EVALUATION OF MECHANICAL AND INTERFACIAL PROPERTIES OF KENAF FILLER/POLY (LACTIC ACID) COMPOSITES

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By

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DECLARATION

I hereby declare that I have conducted, completed the research work and written the dissertation entitled '**Evaluation of mechanical and interfacial properties of kenaf**

filler/poly (**lactic acid**) **composites'**. I also declare that it has not been previously submitted for the award of any degree and diploma or other similar title of this for any other examining body or University.

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LIST OF SYMBOLS

ε _c	Elongation at break value for composites
εpla	Elongation at break value for pure PLA
G	the measurement of geometry, distribution, and loading conditions of
	the filler
k	Weightage factor
M _C	Young's modulus value for composites
M _{PLA}	Young's modulus value for pure PLA
η_m	stress-partitioning factors for matrix
R _m	the ratio of the filler modulus to the matrix modulus
T_{10}	Temperature when 10 % of weight is lost
T _{cc}	Cold crystallization temperature
Tg	Glass transition temperature
T _m	Melting temperature
T _{max}	Maximum decomposition temperature
To	Onset decomposition temperature
υ	Passion ratio of PLA,
W _i	weight fraction of component <i>i</i>
W_{f}	Weight fraction of kenaf filler
X _c	Degree of crystallinity

$ ho_i$	Density of component <i>i</i> ,
ΔH_{cc}	Enthalpy of cold crystallization
ΔH_m	Enthalpy of heat of fusion
$\Delta H^o{}_m$	Enthalpy of perfect heat of fusion
σ _c	Tensile strength of the composites
σρία	Tensile strength of pure PLA
ф	volume fraction of the kenaf filler, KFA and KFB

LIST OF ABBREVIATIONS

DSC	Differential Scanning Calorimetry	
DTG	Derivative Thermogravimetry	
KFA	50-100 mesh size of kenaf filler	
KFB	Pass through 100 mesh size of kenaf filler	
MFI	Melt Flow Index	
PLA	Polylactic acid	
PLA/KF	PLA filled with kenaf filler	
PLA/10KFA	PLA filled with 10 wt% KFA	
PLA/20KFA	PLA filled with 20 wt% KFA	
PLA/30KFA	PLA filled with 30 wt% KFA	
PLA/10KFB	PLA filled with 10 wt% KFB	
PLA/20KFB	PLA filled with 20 wt% KFB	
PLA/30KFB	PLA filled with 30 wt% KFB	
REB	Relative Elongation at Break	
RTS	Relative Tensile Strength	
RYM	Relative Young Modulus	
TGA	Thermogravimetric Analysis	

PENILAIAN SIFAT MEKANIKAL DAN ANTARA MUKA PENGISI KENAF/POLI (LAKTIK ASID) KOMPOSIT

ABSTRAK

Sifat mekanikal biokomposit ini yang terdiri daripada pengisi kenaf dan matriks Polilaktik asid (PLA) telah dijalankan. Pengayak mekanikal selepas pengeringan menghasilkan dua saiz pengisi kenaf berbeza, KFA (50-100 mesh) dan KFB (melepasi 100 mesh). Menggunakan penyemperit skru tunggal dengan pemuatan zarah berubah antara 10% pecahan berat, 20% pecahan berat, dan 30% pecahan berat untuk setiap saiz pengisi, komposit bio granular dihasilkan menggunakan kaedah pengacuan suntikan. Piawaian ASTM digunakan untuk menilai sifat mekanikal komposit. KFA mempunyai ketumpatan 1.3414 g/cm³ dan nisbah aspek pengisi 2.2, manakala KFB mempunyai ketumpatan 1.3874 g/cm³ dan nisbah aspek 1.9. Apabila beban pengisi bertambah, kekuatan tegangan bahan berkurangan disebabkan oleh lekatan antara muka yang lemah yang disebabkan oleh ikatan kimia. Nisbah aspek dan pecahan berat pengisi dalam komposit PLA/KF mempengaruhi sifat mekanikalnya. Terdapat perbandingan antara data eksperimen dan pelbagai model teori. Data eksperimen dibandingkan dengan model komposit termasuk Nicholais dan Narkis, Turcasanyi, Nielsens, Kernel, dan Halpin-Tsai. Korelasi dengan model Turcasanyi dan Kernel adalah baik, namun kesimpulan Nielsens, Nicholais & Narkis melebihi data eksperimen. Kandungan abu dan kestabilan terma komposit PLA ini dipertingkatkan sedikit berbanding dengan PLA yang digunakan sebagai garis dasar. Untuk biokomposit polimer berprestasi tinggi, pengisi kenaf boleh menjadi tetulang yang boleh diterima.

EVALUATION OF MECHANICAL AND INTERFACIAL PROPERTIES OF KENAF FILLER/POLY (LACTIC ACID) COMPOSITES

ABTRACT

The mechanical properties of this biocomposite constituted of kenaf filler and Polylactic acid (PLA) matrix were examined. Mechanical sieving after drying produced two different kenaf filler sizes, KFA (50-100 mesh) and KFB (pass through 100 mesh). Using a single-screw extruder with variable particle loading between 10 wt%, 20 wt%, and 30 wt% for each filler size, a granular bio-composite was produced. Using an injection moulding method, a specimen was produced. The ASTM standard was utilised to evaluate the composites's mechanical properties. KFA has a density of 1.3414 g/cm³ and a filler aspect ratio of 2.2, whereas KFB has a density of 1.3874 g/cm³ and an aspect ratio of 1.9. As the filler loading grew, the tensile strength of the material decreased due to poor interfacial adhesion caused by chemical bonding. The aspect ratio and weight fraction of filler in PLA/KF composites affected their mechanical properties. There was a comparison between experimental data and various theoretical models. The experimental data were compared to composite models including Nicholais and Narkis, Turcasanyi, Nielsens, Kernel, and Halpin-Tsai. The correlation with Turcsanyi and Kernel's model is good, however Nielsens, Nicholais & Narkis's model conclusions overestimated the experimental data. The ash content and thermal stability of this PLA are marginally enhanced in comparison to the PLA used as a baseline. For high-performance polymer biocomposites, kenaf filler could be an acceptable reinforcement.

CHAPTER 1

INTRODUCTION

1.1 Research Background

Currently, polymer composite is applied in a variety of applications. However, synthetic fibres used as reinforcement in petrochemical-based composites do not degrade biologically in composting or landfill environments, which has a major negative impact on the environment (Zini and Scandola, 2011). Recent government policies focused on footprint reduction and conservation of energy are considered the driving force towards the use of sustainable and renewable bio-based polymers. Thus, societies have switched to green resources to meet the demands of the continuously increasing population that does not affect the functioning ecosystems. (Naser et. al. 2021).

Growing desire in using more environmentally friendly and sustainable materials is a result of increased environmental awareness in the community. Natural fibre reinforced bio-derived polymer matrix composites, often known as bio-composites, have received attention as the subject of numerous research investigations and as a commercial product in the engineering industry over the past few decades (Yousif et al., 2012).

Biocomposites can be used efficiently in a wide range of applications, including mass-produced consumer products with short life cycles or products designed for once or short-term usage before disposal. Biocomposites products can be entirely degraded in the environment or in waste infrastructures such as composting units at the end of their service life without affecting the environment (Mat Taib et. al, 2009). Natural fibres' environmental benefits have had a significant influence on their utilisation. (Xanthos, 2005).

Thus, the use of natural fibre reinforced bio-composites is expanding in numerous industries, including the automotive, building, furniture, and packaging sectors. Natural fibre reinforced bio-composites have a wide range of benefits in addition to being biodegradable, like being lightweight, economical, having great specific strength and high modulus, and minimizing tool wear (Akil et al., 2011). As a result, they are frequently produced for use in car interiors, engine and gearbox covers, door panels, seal backs, and packaging. According to certain studies, the use of natural fibre bio-composites in vehicle parts results in lower carbon dioxide emissions as compared to the usage of alternative thermoplastics with a petrochemical base (Chen, 2005).

The main constituents of composites are matrix, reinforcement and interphase between matrix and reinforcement are shown in Figure 1.1. The matrix is the continuous phase that have a great influence on the overall properties of the composite which it prevent abrasion and formation of new surface flaws as it isolates the fibers from one another and hold the fibers in place. The capability to deform easily under applied load, transferring the load to the fibers and distributing the stress concentration uniformly are the characteristics of a good matrix. The second main component is reinforcement that usually was added to enhance the mechanical properties of the resin system. The interphase in composites is the region that exists between matrix and reinforcement. It can improve wettability of the composite due to their characteristic contiguous region, which sometimes can be reacted with other constituents. Its characteristics are also not described by any component in isolation. The applied load that transferred efficaciously from matrix to the fibers through the interphase can contribute in order to obtain desirable properties in composite. (Rajak et.al, 2019).



Figure 1.1: The constituent of the composite (Cazan et al. 2021)

The combination of natural fibre and thermoplastic is one of the major new areas of development that has shown remarkably high demand in industry. Polypropylene, polyvinyl chloride, and polyethylene are thermoplastics composed of long discrete molecules that melt to a viscous liquid at processing temperature. In addition, unlike thermosetting resin, thermoplastic processing is reversible.

The development of sustainable and friendly biodegradable material in industrial societies leads to biodegradable polymers such as Poly (lactic acid) (PLA) for the raw material of plastic product. In this study, poly (lactic acid) (PLA) is arguably one of the greatest possibilities in this study, as it is a versatile polymer derived from renewable agricultural raw resources (mostly starch and sugar) that are fermented to produce lactic acid (Lim et. al., 2008 & Wallenberger et. al., 2004). By its high cost, restricted availability, and low molecular weight, PLA was primarily used in medical applications like as implant devices, tissue scaffolds, and internal sutures until the last decade (Lim et. al., 2008 & Balkcom et. al., 2002).

PLA is a thermoplastic with a reasonable shelf life for most single-use packaging applications that will hydrolyze to safe natural products when disposed of appropriately. PLA breakdown occurs in two steps: hydrolysis (primary reaction) followed by bacterial attack on the fragmented residue. PLA degrades through hydrolysis into lower molecular weight lactic oligomers, which are then decomposed by microorganisms into water, carbon monoxide, carbon dioxide, and biomass (Figure 1.2).

There are no microorganisms present throughout the hydrolysis process (Garlotta, 2002 & Balkcom, 2002). Hydrolysis is the most important PLA degrading process that happens within two weeks when composted with other biomass, and the material completely disappears within three to four weeks (Wallenberger, 2004).Pure PLA can persist for more than ten years under dry conditions (Huang, 2005). Overall, PLA degrades in 6 months to 2 years, compared to 500 years to 1000 years for typical plastics such as PE and PS (Garlotta, 2002).



Figure 1.2: Hydrolytic degradation of polylactic acid. (Elsawy et. al., 2017)

To deal with recycled or reused materials, commercial uses are being examined for composites generated from natural polymer matrix and a variety of reinforcing fibres. Many matrix polymers and resins derived from renewable raw materials are widely available from renewable and sustainable resources and can be easily developed, synthesised, and manufactured using sustainable methods and disposed of after use via biodegradation. (Wallenberger and Weston, 2004).

The natural fiber is drawing considerable attention as an alternative replacement for synthetic filler of composite applications, due to their cost efficiency, sustainability and comparable performances. Among various type of natural fibers for composite applications, kenaf was selected in this study because of convenient and abundant availability in Malaysia than other types of natural fibers. This is equivalent to a significant increase in research being conducted in relation to the commercialised fibre's technical potential in contrast to other fibre plants obtainable globally. (Radzi et. al., 2018). Kenaf has considered as tropical climate crop with profuse usability. However, this fiber possessed low tolerance to heat and moisture. It also requires precise surface modification via chemical treatment to enhance the resulted matrixfiller interphase and interface interaction. In addition, due to the sustainability issue in material revolution, natural fiber like kenaf has always been selected, as a way forward to promote the utilization of green materials for many demanding engineering applications in composites. (Kamaruddin et. al, 2018).

Synthetic fibres such as carbon fibres are mixed with unsaturated polyester, epoxy, and other thermoplastic polymers such as polypropylene (PP) and polyethylene in conventional fiber reinforced composites (PE). As these composites are made up of two incompatible elements, they can't be easily recycled or reused once they've outlived their usefulness. (Mat Taib et. al., 2009). Synthetic fibres used in the reinforcing of petrochemical-based composites, on the other hand, do not biodegrade in landfills or composting environments, resulting in major contamination (Zini and Scandola, 2011). However, the major aim of adding natural fibre to thermoplastic is to emphasise the significance of natural fibre as comparatively lowcost fillers. Natural fibres can also improve the rigidity of a composite. (Rusyidah et. al., 2021).

As it has an effect on the final composites' physical and mechanical properties, the study of the interaction between the fibre and the polymeric matrix in a composite plays a significant part in the process. Specifically, adhesion, which can be defined as the ability to transfer stresses across an interface, is frequently related to a combination of a number of different factors. These factors include the thickness of the interface, the interphase layer, the adhesion strength, and the surface energy of the fibres.

Several reviews (Petinakis et al. 2013, Gurunathan et al. 2015 and Muturaj et al. 2015) discuss lignocellulose-based composites, including preparation methods and properties, but many don't consider a deep analysis of interfacial adhesion between fibre and matrix or the application of models to explain it, which are useful for predicting and optimising bio-composites' properties. Thus, further analysis of cellulose-based applications is needed. Weak interfacial interaction between polar natural fibres and a non-polar organophilic matrix can reduce bio-composites' final characteristics, thus limiting their industrial use. Hydrophilic natural fibres decrease adhesive to a hydrophobic matrix, resulting in reduction of strength.

This area of study focuses on entirely biodegradable composites in which a biodegradable polymer functions as the matrix and natural fibres act as reinforcing filler. A PLA polymeric matrix was injected with different loading of two types of short cellulosic fibres, with different aspect ratios and analyzed the effect of these had on the final mechanical qualities with the aid of selected model to evaluate the matrix-filler adhesion.

1.2 Problem Statement

Recently, a number of results have been reported for thermoplastic composites. Many different types of organic fillers can be added into polyethylene, polypropylene and other thermoplastic polymers. Several different natural organic fillers such as wood fibers and flour, kenaf fibers, sago, rice starch, cornstarch, henequen fibers, and pineapple-leaf fibers, have been used as fillers in polymer matrices. The addition of fillers into a polymer will affect the mechanical, thermal, and water absorbent properties of the composite. In addition, the properties of the composite will be unlike those of the pure polyethylene because of variations in the filler types, shape, dimension, percentage of filler loading, and presence of coupling agent.

As the advantages of the natural fillers has higher availability, lower cost, biodegradability, wide variety, and high range of mechanical and physical properties, the usage of these type of fiber become more recognizable among the manufacturing industries in various sector such as packaging, building, and automotive industry to replace synthetic fibers. Due to these, several of bio-composite were produced with polymeric biodegradable matrix and their properties were evaluated to reach the desired function in each of the applications. (Bajpai et. al, 2014).

The limitation in the widespread application of natural fibers as reinforcements for polymers is the inconsistency in their properties which depend strongly on the conditions of growth, high moisture absorption, and incompatibility with some polymeric matrices. The approach commonly followed in order to overcome the above limitations is to either surface treat the fibers or add compatibilizers and coupling agents such as maleic anhydride, acetic anhydride, and silanes, which can facilitate better fiber-polymer interfacial interactions and efficient load transfer. However, the absence of compatibilizers and use of as-received kenaf fibers will allow to study the fiber-polymer interactions and elucidate the reinforcing mechanisms.

Then, as PLA exhibits numerous limitations such as low heat stability, water barrier properties, oxygen permeability and mechanical properties, thus it is not suitable for demanding mechanical performance applications unless it is suitably modified. (Tawakkal et . al, 2012) For these reasons, one of the remarkable efforts have been made to enhance the performance of PLA by mixed with appropriate additives or with other polymers that have higher gas barrier properties. Thus, limited work has been carried out to study the improved mechanical and interfacial properties of the kenaf filler/PLA composites in various parameter variables.

It is also apparent that particle/matrix adhesion affects particulate size in polymer filled composites as well-bonded particles can effectively transfer the stress applied to the matrix, and strength becomes an interfacial adhesion component. The strength of a composite and its particle size have an inverse correlation in general, and the goal of this study is to investigate the strength regulated by particle size and its mechanical properties by using the proposed theoretical model to study the adhesion and intermolecular of filler-matrix interaction.

Kenaf is also a lignocellulosic fibre made up primarily of cellulose, hemicellulose, and lignin. Thus, the mechanical capabilities of natural fibre are highly controlled by physical parameters such as cellulose level and fibre structure (Fu et al, 2008). To manufacture kenaf-reinforced polymer composites with excellent performance, it is critical to understand all of the factors influencing the composite's mechanical properties. Many research efforts have been conducted in recent years to determine the parameters that affect the mechanical properties of natural fiberreinforced polymer composites. The impact of filler loading on the mechanical properties of the composite were also the focus of this study.

So in this work, the main focused is on the mechanical properties of the kenaf filler/PLA composite as the filler content is increasing below 40 wt% as the properties of the composites will lower beyond 40 wt% and the interfacial properties of the composite will be investigated in more details according to theoretical strength model by studying on the compatibilization of the polymer matrix and the filler in different filler loading and filler sizes.

1.3 Research Objectives

The objectives of the study are summarized as follow:

1. To investigate the effect of kenaf filler size and filler loading on the mechanical properties of the kenaf filler/Poly (lactic acid) composites.

2. To study the interaction between the kenaf filler and Poly (lactic acid) matrix by using theoretical strength model.

1.4 Scope of study

In this study, the kenaf filler was sieved into two categories of sizes which is 50-100 mesh size (KFA) and pass through 100 mesh size (KFB). Using Fourier Transform Infrared Spectroscopy (FTIR) and a scanning electron microscope, the chemical and morphological features of two distinct sizes were studied. Then, using a single screw extruder and injection moulding, pure PLA and kenaf filler/PLA composites were created. Tensile tests were used to determine the mechanical characteristics of pure PLA and kenaf filler/PLA composites. Thermogravimetric Analyzer and Differential Scanning Calorimetry were used to evaluate the thermal characteristics of pure PLA and kenaf filler/PLA composite (DSC).

1.5 Thesis structure

This thesis is divided into 5 chapters.

Chapter 1 provides general information about the study, such as an introduction, research rationale, research objectives, and thesis structure.

Chapter 2 gives a general understanding of matrices and natural fillers, as well as pertinent literature. It also involves matrix-filler interaction and thermoplastic composite processing methods.

Chapter 3 provides the information of materials and method of composite preparation used in this study. The general description of the characterization methods used also included in this chapter.

Chapter 4 covers the experimental result and discussion of the mechanical and interfacial properties of kenaf filler/PLA composites with different filler sizes and loading.

Chapter 5 draws the conclusion of this study and recommendation for future work

CHAPTER 2 LITERATURE REVIEW

2.1 Biodegradable Polymer

Biodegradation is defined as the degradation of a material caused by biological activity, particularly enzymatic action, resulting in a significant change in the chemical structure of the exposed material and the production of carbon dioxide, water, mineral salts (mineralization), and new microbial cellular constituents, according to ISO, CEN (biomass). It can develop in two distinct ways: aerobic in the presence of oxygen or anaerobic in the absence of oxygen. Biodegradable polymers are a broad category of polymers that are intended to disintegrate in the presence of live organisms. (Lovino et. al., 2007).

There are three major classes of biodegradable polymer based on Figure 2.1:

- Natural polymers: These are polymers that are generated entirely from natural sources. Natural origin biodegradable polymers provide exceptional biocompatibility, however they are not being extensively researched due to undesired features such as antigenicity and batch-tobatch fluctuation.
- Semi-synthetic polymers: These are polymers in which the raw material originates from nature, but polymerization occurs after some chemical modification.

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 iii. Synthetic polymers: These are polymers that have been developed entirely by chemical synthesis. Synthetic origin biodegradable polymers provide a number of advantages over natural polymers, including the capability to change mechanical qualities and modify the rate of decomposition depending on the requirements



Figure 2.1: Classification of biodegradable polymers (Shah & Vasava, 2019)

Biodegradable polymers give a wide range of uses, particularly in the medical field. Polyesters, which are biodegradable synthetic polymers with high biocompatibility and tunable degradation properties, are a popular choice in many biomedical applications. Poly (glycolic acid) and poly (lactic acid) are examples of poly (-hydroxy acids) (Doppalapudi et al., 2014). Due to environmental concerns and the depletion of petroleum supplies, the usage of biodegradable polymers made from renewable resources has recently received much interest.

2.1.1 Poly (lactic acid) (PLA)

PLA is an aliphatic polyester that can be derived from natural resources such as wheat, rice bran, potato starch, corn, and biomass based on lactic acid ($C_3H_6O_6$). PLA (Figure 2.1.1) is derived from annually renewable raw materials. Thus, it is a fully sustainable and biodegradable polymer as PLA-based materials are converted into water and carbon dioxide which are consumed in growing more agricultural products for further conversion to PLA. PLA has good mechanical properties compared to polyethylene terephthalate (PET) and polypropylene (PP). The melt temperature of PLA is safe for natural fibers as the natural fibers do not degrade at the processing temperature.



Figure 2.1.1: Structure of PLA

PLA can be synthesized from lactic acid monomers via a variety of ways including polycondensation, ring-opening polymerization (ROP) of the lactone ring (a lactic acid dimer known as lactide), or direct methods such as azeotropic dehydration and enzymatic polymerization (Figure 2.1.2). Polycondensation is the least costly of these. It does not, however, create PLA with a high molecular weight. Azeotropic dehydrative condensation, or ROP, is commonly used to produce high molecular weight PLA. (Lasprilla et. al. 2012 & Hamad et. al., 2015). The azeotropic

polycondensation method involves removing water molecules using appropriate azeotropic solvents, which manipulates the polymerization process' equilibrium to make relatively high molecular weight polymers at a temperature lower than the polymer's melting point temperature (Hu et. at., 2016).



Figure 2.1.2: Synthesis method of PLA (Shah & Vasava, 2019).

PLA degrades through hydrolysis and random cleavage of the ester link into lactic acid, oligomers, and other water-soluble compounds. Microorganisms breakdown these products into CO₂, water, and biomass. PLA degrades hydrolytically by surface or bulk erosion, and the rate depends on molecular weight, temperature, and pH. Molecular structure, polydispersity index, hydrophilic or hydrophobic character, crystallinity, chemical stability of polymer chains, trace amounts of catalyst, additives, contaminants and softening agents affect PLA breakdown. Polylactide mass loss depends on chirality and composition. (Shah & Vasava, 2019). Despite having desirable features, PLA applications are limited by its poor crystallisation rate, low crystallinity, low heat distortion temperature (HDT), and high cost. Owing to slow crystallisation, PLA processed via injection moulding and extrusion has weak mechanical strength and stiffness above its glass transition (T_g 60°C). Thus, PLA crystallisation kinetics must be improved for practical usage. Several approaches have been used to improve the crystallisation kinetics of PLA, including blending, adding nucleating agents, chemical modification, and/or adding plasticizing agents. (Jiang et. al., 2016)

Owing to its great renewability, biocompatibility, and biodegradability, it is widely used in both commodities and biomedical applications. The constituent monomer of PLA is 2-hydroxypropionic acid, which is a chiral molecule that exists in both D- and L-enantiomeric forms (Figure 2.1.3), and the properties of the polymer are determined by the stereoregularity of monomers in the polymeric backbone; thus, polymer with desired properties can be obtained by varying the amount of enantiomeric stereocenters (D- or L-) in the resulting PLA. Table 2.1.1 shows the general features of lactic acid polymers.



Figure 2.1.3: Two stereoisomers of lactic acid (Avinc and Khoddami, 2009).

Lactic acid polymers	Glass transition temperature T _g (°C)	Melting temperature T _m (°C)	Density (g/cm ³)	Solubility
PLLA	55-80	173-178	1.290	CHCl ₃ , furan, dioxane, and dioxolane
PDLLA	43-53	120-170	1.25	PLLA solvents and acetone
PDLA	40-50	120-150	1.248	Ethyl lactate, THF, ethyl acetate, DMSO, N-xylene, and DMF

Table 2.1.1 Properties of lactic acid polymers (Lasprilla et. al., 2012)

PLA's principal applications in biomedical industries over the previous decade have been limited to sutures, medication delivery, and orthopaedic implants. Nonetheless, PLA is a flexible polymer with numerous potential applications due to its respectable ease of production, mechanical characteristics, and, most importantly, biodegradability. With these properties, PLA is a great material for food packaging and other consumer products. PLA can be used in a variety of applications, including textiles and injection moulded components. (Balkcom, 2002, Garlotta, 2002, & Mat Taib et al., 2009) PLA has numerous potential applications in fibre and non-wovens. It can be easily transformed into a variety of fibre types using conventional meltspinning methods. (Platt, 2006 & Balkcom, 2002)

2.2 Natural fiber

Wood flour, kenaf, hemp, flax starch, sisal, and jute are examples of natural organic cellulose fibres that are numerous, affordable, renewable, exhibit good specific mechanical characteristics, and are totally biodegradable. They've long been recognised as an applicable reinforcing filler or fibre in thermoplastics and thermoset polymers. They can be mixed with plastics to create materials that are more biodegradable while still maintaining the beneficial characteristics of traditional plastics. (Wu, 2009). The physical and mechanical properties of selected natural fibres and glass fibres (the most often used synthetic fibres) are compared in Tables 2.2.

Properties	Natural fibers	Glass fibers
Density	Low	Much higher
Renewability	Yes	No
Suitability for recycling	Yes	No
Wear of tool during processing	Low	High
Health risk during inhalation	Low	High
Biodegradability	Yes	No
Production cost	Moderate	Somewhat higher

Table 2.2: general characteristic of natural and glass fibers (Kaczmar et. al., 2007)

Microfibers' constitute natural fibres, and the direction of their greatest strength distinguishes them. The microfibers, on the other hand, are primarily made up of cellulose and are held together by lignin and hemicellulose. Lignocellulose cells are simple tubes that are 0.7–5 mm long and 30–90 mm in diameter. The fibrous lignocellulose components act as reinforcement in the composites, improving their mechanical properties significantly. (Kaczmar et. al., 2007).

The natural fiber is drawing considerable attention as an alternative replacement for synthetic filler of composite applications, due to their cost efficiency, sustainability and comparable performances. Natural fibres used in polymer composites provide advantages such as low density, biodegradability, the ability to dampen acoustic waves, and the simplicity with which they may be recycled. Incorporating such fibres into the polymer matrix significantly decreases its thermal expansion. The acceptable temperature range in the manufacturing of natural fiberreinforced composites is a limiting factor, which should not exceed 230°C due to risk of fibre degradation. (Garbarczyk & Borysiak., 2004).

2.2.1 Kenaf fiber

Kenaf (Hibiscus cannabinus L.) fibers have a low density (1.2-1.6 g/cm³), outstanding characteristics, are non-abrasive, and have high stiffness properties, and have a significant influence on both the physical and mechanical properties of polymer composites (Rowell et al. 1997). Kenaf, one of Malaysia's many industrial crops, is expected to contribute significantly to the processing industry. This is due to massive increase in research being conducted in relation to the technical potential of commercialised fibre in compared to other fiber plants accessible internationally.

Kenaf is a low-cost renewable material that can be used to commercialize composite materials for the development of high-performance engineering products. Kenaf growth has increased in recent years for two main reasons. For starters, kenaf is capable of absorbing phosphate and nitrogen from the soil. Phosphorus and nitrogen improve crop height, cumulative weed weight, stem diameter, and fibre output. Second, kenaf has a higher photosynthetic rate than other typical trees, therefore it can transforms carbon dioxide to oxygen at a much faster pace (Kuchinda et al., 2001).

The kenaf plant has two different fibres: short fibre derived from the core and long fibre derived from the bast. These two fibres give composites excellent mechanical properties. Although the mechanical capabilities of kenaf fibre are lower than those of standard fibres such as carbon, glass, and Kevlar, kenaf fibre can still be considered a great substitute to those synthetic fibre due to its cost effectiveness and low-density properties. (Rusyidah et. al., 2021)

2.3 Kenaf filler/PLA composite

Aliotta et. al (2019) have demonstrated the biodegradable polymers, Poly(Lactic acid) PLA were reinforced with two different types of cellulosic fibers, BWW40 and 600Be/PU to produce bio-composites which having different percentages of fibers and their mechanical, interfacial and thermal properties were investigated. Both of these properties met the requirement for practical applications as the fiber inside the composite was stable and not degraded until the temperature reached beyond 300 °C and the overall mechanical properties of PLA with addition of cellulose fibers was negligibly improved. Therefore, the Punkánszky's model that was used in this paper

to estimate the adhesion between the matrix and the filler had been verified as there is no significant improvement of strength due to low adhesion between these cellulosic fiber and PLA matrix.

Moreover, Bajpai et. al (2014) carried out the study of biodegradable polymer based composite with the green fibers such as flax, nettle, jute, sisal, kenaf and so on as a potential replacement of synthetic fibers and Polylactic acid has been used for polymer based green composite. They studied about the mechanical characterization of the composite which include their tensile strength, compressive strength, flexural strength, flexural modulus, impact strength and inter-laminar shear strength. They concluded that the mechanical properties of bio-composites depend on a number of parameters such as percentage of fiber content, interfacial characteristics between fiber and matrix, fiber aspect ratio, surface modification of fibers and addition of various additives (coupling agents) to enhance the compatibility between fiber and matrix. Also, the chemical treatment of the fibers, use of fillers and additives and processing techniques are the one of the ways to improve the adhesion characteristic of the matrix and natural fibers.

According to Han et al (2012), the addition of the Kenaf filler on PLA without using any fiber treatment method or compatibilizers showed that the strength of the composite was insignificantly increased with the fiber content compared to Kenaf fiber/PP composites. This is due to the higher melt viscosity of PLA that reduce the fiber-polymer interactions. Thus, the mechanism for the enhancement of strength is closely related to the fiber-polymer interfacial interactions. They had been concluded that the Kenaf fibers was act as nucleating agent in these composites due to changes in the crystallization temperature and melting behavior of PLA. In SEM, it did not indicate any significant differences in the dispersion and distribution of the fibers within the two polymers and did not show any difference in the fiber pull out that reflects the interfacial adhesion between the two composite systems.

Radzi et. al. (2018) explored the influence of kenaf filler reinforcement on injection-molded polypropylene composites. Filler weights were 10%, 20%, 30%, and 40%. The maximum tensile strength for kenaf 20 and 40 mesh core filled composites was achieved at 30 wt%. At 30% filler loading, kenaf bast-filled composites had 19.52 MPa tensile strength. Thus, kenaf bast filled composites had the highest tensile strength values at each loading percentage. Instead, kenaf 20 mesh core filled composites showed the maximum flexural strength, 25 MPa at 20% and 29 MPa at 30%. Compared to kenaf bast filled composites, kenaf 40 mesh core filled composites showed the highest values at 25.35 MPa. Increased filler loadings increased the modulus strength of both kenaf/PP composites.

The properties of the PLA reinforced with natural cellulose that chemically derived from the plant-based kenaf bast fibers has been improved in the study carried out by Tawakkal et.al, 2012. They mentioned that the flexural properties of the bio-composites is increasing as the loading of the natural fibers increased up to 20% of Kenaf derived cellulose (KDC) where it also attributed on improvement of the adhesive characteristic. However, the non-uniform length and diameter as well as aspect ratio were assumed to effect interfacial adhesion as the ability to support stress transmitted from PLA matrix was reduced. As KDC content increased, the elasticity of PLA has been suppressed thus reduce the deformability of the composite and elongation at break. However, the impact strength has been concluded to not significantly affect the composite when KDC loading was increased up to 30wt% and

further addition slightly reduced the strength due to incorporation of the KDC into PLA matrix.

Fu et. al. (2008) study on experimental results and theories of polymer-based particulate micro and nano-composites' mechanical characteristics. The effects of particle size, particle/matrix adhesion, and particle loading on composite stiffness, strength, and toughness are studied. All three parameters, especially particle/matrix adhesion, affect composite strength and toughness. This is expected as strength depends on filler-to-matrix stress transfer and adhesion controls toughness/brittleness. Due to the interplay between these three parameters, particle loading affects composite strength and toughness in various ways. Composite stiffness depends on particle loading, not particle/matrix adhesion, since fillers have a higher modulus than the matrix. There is a critical particle size, usually in nanoscale, below which composite stiffness is considerably enhanced due to the particle size, perhaps driven by the much larger surface areas giving a "nano"-effect. They evaluate existing test data and theoretical and phenomenological models to understand the strengthening, stiffening, and toughening mechanisms in particle composites. Existing phenomenological and empirical/semi-empirical models are used to analyze polymerbased particle composites.

For the theoretical model, Maldhure et al. (2011) concluded that there are various mathematical models that can be proposed for the composites to evaluate the interfacial adhesion between the filler and the matrix. The model that was used in this study was Nicholais & Narkis, Turcsanyi, Nielsens, Kernel, Halpin-Tsai, and Sata-Furukawa model where these later can also verified the compatibility and intermolecular interactions that affected the mechanical properties. Among these, Tursanyi, Nielsen, and Halpin-Tsai can be used to prove the intermolecular interaction of the polypropylene with modified maleic anhydride lignin (PPMALig) composite and polypropylene with modified dichloroethane (PPCELig) composites by their miscibility of the theoretical models to experimental data. The compatibility of PPMALig composites can be referred to the effect of the stiffening from the modified lignin on the PP matrix was greater than PPCELig where it then confirmed some of the intermolecular interaction between filler and matrix.

Divya et al. (2015) studied the micromechanical model to analysed the improvement of the mechanical properties for nanocomposites using both cenospheres and multi-walled carbon nanotubes (MWCNT), where they concluded that the interfacial interaction had been improved. By using the Nicolais and Narkis, Piggot and Leiner, Nielsen, Turcsanyi, Halpin-Tsai, Kerner, Sato-Furukawa and Mori-Tanaka, the interfacial adhesion had been analysed to further evaluate the tensile strength. For relative tensile strength (RTS) the Turcsanyi model, a constant B value (related to interfacial properties) of 4.1 indicates enhanced matrix filler adhesion in this study as the model were closely match with the experimental while Nicholais & Narkis showed the slightly fitting with the experimental curve along with the other models. In relative tensile modulus (RYM), they conclude that Kerner and Halpin-Tsai model was the model that match closely compared to Sato-Furukawa and Mori-Tanaka as this indicates the effectively interfacial adhesion of cenospheres and MWCNT with the compatibilizer.