



Review Article

A Review: Fibres, Polymer Matrices and Composites

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ABSTRACT

The growing interest, environmental consciousness and high performance demands on engineering have led to extensive research and development of new and improved materials. Among the most commonly used natural fibres are kenaf, oil palm, sugar palm, pineapple leaf fibre, flax, hemp, sisal, coir and jute. These fibres are used to reinforce thermoplastic polymer matrices such as polystyrene (PS), polypropylene (PP), polyethylene (PE) and polyvinyl chloride (PVC). Meanwhile, phenolic, unsaturated polyester vinyl ester and epoxy resin are for thermosetting polymer matrices. The objective of this paper is to solicit works that cover major class of natural fibres, thermosetting polymers matrices, which detail about unsaturated polyester resin and hybrid biocomposites industry.

Keywords: Natural fibres, modification, polymer matrices, unsaturated polyester, hybrid biocomposites

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INTRODUCTION

Composite is the combination of two or more elements in any form and for a variety of uses. The concept of composite materials is simply that the action of combining different materials often produces a new material with superior performance beyond that exhibited by their individual constituents. The applications of composite materials have grown steadily throughout the years, penetrating and conquering newer markets nowadays.

Modern and established composite materials constitute a significant proportion of these engineered material markets ranging from daily products to sophisticated niche applications. These advanced materials found usage in construction, military, automotive and aerospace industries. Their technologies are especially attractive due to their advantage over most existing materials like metal, as it possesses high specific strength, low density, light weight, enhanced corrosion and temperature capability. An added advantage, which is also its contribution to nature, is being a biodegradable and green product (Ishak et al., 2013; Nair et al., 1996).

Synthetic fibre-like glass, aramid and carbon fibre are widely used as reinforcement fibres in composites, which have been proven to be good mechanical strength enhancer. One disadvantage of synthetic fibre is that without proper handling, it may cause skin irritation causing it to be dangerous to human health. The desired tensile strength and young modulus of glass fibre are visibly much higher than that of the natural fibres. However, the difference in the characteristics of glass and natural fibres is considered as the most important when their applications and costs are taken into account (Bledzki & Gassan, 1999).

Natural fibre, as a reinforcement fibre, has become a popular alternative due to their abundance and significant performance in term of strength and stiffness enhancement, apart from being readily available from renewable sources and producible with low investments, low wear of tooling and skin irritation, environmental-friendly, and may act as a good substitute to synthetic fibre. Natural fibre composites are essentially a plant fibre embedded within a thermoset or thermoplastic polymer. The density of these natural fibres is similar as their plastic counterparts, which are usually 40–50% lower than the density of glass fibre (Rajan & Curtin, 2015; Saba et al., 2014). Therefore, polymeric materials could be reinforced or filled without having significant effects on their density. Finally, natural fibres, such as kenaf, oil palm, sugar palm, pineapple leaf, flax, hemp, sisal, coir and jute, have attracted the attention of most scientists and technologists for their widespread applications.

NATURAL AND SYNTHETIC FIBRES

Natural Fibre Sources

An increase in the demands for engineering materials has prompted enthusiastic broad research and development of new and improved materials especially from the composites industry. Since most natural fibres used today are being researched to the leading edge of material technology, the development enabling their use in advanced applications is becoming harder to be ignored.

Recently, there has been an expansion for research into creating new materials with high performance and desirable properties at an affordable cost from renewable agriculture based materials. This growth of concern on the utilisation of alternative sources of natural fibre was contributed by the increasing awareness towards eco-friendly, renewable and biodegradable materials. All these are for purpose of reducing dependency on non-renewable natural resources, as there is short supply of petroleum based polymer and to generate their replacement (Sahari et al., 2013).

The primary advantages of natural fibres over synthetic fibres are their abundance sources and relatively low cost, low mass and specific density, high specific strength, renewability and

biodegradability (Mohanty et al., 2002; Sahari et al., 2013). The present use of the biodegradable term in natural fibre composite actually refers to the utilisation of natural sources in the polymer industry, which reduces the dependency on petroleum resources and carbon emissions (CO₂) due to the decreasing need for plastic burning (Sahari et al., 2013).

In this cutting-edge technology era, the use of natural fibre composites offers an environmental gain opportunity via reduction of dependency on petroleum-based materials. Figure 1 offers an outlook on the application of natural fibre reinforcement in the automotive industry in Germany and Austria between 1996 and 2002. It was forecasted that the introduction of natural fibres into composites would then alter the global composites trend and serve as a better choice in appropriate construction material selection.

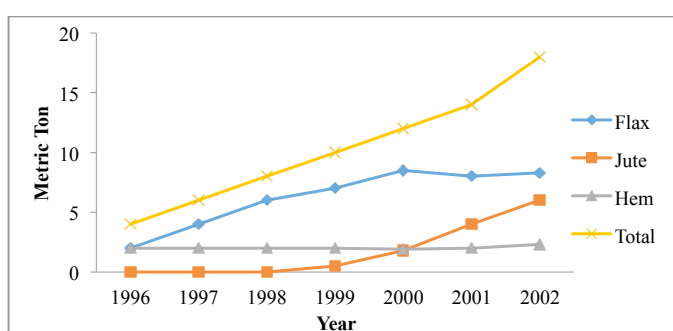


Figure 1. Utilisation of natural fibre reinforced composite for the automotive industry in Germany and Austria 1996 and 2003 (Karus et al., 2003)

The industrial use of natural fibres was not driven only by cost reduction, but also by other issues related to overall environmental awareness. In Europe, the EU “end-of-life vehicle” directive imposes that 85% of all vehicle component weight should be recyclable by 2005, which would later be increased to 95% by 2015 (Madsen & Lilholt, 2003). Mercedes Benz, with its own renovation engineering recently achieved around 20% of car weight reduction by implementing hybrid flax and sisal reinforced thermoset composites for their door panels (Mohanty et al., 2005). Figure 2 shows how natural fibres such as flax, hemp, sisal, wood and other natural fibres reinforced polymer composites are utilised in the automotive applications to produce up to 50 components of the Mercedes Benz E-class series, with the inner door made specially from kenaf reinforced PP composites.

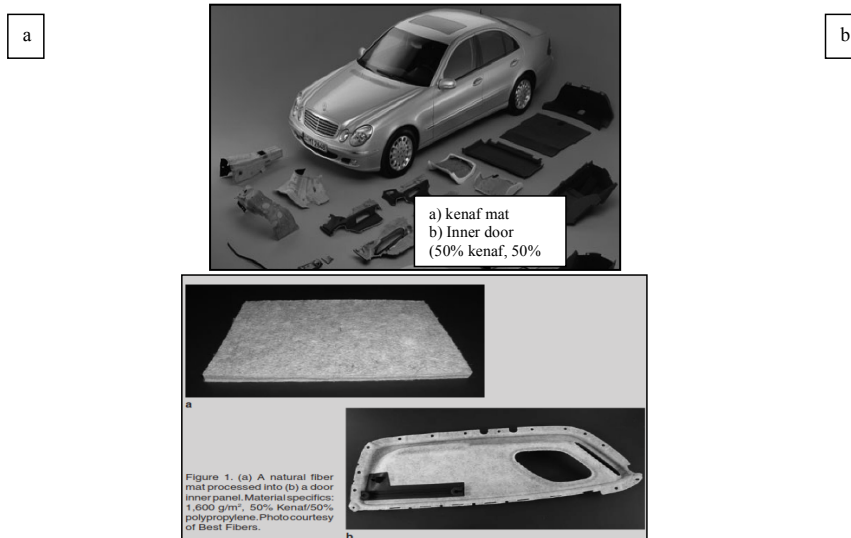


Figure 2. (a) Natural fibre reinforced composites in automotive applications, (b) inner door made of kenaf reinforced PP composites (Mohanty et al., 2005)

The major market identified for the applications of green composites are the replacement of fibre glass and steel in the automotive components. These are the trim parts in dashboards, door panels, parcel shelves, seat cushion and cabin linings. For example, flax fibres used in car disk brakes to replace asbestos fibres, and the use of hybrid kenaf, i.e. glass reinforced epoxy composites, as car bumper beam (Bismarck et al., 2006; Davoodi et al.; 2010).

In addition, natural fibre composite has been used for panelling materials in buildings, furniture components, moulded and pultruded sections for various other applications. The use of natural fibre composites is not limited to the automotive industry only. Natural fibre reinforced composites are being widely used in a large number of applications such as for the construction and aerospace industries. Figure 3 shows the percentage of natural fibres usage in different applications.

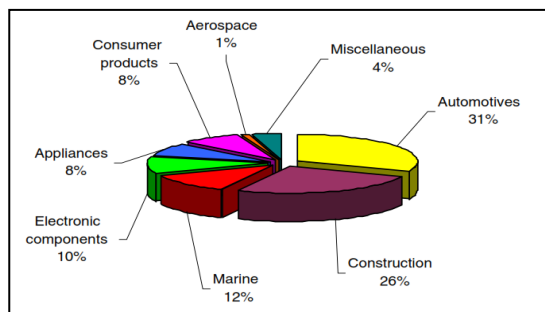


Figure 3. The use of natural fibre reinforced plastic composites in 2002
Sources: Ashori, 2008; John & Thomas, 2008

The physical and mechanical properties of these natural fibres are determined by their chemical and physical composition such as the structure of the fibres, cellulose content, their inherent microfibrillar angle and cross section and degree of polymerisation. Swelling of the fibres, due to moisture absorption, has been a major drawback for natural fibres, causing a weak bonding to the fibre-matrix interaction in the composites (Cheung et al., 2009).

Classification and Limitations of Natural Fibres

Fibres can be classified into two main groups; natural and man-made. A diagram with a classification of various fibres from plants, animals, minerals and synthetic fibres is shown in Figure 4. Currently, most industries utilise fibre glass, which is also known as man-made fibre (synthetic fibre) as the reinforcement in composite. A recent study, however, has examined the development of natural fibres such as kenaf, oil palm, sugar palm, pineapple leaf, flax, hemp, sisal, coir and jute as the alternatives for conventional reinforcement materials (Saba et al., 2014).

According to Gurunathan et al. (2015), all plant fibres are composed of cellulose, while animal fibres consist of proteins (hair, silk and wool). Plant fibres includes bast (or stem or soft sclerenchyma) fibre, leaf or hard fibre, seed, fruit, wood, cereal straw and other grass fibres. The plant fibres have three major compositions known as cellulose, hemicellulose and lignin. Meanwhile, plant fibres also contain minor components consisting of pectin, waxes and water-soluble substances (Bledzki et al., 1999). Faruk et al. (2012) have listed the chemical compositions of cellulose, hemicellulose, lignin and wax content as having some common natural fibres presence.

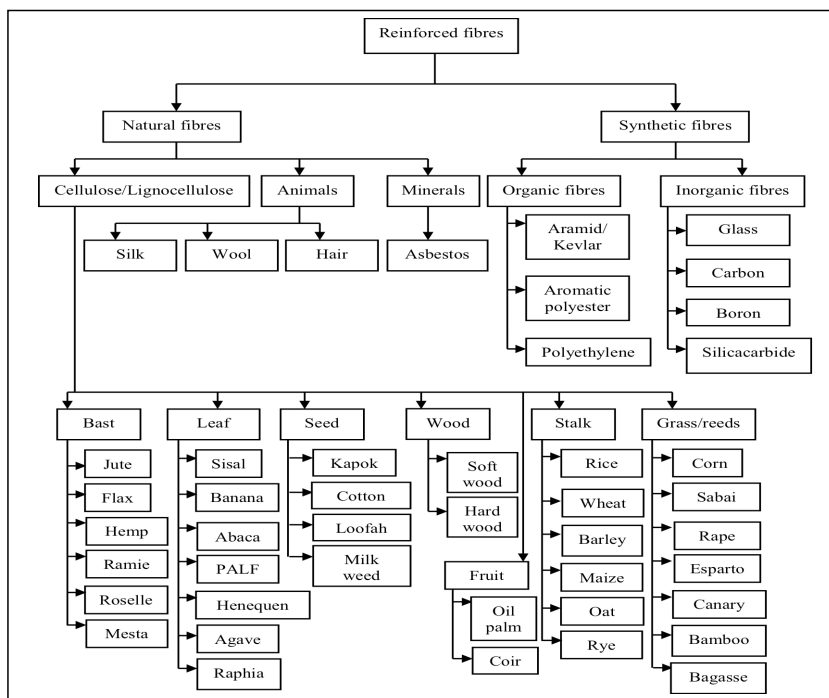


Figure 4. Classification of natural and synthetic fibres (Gurunathan et al., 2015)

In tropical countries like Malaysia, fibrous plants are available in abundance; and at least some of them are agricultural crops. Table 1 compares the mechanical properties of different natural fibres with their synthetic fibre counterpart. The tensile strength, elongation at break and young modulus of the single fibres depend on the chemical compositions, size, shape, orientation and thickness of their cell walls. Figure 5 shows the factor influencing performance of natural fibre reinforced composites.

Table 1
The physical and mechanical properties of selected natural fibres and synthetic fibres (Ishak et al., 2013; Satyanarayana et al., 2009; Koronis et al., 2013; Ku et al., 2011)

Fibres	Density (g/cm ³)	Tensile strength (MPa)	Elongation at break (%)	Young Modulus (GPa)
Sugar Palm	1.29	15.5-290	5.7-28.0	0.5-3.37
Bagasse	1.5	290	-	17
Bamboo	1.25	140-230	-	11-17
Flax	0.6-1.1	345-1035	2.7-3.2	27.6
Hemp	1.48	690	1.6-4	70
Jute	1.3	393-773	1.5-1.8	26.5
Kenaf	1.45	930	1.6	53
Sisal	1.5	511-535	2.0-2.5	9.4-22
Ramie	1.5	560	2.5-3.8	24.5
Pineapple	0.8-1.6	400-627	14.5	1.44
Coir	1.2	175	30	4-6
E-Glass	2.5	2000-3500	0.5	70
S-Glass	2.5	4570	2.8	86
Aramid	1.4	3000-3150	3.3-3.7	63.0-67.0

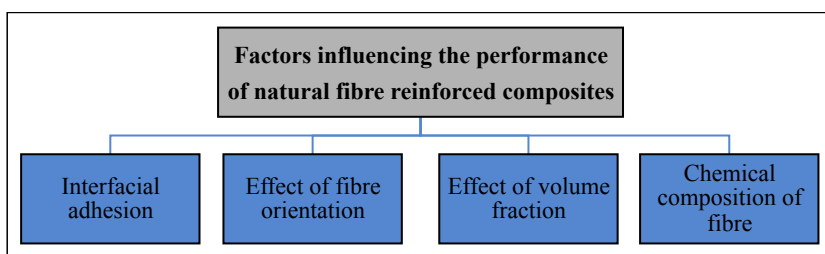


Figure 5. Factors influencing the performance of biocomposites (source: Shalwan & Yousif, 2013)

According to Ishak et al. (2011), one of the limitations of natural fibre is its hydrophilic nature due to the presence of hydroxyl (OH) group throughout the structure especially for cellulose (repeated unit of glucose in plants) and hemicellulose portions. When natural fibre is exposed to high humidity environment, these hydroxyl groups attract water molecules by chemical interaction called hydrogen bonding. Figure 6 shows the cellulose structure that is rich with OH group (inter-fibrillar region of the fibre, hemi-cellulose and lignin).

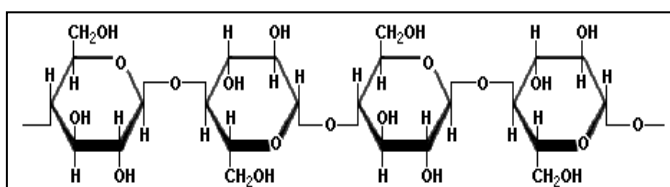


Figure 6. Cellulose structure (Brown, 2004)

Besides the aforementioned risk, an additional risk of using plant fibres is the poor compatibility between fibres and polymer matrix, which results in a non-uniform dispersion formation of fibres within the matrix, and thus generates poor interfacial bonding properties. Most polymers, especially thermosetting, are non-polar ('hydrophobic', water repelling) substances, which are compatible with polar ('hydrophilic', water attracted) wood fibres, and therefore cause poor adhesion between the fibre and matrix interaction surfaces.

Modification of Natural Fibres

The main purpose of modification of natural fibres is to improve the interfacial adhesion and compatibility with the polymer matrix. This is due to the fact that natural fibres consist of cellulose, hemicellulose, pectins and lignin, which are in hydroxyl groups and tend to be strong hydrophilic materials. The significant issue of compatibility between natural fibre and the matrix is the weak interface area due to the different polarity interfaces.

According to Shalwan and Yousif (2013), chemical treatment such as bleaching, acetylation and alkali treatment may improve the interfacial adhesion between fibre and matrix by cleaning the surface of the fibres from impurities. As a result, the roughness of the fibres surface increases and disrupts the moisture absorption process by removing the layer surface of the OH groups in fibre.

In order to improve the affinity and adhesion between the fibres and polymer matrices in production, chemical 'coupling' or 'compatibilising' agents were employed (Kim et al., 2006). Chemical coupling agents are substances, typically polymers that are used in small quantities, to treat the surface of a molecule to enable bonding between the fibre surface and polymer matrix.

The coupling form includes covalent bonds, secondary bonding (such as hydrogen bonding and Van der Waals' forces), polymer molecular entanglement and mechanical inter-blocking (Lu et al., 2006). Therefore, chemical treatment can improve the strength and interfacial adhesion of natural fibres. Some compounds are known to promote adhesion by chemically coupling the adhesion to the material such as sodium hydroxide, silane, acetic acid, acrylic acid, isocyanates, potassium permanganate, peroxide and other coupling agent.

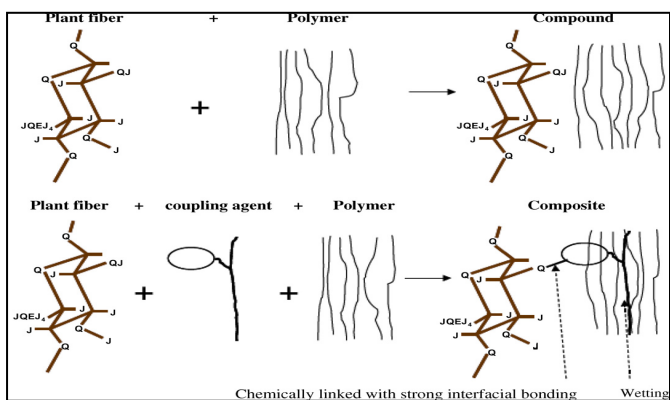


Figure 7. The mechanism of coupling agent between hydrophilic fibre and hydrophobic matrix polymer (Liu et al., 20046)

Bachtiar et al. (2008; 2009; 2010) studied the effects of different concentrations of NaOH (alkaline treatment or mercerisation) on sugar palm reinforced epoxy composites. These fibres were subjected to alkali solution by soaking with 0.25M and 0.5M of NaOH under 1, 4, and 8 hrs of soaking time, respectively. They concluded that the treatment might have increased their tensile strength and modulus by up to 16%, flexural strength and modulus increment up to 24% and 147%, respectively, and impact strength up to 29%. The increase of those properties might be due to the reduction of hydrophilic properties within the sugar palm fibres after being treated with NaOH. It was also thought to have facilitated better interfacial bonding between the palm fibre and epoxy matrix (Ishak et al., 2013).

Synthetic Fibre Sources

With excellent mechanical properties such as high strength and stiffness, and low density, these characteristics enable high carbon and aramid (e.g., Kevlar) to be the commonly used as fibre reinforcement for composites. However, due to their high cost, most researchers and scientists have shifted to using glass fibre instead. Glass is defined by the ASTM standard (C167-71) as an inorganic product of fusion which has been cooled to a rigid condition without crystallisation. In the amorphous state, it is isotropic and has a glass transition point rather than a melting point. Raw materials of glass fibre are silicates, soda, clay, limestone, boric acid, fluorspar or various metallic oxides, which are blended to form a glass batch, which is melted in a furnace and refined during lateral flow to the narrow chamber.

The benefits of using glass fibre include lower cost, moderate stiffness to weight ratio, and ease of manufacture. However, their drawbacks include an increasing wear on the processing machinery due to their abrasive properties (Wambua et al., 2003). Health risk is also another shortcoming, as well as its disposal issue in which glass fibre could not be incinerated as its ash residues could lead to furnace damage.

The only feasible way to dispose glass fibre is to dump the waste in landfill, which may result in another issue of high costs associated with establishment of landfill, machine, labour

and other related operation costs (Bos, 2004). Table 2 shows the classification of glass fibres and their applications.

Table 2

The classifications and applications of glass fibre with designation letter (Wallenberger et al., 2001)

Letter of designation	Properties	Applications
E, electrical	Low electrical conductivity	Electrical applications and widely used as a reinforcement for plastic and composites
S, strength	High strength	Aerospace applications
C, chemical	High chemical durability	Battery plate wrappers and chemical filters
A, alkali	High alkali or soda lime glass	Window glass, bottles and containers
D, dielectric	Low dielectric constant	Electrical and construction applications

In addition, glass fibre has perfect elasticity where glass fibres often follow Hooke's law, while typical glass fibres have a maximum elongation of 5% at break. They also have low coefficient of thermal expansion and high thermal conductivity, outstanding dimensional stability and good chemical resistant (Altan, 2004). As a result, glass fibres are commonly used as a reinforcement material in structural composites such as aircraft parts, boats, automotive parts and other high-end applications (Wallenberger et al., 2001).

POLYMER MATRICES

The matrix serves five important roles of: (1) holding the reinforcement phase in place, (2) deforming and distributing stress to the reinforcement under applied loads or stress, (3) binding the fibres together and transferring load to the fibres and providing rigidity and shape to the structure, (4) isolating the fibres so that individual fibres can act separately and stopping or slowing the propagation of cracks, and (5) providing protection to the reinforced fibres against chemical attack and mechanical damage (Callister et al., 2012; Mazumdar, 2001).

According to Smith and Hashemi (2003), there are two major types of matrices, namely thermoplastics and thermosetting. The composite requirement and application are the determining factors for the matrices selection criteria. For example, in a case where chemical resistance with high temperature resistance is required, thermoset matrices will be more suitable. On the other hand, thermoplastics are preferred when the composite requires high damage tolerance and recyclable.

According to Holbery and Houston (2006), the primary thermoset resins used in natural fibre composites at present are phenolic, unsaturated polyester, vinyl ester and epoxy resin. Table 3 shows a comparison between typical thermoset properties and the most favourable thermosetting resin widely used in various applications.

Table 3

Mechanical properties of phenolic resin with other resins (Leemon et al., 2015; Leman et al., 2008; Sreekala et al., 2001; Iijima et al., 1991)

Type of Resin	Density (g/cm ³)	Tensile strength (MPa)	Tensile Modulus (MPa)
Phenolic (Resole type)	1.19 – 1.2	10	375
Unsaturated polyester	1.2 – 1.5	40 – 90	2000 – 4500
Vinyl ester	1.2 – 1.4	69 – 83	3100 – 3800
Epoxy	1.1 – 1.4	35 – 100	3000 – 6000

Unsaturated Polyester Resin

Unsaturated polyester (UP) resin, which is commonly referred to as polyester resin, is seen as one of the most promising and popular resins, and one of the more versatile synthetic copolymers in composites manufacturing. Unsaturated polyesters are an important class of high performance engineering polymers, which can be found in a number of engineering applications.

They were primarily used in compression moulding (sheet moulding compounds), injection moulding (bulk moulding compounds), resin transfer moulding (RTM), pultrusion, filament winding and hand lay-up process (Vilas et al., 2001). It was found that 85% of the fibre reinforced polymer (FRP) products like boats, car and aircraft components and chairs were manufactured using polyesters (Devi et al., 19978).

Unsaturated polyester is a thermosetting polymer based which contains ester functional group in their main chain and it is dissolved in styrene monomer that copolymerises and causes the resin to cure. The unsaturated nature of the backbone provides sites for reaction with double bonds in styrene monomer through peroxide initiators, leading to the formation of three-dimensional network (Malik et al., 2000). Figure 8 shows the molecular structure of unsaturated polyester with positions of ester group and reactive sites.

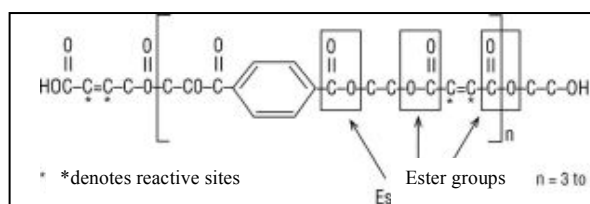


Figure 8. Positions of the ester group (C=O-O-C) and reactive sites (C* = C*) attached within the molecular structure of unsaturated polyester

The most frequently catalyst used is methyl ethyl ketone (MEKP) or benzoyl peroxide (BPO) with the amount varies from 1-2%. It was added to the resin system in order to initiate the polymerisation reaction. The catalyst does not take part in the chemical reaction but simply activates the process for curing process.

In order to enable reaction to take place at ambient temperature and decrease process time, an accelerator is added to the catalysed resin. Without the presence of catalyst, accelerators have

small effect on resin reaction. Therefore, the polyester manufacturer normally adds accelerator into the resin to create a 'pre-accelerated' reaction. The cobalt based accelerator expedites the catalyst to initiate the chemical reaction between the resin and styrene monomer.

In the presence of polyester resin, the catalyst will decompose to form free radicals that attack the unsaturated groups (C=C) to start up the polymerisation process. Meanwhile, the rate of polymerisation is proportional to the processing temperature and amount of catalyst being used (Dholakiya, 2012).

Unsaturated polyesters are capable of producing very strong bonds with other materials, with good toughness and crack resistance capability (Willis et al., 2003, pp. 475-479). Unlike the compatibility interaction between polar UP and polar natural fibres, non-polar thermoplastics are not compatible with natural fibres, which resulted in a poor interfacial interaction. In order to enhance this poor interfacial adhesion, a coupling agent was used in small quantities to treat the fibrous surface (Naguib et al., 2015; Schneider et al., 1985). Table 4 shows the test methods according to the ASTM standard for characterising the mechanical properties of unsaturated polyester resin.

Table 4

The ASTM test methods for characterising the mechanical properties of unsaturated polyester resin

Properties	ASTM test method
Tensile strength, modulus and % elongation	D3039/D638
Flexural strength and modulus	D790
Compressive strength, modulus and % compression at break	D695
Izod impact	D256
Heat distortion	D648
Barcol hardness	D2583

The advantages of unsaturated polyester are its dimensional stability and affordable cost. Other advantages include ease of handling, processing, fabricating and good balance of mechanical, electrical and chemical properties. Some special formulations offer high corrosion resistance and fire retardant. This resin is probably the best value for a balance between performance and structural capabilities (Mathews et al., 1994; Mishra et al., 2003; Tuttle, 2004).

NATURAL FIBRE REINFORCED COMPOSITES

Hybrid Fibre Reinforced Composites

Hybridisation is a design strategy for fibre reinforced composite materials through an incorporation of two or more fibres within a single matrix with different classes to manipulating the desired properties. It can be from a combination of natural-natural fibres, natural-synthetic fibres and synthetic-synthetic fibres (Mishra et al., 2003). Hybrid biocomposite materials are usually quite challenging to manufacture due to their different properties. They could be made from combinations of polymer, elastomer, metal, ceramics, glasses, and plant, and can

be produced in the forms of composite, sandwich laminates, lattice and segmented (Ashby et al., 2013).

According to Caseri and Walter (2007), the interaction could be either weak because of Van de Waals, hydrogen bonding and weak electrostatic interactions, or strong because of the chemical interactions between the different components. Besides that, the designation of the hybrid biocomposite also can affect their mechanical strength and load carrying. Furthermore, hybridisation with glass fibres could improve the mechanical properties of biocomposites. However, hybrid biocomposites are generally limited up to 50% of fibre loading (Maleque et al., 2007; Sanadi et al., 2001). Figure 9 shows an example of the mechanism of hybridisation conducted by Atiqah et al. (2014).

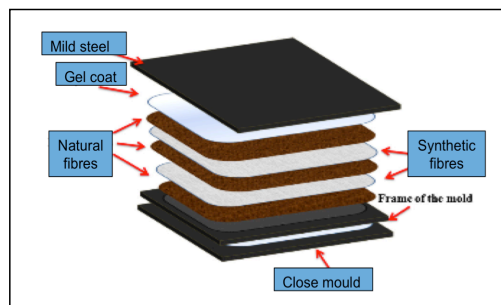


Figure 9. The mechanism of hybridisation components of natural fibres, synthetic fibres and polymer matrix (Atiqah et al., 2014)

According to Thwe and Liao (2002), the advantage of hybridisation of different fibres is that the fibre could complement the limitation with another fibre. As a result, a balance of mechanical performance and cost reduction for engineering applications could be achieved (Reddy et al., 2008).

Meanwhile, Jawaid et al. (2013) studied the mechanical behaviour of hybrid biocomposites based on jute and oil palm fibre. It was found that the use of hybrid system increased the tensile strength and modulus, and lowered the damping effect of the oil palm-epoxy composite. This was due to the good fibre-matrix interface bonding and the increase in stress-transfer effectiveness.



Figure 10. Mini boat made from hybrid sugar palm fibre with glass fibre reinforced unsaturated polyester composites (Ishak et al., 2011)

Ishak et al. (2013) developed a composite boat made of sugar palm and glass fibres. The boat, shown in Figure 10, was made of 2 layers of the fibres which were woven sugar palm and glass fibres reinforced with unsaturated polyester resin. The dependence of glass fibre has reduced the weight of the boat up to 50%. It should be noted that the density of sugar palm is approximately 1.22–1.29 kg/m³ compared to the density of glass fibre, which is 2.5 kg/m³.

An investigation was done by Ahmed et al. (2008) to study the effects of stacking sequence on the mechanical properties of woven jute and glass fabric reinforced polyester hybrid biocomposites. It was found that layering sequence affects the flexural and inter-laminar shear properties more than the tensile properties. They also concluded that hybrid laminates, with two glass plies on each side, were the optimum combination for a good balance between mechanical properties and also cost incurred.

Mishra et al. (2003) stated that various research has shown that the hybrid biocomposites behaviour is a function of weighted sum of individual components, with favourable balance between the benefits and drawbacks of the composite materials. There are a few factors affecting the mechanical properties of hybrid biocomposites, which include hybridisation design, nature of the matrix, fibre length, fibre compositions and fibre-matrix interface.

CONCLUSION

There are lot of beneficial natural fibre sources in a wide range of applications in the composite industry. It is worth mentioning that the performance of natural fibre reinforced composites can be tailored through hybridisation by employing an appropriate amount of synthetic fibres. In addition to cost effectiveness balance, a balance between environmental impacts and desired performance could be achieved by designing the composite based on the product requirements.

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