EFFECT OF MALEIC ANHYDRIDE GRAFTING ON PROPERTIES OF BAMBOO AND JUTE FIBRES WOVEN FABRIC REINFORCED POLYLACTIC ACID COMPOSITES

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LIST OF ABBREVIATIONS AND SYMBOLS

s	second
min	minute
BC	before Christ
AD	Anno Domini
mm	millimetre
cm	centimeter
m	meter
psi	pound per square inch
Pa	pascal
kPa	kilopascal
MPa	megapascal
GPa	gigapascal
mg	milligram
g	gram
kg	kilogram
ha	hectare
C	carbon atom
O	oxygen atom
H	hydrogen atom
O_2	oxygen
CO_2	carbon dioxide
MA	maleic anhydride
DCP	dicumyl peroxide

PLA	polylactic acid
PLA-g-MA	maleated polylactic acid
UPE	unsaturated polyester
HDPE	high density polyethylene
T_{onset}	initial decomposition temperature
T_{50}	temperature at which 50 % of material had thermally decomposed
T_{peak}	temperature at which weight loss rate is maximum
T_g	glass transition temperature
T_m	melting temperature
T_{cc}	cold crystallization temperature
X_c	crystalline fraction
%	percentage
wt%	weight fraction
E’	storage modulus
Tan δ	damping factor
3D	three dimensional
$	dollar sign
HT	Halpin-Tsai
ROM	rule of mixture
°C	degree Celsius
FTIR	Fourier Transform Infrared Spectrometer
ASTM	American Society for Testing and Materials
J	joule
ml	millilitre
kV	kilovolt

Hz	Hertz
°	degree
α	fibre twist angle
rpm	revolutions per minute
~	approximately
≤	less than or equal to
+	positive
–	negative
ATR-FTIR	Attenuated Total Reflectance-Fourier Transform Infrared Spectrometer
SEM	Scanning Electron Microscope

KESAN PENCANGKUKAN MALEIK ANHIDRIDA TERHADAP SIFAT-SIFAT KOMPOSIT ASID POLILAKTIK DIPERKUAT FABRIK TENUN GENTIAN BULUH DAN JUT

ABSTRAK

Polilaktik acid (PLA) mesra alam dan mempunyai sifat tegangan yang baik. Tetapi, kekuatan impak PLA agak lemah. Justeru, fabrik serat buluh dan jut yang mempunyai kekuatan mekanikal yang tinggi digunakan sebagai pengukuh. Dalam kajian ini, fabrik serat buluh/PLA dan fabrik serat jut/PLA komposit dihasilkan dan dikaji. Pertama, kesan kandungan serat; 20 dan 30 wt% terhadap sifat komposit. Keputusan menunjukkan 30 wt% komposit mempunyai kestabilan terma yang rendah, tetapi mempunyai sifat penyerapan air, terma dan mekanikal yang tinggi. Hal ini disebabkan sifat terma serat yang kurang stabil, hidrofilik secara semulajadi, agent nukleasi dan sifat mekanikalnya yang tinggi. Kedua, komposit fabrik serat buluh/PLA dan komposit fabrik serat jut/PLA dibandingkan. Keputusan menunjukkan komposit fabrik serat buluh/PLA mempunyai kestabilan terma dan modulus yang rendah disebabkan oleh kestabilan terma dan modulus serat buluh yang rendah. Keputusan juga menunjukkan struktuk saling-mengunci fabrik serat buluh padat dan ini membawa kepada dua kejadian. Pertama, banyak tenaga diperlukan untuk mencetuskan fenomena tarikan serat dalam buluh. Justeru, kekuatan impak fabrik serat buluh/PLA komposit adalah lebih tinggi. Kedua, menghalang impregnasi PLA ke dalam fabrik serat buluh. Oleh yang demikian, fabrik serat buluh/PLA komposit mempunyai kekuatan tegangan dan lenturan yang lebih rendah dan penyerapan air yang tinggi. Ketiga, kesan pengacuan mampatan; masa dan tekanan terhadap sifat

mekanikal 30 wt% komposit. Degradasi terma serat berlaku pada 6 atau 9 minit. Justeru, masa 3 minit memberi 30 wt% komposit kekuatan mekanikal yang tertinggi. Fabrik serat buluh rosak pada tekanan 1,000 atau 1,500 psi. Malah, impregnasi PLA bagi fabrik serat jut dengan lebihan serat pada 500 atau 1,000 psi juga tidak mencukupi. Justeru, tekanan 500 dan 1,500 psi memberi sifat mekanikal tertinggi untuk kedua-dua 30 wt% fabrik serat buluh/PLA dan fabrik serat jut/PLA komposit. Keempat, kesan MA dan DCP pada cangkukan MA untuk PLA-g-MA. Keputusan menunjukkan peningkatan kandungan MA dan DCP sehingga 6.0 dan 0.9 wt% menghasilkan cangkukan MA tertinggi bagi PLA-g-MA. Hal ini dikaitkan dengan bilangan makroradikal PLA dan MA yang lebih tinggi. Kelima, kesan kandungan PLA-g-MA; 3, 5 dan 10 wt% terhadap sifat 30 wt% komposit. Kajian menunjukkan PLA-g-MA menjejaskan kestabilan terma untuk 30 wt% komposit disebabkan oleh sifat terma PLA-g-MA yang kurang stabil. Namun, PLA-g-MA meningkatkan lekatan antara serat-PLA dan seterusnya meningkatkan sifat penyerapan air, terma, mekanikal, dan dinamik mekanikal 30 wt% komposit. Kesimpulannya, komposit ini berpotensi digunakan untuk aplikasi dalaman seperti bahagian automotif dalaman.

EFFECT OF MALEIC ANHYDRIDE GRAFTING ON PROPERTIES OF BAMBOO AND JUTE FIBRES WOVEN FABRIC REINFORCED POLYLACTIC ACID COMPOSITES

ABSTRACT

Polymers like polylactic acid (PLA) is environmentally friendly with good tensile properties. However, impact strength of PLA is low. Hence, mechanically stronger bamboo and jute fibre fabrics were used as reinforcement. In this study, bamboo fibre fabric/PLA and jute fibre fabric/PLA composites were fabricated and studied. Firstly, effect of fibre content; 20 and 30 wt% on the properties of composites. Results showed that 30 wt% composites have lower thermal stability, higher water absorption, thermal and mechanical properties. These are attributed to fibre being less thermally stable, hydrophilic in nature, a nucleating agent and mechanically stronger. Secondly, bamboo fibre fabric/PLA and jute fibre fabric/PLA composites were compared. Results showed that bamboo fibre fabric/PLA composite have lower thermal stability and modulus due to lower thermal stability and modulus of bamboo fibre. Results also showed that the interlocking structure of bamboo fibre fabric is tighter and this led to two outcomes. First, more energy required for bamboo fibre pull out to occur. Hence, impact strength of bamboo fibre fabric/PLA composite is higher. Second, hinder PLA impregnation of bamboo fibre fabric. Hence, bamboo fibre fabric/PLA composite have lower tensile and flexural strength and higher water absorption. Thirdly, effect of compression moulding; time and pressure on the mechanical properties of 30 wt% composites. Thermal degradation of fibre occurred at 6 or 9 min. Hence, 3 min led to highest mechanical properties of 30 wt% composites.

Bamboo fibre fabric was damaged at 1,000 or 1,500 psi. Insufficient PLA impregnation of jute fibre fabric with more fibre entanglement at 500 or 1,000 psi. Hence, 500 and 1,500 psi led to highest mechanical properties of 30 wt% bamboo fibre fabric/PLA and 30 wt% jute fibre fabric/PLA composites. Fourthly, effect of MA and DCP content on the MA grafting of PLA-g-MA. Results showed that increasing MA and DCP content until 6.0 and 0.9 wt% resulted in highest MA grafting of PLA-g-MA. These are attributed to higher number of MA and PLA macroradicals. Fifthly, effect of PLA-g-MA content; 3, 5 and 10 wt% on the properties of 30 wt% composites. PLA-g-MA adversely affect the thermal stability of 30 wt% composites due to less thermally stable PLA-g-MA. However, PLA-g-MA improved the interfacial adhesion of fibre-PLA and this further improved the water absorption, thermal, mechanical and dynamic mechanical properties of 30 wt% composites. In conclusion, these composites have the potential for indoor applications such as interior automotive parts.

CHAPTER 1

INTRODUCTION

1.1 Background

A composite is made up of two or more constituent materials. These constituent materials are distinctly different from each other in their chemical, physical, thermal and mechanical properties. The combination of these constituent materials produces a composite exhibiting property superior than its individual constituent material property. This led to the wide application of composite by humans in their everyday life. In 1500 BC, composite made up of straw and mud was used in the construction of strong and durable buildings by the Egyptian and Mesopotamian settlers. In 1200 AD, the Mongols used composite made up of animal glue, bone and wood in the construction of a strong and light weight composite bow (Ngo, 2020).

Later in 1900s, the successful development of synthetic fibre and petroleum-based polymer led to the introduction of composite made up of synthetic fibre and petroleum-based polymer. This synthetic fibre/petroleum-based polymer composite was used in the construction of commercial boat hull in 1946, a full automotive body in 1947, a pedestrian bridge located in Aberfeldy, Scotland and a bridge deck located in West Virginia and Kansas, United States during the 1990s. In 2009, synthetic fibre/petroleum-based polymer composite was used in the construction of a commercial airliner, Boeing 787 Dreamliner. At present, synthetic fibre/petroleum-based polymer composite is widely used in the manufacture of aerospace components, sporting equipment, consumer and industrial products (Ngo, 2020).

The major driving force behind the diverse application of synthetic fibre/petroleum-based polymer composite in our everyday life is its customizability to meet specific final properties required of a product. This is done by selecting and

combining the appropriate synthetic fibre/petroleum-based polymer composite constituents to ensure its final properties meet the product specification (Girijappa et al., 2019). One such example is by aligning high stiffness synthetic fibre in a defined direction within synthetic fibre/petroleum-based polymer composite to ensure its final properties meet the hockey stick or kayak paddle specification that requires high stiffness in a defined direction. Another example is the selection of fire-resistant petroleum-based polymer in synthetic fibre/petroleum-based polymer composite to ensure it is fire-resistant according to the product specification.

Other advantages of synthetic fibre/petroleum-based polymer composite are its high strength to weight ratio and high stiffness to weight ratio. This makes it suitable in the manufacture of transportation components as it provides structural stability and increases transportation fuel efficiency (Girijappa et al., 2019). However, the issue with synthetic fibre/petroleum-based polymer composite is it is made up of non-renewable and non-environmentally friendly materials, synthetic fibre and petroleum-based polymer. Consequently, a non-renewable and non-environmentally friendly synthetic fibre/petroleum-based polymer composite that is harmful to the environment at a time when there is a growing awareness worldwide to the environmental problems we are currently facing (Das and Chaudhary, 2021; Rajeshkumar et al., 2021).

This has prompted an increase interest in renewable and environmentally friendlier natural fibre/polymer composite made up of natural fibre and polymer derived from biological origin (Ilyas et al., 2021). Another advantage of natural fibre/polymer composite is its customizability to meet specific final properties required of a product, high strength to weight ratio and high stiffness to weight ratio similar to the advantages offered by synthetic fibre/petroleum-based polymer composite. These advantages of natural fibre/polymer composite allow it to be

considered as an alternative material to synthetic fibre/petroleum-based polymer composite (Stefaniak and Masek, 2021). Hence, narrowing down the focus of this thesis specifically to natural fibre/polymer composite.

In natural fibre/polymer composite, its constituent natural fibre is mechanically stronger than its other constituent, polymer. Since natural fibre is mechanically stronger than polymer, natural fibre is also known as a reinforcement material within natural fibre/polymer composite. Additionally, natural fibre is inexpensive, abundantly available, renewable, environmentally friendly, has a low density and cause less abrasive damage to natural fibre/polymer composite processing equipment (Kandemir et al., 2021; Karimah et al., 2021). Examples of natural fibre include bamboo fibre and jute fibre.

Bamboo fibre is extracted from bamboo. Bamboo is a fast-growing plant, able to grow by up to 5 cm per hour and 18 m in 3 months, allowing bamboo to yield a large amount of bamboo fibre in a short time (Zhang et al., 2020). Bamboo is also easy to cultivate with around 63 types of bamboo found in the peninsular Malaysia, ensuring a constant supply of bamboo fibre in Malaysia (Radzi et al., 2022). Apart from that, the added benefit of cultivating bamboo is it is able to absorb about 1.83 kg of atmospheric CO_2 in a month as well as soil contaminants like waste water from human activities and heavy metals during its lifespan (Emamverdian et al., 2020).

As for the other natural fibre, jute fibre, it is extracted from jute. Jute needs very little water and chemicals like fertilizers, herbicides and pesticides during its cultivation, making jute easier to cultivate for its jute fibre (Samanta et al., 2020). Similar to bamboo, jute is also a fast-growing plant, requiring about 3 to 4 months before jute fibre can be extracted from it. The other similarity of jute to bamboo is,

jute can also absorb atmospheric CO_2 at about 13,600 kg per ha and at the same time release about 9,900 kg per ha of O_2 to the atmosphere (Sarkar et al., 2016).

In natural fibre/polymer composite, its other constituent, the renewable and environmentally friendlier polymer, also known as a matrix material, functions as a binder that holds the natural fibre/polymer composite together. Example of this polymer is polylactic acid (PLA). In addition to being renewable and environmentally friendlier, PLA has better tensile and flexural properties compared to other commonly used polymers like polypropylene, polyester, unsaturated polyester, polyethylene and high impact polystyrene. PLA also has desirable thermal stability, can be processed using either extrusion, injection moulding, blow moulding, compression moulding or fibre spinning and reprocessed again for the second time as PLA is a thermoplastic (Albuquerque et al., 2021; Teixeira et al., 2021; Patel et al., 2022). The combination of PLA with either bamboo fibre or jute fibre results in a renewable and environmentally friendlier bamboo fibre/PLA composite and jute fibre/PLA composite that has all the good attributes of their constituents.

However, the issue with bamboo fibre/PLA composite and jute fibre/PLA composite are the incompatibility between bamboo fibre with PLA and jute fibre with PLA respectively. The hydroxyl groups present on the surface of bamboo fibre and jute fibre makes bamboo fibre and jute fibre highly hydrophilic in nature while PLA is hydrophobic in nature. This causes poor interfacial adhesion between bamboo fibre with PLA and jute fibre with PLA in bamboo fibre/PLA composite and jute fibre/PLA composite. As a result of their poor interfacial adhesion, stress cannot be transferred efficiently from the mechanically weaker PLA to the mechanically stronger bamboo fibre or jute fibre when bamboo fibre/PLA composite and jute fibre/PLA composite is under mechanical stress. Subsequently, compromising the mechanical properties of

bamboo fibre/PLA composite and jute fibre/PLA composite (Balla et al., 2019; Karthi et al., 2020).

Hence, it is vital to improve bamboo fibre/PLA composite and jute fibre/PLA composite interfacial adhesion as it affects their mechanical properties. Literature has suggested two solutions to this problem, the first solution is bamboo fibre and jute fibre surface modification prior to bamboo fibre/PLA composite and jute fibre/PLA composite fabrication (Balla et al., 2019). Examples of fibre surface modification include thermal plasma treatment, non-thermal plasma treatment, alkaline treatment and acetylation treatment among others. Nonetheless, the drawback of these fibre surface modification far outweighs their advantage. Thermal plasma treatment causes damage to fibre-based materials while non-thermal plasma treatment is expensive (Ferreira et al., 2019). Alkaline treatment and acetylation treatment may cause fibre degradation, adversely affecting fibre mechanical properties (Latif et al., 2018).

This led to the second solution, chemically modifying PLA into maleated polylactic acid (PLA-g-MA) and using it as a compatibilizer in bamboo fibre/PLA composite and jute fibre/PLA composite (Balla et al., 2019). There are two advantages of this solution, firstly it does not produce any effluent as a by-product and secondly, a two-fold increase in the mechanical strength of natural fibre/polymer composite compared to silane treatment (Koohestani et al., 2018). This solution was also successfully used to improve natural fibre/PLA composite interfacial adhesion and mechanical properties. Zhang et al., (2017) reported that the addition of PLA-g-MA as compatibilizer improved wood fibre/PLA composite tensile strength and flexural strength. Ghasemi et al., (2018) reported that the addition of PLA-g-MA as compatibilizer improved linter pulp fibre/PLA composite tensile strength and impact strength.

Others used PLA-g-MA as compatibilizer in rice hull fibre/PLA composite (Dimzoski et al., 2008), wheat straw fibre/PLA composite (Nyambo et al., 2011), flax fibre/PLA composite (Arias et al., 2013; Aitor et al., 2020), hemp fibre/PLA composite (Gunning et al., 2014), cotton fibre/PLA composite (Ghasemi et al., 2018) and sisal fibre/PLA composite (Aitor et al., 2020). Yet, there is a lack of literature on PLA-g-MA as compatibilizer in bamboo fibre/PLA composite and jute fibre/PLA composite. Thus, PLA-g-MA was used as compatibilizer in bamboo fibre/PLA composite and jute fibre/PLA composite in this study.

1.2 Problem Statement

In natural fibre fabric, natural fibres are twisted into yarns and yarns are weaved into natural fibre fabric oriented at 0 ° and 90 ° direction (Arifuzzaman Khan et al., 2016). The biaxial orientation of yarns in natural fibre fabric will impart a high axial strength at 0 ° and 90 ° direction to composite (Aisyah et al., 2021). Additionally, the tight interlocking structure of weaved yarns in natural fibre fabric will further increase the axial strength of composite (Jauhari et al., 2015). With this in mind, bamboo fibre fabric and jute fibre fabric were used as reinforcement in this study.

Equally important is the selection of natural fibre content in composite. Literature reported that 20 wt% fibre content maximized the impact strength of bamboo fibre/PLA (Liu et al., 2019) and jute fibre/PLA composite (Gunti et al., 2018). Literature had also reported that 30 wt% fibre content slightly reduced the impact strength of bamboo fibre/PLA composite by –10.38 % (Liu et al., 2019). However, 30 wt% fibre content increased the tensile strength and tensile modulus of bamboo fibre/PLA composite by +7.40 % and +25.13 % (Sujaritjun et al., 2013; Liu et al., 2019). With this in mind, 20 wt% and 30 wt% fibre content were selected to maximize

the mechanical properties of bamboo fibre fabric/PLA and jute fibre fabric/PLA composite in this study.

To fabricate composite, injection moulding and compression moulding are two commonly used fabrication method in literature (Tokoro et al., 2008; Depuydt et al., 2019). In injection moulding, mechanical stress applied onto fibres during melt-blending will shorten the length and width of fibres by –81.98 % and –66.67 % (Gallos et al., 2017; Albrecht et al., 2018). Unlike injection moulding, compression moulding allows natural fibre fabric to retain its structure in the fabricated composite (Ismail et al., 2015; Rajak et al., 2019). Additionally, compression moulding is a relatively simple and low-cost fabrication method with short cycle time and high degree of reproducibility (Wang et al., 2019; Greene, 2021). With this in mind, compression moulding was used to fabricate bamboo fibre fabric/PLA and jute fibre fabric/PLA composite in this study.

In compression moulding, temperature, time and pressure will affect the mechanical properties of fabricated composite. In the case of temperature, Nurul Fazita et al., (2014) suggested 160 °C. At 160 °C, the viscosity of PLA reaches the recommended viscosity at 100 Pa/s to facilitate PLA impregnation of fabric without disorienting the yarns in fabric. With this in mind, 160 °C was used to fabricate bamboo fibre fabric/PLA and jute fibre fabric/PLA composite in this study.

In the case of time, Katayama et al., (2006) and Tanaka et al., (2014) suggested 10 min. However, 10 min is too long and led to discoloration of bamboo fibre fabric into darker colour in this study. The discoloration of fibre into darker colour is an initial sign of the thermal degradation of fibre (Fan and Naughton, 2016; Hart and Summerscales, 2017). In the case of pressure, Nurul Fazita et al., (2013) suggested 50

psi to 150 psi. However, 50 psi to 150 psi is too low to facilitate PLA impregnation of fabric in this study. Hence, compression moulding; time and pressure were optimized to maximize the mechanical properties of fabricated bamboo fibre fabric/PLA and jute fibre fabric/PLA composite in this study.

In natural fibre/PLA composite, hydrophilic natural fibre and hydrophobic PLA is incompatible. This incompatibility causes poor interfacial adhesion between natural fibre and PLA. Subsequently, compromising the mechanical properties of natural fibre/PLA composite. To improve the interfacial adhesion and mechanical properties of natural fibre/PLA composite, literature suggested chemically modifying PLA into PLA-g-MA and using PLA-g-MA with highest MA grafting as compatibilizer.

Nyambo et al., (2011) reported that PLA-g-MA with 1.6 %, 1.7 % and 3.8 % MA grafting as compatibilizer improved the tensile strength of wheat straw fibre/PLA composite by +11.40 %, +18.59 % and +21.87 %. Csikos et al., (2015) reported that PLA-g-MA with 0.25 %, 0.70 %, 1.61 % and 2.40 % MA grafting as compatibilizer improved the tensile strength of wood fibre/PLA composite by +5.37 %, +8.18 %, +10.97 % and +17.78 %. With this in mind, PLA-g-MA with highest MA grafting was used as compatibilizer in bamboo fibre fabric/PLA and jute fibre fabric/PLA composite in this study.

To synthesize PLA-g-MA with highest MA grafting, Rigolin et al., (2019) suggested optimizing MA and DCP content. If MA content is too low, there will be an insufficient amount of MA to react with PLA. If MA content is too high, MA will homopolymerize instead of reacting with PLA (Asmak et al., 2020). If DCP content is too low, there will be an insufficient amount of DCP to facilitate reaction between MA

and PLA. If DCP content is too high, DCP will cause chain scission or cross linking of PLA instead of facilitating reaction between MA and PLA (Pan et al., 2018). Consequently, MA and DCP content that is too low or too high will ultimately lead to lower MA grafting of PLA-g-MA. With this in mind, MA and DCP content were optimized to maximize the MA grafting of the synthesized PLA-g-MA to be used as compatibilizer in bamboo fibre fabric/PLA and jute fibre fabric/PLA composite.

PLA-g-MA had been used as compatibilizer in literature to improve the interfacial adhesion and mechanical properties of natural fibre/PLA composite. However, there is a lack of literature on PLA-g-MA as compatibilizer to improve the interfacial adhesion and mechanical properties of natural fibre fabric/PLA composite. This study addresses the research gap by using PLA-g-MA as compatibilizer to improve the interfacial adhesion and mechanical properties of bamboo fibre fabric/PLA and jute fibre fabric/PLA composite.

As for the hypothesis of this study, there are two hypotheses in this study. Bamboo and jute fibre fabric are cellulose-based natural fibre similar to other cellulose-based natural fibre like flax, hemp, wood and sisal fibre used to reinforce PLA in literature. All of this literature reported that PLA-g-MA as compatibilizer improved the interfacial adhesion and mechanical properties of cellulose-based natural fibre/PLA composite. Hence, the first hypothesis of this study is PLA-g-MA as compatibilizer will also improve the interfacial adhesion and mechanical properties of cellulose-based bamboo fibre fabric/PLA and cellulose-based jute fibre fabric/PLA composite.

According to literature, bamboo and jute fibre have different thermal stability, physical and mechanical properties. Since bamboo and jute fibre have different

properties, bamboo fibre fabric and jute fibre fabric will also have different properties. Subsequently, bamboo fibre fabric/PLA and jute fibre fabric/PLA composite will also have different properties. Hence, the second hypothesis of this study is bamboo fibre fabric/PLA and jute fibre fabric/PLA composite will also have different thermal stability, physical, mechanical and dynamic mechanical properties.

1.3 Research Objectives

1) To investigate the effect of bamboo and jute fibre fabric content; 20 wt% and 30 wt% on the thermal stability, physical, thermal, and mechanical properties of bamboo fibre fabric/PLA and jute fibre fabric/PLA composites.

2) To compare the thermal stability, physical and mechanical properties of bamboo fibre fabric/PLA and jute fibre fabric/PLA composites.

3) To investigate the effect of compression moulding factors; time and pressure on the mechanical properties of 30 wt% bamboo fibre fabric/PLA and 30 wt% jute fibre fabric/PLA composites using Taguchi experimental design.

4) To investigate the effect of grafting reaction factors; MA and DCP content on the MA grafting of PLA-g-MA using Taguchi experimental design.

5) To investigate the effect of PLA-g-MA content; 3 wt%, 5 wt% and 10 wt% on the thermal stability, physical, thermal, mechanical and dynamic mechanical properties of 30 wt% bamboo fibre fabric/PLA-g-MA/PLA and 30 wt% jute fibre fabric/PLA-g-MA/PLA composites.

CHAPTER 2

LITERATURE REVIEW

2.1 Materials

2.1.1 Natural Fibre

Interest in natural fibres over synthetic fibres as polymeric reinforcement stems from increasing environmental awareness worldwide. Other than that, there are also other favourable factors favouring natural fibres over synthetic fibres and these factors are summarized in Table 2.1. Natural fibres can be further categorized based on their origin into plant, animal and mineral. Their main constituent is cellulose, protein and silicates respectively.

Table 2.1: Comparison between natural and synthetic fibres

		Natural Fibres	Synthetic Fibres
Cost	Production Cost	Lower production cost	Higher production cost
	Cost per Weight	Lower cost per weight • 200 – 1,000 $/ton	Higher cost per weight • glass fibres, 1,200 – 1,800 $/ton • carbon fibres, 12,500 $/ton
Properties	Durability	Lower durability	Higher durability
	Quality	Non-homogeneous quality	Homogeneous quality
	Density	Lower density • 0.9 – 1.6 g/cm^3	Higher density • glass fibres, 2.5 g/cm^3 • carbon fibres, 1.8 – 2.0 g/cm^3
	Moisture Resistance	Lower moisture resistance	Higher moisture resistance
	Tensile	Lower tensile strength • 14 – 1,200 MPa	Higher tensile strength • glass fibres, 2,000 – 3,000 MPa • carbon fibres, 3,500 MPa
		Lower tensile modulus • 9 – 70 GPa	Higher tensile modulus • glass fibres, 70 GPa • carbon fibres, 228 GPa

Table 2.1: Continued

		Natural Fibres	Synthetic Fibres
Properties	Specific Tensile	Specific tensile strength • 9.2 – 923.1 $MPa.g^{-1}cm^3$ Comparable to glass fibres Lower than carbon fibres	Specific tensile strength • glass fibres, 800 – 1,200 $MPa.g^{-1}cm^3$ • carbon fibres, 1,750 – 1,944 $MPa.g^{-1}cm^3$ Higher than natural fibres
		Specific tensile modulus • 5.2 – 48.3 $GPa.g^{-1}cm^3$ Higher than glass fibres Lower than carbon fibres	Specific tensile modulus • glass fibres, 28 $GPa.g^{-1}cm^3$ Lower than natural fibres • carbon fibres, 127 – 114 $GPa.g^{-1}cm^3$ Higher than natural fibres
Processability	Processing Temperature	Lower processing temperature • ≤ 200 °C	Higher processing temperature • glass fibres, < 300 °C • carbon fibres, < 450 °C
	Abrasive Damage to Processing Equipment	Lesser abrasive damage	More abrasive damage
Environmental	Energy Consumption for Production	Require lesser energy • 4 GJ/ton	Require more energy • glass fibres, 30 GJ/ton • carbon fibres, 130 GJ/ton
	CO_2 Emission during Production	Lower CO_2 emission	Higher CO_2 emission
	Biodegradability and Recyclability	Biodegradable and recyclable	Non-biodegradable and difficult to recycle

$ (dollar sign); g (gram); cm (centimetre); MPa (megapascal); GPa (gigapascal); °C (degree Celsius); GJ (gigajoule); CO_2 (carbon dioxide)

References: Feih et al., (2009); Mukhtar et al., (2016); Pickering et al., (2016); Ali et al., (2018); Pecas et al., (2018); Sanjay et al., (2019)

Mineral fibres like asbestos are no longer used and are even banned in several countries due to their carcinogenic nature while animal fibres like silk exhibits high strength but they are relatively costly, less readily available and exhibit low stiffness. Unlike animal fibres, plant fibres exhibit high strength and stiffness and are more

readily available as they can be grown in most places worldwide and harvested after short periods (Pickering et al., 2016).

2.1.2 Bamboo Fibre

Bamboo plant is classified under family *Poaceae* and subfamily *Bambusoideae*, has three advantages compared to other plants where natural fibres are sourced from. Firstly, bamboo plant is a fast-growing plant that can grow at around 21 cm per day and will reach maturity in 3 years (Rocky and Thompson, 2018). Secondly, bamboo plants have the ability to absorb carbon dioxide emitted by two cars on a daily basis during the growth of bamboo plants (Emamverdian et al., 2020). Thirdly, bamboo plants are abundantly available at 31 million hectares of bamboo forest land worldwide; 3.1 million hectares in Africa, 3.1 million hectares in America and 24.8 million hectares in Asia (Figure 2.1).

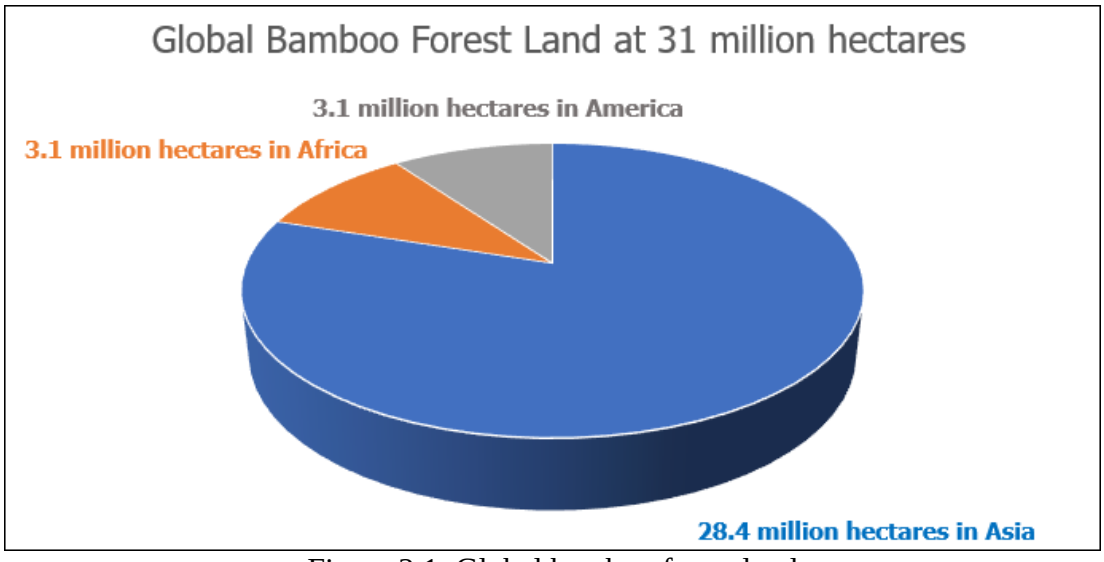

Figure 2.1: Global bamboo forest land

References: Lobovikov et al., (2007); Emamverdian et al., (2020)

The first and third advantage of bamboo plants led to higher yield of bamboo fibres at 27,216 million kg compared to other natural fibres like jute, kenaf, flax, sisal, hemp, coir, ramie and abaca (Table 2.2). The second advantage of bamboo plants led

to a reduction in the effects of global warming and climate change during the growing phase of bamboo plants.

Bamboo fibres are harvested from the stem wall of bamboo plant (Figure 2.2). The chemical constituent of the harvested bamboo fibres is cellulose at 26 to 43 wt%, hemicellulose at 30 wt% and lignin at 21 to 31 wt% (Syduzzaman et al., 2020). The cellulose content in bamboo fibres will impart a high tensile strength and high tensile modulus to bamboo fibres at 441.0 MPa (Shito et al., 2002) and 35.9 GPa (Ali et al., 2018). These bamboo fibres also have a low density at 1.52 gcm^{-3}. Since bamboo fibres have high tensile strength coupled with low density as well as high tensile modulus coupled with low density, bamboo fibres will have high specific tensile strength as well as high specific tensile modulus.

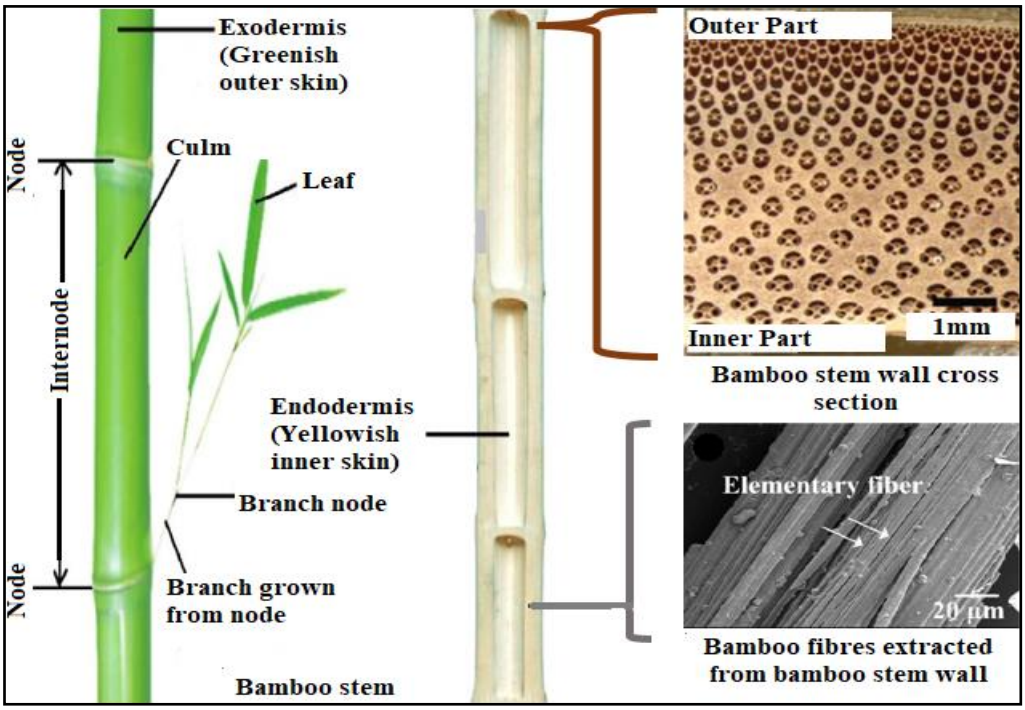

Figure 2.2: Different parts of bamboo stem

References: Osorio et al., (2018); Rocky and Thompson, (2018); Zhang et al., (2018)

Based on Table 2.2, the specific tensile strength of bamboo fibres at 290.1 MPa per gcm^{-3} is higher than coir and abaca fibres. The specific tensile modulus of bamboo

fibres at 23.6 GPa per gcm^{-3} is higher than sisal, coir and ramie fibres. Other than that, the price of bamboo fibres at $ 0.40 per kg is cheaper than kenaf, flax, sisal and hemp fibres. According to literature, bamboo fibres also have antibacterial and antifungal properties that will enhance the durability of bamboo fibres (Nayak and Mishra, 2016; Azeez and Orege, 2018). All the advantageous properties of bamboo fibres had attracted the interest of researchers to seriously consider bamboo fibres to replace synthetic fibres as polymeric reinforcement material (Porras and Maranon, 2012; Nurul Fazita et al., 2013).

Table 2.2: The production, price, specific tensile strength and specific tensile modulus of natural fibres

Natural Fibres	World Production (10^6 kg)	Price/Kg ($)	Mechanical Properties	
			Specific tensile strength (MPa/gcm^{-3})	Specific tensile modulus (GPa/gcm^{-3})
Bamboo	27,216	0.40	290.1	23.6
Jute	2,087	0.31	343.3	41.0
Kenaf	880	0.50	230.8 – 923.1	16.9 – 46.2
Flax	753	0.96	351.7 – 627.6	34.5 – 48.3
Sisal	343	0.51	69.0 – 551.7	6.2 – 15.2
Hemp	194	0.99	209.8 – 531.5	21.0 – 42.0
Coir	91	0.35	87.0 – 173.9	5.2
Ramie	91	1.97	590.3	14.8
Abaca	64	2.08	9.2	27.0

kg (kilogram); $ (dollar sign); MPa (megapascal); GPa (gigapascal); g (gram); cm (centimetre)

References: Faruk et al., (2012); Ali et al., (2018)

2.1.3 Jute Fibre

Jute plant is classified under family *Malvaceae* and genus *Corchorus*. Jute plant has three advantages compared to other plants where natural fibres are sourced from. Firstly, jute plant is a fast-growing plant that will reach maturity in 3 to 4 months with very little need for water, fertilizers, herbicides and pesticides (Salman, 2020). Secondly, a hectare of land cultivated with jute plants can absorb 13,000 kg of carbon

dioxide from the atmosphere and release 10,000 kg of oxygen back into the atmosphere (Tanjinul et al., 2021). Thirdly, jute plants are cultivated on a large scale in India and Bangladesh while other countries like China, Nepal, Thailand and Pakistan cultivate jute plants on a smaller scale (Summerscales et al., 2010).

The first and third advantage of jute plants led to higher yield of jute fibres at 2,087 million kg, higher than the yield of kenaf, flax, sisal, hemp, coir, ramie and abaca fibres (Table 2.2). The second advantage of cultivating jute plants for jute fibres led to a reduction in the effects of global warming and climate change during the cultivation phase of jute plants.

Jute fibres are harvested from the stem of jute plant. The chemical constituent of the harvested jute fibres is cellulose at 61.0 to 71.5 wt%, hemicellulose at 13.6 to 20.4 wt% and lignin at 12.0 to 13.0 wt% (Syduzzaman et al., 2020). The cellulose content in jute fibres will impart a high tensile strength and high tensile modulus to jute fibres at 460.0 MPa (Shito et al., 2002) and 55.0 GPa (Ali et al., 2018). These jute fibres also have a low density at 1.34 gcm^{-3}. Since jute fibres have high tensile strength coupled with low density as well as high tensile modulus coupled with low density, jute fibres will have high specific tensile strength as well as high specific tensile modulus.

Based on Table 2.2, the specific tensile strength of jute fibres at 343.3 MPa per gcm^{-3} is higher than bamboo, coir and abaca fibres. The specific tensile modulus of jute fibres at 41.0 GPa per gcm^{-3} is higher than bamboo, sisal, coir, ramie and abaca fibres. Other than that, the price of jute fibres at $ 0.31 per kg is cheaper than bamboo, kenaf, flax, sisal, hemp and coir fibres. All the advantageous properties of jute fibres had attracted the interest of researchers to consider jute fibres in the fabrication of

polymer composites. According to Sanjay et al., (2019), these jute fibre/polymer composites have several potential applications in the field of aerospace, automotive, marine, building and construction, sporting goods, leisure goods and others (Table 2.3).

Table 2.3: Potential applications of jute fibre/polymer composite

Field	Application
Aerospace	tails, wings, propellers and helicopter fan blades
Automotive	door frames, door shutters, window frame and mirror casing
Marine	boat hulls and fishing rods
Building and Construction	roofing sheets, bricks, furniture panels, storage tanks and pipelines
Sport and Leisure Goods	ice skating boards, bicycle frames, baseball bats, tennis racket, fork, helmet and post-boxes
Electronic Appliances	laptop and mobile cases, chip boards, projector and voltage stabilizer cover
Others	pipes carrying coal dusts, weapons construction, textiles, industrial fans, paper and packaging

References: Sanjay et al., (2019)

2.1.4 Natural Fibre Fabric

Natural fibre plain weaved fabric at the macroscopic level is made up of weaved natural fibre yarns at the mesoscopic level. Natural fibre yarns at the mesoscopic level are made up of twisted short natural fibres at the microscopic level. In other words, to produce natural fibre plain weaved fabric, short natural fibres are initially mechanically extracted (decortication and retting) or chemically extracted (solution spinning) from the plant (Sanjay et al., 2019). The extracted short natural fibres were later twisted together into natural fibre yarns. Finally, the twisted natural fibre yarns were weaved together into natural fibre plain weaved fabric (Strugala et al., 2018). The entire process from the plant, bamboo or jute to the production of

natural fibre plain weaved fabric, bamboo plain weaved fabric or jute plain weaved fabric is illustrated in Figure 2.3.

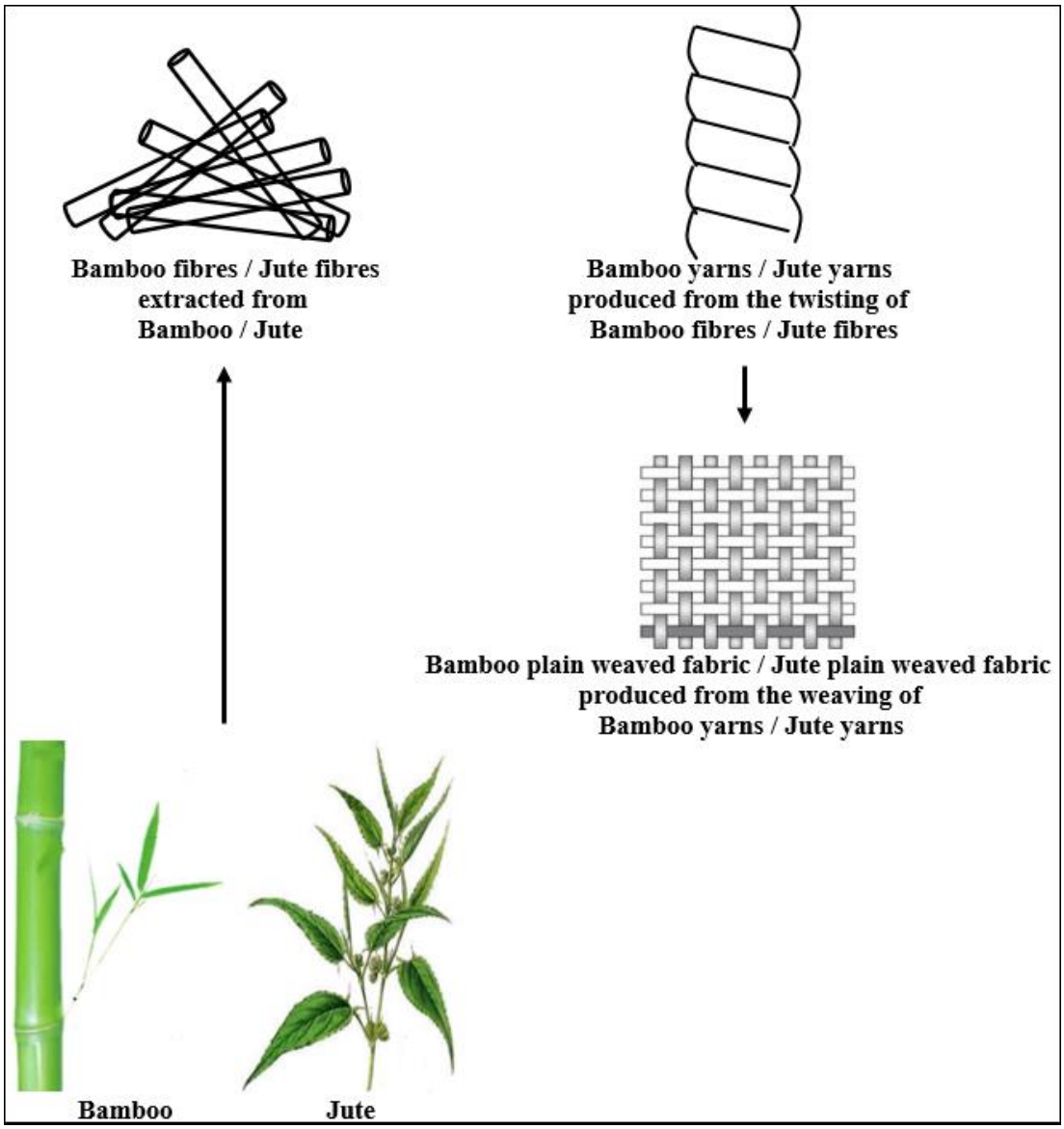

Figure 2.3: Production of natural fibre plain weaved fabric from plant

References: Islam, (2019); Rocky and Thompson, (2018); Sanjay et al., (2019)

2.1.5 Polymer

Polymers can be categorized into petroleum-based polymers and non-petroleum-based polymers. Petroleum-based polymers are produced from non-renewable petroleum resources while non-petroleum-based polymers are produced from renewable natural resources. The renewable natural resources can either be

directly from micro-organisms, plants and animals or are chemically synthesized from biological components such as fats, sugars, proteins, amino acids and vegetable oils (Sunita et al., 2018). These renewable natural resources are biodegradable. Since non-petroleum-based polymers are produced from renewable natural resources, the biodegradability of renewable natural resources is transferred to the non-petroleum-based polymers. The biodegradable nature of non-petroleum-based polymers allow micro-organisms in the environment to degrade these polymers into carbon dioxide and water (Mohan et al., 2016).

Hence, polymer products made of non-petroleum-based polymers like textile apparels, automotive interior components and food packaging can be disposed into the environment at the end of their life cycle without polluting the environment (Albuquerque et al., 2021). There are also other applications that will require polymer to have a short-life span. One of it is mulching film. Conventional mulching film is usually made of petroleum-based polymers. Conventional mulching film is used to cover up the surface of the soil around the cultivated crop to prevent weeds from growing. However, the issue with conventional mulching film is its long-life span, planters will have to manually remove them after each growing season.

On the contrary, mulching film made of non-petroleum-based polymer that is biodegradable has a short-life span and will biodegrade by itself without the need for the planters to manually remove them after each growing season. One such example of non-petroleum-based polymer is PLA. Other than being environmentally friendlier, PLA has comparable or better specific mechanical properties compared to other more often used polymers in the field of natural fibre/polymer composite (Table 2.4).

Table 2.4: Specific mechanical properties of polymers used in natural fibre/polymer composite

Polymers	Specific Mechanical Properties				
	Tensile Strength, MPa/ $g.cm^{-3}$	Tensile Modulus, GPa/ $g.cm^{-3}$	Flexural Strength, MPa/ $g.cm^{-3}$	Flexural Modulus, GPa/ $g.cm^{-3}$	Impact Strength, kJ/m^{-2}/ $g.cm^{-3}$
Epoxy	66.32	2.32	79.91	2.10	3.44
Vinyl Ester	76.00	3.00	155.00	3.50	16.00
High Impact Polystyrene	26.52	1.72	57.67	1.86	0.11
Polyester	9.92	1.15	39.34	1.80	1.44
Unsaturated Polyester	19.74	0.50	61.58	1.16	1.62
Low Density Polyethylene	10.49	0.42	11.24	0.22	–
High Density Polyethylene	33.04	1.44	33.04	1.25	0.07
Modified Acrylic	52.58	2.19	79.99	–	0.03
Polylactic Acid	49.88	2.73	84.68	3.00	0.03
Polypropylene	24.06	1.13	27.22	1.12	0.22

MPa (megapascal); GPa (gigapascal); g (gram); cm (centimetre); kJ (kilojoule); m (meter)

References: Devi et al., (1997); Gowda et al., (1999); Zhou et al., (2007); Liu et al., (2012); Nurul Fazita, (2014); Klimek-McDonald et al., (2018); Hexion; ICOM Composites; Laminated Plastics Distributors & Fabricators; Curbell Plastics

2.1.6 PLA

The building block of PLA is lactic acid, hence the need to understand lactic acid production before proceeding to PLA production and PLA application in this section. Lactic acid can be produced either by bacterial fermentation or chemical synthesis. Out of these two methods, bacterial fermentation is preferred (Casalini et al., 2019; Singhvi et al., 2019) as chemical synthesis is costly, has limited capacity and is unable to produce the desired lactic acid stereoisomer. To produce PLA, lactic acid

will undergo polymerization *via* any one of these three methods; direct condensation polymerization, direct polycondensation in an azeotropic solution or polymerization through lactide formation (Figure 2.4).

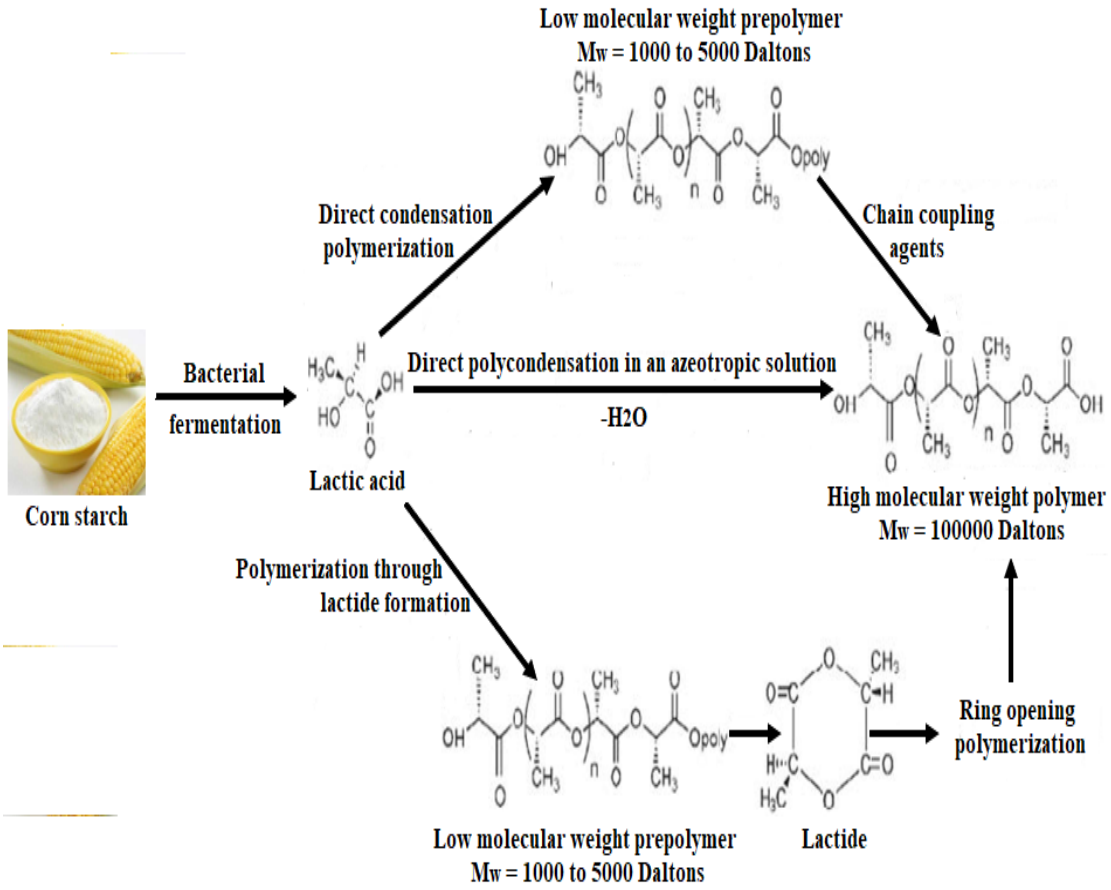

Figure 2.4: Synthesis methods of PLA

References: Jamshidian et al., (2010); Lopes et al., (2014)

For the first method, lactic acid will initially undergo esterification in some solvents and later undergo progressive vacuum at high temperatures to produce high molecular weight PLA. As for the second method, lactic acid will initially undergo distillation in an azeotropic solution at low distillation pressures and later filtered to produce high molecular weight PLA. However, the first and second method produce high molecular weight PLA that contain impurities. Unlike the first and second method, the third method produces high molecular weight PLA at milder conditions without using any solvents. The third method also produces high molecular weight

PLA that does not contain impurities. For these reasons, the third method was used by the industry to produce high molecular weight PLA on a larger scale.

The high molecular weight PLA that is produced are transparent, resistant to photodegradation and has desirable properties like gas barrier, tensile and flexural (Albuquerque et al., 2021). These properties of PLA allow it to be used in a variety of sectors such as in the biomedical sector (tissue engineering, drug delivery and implants), textile sector (apparels) and automotive sector (car interior components). Other than that, PLA is also used in the agriculture sector (mulch films) and food packaging sector (bags, bottles, containers and films) (Rajeshkumar et al., 2021). However, the high molecular weight PLA also has its own shortcoming like less than desirable impact strength, limiting the application of PLA in other sectors. To further widen the current application of PLA, literature had suggested reinforcing PLA with natural fibres to improve the less than desirable impact strength of PLA (Ilyas et al., 2021).

2.2 Effect of Fibre Content on the Properties of Bamboo Fibre/PLA and Jute Fibre/PLA Composites

2.2.1 Water Absorption of Bamboo Fibre/PLA and Jute Fibre/PLA Composites

The water absorption of PLA will initially show a slight increase. Once the saturation point is reached, the water absorption of PLA will remain constant throughout the entire duration of the experiment. Gunti et al., (2016) found that the water absorption of PLA reached the saturation point after 12 hours of immersion, increasing the water absorption of PLA to 0.72 %. Past this saturation point, the water absorption of PLA remained constant at around 0.72 to 0.87 % throughout the entire duration of the experiment from 12 to 48 hours of immersion. Nurul Fazita et al., (2014) found that the water absorption of PLA reached the saturation point after 1 day

of immersion, increasing the water absorption of PLA to 0.59 %. Past this saturation point, the water absorption of PLA remained constant at around 0.59 to 0.72 % throughout the entire duration of the experiment from 1 to 30 days of immersion. From the findings made in literature, the water absorption of PLA is low due to PLA being hydrophobic in nature (Nurul Fazita et al., 2014).

As for natural fibre/PLA composite, its water absorption will initially show a significant increase. Once the saturation point is reached, the water absorption of natural fibre/PLA composite will remain constant throughout the entire duration of the experiment. Gunti et al., (2016) found that the water absorption of 25 wt% jute fibre/PLA composite reached the saturation point after 12 hours of immersion, increasing the water absorption of 25 wt% jute fibre/PLA composite to 9.14 %. Past this saturation point, the water absorption of 25 wt% jute fibre/PLA composite remained constant at around 9.14 to 9.95 % throughout the entire duration of the experiment from 12 to 48 hours of immersion. Nurul Fazita et al., (2014) found that the water absorption of 35 wt% bamboo fibre/PLA composite reached the saturation point after 15 days of immersion, increasing the water absorption of 35 wt% bamboo fibre/PLA composite to 11.09 %. Past this saturation point, the water absorption of 35 wt% bamboo fibre/PLA composite remained constant at around 11.09 to 13.05 % throughout the entire duration of the experiment from 1 to 30 days of immersion. From the findings made in literature, the water absorption of natural fibre/PLA composite is high due to natural fibre being hydrophilic in nature (Lee and Wang, 2006).

2.2.2 Thermal Stability of Bamboo Fibre/PLA and Jute Fibre/PLA Composites

Literature had found that PLA only undergo single stage thermal degradation (Lee and Wang, 2006; Goriparthi et al., 2012; Hu et al., 2018). Unlike PLA, natural fibres such as bamboo fibres and jute fibres undergo three stage thermal degradation

based on the thermal gravimetric analysis conducted by Lee and Wang, (2006), Goriparthi et al., (2012) and Nurul Fazita et al., (2014). The three stages of natural fibre thermal degradation are attributed to moisture loss at 30 to 110 °C, hemicellulose at 280 °C and cellulose at 360 °C according to Goriparthi et al., (2012). However, Lee and Wang, (2006) attributed the three stages of natural fibre thermal degradation to hemicellulose at 250 to 300 °C, cellulose at 300 to 400 °C and lignin at ~ 420 °C.

In contrast, Nurul Fazita et al., (2014) found that bamboo fibres in fabric form only undergo single stage thermal degradation. The single stage of bamboo fabric thermal degradation is attributed to cellulose at ~ 273 °C. The removal of hemicellulose and lignin in bamboo fibres during the degumming process of bamboo fibres prior to the processing of bamboo fibres into bamboo fabric resulted in the bamboo fabric consisting mainly of cellulose (Subash and Muthiah, 2021). In the case of natural fibre/PLA composite, Hu et al., (2018) reported single stage thermal degradation but Lee and Wang, (2006) reported two stage thermal degradation. The two stages of natural fibre/PLA composite thermal degradation are attributed to PLA at 280 to 340 °C and natural fibre at ~ 350 °C.

Other than the thermal degradation stages of a material, thermal gravimetric analysis also provides information on the thermal stability of a material. The thermal stability is expressed as T_{onset} (initial decomposition temperature), T_{50} (temperature at which 50 % of material had thermally decomposed) and T_{peak} (temperature at which weight loss rate is maximum). The higher these values are, the better, as higher values of T_{onset}, T_{50} and T_{peak} indicates better thermal stability of the material analysed.

Based on Table 2.5, the thermal stability of PLA is better than bamboo fibres (Lee and Wang, 2006; Nurul Fazita et al., 2014; Wang et al., 2014) and jute fibres