

**NATURAL WEATHERING AND
HYGROTHERMAL AGING STUDIES OF
JUTE/GLASS FIBRES REINFORCED
UNSATURATED POLYESTER HYBRID
COMPOSITES**

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UNIVERSITI SAINS MALAYSIA

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JUTE/GLASS FIBRES REINFORCED
UNSATURATED POLYESTER HYBRID COMPOSITES**

by

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LIST OF SYMBOLS

π	pie
%	percentage in 100
\pm	tolerance
cm^{-1}	wave number
g/cm^3	density in gram per centimetre cube
g/m^2	gram per meter square
h	hour
kPa	kilo pascal
MJ/kg	mega joule per kilogram
mm	milimeter
mm/min	milimeter per minute
mm^2	milimeter square
$\text{MPam}^{1/2}$	mega pascal square root of meter
$^\circ$	degree
$^\circ\text{C}$	degree Celsius
phr	part per hundred parts of resin

LIST OF ABBREVIATIONS

Δa^*	difference between the initial and final values of a^*
Δb^*	difference between the initial and final values of b^*
ΔE	total colour change
ΔL^*	difference between the initial and final values of L^*
a	notch length
a^*	chromaticity coordinates of red-green axis
Al	aluminium
Al_2O_3	aluminium oxide
AR	as-received
ATR	Attenuated Total Reflectance
B	boron
b^*	chromaticity coordinates of yellow-blue axis
C	corrosion
Ca	calcium
CaO	calcium oxide
cf.	compare or consult
CI	carbonyl index
D	dextro
D	diffusion coefficient
DCPD	dicyclopentadiene
E	electrical
etc	et cetera (and other things)
exp	exponential

Fe	ferum
F_{\max}	maximum force
F_Q	the load at crack growth initiation
FRPs	fibre reinforced plastics
FTIR	Fourier transform infra-red
GF	glass fibre
GF/UP	glass fibre-reinforced unsaturated polyester
GP	general purpose
GPa	gega pascal
h	thickness
HA	Hygrothermal Aging
i.e.	that is
ILSS	interlaminar shear strength
JF	jute fibre
JF/GF/UP	jute/glass fibre-reinforced unsaturated polyester
JF/UP	jute fibre-reinforced unsaturated polyester
K_2O	potassium oxide
K_c	critical stress intensity factor
L^*	lightness
LEFM	Linear Elastic Fracture Mechanics
Li_2O	lithium oxide
MEKP	methyl ethyl ketone peroxide
MgO	magnesium oxide
M_m	maximum water uptake
MPa	mega pascal

Na	sodium
Na ₂ O	sodium oxide
NW	natural weathering
OH	hydroxyl group
PMCs	Polymer matrix composites
RD	re-dried
RTM	resin transfer moulding
S	silica
SEM	Scanning Electron Microscopy
SENT	single-edge notched tensile test
SiO ₂	silicone dioxide
t	time
UP	unsaturated polyester
UV	ultraviolet
UV-B	ultraviolet radiation in the range of 290-320 nm
W	width of the specimen
W _g	weight of glass fibre
W _j	weight of jute fibre
W _o	weight of the specimen
W _r	weight of resin
wt	weight
W _t	weight at any specific time of immersion
ρ _g	density of glass fibre
ρ _j	density of jute fibre
ρ _r	density of resin

**KAJIAN PENCUACAAN SEMULAJADI DAN PENUAAN HIDROTERMA
BAGI KOMPOSIT HIBRID POLIESTER TAK TEPU
DIPERKUAT GENTIAN JUT/KACA**

ABSTRAK

Kajian terhadap ketahanan komposit polyester tak tepu (UP) mengandungi dua bahagian. Bahagian pertama adalah untuk menyiasat pengaruh pencuacaan semulajadi terhadap sifat-sifat mekanikal dan morfologi matrik UP dan komposit-komposit diperkuat gentian jut (JF), gentian kaca (GF) dan gentian hibrid jut/kaca (JF/GF). Matrik UP disediakan dengan kaedah tuangan manakala komposit disediakan dengan kaedah pembasahan lapisan. Untuk kajian pencuacaan semulajadi, spesimen telah didedahkan kepada cuaca iklim tropika selama sehingga dua belas bulan ($T_{\max}=31.2\pm 3.8^{\circ}\text{C}$, $T_{\min}=23.7\pm 2.7^{\circ}\text{C}$, $\text{RH}=79.7\pm 18.0\%$) di Pulau Pinang, Malaysia. Tahap degradasi komposit terhadap pencuacaan telah diukur melalui sifat mekanikal, fizikal dan morfologi komposit. Ketahanan mekanikal spesimen terhadap pencuacaan semulajadi telah diukur melalui sifat-sifat kelenturan, regangan, keliatan patah dan kekuatan ricih interlaminar (ILSS). Pengurangan kekuatan mekanikal spesimen komposit JF/UP adalah disebabkan oleh degradasi pada permukaan yang terdedah dan antaramuka matrik/gentian melalui kombinasi faktor persekitaran dimana ia diterangkan oleh mikrograf serta pemudaran warna pada kawasan yang terdedah. Prestasi mekanikal dan daya tahan komposit hibrid dipertingkatkan menerusi pendedahan terhadap pencuacaan semulajadi. Tiada perubahan kimia yang ketara pada spesimen selepas terdedah kepada

cuaca semulajadi berdasarkan Inframerah Transformasi Fourier (FTIR). Namun, penurunan Index Karbonil (CI) terhadap pencucuan mungkin disebabkan oleh degradasi dan kelunturan lignin pada JF. Bahagian kedua kajian ini adalah untuk mengkaji daya tahan komposit terhadap penuaan hidroterma (HA). Untuk HA, spesimen telah direndamkan di dalam air suling pada suhu 30°C, 60°C dan 90°C selama tiga bulan. Pola penyerapan air untuk spesimen pada rendaman 30°C dan 60°C telah mengikuti perilaku “Fickian”, manakala spesimen pada rendaman 90°C mempamerkan perilaku bukan “Fickian”. Komposit JF/UP menunjukkan penyerapan air yang paling tinggi pada semua suhu rendaman dibandingkan dengan spesimen lain kerana sifat hidrofilik pada JF. Sebaliknya, komposit GF/UP menunjukkan penyerapan air yang boleh diabaikan semasa HA kerana sifat hidrofobik pada GF. Nilai pekali difusi, D amat bergantung pada suhu rendaman. Namun, komposit hibrid JF/GF/UP menunjukkan nilai D yang paling rendah pada semua suhu rendaman. Sifat-sifat kelenturan dan ILSS matrik UP berkurang secara mendadak semasa HA pada suhu 90°C. Mekanisme kegagalan digabungkan oleh degradasi matriks melalui hidrolisis, penarikan keluar gentian, degradasi dan penyahikatan gentian. Penghibridan JF dan GF telah menunjukkan kesan sinergi dalam meningkatkan rintangan terhadap penyerapan air dan daya ketahanan sifat-sifat kelenturan dan ILSS terhadap HA. Mikrograf SEM menunjukkan komposit-komposit JF/UP, GF/UP and hibrid JF/GF/UP mengalami kerosakan teruk pada permukaan yang terdedah, retakan dan degradasi matrik pada antaramuka matrik/gentian dan menjadi lebih ketara terhadap HA pada suhu 90°C.

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ABSTRACT

The research on the durability studies of unsaturated polyester (UP) based composites consists of two parts. The first part is mainly to investigate the effects of natural weathering on the mechanical and morphological properties of UP matrix as well as jute fibre- (JF), glass fibre- (GF) and jute/glass hybrid (JF/GF) fibre-reinforced UP composites were investigated. The UP matrix was prepared by casting method, whereas the composites were fabricated by a hand lay-up technique. For the natural weathering study, specimens were exposed for up to maximum of twelve months under tropical climatic conditions ($T_{\max}=31.2\pm 3.8^{\circ}\text{C}$, $T_{\min}=23.7\pm 2.7^{\circ}\text{C}$, $\text{RH}=79.7\pm 18.0\%$) in Pulau Pinang, Malaysia. The mechanical, physical and morphological properties of composites were used to measure the extent of degradation in the composites upon weathering. The flexural, tensile, fracture toughness and interlaminar shear strength (ILSS) properties were evaluated to measure the mechanical retentionability of the naturally weathered specimens. The mechanical strength reduction of the naturally weathered JF/UP composite specimens were due to degradation of the exposed surface and the fibre/matrix interface by a combination of environmental factors which was revealed by SEM micrographs and colour fading of the exposed surfaces. The hybridisation of JG and GF has improved the mechanical performance and has also enhanced the durability of the hybrid composites upon exposure to natural weathering. Fourier transform

infra-red (FTIR) revealed that no significant chemical changes of the specimens upon exposure to NW. However, the JF/UP composites show decreasing of Carbonyl Index (CI) upon weathering may be attributed to the degradation and leached of the lignin in JF. The second part of the research was to study the durability of the composites upon hygrothermal aging (HA). For the HA, specimens were immersed in distilled water at 30°C, 60°C and 90°C for a period of three months. The water absorption pattern of hygrothermally aged specimens immersed at 30°C and 60°C followed Fickian behaviour, whereas specimen immersed at 90°C exhibited non-Fickian behaviour. The JF/UP composite showed the highest water uptake at all immersion temperatures as compared to other specimens due to hydrophilic nature of the JF itself. In contrast, the GF/UP composite showed negligible water uptake upon HA due to the hydrophobic nature of the GF itself. Values of diffusion coefficient, D were observed to be strongly dependant on immersion temperature. However, the JF/GF/UP hybrid composites showed the lowest D values at all immersion temperatures. The flexural and ILSS properties of the UP matrix and its composites were dramatically reduced after HA at 90°C. The failure mechanism is combination of matrix degradation via hydrolysis, fibre pull out, fibre degradation and fibre debonding. Hybridisation of JF and GF has shown a synergistic effect in terms of increasing the resistance against water uptake and superior retention ability of flexural and ILSS performances against HA. SEM micrographs revealed that severe deterioration on exposed surfaces, matrix cracking and degradation at fibre/matrix interface in JF/UP, GF/UP and JF/GF/UP hybrid composites became prominent upon HA at 90°C.

CHAPTER 1

INTRODUCTION AND PROBLEM STATEMENT

1.1 Introduction

Synthetic fibres such as nylon, rayon, aramid, glass and carbon are used extensively for the reinforcement of plastics (Albuquerque *et al.*, 2000). Nevertheless, these materials are expensive and non-renewable. Over the recent years, there is an increasing interest in natural fibres as a substitute for glass fibre (GF) mainly because of their low specific gravity, low cost, availability, renewability, biodegradable nature and low price as well as their satisfactory mechanical properties (Joshi *et al.*, 2004). This makes them an attractive ecological alternative to the glass, carbon and man-made fibres used for the manufacture of composites which can lead to the development of ecologically friendly composites (Al Kafi *et al.*, 2006; Montazer & Salehi, 2008). The ecological balance of natural fibre-reinforced composites points to their lower environmental noxiousness compared to the reinforcing materials used hitherto. Lignocellulosic fibres like sisal, jute, hemp, flax, coir, pineapple and banana (Bismarck *et al.*, 2005) have been used as reinforcements in thermoset matrices. They are currently used in transportation, military applications, the building and construction industries, electronics, packaging and consumer products, etc. (Anandjiwala & Blouw, 2007; Peponi *et al.*, 2008). Among natural fibres, jute fibre (JF) is of particular interest in that its composites have high impact strength, moderate tensile and flexural strength properties compared to other lignocellulosic fibres (Wollerdorfer & Bader, 1998). JF is a natural plant fibre, composed mainly of 58-63% cellulose, 20-22% hemicellulose, 12-15% lignin (Sinha & Rout, 2008) and a small proportional of pentosans, extractives and waxes. JF based composites have

been widely using in the transportation, military applications, food grain silos, wood substitute, low cost housing units, roofing, pipes, packaging and consumer products, etc (Anandjiwala & Blouw, 2007; Kozlowski, 2000; Peponi *et al.*, 2008). Extensive studies have been carried out to fabricate jute/epoxy, jute/polyester and jute/phenol-formaldehyde composites for applications such as low-cost housing materials, silos for grain storage and small fishing boats (Bismarck *et al.*, 2005; Suddell & Evans, 2005).

1.2 Problem Statement

Despite the advantages of natural fibres based polymer composites, natural fibres are environmentally sensitive, and have relatively poor moisture resistance, which causes degradation to the strength of the natural fibre composites. These disadvantages limit the use of natural fibres in most of the applications. Studies of environmental effects on the mechanical and physical properties of some natural fibre composites, such as jute, sisal and kenaf, etc., have been reported (Albuquerque *et al.*, 2000; Leman *et al.*, 2008; Singh *et al.*, 2000; Thwe & Liao, 2003; Wang *et al.*, 2006; Wollerdorfer & Bader, 1998).

The effects of natural weathering on natural fibre-reinforced unsaturated polyester (UP) composites and GF/UP composites have raised concerns in terms of their durability, including their resistance to ultraviolet (UV), temperatures fluctuation and dimensional stability (Bengtsson *et al.*, 2007; Gu, 2008; Mouzakis *et al.*, 2008; Stark & Matuana, 2004).

In addition to this, natural fibres based composites also suffer from some limitation such as poor retention ability when expose to moisture and heat (Alsina *et al.*, 2007; Dhakal *et al.*, 2007; Singh *et al.*, 2000). Composite generally absorb

moisture to some measurable degree when immersed directly in water or when exposed to atmospheric moisture, affecting the immediate or the long term performance of the materials (Albuquerque *et al.*, 2000). The water absorption can have both reversible and irreversible effects on properties and performances of natural fibre based composites. Since fibres (organic and inorganic) and polymer matrix exhibit different properties in terms of water absorption, type of fibres reinforced into the polymer matrix, fibre distribution, fibre loading, fibre/matrix interface and sample preparation are the key for overall moisture absorption of composites (Dhakal *et al.*, 2007; Marais *et al.*, 2000; Wang *et al.*, 2006).

The influence of the water absorption and HA on the mechanical properties of the JF/UP composites has been reported in previous studies (Aquino *et al.*, 2007; Athijayamani *et al.*, 2009; Md Akil *et al.*, 2009). In order to overcome the weakness of the JF/UP composite, combination of JF and GF in same UP matrix can produce a hybrid composite that take full advantage of the best properties of the constituents (synergistic effect), and thereby an optimal, superior but economical composite can be obtained. Hybrid composites can be prepared by combining two or more different types of fibres in a common matrix. They offer a range of properties that cannot be achieved with a single kind of reinforcement. Studies on the hybridisation effect of GF with natural fibres have been reported in the literature (Abdul Khalil *et al.*, 2007; Ahmed *et al.*, 2006).

So far, only a few studies have been reported on the effect of natural weathering on the performance of JF/GF/UP hybrid composites and these studies were focused on Mediterranean, oceanic, season and monsoon temperature climates. Studies reporting on the performance of composites in a tropical climate (hot and wet) are rather limited. Furthermore, it is also rare to find from the literature that shows

that effects of HA at high temperatures on the performance of JF based UP composites.

1.3 Research Objectives

This study is concerned with the durability of the JF and GF based UP composites.

The primary objectives of this study are:

- ❖ To study the durability (physical and mechanical properties) of the monotonic fibre-reinforced UP composite, i.e. JF/UP and GF/UP composites exposure to tropical weathering for a period of twelve months. The mechanical, physical and morphological properties of the composites were used as measures of the extent of degradation in the composites upon weathering.
- ❖ To study the effect of hybrid fibre (JF/GF) on the mechanical and physical performance of JF/GF/UP hybrid composite as compared to the monotonic JF/UP and GF/UP composites.
- ❖ To study the durability (physical and mechanical properties) of the JF/UP, GF/UP and JF/GF/UP hybrid composites exposure to HA at 30°C, 60°C and 90°C for a period of three months. The mechanical, physical and morphological properties of the composites were used as measures of the extent of degradation in the composites upon HA.

CHAPTER 2

LITERATURE REVIEW

2.1 Polymer Matrix

2.1.1 *Thermoset Materials*

Thermoset materials are the polymers compounds that decompose before they can be melted or reshaped. Thermoset consist of a three-dimensional network polymer containing cross-linked molecules that does not soften when heated. Thermoset polymers are used as the matrix for filled plastics and fibre-reinforced. Examples of thermosetting resins include epoxy, polyester, polyurethane, phenolics and vinyl ester (Carracher, 2007; Franco, 2005).

2.1.1.1 *Polyester*

The initial polyester formation actually occurred early is attributed to Gay Lussac and Pelouze in 1833 and Berzelius in 1847. These polyesters are called glyptals and alkyds, and they are useful as coatings materials and not for fibre production. Glyptal polyesters were commercially produced in 1901 by heating glycerol and phthalic anhydride. Since the secondary hydroxyl is less active than terminal primary hydroxyls in glycerol, the first product formed at conversions of less than 70% is a largely linear polymer (Carracher, 2007, 2008). Alkyds were synthesized by Kienle in the 1920s from trifunctional alcohols and dicarboxylic acids. The term alkyd is sometimes used to describe all polyesters produced from polybasic acids and a polyhydric alcohol. The terms “saturated” and “unsaturated” polyesters have been used to distinguish between those alkyd polyesters that are saturated from those that contain unsaturation.

The majority of dinghies, yachts and workboats are built with composites reinforced with glass fibre (GF) having polyester resin as the matrix material. The cross-linking occurs either at room temperature using methyl ethyl ketone peroxide (MEKP) or at elevated room temperatures in press mouldings using benzoyl peroxide. A whole range of polyesters is made from different acids, glycols, and monomers, all having varying properties. There are two principal types of polyester resins used as a standard laminating system in the composites industry. Orthophthalic polyester resin is the standard economic resin commonly used, and it yields highly rigid products with a low heat resistance property. Isophthalic polyester resin is now becoming the preferred material in the marine industry, where its superior water resistance is desirable (Ray & Rout, 2005). Figure 2.1 shows the typical polymerization processes of ester and polyester.

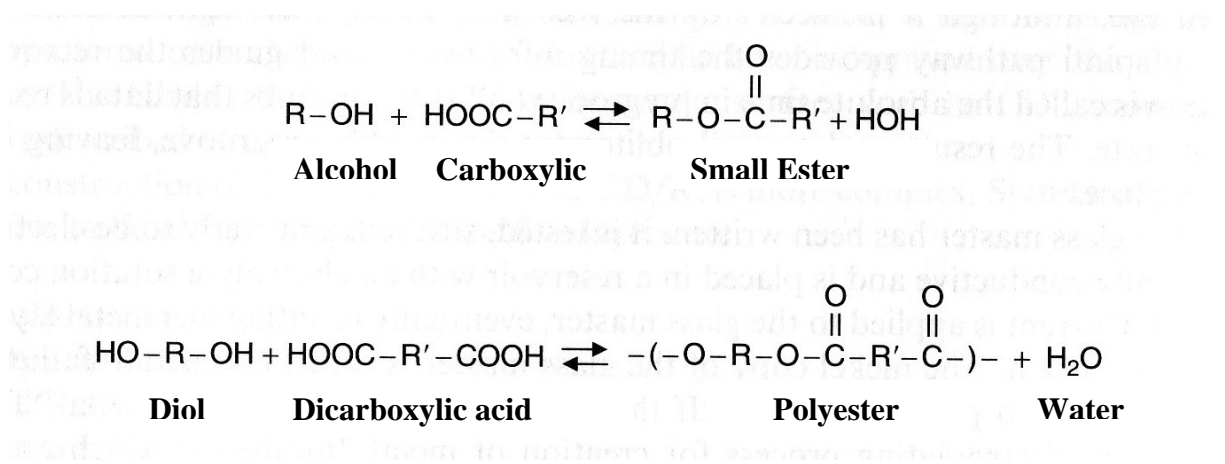


Figure 2.1: Typical polymerization process of ester and polyester (Carracher, 2008).

2.1.1.2 Unsaturated Polyester

Unsaturated polyester (UP) resin is a thermoset capable of being cured from a liquid to a solid state when subjected to appropriate conditions. UP consists of low molecular weight fumarate esters containing various chemical structures designed for their specific cost and performance purposes. The two most important features of UP

are the fumarates, which provide the active sites for radical cross-linking with the diluent monomer and the random, low molecular weight, irregular nature of the rest of molecule, which provide necessary solubility in the diluent monomer. For most commercial resins, the diluent is styrene monomer, but is possible to use other vinyl monomers such as methyl styrene and alkyl methacrylate monomers. These diluents serve two vital roles for the system. They reduce viscosity so the resins can process, and they cross-link with the double bonds in the polyester (Carracher, 2008).

UP are prepared in two steps; synthesis of low molecules weight linear polyester (700-4000) containing carbon-carbon double bonds in the polymer chains, and is dissolved in a vinyl monomer and stabilized with an inhibitor (Feldman & Barbalata, 1996). Typically, UP are prepared through a classical esterification process; e.g. a dihydroxy compound or mixtures of dihydroxy compounds are treated with maleic anhydride and/or together with other dicarboxylic acids such as aromatic or aliphatic dicarboxylic acids under elevated temperature to remove the water produced during esterification process (Carracher, 2007, 2008). Although various catalysts will catalyze this esterification reaction, there is enough carboxylic acid in the mixture so that it is not necessary to add extra catalyst.

Figures 2.2 and 2.3 show the preparation of UP by esterification and condensation polymerization process. Esterification process was prepared by reaction between maleic anhydride and propylene glycol while condensation process was prepared by reaction between fumaric acid and ethylene glycol (Feldman & Barbalata, 1996). When used in applications, the resin composition is mixed with a catalyst system (peroxide), and cross linked by the copolymerization of the unsaturated double bonds in the polyester with the vinyl monomer used as solvent. This can occur at room temperature, under heat (60°C -90°C) or by UV or visible

light radiation. The resin begins to cure as soon as the initiator is added, the speed of reactivity depending on the temperature, resin and catalyst reactivity. Figure 2.4 shows the process of UP resin; from preparation of resin by condensation process to a cured composite.

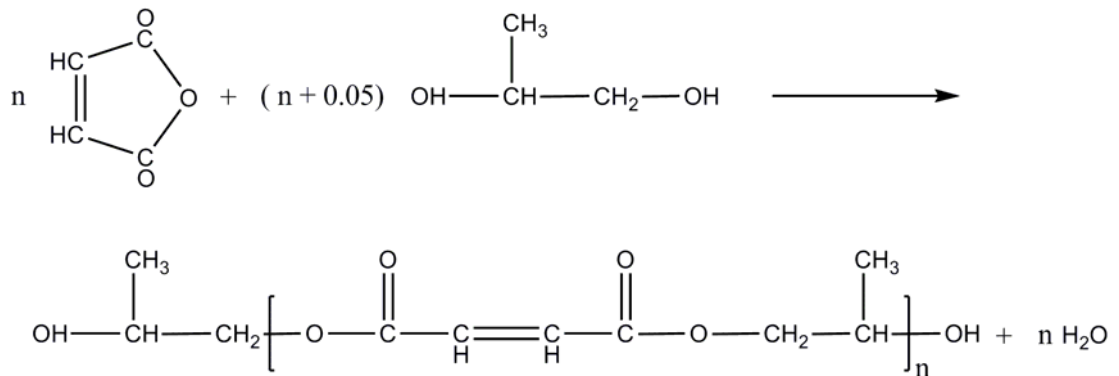
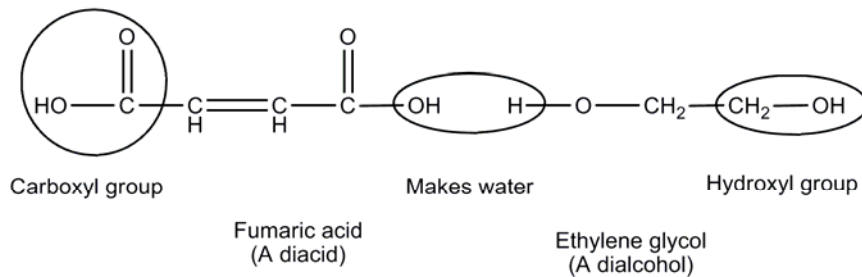
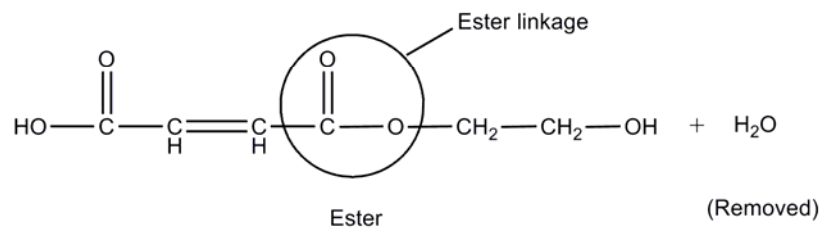


Figure 2.2: Esterification process of the UP resin by maleic anhydride and propylene glycol (Feldman & Barbalata, 1996).

Reactants



Dimer formation



Chain growth

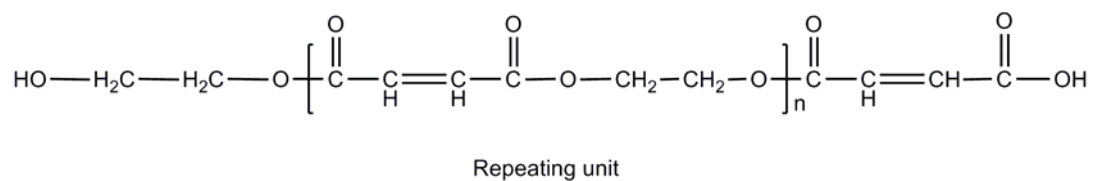


Figure 2.3: Schematic of condensation polymerization of an UP resin (Astrom, 1997).

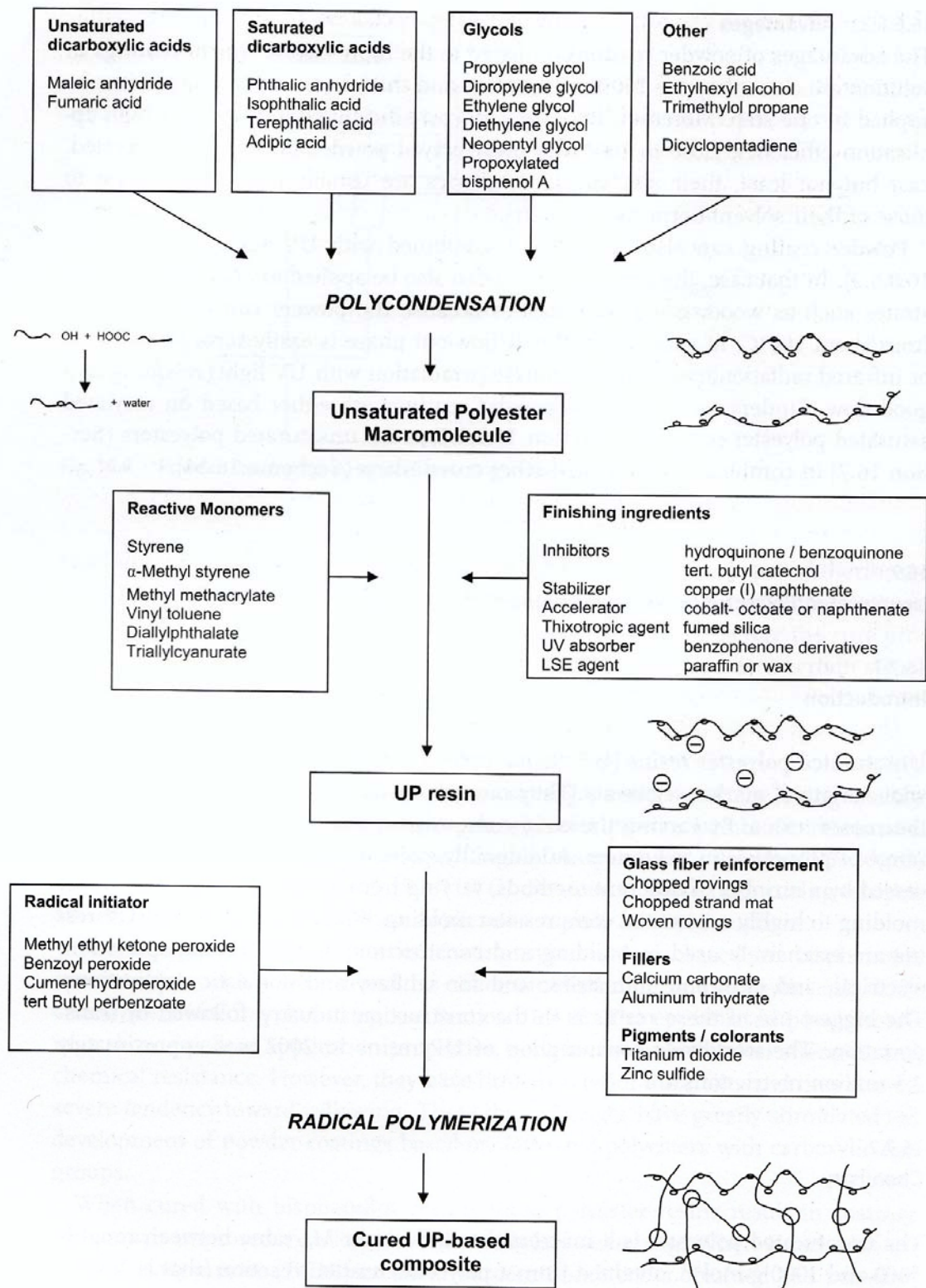


Figure 2.4: Schematic process of UP resins: from raw materials to cured composites (Meyer & Keurentjes, 2005).

Aromatic polyesters had been successfully synthesized from the reaction of ethylene glycol and various aromatic diacids but commercialization awaited a ready

inexpensive source of aromatic diacides. An inexpensive process was discovered for the separation of the various xylene isomers by crystallization. The inexpensive xylene isomers allowed the formation of terephthalic acid through the air oxidation of the p-xylene isomer (Carraher, 2008).

In modern day, there are three basic types of UP resins:

- ❖ The phthalic anhydride, maleic anhydride and glycol resins. These are generally referred to as “general-purpose orthophthalic resins”, or simply “GP Ortho resins”.
- ❖ The isophthalic acid, maleic anhydride and glycol resins. These are generally referred to as “isophthalic resins”, or simply “iso resins”.
- ❖ The dicyclopentadiene (DCPD)-capped resins, or simply “DCPD resins”.

Basic chemical structures of UP monomers are shown in Figure 2.5.

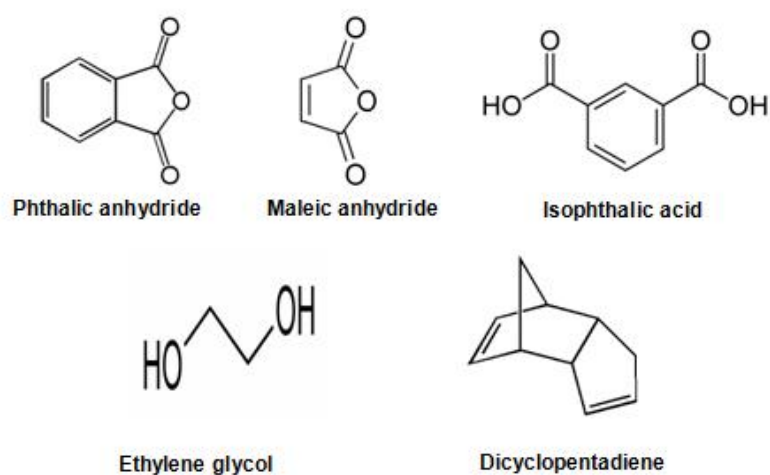


Figure 2.5: Chemical structure of phthalic acid anhydride, maleic anhydride, isophthalic acid, ethylene glycol and dicyclopentadiene (Carraher, 2007, 2008).

The UP are extremely versatile in properties and applications and have been a popular thermoset used as polymer matrix in composites (Al Kafi *et al.*, 2006; Albuquerque *et al.*, 2000; Dhakal *et al.*, 2007; Errajhi *et al.*, 2005; Khashaba, 2003).

This matrix has been used for many years in broad technology fields such as naval construction, offshore applications, waterlines and building construction (Gu, 2008; Mouzakis *et al.*, 2008).

2.2 Fibres

2.2.1 Natural Fibres

Wood, agricultural residues, and several plants, such as sisal, jute, ramie, coconut tree and pineapple, are examples of lignocellulose sources. Lignocellulosic materials consist mainly of cellulose, hemicelluloses and lignin. The amounts of these three constituents vary considerably among the different lignocellulosic fibres. For instance, flax has a relatively high cellulose content and low lignin content; whilst jute has relatively high lignin content. Composition differences occur between the bottom and top of the stem and composition changes during growth; the lignin content increases particularly at the end of the plant growth (Pott, 2004). Lignocellulosic materials can contribute significantly to lumber and agricultural industries by developing high performance materials and composites (Frollini *et al.*, 2004).

2.2.1.1 Cellulose

Cellulose was originally “discovered” by Payen in 1838 (Hamad, 2002). For thousands of years impure cellulose formed the basis of much of our fuel and construction systems in the form of wood, lumber (cut wood), and dried plant material; served as the vehicle for the retention and conveying of knowledge and information in the form of paper, and clothing in the form of cotton, ramie, and flax. Cellulose is a homo-polysaccharide of β -D-glucopyranose units which are linked

together by (1-4) glycosidic bonds. Figure 2.6 shows the structure of cellulose. Cellulose is a polydisperse polymer with an average degree of polymerization in the general range of 3,500-36,000. Native cellulose is widely distributed in nature and is the principle constituent of cotton, kapok, flax, hemp, jute, ramie and wood.

Cellulose comprises more than one-third of all vegetable matter and is the world's largest renewable resource. Cellulose is not found in a pure form, but is rather associated with other materials such as lignin and hemicelluloses (Chawla, 1998). Cotton fibre, for example contains 80-85% cellulose, while other vegetable fibres such as ramie, hemp, jute, flax and sisal have between 60-75% cellulose. Highly oriented crystalline cellulose has a density as high as 1.63 g/cm^3 , while amorphous cellulose has a density as low as 1.47 g/cm^3 (Caraher, 2008). As a consequence of high oriented fibrous structure and strong hydrogen bonds, cellulose has a high tensile strength and insoluble in most solvents. For an organic polymer, cellulose is remarkably resistant to chemical attack. Fairly severe conditions of acidity, alkalinity or temperature are usually required before degradation can take place (Hamad, 2002).

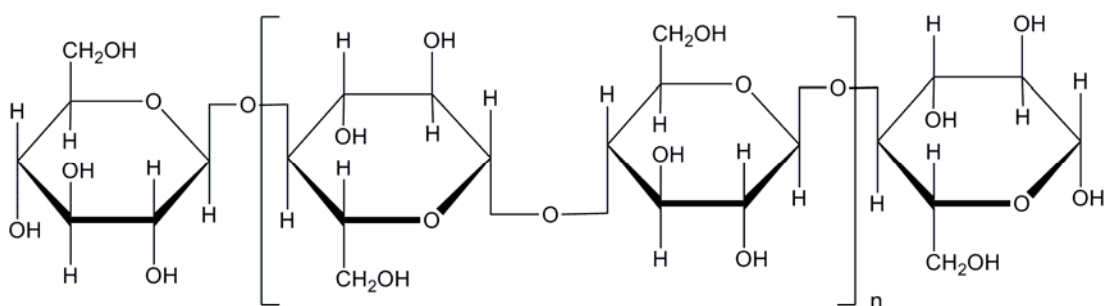


Figure 2.6: Chemical structure of cellulose (Caraher, 2008).

2.2.1.2 Hemicelluloses

The heterogeneous polysaccharide components of the cell wall other than cellulose are called the hemicelluloses. Hemicelluloses, which are amorphous, branched polymers composed of two or three monosaccharide residues and have much lower degrees of polymerization (150-200) than cellulose, are a group of non-structural, low molecular weight, mostly heterogeneous polysaccharide which are unrelated to cellulose. They are formed biosynthetically by a separate route, and are not as the name seems to imply, biosynthetic precursors to cellulose. The structure and composition of hemicelluloses vary with three species, and two major groups exist in gymnosperms: xylans and glucomannans (Hamad, 2002).

Hemicelluloses in wood have been described as cementing agents that bind micro fibrils together and prevent splits as wood fibres are twisted, bent and/or stretched in service. Swollen in water, they render the surfaces of fibres tacky during paper formation and seem to form strong bonds upon drying. The greater the amount of these hemicelluloses in delignified fibres, the more pronounced is the degree of fibre swelling. Hemicelluloses are less resistant to chemical attack than cellulose. Hemicelluloses are relatively easily hydrolyzed by acids to their monomeric components consisting of D-glucose, D-mannose, D-xylose, L-arabinose, and small amounts of L-rhamnose in addition to D-glucuronic acid, 4-O-methyl-D-glucuronic, and D-galacturonic acid (Pott, 2004; Sjostrom, 1981). These polymers are thus recognized as being important with respect to swelling, beating and bond formation of fibres in the pulping and paper making processes; in the cell wall, they are oriented parallel to the cellulose chains..

2.2.1.3 Lignin

The third major component of wood is lignin. Lignin is a complex, 3 dimensional, cross-linked networks composed of phenyl propane-type monomers. Since lignin is produced as a by-product of plant metabolism, it is located both within and between wood fibres. High tough amorphous nature of lignin serves to reinforce wood fibres by increasing the rigidity of the cell wall. Lignin exists between carbohydrate constituents of the cell wall in a lamellar type of structure. Units present in lignin and structure of lignin are shown in Figures 2.7 and 2.8. Lignin which is generally insoluble in neutral solvents however can be degraded under acidic or alkaline conditions. (Hamad, 2002). Lignin can be isolated from extractive-free wood as an insoluble residue after hydrolytic removal of the polysaccharides. Alternatively, lignin can be hydrolyzed and extracted from the wood or converted to a soluble derivative. Klason lignin is obtained after removing the polysaccharides from extracted (resin-free) wood by hydrolysis with 72% sulfuric acid. The polysaccharides may be also removed by enzymes from finely divided wood meal with remaining its original structure while extracted by using dioxane containing water and hydrochloric acid, but considerable changes in its structure (Sjostrom, 1981).

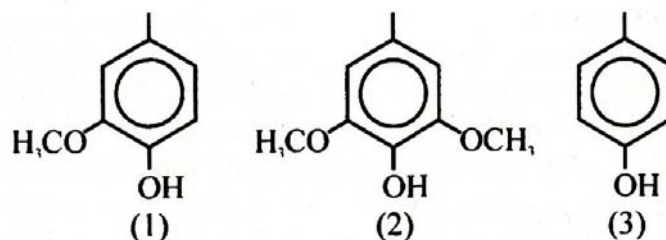


Figure 2.7: Units present in lignin: (1) guaiacyl, (2) syringyl and (3) p-hydroxyphenyl (Frollini *et al.*, 2004).

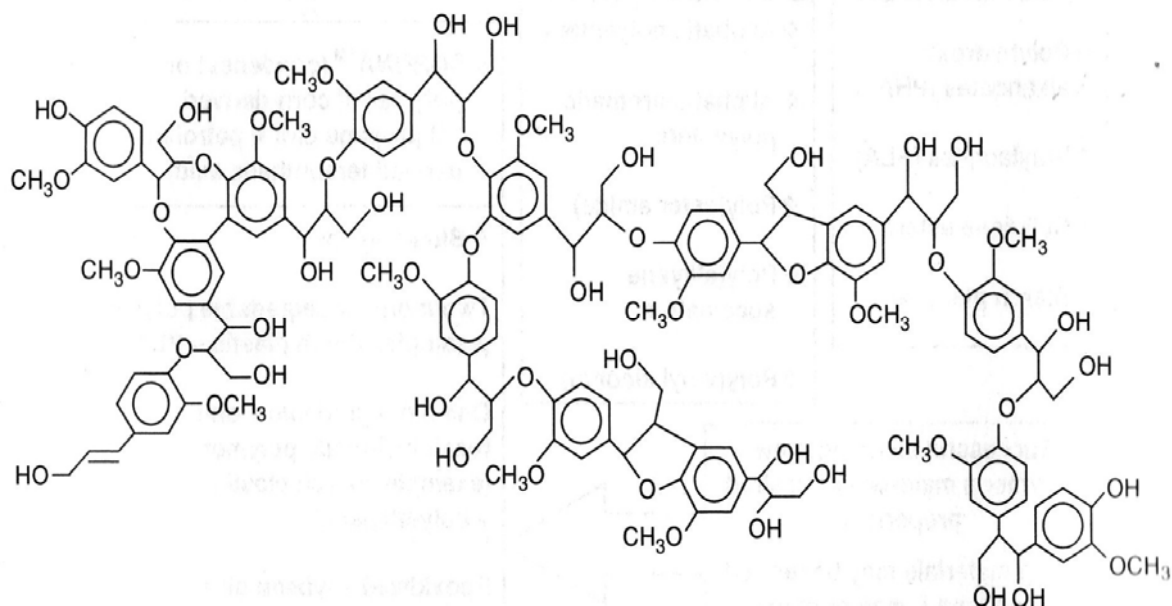


Figure 2.8: Representative structure of lignin (Mohanty *et al.*, 2005).

2.2.1.4 Extractives

The various minor constituents of wood which are soluble in organic solvents are collectively classified as extractives. Two major categories exist, namely wood resins and polyphenols. The former includes resin acids such as abietic acids, triglycerides, long chain alcohols and sterols. On the other hand, the polyphenols are comprised of lignans, or dimeric phenyl propane compounds, tannins, flavones, catechins and stilbene derivatives. The variety and types of extractives differ considerably from one species to another (Hamad, 2002).

2.2.1.5 Jute Fibre

Jute is a long, soft, shiny vegetable fibre that can be spun into coarse, strong threads. It is produced from plants in the genus *Corchorus* (*Corchorus olitorius* and *Corchorus capsularis*), family Tiliaceae. It is produced mainly in Bangladesh, India and Brazil. In the trade there are usually two names of jute, White and Tossa. *Corchorus capsularis* is called White Jute and *Corchorus olitorius* is called Tossa Jute.

In India & Bangladesh Roselle is usually called Mesta. Jute fibre (JF) is finer and stronger than Mesta and are, therefore, better in quality (International Jute Study Group, 2003).

The JF comes from the stem and ribbon (outer skin) of the jute plant. The fibre is first extracted by retting from woody part of the stem. The retting process consists of bundling jute stems together and immersing them in low, running water. There are two types of retting: stem and ribbon. The action involves water, micro organisms, and enzymes, and takes between 5 and 30 days to completion, depending on the temperature of the water. After the retting process, stripping begins. In the stripping process, non-fibrous matter is scraped off, then the workers dig in and grab the fibres from within the jute stem (Chawla, 1998).

Jute is one of the cheapest natural fibres and is second only to cotton in amount produced and variety of uses. The JF is composed primarily of the plant materials cellulose (major component of plant fibre) and lignin (major components wood fibre). It is thus a lignocellulosic fibre that is partially a textile fibre and partially wood. It falls into the bast fibre category (fibre collected from bast or skin of the plant) along with kenaf, industrial hemp, flax (linen), ramie, etc (Chawla, 1998; International Jute Study Group, 2003). The industrial term for jute fibre is raw jute. The fibres are off-white to brown, and 1–4 meters (3–12 feet) long.

The JF have a multicellular structure, composed of micro fibrils and the cross section is highly non-uniform. The JF is of particular interest in that its composites have high impact strength and which in addition to having moderate tensile and flexural strength properties compared to other lignocellulosic fibres (Wollerdorfer & Bader, 1998). Despite the attractiveness of JF reinforcement polymer composites, several disadvantages of jute, such as poor wet ability, poor

fibre-matrix adhesion, intrinsic polarity due to the presence of hydroxyl and carbonyl groups in their structure, and low moisture resistance make it insufficient for proper reinforcement (Ahmed *et al.*, 2006; Aquino *et al.*, 2007, Dhakal *et al.*, 2007; Md Akil *et al.*, 2009; Uma Devi *et al.*, 2004). Table 2.1 shows chemical composition of JF and some other natural fibres for natural fibres reinforced polymer composites.

Table 2.1: Chemical composition and moisture content of few natural fibres (Adopted from Bismarck, 2005).

Fibre	Cellulose (wt %)	Hemicelluloses (wt %)	Lignin (wt %)	Pectin (wt %)	Moisture Content (wt %)
Flax	71	18.6-20.6	2.2	2.3	8-12
Hemp	70-74	17.9-22.4	3.7-5.7	0.9	6.2-12
Jute	61-71.5	13.6-20.4	12-13	0.2	12.5-13.7
Kenaf	45-57	21.5	8-13	3-5	-
Ramie	68.6-76.2	13.1-16.7	0.6-0.7	1.9	7.5-17
Sisal	66-78	10-14	10-14	10	10-22
Banana	63-64	10	5	-	10-12
Cotton	85-90	5.7	-	0-1	7.85-8.5
Coir	32-43	0.15-0.25	40-45	3-4	8

2.2.2 Glass Fibre

Glass fibre (GF) is by far the world's most important continuous inorganic composite reinforcing fibre. Commodity or general purpose products are characterized by universal applicability, large sales volumes, and low unit cost. They present nearly 99% of commercial GF market. The remaining GFs are niche or special purpose products, characterized by special, and therefore premium, properties, small sales volumes, and high unit costs. They present 1% of the GF market. Due to its extensive usage, it is available in a wide variety of configurations, including

continuous filaments with twisted or parallel strands, short chopped fibres, woven cloth, roving and preregs (Miller, 1996).

In principle, five generic fibre-forming processes are known to yield glass or glass ceramic fibre from strong, fragile and in viscid melts. They are downdrawing fibres from preforms, drawing fibres from bushing tips, updrawing fibres from their melts, extruding fibres into a chemically reactive environment and lastly forming fibres in a containerless laser melt process (Wallenberger, 2000). These processes are schematically shown in Figure 2.9.

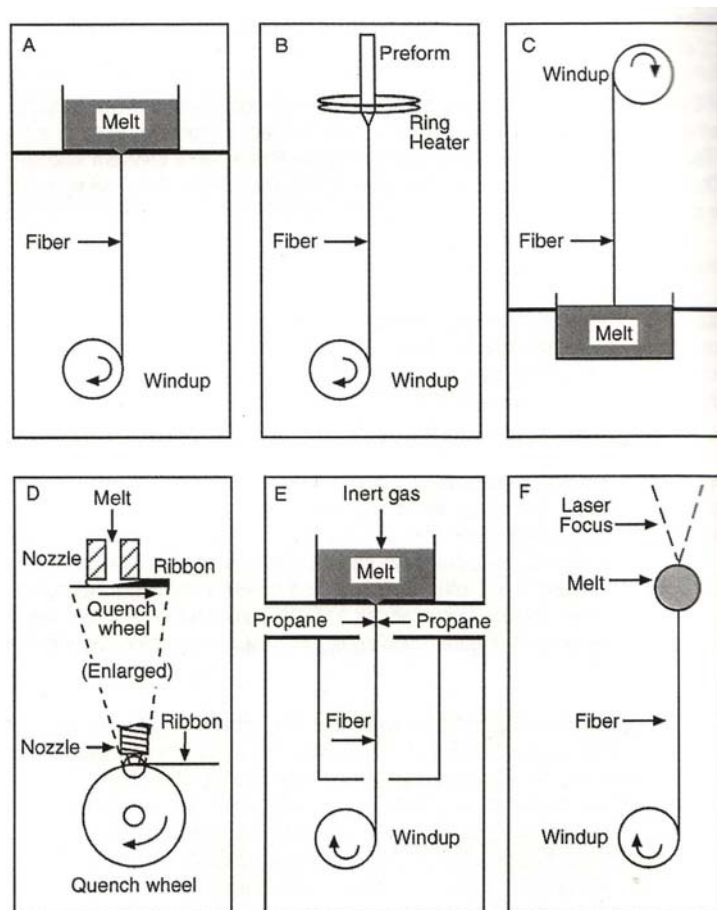


Figure 2.9: Generic fibre forming processes. Drawing fibres from a bushing (A), downdrawing fibres from a preform (B), updrawing fibres from a supercooled melt (C), rapid solidification of a metal ribbon on a quench

wheel (D), extruding fibres from a bushing into a chemically reactive environment (E), and drawing fibres from an acoustically levitated, therefore, containerless melt (F) (Wallenberger, 2000).

Silica based GF has been around for a long time. Common GF is readily available commercially in a variety of different chemical compositions. Typical general and special purpose fibre compositions contain ~50-60 % silica and contain a host of other oxides of Ca, B, Na, Al, Fe, etc. Table 2.2 gives the compositions of some commonly used GFs in composites. The designation E stands for electrical as E glass is a good electrical insulator besides having good strength and reasonable Young's modulus; C stands for corrosion as C glass has a better resistance to chemical corrosion; S stands for higher silica content and S glass is able to withstand higher temperatures than others. It should point out that more than 90% of all continuous GF produced is of E-glass type (Chawla, 1998; Miller, 1996). Typical mechanical properties of different GFs are summarized in Table 2.3.

Table 2.2: Chemical composition of some common GFs (Chawla, 1998).

Compound	E-glass	C-glass	S-glass
SiO ₂	55.2	65.0	65.0
Al ₂ O ₃	8.0	4.0	25.0
CaO	18.7	14.0	-
MgO	4.6	3.0	10.0
Na ₂ O	0.3	8.5	0.3
K ₂ O	0.2	-	-
Li ₂ O	7.3	5.0	-

Table 2.3: Typical properties of some GFs (Chawla, 1998).

Glass Fibre Type	Density (g/cm ³)	Tensile Strength (GPa)	Young's Modulus (GPa)
E	2.54	1.7-3.5	69-72
S	2.48	2.0-4.5	85
C	2.48	1.7-2.8	70

2.2.3 Comparison between Natural and Glass Fibres

The technological advantages of natural fibre reinforced plastics parts are primarily based on the potential for weight reduction, good processing and machining properties. Natural fibres have about 40% lower density than GF. During ecological evaluation, eco-balance, natural fibres also offer advantages in comparison with GF (Al Kafi *et al.*, 2006; Joshi *et al.*, 2004; Montazer & Salehi, 2008). The energy consumption to produce a natural fibre mat (9.7 MJ/kg), including cultivation, harvesting and fibre digestion, amounts to just less than one-fourth of the energy required to produce a GF mat (54.8 MJ/kg). From the standpoint of occupational medicine, natural fibre reinforced plastics have the advantage of not causing allergies, skin irritation and silicoses, contrary to GF dusts (Schlosser, 2004). Table 2.4 shows mechanical properties of some natural fibres as compared to E class GF. Typically, specific modulus of flax, hemp, jute, ramie and sisal fibres are comparable or even higher than E class GF.

2.3 Polymer Composites

Composites are defined as materials consisting of two or more distinct phase with an interface between them. This definition is basically used for materials containing reinforcements characterized by a high aspect ratio, as in the case for fibres, platelets and flakes. Polymer matrix composites (PMCs), also referred to as