LIGNOCELLULOSE BASED HYBRID LAMINATE COMPOSITE: PHYSICAL, MECHANICAL AND FLAMMABILITY PROPERTIES OF OIL PALM FIBER/GLASS FIBER REINFORCED EPOXY RESIN.

by

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LIST OF ABBREVATIONS

ABS	Acrylonitrile-Butadiene Styrene
PBT	Poly(Para Phenylene Benzobisimidazole)
PEEK	Polyetheretherketone
PEK	Polyetherketone
PPS	Polyphenylenesulphide
EFB	Empty fruit bunch
GF	Glass Fiber
CSM	Chopped strand mat

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KOMPOSIT LAMINAT HYBRID BERASASKAN LIGNOSELULOSA: SIFAT-SIFAT FIZIKAL, MEKANIKAL DAN KEBOLEHBAKARAN KOMPOSIT GENTIAN KELAPA SAWIT/GENTIAN KACA DIPERKUATKAN RESIN EPOKSIDA.

ABSTRAK

Dalam kajian ini, sifat-sifat mekanikal, fizikal dan kebolehbakaran komposit laminat hibrid berasaskan lignoselulosa telah di kaji. Daripada kajian tersebut telah diperhatikan bahawa penambahan gentian kaca dalam komposit gentian kelapa sawit telah meningkatkan sifat-sifat tensil, hentaman dan fleksural komposit tersebut. Kesan hibrid negatif yang telah diperhatikan untuk kekuatan tensil, modulus Young, kekuatan fleksural dan modulus fleksural adalah disebabkan oleh struktur laminasi hibrid komposit. Walau bagaimana pun, kesan positif hibrid telah diperhatikan untuk pemanjangan dan kekuatan hentaman hibrid komposit berasaskan lignoselulosa. Kekuatan hentaman komposit lignoselulosa yang dihentam pada lapisan gentian kaca adalah lebih tinggi daripada komposit yang dihentam pada lapisan gentian kelapa sawit. Walau bagaimana pun, kekuatan fleksural yang tinggi telah dipamerkan oleh komposit hibrid lignoselulosa apabila beban dikenakan pada lapisan gentian kelapa sawit. Di samping itu, penambahan gentian kaca telah pun mengurangkan kandungan rongga, sifat-sifat penyerapan air dan mengurangkan kebolehbakaran komposit berasaskan lignoselulosa.

Kesan penyusunan lapisan terhadap sifat-sifat mekanikal, fizikal dan keupayaan terbakar komposit laminat hibrid berasaskan lignoselulosa telah di kaji dan dibandingkan dengan komposit hibrid dua lapisan EFB/GF. Komposit hibrid EFB/GF/EFB dan GF/EFB/GF telah mengandungi kandungan rongga yang kurang berbanding komposit dua

lapisan EFB/GF. Kekuatan fleksural dan modulus fleksural komposit hibrid GF/EFB/GF adalah lebih tinggi daripada komposit hibrid dua lapisan EFB/GF. Modulus fleksural komposit GF/EFB/GF juga adalah jauh lebih tinggi daripada modulus fleksural komposit gentian kaca. Disamping itu, kekuatan hentamam komposit EFB/GF/EFB adalah lebih tinggi daripada komposit dua lapisan EFB/GF. Selain itu, komposit hibrid EFB/GF/EFB dan GF/EFB/GF telah mempamerkan kekuatan hentaman yang lebih tinggi dari komposit dua lapisan EFB/GF. Akan tetapi, kesan penyusunan lapisan didapati tidak mempengaruhi kebolehbakaran komposit hibrid lignoselulosa.

Komposit hibrid EFB/GF/EFB dan GF/EFB/GF telah diperkuatkan dengan gentian kaca teranyam dihasilkan dan sifat-sifat mekanikal, fizikal dan kebolehbakaran komposit tersebut dibandingkan dengan hibrid komposit yang diperkuatkan tikar gentian kaca terpotong (chopped strand mat). Didapati bahawa komposit hibrid diperkuatkan gentian kaca teranyam mempunyai kekuatan hentaman dan kebolehbakaran yang rendah berbanding hibrid komposit diperkuatkan gentian kaca terpotong. Tiada peningkatan yang ketara diperhatikan pada kekuatan fleksural dan modulus fleksural bagi komposit hibrid diperkuatkan gentian teranyam. Walau bagaimana pun, kandungan rongga yang tinggi dalam komposit hibrid teranyam mengakibatkan keupayaan penyerapan airnya lebih tinggi berbanding komposit hibrid diperkuatkan gentian kaca terpotong.

ABSTRACT

The mechanical, physical and flammability properties of the lignocellulose hybrid laminate composites were studied in this research. It was observed that the addition of glass fibers into the oil palm fiber composite enhanced the tensile, flexural and impact properties of the lignocellulose composites. Owing to the interlaminated structure of the composite, a negative hybrid effect was observed for the tensile strength, Young's modulus, flexural strength and flexural modulus. However, a positive hybrid effect was observed for the elongation at break and the impact strength of the lignocellulose hybrid laminate composites. The lignocellulose hybrid laminate composites, which were impacted at the glass fiber layer exhibited higher impact strength than when impacted at the oil palm fiber layer. On the other hand, the hybrid composites exhibited higher flexural strength when loaded at the oil palm fiber layer. Furthermore, the hybridization of the oil palm fiber composite with glass fibers reduced the void content, the water absorption properties and enhanced the flammability properties of the lignocellulose composite.

The effect of stacking sequence on the mechanical, physical and flammability properties of the lignocellulose hybrid laminate composites were studied and was compared with the EFB/GF hybrid bi-layer laminate composite. The stacking sequence of EFB/GF/EFB and GF/EFB/GF hybrid composites reduced the void content of the hybrid composite compared to the EFB/GF bi-layer laminate composite. The GF/EFB/GF hybrid composite exhibited an enhanced flexural strength and modulus as compared to the bi-layer laminate composite. The flexural modulus though was higher than the glass fiber composite. The impact strength of the EFB/GF/EFB hybrid composite was higher than the EFB/GF hybrid composite. Besides that, the EFB/GF/EFB and GF/EFB/GF hybrid composites exhibited improved water absorption properties compared to the EFB/GF

hybrid bi-layer laminate composite. However the effect of stacking sequence did not seem to have contributed to the flammability properties of the lignocellulose hybrid laminate composites.

The EFB/GF/EFB and GF/EFB/GF hybrid composites were reinforced with a plain weave glass fabric and the mechanical, physical and flammability properties of the hybrid composites were compared with the chopped strand mat reinforced hybrid composites. The woven fabric reinforced hybrid composites exhibited enhanced impact strength and improved flammability properties compared to the chopped strand mat hybrid composites. Significant improvement in the flexural properties were not observed in the woven fabric reinforced hybrid composites. The high void content of the woven fabric reinforced hybrid composites resulted in an increase in the water absorption behavior of the composites compared to the chopped strand mat reinforced hybrid composites.

CHAPTER 1

INTRODUCTION

1.1 Lignocellulose Fiber Composites: Overview

In recent years, owing to the increased environmental awareness, the usage of lignocellulosic fibers as a potential replacement for synthetic fibers such as carbon, aramid and glass fibers in composite materials have gained interest among researchers throughout the world. Extensive studies which have been done on lignocellulosic fibers such as sisal (Joseph *et al.*, 1996 and Oksman *et al.*, 2002), jute (Pal *et al.*, 1984, 1988 and Albuquerque *et al.*, 2000), pineapple (George *et al.*, 1998; Devi *et al.*, 1997 and Mishra *et al.*, 2001), banana (Pothan *et al.*, 1997 and Joseph *et al.*, 2002) and oil palm empty fruit bunch fibers (Rozman *et al.*, 1998; Hill *et al.*, 2000 and Abdul Khalil *et al.*, 2001) have shown that lignocellulosic fibers have the potential to be an effective reinforcement in thermoplastics and thermosetting materials.

According to Bledzki *et al.* (1999), Wambua *et al.* (2003) and Mishra *et al.* (2000), lignocellulose fibers offer several advantages over their synthetic fiber counterparts. Lignocellulose fibers are abundant in nature, renewable raw material and a low cost material. Owing to their low specific gravity, which is about 1.25-1.50 g/cm^3 compared to synthetic fibers, especially glass fibers which is about 2.6 g/cm^3, lignocellulose fibers are able to provide a high strength to weight ratio in plastic materials. The usage of lignocellulose fibers also provides a healthier working condition than the synthetic fibers. This is due to the fact that, the glass fiber dust from the trimming and mounting of glass fiber components causes skin irritation and respiratory diseases among workers. Besides that, the less abrasive nature of the lignocellulose fibers offers a friendlier processing environment as the wear of tools could be reduced. Furthermore, lignocellulose fibers offer good thermal and insulating properties, easily recyclable and are biodegradable especially when used as reinforcement in a biopolymer matrix.

These advantages have gained interest in the automotive industry where materials of lightweight, high strength to weight ratio and minimum environmental impact are required. Automotive giants such as DaimlerChrysler are using flax/sisal fiber mat embedded in an epoxy matrix for the door panels of the Mercedes Benz E-class model (Gayer and Thomas, 1996). Coconut fibers bonded with natural rubber latex are being used for seat cushions in the Mercedes Benz A-Class model (Deem, 2003). Cambridge Industry (an automotive industry in Michigan, USA) is making flax fiber reinforced polypropylene for Freightliner Century COE C-2 heavy trucks and also rear shelf trim panels of the 2000 model Chevrolet Impala (Sherman, 2003). Besides the automotive industry, lignocellulosic fiber composites such as jute fiber reinforced polyester have also found application in the building and construction industries such as panels, ceilings and partition board.

However, lignocellulose fiber composite products are still limited to the interior of cars that are not exposed to strong mechanical impacts and non-structural components compared to synthetic fiber composites which are used widely in high performance engineering applications such as in the aerospace industry. This is because lignocellulosic fiber composites have low strength properties, poor moisture resistance, poor microbial and fire resistance and low durability properties.

Therefore, through hybridization of the lignocellulosic fibers with a stronger and more corrosion resistant synthetic fiber such as glass fibers, the strength, stiffness, moisture and fire resistant behavior of the lignocellulosic composite can be improved significantly.

1.2 Objectives of The Research

Hybridization (intermingled system) of lignocellulose fibers with glass fibers have been studied extensively by researchers throughout the world. However, in this study a different type of lignocellulose based hybrid composite which is an intraply or interlaminated hybrid composite was produced. Studies on lignocellulose based hybrid laminate (interply) composite are still new and not much research has been published. Therefore, an attempt was undertaken by our research group to understand the mechanical and physical properties of a lignocellulose based hybrid laminate composite. The main objectives of this research are summarized below:

1) To study and understand the effect of glass fiber loading on the mechanical, physical and flammability properties of oil palm fiber/glass fiber hybrid bi-layer laminate composites.
2) To study the effect of stacking sequence on the mechanical, physical and flammability properties of the oil palm fiber/glass fiber hybrid laminate composites. The mechanical physical and flammability properties of the EFB/GF/EFB and GF/EFB/GF hybrid composites were compared with the EFB/GF bi-layer hybrid composite.
3) To study the effect of woven glass fiber fabric as a reinforcement in EFB/GF/EFB and GF/EFB/GF hybrid composites.

CHAPTER 2

LITERATURE REVIEW

2.1 Composite Materials.

In the new millennium, composite materials are considered as a new and important class of engineering materials.

The earliest example of a composite material can be traced back in the third millennium BC when Egyptians made bricks from clay reinforced with straw (Astrom, 1977 and Matthews *et al.*, 1999). The Mongolians though produced bows which were made from a combination of wood, animal tendons and silk.

Composite materials can be divided into natural occurring composite and synthetic composites. Many natural occurring materials are classified as composite materials. Examples of natural composites are wood which is made up of cellulose molecules in a lignin matrix, teeth and bone which are composed of hydroxyapetite in a matrix of collagen, insect exoskeleton to name a few. Examples of synthetic composites are concrete (combination of stone and cement), asphalt and also glass fiber reinforced unsaturated polyester matrix.

2.1.2 Definition of Composite

Nowadays, composite materials are used in a wide variety of applications. According to Astrom (1997), the noun composite is derived from the Latin verb *componere* which means to put together. Therefore, a composite material can be considered as a material which is formed when two or more chemically distinct constituents are combined together on a macro scale (Astrom, 1977; Schwartz, 1992 and Jordan, 2002). Schwartz

(1992) further stressed that the constituents in a composite material can be physically identified and exhibit an interface between one another. A more detailed description of a composite material was given by Agarwal *et al.* (1981) who defined a composite as a material that consists of one or more discontinuous phases which are usually hard and strong embedded in a continuous phase. The continuous phase is called the matrix while the discontinuous phase is termed the reinforcement material.

2.1.3 Classifications of Composite Materials

Based on the definition of a composite material, composites can be produced by any combination of two or more materials which can be metallic, organic or inorganic. Schwartz (1992) cited that the most widely used constituent forms in a composite material are fibers, particles, laminae or layers, flakes, fillers and matrixes.

Generally, composite materials are classified based on the morphology of reinforcement and also on the matrix material. Classifications of composite materials according to the reinforcement forms are particulate reinforced composites, fiber reinforced composites and structural composites. Particles by definition are non-fibrous in nature and have roughly equal dimensions. Common shapes of particles used as reinforcements in composites are spherical, cubical, tetragonal, platelet or of other regular or irregular shapes (Agarwal, 1981 and Mariatti, 1998). Fiber reinforced composites are composed of reinforcing fibers which are a characterized as a long fine filament with an aspect ratio of greater than 10 . Glass, carbon, aramid, boron and cellulose fibers are widely used as reinforcement in composite materials. Structural composites though, consist of laminate and sandwich composites, which are used in structural engineering applications.

In addition, classification of composite materials based on the type of the matrix can be grouped into there main categories such as metal matrix composites (MMC), polymer matrix composites (PMC) and ceramic matrix composites (CMC) (Schwartz, 1992; Hull *et al.*, 1996 and Reinhart, 1987). According to Hull *et al.* (1996) most composites in industrial use are based on polymeric matrices. The focus of this research is mainly on polymer matrix composite.

2.2 Fiber Reinforced Plastic Composites

Fiber reinforced plastic composites or commonly known as FRP are now competing with traditional materials such as steel, wood, aluminum, and concrete in various engineering applications. Fiber reinforced plastics composites have found applications in automobiles, boats, aircrafts and as construction materials. This is because unlike the conventional materials mentioned above, fiber reinforced plastic composites have excellent specific mechanical properties (high strength to weight ratio), corrosion resistance and are low cost. The properties of a fiber reinforced plastic composites are mainly governed by the fiber, matrix and interface. Thus, the following chapter would deal in detail with the functions and characteristics of these components.

2.2.1 Matrices

Matrix can be easily defined as a material where the reinforcing system of a composite is embedded. The matrix serves as a binder which holds the reinforcing materials in its place. Besides that, when a composite is subjected to an applied load, the matrix deforms and transfers the external load uniformly to the fibers (Astrom, 1977; Mariatti, 1998). The matrix also provides resistance to crack propagation and damage tolerance owing to the plastic flow at crack tips (Shwartz, 1992). Furthermore, the matrix also functions to protect the surface of fibers from adverse environmental effects and abrasion especially during composite processing.

Plastic matrices can generally be classified into two major types which are thermoplastics and thermosets. The selection criteria of the matrices depend solely on the composite end use requirements. For example, if chemical resistance together with elevated temperature resistance is needed for a composite material then thermoset matrices are preferred than thermoplastics. Whereas, if a composite material with high damage tolerance and recyclability is needed then thermoplastics are preferred.

2.2.1.1 Thermoset

Thermoset resins are usually liquids or low melting point solids in their initial form. This liquid resin is then converted to a hard rigid solid by chemical cross-linking through a curing process which involves the application of heat and the addition of curing agents or hardeners. Once cured, a tightly bound three dimensional network structure is formed in the resin and hence the resin cannot be melted, reshaped and reprocessed by heating (Hull *et al.*, 1996 and Matthews *et al.*, 1999). Therefore, during composite manufacturing, the impregnation process followed by the shaping and solidification should be done before the resin begins to cure (Mariatti, 1998). Thermoset resins are brittle at room temperature and have low fracture toughness. On the other hand, owing to its three dimensional cross linked structure, thermoset resins have high thermal stability, chemical resistance, high dimensional stability and also high creep properties (Matthews *et al.*, 1999 and Shwartz, 1992).

Among the most common thermosetting resins used in composite manufacturing are unsaturated polyesters, epoxies, vinyl esters and phenolics. In this research, epoxy resin was preferred over unsaturated polyester resins which are used widely in composite industries. This is because epoxy resins exhibit better structural characteristics such as high stiffness and strength properties and have also excellent water and chemical resistance, good thermal stability, good adhesion to a substrate and low shrinkage during cure (Matthews *et al.*, 1999; Hull *et al.*, 1996; Dominick, 1997 and Shwartz, 1992).

2.2.1.2 Thermoplastic

Thermoplastic resins are linear or branched polymers which remain as a solid at room temperature. Unlike thermosets, thermoplastics do not form a three dimensional cross linking network. The monomer units in the thermoplastic are held by a weak Van der Wall's forces which are easily broken by heat and stress (Dominick, 1997). As a result, thermoplastics are able to melt when heated and becomes a solid when cooled to room temperature.

Thermoplastics can be classified into two classes which are commodity plastics and engineering plastics. Commodity plastics such as polyolefins, styrenics, acrylics and vinyls possess moderate mechanical and thermal properties. Thus, they are normally used in applications with less service requirements. Engineering plastics though are able to perform at elevated temperatures and under high load bearing. Examples of engineering plastics are ABS, PBT, Nylon, PEEK, PEK and PPS. The costs of engineering plastics are much higher than the commodity plastics.

Thermoplastic resins offer several advantages over their thermoset resin counterpart. Thermoplastic products can be recycled owing to the nature of the thermoplastics which can be repeatedly heated and shaped (Ruzaidi, 1999 and Matthews *et al.*, 1999). In addition, thermoplastics offer improved fracture toughness and low moisture absorption behavior. The processing time of thermoplastics is shorter than most thermosets as the processing of thermoplastics only involve melting, shaping and cooling which can be achieved in a matter of few seconds whereas thermosets would take several hours to days to fully crosslink (Astrom, 1977 and Leong, 2003). Moreover, imperfect thermoplastic products can be reprocessed and flash or unused thermoplastics can be used for other applications.

However, unlike thermoset resins, thermoplastics have low thermal and chemical resistance. Therefore, thermoset resins mainly dominate as matrices in structural composite applications. Table 2.1 summarizes some typical properties of thermoset and thermoplastic resins.

Table 2.1: Some typical properties of thermoset and thermoplastic resins

	Epoxy	**Polyester**	**Nylon (6.6)**	**Polypropylene**
Density (g/cm^3)	1.1-1.4	1.1-1.5	1.1	0.9
Tensile Strength (MPa)	35-90	45-85	60-70	25-38
Young’s Modulus (GPa)	2.1-6.0	1.3-4.5	1.4-2.8	1.9-1.4
Strain at break (%)	1-6	2	40-80	300

2.2.2 Fiber Reinforcement

Reinforcement is the disperse phase in a composite material and its primary function is to carry the structural load subjected on the composite. Dominick, (1997) added that besides carrying the structural load, reinforcements also function to increase the strength and stiffness to density ratio of a composite, increase the resistance to corrosion, fatigue, creep, stress rupture and reduce the coefficient of thermal expansion of composite materials. Furthermore, fibers are able to retard the propagation of cracks and hence increasing the toughness of the composite material.

Based on previous literatures, it is noted that reinforcing fibers can be classified according to their origin, length, physical structure and chemical structure (Dominick, 1997; Lilholt and Lawther, 2002 and Fried, 1995). For the sake of simplicity and better understanding, classification of fibers according to their origin is adopted in this study. As shown in Figure 2.1, reinforcing fibers can be classified into natural fibers and synthetic fibers. Natural fibers which are obtained from natural resources include animal fibers, lignocellulose fibers and mineral fibers. Lignocellulose fibers will be discussed in detail in the following section (refer to section 2.4). Silk, wool and hair are examples of animal fibers which are basically made up of protein and keratin (Fried, 1995). Asbestos which is a fibrous mineral found in rocks, is the most widely used mineral fiber in composite materials. This is because asbestos fibers are low cost and have also superior strength and modulus. However since asbestos fibers are hazardous to the health, the usage of asbestos in composites have been significantly reduced (Dominick, 1997).

Synthetic fibers are man made fibers which can be further categorized as organic fibers and inorganic fibers. Organic fibers include aramid (eg. Kevlar), polyethylene (eg. Spectra and Dyneema), polypropylene, polyester and polyacrylonitrile. Glass, carbon, alumina and boron are examples of inorganic fibers which are more established in composite materials than organic fibers.

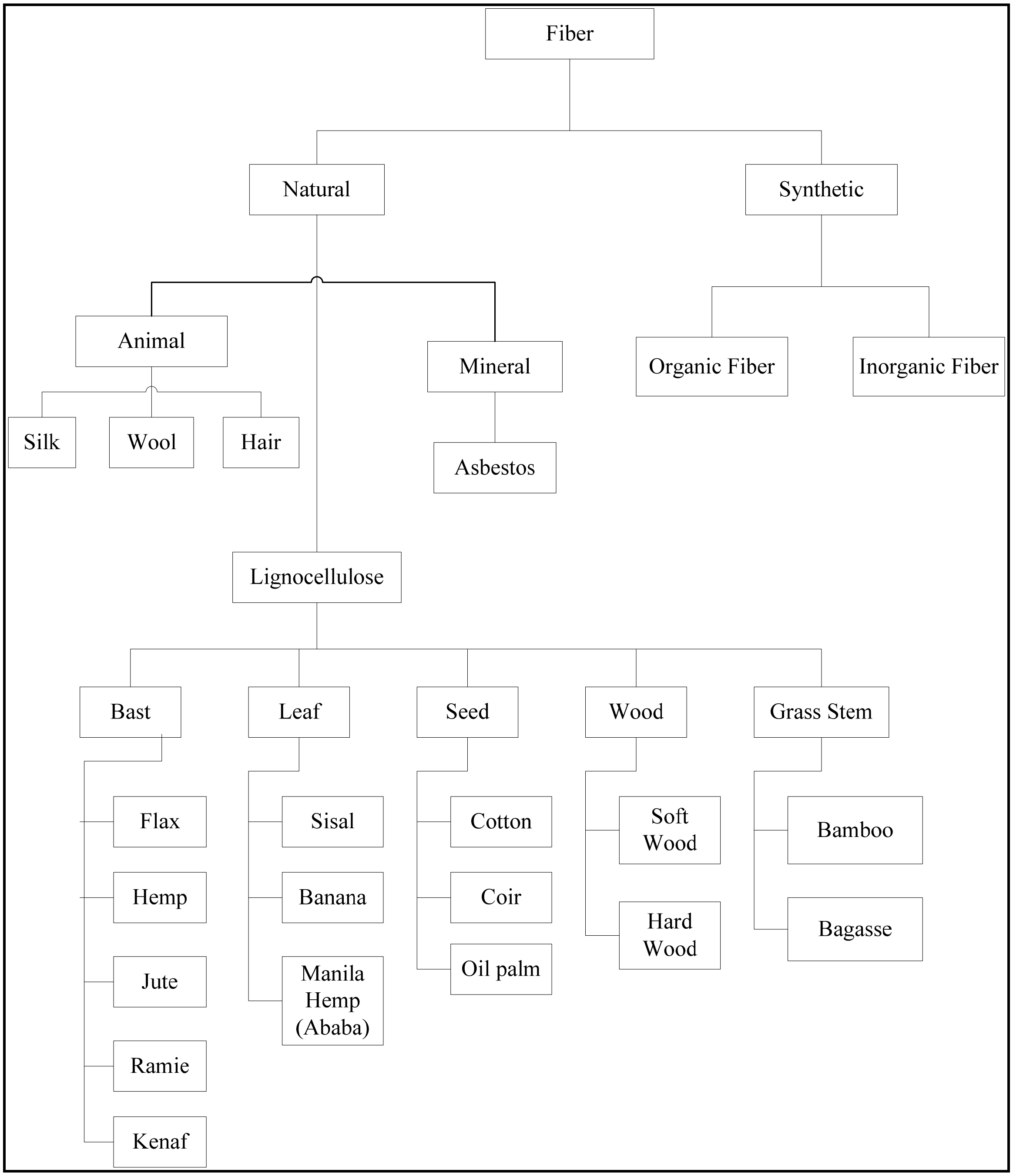


Figure 2.1: Classifications of fibers (Lilholt and Lawther, 2002)

Glass fibers are extensively used as reinforcements in plastic materials and according to Dominick (1997), glass fibers represent about 90 wt.% consumption of all reinforced fibers used in plastic materials world wide. The major ingredient of glass fibers is silica (SiO_2). Glass fibers have good mechanical properties such as high strength and good impact properties, chemical resistance, good electrical and thermal insulating properties and the cost of production is low compared to other synthetic fibers (Astrom, 1977; Fried, 1995 and Matthews *et al.*, 1999). There are several different types of glass fibers available in the market and the most common ones are the E, A, S, and C glass fibers. The "E" glass fibers are the general purpose grade glass fibers and have excellent electrical and durability properties. The "S" glass fibers offer high strength and modulus and heat resistance. The "C" type of glass fibers though, exhibit high chemical resistance while the "A" glass fibers exhibit high alkali resistance. Therefore, the selection of the type of glass fibers needed is mainly dependent on the end use of the composite material.

2.2.2.1 Reinforcement Forms

Fiber reinforcements are available in a variety of forms to serve a wide range of processes and end product requirements. Reinforcement forms include rovings, chopped strands, mats and fabrics. The characteristics of the reinforcement forms are discussed below:

(a) Rovings

Rovings consist of many individual continuous fiber strands wound into a spool. Fiber rovings are mainly used in continuous composite molding processes such as filament winding and pultrusion. Prepregs can also be formed by impregnating the rovings with a polymeric resin (Mallick, 1988; Jones, 1994 and Norwood, 1994).

(b) Chopped strands

Chopped strands or also known as chopped fibers are produced by cutting the continuous fiber rovings into shorter lengths. The fiber length ranges from 3.2 to 12.7mm. Chopped strands are used in injection molding process.

(c) Mats

Fiber mats which are known as non-woven fabrics are the most commonly used fiber form in composite manufacturing. Chopped strand mat and continuous strand mat are two distinct forms of fiber mats. Chopped strand mats (CSM) are produced by dispersing uniformly chopped fibers with an average length of about 25mm together with a polymeric binder onto a thin film. These mats provide equal strength in all directions and is used primarily for hand-lay up processing (Mallick, 1988 and Astrom, 1977). Continuous strand mats though, are formed by swirling continuous fiber strands in a moving film and then applying a polymeric binder to hold the mat together. Continuous strand mats are now finding usage in resin transfer molding (RTM) and matched die-molding (Norwood, 1994).

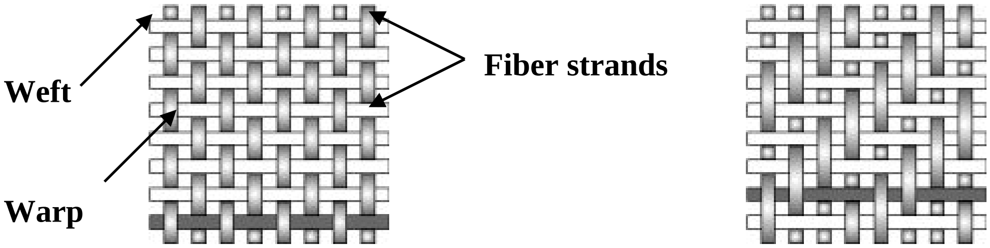
(d) Fabrics

Fabrics are made up of long continuous fibers which are oriented along two perpendicular directions (Gay, 2003). These fibers are held together by mechanical interlocking of the fiber themselves or by a polymeric binder. Fabrics exist in many different forms such as woven fabrics, unidirectional fabrics, hybrid fabrics, braided fabrics and knitted fabrics. However, in this study the nature and properties of woven fabrics would be discussed.

Woven fabrics are formed by interlacing two or more fiber strands in the warp (longitudinal) and weft (transverse) directions in a weave style (Astrom, 1977, Mariatti, 1998). Woven fabrics provide enhanced mechanical properties in the 0^0 and 90^0 directions.

The weave style in a woven fabric influences the formability, surface smoothness and fabric stability. The commonly used woven fabric weave styles are plain weave, twill weave, satin weave and basket weave (refer to Figure 2.2). Among these weave styles, plain weave fabric is widely used in general composite applications owing to its easy handling characteristics.

On the whole, the selection of a fiber form is mainly dependent on the type of composite processing and also on the end use of the composite product



(a) Plain weave **(b)** Twill weave

(c) Satin weave **(d)** Basket weave

Figure 2.2: Schematic diagram of different woven fabric styles

2.2.3 Fiber Matrix Interface

Besides the properties fiber and matrix, the fiber-matrix interface which results from the interaction of the matrix and the surface of the reinforcing fibers, plays an important role in determining the properties of a composite material. According to Chawla (1987), the interface consists of surface layers of fiber and matrix and any layers of material existing between these surfaces. He also explained that the internal surface area occupied by the interface in a composite containing a reasonable fiber volume fraction is quite extensive which is about 3000 cm^2/cm^3.

The main function of an interface is to transfer effectively the applied load on the composite from the matrix to the fibers. The interface also protects the fiber surface from environmental degradation (Chawla, 1987; Astrom, 1977, Mariatti, 1998). The ability of the interface to act effectively as a load transferring medium between the matrix and fibers is dependent on the fiber-matrix interfacial bonding or adhesion. A strong interfacial adhesion would enhance the interlaminar shear strength, delamination resistance, fatigue properties and corrosion resistance of a composite (Astrom, 1977). However, strong interfacial adhesion would result in low damage tolerance properties of the composite material. The adhesion between the fiber and matrix occurs through various mechanisms such as chemical adhesion, mechanical adhesion, interdiffusion and electrostatic attraction (Astrom, 1977; Chawla, 1987 and Mariatti, 1998). Therefore, by understanding these fiber-matrix adhesion mechanism, the degree of adhesion between the fiber and matrix could be controlled in order to obtain the desired properties in a composite material.

2.3 Lignocellulose Fiber Composite

Among the natural fibers mentioned in the earlier section, lignocellulose fibers are used widely in fiber reinforced plastic composites. Lignocellulose fibers are derived from plants and their main chemical component is cellulose.

2.3.1 Classification of Lignocellulose Fibers

As seen in Figure 2.1, lignocellulose fibers can be classed according to the their source in plants such as bast fibers, leaf fibers, seed fibers, wood fibers and grass stem fibers (Rowell, 1995; McGovern, 1987 and Lilholt and Lawther,2002).

Bast fibers or also known as stem fibers are from the inner bark of the plant stem and extend along the length of the stem. These fibrous strands serve to strengthen the stem of plants. Bast fibers are multicelled in structure which consist of a number of single fibers bundled together. Examples of bast fibers are jute, flax, hemp, kenaf and ramie (McGovern, 1987 and Lilholt and Lawther, 2002).

Leaf fibers include sisal, henequen, abaca, pineapple, banana and Manila hemp. Leaf fibers extend longitudinally of the length of the leaf and contribute to the strength of the leaf. Leaf fibers are also multicelled fibers similar to bast fibers.

Seed fibers such as cotton, coir, oil palm and kapok are obtained from the fruits of the plants. Among the seed fibers, cotton is the most famous fiber and is used in the textile industry all over the world. According to McGovern (1987), all seed fibers are single celled fibers. However in a recent work, Rozman *et al.* (2001) concluded that oil palm fibers do exist as fiber bundles.

Wood fibers are found from the bark of a tree. Wood fibers can be divided into hardwood fibers which are from hardwood trees and softwood fibers which are from softwood trees. Wood fibers are singled cell fibers and are short fibers compared to the long bast and leaf fibers (Lilholt and Lawther, 2002).

Grass fibers include bamboo, bagasse, cereal straw and reed canary grass. The most widely used grass fibers as reinforcement in a composite material are the bamboo fibers. Most grass fibers are short fibers similar to wood fibers (Moran, 2003).

2.3.2 Chemical Composition of Lignocellulose Fibers

The basic chemical components of a lignocellulose fiber are cellulose, hemi-cellulose, lignin and pectin. Pectin is usually not found in wood tissues because the secondary wall thickening replaces almost all of the pectin with lignin (Lilholt and Lawther, 2002). Waxes and water soluble substances are also found in small amounts in the cell wall of a lignocellulose fiber. The chemical compositions of a lignocellulose fiber vary according to the species, growing conditions, method of fiber preparations and many other factors (Bledzki *et al.*, 1999). Chemical compositions of lignocellulose fibers are shown in Table 2.2

Table 2.2: Chemical composition of some common lignocellulose fibers (Bledzki *et al.*, 1999; Sreekala *et al.,* 1997)

Fibers	Cellulose(%)	Hemicellulose(%)	Lignin(%)	Pectin(%)
Oil palm empty fruit bunch fiber(EFB)	19	-	19	2
Coir	32-43	0.15-0.25	40-45	-
Banana	63-64	19	5	-
Sisal	66-72	12	10-14	0.8
Jute	64.4	12	11.8	4-10
Pineapple	81.5	-	12.7	
Flax	71.2	18.6	2.2	5-12

2.3.2.1 Cellulose

Cellulose is the main component in lignocellulose fibers and is the reinforcing material within the cell wall. Cellulose is a linear crystalline condensation polymer consisting of D-anyhydroglucopyranose units held together by β-1,4-glycosidic bonds (see Figure2.3). Cellulose is a high molecular weight homopolymer of glucose and it is laid down in microfibrils where extensive hydrogen bonding between the cellulose chains produces a strong crystalline structure (Lilholt and Lawther 2002). According to Bledzki *et al.* (1999) and Daniel (1985), cellulose can be characterized as cellulose I, cellulose II, cellulose III and cellulose IV based upon their physical crystal structure. Furthermore, the mechanical properties of lignocellulose fibers depend on the type of cellulose whether it is cellulose I or cellulose II because each type of cellulose has its own geometrical conditions, which influences the mechanical properties.

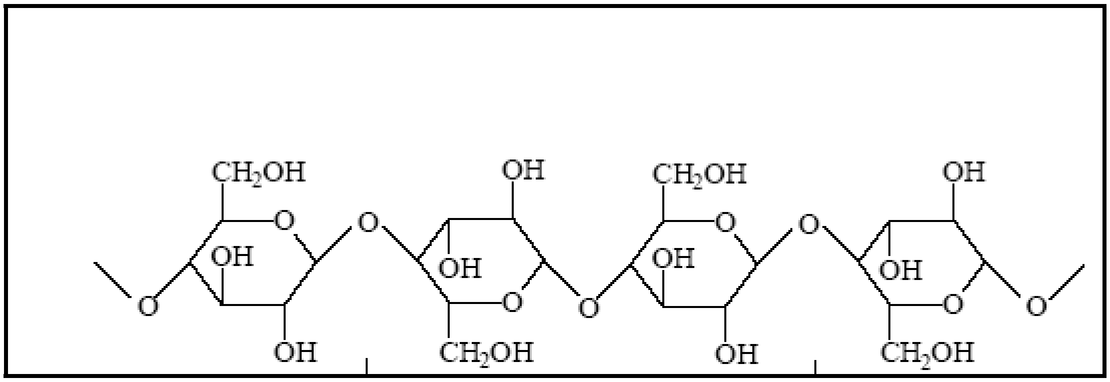


Figure 2.3 Molecular structure of cellulose

2.3.2.2 Hemicellulose

Hemicellulose is not a form of cellulose and is a copolymer of a group of polysaccharides consisting of glucose, mannose, xylose, galactose and arabinose. Unlike cellulose, hemicellulose is of low molecular weight, amorphous and exhibits chain branching. Owing to its amorphous morphology, hemicellulose is partially soluble in water. Besides that, the constituents of hemicellulose vary from plant to plant (Nevell, 1985).

2.3.2.3 Lignin

Lignin, which is generally regarded as an adhesive in the cell wall, is a hydrocarbon polymer consisting of aliphatic and aromatic components (Nevell, 1985 and Bledzki *et al.*, 1999). The structure of lignin is shown in Figure (2.4). Lignin has a disordered structure and is formed through ring opening polymerization of phenyl propane monomers. Lignin also provides rigidity, hydrophobicity and decay resistance to the cell wall of lignocellulose fibers.

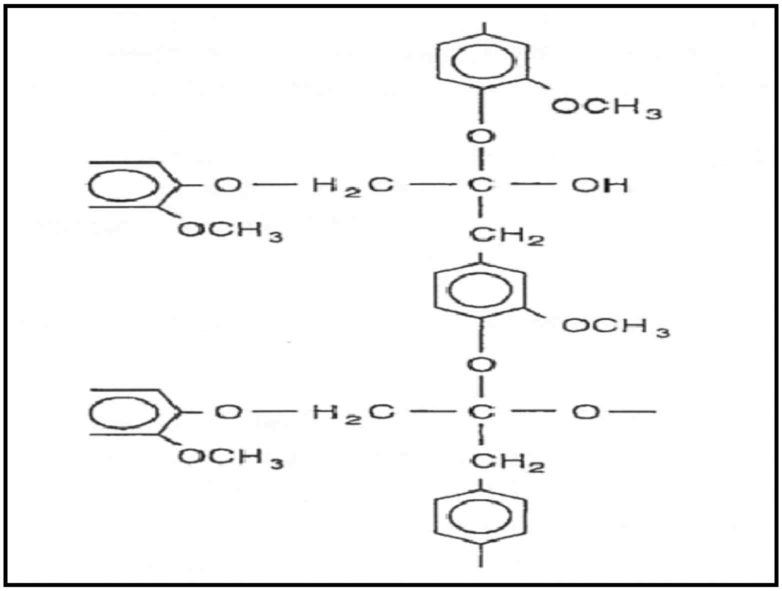


Figure 2.4: Molecular structure of lignin

2.3.2.4 Pectin

Pectin is a polysaccharide consisting polygalacturon acid. Lilholt and Lawther (2002), cited that pectin is the major matrix component within the cell wall of lignocellulose fibers especially in non-wood fibers.

2.3.2.5 Waxes

Waxes consist of different type of alcohols which are insoluble in water and also in certain acids such as palmitic acid, oleaginous acid and stearic acid. They can only be extracted from the fibers by using organic solvents.

2.3.3 Physical and Mechanical Properties of Lignocellulose Fibers

The diameter of lignocellulose fibers normally varies in the range of 0.015×10^4 to 0.05×10^4 μm. The densities of lignocellulose fibers are in the range of 1.25 to 1.55 g/cm^3. The mechanical properties of lignocellulose fibers are summarized in Table 2.5. Based from the Table it could be observed that bast and leaf fibers are the strongest among the lignocellulose fibers with a high tensile strength and modulus of elasticity. However, owing to their low extensibility, bast and leaf fibers have poor toughness properties compared to seed fibers such as oil palm fiber and coir fibers. According to Sreekala *et al.* (1997) and Bledzki *et al.* (1999), the mechanical properties of lignocellulose fibers depend on the fibrillar structure, spiral angle of the micro-fibrils and the cellulose content.

The relationship between the strength of the lignocellulose fibers with the microfibrillar angle and cellulose content is given by equation 2.1

$$\sigma = -334.005 - 2.830\theta + 12.22W \qquad (2.1)$$

where σ is the fiber strength, θ is the microfibrillar angle and W is the cellulose content. Furthermore, the elongation at break ε can be correlated with the microfibrillar angle θ based on equation 2.2 below :

$$\varepsilon = -2.78 + 7.28 \text{ x } 10^{-2}\,\theta + 7.7 \text{ x } 10^{-3}\,\theta \qquad (2.2)$$

Table 2.3: Mechanical and physical properties of lignocellulose fibers and synthetic fibers (Sreekala *et al.,* 1997; Bledzki *et al.,* 1999 and Lilholt and Lawther, 2002)

Fibers	Density	Stiffness(GPa)	Strength(MPa)	Strain(%)
Glass fibers	2.56	72	3530	4.8
Carbon	1.4	235-827	2200-4410	0.27-1.5
Oil palm empty fruit bunch fibers	0.7-1.55	2.0	248	14
Flax	1.5	27.6	345-1035	2.7-3.2
Jute	1.3	26.5	393-773	1.5-1.8
Sisal	1.5	9.4-22.0	511-635	2.0-2.5
Banana	1.4	7-20	500-700	1-4
Pineapple	1.44	35-80	400-1600	0.8-1.6
Softwood	1.4	10-50	100-170	-
Hardwood	1.4	10-70	90-180	-

According to Bledzki *et al.* (1999), high cellulose content and smaller spiral angle would result in the increase of the lignocellulose fiber strength. On the other hand, Lilholt and Lawther (2002), stressed that the strength of lignocellulose fibers are strongly affected by various factors such as the structure of the fibers, orientation of molecular chains, imperfection or defects in the fibers and the degree of polymerization.

The regions with non-crystalline structure such as the amorphous regions in the cell wall of the lignocellulose fibers are considered as weak points owing to low number of chains per cross sectional area which are unable to withstand the stresses effectively. The orientation of the crystalline components influences the strength of the fibers. High strength fibers are normally achieved with alignments within about 5^0 of perfect orientation. The imperfection in the fiber structure such as the presence of fiber bundles which results in non uniform stress distribution, traces of oil especially found on oil palm fibers and cracks on the fiber surface reduces the strength of the lignocellulose fibers. The strength of highly oriented lignocellulose fibers are affected by the degree of polymerization. Lilholt and Lawther (2002) also explained that the strength of the fibers is inversely proportional to the degree of polymerization.

2.3.4 Oil Palm Empty Fruit Bunch Fiber and Its Composites

Oil palm empty fruit bunch fibers (EFB) which are derived from an oil palm tree (*Elais guineensis*) component namely the empty fruit bunch were used as a reinforcement in this study rather than other lignocellulosic fibers because oil palm trees are abundant and are also an important commercial plant in Malaysia. The total area of oil palm plantation in Malaysia is about 2.5 million hectares (Mohammad *et al.*, 2000). The oil palm fruits are processed in mills and crude palm oil is extracted from these fruits. The oil palm industry in Malaysia produces about 10.5 million tones of crude palm oil per annum and produces a massive amount of biomass waste. One of the bio mass waste produced is the empty fruit bunches which are left behind after removal of oil palm fruits for the oil refining process at the oil refineries. Based on a report by Tanaka (2003), 16 million tons per annum of empty fruit bunches were discharged from each oil palm refinery in the country regularly in the year 2000. The empty fruit bunches are then used as boiler feedstock in the oil mill and are also left to mulch and degrade as soil fertilizers in the field while the majority of the empty fruit bunches are unutilized. According to Kim (2003), the oil palm empty fruit bunch which are disposed in the oil palm estates as soil fertilizers, takes a long time to break down and hence during the rainy season it provides an ideal condition for fungi to grow which is the main cause of ganoderma disease. Therefore by using the oil palm empty fruit bunch fibers which were extracted from the empty fruit bunches as a reinforcement in composite materials, the biomass waste generated by the oil palm industry can be reduced significantly.

The usage of oil palm empty fruit bunch fibers as a reinforcement in composite materials have gained interest among scientists especially those in the South East Asia region where oil palm is a major industrial cultivation. Pioneer work on the oil palm fibers were done by Sreekala *et al.* (1997). They conducted a thorough study on the morphology, chemical composition, surface modifications and mechanical properties of oil palm fibers alone. The thermal behavior of oil palm fiber reinforced phenol-formaldehyde composites were studied by Agarwal *et al.* (2000). They reported that surface modifications of oil palm fibers by alkali, potassium permanganate and peroxide treatments enhanced the thermal stability of the lignocellulose composites. Moreover, composites treated by peroxides exhibited the highest thermal stability compared to the other chemical treatments.

Hill and Abdul Khalil (2000a) who studied the effect of acetylation and the usage of silane and titanate coupling agents on the mechanical properties of oil palm fiber reinforced polyester composite, concluded that chemical treatment of the fibers improved the mechanical properties of the composites. Furthermore, in a separate study, Hill and Abdul Khalil (2000b) reported that acetylation of the oil palm fibers resulted in good retention of mechanical properties of the oil palm fiber composite during soil and water exposure tests. Recently, Abdul Khalil *et al.* (2002a) reported that, oil palm flour filled polyester composite offered better mechanical and chemical resistant properties compared to calcium carbonate filled polyester composite.

Oil palm empty fruit bunch filled thermoplastic composites were studied by Rozman *et al.* (1998). In their study, they observed that as the oil palm empty fruit bunch filler loading increased in the high density polyethylene, the modulus of elasticity and modulus of rupture of the composite increased and decreased respectively. The tensile and impact properties were found to decrease with the increasing oil palm empty fruit bunch filler in the composite. In addition, smaller sized empty fruit bunch filler particles displayed higher modulus of elasticity and modulus of rupture compared to larger filler particle size. In order to improve the compatibility between lignocellulose fibers and polypropylene matrix, Rozman *et al.* (2001a) used hexamethylene diisocyanate (HMDI) modified lignin as coupling agents in oil palm empty fruit bunch filled polypropylene composite and observed a significant increase in the flexural strength of the composite. Besides working on thermoplastic composites, Rozman and co-workers (2001b) also produced oil palm empty fruit bunch-polyurethane composites by reacting the oil palm fibers and polyethylene glycol (PEG) with diphenylmethane diisocyanate (MDI). It was found that the tensile properties were influenced by the percentage of OH groups of the oil palm fibers together with the reinforcing effect of the oil palm fibers.