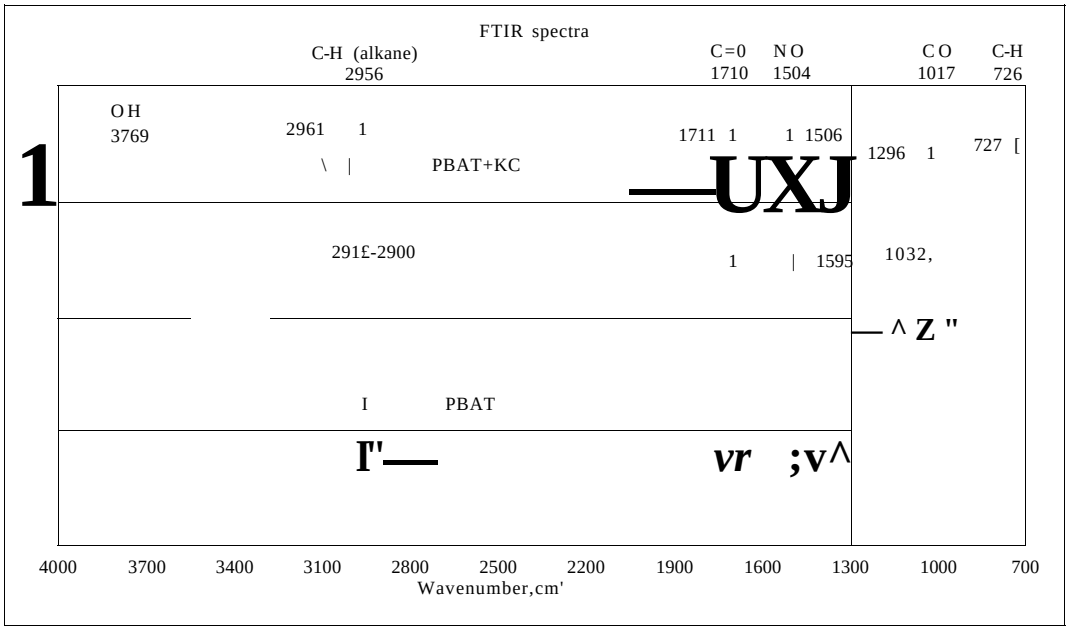

Figure 4. 17 FTIR Spectra Wavenumber (cm^{-1}) Graph of PBAT + KC

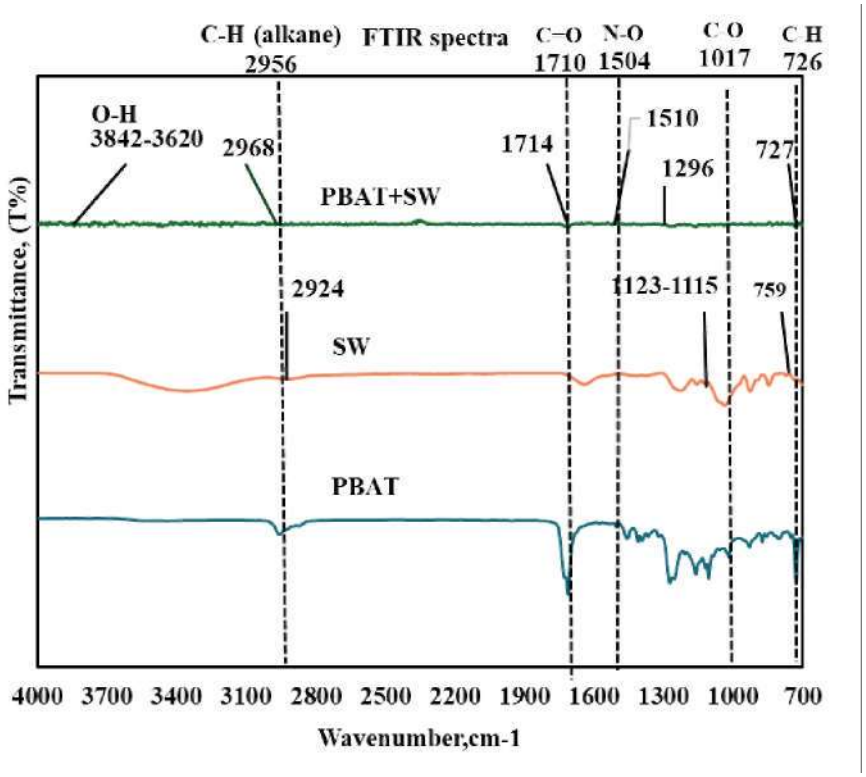

Figure 4. 18 FTIR Spectra Wavenumber (cm^{-1}) Graph of PBAT + SW.

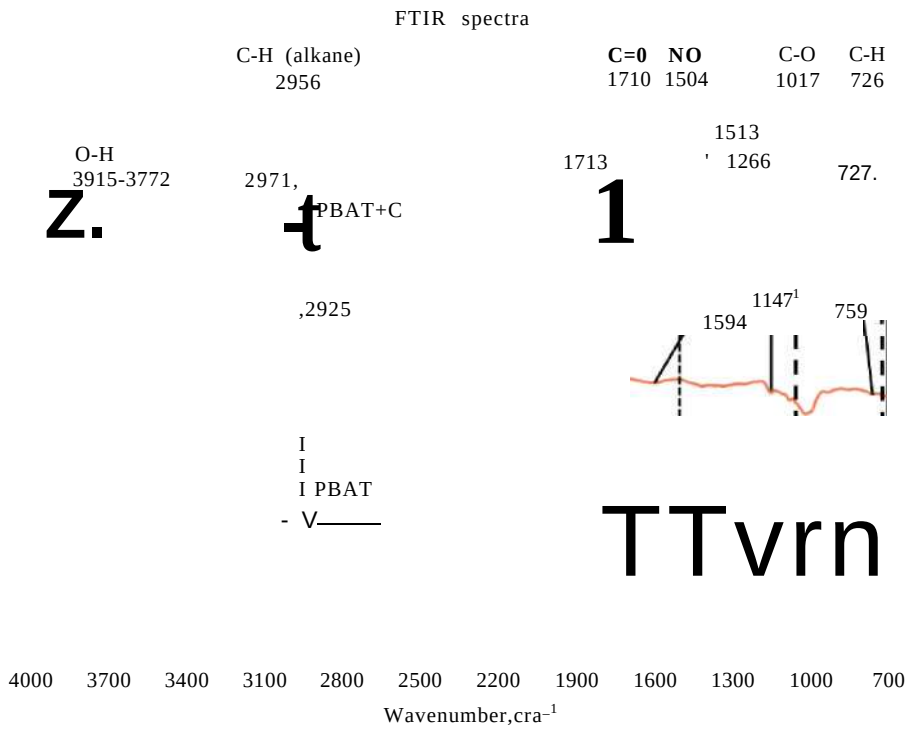

Figure 4. 19 FTIR Spectra Wavenumber (cm"[1]) Graph of PBAT + C

The infrared spectral data for PBAT (Figure 4.16 to Figure 4.19), particularly at 726 cm-1, signifies the bending vibration related to the CH-plane of the benzene ring (Conterosito et al., 2025; Li et al., 2024). The peak at 1017 cm"[1] is linked to the bending vibration of adjacent hydrogen atoms on the phenyl ring (Nomadolo et al., 2024a; Som & Harnkarnsujarit, 2024). The peak at 1409 cm"[1] is associated with the O-H bending vibration (da Rocha et al., 2024a; Pattaraudomchok et al., 2024). The peak at 1504 cm" [1] relates to the skeleton vibration of the benzene ring (Techawinyutham et al., 2024; Venkatesan et al., 2024). The peak at 1710 cm"[1] is indicative of the C=0 stretching vibration, while the peak at 2956 cm"[1] corresponds to the C-H asymmetric stretching vibration (da Rocha et al., 2024b; Nomadolo et al., 2024b).

The absorption spectrum of PBAT bio-composites indicated an upward shift of the CH-plane vibration of the benzene ring from 726 to 727 cm"[1] across all samples. The data for PBAT/KF showed an up-shift in the C-0 symmetric stretching vibration from 1017 cm"[1] to 1256 cm"[1]. ForPBAT/KC, the shift was observed at 1267 cm"[1], while PBAT/SW exhibited a shift at 1296 cm"[1], and PBAT/C at 1266 cm"[1]. The addition of KF, KC, S, and C resulted in a slight alteration of the band at 2956 cm^{-1}. The

incorporation of bio-fillers leads to a minor modification of the band at 2956 cm , whereas PBAT bio-composites displayed new peaks within the range of 3933 to 3412 cm^{-1}, which are ascribed to alcohol O-H stretching primarily ascribes to presences of chemical compositions (carbohydrates, cellulose, hemicellulose, and lignin) from bio-fillers due to the chemical reaction between hydroxyl group from bio-fillers and carbonyl group from PBAT. A similar finding found from Miescher et al. (2025) where a broad bands shows from 3600 to 3000 cm"[1] in which ascribe as O-H stretching while utilizing a potato peeled based PBS/PBAT bio-composites.

The evaluation of hydrogen bond energies for PBAT and its modified structure included the analysis of peaks between 1710 and 1713 cm^{-1}, which corresponded to free and bonded C=0 groups. A similar finding found by Botta, Teresi, et al. (2021) in which the presence of peaks ranging 1744 cnT[1] in which ascribe as the carbonyl, C=0 groups as the reinforcement of PBAT with bio-char. The hydrogen bond coefficients increased for all PBAT bio-composites, indicating a higher prevalence of hydrogen bonds between the blends and the PBAT C=0 groups. This enhancement supports the hypothesis that bio-fillers function as proton donors, while PBAT acts as a proton acceptor, indicating a notable electrostatic attraction between the components. The findings indicate the formation of a rigid hydrogen-bonded network in PBAT bio-composites, which is associated with an effective percolating network. The FTIR results align with the mechanical tests performed.

4.5 Scanning Electron Micrograph (SEM)

SEM images of the bio-composites were obtained to analyse the distribution of KF, KC, C, and SW within PBAT and to assess the morphological structure of the bio-composites. The image analysis of the bio-composites (Figure 4.20) reveals randomly dispersed bio-fillers, along with the presence of gaps between these components and the matrix. The random arrangement of the bio-fillers within the matrix, coupled with insufficient interaction, can lead to diminished mechanical properties. This aligned with the tensile strength results, indicating that the incorporation of bio-fillers diminished the tensile strength of the bio-composites. Previous studies indicate comparable

performance when utilizing bamboo powder in compositions with PBAT (Ye et al., 2024a) and blending rice straw microparticles with the PLA/PBAT matrix (Jubinville et al., 2024b).

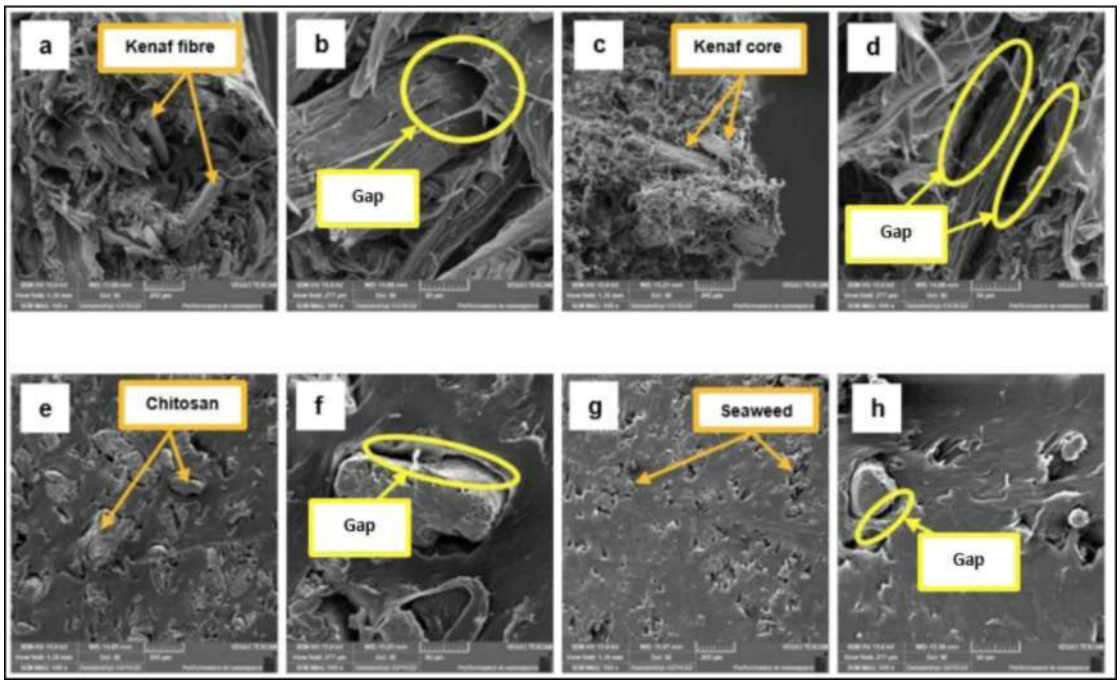

Figure 4. 20 SEM Micrographs of the Fracture Tensile Test Surface (a) (b) PBAT/KF (c) (d) PBAT/KC (e) (f) PBAT/C (g) (h) PBAT/SW

KF, KC, SW and C exhibited comparable tensile modulus, as shown in Figure 4.3. The PBAT/KC and PBAT/ KF bio-composite demonstrated slightly greater stiffness compared to PBAT/KC and PBAT/SW which, conversely, exhibited higher resistance and ductility. The stiffness and breaking properties of PBAT bio-composites are significantly influenced by the insufficient interaction which aligns with prior studies on lignocellulosic fibres and organic fillers (Hariprasad et al., 2020b; Kamarudin et al., 2022; Lang et al., 2022; Letwaba, 2024).

A comprehensive analysis of bio-composite morphology revealed that the densification of composites was quantitatively evaluated by observing the changes in porosity (Figure 4.20 a and c) and intraphase (Figure 4.20 e and g) following the hot press process. The results demonstrate that densification primarily occurs due to hot pressing, as materials from the extrusion and pelletizing stages exhibited lignocellulosic fibres and organic fillers that were nearly entirely coated with PBAT, which was

incorporated by polymer chains during compression molding. In addition, the types of filler primarily consist of elliptical and uniform spherical shapes that create a circular cross-section. The ability of PBAT coated the fillers, along with potential alterations in the fillers (e.g., aspect ratio) or in the PBAT matrix (e.g., crystallinity and crosslinking), may directly result from melt-processing. This phenomenon may be attributed to the chemical affinity between the aromatic domains of fillers and the matrix, specifically the terephthalate units of PBAT. These characteristics are essential for understanding the mechanical performance of bio-composites.

The aspect ratio is a critical parameter influencing the properties of a bio-composite material. The observed features can be elucidated by noting that tensile modulus generally increases with bio-fillers content, thereby enhancing the probability of rigidity occurrences. Collectively, these results demonstrate that under the specified operating conditions, KF, KC, SW and C exhibited uniform dispersion and built interfacial adhesion. However, they differed in actual aspect ratio and the ability to form intraphase, suggesting they may impart distinct mechanical behaviour to PBAT.

CHAPTER 5
CONCLUSION AND RECOMMENDATIONS

5.1 Conclusion

This study investigates the effects of kenaf fibre (KF), kenaf core (KC), seaweed (SW), and chitosan (C) on the chemical structure, crystallisation behaviour, thermal properties, microscopic morphology, and tensile properties of PBAT bio-composites. All evaluation and determination assessment has been done to bio-composite sample to assess the characterization of the PBAT bio-composites.

The incorporation of bio-fillers resulted in enhanced stiffness of PBAT with KF30 (652 MPa) as the highest of tensile strength, attributed to the significant interactions between the carbonyl groups of the PBAT matrix and the hydroxyl groups of the bio-fillers. Furthermore, the increase in the quantity of bio-fillers resulted in a non-homogeneous dispersion, consequently diminishing the tensile strength and elongation at break of the bio-composites.

The dynamic-mechanical analysis demonstrated notable increases in storage modulus (E') for all bio-composites with KFIO bio-composite as the highest peak of storage modulus (E') recorded, suggesting that the incorporation of bio-fillers resulted in substantial improvements in the tensile properties of the PBAT matrix. The peak value of the loss modulus (E'') increased with the addition of bio-fillers, while the glass transition temperature (T_g) exhibited a slight decreased. The decrease in T_g may result from the liberation effect of bio-fillers, which enhances the chain mobility of the amorphous regions in PBAT composites near the bio-filler particles.

The TG and DTG curves of the bio-composites indicated that the mass losses observed in all samples at temperatures below 200 °C can be attributed to the evaporation of adsorbed and bonded water. The variation in thermal behaviour of the bio-composites is likely attributable to the differing chemical compositions of the bio-fillers. Consequently, the varying mineral concentrations detected in the bio-fillers might originate from these sources. The thermal stability of PBAT bio-composites

influenced by the chemical composition of each bio-fillers utilized. The following order of PBAT bio-composite stability is PBAT/KF > PBAT/KC > PBAT/C > PBAT/SW.

The thermal behaviour of bio-composites containing bio-fillers was influenced by the incorporation of bio-fillers into the polymeric matrix. The capacity of the bio-filler to induce crystallisation in PBAT during thermal processing depended on the surface area and composition of the particles. The utilisation of bio-filler, distinguished by its minimal surface area and varied composition, influenced the crystallisation kinetics of PBAT. This suggests that the bio-filler functions as nucleating agents, affecting the morphology of PBAT at the interface between the bio-filler and the PBAT matrix.

The interaction between bio-fillers and the PBAT matrix was analysed using SEM. The images revealed the presence of bio-fillers, with no distinct gaps observed between the bio-fillers and the PBAT matrix, indicating effective contact between the two components. This interaction can help dissipate energy from external stresses through friction between particle-particle and particle-polymer at the fibre-matrix interface.

Finally, it is possible to say that KF, KC, SW and C demonstrated favourable thermo-mechanical and physical properties in the bio-composites examined in this study. Their enhanced thermo-mechanical properties across a broad temperature range positioned these bio-fillers as highly suitable for medical applications and various environmental industrial uses, such as packaging, without requiring any organic modifiers or compatibilizers for effective dispersion within PBAT. This effort aims to broaden the applicability of PBAT and offer a viable approach for utilising biodegradable materials as alternatives to conventional plastics.

5.2 Future Work Recommendations

This study demonstrated that the integration of KF, KC, SW and C into a PBAT matrix results in green composites with enhanced properties. The enhancement of interface compatibility among KF, KC, SW, C and PBAT is a critical issue for maximizing the potential of these bio-composites across various applications.

Therefore, additional potential enhancements could be made to these bio-composites, such as incorporating a plasticizer into the matrix to improve its ductility properties. This discovery holds significant potential to serve as a guide and reference for future studies aimed at addressing industrial demand and advancing research efforts. This discovery further illustrates the potential applications of these bio-composites, highlighting their role in improving the production of high-quality materials and contributing to advancements in sustainable chemistry.

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APPENDICES

APPENDIX A

Sample Fabrication Calculation

Melt blending using Rheomixer - 80 g for a single mixer

10wt%

$\frac{10}{100} \times 80 = 8g$

20wt%

$\frac{20}{100} \times 80 = 16g$

30%

$\frac{30}{100}$ **X 80 = 24#**

Composition **of bio-filler blended with PBAT matrix**

Sample code	PBAT wt%	10 wt%	20 wt%	30 wt%	Total weight blend
PBAT	80g	-	-	-	80g
KF10	72g	**8g**	-	-	80g
KF20	64g	-	16g	-	80g
KF30	56g	-	-	24g	80g
KC10	72g	**8g**	-	-	80g
KC20	64g	-	16g	-	80g
KC30	56g	-	-	24g	80g
SW10	72g	**8g**	-	-	80g
SW20	64g	-	16g	-	80g
SW30	56g	-	-	24g	80g
C10	72g	**8g**	-	-	80g
C20	64g	-	16g	-	80g
C30	56g	-	-	24g	80g

Hot pressing in a steel cast mould:

Target density = 1000kg/m^3

Mould size = 300 mm length, 300 mm width, 3mm thickness (Volume = 270 cm^3)

$$Density = \frac{Mass}{Volume}$$

Mass = 1 g/cm^3 x 270 cm^3

= 270g

AUTHOR'S PROFILE

Muhamad Haikal Bin Hamdan obtained his Diploma in Wood Industry in 2021 from Universiti Teknologi MARA, Pahang and Bachelor of Science in Furniture Technology (Hons.) in 2023 from Universiti Teknologi MARA, Pahang, MSc in Wood Science and Technology (2023 - now) from Universiti Teknologi MARA, Pahang. His Master thesis involves several analyses in determining the compatibility of several bio-filer reinforced with biodegradable polymer, Poly (Butylene Adipate-co-Terephthalate).

LIST OF PUBLICATION

Peer Reviewed

1. Hamdan, M.H., Sarmin, S. N., Abdul Jalil, A.M., Abdullah, Z.A. and Mohammad, N.A. (2024). Tensile and Morphological Properties of Biodegradable Composites Reinforced with Kenaf Core Fibres. *Science Letters,* 18(4), 7-11. **(Published - MyCite)**
2. Hamdan, M.H., S. N., Salim, N., and Jawaid, M. (2025). Evaluation on tensile and morphological properties of chitosan in PBAT bio-composite. *Jurnal Teknologi.* **(Published - Q3 IF 0.7)**
3. Hamdan, M.H., Sarmin, S. N., Jawaid, M., Ismail, A.S., Fouad, H. and Salim, N. (2025). Investigating the Influence of Varied Seaweed Filler Content on the Thermal and Tensile Properties of poly (butylene adipate-co-terephthalate) Bio-composite. In Materials Science Forum, Trans Tech Publications Ltd.

(Published - SCOPUS)

4. Hamdan, M.H., Sarmin, S.N., Karim, Z., Jawaid, M., Ismail, A.S. and Salim, N., 2026. Impact of seaweeds on tensile, thermal and viscoelasticity behavior of polybutylene adipate terephthalate-based composites. Scientific Reports. **(Published - Ql)**

Conference Proceedings

1. Hamdan, M.H., Sarmin, S. N., Jawaid, M., Ismail, A.S., Fouad, H. and Salim, N. (2025). Investigating the Influence of Varied Seaweed Filler Content on the Thermal and Tensile Properties of poly (butylene adipate-co-terephthalate) Bio-composite. In Materials Science Forum, Trans Tech Publications Ltd. *International Symposium on Advanced Polymeric Materials (ISPM 2024), UNIMAP Per lis.*

2. Hamdan, M.H., Sarmin, S. N., Abdul Jalil, A.M., Abdullah, Z.A. and Mohammad, N.A. (2024). Tensile and Morphological Properties of Biodegradable Composites Reinforced with Kenaf Core Fibres. *International Science, Engineering and Technology Colloquium (ISETC) 2024, UiTMPerak*

Innovation Competition

1. Green Packaging: Bioplastic from Seaweed (2025). International Invention, Innovation & Entrepreneurship (i3EC2025) - **Gold Award**
2. Green composites for pharmaceutical applications (IBIEC 2025) - **Silver Award**
3. Eco-composites reinforced with kenaf core (2024). Malaysia Invention & Innovation Expo 2024 (MUX 2024) - **Silver Award**
4. Biodegradable composites reinforced with seaweed (2024). Malaysia Invention & Innovation Expo 2024 (MUX 2024) - **Bronze Award**
5. Chito-Biocomposite (2024). International Innovation ARSVOT Malaysia 2024 (IAM2024) - **Bronze Award**

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1. PBAT Eco-composite reinforced with organic filler **(CRLY2024C02591)**