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The preparation and properties of composites reinforced with natural fibres

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The Preparation and Properties of Composites Reinforced with Natural Fibres

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By

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Abstract

Woven flax reinforced and woven glass roving reinforced unsaturated polyester composites were fabricated at various fibre volume fractions. At equivalent fibre volume fractions, the warp tensile and flexural properties, as well as the Charpy impact strengths of both types of reinforced composites were compared. It was found that warp flexural and tensile strengths of woven flax reinforced composites were lower than the strengths exhibited by the woven glass composites. The specific stiffness of woven flax composites were also observed to be of a lower value than the glass reinforced composites specific moduli. The Charpy impact strengths exhibited by the glass composites were also a higher value.

The Tex (size) of the weft yarns within woven flax fabric were investigated to establish how they affect the warp and weft mechanical properties (flexural, tensile and Charpy impact strength) of epoxy composites when the flax fabric was used as reinforcement in two simple geometries. It was found that the warp and weft mechanical properties of woven flax fabric reinforced epoxy composites were influenced by the Tex of the weft yarn reinforcement. The stacking sequence used to arrange the woven flax reinforcement also influenced the mechanical properties of the composites. In addition, epoxy composites were fabricated containing 12 different types of woven flax fabric. An investigation was performed to establish the influence the weave type architecture of woven flax reinforcement has upon the flexural properties and Charpy impact strength of epoxy composites. The warp and weft flexural properties and Charpy impact strengths were observed to be significantly different in composites reinforced with plies of woven flax fabric that consisted of highly crimped warp yarns. It was also observed that the weave type of woven flax fabric reinforcement used in composites did affect their warp flexural properties, this was dependent upon the frequency of warp yarn crimping.

A study was undertaken to gain an understanding of the micromechanical processes that cause non-linear behaviour in flax fibre reinforced unsaturated polyester composites. Unidirectional composite bars reinforced with high quality flax sliver were fabricated at various fibre volume fractions. Tensile, flexural and Charpy impact tests were performed. Composite properties exhibited a linear rule of mixtures relationship with fibre content up to a fibre volume fraction of 60%. The response to straining of flax reinforced unidirectional composites bars under tensile loading conditions were recorded and analysed. Though exploring the deformation behaviour of flax composites *via* loading and unloading behaviour and acoustic emissions analysis, it was found that they undergo yielding at low values of stress and strain and that the reinforcing fibres are at the route of this behaviour. The existence of micro-compressive defects along the lengths of flax fibres and the effect the defects have upon stress-transfer are the likely cause of the observed deformation behaviour.

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Abbreviations

(In order of appearance)

PMC	Polymer Matrix Composite
UK	United Kingdom
USA	United States of America
GJ	Giga Joule
EU	European Union
ARL	Aero Research Limited
RH	Relative Humidity
dp	Degree of Polymerisation
LPC	Lignin-Polysaccharide Complex
S_1	Outer Layer of Secondary Cell Wall
S_2	Middle Layer of Secondary Cell Wall
S_3	Inner Layer of Secondary Cell Wall
H	Hours
CMC	Carboxymethyl Cellulose
PVA	Polyvinyl Alcohol
PEEK	Polyether-Ether-Ketone
MDF	Medium Density Fibreboard
kJ	Kilo Joule
ROM	Rule of Mixture
$[]_S$	Symmetrical
RTM	Resin Transfer Mould
CSM	Chopped Strand Mat
SEM	Scanning Electron Microscope
EFB	Oil Palm Empty Fruit Bunch Fibre
VARTM	Vacuum Assisted Resin Transfer Moulding
ESEM	Environmental Scanning Electron Microscope
CCD	Close Circuit Digital Camera

SFFT	Single Fibre Fragmentation Tests
N	Newton
IMS	Industrial Methylated Spirits
TLC	Thin Layer Chromatography
Diff	Difference
Min	Minute
Co	County
UnM	Unmodified Flax Sliver Reinforced Unidirectional Composite
MeA	Methacrylic Anhydride Modified Flax Sliver Unidirectional Composite
PrA	Propionic Anhydride Modified Flax Sliver Unidirectional Composite
WPG	Weight Percent Gain
OH	Hydroxyl
AE	Acoustic Emissions

Nomenclature

(In order of appearance)

Symbol	*Unit*	*Parameter*
Tex	(g km^{-1})	Linear density of fibres/filaments or yarns
kTex	(kg km^{-1})	Linear density of fibres/filaments or yarns
mTex	(mg km^{-1})	Linear density of fibres/filaments or yarns
σ_c	(MPa)	Composite applied stress
V_f	(-)	Fibre volume fraction
$\overline{\sigma}_m$	(MPa)	Volume-averaged matrix stress
$\overline{\sigma}_f$	(MPa)	Volume-averaged fibre stress
τ_i	(MPa)	Interfacial shear stress
τ_{i^*}	(MPa)	Critical interfacial shear stress
σ_f	(MPa)	Axial fibre stress
E_f	(GPa)	Fibre Young's modulus
ε_1	(-)	Applied composite strain
S	(-)	Fibre aspect ratio
R	(m)	Fibre radius
L	(m)	Fibre half length
n	(-)	Dimensionless constant
x	(m)	Axial distance from the fibre mid-point
E_m	(GPa)	Matrix Young's modulus
ν_m	(-)	Matrix Poisson's ratio
σ_{1^*}	(MPa)	Composite stress at the onset of inelastic behaviour
τ_{if}	(MPa)	Frictional interfacial shear stress
S_c	(-)	Critical fibre aspect ratio
σ_{fu}	(MPa)	Fibre ultimate tensile stress

Θ	(°)	Contact angle
γ_{SV}	(J m^{-2})	Surface energy of the solid-vapour interface
γ_{SL}	(J m^{-2})	Surface energy of the solid-liquid interface
γ_{LV}	(J m^{-2})	Surface energy of the liquid-vapour interface
V_m	(-)	Matrix volume fraction
M_f	(kg)	Mass of fibre
V_c	(m^{-3})	Volume of composite
ρ_f	(kg m^{-3})	Fibre density
M_c	(kg)	Mass of composite
ρ_r	(kg m^{-3})	Density of cured resin
X_c	(-)	Composite property
X_f	(-)	Fibre property
X_m	(-)	Matrix property
E_1	(GPa)	Axial composite modulus
E_2	(GPa)	Transverse composite modulus
ξ	(-)	Adjustable parameter
σ_{1u}	(MPa)	Critical axial tensile stress of composite
σ_{2u}	(MPa)	Critical transverse tensile stress of composite
τ_{12u}	(MPa)	Critical shear stress of composite
ε_{fu}	(-)	Fibre strain at failure
ε_{mu}	(-)	Matrix strain at failure
σ_{mfu}	(MPa)	Matrix stress at the onset of fibre fracture
σ_{fmu}	(MPa)	Fibre stress at the onset of matrix cracking
σ_{ϕ}	(MPa)	Off-axis tensile failure stress of composite
ϕ	(°)	Loading angle
Wt	(-)	Fibre weight fraction

V_{vf}	(-)	Void volume fraction
σ_{uc}	(MPa)	Composite tensile stress at break
$V_{f\,\min}$	(-)	Minimum fibre volume fraction
E	(GPa)	Young's modulus
E_c	(GPa)	Composite flexural Young's modulus
ρ	(kg m^{-3})	Density
N	(-)	Cumulative event counts

1 INTRODUCTION

1.1 Background

Composite materials are constructed of at least two components. Typically there is a strong stiff material often elongated in shape, referred to as the 'reinforcement' embedded in a weaker compatible material known as the 'matrix'. Composite materials often demonstrate different properties to that of the parent materials used in their fabrication. Properties are usually improved so composites can qualify for higher-performance applications than the parent materials could achieve in isolation.

Generally, polymer matrix composites (PMC's) are reinforced with carbon-based or graphite fibres, glass-based fibres, boron fibres or synthetic polymeric fibres such as Kevlar, in either a thermosetting or thermoplastic polymer matrix. The above composites are established as workable engineering materials and are widely used for structural purposes (Matthews & Rawlings, 1993). Aircraft, automobile, leisure, electronic and medical industries rely on fibre-reinforced polymers.

World War II brought about large-scale exploitation of polymer matrix composites due to the demand from military applications. Rapid growth of the industry then followed with development in the UK of carbon fibres and in the USA of boron fibres in the 1960's. Carbon and boron fibres gave a significant increase in the stiffness of composites over the well-established glass fibre containing materials.

Nature has formed its own composite structures such as bone, shells and wood. In the case of wood, cellulose microfibrils act as the reinforcement bonded together in a matrix of lignin and hemicellulose. Currently there is a growing interest in utilizing plant fibres other than wood as reinforcement in thermoplastic and thermosetting polymer matrix composites. Man-made materials such as E-glass, carbon and aramid are expensive compared to natural fibres like flax (*Linum usitatissimum*), hemp (*Cannabis sativa*) and

jute (*Corchorus* sp.). The cost of aramid and carbon fibres is largely attributed to the high energy and raw material costs (Robson *et al.,* 1993). Plant fibres are also favoured due to lower density; and because they are often claimed to be recyclable, biodegradable and carbon dioxide neutral. Organic natural fibres such as flax, hemp and others have the potential to substitute glass fibre in many applications. These fibres are not fully exploited materials and are still limited to lower value products (Kohler and Kessler, 1999).

1.2 Why Look at Plant Fibres?

Environmental and economic concerns regarding existing well-established composite materials have brought about the incentive to use renewable resources. A resource that offers numerous types of usable fibres or fibre bundles is the plant kingdom.

Table 1.1 World annual productions of some plant fibres and viable growing regions (*Source*: FAOSTAT database, 2001).

Fibre type	*Plant Family*	*World Production (Metric tonnes)*	*Region*
Jute (*Corchorus sp.*)	*Tiliaceae*	2668832	World
Coir (*Cocos nucifera*)	*Arecaceae*	631790	World
Flax (*Linum usitatissimum*)	*Linacceae*	588221	UK/ Europe/World
Sisal (*Agave sisilana*)	*Agavaceae*	357401	World
Ramie (*Boehmeria nivea*)	*Urticaceae*	178950	World
Abaca (*Musa textiles*)	*Musaceae*	104430	World
Hemp (*Cannabis sativa*)	*Cannabaceae*	58611	UK/ Europe/World

Fibres occur either in fruits (cotton, kapok and coir), stems (jute, hemp, flax) or leaves (sisal, abaca). Table 1.1 shows world production of some plant fibres together with their world locality in 2001.

1.2.1 Costs

The European market for fibreglass composites is about 300,000 tonnes, approximately one third for automotive applications, at a cost of about £5.50 per kg (8.75 EUR per kg) for mats (Ellison and McNaught, 2000). At current market prices of 30 to 35 pence per kg (0.47 to 0.55 EUR per kg), the use of natural fibres in composite blends realises significant cost reduction possibilities. A tonne of plant fibres costs between $200 and $1000 US dollars (193 to 967 EUR per tonne). Glass fibre costs between $1200 and $1800 US dollars per tonne (1160 to 1741 EUR per tonne) (Bolton, 1995). Prices of flax fibres suitable for composites used in the German automotive industry fluctuate between DM 0.90 to 1.50 kg (0.46 to 0.76 EUR per kg). The price fluctuation is due to supply shortages, oversupply and textile fashion changes (Karus, 2000). The market price of long scutched flax fibre in the Netherlands is between €1300 and €1700 per tonne, short fibre (tow) €100 to €150 per tonne and shives costing €20 to €40 per tonne (Van Dam, 1999).

A comparison between prices of three man-made and four natural fibres is shown in Table 1.2. The table illustrates that all four-plant fibres cost less than E-glass and considerably less than aramid or carbon fibres.

Prices for natural fibres vary depending on the extent of processing and fibre quality. Another factor that can influence the price of natural fibres, are treatments that the fibres may require to enable composite production. This may involve chemical modification and/or surface treatment to the fibres. Processing the fibres into a workable form such as a non-woven mat or weaving to create a woven textile may be necessary for use as a composite reinforcement.

Table 1.2 Comparison between the costs of man-made and natural fibres (*Source*: Ivens *et al.*, 1997).

Fibre Type	*Cost (Euro/Kg)*
Carbon	30-50
Aramid	20-35
E-glass	1.5-2.5
Flax	0.5-1
Hemp	0.5-1
Jute	0.5
Sisal	0.5

1.2.2 Environmental considerations

Natural fibres have a major advantage over synthetic fibres in terms of environmental impact. Renewable resources consume CO_2 during growth and liberate CO_2 during disposal, (composting/incineration). For this reason plant fibres are often stated to be CO_2 neutral. Natural fibres only require small energy inputs for processing. The embodied energy content of plant fibres is around 4 GJ/tonne, for glass fibre, the most commonly used reinforcement in polymer matrix composites the energy content is around 30 GJ/tonne. Kevlar has an embodied energy content of 25 GJ/tonne and carbon fibre 130 GJ/tonne (Bolton, 1994). Incineration allows some of the sun's energy stored in plant fibres to be recovered after the product's life. The above methods of disposal are not available for glass fibre and the only reasonable method is landfill. However in 1999 the United Kingdom adopted the European Union Landfill Directive (1999/31/EC) that will drastically change the way in which waste is disposed. In the United Kingdom, synthetic fibres may be subject to extra costs after service by means of the landfill tax that currently is at £11 per tonne, but is set to rise by £3 per tonne per year from 2005/6.

Plant fibre crops offer an alternative use of land for farmers. Intensive farming has led to the over production of some food in Europe. Plant fibre crops could be used as break crops in food crop agricultural systems. Fibre hemp is one of the most beneficial crops to plant before cultivating cereal crops. Farmers planting wheat after hemp frequently experience yield increases of 10-20% (Bócsa and Karus, 1998). Fibre hemp crops also eradicate weeds and help retain soil moisture for the successive crop as a result of good ground-shading cover. This makes the soil easier to work and the farmer is less dependent on herbicides (Bócsa and Karus, 1998).

In terms of the working environment, the processing of plant fibres brings about less health risks than synthetic fibres for workers (Stamboulis *et al.*, 2001, Ivens et al., 1997; Bolton, 1994).

1.2.3 Fibre properties

Typical physical properties of synthetic fibres used in polymer matrix composites, and some natural fibres, are given in Table 1.3. Tensile properties of natural fibres generally do not compare well to that of synthetics. Plant fibres have densities usually between 1400 and 1500 kg m^{-3}, the densities of carbon and E-glass are significantly higher. In terms of specific properties, some natural fibres are competitive with E-glass on a stiffness basis. Specific properties are used for design and are obtained by dividing the mechanical property by the specific gravity. Apart from pineapple, plant fibres listed in Table 1.3 are inferior to E-glass in specific tensile strength. This negative aspect is not necessarily a problem since many composites are designed based on stiffness. The tensile strength of E-glass fibres presented in Table 1.3 are realistic if the fibres are tested immediately after production. The likely tensile strength of E-glass fibres is approximately 1500 MPa. The significant reduction in tensile strength is due to damages that occur to the fibres during possessing *e.g.* abrasion with other fibres and.

Table 1.3 Mechanical properties of synthetic fibres used in composites and some natural fibres (*Source*: Ivens *et al.*, 1997).

Fibre Type	*Tensile strength (MPa)*	*Specific tensile strength (MPa)*	*Young's modulus (GPa)*	*Specific Young's modulus (GPa)*	*Failure strain (%)*	*Density (kg m^{-3})*
E-glass	2500-3500	977-1367	70-73	27-29	3	2560
Carbon	2500-6000	1429-3158	220-700	126-368	0.5-2.0	1750-1900
Aramid	3500-4000	2431-2778	85-135	59-94	3-5	1440
Flax	500-900	357-600	50-70	36-47	1.3-3.3	1400-1500
Sisal	80-840	55-579	9-22	6-15	3-14	1450
Jute	200-450	143-321	20-55	14-39	2-3	1400
Hemp	310-750	209-536	30-60	20-41	2-4	1480
Banana	530-750	379-536	7-20	5-14	1-4	1400
Pineapple	400-1600	278-1111	34-82	24-57	0.8-1.6	1440
Cotton	300-600	200-400	6-10	4-7	6-8	1500

1.3 Glass fibre markets

Advantages such as cost and performance have ensured that 99% of all fibres used within the composites industry are glass fibres rather than 'exotic' fibres like aramid, carbon and boron. During 1996, the global glass fibre industry was worth about $4.3 billion and 2.3 million tonnes in size (Owens Corning, 1996). Western Europe is the second largest market for glass fibre after North America, with France, Germany and Italy as the dominant countries. The main uses of glass fibre composites are given in Figure 1.1.

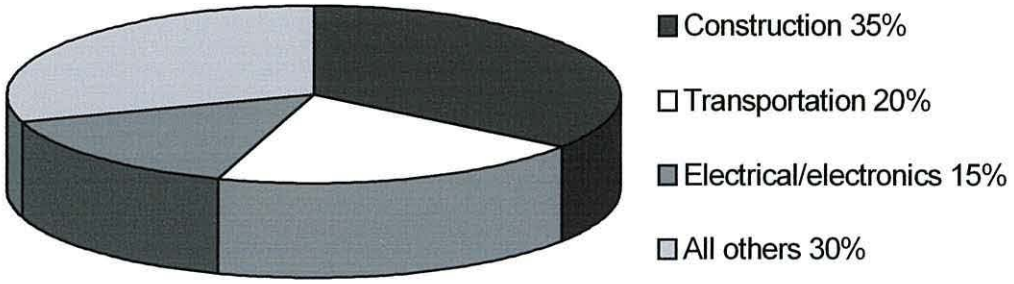


Figure 1.1 Global end-use segments for glass fibre composites in industry *(Source*: Owens Corning, 1996).

1.3.1 Feasibility of replacing glass

As Section 1.2.3 summarised, some plant fibres do compare favourably with E-glass fibres in terms of stiffness and strength, especially when the specific properties are considered. It is also often quoted that flax is available at a lower cost than glass fibres (Bolton, 1995; Ivens *et al.,* 1997; Ellison and McNaught, 2000; Brouwer, 2000).

It has been reported that Germany produced 1800 tonnes of flax fibre composite components for the automotive industry in 1996, and had increased production to 11000 tonnes by 1999 (Flake, 2000). It is clear that there is increasing industrial interest in plant fibres for technical uses, whether to create new products or to replace existing components. Smeder and Liljedahl (1996) identified the most important general properties for flax fibres in technical uses by focusing on market opinions in Sweden. Out of 11 feasible applications of flax, reinforcement was the main function of interest. Industry saw that the main problem was achieving a uniform distribution of the fibre in the material. The study also found that the cost of producing flax fibre and the advantages that flax might have in a new application, has been unfavourable for flax fibres in many cases. Two applications where flax fibres could achieve a niche in the

market are structural materials in transportation products, such as cars and aircraft, where weight is important and consumer products where environmental considerations are important (Riedel *et al.,* 1999).

A possible marketing opportunity is the development of plant fibre reinforced materials with biopolymeric matrices made of derivatives from cellulose, starch, lactic acid, *etc.* (Herrmann *et al.,* 1998). These so called 'biocomposites' are completely made from renewable resources and are biodegradable and can be fully integrated into natural cycles, even combusted for energy recovery. Man-made reinforcements and matrices contain compounds that are not biodegradable and these tend to be land filled or combusted depending on components. Due to the creation of slag waste, glass fibre cannot be effectively incinerated. Environmental concerns over conventional composites may help boost the market share for plant fibres. In the United States of America in 1988 there were 8000 landfill sites compared to only 2314 in 1998 (Netravali and Chabba, 2003). Limitations such as a shortage of space to landfill can only benefit materials that have other disposal alternatives.

However, problems arise when using plant fibres instead of glass fibres for reinforcement in composites. These are associated with composite production, performance in service and product life span and also with the agronomy and fibre processing, which must be dealt with before large scale exploitation is possible. The supply of the fibre raw material to industry has to be secure, constant and not subject to large price fluctuations to allow for future production strategies to be formed. The raw material has to have a consistent quality. For example, fibre quality of flax may vary with different successions of flax grown, climate conditions during growth, farm practice (*e.g.*, fertiliser usage, sewing density, desiccation timing and methods), and site quality. Flax that has been dew retted, stand retted, water retted, enzyme retted or steam exploded will all potentially have very different properties. Decortication processing parameters that retted flax is subjected to can further alter the fibre characteristics. Raw material feedstock to industry may have a constantly changing quality, causing numerous unforeseen difficulties for applications of quality control. For plant fibres to expand their market share within the European Union

(EU), Karus (2000) believes that a quality management scheme from cultivation to harvesting, through processing is required to ensure reproducible fibre qualities.

1.4 History of natural fibre reinforced composites

Natural fibres were used thousands of years ago in composite materials. The ancient Egyptians invented papier-mâché and the Inca and Maya used natural fibres as reinforcement in clay pottery to prevent the propagation of cracks (Gordon, 1976; McMullen, 1984). Composites, either reinforced with man-made or natural fibres bound in a synthetic resin, did not feature to any great extent before the 20th century and it was in 1909 that the composite industry was truly born. A resin referred to as 'Bakelite' was invented in 1909 and it was found that by adding wood flour the properties could be increased (McMullen, 1984). Phenol-formaldehyde the basis of Bakelite is a weak brittle resin but the addition of string and rag can increase its toughness and durability. Early composite materials were known then as 'reinforced plastics' and engineers in the 1930's were interested in using such materials instead of metals in aircraft structures (McMullen, 1984).

During the 1930's a composite was developed, which was reinforced with natural fibre by Aero Research Limited (ARL) (Bishop, 1997; McMullen, 1984). The composite material was referred to as 'Gordon Aerolite' and was one of the first structural man-made composites. The development of Gordon Aerolite arose from work Norman de Bruyne performed at ARL when trying to utilise cotton fibres to reinforce cured phenolic mouldings (Bishop, 1997). Norman de Bruyne abandoned the research work because of problems with the uniformity of composites, but Mr. Gordon found a solution by replacing cotton fibre with unidirectional flax fibres. Gordon Aerolite consisted of unidirectionally aligned flax thread in an unbleached state impregnated with phenolic resin (McMullen, 1984). A collection of impregnated threads was put together to form a 'skein' and these were hot press moulded to produce the final composites. Gordon Aerolite was used in the fabrication of a wing spar for the Bristol *Blenheim* and an

experimental fuselage for the Spitfire fighter because of concerns regarding aluminium supplies during the Second World War. Shortages of materials for the construction of the aircraft never occurred during war and the flax based fuselage was not required and the research discontinued (Aero Research Ltd., 1945). However, it is worth mentioning the properties of the material used in its construction.

The following details of Gordon Aerolite unless otherwise stated are from technical notes from Aero Research Ltd., (1945). Gordon Aerolite was comprised of untwisted fibres of flax impregnated with phenolic resin and was made on a machine into bands 15 cm in width. Sheets of the material could then be manufactured by placing the bands edge to edge to each other to form a ply and a second ply could then be over laid at an 90° angle (*i.e.* $[0/90/0]_s$). Once the correct number of plies had been stacked in order to achieve a desired thickness they were hot pressed to bond them into a single laminate. The material exhibited an ultimate tensile strength of 482 MPa and a Young's modulus of 48.2 GPa. The strength and stiffness along and across the laminate was equal but at 45° the strength and stiffness was only one half that along the fibre direction. A density of 1361 kg m^{-3} ensured that Gordon Aerolite achieved a specific tensile strength at 0° and 90° the same as that of duralumin, the material that it would be replacing in the event of shortages. The specific tensile stiffness at 0°, 90° and shear at 45° was about three quarters those of duralumin. Gordon Aerolite could be manufactured with a fibre volume fraction of 75%. This fibre volume fraction value is reported by Livingston Smith (1945) and for a unidirectional composite it is approaching the theoretical maximum. Livingston Smith, (1945) reports that the compressive strength of Gordon Aerolite parallel to the fibres was 200 MPa and 95 MPa 90° to the fibre axis. Shear strength was 37.8 MPa and Livingston Smith (1945) proposed that orientating some of the fibres within the laminate at 45° might have increased this figure. Aero Research Ltd., (1945) had constructed a Spitfire fuselage from flax fibre that had the same weight as the production fuselage in light alloy and passed flight standards for the time.

Along with the development of Gordon Aerolite, natural fibres were being utilised in the form of high-grade Kraft paper impregnated phenolic resin (McMullen, 1984). Cellulose

composites during the war were used in the construction of aircraft drop tanks, which saw service, but a number of other applications were also made such as a seat for the 'Spitfire'. Apart from the seat and drop tanks, cellulose based composites did not see service in structural applications on aircraft. This was due to the hygroscopic nature of the material and the high moulding pressures required in achieving adequate properties due to voids (McMullen, 1984; Livingston Smith, 1945).

Livingston Smith's, (1945) paper details how cellulose fibres may have a specific strength four or five times that of metals and how their incorporation can benefit the strengths of resins. Manila hemp paper based laminate bound with a phenol-formaldehyde resin achieved a tensile strength of approximately 186 MPa and Young's modulus of 13.7 GPa. Fabric based laminated sheets were also fabricated using cotton and tested in both warp and weft directions. Expensive high-grade cotton laminates often displayed less strength than the laminated paper but Livingston Smith (1945) reports that they had a greater resistance to shock (*i.e.* greater toughness). Brown (1947) also suggests that paper based materials were stronger than the fabric reinforced laminates but the impact strength of fabric laminates was two and half time that of spruce. At the time of Livingston Smith's paper (1945), cellulose-based composites of either paper or fabric were established and produced commercially along with research to establish and improve properties.

At this time natural fabric based composites were exhibiting higher strengths than the unreinforced resin but the improvement to stiffness was not that significant (Livingston Smith, 1945). The amount of crimp and twist in the yarns of the fabric were seen as factors that affected the property of stiffness.

The following section details the summary produced by Brown (1947) of the significant factors that affect the mechanical properties of woven reinforced composites, based on his experience. The fabric strength has little influence upon the strength of the composite, but the strength and stiffness of the fibre from which the fabric is composed has a significant influence on the composite. Composite performance is enhanced when

the fibres used in the fabric have high moduli and tensile strengths. Woven natural fibre requires scouring to remove sizes that otherwise prevent resin penetration in yarns. Desirable features of fabrics include soft yarns with minimum twist, finely woven with minimum crimp. Minimum yarn twist confers greater ease of resin penetration resulting in greater composite strengths and a low amount of crimp ensures that yarns lie as straight as possible. Brown (1947) also points out that if properties are desirable in a preferred direction then unidirectional fabrics can be used. Resins used should have high tensile strength and moulding pressures should be high, so that consolidation occurs between the two phases and the maximum density is obtained.

The developments of glass, carbon, boron and synthetic fibres, superior resins and manufacturing systems saw the decline in the use of long natural fibres. Industry and consumers demanding materials with high strength and stiffness to weight ratios, with low cost have also contributed to the decline. However, natural fibres have recently re-emerged as a subject for renewed research interest.

1.5 Current situation of plant fibre reinforcement for PMC

Many academic research projects utilising plant fibre reinforcement within thermosetting or thermoplastic polymers have been undertaken and reported. There is a great deal of literature on the mechanical and physical properties of plant fibres and types of composites that can be produced from them when incorporated with synthetic polymer resins such as polyester or epoxy. An area of research that has generated a great deal of interest is the potential use of renewable polymers as matrices for plant fibre reinforcement. For example Mwaikambo and Ansell (2003) used a cashew nut shell liquid matrix to fabricate non-woven and unidirectional hemp fibre composites. Other researches have explored the potential of improving the suitability of plant fibres for PMCs through various fibre treatments. The rationale for treating natural fibres is to reduce the number of disadvantages associated with them, for example the hydrophilic nature of the cell wall polymers that leads to undesirable changes in mechanical and

dimensional properties as well as degradation by decay organisms. Hill *et al.,* (1998) have investigated chemical modification through acetylation on coir, oil palm fibre, flax and jute fibres using acetic anhydride. Bisanda and Ansell (1991) modified sisal fibres by mercerisation and a silane treatment to improve adhesion characteristics and moisture resistance. Un-modified and modified fibre was used to fabricate unidirectional sisal-epoxy composites. Industry has also been active with research into the use of natural fibres in polymer matrix composites. An example is DaimlerChrysler who have succeeded in making under floor encapsulation panels for vehicles from natural fibre reinforced polymers (DaimlerChrysler, 2001). A large proportion of literature is concerned with composites reinforced with flax, hemp and jute fibre in the form of non-woven or unidirectional mats bound with a thermosetting polymer matrix. Oksman (1999) has reported on the mechanical properties of unidirectional epoxy composites reinforced with flax. Roe and Ansell (1985) fabricated unidirectional jute reinforced polyester composites. O'Dell (1997) has used jute fibre non-woven mats and Sèbe *et al.,* (1999 and 2000) used hemp fibre non-woven mats to reinforce resin transfer moulded unsaturated polyester composites. Hughes *et al.,* (1999) also used hemp in the form of non-woven felted mats as reinforcement in an unsaturated polyester resin matrix. O'Dell (1997), Sèbe *et al.,* (1999) and Hughes *et al.,* (1999) all compared the mechanical properties of composites reinforced with natural fibre against composites containing glass fibre reinforcement. O'Dell (1997) concluded that the jute fibres could be processed just as well as glass fibres in resin transfer moulding and that the flexural strength and tensile and flexural modulus of non-woven jute fibre composites were of the same order of magnitude as glass fibre-reinforced composites, although they were slightly lower. However, O'Dell (1997) states that the Izod impact of non-woven jute fibre composites is an order of magnitude lower than the glass fibre composites. Sèbe *et al.,* (1999) found that the impact properties of hemp composites also were insufficient to compete with glass in structural applications. Hughes *et al.,* (1999) also found that both tested properties (flexural, Charpy impact strength) of non-woven hemp reinforced polyester composites were always inferior when compared with glass chopped strand mat reinforced polyester composites.

The fibre architecture of non-woven plant fibre reinforcement has undoubtedly been a factor in their poor performance. However the use of plant fibres in polymer matrix composites in the form of woven fabric reinforcement may improve properties, especially toughness. Currently there is very little literature on composites that are reinforced with a woven reinforcement produced from natural fibres. Woven materials are composed of yarns, strands or fibres that are weaved together to form a fabric. Yarns, strands or fibres are interlaced in a regular pattern or weave style. The woven fabric is comprised of warp yarns, strands or fibres in a lengthwise direction with weft yarns, strands or fibres interlacing at 90 degrees. Mechanical interlocking of warp and weft fibres maintains the fabric's integrity.

The purpose of the research undertaken during this PhD was to further extend work that has taken place at University of wales Bangor since the early 1990's. This has been largely concerned with utilising non-woven felts as composite reinforcement. The work reported herein was confined to studying woven fabric reinforcements with some unidirectional composites studied in addition.

1.6 Justification

The research conducted for this thesis is novel, as much of the research into plant fibre reinforced thermosetting polymer matrix composites has not been aimed at woven reinforcements. The research that has been undertaken and reported previously has not looked at the influence that weft yarn structure has upon the mechanical properties of woven flax reinforced thermosetting composites when tested in either the warp or weft direction (Chapter 4). In addition, the effects that different weave types of woven flax reinforcement have upon the warp and weft mechanical properties of composites has also been studied and reported on in this work, because of the lack of information in the current literature (Chapter 5). As it has been reported that many natural fibre reinforced composites show yielding or non-elastic behaviour at relatively low levels of strain and stress it was deemed necessary to investigate the underlying causes of this behaviour.

Unidirectional flax composites were fabricated and used to investigate this phenomenon (Chapter 6). Unidirectional type composites are not as complex as composites containing woven reinforcement and it was for this reason why this type of composite system was used to investigate non-linear behaviour.

1.7 Background and rationale of the study

At present, research into the use of natural fibres for reinforcement in PMC's has mainly concentrated on using the fibres in a non-woven or in an aligned form (Dweib *et al.*, 2004; O'Donnell *et al.*, 2004; Mwaikambo and Ansell, 2003; Van de Weyenberg *et al.*, 2003; Shawkataly and Ismail, 2001; Hill and Shawkataly, 2000; Hepworth *et al.*, 2000; Hughes, 2000; Oksman, 1999; Bos and Van den Oever, 1999; Hughes *et al.*, 1999; Sèbe *et al.*, 1999; Hill *et al*, 1998; Devi *et al.*, 1997; O'Dell, 1997; Bisanda and Ansell, 1991; Sanadi *et al.*, 1986; Roe and Ansell, 1985).

There has been very little work undertaken that has investigated PMC's with natural fibre reinforcement in a woven form. The published material found and reviewed mainly deals with polyester composites reinforced with various woven jute fabrics (Maffezzoli *et al.*, 2004; Gassan, 2002; Gowda *et al.*, 1999; Ghosh and Ganguly, 1993). Mohanty *et al.*, (2004) also investigated woven jute fabric reinforced composites but they consisted of a thermoplastic biodegradable polyester matrix. Banana-cotton fabric reinforced polyester composites mechanical properties were compared to CSM glass fibre reinforced polyester composites by Satyanarayana *et al.*, (1996). Much of the work that has been undertaken using woven jute fabric as reinforcement in polyester composites has focused on either investigating composites mechanical properties or fatigue behaviour (Gowda *et al.*, 1999; Gassan, 2002) or the effect of chemically modifying the jute fibre within the woven material in an effort to improve its suitability as reinforcement (Ghosh and Ganguly, 1993). Gowda *et al.*, (1999) stated that 'no single group of researchers has completely characterised the mechanical properties of jute fabric reinforced polyester composites'and Gassan, (2002) stated that there was 'limited information available'.

Due to the apparent lack of information available on woven plant fibre reinforced PMC's composites it was felt necessary to conduct research into these types of reinforced composites. Flax fibre is a widely available plant fibre that possesses very good mechanical properties when compared to other natural fibres and some man-made fibres. It is used presently and has been used historically in the production of linen. Considering the previous research undertaken it is timely to explore the potential of woven flax for the use as reinforcement in PMC's, as certain properties may be enhanced due to the bi-directional nature of the reinforcement and because the fabric reinforcement has its own integrity given to it by the interlacing and twisting of yarns. Furthermore, it is necessary to explore the mechanical properties of composites reinforced with woven flax fabric and compare their properties to equivalent conventional composites reinforced with man-made fibres. Certain variables, such as the Tex of the yarns within fabrics and the weave type that the fabric has been woven to form may influence the properties of PMC's and therefore these issues should be investigated. Both of these issues, to the best of my knowledge, have not been investigated when woven flax fabric is used as reinforcement in a thermosetting matrix composite system.

As mentioned above, many studies have been undertaken to investigate the mechanical properties of unidirectional composites reinforced with natural fibre. Researchers have also analysed the stress-transfer at the fibre to matrix interface and the deformation behaviour of individual fibre micro-tensile composites (Eichhorn and Young, 2003 and 2004). Hughes (2000) has studied the effect that fibre defects have upon the stress-strain field in the surrounding matrix of single fibre epoxy composites. Hughes (2000) and Hughes *et al.*, (2002) also studied bast fibre reinforced unsaturated polymer composite systems and observed non-linear behaviour to occur at low values of stress and strain.

Few studies however, have been undertaken that consider the nature of the deformation behaviour of natural fibre thermosetting polymer composite systems and how this relates to the structural applications of these materials. Therefore an investigation studying the mechanical properties and more importantly the deformation behaviour of flax fibre

reinforced unidirectional unsaturated polyester composites was deemed necessary in order to gain an understanding of the microstructural processes operative.

2 SPECIALISED LITERATURE REVIEW

2.1 Plant fibre types

Fibres occur in plants in their seed: *e.g.* cotton *(Gossypium barbadense)*, coir *(Cocos nucifera)*, kapok *(Ceiba pentandra)*; the stem *e.g.* flax *(Linum usitatissimum)*, hemp *(Cannabis sativa)*, ramie *(Boehmeria nivea)*, kenaf *(Hibiscus cannabinus)*; or the leaves *e.g.* sisal *(Agave sisilana)*, abaca *(Musa textiles)*. World annual production and locality of popular plant fibres is shown in Table 1.1 on page 2.

Stem fibres are referred to as 'bast fibres'. Bast fibres form the fibrous bundles in the inner bark of the stems which help to hold the plant erect. Bast fibres such as flax offer strength and stiffness and can be grown in temperate and sub-tropical regions of the world. *Linum usitatissimum* is an important fibre in the textile industry because it can be spun into a yarn for linen. Jute is the most commonly used bast fibre globally, having reasonable strength and a degree of resistance to rot. Jute is used for many applications due to its abundance and low cost it has become an important fibre for sacks and packing cloths. Good quality jute fibre is used for curtains and furnishing fabrics. Hemp fibres are used throughout the world. Hemp fibre has been used to make fine fabrics similar to linen but is currently used for coarse fabrics such as sacking and canvas.

Leaf fibres occur in monocotyledenous plants and provide strength to the leaves. Leaf fibres are often coarser than bast fibres and have achieved great commercial importance for use as ropes, cordage and textile fabrics. Sisal fibres can be 60-120 cm in length and are strong and stiff. The stiffness of sisal has limited its uses but it still remains as one of the most valuable cordage fibres. It is used extensively for baler twine and sacks. Strength and sisal's ability to take up acid dyestuffs has made it an attractive fibre for textile uses, such as matting and rugs. Commercial abaca fibre is another leaf fibre from the *Musaceae* family of plants. Good quality abaca fibre can have strands up to 4.5 meters in length. The fibres are strong and flexible and salt water tolerant. High-grade

paper, such as teabags and stencil tissue is the main market of abaca fibre but there are other smaller markets such as fishing nets and ropes.

Cotton, the most famous and important plant fibre is an example of a seed fibre (Riccio and Orchard, 1999). Cotton fibre is very strong especially when wet. Coir is another seed fibre that comes from the husks of coconuts. It consists of short thick walled fibres that are used for brushes, matting and cordage.

2.2 Structure of flax and hemp plants

Flax and hemp are annual plants that produce long stalks, which can grow to a height of 90 to 130 cm and 150 to 300 cm, respectively (Bócsa and Karus, 1998; Catling and Grayson, 1982; Jarman, 1998; Benjamin and Weenen, 2000). Stalks of hemp are hexagonally shaped and are 4 to 26 mm in diameter depending on the plant spacing during growth and sex of the plant. Hemp male plants can be 10 to 15% taller than female plants but have thinner stalks and often grow fewer branches (Bócsa and Karus, 1998). Leaves on hemp are fine pinnate shaped containing several pinnations (the number depends on variety). Flax stems are 4 to 5 mm in diameter, are smooth and free of hairs (Catling and Grayson, 1982). Leaves are narrow and oval shaped, tapering to a point at each end. Dicotyledonous plants such as flax and hemp contain bast fibres that are bundles or strands beneath the epidermis and cortex layers as Figure 2.1 shows. Bast fibres are extremely useful materials for textiles, ropes and many other products after retting and decortication processes which are explained in a later section.

Breaking the stem structure down, it is possible to obtain the fibre bundles or technical fibres (bast fibres) as some literature refers to them. Flax fibre bundles that are only a few centimetres long are referred to as 'tow fibre' but bundles can be up to 1 m long and these are called 'line fibre'. Fibre bundles from hemp can be up to 2 m in length (Cook, 1993). Details of the dimensions of individual fibres (fibre ultimates) within bundles can be found detailed in Section 2.3.

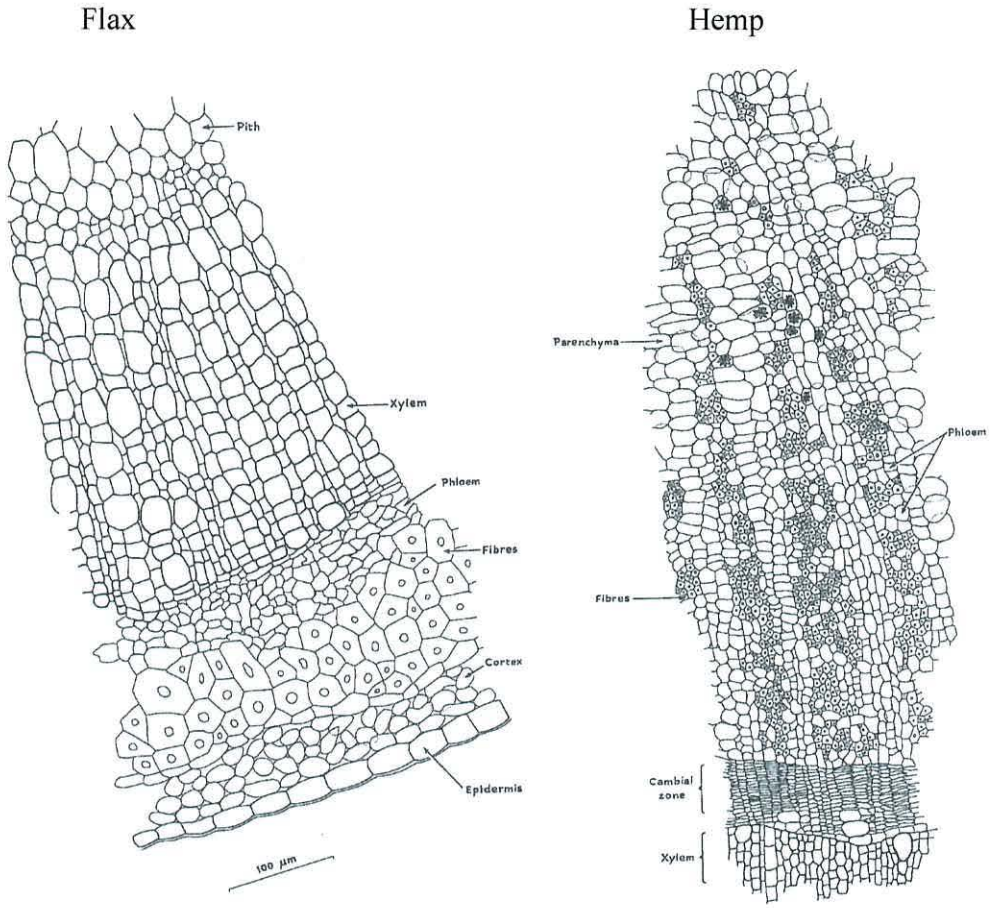


Figure 2.1 Transverse sections of flax and hemp stems (*Source*: Catling and Grayson, 1982).

2.3 Properties of flax and hemp fibres

Bast fibres from flax and hemp have the potential to be a good reinforcement for composites. They display highly elongated fibre cells (good aspect ratio) and have a lower density than glass fibre and can exhibit acceptable mechanical properties, especially stiffness. However plant fibres are not uniform nor cylindrical nor smooth surfaced. They are also variable, not water resistant nor decay resistant.

Numerous problems arise when obtaining values for plant fibre physical and mechanical properties. It is important to know whether the mechanical properties reported refer to

fibre bundles (groups of fibres bunched together) or fibre ultimates (single fibre). Fibre variability is often high, even when large numbers are tested (Table 2.2 on page 23). Possible explanations for this are that properties may change depending on the location in the plant the fibres are taken from; maturity of plant; the farm practice used; variety of flax/hemp; growing conditions and the type of retting and processing implemented. As Morvan *et al.,* (2003) described in a flax review paper, the fibre diameter is larger in the middle part of the stem rather than the slower growing top and bottom regions of the stem. Fibre diameter of the succession of flax used (*Var.* Natasha) was also found to be different depending on the development stage of the plant. Table 2.1 shows flax fibre diameters from different stem locations and development stages of the plant.

Table 2.1 Average flax fibre diameters from three stem locations and three development stages (*Source*: Morvan *et al.*, 2003).

	Fibre diameter (μm)		
Development stage	*Stem bottom*	*Stem middle*	*Stem top*
Flowering/capsulation	11	9	7
Mature capsule	26	19	16
Seed maturation	30	16	15

Catling and Grayson (1982) measured the diameter of single flax fibres and found that the average was 19 μm with a range of 11.68–31.96 μm. Average hemp fibre diameters were 30 μm with a range of 16.27-67.10 μm. Olesen and Plackett (1999) estimated fibre diameters of single flax and hemp fibres finding that the average for flax was also 19 μm and hemp 25 μm. Due to the non-uniformity of the cross sectional shape of flax and hemp fibres, the diameter is difficult to measure accurately.

Olesen and Plackett (1999) found that the average length of flax single fibres was 33 mm from a range between 9-70 mm. Hemp fibre length was 25 mm on average from a range

of 5-55 mm. Catling and Grayson (1982) found the mean length of flax fibres to be 7.9 mm with a range of 1.6-24 mm. Hemp single fibre length was 8.46 mm with a range of 1-34 mm. Morvan *et al.,* (2003) reports that the average flax fibre length is between 20 to 50 mm. Large ranges of fibre lengths exist for flax and hemp within published results and between published results that may be explained by the factors mentioned earlier in the section.

Obtaining mechanical properties of flax and hemp fibres can be a difficult task, especially when extracting fibre ultimates without causing damage to the fibre. Fibres extracted by hand in a laboratory will have different properties to fibres that have been obtained mechanically. It is extremely difficult to measure the cross sectional area of fibres and it is likely that a high variation will exist within a test population.

Davies and Bruce (1998) showed the effects of environmental conditions and mechanical damage induced during processing, on the strength and stiffness of flax and nettle (*Urtica dioica*) fibre. Fibre was obtained from dew retted French flax and nettles were collected from local parks. Nettles were air dried for one month. Fibres from both plant types were extracted by hand with care taken not to apply a tensile stress. A mean cross sectional area was calculated for each fibre tested under a light microscope. Single fibres mounted on cardboard with an 8 mm gauge length were placed into grips on a testing machine in a test chamber. Time was allowed for the fibres to equilibrate with the conditions in the test chamber. It was found that the mean strain to failure for flax was 1.33%, and for nettle fibres it was 1.65%. Mean failure stress for flax fibres was 621 MPa with a mean modulus of 51.7 GPa. Nettle's mean failure stress was 368 MPa with a modulus of 25.5 GPa. Fibre damage was measured by looking at the fibres with a polarizing microscope under crossed polars. It was believed that undamaged fibres appeared dark but damaged areas of fibres appeared bright. A qualitative measure of damage was recorded by measuring the proportional area of the bright regions of the fibre. This does not measure the severity of the damage. Higher proportions of fibre damage caused decreases in the moduli for both fibre types. As relative humidity increased the fibre's stiffness decreased for a given amount of fibre damage. The range

of relative humidity that fibres were tested in was 30 to 70%. It was found that flax modulus decreased with increased relative humidity at a rate of 0.39 GPa/%RH. Fibre strength was found to decrease, with an increase in fibre damage. However, fibres can contain weak points where failure occurs that does not appear damaged under inspection.

Table 2.2 Mechanical properties of flax and hemp fibre from different publications.

Fibre type	*Tensile strength (MPa)*	*Young's modulus (GPa)*	*Elongation at break (%)*	*Reference*
Flax	345 - 1035	27.6	2.7 – 3.2	Netravali and Chabba, 2003
	1216	-	4.1	Jarman, 1998
	500 - 900	50 - 70	1.3 – 3.3	Ivens *et al.*, 1997
	800 - 1500	60 - 80	1.2 – 1.6	Brouwer, 2000
	1100	100	2.4	Bledzki *et al.*, 1996
	2000	85	-	Bolton, 1994
Hemp	690	-	1.6	Netravali and Chabba, 2003
	1235	-	4.2	Jarman, 1998
	310 - 750	30 - 60	2-4	Ivens *et al.*, 1997
	550 - 900	70	1.6	Brouwer, 2000
	690	-	1.6	Bledzki *et al.*, 1996
	895	25	-	Bolton, 1994

Mwaikambo and Ansell, (2003), tested hemp fibre bundles that had been mercerised in concentrations of caustic soda ranging from 0.8–8% to change their surface morphology. No information was available about prior processing conditions that the hemp had undergone. Fibre bundles were tested with a gauge length of 19 mm. The specific tensile strength of the fibre bundles increased as the fibre bundle diameter decreased following higher concentrations of caustic soda treatments. The optimum concentration was found

to be 6% for tensile strength and 4% for fibre modulus. Treated fibre bundles achieved a tensile strength of 1064 MPa and a Young's modulus of 65 GPa. Untreated fibre bundles had a tensile strength of 591 MPa and a Young's modulus of 38 GPa. Increases in mechanical properties are due to the swelling of cell walls and partial removal of hemicellulose that helps break alkali-sensitive bonds between components within the fibre. Caustic soda makes the fibres more consistent, improves stress transfer between single fibres and creates new hydrogen bonds.

Table 2.2 shows mechanical properties of flax and hemp fibres from different publications. Notice the large variations that exist due to some of the factors mentioned in this section.

2.3.1 Chemical constituents of bast fibres

Kohler and Kessler (1999) describe fibres as 'complex natural composites' whose properties depend on the degree of polymerisation of cellulose, arrangement of fibrils, related crystallinity, and the amount of non-fibrous molecules such as hemicellulose, pectin and lignin. Table 2.3 shows the percentages of chemical constituents as published by various sources.

Table 2.3 shows that a great deal of variation exists between published results concerning the amounts of individual components. It can be assumed that the variation exists due to differences in flax and hemp brought about by conditions during growth and the variety used. Methods used to obtain results may also affect the outcome. Olesen and Plackett (1999) and Garcia-Jaldon *et al.*, (1998) have reported high quantities of pectin. The percentages of pectin within flax and hemp fibres from the other sources reported in Table 2.3 are considerably lower. Olesen and Plackett (1999) made 'qualified guesstimates from the published data' to derive their percentages of chemical constituents of flax fibre. Garcia-Jaldon *et al.*, (1998) state that the amount of cellulose is

overestimated in some literature and that the amount of pectin is underestimated, compared to their findings.

Table 2.3 Chemical compositions of flax and hemp fibres published from various sources.

Fibre type	*Cellulose (%)*	*Hemicellulose (%)*	*Pectin (%)*	*Lignin (%)*	*Other (%)*	*References*
Flax	68 – 85	10 – 17	5 - 10	3 – 5	1 – 2	Olesen and Plackett, 1999
	64.1	16.7	1.8	2	5.4	Jarman, 1998
	76	-	11*	-	-	Edwards *et al.*, 1997
	71	18.6	2.3	2.2	1.72	Bledzki *et al.*, 1996
	81	14	2	3	-	Bolton, 1994
Hemp	67	16.1	0.8	3.3	2.8	Jarman, 1998
	74.4	17.9	0.9	3.7	0.8	Bledzki *et al.*, 1996
	74	18	1	4	-	Bolton, 1994
	55	16	18	4	7	Garcia-Jaldon *et al.*, 1998

*Includes lignin and pectin.

2.3.1.1 Cellulose

Cellulose is the major constituent of flax and hemp fibre cells. Cellulose is comprised of β-D-anhydroglucopyranose units ($C_6H_{12}O_5$) that are linked together to from a long thin filament structure. Each unit (monomer) is rotated 180° to maintain a straight chain (Desch and Dinwoodie, 1996). Linkages between anhydroglucopyranose units occur through condensation reactions between adjacent carbon 1 and 4 positions. These linkages are referred to as 'β- (1→4)-glycosidic bonds' (Focher, 1992; Fengel and Wegener, 1989). Two anhydroglucose units are linked form a 'cellobiose unit' that has a length of 1.03 nm (Fengel and Wegener, 1989). Figure 2.2 shows part of the molecular

chain of cellulose. The number of monomers linked together refers to the degree of polymerisation (dp) of a polymer; Fengel and Wegener (1989) state that for plant cellulose the dp range is 7000 to 15000.

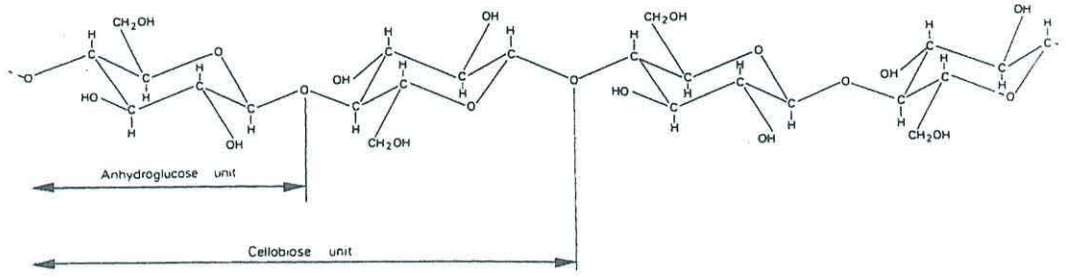


Figure 2.2 Section of the molecular chain of cellulose, showing four glucose units (*Source*: Desch and Dinwoodie, 1996).

The dp of cellulose for retted flax is normally within the range of 2500-3000, depending on growing and retting conditions (Focher, 1992). Covalent bonds join anhydroglucose units together giving strength to the length of the chain. Hydroxyl groups along the cellulose polymer can form two different hydrogen bonds depending on the location on the glucose monomers. Bonds can be formed that are called 'intramolecular linkages'. These are when hydrogen bonds occur within a cellulose molecule. The other bond type, 'intermolecular linkages' are hydrogen bonds that form between adjacent cellulose polymers (Fengel and Wegener, 1989). Intramolecular linkages help stiffen the chain whilst intermolecular linkages give rise to supramolecular structures (Fengel and Wegener, 1989). Intermolecular linkages form cellulose molecules into sheets that can pack together to create a crystalline structure. These cellulose chains create the reinforcement structure 'microfibrils'. Desch and Dinwoodie (1996) state that there are 48 molecular chains of crystalline cellulose at the core of microfibrils in the secondary cell wall that have a cross section of 5x3 nm. Microfibrils have a cross section of 10x5 nm. The cellulose surrounding the core of the microfibril is present in a non-crystalline state (amorphous cellulose) as well as other molecules such as hemicellulose and lignin (Desch and Dinwoodie, 1996). Fengel and Wegener (1989) state that for the bast fibre 'ramie' about 80–70% of cellulose is in a crystalline state.

2.3.1.2 Hemicellulose

Hemicellulose differs from cellulose by having a lower molecular weight (shorter chains), it is comprised of various sugar units, and is often non-linear and branched in structure (Bolton, 1994; Fengel and Wegener, 1989; Focher, 1992). Monomeric units joined by various glycosidic linkages form the backbone of hemicellulose. The monomeric units consist of D-glucose, D-galactose, D-mannose, D-xylose and L-arabinose with glucuronic and galacturonic units present (Focher, 1992). The major non-cellulosic polysaccharides in flax fibre bundles are xylans, mannans and galacturonans (Focher, 1992). The dp of hemicellulose is between 150 to 200 monomers in each molecule (Desch and Dinwoodie, 1996). Hemicelluloses are found in the middle lamella, primary wall and in the secondary cell wall where it is bonded with cellulose and lignin to form the thickest cell wall layer (Focher, 1992). Structure and quantities of the chemical constituents of the secondary cell wall region often determine the mechanical properties of fibres. It is known (Fengel and Wegener, 1989) that lignin is not just present throughout the cell walls but is bonded to the polysaccharides. The hydrophilic nature of the surface of cellulose is not compatible with hydrophobic lignin limiting H-bonding. Hemicellulose acts as a coupling agent between crystalline cellulose and lignin creating a lignin-polysaccharide complex (LPC). Hemicellulose can covalently bond to lignin and hydrogen bond to cellulose. Hydroxyl groups from the different sugars of hemicellulose cannot bond with all hydroxyl groups on cellulose because some are misaligned and others have been substituted with acetyl groups (steric hindrance). The imperfect bond that occurs between cellulose and lignin *via* hemicellulose allows stress transfer to the microfibrils but creates a weak interface in the structure that allows failure to occur giving the overall structure the toughness required.

2.3.1.3 Pectins

Pectic substances are found in the middle lamella and primary cell walls and are acid polysaccharides with a high molecular weight. Pectic substances have a linear main chain of (1→4) linked α-D-galacturonic acid (Focher, 1992). Regular intervals of α-(1→2) and α- (1→4) bonded rhamnose units branch from this chain. Arabinose and galactose molecules are present as side chains with small amounts of xylose and glucose monomers (Fengel and Wegener, 1989). Pectins along with hemicellulose bind cell wall layers (Focher, 1992; Mooney *et al.,* 2001). Removal of pectic material facilitates the separation of fibre bundles from the stem. The removal or degradation of pectic substances is extremely important during retting processes if fibre bundles are to be obtained. The chemical composition of pectic substances in flax fibre varies within the location of the plant tissues, fibre variety, and the type and duration of retting (Focher, 1992). Mooney *et al.,* (2001) believes that the characterisation of pectic polymers after retting is of significant interest because pectic substances are thought to have a structural role in binding cells, thus contributing to the tensile strength of fibre bundles and having an influence on the dye-ability of flax for textiles.

2.3.1.4 Lignin

Lignin is a complex, highly cross linked, non-crystalline, aromatic polymer with a molecular weight obtained from extracted material to be as high as 11000 (Desch and Dinwoodie, 1996). The constituents for all lignins are *p*-coumaryl, coniferyl and synapyl alcohols that are formed by dehydrogenative radical polymerisation (Focher, 1992). Lignin content in wood according to Fengel and Wegener (1989) is between 20 to 40% but in flax fibre it ranges from 2-5% depending on retting and contamination levels from parenchyma cells present (Focher, 1992). Lignin levels are also believed to decrease during fibre processing and in bleached flax fibre the lignin level is very low (Focher, 1992).

2.3.2 Structure of the cell wall

Kohler and Kessler (1999) description of fibres as being 'complex natural composites' is indeed very accurate. Within the cell walls, microfibrils constructed from crystalline cellulose act as reinforcement bound within a matrix of lignin with amorphous cellulose, hemicellulose and pectins acting as a binder between the two phases. Plant fibre cells have two main layers: the primary wall and the secondary wall. The secondary wall can be sub-divided into three layers referred to as the 'outer layer (S_1), middle layer (S_2) and the inner layer (S_3)'. Figure 2.3 shows a simplified structure of a cell wall (wood), and it also highlights the difference of the angle of orientation of the microfibrils between each major layer.

The primary cell wall located next to the pectin rich middle lamella is thin and consists of randomly arranged microfibrils (Desch and Dinwoodie, 1996). Microfibrils in the S_1 layer run parallel to one another in two distinct spirals with an angle between 50° to 70° to the vertical axis (microfibillar angle), (Desch and Dinwoodie, 1996). The S_1 layer accounts for about 10% of the cell wall thickness. The thickest layer within the cell wall is the S_2 layer that takes up about 85% of cell volume (Desch and Dinwoodie, 1996). Microfibrils in the S_2 layer lie parallel to each other in a spiral formation. The microfibrillar angle in wood has been measured to be between 10° and 30° in the S_2 layer (Desch and Dinwoodie, 1996). The microfibrillar angle in the S_2 layer of flax fibres is almost parallel to fibre axis (Olsen and Plackett, 1999; Morvan *et al.,* 2003; Girault *et al.,* 1997). The longitudinal strength and stiffness of fibres correlate with the microfibrillar angle.

The smaller the angle of the orientation of microfibrils (close to the longitudinal axis of fibre) in the S_2 layer results in higher mechanical properties. An example from Bledzki *et al.,* (1996) is that the microfibrillar angle measured in flax was 10° whilst in sisal it was 20°. Mechanical properties from both sisal and flax sourced from Ivens *et al.,* (1997) shown in Table 1.3 on page 6, clearly show that the tensile strength and Young's modulus of flax is higher than sisal. This may be due to a whole host of other factors

such as cellulose content but undoubtedly the microfibrillar angle of the thickest layer within the cell wall plays a crucial role. The S_3 layer is similar to the S_1 layer but it only accounts for 1% of the thickness of the cell wall (Desch and Dinwoodie, 1996).

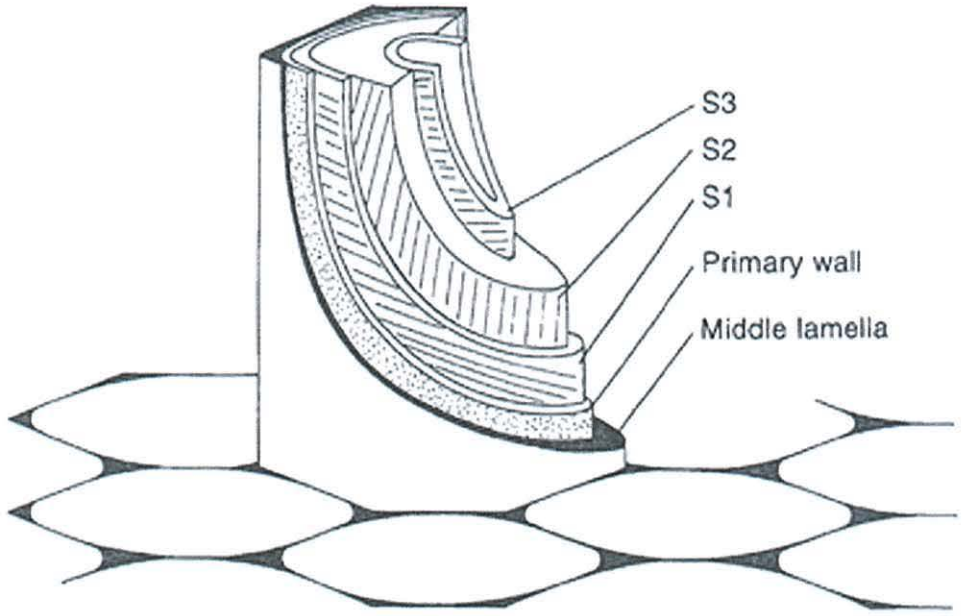


Figure 2.3 Structure of the cell wall, highlighting the differences in microfibillar angle between layers (*Source*: Brett and Waldron, 1996).

2.4 Properties of fibres that enable them to be reinforcement in composites

The properties of fibres used as reinforcement for composites are an important aspect in the design of materials. Properties of natural fibres may influence the manner in which the fibres are used as reinforcement and the mechanical properties of the composite. Glass fibres have features that enable them to be used as a good reinforcement in composites such as their high aspect ratios (length: width), uniformity, cylindrical shape and smooth surface. High aspect ratios allow fibres to be aligned for use in composites and give a greater surface area for adhesion. E-glass fibres have a density of

approximately 2500 kg m^{-3} a Young's modulus of 73 GPa and a tensile strength of 3.5 GPa (Anderson *et al.,* 1990). E-glass fibres also have a failure strain of approximately 2.6% (Hull and Clyne, 1996). Polyesters and epoxy resins are commonly used with E-glass fibres and their failure strain is approximately 2% and epoxy resins range from 1-6% (Hull and Clyne, 1996). E-glass fibres have similar failure strains to these resins, which is necessary if stress is to be transferred to the reinforcement (E-glass fibres). If this were not the case then matrix failure would occur prior to the reinforcement working effectively.

2.5 Harvesting, retting and processing of bast fibres

The harvesting and retting method used will influence the quality of the fibre gained. Processing fibre can also alter characteristics of the fibre and determine the end-use. The following sections summarise the main routes that fibres such as hemp and flax can follow from the field to an end product.

2.5.1 Harvesting

The valuable part of flax grown for fibre production lies within the stem where fibres run the entire length. To obtain the maximum yield, flax is not cut from the ground but is pulled (Sultana, 1992). This process can be done by hand or with a harvesting machine. Determining the time of harvest of hemp is important with respect to the quantity and quality (including fineness) of the yield gained (Bócsa and Karus, 1998). Flax fibre can be harvested when one third of the stem has turned yellow and the plant has reached maturity (Lianshu and Sharma, 1992).

2.5.2 Retting

Usually after harvesting the process of retting of the stem is required. Retting is the separation of fibre bundles from the rest of the plant by the degradation of substances surrounding the fibres by enzyme activity or thermochemical hydrolysis. This controlled rotting process involves bacteria and fungi releasing enzymes that convert the cells of parenchyma, epidermis and bark into a slimy material leaving the fibre bundles exposed (Van Sumere, 1992). Pectic enzymes alone can separate fibre cells (Chesson, 1978). A number of retting methods are described in the following sections.

2.5.2.1 Dew-retting

Dew-retting (also known as ground or field retting) is a process when the flax stems are laid on the ground and allowed to ret. It is a low-cost method but at the expense of not being a totally controllable method (Sultana, 1992). Swaths are bundles of stems that are collected after harvesting and it is in these swaths that dew-retting occurs. Stems that are 90 cm high require swaths 105-110 cm in width (Sultana, 1992). Retting does not progress homogeneously in the swaths so they require turning at least once (Sultana, 1992). Dew retting can be done straight after harvesting or flax stems can be collected, dried and dew retted when conditions are more favourable (Van Sumere, 1992). Fungi produce the enzymes responsible for the separation of fibres from within the stems. The enzymes can complete the separation in 3 to 8 weeks depending on the time of year. It is a great skill in determining if the retting process has reached its optimum but it is normally terminated when stems have a greenish-black colour over the entire length (Van Sumere, 1992). Dew-retting produces higher fibre yields than water retting but does result in lower quality flax fibres.

2.5.2.2 Stand-retting

Stand-retting is a pre-harvest retting system. An application of glyphosate is applied to standing growing flax causing the plants to rapidly die and desiccate within 1-2 weeks (Sharma *et al.,* 1992). Fungi start to grow; releasing microbial enzymes and the standing stems start to ret in a similar fashion as dew-retted flax stems. The amount of glyphosate used and the timing affects the fibre yield, which can be greater or less than dew-retted fibre. Difficulties in spraying crops and applying glyphosate to individual plants, loss of seed, timing, weather variability and possible fibre yield reductions do not make stand retting economically viable (Easson and Long, 1992).

2.5.2.3 Water-retting

Stems are collected and placed in heated tanks or pits between 30 to 40°C and retted for 3 to 7 days (Van Sumere, 1992; Cook, 1993). It is expensive but a better method for producing finer fibres than dew-retting and can be operated all year round. One tonne of flax stems are mixed with 10-11 tonnes of water at 20°C. It is left for 6-8 hours to leach inorganic salts, colouring matter, and soil from roots (Van Sumere, 1992). The water is then drained away and the tank or pit refilled with warm water (40°C) and the retting process begins. Bacteria are present during water retting and the blowing of air into the tanks accelerates the process (Van Sumere, 1992). Once retting is completed, the waste can be pumped away into a holding tank. The environment surrounding a retting tank/pit is subjected to strong and unpleasant smells from the organic acids produced.

2.5.2.4 Steam-retting

This process takes place in autoclaves and involves the retting of stems in cold-water and under steam pressure. After the initial treatment with cold-water, steam is pumped into

the autoclaves and the non-cellulosic substance bonding the fibres weaken due to thermochemical hydrolysis (decomposition of chemical compounds reacting with water), (Mukhin, 1992). Mukhin, (1992) reports that the fibre is light in colour, coarse, has a lower spinning limit and is used in the textile industry only as a blend with other fibres.

2.5.2.5 Enzyme-retting

Enzymes produced by fungi and bacteria which are present during conventional types of retting such as dew, stand and water-retting can be replaced by industrially obtained plant cell wall degrading enzymes, such as pectinases, hemicellulases and cellulases (Van Sumere, 1992). A liquid known as 'Flaxzyme' has been developed for the enzyme-retting of flax (Van Sumere, 1992). Enzyme-retting is advantageous over conventional methods, due to the added control over the process and the time saved during the retting. Van Sumere (1992) has tested different procedures and concentrations of Flaxzyme and found that retting at 40°C can be completed within a few hours and at ambient temperatures in about 3 days. The process of enzyme-retting is similar to water-retting. Stems are placed into tanks and water added with the correct concentration of enzymes. Enzyme solution can be recovered by passing retted stems through rubber rollers to squeeze out excess solution. (Van Sumere, 1992). It is believed by Van Sumere (1992) that enzyme retting will decrease the wastage of fibre that occurs during dew-retting, especially in regions where weather conditions are not always favourable and save energy when compared to water-retting.

2.5.3 Scutching of flax and hemp

After retting, the fibre bundles have to be separated from the woody parts and cortical parenchyma by an operation known as 'scutching' sometimes referred to as 'decortication'. Well-retted stems are easier to scutch than under-retted, which prove to

be difficult and the fibre bundles often have a greater amount of woody material attached (Sultana, 1992a). The strength of over retted fibre is low and scutching has to be carried out with care (Sultana, 1992a). Scutching involves two operations, the first is 'braking' and the second is 'beating'. Braking aims to crush woody components within the stem into small pieces named 'shives', which are approximately 1cm in length. Beating then aims to remove the shives by tangential scraping of the broken stems (Sultana, 1992a; Cook, 1993). Two types of fibre are gained from scutching, long fibre and tow fibre (short fibre). Various machines have been developed for scutching since World War II but with all the machines the stems have to be processed lengthwise to obtain long fibre. After scutching, the flax is in the form of very coarse fibre strands (many fibre bundles linked), often containing some shives (Ross, 1992).

2.5.4 Hackling

Hackling is required for scutched flax if it is to be used in the textile industry. Hackling splits the fibre bundles that make up the fibre strands into finer fibre bundles (Ross, 1992). Ross (1992) and Cook (1993) describe the three objectives of hackling:

1 Disentangle and straighten out fibres,
2 Separation of fibre bundles whilst maintaining length and,
3 Cleaning any shives that have remained from scutching.

Different hackling machines exist but they all work with the principle that fibre strands are hackled (combed) through pinned sheets (hackles) first by the 'root side' then they are turned and combed from their 'top side' (Ross, 1992). The hackles can have different sizes depending on the quality of fibre being processed. Long fibre bundles break down unavoidably into shorter sections which are often tangled, referred to as the tow fibre. The yield of line fibre (long straight fibre bundles) produced is often in the range of 55 to 65% with a throughput of 40 to 65 kg/hectare (Ross, 1992). Line fibre at the end of hackling machines is often transferred onto an automatic spread-board. The spread-board

overlaps individual fibre bundles converting them into a continuous length known as a 'sliver'. Flax sliver is then made up to known lengths and packaged. Flax sliver alone has some structural integrity but must be handled carefully to prevent it breaking into shorter lengths.

2.5.5 Non-woven felted mats

Air laid, needle punched, non-woven felted mats are created by making a fine web of chopped decorticated fibre and combing it between rollers that are equipped with fine teeth, a practice known as 'carding' (see Section 2.5.6). Many layers of the web are built up to form a mattress. The masses of fibres are then condensed with nip rollers and needle punched to secure the compressed mattress into a workable felted material. A more detailed description of the manufacture of non-woven reinforcement can be found in Wagner (1988).

2.5.6 Spinning

Before spinning, flax sliver is subjected to a process known as 'drafting', which draws out sliver by allowing fibres to slip past each other (Jarman, 1998). Sliver is drafted through pins that split the fibre bundles further. The fibre bundle length decreases, but the number and fineness of the fibres increases (Ross, 1992). Several flax slivers may be brought together to improve the regularity of the sliver during drafting in a process known as 'doubling' (Ross, 1992; Jarman, 1998). Rove is the term for flax sliver that has been given a loose twist. Rove sliver, usually, has not been doubled so the twist is necessary as it gives extra integrity to the structure that it will require during spinning (Ross, 1992). Twisted rove is then wound onto a bobbin. Wet spinning of flax is a process where fibre passes through a hot water trough so the residual pectins on the fibre are softened. They are then passed through metal fluted rollers that split the fibres along

their length by applying pressure. The fibres are then subjected to transverse rupture as the yarn is spun from shorter and finer fibres than the incoming twisted rove (Ross, 1992). It is thought that wet spun yarns are stiffer than dry spun yarns due to the residual pectins on the surface of fibres re-setting and adjoining adjacent fibres (Ross, 1992). To a degree, yarn quality is dependent on the sub-division of fibres during hackling, greater sub-division leading to higher spinning quality (Archibald, 1992). Hot air dryers are used at a temperature of 65°C to dry wet spun yarns; higher temperatures would alter strength and dye uptake (Ross, 1992.). Dry spinning of flax yarn is usually used when processing the tow fibre. Tow fibre must pass through a process known as 'carding' before it can be eventually dry-spun from sliver produced from carding. Ross (1992) describes four objectives of carding:

1 Opens lumps of fibre,
2 Partially straightens the fibres,
3 Removes dirt and tangled fibres,
4 Forms continuous lengths of sliver.

For both methods of spinning, yarns are then wound on automatic winding machines and any yarn faults are removed.

2.5.7 Weaving

Woven fabrics are produced from the weaving of 'warp yarns' (0°) and 'weft yarns' (90°) in a regular pattern or weave style. Interlocking of the yarns gives integrity to the structure. Flax/hemp weaving firstly involves the preparation of the warp yarns. Warping is achieved by putting packages of flax/hemp yarns on a creel (rack holding bobbins) that are wound in parallel on a warper's beam. Several of the yarns are then assembled in a sizing process. Sizings are adhesives and lubricants that are coated onto yarns to help decrease the amount of yarn damage caused by abrasion between other yarns and parts of machinery (Vangheluwe and Kiekens, 1992). Sizing is explained in

further detail in Section 2.5.9 on page 42. Yarns must be wound in parallel and an equal tension must be applied to all the yarns during both types of warping. Figure 2.4 shows a schematic drawing of a loom.

1. Warp beam
2. Warp yarns
3. Back rest
4. Shaft
5. Shaft
6. Reed
7. Filling or weft yarn
8. Filling bobbin
9. Fabric
10. Breast beam
11. Weft insertion
12. Cloth beam

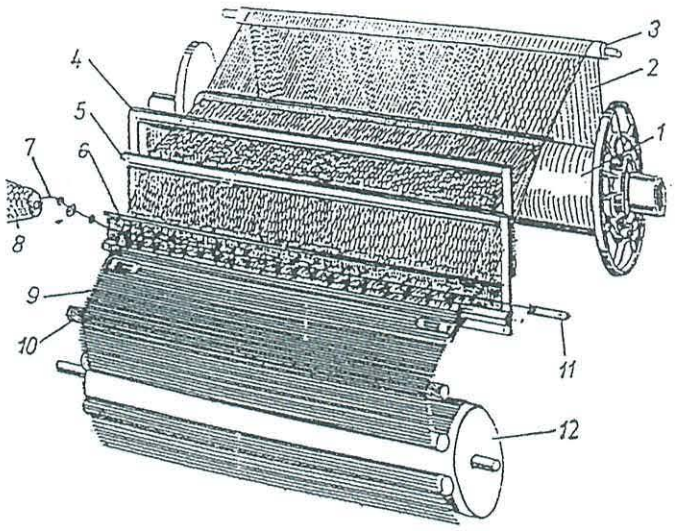


Figure 2.4 Schematic drawing of a loom (*Source*: Vangheluwe and Kiekens, 1992).

As the warp beam unwinds, warp yarns travel vertically over the 'back rest', which bends them to a horizontal travelling direction. On each warp end (end of warp yarn) there are fine metal plates known as 'droppers'. If a warp yarn were to fail then the droppers fall and the loom stops. A process known as 'shedding' creates a 'shed', which is an opening where a weft yarn can be inserted into the warp sheet. There are three methods available to create sheds but a useful way is the Jacquard mechanism. This technique allows each warp yarn to be pulled up or down separately which enables a vast number of weave patterns to be produced (Vangheluwe and Kiekens, 1992). As the warp yarn passes the section where the weft yarns are inserted, the fabric then winds onto the cloth beam.

2.5.7.1 Types of weave

Woven flax is mainly weaved into plain (tabby weave), twill and satin weave styles but many more weaving types can be obtained by interlacing these three patterns (Vangheluwe and Kiekens, 1992). According to Vangheluwe and Kiekens (1992) about 70% of woven flax is plain weaved, 20% is twill and satin and 10% are fancy weave types. Figure 2.5 schematically shows some weave types with warp yarns displayed in shades of dark grey and weft yarns displayed in white.

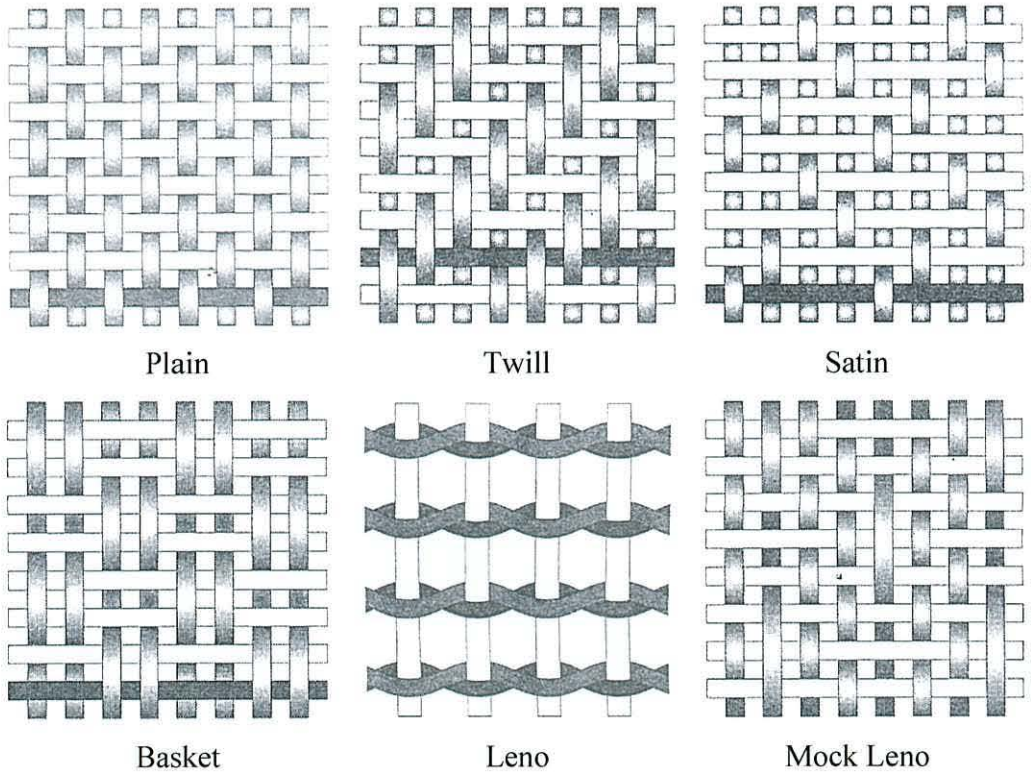


Figure 2.5 Schematic representation of six weave styles.

The warp yarns in Figure 2.5 are vertical (apart from 'Leno'); warp yarns are the yarns that are crimped over and under the weft yarns (horizontal yarns in Figure 2.5 apart from

'Leno' weave). Weft yarns are often straighter than warp yarns as they are inserted between the warp yarns during weaving.

Drape (the ability of a fabric to conform to a complex surface), surface smoothness and stability of a fabric are controlled primarily by the weave style. The weight, porosity and wet-out characteristics are determined by the thickness of yarn and the number of yarns per given area. Crimp is a term used to describe the waviness or distortion of a yarn due to the interlacing in the fabric.

Different weave styles have different amounts of crimp, for example satin weave has far fewer crimps than a plain weave. The size of yarns within linen can be described by the 'Tex system', which is commonly used throughout the industry. The Tex system is an expression of linear density (mass per unit length) of fibres, filaments, yarns and other linear textile material. The basic unit is a 'Tex', which is the mass in grams of one kilometre of the product. Tex is a recognized SI unit and it is often found in combinations like the following:

- Kilogram per kilometre (kTex),
- Milligram per kilometre (mTex).

Flax yarns produced from a range of spinning techniques can be formed into yarns of various linear densities often ranging from 11-400 Tex (Kernaghan and Kiekens, 1992). Coarse dry spun yarns range from 400 to 165 Tex whereas the fine dry spun yarns range from 165 – 65 Tex. Wet spinning of yarns allows finer yarns to be spun and they range between 120 to 20 Tex (Kernaghan and Kiekens, 1992). Rosiak and Przybyl (2003) believe that the amount of twist within yarns of various materials is one of the most important morphological yarn features that influence mechanical properties such as breaking strength. Within flax woven fabric it is often the case that the warp yarns are different to the weft yarns with respect to the size. Weave type, yarn linear density, twist, and number of yarns within a given distance in the woven fabric all influence the properties. Flax woven fabrics are sometimes referred to as 'grey'. Woven grey goods

are in the same condition as they leave the loom. Flax woven grey fabrics have not been bleached, dyed nor had any other finishing treatments given to them. However as mentioned in Section 2.5.8, bleaching can take place before weaving.

2.5.8 Bleaching

Bleaching can be described as a process that aims to remove colouration in the material. Kernaghan and Kiekens (1992) describe two methods that bleaching agents can be used to achieve the aim, one is *via* chemical action by oxidative or reductive means that will solubilise colouring materials and/or make them accessible for later processing, the other is modification of colouring matter *in situ*. Chemical modification may cause the colouring matter to lose its ability to absorb visible light, and prevent discoloration such as yellowing with age. Flax fibre colouration is from inorganic pigments, organic compounds and complexes incorporating metal cations (Kernaghan and Kiekens, 1992). Bleaching of flax can be a one or two-stage operation, it can have a pre treatment or just a single treatment at various stages throughout processing. Bleaching can be implemented when flax is in the form of sliver, rove, yarn or fabric. Flax bleached as a rove may have a secondary bleaching when it has been weaved into a fabric. Kernaghan and Kiekens (1992) describe in detail many of the treatments that are available for bleaching and dying of flax. Kernaghan and Kiekens (1992) suggest there are five treatments that may be used individually or as combinations that can bleach flax fabric. These include:

1. Alkaline boil, or scour, of variable severity,
2. Acidic chlorination,
3. Sodium hypochlorite solution,
4. Hydrogen peroxide bleach,
5. Sodium chlorite bleach.

Bleaching can have an effect on the properties of flax. Focher *et al.,* (1992) shows results gained with linen yarns that were bleached with hydrogen peroxide (H_2O_2) and treated

with increasing concentrations of sodium hydroxide (NaOH) (0.5-2%) simulating conditions of industrial scouring. It was found that the yarns breaking load did not decrease compared with a control but weight loss increased with NaOH concentration. A gradual decrease in the degree of polymerisation of cellulose was also noted. Kernaghan and Kiekens (1992) state that modern bleaching agents are capable of a bleaching action with hardly any depolymerisation of cellulose. Yarn scouring and bleaching actually improves weaving performance of both dry and wet spun yarns (Kernaghan and Kiekens, 1992). Kernaghan and Kiekens (1992) exposed top quality and under-retted flax roves to a short two-stage bleaching treatment of chlorination/peroxide and caustic boil/peroxide. It was found that both roves were capable of making yarns with high tensile strength. The bleaching of flax undoubtedly is advantageous for many processing operations and to the physical appearance of flax fabric, especially when the flax used has come from different processing backgrounds. The tensile strength of bleached yarns seems to change very little, but the chemical constituents of the fibre are likely to change (Focher *et al.,* 1992).

2.5.9 Sizing

As previously mentioned, yarns are sometimes coated with a size before weaving. The flax industry has utilised sizing solutions developed for the sizing of cotton yarns. Natural starches and modified starches are used, as well as synthetic polymer based sizing agents (Vangheluwe and Kiekens, 1992). Synthetic polymer based sizing agents include sodium carboxymethyl cellulose (CMC), polyvinyl alcohol (PVA), polyacrylic derivatives, vinyl co-polymers, acrylates, and polyesters (Kernaghan and Kiekens, 1992). Drying of yarns after sizing is an important part of the sizing operation. Vangheluwe and Kiekens (1992) believe that sized warp yarns must be kept under an equal tension over the width of the warp during drying, and the tension applied should be very low to prevent irreversible elongation occurring that would reduce strain at failure of dried sized warp yarns (approximately 2% normally). Prior to bleaching it may be necessary to remove size from yarns as the presence and composition of sizing agents affect some

bleaching processes. The Northern Ireland flax industry commonly uses synthetic blends from acrylates, vinyl co-polymers and PVA that can be removed by detergent scouring (Kernaghan and Kiekens, 1992). Starch based sizes can be difficult to remove completely with detergent scouring according to Kernaghan and Kiekens (1992) but is possible by using enzymatic means.

2.6 Resins

As previously mentioned, one phase of polymer matrix composites is either a thermoplastic or thermosetting resin system. In the following sections, details of thermoplastic and thermosetting resins will be described with particular attention paid to the structure and properties of epoxy and polyester resins.

2.6.1 Thermoplastic polymers

Thermoplastic resins are solid at room temperature but at elevated temperatures readily flow under an applied stress. Thermoplastics can be repeatedly heated and re-moulded and cooled but this may be detrimental to the properties due to a reduction in molecular weight of the polymer (Matthews and Rawlings, 1993). Unlike thermosetting resins, thermoplastic resins are not cross-linked. Thermoplastics consist of high-molecular-weight polymer chains. The strength and stiffness of the resin is derived from the chemical properties of the monomer units and the entanglement of the polymer chains. Bonding between chains is achieved by weak Van der Waals forces, which are easily broken by thermal activation or applied stress. Thermoplastic polymers that have a semi-crystalline structure are ductile and can undergo large plastic deformations before final fracture. Polyether-ether-ketone (PEEK), polypropylene and nylon are all examples of thermoplastic polymers that have a higher resistance to crack propagation than thermosetting resins because of their structure (Hyer, 1998; Hull and Clyne, 1996).

Thermoplastic polymers that have an amorphous structure such as polystyrene are brittle and therefore exhibit a lower toughness than semi-crystalline thermoplastics.

2.6.2 Thermosetting resins

Thermosetting polymers or thermosets are liquid resins that undergo an irreversible chemical change when they are heated, called 'curing'. Application of heat and/or, pressure and/or, a catalyst initiates chemical cross-linking (curing). The three-dimensional network structure formed sets the final shape. Cross-link density (number of cross-links per unit volume) and length affects properties such as rigidity, strength, solvent resistance and thermal stability (Arnold *et al.,* 1992. & Hull & Clyne, 1996; Anderson *et al.,* 1990). High density cross-linking leads to restricted molecular movement that causes brittleness, low strains to failure and poor impact and fracture toughness. Examples of thermosetting polymers are epoxy, cyanates, unsaturated polyesters, polyimides, phenolic resins and urea-formaldehyde.

A comparison between thermoplastic and thermosetting resins environmental stability and dimensional properties is presented in Table 2.4, whilst Table 2.5 shows typical mechanical properties from a selection of both types of resins.

2.6.2.1 Epoxy resin

Epoxy resins are mainly used for advanced composite materials, such as structural aerospace composites. Expoxies offer excellent mechanical properties, retention of mechanical properties in hot and moist environments, and good chemical resistance. They exhibit good adhesion to a wide range of fibres but they are more expensive and viscous than polyester resins making impregnation of woven fabrics more difficult (Matthews and Rawlings, 1993). One advantage of epoxy resins is that curing can

involve two or more stages allowing more time before being moulded into the final shape.

Table 2.4 Comparison between environmental stability and dimensional properties of thermosets and thermoplastic polymers (*Source*: Hull and Clyne, 1996).

Property	*Thermosets*		*Thermoplastics*		
	Epoxy resin	*Polyester resin*	*Nylon 6.6*	*Polypropylene*	*PEEK*
Melting temperature (°C)	-	-	265	164	334
Distortion temperature (°C)	50-200	50-110	120-150	80-120	150-200
Shrinkage on curing (%)	1-2	4-8	-	-	-
Water absorption (24h @ 20°C) (%)	0.1-0.4	0.1-0.3	1.3	0.03	0.1

Table 2.5 Mechanical properties of different types of matrix (*Source*: Hull and Clyne, 1996).

Matrix	*Density ($Mg\ m^{-3}$)*	*Young's modulus (GPa)*	*Tensile strength (GPa)*	*Failure strain (%)*
Thermosets				
Epoxy resins	1.1-1.4	3.6	0.035-0.1	1-6
Polyesters	1.2-1.5	2.0-4.5	0.04-0.09	2
Thermoplastics				
Nylon 6.6	1.14	1.4-2.8	0.06-0.07	40-80
Polypropylene	0.90	1.0-1.4	0.02-0.04	300
PEEK	1.26-1.32	3.6	0.17	50

The most common epoxy resin (90% of world production) is based on the reaction of epichlorohydrin and bisphenol 'A' as shown in Figure 2.6 (Hyer, 1998; Arnold *et al.*, 1992; Elias, 1993). Varying the proportions of epichlorohydrin and bisphenol 'A' can alter properties such as viscosity and melting point. The molecular weight of the resin is increased when the proportion of epichlorohydrin is reduced (Norwood, 1994).

Cross-linking is accomplished by adding a curing agent (catalysts, hardeners or activators) that reacts with the epoxide (epoxy) or hydroxyl groups. The curing agent may become incorporated into the network structure hence resultant properties of resin are dependent on curing agent used. Generally epoxies are stiffer and stronger, but more brittle than polyesters (Matthews and Rawlings, 1993). However recent epoxies are tougher than unsaturated polyesters due to advanced epoxy formulations (Hull & Clyne, 1996).

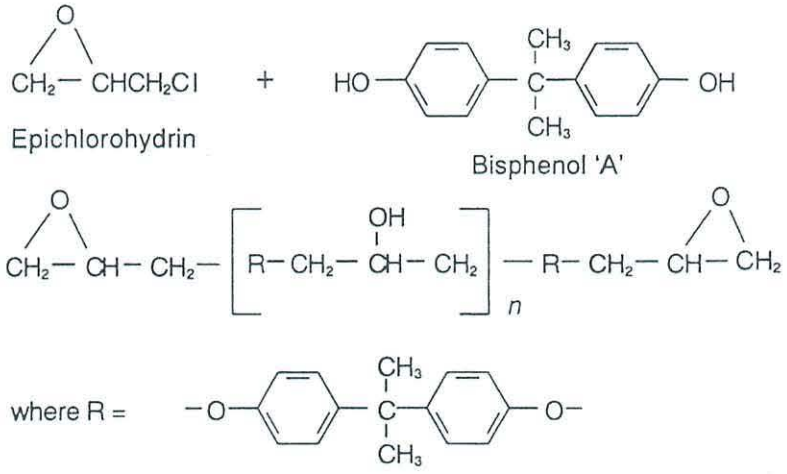


Figure 2.6 Schematic representation of an epoxide based upon epichlorohydrin and bisphenol 'A' (*Source*: Norwood, 1994).

2.6.2.2 Polyester resins

Unsaturated polyester resin is by far the most widely used thermosetting resin in the automotive, construction and in general composite applications market (Hyer, 1998; Arnold *et al.,* 1992; Norwood, 1994). Poor impact and hot/wet mechanical properties, limited shelf life and high shrinkages during curing prevent its use for high-performance applications (Arnold *et al.,* 1992). Shrinkage during curing can be between 4 to 8% (Matthews and Rawlings, 1993). However polyesters are inexpensive and have low viscosities, which are beneficial in fabrication processes and lead to easier impregnation into reinforcement fibres. Unsaturated polyester resins consist of polymer chains dissolved in an organic solvent. Styrene is an organic solvent that is often used with polyesters because it is inexpensive, compatible with polyester, reduces viscosity and acts as a cross linking agent. The addition of catalysts and accelerators causes polyesters to form a solid three-dimensional structure. Catalysts such as organic peroxides cause free radical co-polymerisation to occur throughout the polyester either at ambient or elevated temperatures. Polyesters are formed by reacting saturated dialcohols (glycols) with a mixture of unsaturated and saturated dibasic organic acids (or anhydrides). By elimination of water between the acids and the glycols, ester linkages are formed that produce long chain molecules, consisting of alternating units of acid and glycol. Figure 2.7 shows a polyester polymer chain.

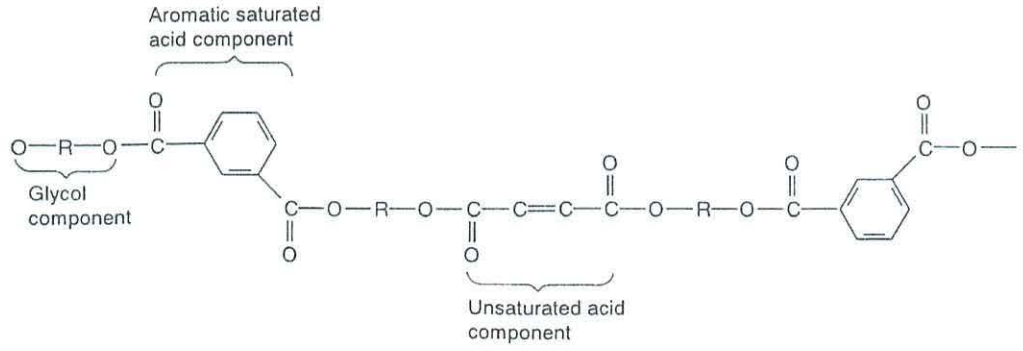


Figure 2.7 Polyester polymer chain (*Source*: Norwood, 1994).

Properties and structure of cured resin depends on the frequency of carbon-carbon double bonds situated on the chain.

2.7 Composite materials

The following sections aim to briefly review composite materials by encompassing aspects of the reinforcement, interface, fibre architecture, properties and deformation, toughness and failure modes.

2.7.1 Reinforcement processes

2.7.1.1 Load sharing

Composed of at least two phases, composites must be able to share loads between the matrix and reinforcement. From point to point, stress along fibres can vary substantially, especially when reinforcement is in the form of short fibres (Hull and Clyne, 1996). Proportions of an external load applied that each phase experiences (assuming unidirectional composite reinforced with continuous fibres with equal strain occurring for both fibres and matrix) can be estimated by volume-averaging the loads related with them as seen in Equation 2.1.

Equation 2.1 $$\sigma_c = V_f \overline{\sigma}_f + (1 - V_f)\overline{\sigma}_m$$

Where: σ_c Applied stress of the composite.

V_f The fibre volume fraction. This is the volume of fibre present in the composite as a fraction of the

total volume of the composite. Often expressed as a percentage.

$\overline{\sigma}_m$ Volume-averaged matrix stress.

$\overline{\sigma}_f$ Volume averaged fibre stress.

Whilst composites display elastic behaviour (stress is proportional to strain) a proportion of the load is carried by the reinforcement and the remainder by the matrix. Irrespective of the amount of applied load, the proportion that is borne by each phase depends on the following factors (Hull and Clyne, 1996). The proportions depend upon the volume fraction, shape and orientation of the reinforcement as well as the elastic properties of both phases. A higher proportion of a load applied to a composite is likely to be carried by the reinforcement than the matrix, if it is working efficiently and stress is transferred. This is often the aim of many composites, as the reinforcement is usually stronger and stiffer than the matrix.

2.7.1.2 Elastic stress transfer

Stress transfer is an essential process that allows composites to transfer stresses from the matrix to the reinforcement. Composite reinforcement is usually the load-bearing phase and inadequate transfer of stress would result in matrix failure. Stress is transferred across the interface between matrix and fibre *via* shear stresses (τ_i). Figure 2.8 is a schematic representation of the shear lag model proposed by Cox (1952).

Figure 2.8 shows two composite systems, one being a fibre embedded in a matrix that is unstressed (a) and the second is a fibre embedded in a matrix that is subjected to a tensile stress parallel to the fibre (b). System (b) is behaving elastically (no slippage is occurring between the fibre and matrix). It can be seen in Figure 2.8 that as the composite is strained, the matrix deforms around the fibre but to a higher degree around the fibre ends. Deformation of matrix leads to shear stress (τ_i) at the interface. Due to the distortion of

the matrix being greater at the fibre ends, the shear stresses are higher at these regions than the centre of the fibre. The axial stresses in the fibre (σ_f) will be zero at the fibre ends but will increase as the distance from the fibre ends increases, due to the shear stresses in the interface decreasing as the distance from the fibre ends increases. Hull and Clyne (1996) have shown that during elastic stress transfer, the axial stress distribution along a fibre (σ_f) can be given by Equation 2.2.

Equation 2.2 $$\sigma_f = E_f \varepsilon_1 [1 - \cosh(nx/r)\operatorname{sech}(ns)]$$

Where:

E_f	The fibre Young's modulus.
ε_1	The applied composite strain.
s	The fibre aspect ratio, defined as *L/r* (where '*L*' is the fibre half length and '*r*' is the fibre radius).
n	Is a dimensionless constant.
x	The axial distance from the fibre mid-point.

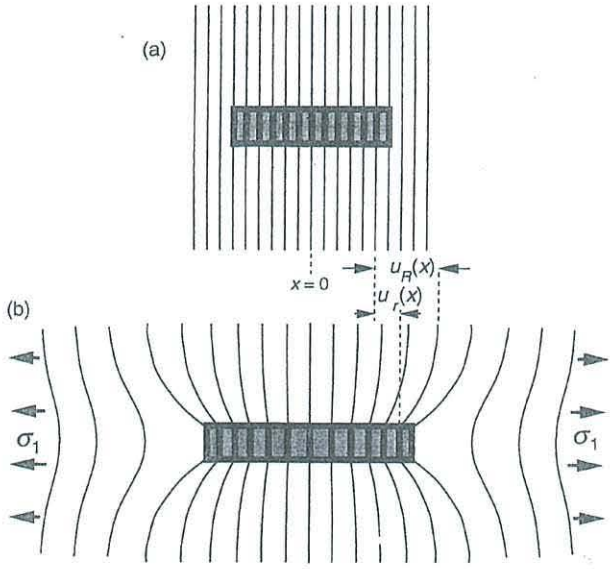


Figure 2.8 Schematic representation of a 'Cox-type' shear-lag model (*Source*: Hull and Clyne, 1996).

The value of n can be obtained by using Equation 2.3. Hull and Clyne (1996) state that the value of n, does not vary widely, it is typically about 0.1 for polymer matrix composites and 0.4 for metal matrix composites.

Equation 2.3

$$n = \left[\frac{2E_m}{E_f(1-v_m)\ln(1/V_f)} \right]^{\frac{1}{2}}$$

Where: E_m The matrix Young's modulus.

v_m The matrix Poisson's ratio.

Hull and Clyne (1996) also describe Equation 2.4 that shows the variation of interfacial shear stress along a fibre (τ_i).

Equation 2.4

$$\tau_i = \frac{n\varepsilon_1}{2} E_f \sinh(\frac{nx}{r})\text{sech}(ns)$$

Both Equation 2.2 and Equation 2.4 allow for predictions about stress distributions along the length of the fibre by taking into account the aspect ratio, the applied strain and the elastic properties of the matrix and reinforcement. Both equations are dependent on the fibre/matrix modulus ratio, the applied strain and the fibre volume fraction.

An example of predicted σ_f and τ_i over a range of aspect ratios (s = 5, 10, 25, 50 and 100) is presented in Figure 2.9 and 2.10, respectively. The example is based on a hypothetical polymer matrix composite, with a fibre Young's modulus of 76 GPa (approximately E-glass) and a matrix Young's modulus of 3.5 GPa (approximately polyester resin). The hypothetical composite has a fibre volume fraction of 50% and an applied strain of 0.1%. The value of n, the dimensionless constant was calculated and found to be 0.31, assuming the Poisson's ratio of the matrix is 0.38.

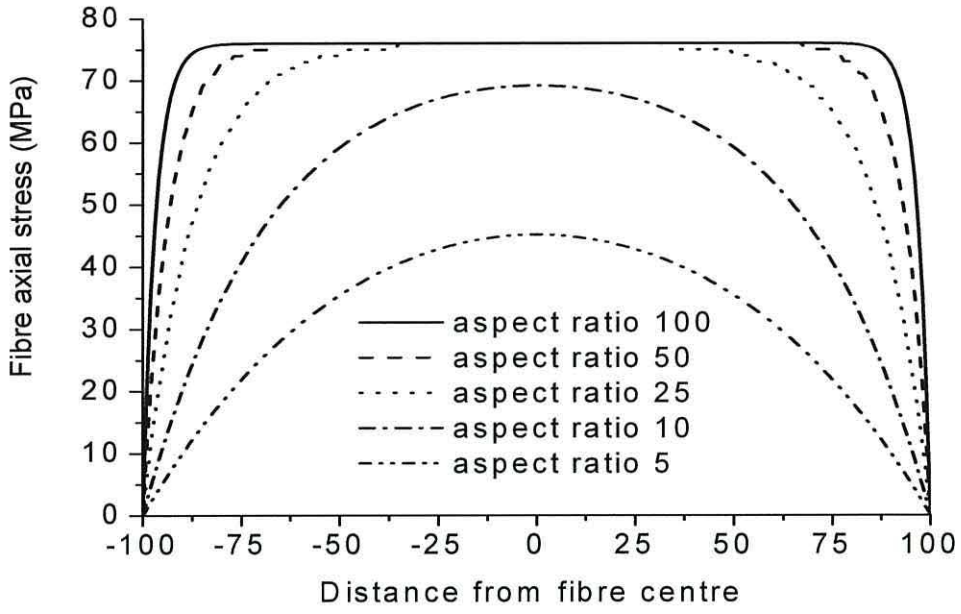


Figure 2.9 Theoretical shear-lag predictions of the variation of axial fibre stress along fibres with varying aspect ratios and an applied strain of 0.1%.

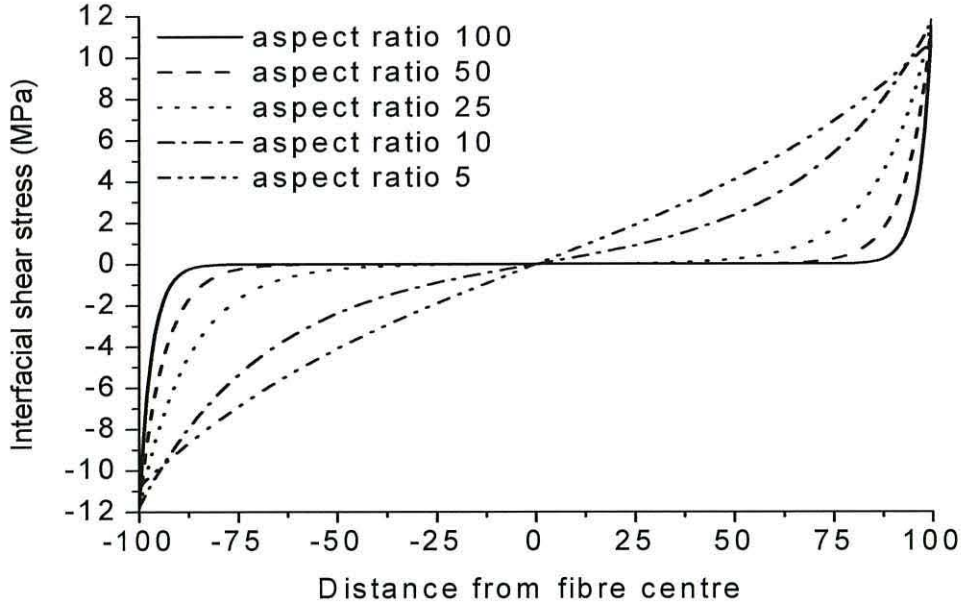


Figure 2.10 Theoretical shear-lag predictions of the variation of interfacial shear stress along fibres with varying aspect ratios and an applied strain of 0.1%.

The n value calculated for the hypothetical composite is near 0.4, a value that Hull and Clyne (1996) believe to be appropriate for metal matrix composites. The n value of 0.1, stated by Hull and Clyne (1996) for polymer matrix composites, is thought to be for lower fibre volume fractions (~10%), which are generally inappropriate for many composites.

2.7.1.3 Stress transfer aspect ratio

Figure 2.9 shows that the axial fibre stress is zero at the fibre ends and increases to a maximum at the fibre mid-point. The maximum axial fibre stress is determined when the fibre strain is equal to the matrix strain. In the example presented above, it is clear that fibres with aspect ratios of 100, 50 and 25 reach the maximum axial fibre stress at a relatively short distance from the fibre end, whilst a fibre with an aspect ratio of 10 just falls short of reaching the maximum axial fibre stress possible. The interfacial shear stress for the fibre that has an aspect ratio of 100 will be zero, as Figure 2.10 shows, when the axial fibre stress is at a maximum. This leads to the notion of a critical value for the stress transfer length. The critical value for the stress transfer length is the value of s where the maximum axial fibre stress is reached, for the applied strain on the composite. The reinforcing efficiency of fibre decreases as the fibre length is reduced, since this increases the proportion of the total fibre length that is not fully loaded. In the example above, the fibre with an aspect ratio of 5 does not even reach 60% of the theoretical maximum axial stress and thus is not providing efficient reinforcement. Composites comprised of continuous aligned fibres are in an equal strain condition with respect to stress (matrix and reinforcement) when a load is applied parallel to the fibres, providing there is no interfacial sliding.

2.7.1.4 Inelastic processes

The above models have been based on composite systems that behave elastically and in that situation the bonding between matrix and fibre are assumed to be perfect, and slippage of the reinforcement will not occur when the composite is strained. During the straining of a composite there are several factors that may cause the onset of inelastic behaviour. Hull and Clyne (1996) describe that plastic deformation of the matrix, fibre/matrix debonding (possibly leading to frictional sliding at the interface), formation of cavities/cracks in the matrix and the fracture of fibres cause the onset of inelastic behaviour. Plant fibres exhibit linear elastic, viscoelastic and plastic behaviour. Due to the interfacial shear stresses being higher at the fibre ends it can be assumed that matrix cracking and other failures will occur preferentially at these regions. Any of these actions occurring will change the stress distribution throughout the composite thus causing the linear stress strain relationship to end.

Inelastic behaviour caused by matrix plasticity or interfacial sliding is expected to occur when a critical value of interfacial shear stress is exceeded. The critical interfacial shear stress (τ_i.) can be obtained by setting x equal to half the fibre length (L) in Equation 2.4. The example displayed in Section 2.7.1.2 of a hypothetical composite with various fibre aspect ratios had a τ_i. of 11.79 MPa for a fibre with an aspect ratio of 100 and a τ_i. of 10.77 MPa for a fibre with an aspect ratio of 5. Predictions of the composite strain at which the onset of inelastic behaviour will occur (ε_1.) are possible by using Equation 2.5 (Hull and Clyne, 1996).

Equation 2.5

$$\varepsilon_{1}. = \frac{2\tau_i. \coth(ns)}{nE_f}$$

Using Equation 2.5 with the τ_i. value obtained from Equation 2.4 the composite strain at which the onset of inelastic behaviour starts is 0.99% for reinforcement with an aspect

ratio of 100. Inelastic behaviour starts at a strain of 0.10% for a composite comprised of fibres with an aspect ratio of 5.

Equation 2.6 calculates the composite stress at which the onset of matrix plasticity or interfacial sliding is expected to occur.

Equation 2.6

$$\sigma_{1^*} = \frac{2\tau_{i^*}}{nE_f}\left[V_f E_f + (1-V_f)E_m)\coth(ns) - \frac{V_f E_f}{ns}\right]$$

Where: σ_{1^*} The composite stress at the onset of inelastic behaviour.

Referring back to the example composites from Section 2.7.1.2 on page 49, it can be calculated that the predicted composite stress at which inelastic behaviour occurs is 38.52 MPa for the composite containing reinforcement with an aspect ratio of 100 and 14.09 MPa for the composites consisting of reinforcement with an aspect ratio of 5.

The stress value predicted by Equation 2.6 is the value where matrix plasticity or interfacial sliding is expected to occur in small localized regions of the composite. This stress value is considered to be the point where the stress-strain curve will deviate from linearity (Hull and Clyne, 1996).

As composites are strained and the stress-strain behaviour changes to an inelastic response, there is a possibility that fibre fracture may occur within the composite. Equation 2.2 on page 50 describes the stress distribution along the fibres, whereas Equation 2.7 gives the peak stress within the fibre at the onset of interfacial sliding or matrix yielding. Equation 2.7 is a direct method for calculating the peak stress that the fibre will endure; assuming that the maximum stress applied to the fibre is limited by the critical interfacial shear stress.

Equation 2.7

$$\sigma_{f0} = \frac{2\tau_i}{n}[\coth(ns) - \operatorname{cosech}(ns)]$$

The peak stress in the fibre at the onset of inelastic behaviour, obtained from either Equation 2.2 or Equation 2.7, is extremely useful in determining if fibre fracture is likely to occur before fibre/matrix debonding or plastic deformation of the matrix. In general, a fibre reinforcement is selected so that it can withstand the maximum stress transferred as the applied load of the composite increases and other microstructural failures occur before fibre fracture.

If plastic deformation of the matrix has not occurred, but failure events such as fibre/matrix debonding have taken place, the stress is transferred at the interface *via* friction. Interfacial shear stresses and the axial stress distribution may be similar to that shown in Figure 2.11. The model presented assumes that the frictional interfacial shear stress (τ_{if}) is zero at the centre and constant near the ends of the fibre.

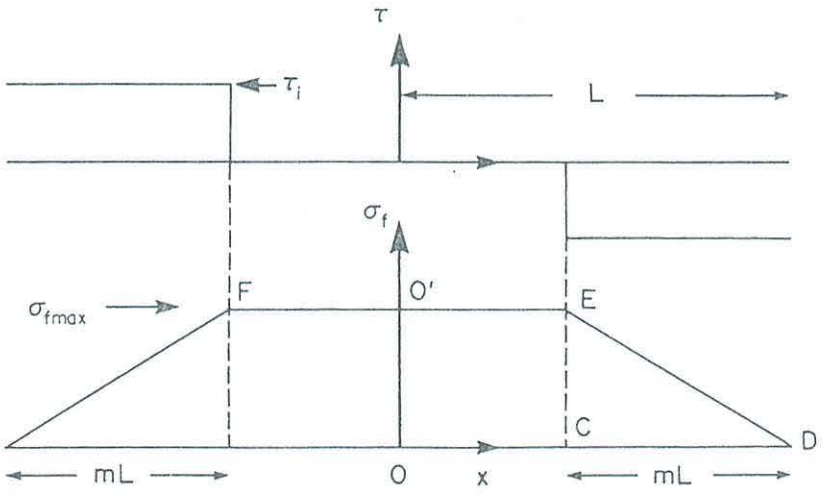


Figure 2.11 Distribution of axial fibre stress and frictional shear stress along a single fibre (*Source*: Piggott, 1980).

It is plausible that both inelastic and elastic stress transfer processes occur simultaneously. High interfacial shear stress at the fibre ends may cause fibre/matrix debonding in these areas, but the middle region of the fibre may still be firmly bonded to the matrix. Increasing the strain on a composite will cause interfacial decoupling to spread along the length of the fibre, thus increasing the tensile stress in the fibre as the interfacial shear stresses increase. Providing the maximum interfacial shear stress is constant along the length of the fibre, fracture of fibre becomes possible. A critical aspect ratio (S_c) can thus be identified. The critical aspect ratio can be viewed as the point where the central axial stress in the fibre equals the ultimate tensile strength of the fibre (σ_{fu}), thus allowing fibre fracture to occur. Hull and Clyne (1996) and Piggott (1980) have shown that S_c can be obtained by Equation 2.8, providing stress transfer by friction is considered.

Equation 2.8

$$S_c = \frac{\sigma_{fu}}{2\tau_i}$$

So far, only composite systems containing aligned fibres have been considered with a tensile stress applied along the fibre axis. This is obviously not the case in many circumstances where composites will experience some degree of off-axis loading. Composites consisting of aligned fibres may encounter stress-applied normal to the fibre axis and in this case they are weak (composite transverse strength is low). The transverse strength can be less than the strength of the matrix and fibres subsequently have a low reinforcing efficiency. The presence of fibres may have a detrimental effect on the transverse strength of the matrix (Anderson *et al.*, 1990).

2.7.2 Interface

A good adhesion between the fibre and matrix is extremely important, not only for the purpose of elastic stress transfer (as discussed in Section 2.7.1.2 on page 49) but on the

composite properties when subjected to loads in directions transverse to the fibre, aligned to the fibre and on the overall shear properties of the composite. The bonding between matrix and fibre also is a factor controlling the rate of degradation of a composite material exposed to adverse environments (Anderson *et al.,* 1990). An example would be a glass fibre polymer matrix composite exposed to water and the effect of osmosis on its structure and properties or untreated medium density fibreboard (MDF) used in an outside application. Although a good bond between the two phases of a composite is necessary to ensure the material has the required properties, it may also be required that the uncoupling of phases occurs in certain conditions. Gordon (1976) describes, the interface acting as a crack stopper by debonding ahead of an oncoming crack, thus diverting the crack as it reaches that part of the interface. This does create two extra cracks at right angles to the original one, but these do not have the tendency to propagate. Due to the ability of the interface acting as a crack stopper, the toughness of a composite can be improved. A strong interface with strong bonding would allow a crack to propagate though the interface into the fibre and cause brittle failure of the reinforcement.

Effectiveness of adhesion depends on the ability of the liquid matrix to wet the surface of the substrate, to which it is to bond. The behaviour of a liquid droplet on a material's surface is controlled by the strength of the interaction between the liquid and solid phases. Achieving good adhesion is possible when there is a strong interaction between the phases, due to them being chemically compatible. Surface energy determines how rapidly the resin will the substrates surface (viscosity of resin has a minor effect). It is possible to determine how well a liquid drop can wet a surface by measuring the contact angle (θ) produced. Compatibility (contact angle) between the resin and substrate determines how effectively the resin wets the surface. The contact angle formed is dependent on the strength of the interaction between the molecules in the liquid (cohesion), the strength of the interaction between the molecules of the solid (cohesion) and the strength of the interaction between the molecules of the liquid and the solid (adhesion). Figure 2.12 shows a contact angle, liquids that produce contact angles greater than 90° are referred to as non-wetting liquids. Accurate measurement of a contact angle is difficult when the solid material is rough or porous. The Young equation obtained by

considering the thermodynamics of the wetting process can be seen in Equation 2.9 (Pizzi and Mittal, 1994).

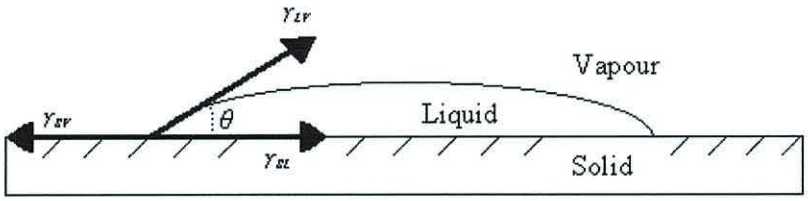


Figure 2.12 Schematic representation of a resin droplet resting on a solid surface and the subsequent contact angle (θ) produced.

Equation 2.9 $$\gamma_{SV} = \gamma_{SL} + \gamma_{LV} \cos\theta$$

Where:
- γ_{SV} The surface energy of the solid-vapour interface.
- γ_{SL} The surface energy of the solid-liquid interface.
- γ_{LV} The surface energy of the liquid-vapour interface.

When $\theta = 0$ in Equation 2.9 then complete wetting has occurred, hence the solid-vapour interface surface energy is equal or greater than the sum of the liquid-vapour surface energy and the solid-liquid interface surface energy ($\gamma_{SV} \geq \gamma_{SL} + \gamma_{LV}$). Wetting can occur easily when the surface energy of the solid-vapour interface is far greater than the liquid-vapour interface (Hull and Clyne, 1996). Wetting of a polar substrate by water may be inhibited if it is not free of release agents, machine oil, grease, *etc.* (Kinloch, 1987).

The work of adhesion (W_a) is shown in Equation 2.10, this is often termed the Dupré equation.

Equation 2.10 $W_a = \gamma_{SV} + \gamma_{LV} - \gamma_{SL}$

Where: W_a Work of adhesion

As seen from the above, the compatibility of molecular forces between the two phases being bonded is essential, in terms of creating an intimate molecular contact between adhesive and substrate. Molecular forces are the main mechanism of adhesion and this is referred to as the 'adsorption theory' of adhesion (Kinloch, 1987). There are three other mechanisms of adhesion:

1. mechanical interlocking,
2. diffusion theory,
3. electronic theory.

Out of all the above theories, the adsorption theory is the most relevant in adhesion science (Kinloch, 1987). The theory basically states that providing there is an intimate molecular contact at the interface, adhesion will occur because of the inter-atomic and inter-molecular forces, which are established between the atoms and molecules in the surface of the liquid matrix and substrate. Table 2.6 shows the bond types and associated bond energies that are concerned with the adsorption theory. The inter-molecular forces mostly active on the surfaces are Van der Waals forces but other secondary forces such as hydrogen bonds may also be present. Chemical bonds can also form across the interface and are referred to as 'primary bonds'. Donor –acceptor bonds can also form across the interface.

Mechanical interlocking (keying) is when the adhesive penetrates into the irregularities of the substrate surface. However it is difficult to measure if true mechanical interlocking has occurred, or if other factors such as the creation of an improved interfacial contact by giving a greater area for other adhesion mechanisms to work. Hull and Clyne (1996) believe there may be a contribution to the strength of the interface from the surface roughness of the fibres, providing good wetting has occurred.

Table 2.6 Bond types and bond energies associated with the adsorption theory of adhesion (*Source*: Kinloch, 1987)

Type	*Bond energy (kJ mol^{-1})*
Primary bonds	
Ionic	600-1100
Covalent	60-700
Metallic	110-350
Donor-acceptor bonds	
Bronstead acid-base interactions	Up to 1000
Lewis acid-base interactions	Up to 80
Secondary bonds	
Hydrogen bonds	Up to 40
Van der Waals bonds	0.08-40

2.7.3 Fibre microstructure

2.7.3.1 Volume fraction

As seen from Equation 2.1 onwards, many calculations on composite materials are based on volume fractions of the constituents. Matthews and Rawlings (1994) state that the most important factor affecting composite properties are the proportions of the constituents within a composite, expressed as a volume fraction (V_f). The fibre volume fraction of a composite can be obtained using Equation 2.11. The volume fraction of the matrix (V_m) can be expressed as $1-V_f$ (assuming no voids).

Equation 2.11

$$V_f = \frac{M_f}{V_c \rho_f}$$

Where: M_f The mass of the fibre.

V_c The volume of the composite.

ρ_f The bulk density of the fibre.

Equation 2.11 obviously requires knowledge of the bulk density of the fibre. For synthetic materials, the bulk density of reinforcement can be easily obtained and they will also exhibit low variations. Many synthetic reinforcements are also solid. Plant fibres being irregular, may have large variations of bulk density due to the same factors referred to in Section 2.3, they also contain inherent voids (lumen).

The fibre volume fraction can also be obtained from Equation 2.12, which does not require knowledge of the bulk density of the fibre (Roe and Ansell, 1985). The expression takes account of porosity associated with the fibre and allows for a value of bulk density of the fibre to be derived. The volume fraction can be obtained by knowing the mass of the constituents and density of the cast resin.

Equation 2.12

$$V_f = \frac{V_c - \frac{(M_c - M_f)}{\rho_r}}{V_c}$$

Where: M_c The mass of the composite.

ρ_r The density of the cured matrix.

Roe and Ansell (1985) state the value obtained using Equation 2.12 'is not a volume fraction in the strictest sense' but is a most practiced value in evaluating practical composites as the figures that are required for Equation 2.12 can be easily measured and calculated.

Hughes (2000) manufactured several composites comprised of jute and hemp bound with a polyester matrix at various fibre volume fractions. Using both Equation 2.11 and Equation 2.12 to evaluate the fibre volume fractions associated with the composites. For Equation 2.11 he used three density values, one was the density of the cell wall (assumed to be 1500 kg m^{-3}), the other two were bulk densities for each fibre type used, taken from Ivens *et al.*, (1997). The bulk density for hemp was 1480 kg m^{-3} and for jute 1450 kg m^{-3}. Hughes (2000) found that Equation 2.11 used with the density of the cell wall produced the lowest estimate of fibre volume fraction. Equation 2.11, when used with the values provided from the literature and Equation 2.12 both assume a certain degree of fibre porosity, thus higher estimates for fibre volume fraction were obtained. However Hughes (2000) states that Equation 2.11 does not allow for the fact that during manufacture, resin enters the lumen whereas Equation 2.12 does take this into account. Hughes (2000) concluded that the differences between the fibre volume fractions obtained from using both equations were small, usually less than 2% for individual composites. This low difference is within the range of experimental error and it was concluded that void space is minimal.

As previously mentioned in this section, the proportions of constituents within the composite are important when considering the material's properties. By using the volume weighted mean of properties from each phase within a composite a relationship known as the 'Rule of mixtures' (ROM) can be expressed, as shown in Equation 2.13. The rule of mixtures can be used to describe a number of material properties as Equation 2.1 used ROM to describe the applied composite stress. The ROM (Equation 2.13) can be used to predict other properties such as composite density, strength, stiffness, *etc.* providing the properties of individual components are known. The ROM relationship according to Hull and Clyne (1996) should be accurate for indicating a composite stiffness, providing the reinforcement is long enough for the equal strain assumption to apply.

Equation 2.13 $$X_c = V_f X_f + (1 - V_f) X_m$$

Where: X_c The composite property.

X_f The fibre property.

X_m The matrix property.

Sanadi *et al.,* (1986) used the rule of mixtures to predict the tensile strength and modulus of composites comprised of continuous unidirectional sun-hemp (*Crotalarea juncea*) fibres bound with a polyester matrix. The composites were made with various fibre volume fractions (0-40%); laminates with low fibre volume fractions were fabricated with a lay-up process, whereas laminates with high volume fractions were manufactured with a pultrusion technique. Sanadi *et al.,* (1986) also carried out tensile testing of individual sun-hemp fibres. It was found that there was a good correlation between experimental values and the values produced from using the ROM for predicting composite tensile modulus and strength. The ROM follows the tensile modulus very closely, but Sanadi *et al.,* (1986) concludes that small discrepancies can be attributed to the difficulties in computing the exact cross sectional area of sun-hemp fibres. Obviously the fibre properties are required for the ROM formula, but because of the irregular shape of plant fibres, variation is likely to be high, as Section 2.3 on page 20 states.

2.7.3.2 Fibre architecture

It is important to discuss the fibre architecture, as composite properties are dependent on the orientation of fibres, and whether the fibres are short or long. Firstly, considering a composite system with long (continuous) fibres that run parallel to each other; this system is referred to as a 'unidirectional lamina'. A unidirectional lamina can be referred to as a 'ply' and a stack of laminae is called a laminate. Composites can comprise a single ply, or laminates with the same or different orientations. Figure 2.13 shows two laminates, one consisting of 6 plies that are at 90° to each other, whereas with the second laminate each ply is at different angle to the previous, apart from the two centre plies. Both stacking sequences shown in Figure 2.13 are symmetrical about the mid plies.

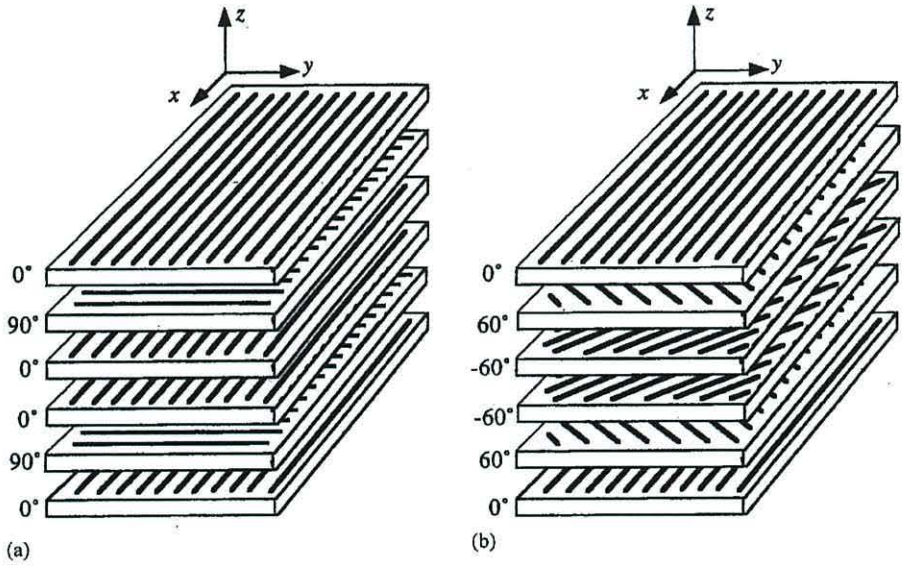


Figure 2.13 Schematic of the arrangements of lamina within two laminates (*Source*: Hull and Clyne, 1996).

In order to describe stacking sequences, the angle of each ply is denoted from the x direction as Figure 2.13 shows. Stacking sequence (a) in Figure 2.13 can be written as $[0/90/0_2/90/0]$. The subscript indicates that the middle plies are repeated twice. As previously mentioned the laminates are symmetrical and a subscript of 'S' is denoted, thus allowing for stacking sequence (a) to be simplified to $[0/90/0]_S$. Laminate (b) can be described as $[0\pm60]_S$.

Fibres, natural or synthetic, can be assembled into a woven fabric (roving) form. Characterisation of woven fabrics requires descriptions about weave type, yarn linear density (Tex system) and the number of yarns within a given distance. It may be necessary to describe warp and weft yarns separately, as they can often be different. Section 2.5.7.1 has already discussed possible weave types for plant fibres and summarised the Tex system for expressing the sizes of yarns. However, it is important to describe weave types by a numerical system. For instance the plain weave shown in Figure 2.5 can be written as $^1/_1$. In this instance, warp yarns pass over one weft yarn, then under one weft yarn. A weave type such as a $^1/_5$ will be woven so the warp yarns pass

over 5 weft yarns then under a weft yarn, then over 5 again. Hull and Clyne (1996) state that woven structures cause pockets of matrix to occur at the warp and weft cross-over points. As a consequence of these numerous cross-over points, the maximum fibre content is less than can be achieved with fully aligned (unidirectional) composites. Stacking sequences for woven laminates can be described with the same system used for unidirectional lamina. However, since there are yarns at 90° to each other within a woven ply, the angle associated with the orientation of lamina from the x direction must only refer to one direction of yarn (warp or weft). Plant fibres within a yarn will not lie straight and aligned; the orientation is changing continuously in three dimensions. It is likely that their orientation will mimic a helical structure like a spring, due to the twist given to the yarns.

2.7.4 Elastic deformation of unidirectional composites

The following two sections discuss models that can predict axial and transverse Young's modulus of composites comprised of continuous aligned fibres. Laminae within the composites are in the same direction $[0]_S$.

2.7.4.1 Axial stiffness

As previously mentioned in Section 2.7.3.1 on page 61, the ROM relationship can be used to express the axial stiffness of a unidirectional continuous fibre laminate loaded parallel to the fibre axis. The composite is treated with the assumption that as stress is applied in the direction of the fibres, there is an equal strain for the matrix and reinforcement that is equal to the composite strain. This 'equal strain model' for the prediction of axial modulus of a composite (E_1) also referred to as the 'Voigt model' is presented in Equation 2.14 (Hull and Clyne, 1996).

Equation 2.14 $$E_1 = V_f E_f + E_m(1 - V_f)$$

Where: E_f The modulus of the fibre.

E_m The modulus of the matrix.

2.7.4.2 Transverse stiffness

When composites experience loads transverse (or normal) to the aligned continuous fibres, it often causes the matrix to creep because it bears the highest stresses (Hull and Clyne, 1996). The Voigt model in Section 2.7.4.1 is an equal strain treatment; composites subjected to transverse loads are unlikely to have an equal strain between constituents. An 'equal stress model' referred to as the 'Reuss model' can be used to describe transverse properties. Hull and Clyne, (1996) state that the model gives a poor approximation of transverse modulus because of the non-uniform distribution of stress and strain. As mentioned in Section 2.7.1.4 on page 54, the reinforcing efficiency of fibres is reduced and their presence can be detrimental to the performance of the composite. The fibres can act as stress concentrators, which may initiate localised inelastic behaviour, as also discussed in 2.7.1.4 (Hull and Clyne, 1996). A prediction for transverse composite modulus (E_2) that is not based on elasticity theory and broadly takes account of enhanced fibre load bearing, relative to the equal stress assumption is the semi-empirical model by Halpin and Tsai (1967). Hull and Clyne (1996) believe that the Halpin and Tsai prediction (Equation 2.15) is the most successful model.

Equation 2.15 $$E_2 = \frac{E_m(1 + \xi\eta V_f)}{(1 - \eta V_f)}$$

Where: $$\eta = \frac{\left(\frac{E_f}{E_m} - 1\right)}{\left(\frac{E_f}{E_m} + \xi\right)}$$

ξ It is an adjustable parameter.

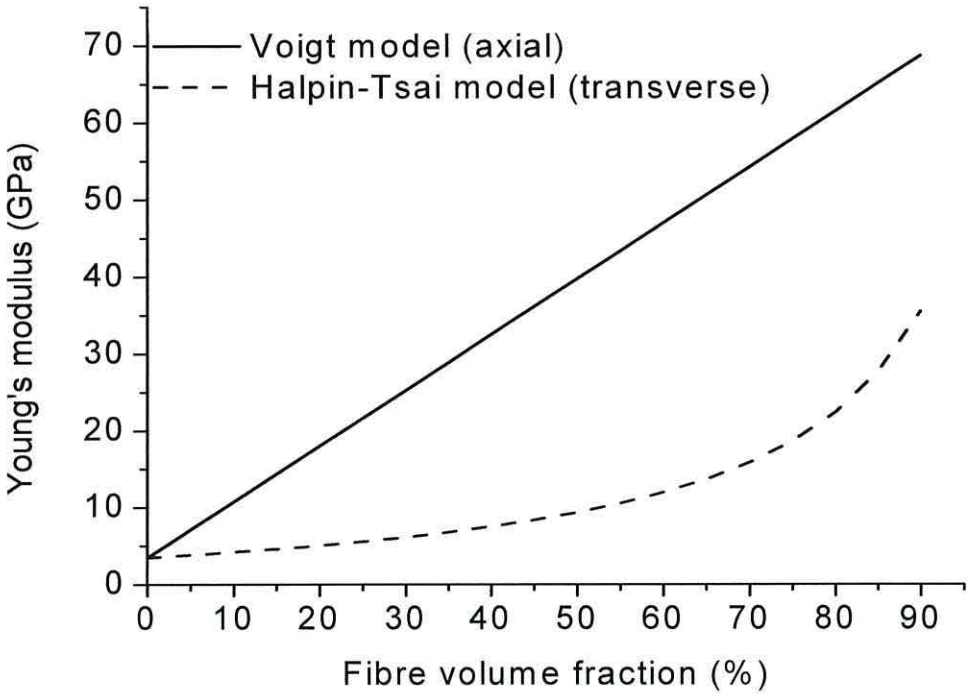


Figure 2.14 Predicted variation of axial and transverse composite moduli against fibre volume fraction.

The Halpin and Tsai model for predicting the transverse modulus and the Voigt model for predicting the axial modulus of a composite is represented in Figure 2.14 using the parameters for the hypothetical composite in Section 2.7.1.2 on page 49. The adjustable parameter for use in Equation 2.15 is assumed to be 1.

2.7.5 Failure modes of continuous fibre composites

Unidirectional composites when subjected to stress may fail if critical values of stress are exceeded whether it being by axial tensile stress (σ_{1u}), transverse tensile stress (σ_{2u}) or shear stresses (τ_{12u}) of the composite. It is possible that the failure mode may be only one of the above or a combination of two or three basic failure processes. Figure 2.15 shows the main failure modes (Hull and Clyne, 1996). Failure arising because of shear or transverse stresses causes a fracture parallel to the fibres, either the fracture can stay in the matrix and follow an interface or fracture can occur within the fibre. Failure as a result of a tensile stress applied parallel to the reinforcement can cause fracture perpendicular to the fibres, thus fibre and matrix fracture occurs.

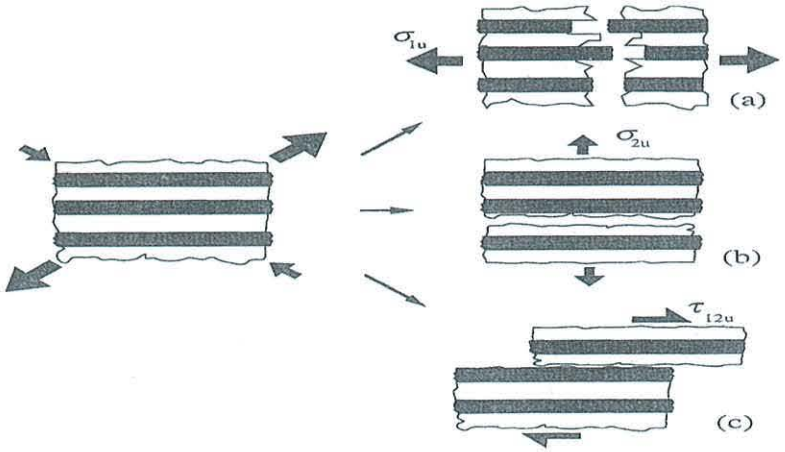


Figure 2.15 Schematic representation of the failure modes for a unidirectional composite due to (a) axial fibre stress, (b) transverse fibre stress and (c) shear stress (*Source*: Hull and Clyne, 1996).

2.7.5.1 Axial tensile failure

Two cases of failure can be identified for unidirectional composites, providing both phases are behaving elastically and exhibit brittle modes of failure when subjected to stresses parallel to the fibre axis. Axial tensile failure may occur with the reinforcement

failing before the matrix because the fibre strain to failure (ε_{fu}) is lower than the matrix strain to failure (ε_{mu}). The second is the opposite of the previous scenario, thus the matrix has a lower strain to failure than the fibre ($\varepsilon_{mu} < \varepsilon_{fu}$).

When fibre failure occurs before matrix fracture, fibres will break into shorter lengths within the composite, thus lowering the amount of stress that can build up along their length. The fibres may reach or drop below their critical aspect ratios. At the point where fibres have dropped below their critical aspect ratio, the matrix is thought to be carrying the entire load. According to Hull and Clyne (1996) when fibre failure occurs before matrix failure, the composite axial failure stress (σ_{1u}) can be given by Equation 2.16, providing the matrix fails whilst the fibres are still carrying some load.

Equation 2.16 $$\sigma_{1u} = V_f \sigma_{fu} + (1 - V_f)\sigma_{mfu}$$

Where: σ_{mfu} The matrix stress at the onset of fibre fracture.

If the matrix has a lower strain to failure than the fibre, axial composite stress can be given by the ROM relationship (Equation 2.1) providing matrix strain at failure has not been reached. As the strain of expected matrix failure is passed, the matrix will undergo microcracking and this can cause a 'knee' on the stress strain curve (Hull and Clyne, 1996). The composite at this point is still continuous to strain and the matrix cracks but there is little increase in composite stress. The breaking of the matrix leads to a higher proportion of load being borne by the fibre. If the assumption is made that the fibres are carrying the entire load, then the composite axial failure stress can by given by Equation 2.17.

Equation 2.17 $$\sigma_{1u} = V_f \sigma_{fu}$$

However if fibre fracture starts to occur before the matrix has broken enough to allow the fibres to carry the entire load, then the axial strength of the composite can be given by:

Equation 2.18 $$\sigma_{1u} = V_f \sigma_{fmu} + (1 - V_f)\sigma_{mu}$$

Where: σ_{fmu} The fibre stress at the onset of matrix cracking.

Hull and Clyne (1996) state that these predictions of composite axial stress are gross simplifications. After cracking, the matrix still carries a portion of the load and fibres that are fractured and broken into shorter lengths also carry a share of the load and the process of stress transfer continues after fracture of any one of the phases. The above models also presume that the fibre strength is constant along the length of the fibre and failure occurs in isolation. Hull and Clyne (1996) make the point that a fibre will firstly fail at its weakest region when it is subjected to a stress parallel to it's axis. This is a major consideration when applied to heterogeneous natural fibres.

2.7.5.2 Transverse failure

Numerous factors affect the transverse strength of composites, such as the presence of voids, the fibre distribution and the nature of the interfacial bonding between phases. The transverse strength of an aligned long fibre composite is often less than the unreinforced matrix, and has a reduced strain to failure (Hull and Clyne, 1996; Anderson *et al.,* 1990). Hull and Clyne (1996) describe a cross-ply laminate in which the plies that are transverse to the load start to crack before the parallel plies, even when they are carrying less load. As previously mentioned in Section 2.7.4.2 on page 67, the presence of fibres can be detrimental to the composite's transverse performance. High stresses and strains develop in the matrix as the composite is loaded transversely and due to the orientation of the fibres, they do not act as very effective reinforcement and thus do not add to the

composite strength. Cracks form at the interface and then propagate to other highly stressed sections of the matrix, providing that the interface is weak.

2.7.5.3 Shear strength

The shear strength of aligned long fibre composites is controlled by the shear strengths of the matrix and fibre and the interfacial shear strength between the fibre and matrix (Hyer, 1998; Piggott, 1980). Dependence on the above factors causes the shear strength to be low, thus aligned composites tend to fail in shear (Piggott, 1980). Shear failure can occur in any one of three shearing planes, as Figure 2.16 depicts. Hull and Clyne (1996) state that there is no simple analytical expression that is able to predict the effect of fibre volume fraction on the shear strength of composites.

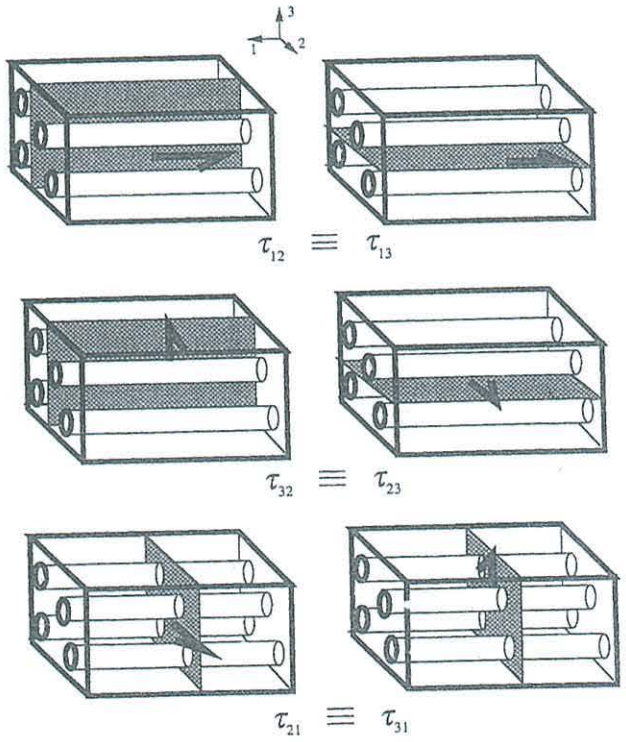


Figure 2.16 Possible planes in which shear stresses act and cause failure in a unidirectional composite (Source: Hull and Clyne, 1996).

2.7.6 Toughness

Toughness can be described as the material's ability to resist the propagation of cracks. Obviously materials require adequate toughness for their safe use and to increase their service life before cracks or crack-like defects propagate sufficiently to cause failure. Cracks or crack-like defects within composite materials arising from internal stresses, straining from an applied load or manufacturing defects cause localised stress concentrations. The severity of the stress concentrations formed depends on the size and shape of the cracks responsible. Sufficiently stressed sound composite material close to the tips of the cracks can fail. This can be localised or lead to the complete failure of the composite. Cracks can only propagate with the release of stored strain energy in the material and any external energy provided from the loading system (Hull and Clyne, 1996; Gordon, 1976). The 'work of fracture' is a measure of the work required to propagate a crack through a unit area of material. According to Piggott (1980) the work of fracture for glass is 0.005 kJ m^2 and for wood, across the grain it is 20 kJ m^2. Piggott (1980) summarises that ductile materials are in general tough and hard materials are usually brittle. Gordon and Jeronimidis (1980) make the point that stiff materials are more likely to break under dynamic conditions because there is less strain energy stored at any given stress. Gordon and Jeronimidis (1980) also suggest that for composites, it is a constant conflict to combine a high work of fracture with the best strength and stiffness properties. It is usually true that toughness can be improved to the detriment of stiffness or *vice versa*. Tough materials are those that are able to absorb energy as cracks advance or relieve the stress concentration at the crack tip (crack blunting). Ductile materials often have the ability to achieve the above, whereas brittle materials like glass fail with low fracture energies because they do not contain adequate mechanisms to prevent crack propagation. However two brittle materials such as glass and cured polyester resin when incorporated into a composite can achieve a reasonable toughness. Gordon (1976) and Cook and Gordon (1964) have shown that the mixture of materials such as the above creates numerous interfaces that facilitate energy adsorption and crack-blunting mechanisms providing the bond between phases is not too strong.

The interface as a mechanism to inhibit the propagation of cracks is reported in Section 2.7.2 on page 57. The stressed interface ahead of a crack (primary crack) can rupture, thus diverting the crack from entering the fibre; this can create secondary cracks that then run along the interface in both directions. Blunting of the tip of the crack may also occur. This action of crack diversion, results in energy being absorbed as new crack surfaces are created. Cook and Gordon (1964) state that in order for the interface to act as an efficient crack stopping mechanism, the adhesive strength of the interface has to be less than 20% of the matrix strength. Energy can also be absorbed in large quantities when interfacial frictional sliding occurs (Hull and Clyne, 1996), obviously some debonding will have occurred prior to this event. Fibres are pulled out from the matrix and the energy absorbed in the process depends on the distance they slide, interfacial roughness and contact pressure. Interfacial debonding according to Piggott (1980) usually has a low value work of fracture (approximately 0.5 kJ m^{-2}) and does not contribute greatly to the overall scheme; unlike frictional sliding and fibre pull-out, that can absorb large amounts of energy and are likely to be the most significant toughness enhancing mechanism for polymer matrix composites.

Other events that contribute to the composite toughness by absorbing energy include fracture and deformation of the matrix and fibre fracture. Thermosetting matrices such as an epoxy resin tend to have low fracture energies, between 0.1 to 0.3 kJ m^{-2} (Hull and Clyne, 1996), thus the matrix contribution to composite toughness is limited. However, the above values are for an unreinforced matrix and it is not reasonable to expect the same fracture energies for a composite. Fracture energies of glass and carbon fibres are also low and their contribution to the composite's work of fracture is unimportant, but natural fibres can (in theory) make a significant contribution (Hull and Clyne, 1996). For example wood across the grain has a fracture energy between 8 and 20 kJ m^{-2} (Hull and Clyne, 1996). The high toughness of wood may be due to deformation of fibres (tension buckling) that possibly occurs during loading because of the structural organization of the cell walls (mainly the S_2 layer). The deformation of the structure adsorbs a very large amount of energy (Gordon and Jeronomidis, 1980). When composites were fabricated with reinforcement containing elements that mimic the structure of natural composites

such as wood, Gordon and Jeronmidis (1980) found that is was possible to produce works of fracture comparable to ductile metals. Along with fibre pull-out, fibre fracture of natural fibres may also act as an excellent mechanism for adsorbing energy, thus increasing toughness.

2.7.7 Voids

Voids can occur in all types of composite, but the amount of voids often depends on the process used during fabrication, and the properties of the matrix (Hull and Clyne, 1996; Hyer, 1998). Voids can occur in matrix rich areas, such as the region between laminae, small voids also form adjacent to fibres as a result of deformation or insufficient wetting during fabrication. Voids can be created during fabrication because of air pockets becoming trapped, adsorbed water in the resin vaporising during curing and gaseous by-products released as the cure reaction occurs. Matrix cracking can also lead to void formation as a result of a build-up of residual stresses. Residual stress can arise due to the thermal expansion differences between phases. Matrix cracking is a possibility when the thermal properties of the phases are greatly different. Plant fibres obviously have lumens, which act as voids, unless filled with resin during fabrication or cell collapse occurs, thus closing the void. Piggott (1980) states that voids are usually concentrated at the fibre to matrix interface, where they weaken the composite by acting as stress raisers and sites for the initiation of cracking and debonding. Hyer (1998) also suggests that voids are extremely detrimental to the mechanical performance of composite materials. Hyer (1998) believes that the gas bubbles released due to the curing of resin during fabrication are a cause for delamination between plies within laminates, when a critical amount of gaseous bubbles (voids) collect at the interface.

2.8 Composite manufacture

The aim of the following sections are to describe possible open mould and closed mould manufacturing processes for composites bound with thermosetting resins. Reinforcement such as bast fibres can be obtained in many different formats (sliver, non-woven felted mats, woven fabrics), and some manufacturing routes will be better suited than others. The manufacturing process adopted and the type/style of reinforcement used may affect the reinforcement orientation and volume fraction. Hull and Clyne (1996) believe that during manufacture the following three objectives relating to the microstructure of the composite are important:

1. Fibres are well wetted with resin,
2. Reinforcement is evenly distributed,
3. Fibres are correctly aligned.

The method of composite fabrication influences the properties that the material exhibits (Anderson *et al.,* 1990).

2.8.1 Open mould processes

2.8.1.1 Hand laminating

Hand laminating is the most common form of composite fabrication (Hyer, 1998). The method involves the mixing of resin by hand and applying it with brush or roller to a mould containing reinforcement. Female moulds used in this process are often coated with a release agent. A layer of neat resin referred to, as a 'gel coat' is usually applied to the mould and allowed to partially set. Alternating layers of reinforcement and resin are then added and consolidated with hand rollers. Air bubbles are removed by the action of

rolling, thus the quality of composite is dependent on the skill of the laminator. Composites manufactured in this manner can consist of many layers to obtain the thickness required. During manufacture, layers can be allowed to gel before adding successive layers (Ball, 1994). The addition of core materials, metal inserts and ribs is possible during fabrication. After laminating and resin cure, the composite is removed from the mould.

2.8.1.2 Spray-up

This process involves catalysed resin and fibre being applied onto a mould simultaneously *via* a resin spay gun with an air driven fibre chopper unit attached. Continuous strands of reinforcement are chopped to short lengths and transported to the mould in the resin stream. Consolidation of fibre and resin is achieved using a hand roller. Composites tend to be excessively heavy because of the quantity of resin used and mechanical properties poor due to the short fibres and lack of orientation. However it is a very quick and low cost method.

2.8.1.3 Filament winding

The process involves a continuous strand of reinforcement fed through a bath of catalysed resin and wound onto a rotating mould (former). Resin-soaked reinforcement passes between rollers to remove excess resin before being incorporated into the composite in production. Composites are formed on a male former that is allowed to rotate. A traversing head (referred to also as a moving carriage) carries the resin bath and passes the impregnated fibre reinforcement onto the former. Rotation speed and traversing head angle/speed can be changed, thus influencing the fibre orientation and thickness formed. Tension applied to the reinforcement helps it to consolidate and achieve the desired fibre content. Composites produced *via* filament winding are smooth

inside because of the male former. Piping, pressure vessels and tubing are examples of products that can be manufactured.

2.8.2 Closed mould processes

2.8.2.1 Vacuum bag

The vacuum bag technique involves the application of reinforcement and resin by hand into a female open mould. A release film is placed over the material in the mould. A rubber bag is placed over the mould and the air between the bag and the mould is removed. Atmospheric pressure is applied to the composite, helping consolidation to occur. To ensure complete impregnation of resin into the fibre mat, it is sometimes necessary to hand roll. Lower void contents and higher fibre volume fractions can be achieved than with hand laminating.

2.8.2.2 Pressure bag

This is a similar process to the vacuum bag method, but can produce higher fibre volume fractions and better consolidation of reinforcement (Ball, 1994). High pressure is applied *via* a compressor that consolidates the composites faster and steam or heated air can be added to the bag to achieve quicker curing.

2.8.2.3 Autoclave

Autoclaving involves the processes of vacuum bag and pressure bag combined. A vacuum bag set-up is placed into a heated and pressurised vessel. The composite then has a vacuum applied as well as heat and external pressure enabling high fibre volume fractions to be achieved. The composite is fully impregnated, consolidated and heated to reduce cure time.

2.8.2.4 Leaky mould

Both male and female moulds are used, they are clamped together and the resin and reinforcement take the shape of the void between. The two moulds can be pressed together but they are designed so excess resin can escape. Benefits in using such a mould is that composites can be made with accurate dimensions and have excellent surface finishes (Ball, 1994).

2.8.2.5 Cold press

Reinforcement layers and resin can be added to a female section of a rigid mould that is able to withstand pressures of 2 bar or more (Ball, 1994). Placing the complete mould in a hydraulic press it is possible to achieve resin distribution, impregnation, and removal of air simultaneously. Curing has to be completed before the mould can be released and the composite extracted.

2.8.2.6 Hot press

The hot-press process is essentially the same as the cold press method but with the application of heat to accelerate curing. To allow heat to disperse quickly, metal moulds are used.

2.8.2.7 Resin transfer moulding

Resin transfer moulding (RTM) is a process where the reinforcement is loaded into female and male moulds and sealed. Degassed catalysed resin that is under pressure is allowed to enter the mould and impregnate the fibre. Hyer (1998) states that the resin flow must not be too quick into the mould because fibre wet-out will not occur and the reinforcement in the mould will not retain its shape. The flow is also important in controlling voids. Moulds are capable of being heated and this can be before or after infiltration of resin depending on resin gel times.

2.8.2.8 Vacuum assisted resin injection

Vacuum assisted resin injection is very similar to the resin transfer moulding process described. It differs by the fact that a vacuum is pulled before and during resin injection into the mould. Advantages of pulling a vacuum are that it gives better consolidation to the composite, removes any trapped air (lower void content), and enables a higher fibre volume fraction to be obtained.

2.9 Recent developments on natural fibre and natural fibre reinforced thermosetting PMC's

2.9.1 Flax fibre

Bos and Van den Oever, (1999) reviewed the influence flax fibre structure has upon the properties of the flax fibre themselves. Epoxy and polyester composites reinforced with flax fibres were also investigated. They found that the tensile strength of a technical flax fibre (fibre bundles) is dependent on the clamping length during testing. At clamping lengths above 25 mm, the fibres strength was constant at approximately 500 MPa. Clamping lengths below 25 mm resulted in the tensile strength increasing to 850 MPa. They summarise that the differences observed with fibre strength and different clamping lengths are because the fibre bundles mode of failure changes. Single elementary (single fibre) fibres from broken, scutched and hackled technical flax fibre had a tensile strength of 1500 MPa. Elementary fibres isolated by hand had a tensile strength of 1800 MPa. Bos and Van den Oever, (1999) state, that the decortication process damages fibres, thus lowering their strength. They go on to discuss how elementary fibre behaviour is influenced by its structure. The fibrillar structure of the S_2 layer within the cell wall makes the fibre sensitive to kink band formation under compressive loading. Determination of compressive strength using the loop test revealed that the compression strength of fibre is about 1200 MPa but after fibres have undergone decortication processes the compression strength can be reduced to 0 MPa due to the damage induced.

Bos and Donald, (1999) studied the deformation of retted and mechanically decorticated elementary flax fibres in a modified loop test using an environmental scanning electron microscope (ESEM) which allowed *in situ* mechanical measurements to be made. Straining of fibres induces compressive deformation on the inner side and tensile deformation on the outer side of the fibre. They found that because of differences in chemical composition and morphology of the two cell walls within a flax fibre, there is a

difference in the fibre's deformation behaviour between cell walls. The failure of the primary cell wall occurs in a brittle manner on the tensile face of the fibre loop, whilst the secondary cell wall fails under compression in a lateral direction because of the highly oriented cellulose. They found that prior to failure of the primary cell wall, cracks, bridged by coarse fibrils were formed in the secondary cell wall.

Nechwatal *et al.*, (2003) describe how numerous problems arise when testing single fibres from natural sources, such as the clamping of specimens and the determination of the cross-sections of the fibres. The cross-sectional area of a natural fibre changes along its length and fibres exhibit a polygonal shape. In order to calculate stress, an average fibre area must be given and the method used to calculate its cross-sectional area may not be exact (Baley, 2002). It is very difficult to measure the strain of a single fibre, since as a stress is applied the fibre can slip in the clamps in which it is held and therefore the fibres true elongation is not measured accurately. The strength of elementary fibres decreases with an increasing clamping length because the longer the stressed distance of the fibre the more inhomogenities should be in the stressed fibre segment which weakens the structure (Nechwatal *et al.*, 2003). Hornsby *et al.*, (1997) found no clear relationship between tensile strength and the tensile modulus and gauge length when testing elementary flax fibres with gauge lengths of 10, 15 or 20 mm. Furthermore, Hornsby *et al.*, (1997) states that because flax fibres contain many microfibrils, total failure under stressed conditions caused by the presence of surface or small internal defects would be limited. Nechwatal *et al.*, (2003) also describe that the calculation of fibre modulus from the initial region of the stress-strain curve can also be uncertain. The stress-strain behaviour of natural fibres is not linear, every fibre shows a different shaped initial curve, and thus different tangents can be fitted to the initial region of the curve.

Baley, (2002) studied the tensile behaviour of flax fibre under both static and cyclic loading conditions. Through the use of a SEM, Baley (2002) observed from fractured surfaces of a elementary flax fibres that the microfibrils in the secondary cell wall were directed along to the axis of the fibre and thus the direction of the applied tensile load. Baley, (2002) also states there is often a wide spread of values observed when measuring

the properties of flax fibres due to stress-strain response of the fibres being influenced strongly by conditions of the test. Baley, (2002) calculated the Young's modulus from the slopes of linear parts of stress-strain curves taking into account the dimensions of tested fibres *via* measurements conducted on an optical microscope. It was found that elementary flax fibres displayed plastic deformation after one cycle of loading and unloading. However, an increase in Young's modulus between 60 to 80% was observed between the first cycle and the last cycle of a single flax fibre when it had been loaded and unloaded approximately 200 times (cycles). Chapter 6 of this thesis describes how unidirectional flax fibre reinforced PMCs also show non-linear behaviour when strained. Baley, (2002) states that the observed fibre modulus increase was due to internal structural changes, namely that the angle of the fibrils was decreasing (to the longitudinal axis). During a tensile test in the direction of the fibre, cellulose fibrils orientate towards the direction of the load, prior to them sliding. Baley, (2002) postulates that as fibres are strained, deformation may occur in a number of ways, one being that the fibrils and the non-crystalline regions between fibrils increase in length. As the fibrils extend their volume reduces, as does the non-crystalline material between and any empty space present. Deformation may also occur by the shearing of the non-crystalline region in order with the new configuration of the fibrillar structure. Baley, (2002) states that for fibre deformation to occur in any of the above mechanisms, the stress applied to the fibre has to be above a threshold value, once past this value deformation is mainly controlled by chain entanglements. The failure of elementary fibres occurs by the propagation of a defect followed by the delamination of the fibre.

McLaughin and Tait, (1980) performed an extensive study of the fracture mechanism of fibres from various plant species and observed that Young's modulus and fibre tensile strength increases with decreasing microfibril angle and an increase in cellulose content. Hornsby *et al.*, (1997) as mentioned previously, also investigated the tensile strength and modulus of elementary flax fibres. Although the dimensions of the flax fibres tested were measured *via* a microscope, it was assumed that the fibres were round and possessed a uniform structure throughout their thickness. Hornsby *et al.*, (1997) found that the force-strain curves of flax fibres often displayed two peaks prior to total failure.

This mode of failure is a reflection on the layered wall structure within flax fibres. Testing the fibres to failure, it was observed that the force-strain curves showed curvature towards higher force levels with increasing strain. It was postulated that the modulus increase of the flax fibres, referred to by Hornsby *et al.*, (1997) as 'strain-hardening' was caused by the progressive reorientation of the microfibrils to the axis of applied load. The method Hornsby *et al.*, (1997) used to measure strain only recorded at each 0.1% strain increment thus the force-strain curves presented do not contain many data points. Baley (2002) found that the average Young's modulus of elementary fibres was 54080 MPa, the tensile strength was 1339 MPa with an average elongation at break of 3.27%. Davies and Bruce, (1998) observed elementary flax fibres to have an average static modulus of 51700 MPa, a tensile strength of 621 MPa and failure at an average strain of 1.33%. Both Baley (2002) and Davies and Bruce, (1998) observed large variations in mechanical properties, for example, Baley (2002) recorded the moduli of fibres from 25 GPa to as high as 95 GPa. Lamy and Baley, (2000) showed how the Young modulus of elementary flax fibres depends on the diameter of the fibre; if the diameter of the fibre increases the Young's modulus decreases. The Young's modulus of flax fibres were found to range from 78680 to 39030 MPa. Other reported mechanical properties of flax and hemp fibres are presented in Table 2.2 on page 23. Mwaikambo and Ansell, (2003) also observed that tensile strength of hemp fibres bundles increases as the bundles diameter is decreased following caustic soda treatment (NaOH).

Joffe *et al.*, (2003) tested the tensile properties of enzyme retted flax elementary fibres. Elementary fibres were obtained by hand from bundles and tested on an Instron testing machine at a straining rate of 0.5 mm/min. The gauge length was fixed at 10, 15 or 20 mm depending on the fibre's length. Joffe *et al.*, (2003) used a travelling close circuit digital (CCD) camera to observe the fibres before and during testing. Five diameter measurements were made along fibres lengths and the average taken, hence for analysis it was assumed that flax fibres have a constant diameter along their lengths. The average Young's modulus of the elementary flax fibres was found to be 89 GPa with a standard deviation of 35 GPa and the average tensile strength was recorded at 1100 MPa. The values are in good agreement with the majority of properties presented in Table 2.2 on

page 23 for flax. Joffe *et al.*, (2003) also performed single fibre fragmentation tests (SFFT) on untreated and treated flax embedded in an unsaturated polyester resin. Two fibre treatments were used; fibres were treated with acrylic acid at two concentrations and fibres were also treated with a vinyl trimethoxy silane. Specimens were prepared with a length of 20-30 mm with a width of 2-4 mm and a height of 2-2.5 mm. Testing was performed on a MINIMAT test machine at a rate of 0.1 mm/min. During testing if no new breaks along the fibre length occurred during an increase in strain of 1% the test was stopped (fragmentation saturation level) or if the specimen completely failed. Testing was observed through a microscope with a CCD camera attached. Strains in excess of 7% were recorded for some specimens. Joffe *et al.*, (2003) found through the SFFT that the strength of the flax fibres was between 1000 and 1600 MPa. They also evaluated the interfacial shear strength of the flax fibre polyester interface. The interfacial shear strength of untreated flax fibre and silane treated flax fibre with polyester was found to be on average 18 MPa, whilst for acrylic acid treated fibre at two concentrations the interfacial shear strength was 22 MPa.

2.9.2 Non-woven and Unidirectional Composites

Hughes (2000) produced composites using jute from Bangladesh and hemp grown within the UK as non-woven reinforcement, with a general purpose unsaturated polyester resin. A resin vacuum impregnation process was used to manufacture the non-woven jute and hemp reinforced composites. Glass fibre in the form of chopped strand mat (CSM) was also used as reinforcement and composites fabricated by hand lay-up for comparative studies. Tensile, flexural, Charpy impact strength and fracture toughness results were obtained. It was found that the non-woven jute and hemp fibre non-woven composites had good stiffness and acceptable strength, especially on a specific basis, when compared with glass fibre CSM with corresponding volume fractions. The limiting factor of the natural fibre non-woven composites was their toughness. It was observed that brittle failure occurred at low fibre loadings but at higher volume fractions shear fracture was observed. Using scanning electron microscopy (SEM) it was found that little fibre pull-

out had occurred. Cured resin was found within lumena and laminates had a low void content, suggesting that there was a good fibre to matrix interaction. The brittle fracture and good fibre to matrix bonding were thought to contribute to the low toughness of hemp and jute non-woven composites. Hughes (2000) stated that the low toughness and strength of plant fibre reinforced composites is partly due to the lower inherent strength of the natural fibres. Damage caused by processing and the highly random fibre orientation within felts results in the formation of stress concentrations. Stress concentrations due to fibre damage were thought to have caused microstructural damage when low loads were applied to the composites. The non-woven felted mats exhibited a fibre orientation that was highly random. Hughes (2000) assumes that prior to needle punching, the majority of fibres are aligned in the two planes of the felt. Examination of felted mats after needle punching revealed that a good proportion of fibres were orientated in a third plane (Y direction). It was assumed that the degree of needle punching and/or thickness and grammage of the felt contributed to the misalignment of fibres. Hughes tested this hypothesis and found that strength and stiffness of laminates reinforced with two layers of 350 gm^{-2} hemp was greater than laminates with only one single layer of 700 gm^{-2} material. Hughes believes that the single layer of felt had a greater number of fibres misaligned in order to lock together the felt. The thinner laminates used, did not require so many fibres misaligned to keep the felt together resulting in a more efficient reinforcement. Fracture of non-woven composites produced with a single layer felt occurred at the needle punching points. It was discovered that needle-punched non-woven felts required large external pressures to consolidate the mat. A hot press was used but it was only possible to produce volume fractions of less than 50%. Composites could only be produced to a maximum volume fraction of 20% without pre-pressing. Composites that were made at volume fractions lower than 25% displayed lower strength than unreinforced cast resin.

Sèbe *et al.*, (1999), used hemp fibre in the form of non-woven mats for reinforcement in polyester composites produced by vacuum assisted resin transfer moulding (RTM). Hemp fibre was chemically modified to give reactive vinylic groups on the surface of fibres *via* esterification of hemp hydroxyl groups, using methacrylic anhydride. The

modification of fibres resulted in strong interfacial adhesion between the fibre and matrix. Flexural properties of the RTM composites were found to be acceptable. Unmodified hemp RTM composites above 30% fibre weight fraction (wt) were stiffer than chopped E-glass RTM composites at 15%wt. Toughness was found to be lower than pure polyester resin when composites had 11% hemp fibre weight fraction. At low weight fractions Sèbe *et al.*, (1999), believes that there is a disproportionately high number of critical defects in the form of voids and/or poorly bonded interface regions. Composites with higher weight fractions of fibre overcome the defects and achieve better impact strengths. However, they are poor when compared with glass fibre. It is believed that numerous micro-compressive defects in the form of small kinks found on the microfibillar structure contribute to the low toughness of hemp-reinforced composites (Sèbe *et al.,* 1999; Hughes, 2000; Hughes *et al.*, 1999). Plate 2.1 shows several micro-compressive defects on a single flax fibre.

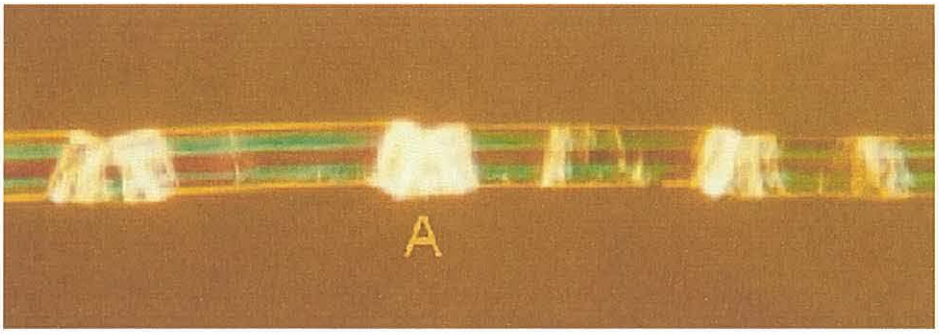


Plate 2.1 Micro-compressive defects in a flax ultimate seen under polarised light (*Source*: Hughes, 2000)

Fractured surfaces of unmodified hemp reinforced composites were compared to surfaces from chemically modified hemp reinforced composites. It was found that some fibre pull-out occurred with unmodified hemp, but modified hemp had a smooth fracture surface with no pull-out. Composites reinforced with modified hemp were much stiffer but had a lower Charpy impact strength. The modification led to a better fibre to matrix interaction that did not affect the load bearing capabilities of the fibres but reduced toughness by preventing fibre pull-out and causing brittle failure.

Shawkataly and Ismail, (2001) investigated the effect of biological exposure upon the properties of acetylated and surface treated plant fibre reinforced polyester composites. Results were compared to cast resin and glass-fibre composites. A major restriction to the successful use of natural fibres in durable composite applications is their high moisture absorption and poor dimensional stability, as well as their susceptibility to rotting (Stamboulis *et al.*, 2001). Shawkataly and Ismail, (2001) used hybrid non-woven mats of oil palm empty fruit bunch (EFB) and coconut (coir) fibre as reinforcement at a 45% fibre loading with polyester resin. The fibre mats were used in an unmodified form but some reinforcement mats were acetylated with acetic anhydride to a 10% weight gain and others were treated with silane or titanate in order to create stable bond between the fibres and the matrix. It was found that after 12 months of exposure to soil at 50% moisture content the acetylated EFB and coir composites showed the highest degree of protection of the natural fibre reinforced composites but it was not as high as the glass CSM composites or the cast resin panels. Shawkataly and Ismail, (2001) state that the unmodified and titanate EFB and coir reinforced polyester composites may be less protected than the acetylated fibre reinforced composites because of poor fibre to matrix bonding that resulted in localised delamination which in turn led to an ingress of fungi. Shawkataly and Ismail, (2001) found that the growth of fungi on or in composites was proportional with the moisture content of specimens. After 12 months of exposure, Shawkataly and Ismail, (2001) observed that the loss in tensile strength and modulus of unmodified coir and EFB reinforced polyester composites was highest. After the unmodified specimens, titanate treated fibre composites had the second highest loss, followed by silane treated fibre composites. Acetylated fibre reinforced polyester composites had the lowest strength and modulus losses. The enhanced compatibility between acetylated fibre and matrix because of the change in characterisation of the fibres surfaces to a more hydrophobic nature lead to an improved wetting of the fibres by the polyester resin and thus a stronger interfacial bond (Hill and Shawkataly, 2000). As well as creating an interface where stress transfer can occur more efficiently between fibres, the acetylated coir and EFB reinforced polyester composites are highly resistant to fungal attack like the glass CSM reinforced composites.

The work of fracture of a small selection of natural and glass fibre PMC's are presented in Table 2.7 on page 90. Comparing the work of fractures of natural fibre reinforced PMC's to those of glass reinforced PMC's displayed in Table 2.7 it is clear that the glass composites are considerably tougher at lower fibre volume fractions. However, it is very difficult to compare values of toughness when the methods used to obtain figures are different (see Section 3.3.7 on page 158). Table 2.8 on page 91 shows the flexural and tensile properties of a selection of natural and glass reinforced PMC's. As can be seen from the table, a few of the natural fibre reinforced composites have comparable properties to glass fibre reinforced properties at similar fibre loadings. The above work on PMC's involving natural plant fibre non-woven materials for reinforcement has demonstrated that they can compete with glass fibre reinforcement equivalents in terms of flexural stiffness and sometimes strength but that toughness remains the limiting factor. The following factors are thought to be reasons for the material's lack of toughness.

- Lack of fibre orientation, especially around needle-punched areas of felt
- Lower strength of fibres due to damage during processing
- Micro-compressive defects along fibres causing stress concentrations
- Stress concentrations due to defects and fibre orientation causing microstructural damage at low loads (possibly prior to testing)
- Good fibre to matrix bonding was thought to contribute to brittle failure
- Fibres have a non-uniform cross section limiting fibre pull-out.

Plant fibre non-woven reinforcement is bulky, making processing difficult and high volume fractions hard to obtain. Without surface treatment, plant fibre reinforced polyester composites display low resistance to decay by fungi, likely to be due to the hydrophilic nature of the fibres.

Table 2.7 Summary of the work of fracture of a number of natural and glass fibre reinforced thermosetting polymer matrix composites.

Type of reinforcement	*Charpy or Izod*	*Notched or Unnotched*	V_f%	*Work of fracture (kJ m^{-2})*	*Source*
Polyester matrix					
Unidirectional jute	Charpy	Notched	60	22	Roe and Ansell, 1985
Unidirectional Sunhemp fibres	Izod	Notched	24	21.5	Sanadi *et al.*, 1986
Randomly orientated pineapple fibre	Charpy	Unnotched	30*	24.2	Devi *et al.*, 1997
Hemp non-woven mats	Charpy	Unnotched	~30	14	Sèbe *et al.*, 1999
Hemp non-woven mats	Charpy	Unnotched	~20	11.5	Hughes *et al.*, 1999
Woven Jute fabric	Charpy	Unnotched	45	29	Gowda *et al.*, 1999
Jute non-woven mats	Charpy	Unnotched	36	14.3	Hughes, 2000
Chopped E-glass	Charpy	Unnotched	~7	34	Sèbe *et al.*, 1999
Glass chopped strand mat	Charpy	Unnotched	~20	80.4	Hughes *et al.*, 1999

** Fibre weight fraction*

Table 2.8 Summary of the mechanical properties of a number of natural and glass fibre reinforced thermosetting polymer matrix composites.

Type of reinforcement	V_f%	*Flexural modulus (GPa)*	*Flexural strength (MPa)*	*Tensile modulus (GPa)*	*Tensile strength (MPa)*	*Source*
Phenolic resin						
Unidirectionally aligned flax thread	75	-	-	48.2	482	Aero Research Ltd. 1945
Polyester matrix						
Unidirectional jute	60	-	-	35	250	Roe and Ansell, 1985
Unidirectional sunhemp fibres	24	-	-	11.2	125	Sanadi *et al.*, 1986
Randomly orientated pineapple fibre	30*	2.76	80.2	2.3	52.9	Devi *et al.*, 1997
Hemp non-woven mats	~30	~6	~98	-	-	Sébe *et al.*, 1999
Hemp non-woven mats	~20	4.72	101.9	-	-	Hughes *et al.*, 1999
Woven Jute fabric						Gowda *et al.*, 1999
Longitudinal (warp)	45	5.1	92.5	7	60	
Transverse (weft)	45	-	-	3.5	35	
Jute non-woven mats						Hughes, 2000
Parallel to fibre direction	~35	7.36	104.4	~9	~69.4	
Perpendicular to fibre direction	~35	5.35	78.2	-	-	
Chopped E-glass	~7	~5.6	~108	-	-	Sébe *et al.*, 1999
Glass chopped strand mat	~20	9.28	233.8	7.9	73.4	Hughes *et al.*, 1999 and Hughes, 2000

** Fibre weight fraction*

Devi *et al.*, (1997) fabricated randomly oriented pineapple leaf fibre reinforced polyester composites at various fibre loadings. Composites contained fibres chopped to different lengths ranging from 5 mm to 40 mm. Devi *et al.*, (1997) observed that the tensile stress-strain behaviour of the composites fabricated at a 30% fibre weight fraction reinforced with different fibre lengths was linear at low strains but non-linear at higher strains. Composites reinforced with pineapple leaf fibres 30 mm in length were found to exhibit optimum tensile properties when compared to the composites reinforced with longer (40 mm) or shorter (20 mm, 10 mm and 5 mm) fibres at the same fibre weight fraction. Due to effective stress-transfer, composites reinforced with 30 mm long fibres had a Young's modulus 180% higher than composites reinforced with fibres at a length of 5 mm and a tensile strength that was 240% higher. As fibre length was increased to 40 mm a reduction in tensile strength and Young's modulus of composites was observed. Devi *et al.*, (1997) state that the decrease in tensile properties was due to fibre entanglement. It was found from observing the fractured composite specimens that long fibres tended to bend or curl during moulding that caused a reduction in the effective length. Devi *et al.*, (1997) found that stressed fibres shorter than 30 mm debonded from the matrix causing failure at low strengths. A good fibre to matrix interface is essential for short fibre reinforced composites if efficient stress transfer is to occur (Baiardo *et al.*, 2004).

Bos and Van den Oever, (1999) fabricated unidirectional polyester and epoxy composites using dew-retted flax fibre as reinforcement. Tensile, flexural and compression tests were preformed on cylindrical specimens measuring 25 mm in length with a diameter of 6 mm. The tensile strength of the unidirectional polyester composites increased with fibre content following a ROM relationship. The compressive strength of the composites did not increase with fibre content but stayed at a similar level to the unreinforced polyester resin (approximately 100 MPa). Bos and Van den Oever, (1999) found that the adhesion between fibre and the matrix was very poor. It was postulated that the thin waxy layer surrounding flax fibres prevented coupling (adhesion) between the two phases. Bos and Van den Oever, (1999) found that the removal of the waxy layer did not significantly improve the unidirectional composites compressive strength. Several compatibilizers were also used in an effort to improve/increase the chemical bonding

between the fibres surface and the polyester resin. As the compatibilizers failed to improve properties, Bos and Van den Oever (1999) concluded that the composites low compressive strength was either caused by lack of adhesion, or by the flax fibres themselves because of the defects (kink bands) that exist along their lengths. The tensile strength of unidirectional epoxy composites also increased with fibre content in a linear fashion (ROM). The compressive strength of the unidirectional epoxy composites was observed to be 120 MPa. Bos and Van den Oever, (1999) removed the fibres waxy layer and observed an increase in the compressive strength of the composites. However, they concluded that the adhesion between epoxy resin and flax was not good. Application of melamine-formaldehyde on to the fibre to act as a compatibilizer resulted in a significant increase in the compressive strength of the epoxy composites. The highest compression strength was observed when 20% (by weight) melamine-formaldehyde was added to the fibres. Bos and Van den Oever, (1999) observed that the adhesion between phases had improved but it was found that melamine-formaldehyde was not just acting as a compatibilizer and attaching to the fibres surface but it was penetrating into the fibres themselves. Such loadings of melamine-formaldehyde onto fibres resulted in the tensile strength of composites reducing. It was postulated that the melamine-formaldehyde within fibres also penetrated kink bands helping to stabilise the fibres under compression loading, thus improving the compressive strength to 300 MPa. The decrease in tensile strength was thought to be caused by stress concentrations within the fibres that led to premature failure. The stress concentrations were caused by the melamine-formaldehyde within the fibres located at the kink bands. Bos and Van der Oever, (1999) concluded that kink bands may impede the use of flax fibres in structural applications, unless alternative decortication methods are developed that reduce there frequency. Keller *et al.*, (2001) also states that fibre damage caused by the decortication process have to be avoided to achieve high quality hemp fibres for industrial uses, such as for the reinforcement of polymers.

Van de Weyenberg *et al.*, (2003) fabricated unidirectional epoxy composites reinforced with flax fibres that had undergone different degrees of processing. Chemical fibre treatments were investigated to improve the adhesion between the fibres and the epoxy

matrix. Van de Weyenberg *et al.*, (2003) utilised flax fibre from the same variety of flax. After harvesting the crop, the flax was split into three parcels. One parcel was green flax and was used as such, the second was partially retted and the third parcel was entirely retted. All three parcels were then scutched, hackled, carded, refined and spun into yarns. Van de Weyenberg *et al.*, (2003) fabricated unidirectional epoxy composites using scutched long flax, hacked slivers and carded slivers from all three retted parcels. Composites were fabricated at a 40% fibre volume fraction. This was determined by weight measurements and assuming that the fibre density equals 1450 kg m^{-3}. Chemical treatments were performed on completely retted scutched long flax fibre. The first of these was the dipping of flax fibre into different concentrations (1, 2 and 3%) of sodium hydroxide (NaOH) in an effort to de-wax, delignify and to bleach the fibre. A pre-impregnation of the fibre with dilute epoxy at different concentrations (1, 3, and 5%) was also performed and the soaking of fibres in silane (1% concentration) in a solution of water and acetone (50/50 by volume) to create a coupling agent. In addition, a combination treatment to the fibre of alkali and dilute epoxy was performed. Tensile tests were performed on the untreated fibre composites and three point bending tests on the treated fibre composites. The longitudinal (fibres in loading direction) and transverse properties of the composites were recorded. It was found that there was a large amount of scatter on the recorded values but the composites reinforced with fibre that had been completely retted displayed the best properties. The composites reinforced with green flax displayed low properties, because the fibres incurred more fibre damage as a result of the higher amount of mechanical labour needed to separate the fibres from the green flax. Van de Weyenberg *et al.*, (2003) thought that because the green flax had not been exposed to microbial activity the fibres were coarser and impure and that these impurities between fibres caused stress concentrations within the composites that led to early fracture. Despite expectations, it was found that composite properties remained the same or even increased with further processing, regardless of the type of retting implied. Refining of the fibre outweighs the damage caused to the fibres during previous processes because fibres have a more orientated structure and contain less areas of pectin that are thought to be weak. Van de Weyenberg *et al.*, (2003) found (as expected) that the transverse flexural properties of all treated composites were higher than the properties

of the untreated composites because of an improved adhesion between the two phases. By removal of pectins with NaOH a 30% increase in longitudinal flexural strength and modulus of composites was achieved compared to the untreated composites. Treating fibres with NaOH has two effects, the first is that it increases the surface roughness of the fibres allowing for better mechanical interlocking and the second is that it increases the amount of cellulose exposed on the surface of the fibre, thus increasing the number of possible reaction sites (Valadez-Gonzalez *et al.*, 1999). Due to resin penetrating in fibres *via* micropores and between elementary fibres, the use of dilute epoxy resin also benefited the composites flexural properties. However, by using a combination of both treatments it was possible to increase the flexural modulus by 60% and the strength by 40% when compared to the untreated composites flexural properties. The strength of the composites could also be improved by treating the fibres with acetone because it removes waxy material allowing a better adhesion with the matrix. However, high concentrations of acetone can lead to fibre damage and a reduction in composite modulus. It was concluded that the properties of the flax reinforced composites were lower than glass fibre equivalents, even after reinforcement had been treated to improve adhesion at the interface.

Hepworth *et al.*, (2000) investigated the properties of high flax and hemp fibre content thermosetting composites. The damage to the fibres, degree of retting, fibre surface treatments and method of fabrication were also investigated. Flax fibre bundles were obtained from retted tissue using a mechanical decorticator and from retted and un-retted stems by hand. Flax and hemp unidirectional epoxy (Ampreg 26 with slow and fast hardeners) and phenolic composites were fabricated in a closed mould. The fibre volume fractions were controlled by adding known weights of fibre and resin and applying different amounts of pressure to the mould. Some hemp and flax fibre was pre-treated with 6 M urea and used for the reinforcement in epoxy composites. In addition, combed decorticated flax fibres were pre-treated with 50% mix of PVA (Unibond) and water. These fibres were then hand pressed to form a mat. Mats were dried to facilitate the curing of PVA and to remove excess water. Epoxy or phenolic resin was then poured over the mat and distributed; the composites were left to cure. Dumbbell shaped

specimens were obtained from the composites and the tensile properties recorded. Transverse sections from the composites were also investigated under a light microscope. It was found that there was good contact between the epoxy resin and hemp and flax fibres, although the resin did not penetrate into the cell lumens. The curing of epoxy resin with a fast hardener reduced the modulus of the composites. Epoxy composites fabricated with urea treated flax fibre had higher moduli and resin had penetrated into fibre bundles. The modulus and strength of epoxy composites reinforced with flax fibre increased with fibre volume fraction. Composites reinforced with strips of hemp and flax fibre tissue exhibited low moduli because of the absence of adhesion between fibre strips and resin in localised places. Resin did not penetrate into strips and adhesion was poor on the outside of strips because of the waxy cuticle. Fibre tissue was obtained after further retting of the hemp stems and composites were fabricated. These composites had a much higher modulus. Hepworth *et al.*, (2000) state that the resin was able to penetrate into the fibre tissue and between bundles because of gaps that had formed from the microbial degradation. The moduli of these composites were greater than the composites reinforced with decorticated fibre. At high fibre volume fractions (70% or above) composites with moduli of up to 26 GPa and a tensile strength of 378 MPa could be produced when flax tissue had been retted and treated with 6 M urea and by replacement of cell water with alcohol and then embedding in a mixture of resin and alcohol. Although these composites had lower moduli and strength than glass fibre CSM composites fabricated in a similar way at a similar fibre volume fraction they were comparable when the specific modulus was compared. Epoxy composites reinforced with PVA treated fibre that had been retted and mechanically decorticated had higher moduli and strengths than any other type of composite fabricated at a fibre volume fraction of 40%. The pre-treated PVA fibre reduced the complexity of processing by enabling preformed mats to be made and also prevented them swelling as epoxy resin was added. Hepworth *et al.*, (2000) state that the epoxy resin penetrated into the fibre structure and made a good bond with the PVA on the fibres. They also report that the critical transfer length of flax fibre cells in epoxy resin is less than 1 cm; the fibres are often longer than this. The length of mechanically decorticated fibre bundles are more than 10 cm long and contain numerous fibres, Hepworth *et al.*, (2000) state that

interfacial strength within these bundles of fibres are probably greater than the interfacial strength between the fibres and the resin. They conclude that the separation of fibre bundles into individual fibres for reinforcement does not matter. Since the mechanically decorticated fibre contains damaged regions which limit the stress carried by the fibre bundles and thereby reduces the modulus and strength of composites, this is why undamaged strips of retted fibre tissue provide a better reinforcement. Hepworth *et al.*, (2000) found that when modulus and strength were plotted against fibre volume fraction a 'J' shaped curve was formed. It was postulated that a linear response was not observed because the pressures used to fabricate composites at low fibre volume fractions were low and as a result fibres may not be as well orientated. The density of the composites also did not follow a ROM relationship. It was found that the production of composites at high fibre volume fractions required the application of high pressure. The pressure causes the fibre cell lumens to collapse and therefore causes an increase in fibre density, this does not reduce the fibre's reinforcement capabilities.

O'Donnell *et al.*, (2004) and Dweib *et al.*, (2004) have used a plant oil based resin (acrylated epoxidised soybean oil) and natural fibre mats to fabricate composites and foam core composite beams. O'Donnell *et al.*, (2004) made composites using vacuum assisted resin transfer moulding (VARTM) techniques; composites with fibre weight fractions ranging from 10 to 50% were achieved. O'Donnell *et al.*, (2004) state that vacuum infused parts have higher void contents than composites fabricated using other methods. The relatively low pressures used may not be sufficient to draw out trapped air and replace it with resin, and the bags used must maintain a seal as well as all the pipe connections into the bags at all times. In addition, the mixing of resin and catalyst often causes bubbles which are then sucked with the resin into the composite part. Dweib *et al.*, (2004) have fabricated composite structural beams with a polyisocyanurate foam core by (VARTM using a soybean oil based resin and various natural fibres including flax non-woven mats. Woven E-glass reinforcement was also used. Beams comprised a top and bottom face sheet with two vertical webs. The face sheets and webs were 6.4 mm thick. Dweib *et al.*, (2004) found that there were no problems with resin flow through the flax mats during fabrication as they had a high porosity. Four-point bending tests were

performed and it was found that the foam core beams reinforced with flax mats failed in a brittle manner. Failure initiated on the tensile face splitting the beam cleanly into two parts. Beams reinforced with woven E-glass failed in a more ductile manner with buckling of the top face sheet because of compression. The flax beams exhibited a failure load of 10.2 kN and a global modulus of 300 MPa whilst E-glass beams failed at 39.3 kN and had a modulus of 1580 MPa. Dweib *et al.*, (2004) are focusing on scaling up the fabrication process in order to manufacture a complete roof for a house, although it is likely that recycled paper and a small quantity of E-glass fibre will be used as reinforcement rather than flax mats as these fibres yielded the highest properties so far.

2.9.3 Yarn and Woven reinforced natural fibre composites

Satyanarayana *et al.*, (1986) used natural fibres in the form of a mat and woven fabric to reinforce composites made by a hand lay-up technique. Banana-cotton fabric was prepared on a normal weaving unit and a general purpose polyester resin was used. Banana-cotton fabric reinforced polyester composites were fabricated with fibre weight fractions ranging from 9 to 18%. A resin rich surface (gel coat) was applied to the composites. Satyanarayana *et al.*, (1986) tested the tensile, flexural and impact resistance of unnotched specimens. Glass fibre CSM reinforced composites containing 25-45% weight of reinforcement were also fabricated and tested. Table 2.9 shows a selection of properties observed by Satyanarayana *et al.*, (1986).

Table 2.9 Properties of cast resin polyester, glass fibre CSM reinforced polyester composites, cotton fabric reinforced polyester composites and banana-cotton fabric reinforced polyester composites as found by Satyanarayana *et al.*, (1986).

Properties	*Polyester resin*	*Glass fibre (CSM) composites*	*Cotton fabric reinforced composites*	*Banana-cotton fabric reinforced composites*		
				9% wt	*14% wt*	*18% wt*
Density (kg m^{-3})	1300	1100 - 1400	1400	-	-	-
Tensile strength (MPa)	41.30	62.06 - 137.90	45 - 62	25.86	30.96	29.50
Tensile modulus (GPa)	2.06	5.5 -12.4	-	1.36	2 – 3.34	1.98
Flexural strength (MPa)	99.69	137.9 – 275.06	89 - 124	52.38	61.24	60.40
Flexural modulus (GPa)	-	-	2.76 – 4.14	-	4.16	-
Impact resistance (kg m^{-1})	-	-	21.4 - 35.6	-	740.5	-

Satyanarayana *et al.*, (1986) postulate, that the low strength of the natural fibre reinforced polyester composites could be due to a lack of bonding between the two phases. SEM micrographs revealed debonding between the fabric and resin. A 50% increase in the modulus of banana-cotton fabric reinforced composites was observed when the weight fraction of reinforcement was increased from 9% to 14%. Satyanarayana *et al.*, (1986) state, that a fibre weight fraction of 14% was the highest achievable with the hand lay-up process. Ghosh and Ganguly, (1993) also found that a general purpose unsaturated polyester resin did not wet the surface of jute fibres because of the fibre's hydrophilic nature. Polyester matrix composites reinforced with woven jute fabric (350 g m^{-2}) had poor strength and a poor environmental resistance. The flexural strength and flexural modulus and maximum strain of unmodified woven jute fabric reinforced polyester composites with a fibre weight fraction of 44% was 89 MPa , 9 GPa and 3.97%

respectively. Unfortunately Ghosh and Ganguly, (1993) do not state whether warp or weft yarns were parallel to the longitudinal axis of the specimens tested. Woven jute fabric was chemically modified by either polyacrylonitrile (PAN) or poly (methyl methacrylate) (PMMA). Various different approaches were used to introduce phenol or resorcinol formaldehyde resin to the fibres to evaluate the ability of the modified jute fabric as reinforcement in polyester based composites. Improved composite strength was observed when modified woven reinforcement was used. At best, modified woven jute composites with the same fibre loading as unmodified composites had a 49.4% higher flexural strength and a 58.9% higher flexural modulus than unmodified composites. The maximum strain at failure for all modified woven jute reinforced polyester composites reduced significantly. Ghosh and Ganguly, (1993) postulate, that modified jute fibre may be more compatible with the polyester matrix which caused an improved adhesion between phases. Jute fibres also had a better resistance to moisture because of the hydrophobic nature of the grafted copolymers.

Gowda *et al.*, (1999) evaluated the mechanical properties of woven jute fabric polyester composites. Prior to spinning, jute fibre is softened and lubricated (sized) in order to minimise fibre breakage and waste. Gowda *et al.*, (1999) used a unidirectional type of jute fabric with a yarn count of 20 × 12. The woven jute consisted of 20 warp yarns with a Tex of 302 and 12 weft yarns with a Tex of 245. Specimens were prepared individually to avoid voids and to minimise cutting. To fabricate specimens a hand lay-up process was implemented; each ply of woven jute was pre-impregnated with a general purpose polyester resin and stacked over the other in a mould that had been coated with PVA to facilitate removal. Care was taken to ensure fabric alignment. Fibre volume fractions of 45% were achieved. Strain was measured *via* a clip on extensometer during testing on a computer controlled servohydraulic testing machine. At least 5 specimens were tested for each property for statistical purposes. The tensile strength and modulus of the yarn and fabric were tested assuming that the cross-sectional area of a yarn is circular. It was found that jute yarn is stiffer than jute fabric because of the initial stretching of the fabric. Ultimate tensile strength of jute fabric was 85 MPa with a tangent modulus (after the slack had been taken up by the system) of 0.8 GPa. Gowda *et al.*, (1999) studied the

tensile stress-strain behaviour of jute laminates and observed that initial portion of the curves were linear. Matrix cracking was thought to have caused non-linear behaviour after the elastic behaviour. However, the first major change in the slope of the curve was thought to be caused by further matrix cracking or the beginning of fibre failure. The first fibre failure occurred at a stress of 26 MPa and further drops in the slope of the stress-strain curves were caused by progressive fibre failures. Little fibre pull out was observed but yarns had fractured. The neat polyester resin had an ultimate tensile strength of 12.1 MPa, an initial tangent modulus of 1.4 GPa and a Poisson's ratio of 0.38. The woven jute reinforced polyester composites exhibited a tensile strength of 60 MPa, an initial tangent modulus of 7 GPa and a Poisson's ratio of 0.25. The tensile strength and modulus obtained from the ROM was 63 MPa and 1.12 GPa. Gowda *et al.*, (1999) postulate that the differences observed between the experimental modulus and the ROM modulus was because of the initial stretching of the fabrics. The composites were also tested in the weft direction and the composite properties were found to be lower. The ultimate tensile strength of weft tested specimens was 35 MPa with a transverse modulus of 3.5 GPa. Gowda *et al.,* (1999) concluded that these differences were because there was a greater amount of yarns in the warp direction than the number of yarns in the weft direction per unit dimension. The longitudinal (warp direction) compressive strength of the composites was recorded at 45 MPa, whilst that of the cast resin was 47.1 MPa. However, because the stiffness of a composite depends on the reinforcement, the modulus of the composites under compressive loading was 2.1 GPa and the cast resin's was 0.94 GPa. Compressive specimens failed in shear, with fibre buckling and the fracturing of fibre to matrix interfaces. Failure of flexural specimens originated on the tensile face and propagated in an upward direction. Gowda *et al.*, (1999) states, that the flexural strength and modulus of these composites are controlled by the strength of the extreme layer of reinforcement. Little or no fibre pull-out and delamination occurred. Non-linear behaviour from the onset of the flexural test was observed. The average flexural strength of the woven jute composites was 92.5 MPa, which is greater than the tensile or compressive strengths of the composites. The flexural modulus was 5.1 GPa. Gowda *et al.*, (1999) used the Charpy impact test to measure the average impact energy per unit area of the composite and cast resin. The average impact energy of the

composites was 29 kJ m^{-2} whilst the resin was lower at 1.76 kJ m^{-2}. Jute composite Charpy impact specimens did not fracture into two pieces. Gowda *et al.*, (1999) concluded that the woven jute polyester composites do not have the mechanical properties of conventional composites but do have better strength than some wood composites.

Jute fibre yarns with a Tex of 280 were dewaxed / desized in methanol-benzene for 24 hours before treatment with NaOH at different concentrations (Gassan and Bledzki, 1999). Unidirectional epoxy composites were also fabricated with untreated and alkali treated fibre yarns. Gassen and Bledzki, (1999) calculated that the void volume content for fabricated composites was a maximum of 3.5%. The tensile strength and modulus of untreated and treated yarns was investigated as was yarn toughness by measuring the area under the load-deflection curve. The tensile and flexural properties of epoxy composites were investigated. Jute fibre yarn treated with aqueous NaOH had a modulus 150% greater than untreated jute fibre yarn and a strength that was 120% higher. The alkali treatment caused a reduction in diameter of approximately 28%. Gassen and Bledzki, (1999) found that the treatment of jute yarns with different concentrations of NaOH and different shrinkages had significant effects on the fibre structure, mechanical properties and fracture behaviour. Fibre shrinkage influenced the pull-out length of cellulose fibrils. Epoxy composites reinforced with NaOH treated fibre yarn had higher strength and stiffness than untreated jute fibre yarn composites. However, Gassan and Bledzki, (1999) state, that the rougher surface of the treated fibre did not improve the adhesion between the fibre and matrix. The Young's modulus of untreated and treated jute fibre yarn composites increased in a linear fashion with fibre content. Untreated and treated jute fibre yarn reinforced unidirectional composites had a Young's modulus approximately 50% and 30% lower than comparable glass fibre epoxy composites.

Mohanty *et al.*, (2000) used jute fibres in the form of hessian cloth and carpet backing cloth to reinforce a thermoplastic biodegradable polyester resin (Biopol). Both jute fibre reinforcements were washed in a detergent solution and distilled water before being dried and fabricated into composites. Two layers of reinforcement material were sandwiched

between three layers of Biopol films. These five layers were heated under different pressures and temperatures to obtain the final composites. Mohanty *et al.*, (2000) used five different surface modifications on both washed fabrics in order to decrease moisture adsorption and increase the wettability of the fibres by the matrix polymer. The tensile, flexural and impact strength of these composites are reported. The effect of fibre content of hessian cloth reinforced Biopol composites was investigated. The hessian cloth used for reinforcement had been defatted with a mixture of alcohol and benzene. Composites with fibre weight fractions ranging from 10 to 42% were investigated. The tensile and impact strength of composites increased between 10 to 25% fibre weight fraction, but beyond this a decrease in tensile and impact strength was observed at a 42% fibre weight fraction. The composite's flexural strength increased up to fibre weight fraction of 15% and then remained constant. Mohanty *et al.*, (2000) postulate that the low values observed for composites mechanical properties at high fibre loadings may be because of the higher number of fibre ends that cause crack initiation and hence potential composite failure. Mohanty *et al.*, (2000a) used jute fabrics (hessian cloth) as reinforcement in biodegradable thermoplastic polyester amides. Because of the presence of hydroxyl and other polar groups on the fibre Mohanty *et al.*, (2000a) also attempted to improve the fibre to matrix interactions *via* alkali treatment and graft copolymerisation. Apart from the effects of surface modifications to the reinforcement fibre and composite mechanical properties Mohanty *et al.*, (2000a) does not discuss the effect that the reinforcements architecture has upon the composites performance.

Gassan, (2002) has studied the fatigue behaviour of natural fibre reinforced polymers of different composite parameters such as fibre type (jute or flax), the quality of the fibre and matrix adhesion, the fibre properties and their content. The specific damping capacity was used to monitor damage and to compare different types of composite. Flax and jute yarns and a woven jute fabric based on yarns with a Tex of 280 were treated by the following:

1. alkaline treatment, followed by a wash in distilled water to influence the fibres strength and modulus;

2. malaeic anhydride- polypropylene fibre treatment to improve the fibre to matrix (polypropylene based composites) adhesion;

3. and a silane treatment to improve the fibre matrix (unsaturated polyester resin) adhesion.

Flax and jute fibre yarns were embedded in epoxy or polyester resin to manufacture unidirectional composites. RTM was used to fabricated woven jute reinforced composites. In addition a film stacking technique was used to fabricate polypropylene based composites. Gassan, (2002) performed fatigue tests with step-wise loading increments, the test was done by holding a mean stress constant for a defined number of cycles and then the mean stress was increased for a defined number of cycles, this was repeated until composite failure occurred. Dynamic stress-strain curves were recorded continuously with the aid of an extensometer. Gassan, (2002) observed that damage initiated for jute reinforced unidirectional epoxy composites at approximately 45 MPa and it progressively got worse until failure at an applied maximum cyclic load of 120 MPa. The specific damping curve of flax epoxy composites was very similar to that of jute but with higher specific damping capacity values. Woven based epoxy composites had a critical (initiation of damage events) applied maximum cyclic load of approximately 20 MPa but obtained higher specific damping capacity values. Gassan, (2002) states that the woven based composites achieved higher specific damping capacity values because woven composites have a more significant viscoelastic character than unidirectional composites. Complete failure of the woven composites occurred relatively soon after the initiation of damage events at load of approximately 40 MPa. It was thought that the differences observed between flax and jute epoxy unidirectional composites behaviour could be caused by the influence their fibre structures have upon the stress-strain behaviour of the composite. As viscoelastic deformation occurs to the fibres, energy is absorbed by internal frictional heat as well as structural and cumulative fibre degradation. Gassan, (2002) states that when the interfacial bonding is weak in a woven composite, debonding and frictional sliding occurs readily upon crack extension, allowing fibres to remain undamaged and able to bridge cracks. Strong interfaces cause

fibre fracture to occur and thus no bridging of cracks because interfacial sliding has not occurred. Higher applied loads were observed for the onset of progressive damage for silane treated jute fibre yarn polyester composites. Matrix cracking and crack propagation were thought to be the main damage mechanisms in brittle composites and it was expected that the silane treated jute composites had a lower crack density for a given load. As the silane treated jute composites did not exhibit the mechanisms that absorb energy such as debonding and frictional sliding they had lower specific damping capacity values than untreated jute polyester composites.

Maffezzoli *et al.*, (2004) has used a general purpose polyester resin and a catalysed cardanol (phenolic based by product of cashew nut industry) based resin as matrices for jute fabric reinforced composites to fabricate flat plane samples. A thermosetting resin containing 40% of cardanol was obtained by adding an epoxy monomer and catalyst. Two different surface treatments were performed on the jute fabric reinforcement. Composite were fabricated containing untreated jute fabric, silane treated fabric and NaOH treated jute fabric in order to improve fibre to matrix wettability and adhesion and promote changes to the fibres chemical composition. It was found that the void content of untreated fibre reinforced cardanol based matrix composites was as high as 15%. Cardanol composites reinforced with NaOH treated jute fabric had void contents of 7% and composites reinforced with silane treated fibres had void contents of 4.3%. The void contents of polyester composites reinforced with untreated and NaOH treated jute fibres were 2.9% and 1.3% respectively. The tensile mechanical properties of jute fabric reinforced polyester and cardanol composites obtained by Maffezzoli *et al.*, (2004) are presented in Table 2.10.

Table 2.10 Tensile properties of jute fabric reinforced polyester and cardanol based matrix composites (Maffezzoli *et al.*, (2004).

Composite Type	*Fibre content (wt. %)*	*Tensile modulus (MPa)*	*Tensile strength (MPa)*	*Elongation to break (%)*
Polyester / untreated jute fabric	26	7324	35.5	1
Polyester / NaOH treated jute fabric	23	9598	35.24	0.8
Cardonal / untreated jute fabric	33	4891	20.6	2.9
Cardonal / NaOH treated jute fabric	26	2006	13.07	6.6
Cardonal / silane treated jute fabric	27	-	9.38	12.02

It is not known what direction Maffezzoli *et al.*, (2004) tested the jute fabric reinforced composites, however from Table 2.10 it is clear that an improvement to the tensile modulus of NaOH treated fibre reinforced polyester composites occurred. This is likely to be because of improved wetting of the fibres by the polymer, the removal of natural and artificial impurities on the fibres surface and an increased number of possible bonding sites leading to a better adhesion. Silane treated fabric affected the cure reaction of the cardonal based matrix, Maffezzoli *et al.*, (2004) state that the matrix turned into a rubbery material.

3 A PRELIMINARY INVESTIGATION INTO THE PHYSICAL AND MECHANICAL PROPERTIES OF WOVEN FLAX REINFORCED POLYMER MATRIX COMPOSITES

3.1 Introduction

The work presented in the following chapter was conducted to gain a fundamental understanding of the physical and mechanical properties of woven flax reinforced unsaturated polyester composites when fabricated on a lab scale. The materials were used in the supplied condition. The aim was to fabricate composites at various fibre volume loadings and by means of tensile, flexural and impact tests determine their mechanical properties and the factors that may influence them. The properties and behaviour of woven flax composites are compared to glass woven roving reinforced polyester composites throughout the following work. The woven glass reinforced composites were fabricated using a laboratory resin transfer moulding machine. Woven flax reinforced polyester composites were also fabricated at various fibre volume fractions using a laboratory resin transfer moulding machine, warp and weft orientated tensile, flexural and impact specimens were tested as composite panels were large enough to obtain a significant number of specimens for each test. Properties of woven flax reinforced polyester composites fabricated with a resin transfer moulding machine are not presented within the thesis.

3.2 Materials and method

3.2.1 Resin

Wresipol 31466, a pre-accelerated, slightly thixotropic (less viscous when worked), general purpose unsaturated polyester resin dissolved in styrene was obtained from Resinous Chemicals Ltd (Appendix 1 details all suppliers and contacts). The recommended catalyst 'Butanox M-50' obtained from Akzo Nobel Chemicals Ltd. was used for curing. The catalyst consisted of 33% methyl ethyl ketone peroxide in dimethyl phthalate. Wresipol 31466 was a suitable resin for a hand lay-up process due to its long gel time and fast cure characteristics. Table 3.1 details the physical and mechanical properties of Wresipol 31466 as determined by the supplier.

Table 3.1 Physical and mechanical properties of Wresipol 31466 unsaturated polyester resin (*Source*: Resinous Chemicals Ltd.).

Property	*Wresipol 31466*
Liquid resin properties	
Typical viscosity @ 25°C (poise)	2.25
Acid value (mg KOH/g)	40
Application time (minutes)	25-30
Fully cured resin properties	
Tensile strength (MPa)	24.4 (3.82)
Tensile modulus (MPa)	3732.8 (265.08)
Load to failure (N)	1080.4 (172.57)
Strain to Break (%)	0.72 (0.13)
Density (kg m^{-3})	1180

Note: figures in parentheses are standard deviations.

3.2.2 Woven flax

Flax woven linen (100%) as shown in Plate 3.1 was obtained from Harry Gilbertson. Unfortunately information regarding the processing of the fabric was unattainable, including details of the yarn Tex from which it was woven. However the fabric appeared to have been bleached and through communication with Harry Gilbertson it was established that a sizing agent had also been used on the yarns. The type of size was unknown although it was believed to contain fatty acids. The woven flax was a fine plain weave ($^{1}/_{1}$) and both warp and weft yarns were identical in size. The fabric count (the number of warp and weft yarns per inch of fabric) was on average 41 warp and 41 weft yarns (41×41).

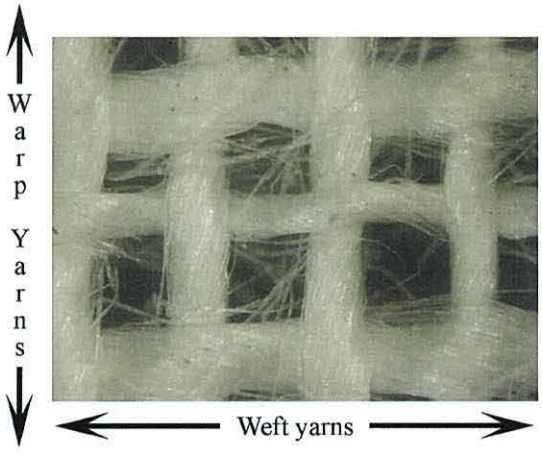


Plate 3.1 Plain weave woven flax fabric.

3.2.2.1 Evaluation of tensile properties of woven flax fabric

Tensile properties of the fabric were calculated by determining the maximum force and elongation at maximum force using the strip method in accordance with BS EN ISO 13934-1:1999. Five specimens 200 mm in length and 25 mm wide were cut from the fabric in both warp and weft directions, and the number of yarn ends (yarns in test direction) was counted. Specimens were not creased and were cut from areas

representative of the whole material. Specimens did not contain the same yarns as other specimens in that particular orientation, whether warp or weft, as a cutting pattern was used to prevent this occurring. All specimens were conditioned to 20°C at 65% relative humidity. Specimens were tested dry at the condition mentioned above on a computer controlled Instron (model 1195) using Merlin software. They were gripped between smooth surfaced self-tightening jaws measuring 25 mm in width and 50 mm in length, thus leaving a sample gauge length of 100 mm. A 1.5 N pretension was applied before the test started at a rate of elongation of 20 mm min^{-1}. The extension of the crosshead and the load recorded from the load-cell was acquired digitally at a frequency of ten data points per second.

3.2.3 Woven glass roving

A standard E-glass woven roving as shown in Plate 3.2 was utilised for the fabrication of glass reinforced composites. The weft fibres lay straight with warp fibres crimping between them, as with the woven flax fabric. Individual continuous glass fibres were combined to form strands or bundles as they were not twisted into yarns. In a warp or weft strand there were approximately 700 to 1000 individual fibres. The stands of fibres were woven into a $^{1}/_{1}$ plain weave type. The fabric count (the number of warp and weft strands per inch of fabric) was on average 8 warp and 8 weft yarns (8×8).

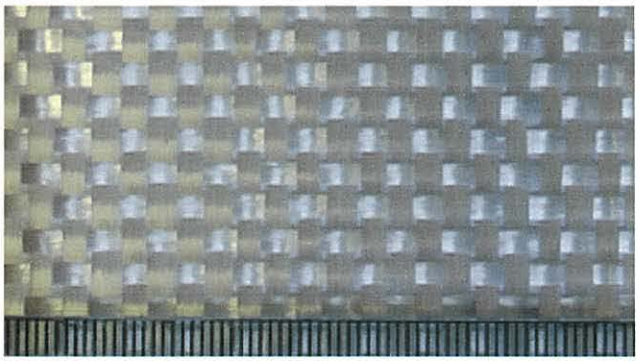


Plate 3.2 E-glass woven roving.

3.2.3.1 Determination of the density of woven glass roving

The density of the woven glass used was determined using an AccuPyc 1330 helium Pycnometer from the Micromeritics Instrument Corporation. A known weight of oven dried glass fibre was placed into a calibrated chamber. The instrument filled the chamber with helium gas at a pressure of 1.26 kgf cm^{-2} ten times, giving an average volume and density of the material tested at a temperature of 24.4°C.

3.2.4 Woven flax composite manufacture

In total, nine flat panelled composites were fabricated measuring approximately 30 cm square with varying thickness, ranging from 2.5 to 4.5 mm. The above dimensions were found to be adequate for obtaining suitable numbers of specimens for testing. The preliminary study into the effect of fibre volume fraction on mechanical properties required composites fabricated with various fibre loadings. A single ply of the woven flax reinforcement used was approximately 0.4 mm thick and different volume fractions were achieved by fabricating composites consisting of multiple woven plies, ranging from 7 to 16 ply. To achieve higher fibre volume fractions, some stacks of woven reinforcement were pressed before composite fabrication in order to consolidate the material, thus reducing the reinforcement's volume. Using a 'Schubert' hot press, stacked plies of woven fabric were pressed at 90°C at approximately 203 kgf cm^{-2} for 1 minute. For all nine composites fabricated the woven reinforcement forming each ply was at the same orientation ($[0°]_S$) and testing was conducted with this in mind. Stacked woven reinforcement for each composite was weighed on an electronic balance.

3.2.4.1 Resin preparation

The required quantity of resin for each composite was mixed with 1% (by weight) of catalyst with a mechanical stirrer for at least 5 minutes. After stirring the catalysed liquid resin was then degassed within a desiccator for at least 5 minutes before use.

3.2.4.2 Resin impregnation into woven reinforcement

Impregnation was achieved within an open plastic bag by drawing catalysed resin over a stack of woven reinforcement using a vacuum. This technique kept styrene emissions to a minimum within the laboratory. Figure 3.1 shows a schematic drawing of the process. The 30 cm square multi plies of woven reinforcement were inserted into a heavy-duty polythene bag. Adjacent to the woven reinforcement, a 30 cm wide 'waste mat' (non-woven jute felt) was inserted and the end of the plastic bag was sealed with tape. The width of the polythene bag often would be greater than 30 cm and in such cases it was rolled back to the edge of the woven reinforcement and secured with 'bulldog clips', thus preventing resin from bypassing the reinforcement and collecting on the waste mat. Bulldog clips were used to seal the polythene bag across its width near to the woven reinforcement which provided a bag at the end of the tube. Catalysed resin was poured into the formed bag and it was sealed with tape and the entire bag laid flat. A vacuum was drawn though a small cut on the surface of the polythene bag above the centre of the waste mat with an 'Edwards' vacuum pump. A suction head was used to cover the cut on the polythene tube and the waste mat prevented the polythene tube from collapsing under vacuum. The vacuum pulled helped impregnate the stack of woven reinforcement with catalysed resin once the bulldog clips across the tube's width were removed. To ensure complete impregnation of the reinforcement a hand roller was used over the surface of the polythene tube. After complete saturation the vacuum was removed and the polythene tube cut open and the impregnated stack of woven plies taken out for moulding.

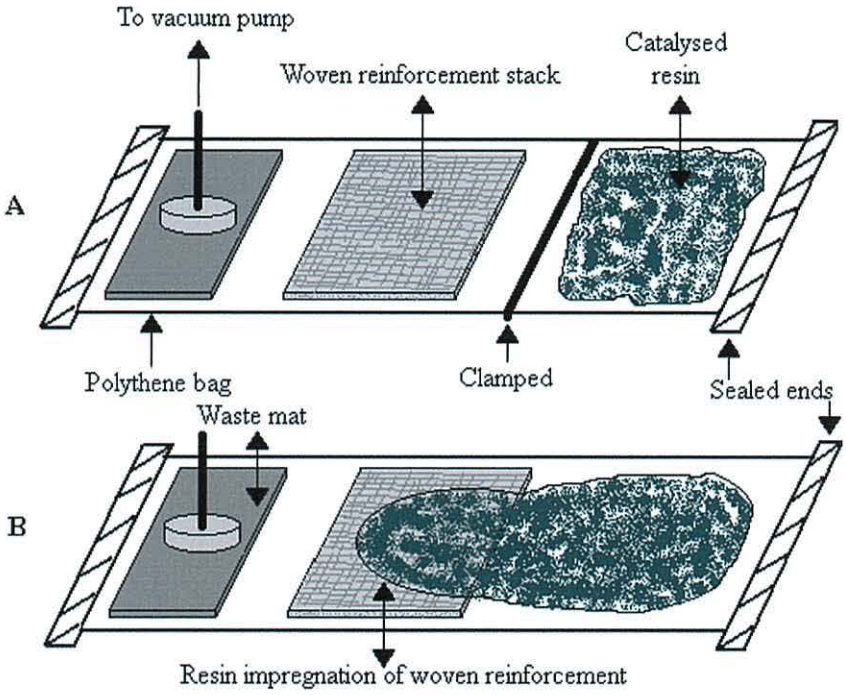


Figure 3.1 Schematic representation of the vacuum impregnation process.

The woven flax composites could have been fabricated using an resin transfer machine (RTM) process. The RTM process was not utilised to fabricate the woven flax composites because of the lack woven flax reinforcement available.

3.2.4.3 Moulding and Curing

Toughened glass plates (400 mm^2 by 15 mm) were coated with a release agent (Paste release wax No 1, Ambersil Ltd.) in accordance with manufacturer's instructions prior to use. Impregnated stacks of woven reinforcement were placed flat between the toughened glass sheets. G-clamps were then used at each corner of the two glass sheets to maintain a slight pressure on the saturated stack of reinforcement. Each G-clamp was tightened to approximately the same amount. Composites were left between the glass sheets for 24 hours at approximately 20°C to cure. After release from the mould, the composites

underwent a post cure in an oven set at 50°C for 5 hours. The composites were not pressed to stops as it had been noticed in earlier attempts of moulding that there was high variation with composite thickness when pressing to stops. This was because the thick glass plates actually flexed, causing the central regions of the composite to be thicker than the edges of the composite, especially areas near the stops *e.g.* composite panel corners. A low variation in thickness was measured for all the composites fabricated and moulded by the experimental method outlined above *e.g.* moulding between glass plates with a low amount of pressure applied.

3.2.5 Preparation of unreinforced resin panels

Modelling clay ('Plasticine') was placed around the edges of a glass sheet (400 mm^2 by 15 mm) forming a barrier about 5 mm in height. Cast resin panels were prepared by pouring catalysed resin onto the glass sheet until flush with the lowest part of the Plasticine barrier. A second glass sheet was then placed over the liquid resin and slightly pushed down evenly into the Plasticine. Section 3.2.4.3 on page 113 details the curing regime applied.

3.2.6 Woven glass composite manufacture

Four woven glass composites were fabricated measuring 300 mm in length and 250 mm in width with varying thickness, ranging from 4.1 to 4.3 mm. Composites were fabricated containing 4, 6, 10 and 12 plies to achieve different fibre volume fractions. All plies of woven reinforcement were stacked in the same direction ($[0°]_S$). Stacked woven reinforcement was weighed on an electronic balance.

3.2.6.1 Fabrication of woven glass reinforced polyester composites

Composites were fabricated using a laboratory Hypaject MK II model (from Plastech, U.K.) RTM that was connected to a 4 by 300 by 500 mm flat panelled mould. Prior to composite manufacture a mould sealer and releasing agent (Frekote mould sealer and Frekote releasing interface, The Dexter Corporation, U.S.A.) was applied according to manufacturer's specifications to the surfaces of both female and male moulds. Two composites were made at the same time as the stacks of woven glass plies only filled half the area of the mould. Rolls of Plasticine were placed around the edges of the mould as previous preliminary experimental work had found that resin would by-pass the reinforcement by flowing through the gaps created at the edge to the vacuum outlet. After mould closure, the required quantity of resin for each composite was mixed with 1% of catalyst with a mechanical stirrer for at least 5 minutes. Resin was then pulled using a vacuum into a 'homogeniser'. The homogeniser further mixed the resin and de-gassed it for approximately 8 minutes before being pressurised for injection (2.03 kgf cm^{-2}). A vacuum was allowed to build to between –0.71 to –0.91 kgf cm^{-2} within the mould prior to resin injection. Resin injection took between 10 to 15 minutes but was controlled so that the resin travelled evenly through the mould. After complete impregnation, the wetted reinforcement was left in the mould to cure at room temperature for 24 hours. Composites underwent a post cure within the same mould. The electric heated mould was set to a temperature of 50°C for 5 hours before opening the mould for composite release. The two composites fabricated were joined together by a section of pure cast polyester resin which was trimmed away using a tile cutter saw and the rolls of Plasticine were also removed from composite edges before weighing.

3.2.7 Measurement of composites

The dimensions and weight of each composite panel and cast resin panel were measured to the accuracy of ±0.1 mm apart from the thickness that was measured at ±0.01 mm.

The thickness of each specimen cut from a composite or resin panel was recorded and the composites thickness was taken to be the mean average from these measurements.

3.2.8 Specimen preparation

Three specimen sizes were required in order to perform the mechanical tests included in the following work. Specimens for all types of testing were produced in an identical manner. Specimens from flax and glass woven reinforced composites and the unreinforced unsaturated polyester resin panels were cut using a 'tile cutter'. It was found that a water lubricated diamond saw gave an excellent surface finish and allowed precise cutting to be achieved. Composites were exposed to water for short periods of time. This was kept to a minimum and specimens and composites were immediately dried using paper towels. Approximately 1 cm of material was trimmed from every composite edge before the cutting of specimens commenced. All specimens from flax and glass woven reinforced composites were cut in the same direction (warp (0°)).

3.2.8.1 Conditioning

Specimens were conditioned prior to testing at 65% relative humidity at a constant temperature of 20°C in a climate controlled room for a minimum of one week.

3.2.8.2 Measurement of specimens

After conditioning, the specimen's weight, length, width and thickness were measured to the accuracy of two decimal places (±0.01) using electronic callipers and balance in the climate conditioned room. The weight and length of specimens were measured once but

the thickness and width of specimens were measured three times and a mean average taken. Specimen's width and thickness were measured at the centre and approximately 1 cm from either end.

3.2.9 Testing

Woven flax and glass reinforced polyester composites are anisotropic but for the preliminary investigation composites were tested in only in one direction. In order to obtain reasonable numbers of specimens for each test performed it was decided that only one direction should be tested. All samples being used for flexural, tensile or impact tests were cut and tested in the warp direction (*i.e.* the warp yarns parallel to the length of the specimens).

3.2.9.1 Flexural testing

All flexural specimens (cast resin and woven reinforced composites) measuring 100 mm in length, 15 mm in width and with a thickness of less than 10 mm were tested in accordance with BS 2782: Part 10: Method 1005: 1977 (EN 63) – Determination of Flexural Properties. Three Point Method. The flexural specimens tested had varying thicknesses ranging from 2.5 to 4.5 mm. Testing was conducted on a computer controlled Instron (model 4301), universal testing machine fitted with a 5 KN load cell. A crosshead speed of 10 mm min^{-1} was utilised and load and extension data acquired digitally at a frequency of 10 per second using 'Series 9®' software.

3.2.9.2 Tensile testing

Tensile specimens were tested in accordance with BS 2782: Part 10: Method 1003: 1997. (EN 61). A type II (rectangular) specimen style was utilised but with modified dimensions. The standard requires that the length of specimens is at least 250 mm. Constrictions such as the size of the composites produced led to a reduction of the specimen length to 200 mm with a width of 20 mm, thus keeping the same aspect ratio. A further modification to the standard concerning the clamping of the specimen was also implemented. The standard requires that specimens be attached to the test machine *via* pin-jointed ends. Utilising pin jointed ends would have eliminated any alignment complications that may have occurred during testing. However, flax specimens were fitted with removable aluminium end tags and clamped using Instron self-tightening jaws. Aluminium end tags and specimens were slightly abraded to prevent slippage occurring during testing. Aluminium end tags were glued onto the surfaces of woven glass reinforced composites as it was found that the composite's smoother surfaces caused slippage during testing. The protected specimens were placed into the jaws and every care was taken to ensure that specimens were aligned before starting. Before testing, an Instron extensometer (for measuring strain) was fitted with clips to the central portion of the specimens within the gauge length (100 mm). The strain measured by the extensometer was used for all stress-strain curves presented herein unless stated otherwise. A computer controlled Instron (model 1195) universal testing machine fitted with a 100 KN load cell was used at a crosshead speed of 10 mm min^{-1} for woven reinforced specimens but for cast resin specimens a slower rate of 2 mm min^{-1} was implemented. Digitally acquired data from the extensometer, load cell and crosshead of the Instron was collected at a frequency of 20 data points per second using 'Merlin®' software.

3.2.9.3 Impact testing

The flat-wise impact properties (direction of blow parallel to the thickness of the specimen with impact on the broad longitudinal surface) of unnotched type 1 specimens were examined using an analogue Zwick 5102 Pendulum impact tester. Testing was conducted in accordance with BS EN ISO 179: 1993 Plastics– Determination of Charpy Impact Strength. Specimens were 80 mm in length and 10 mm in width. A testing span of 62 mm was implemented and a 4 J pendulum was used.

3.2.10 Examination of fractured specimens

The modes of fracture of the specimens were observed visually and through a dissecting microscope. Fractured impact specimens were examined using scanning electron microscopy. Fractured surfaces were cut away from the specimen using a fine toothed band saw. This left the fractured surface with approximately 5 mm of composite material. The flat cut composite material was then secured to aluminium stubs with conducting epoxy adhesive, leaving the fractured surface exposed. The samples were dried in an oven set at 100°C for a few hours before being placed over silica gel for 24 hours. The samples were splutter coated using a Polaron E5000 set to 1.2kV and 10mA. The samples were coated in gold from a pure gold target for 2.5 minutes. A Hitachi S-520 scanning electron microscope (SEM) was set to 12kV and used at various magnifications to record the fractures.

3.2.11 Evaluation of physical properties

3.2.11.1 Measurement of density

Densities of cast resin panels; woven and pre-pressed woven flax reinforced composites were calculated using Equation 3.1.

Equation 3.1 $$\rho_c = \frac{M_c}{V_c}$$

Where: ρ_c The composite density.

Densities from each specimen prepared from the panels or composites were also calculated using the Equation 3.1. The volume of the composite was calculated using the average measurements of the exterior dimensions.

3.2.11.2 Measurement of volume fraction

The volume fractions of the matrix and reinforcement were calculated using both Equation 2.11 on page 62 and Equation 2.12 on page 62. However a slightly modified form of Equation 2.12 was utilised to calculate the volume fractions presented in all figures and tables throughout the work (unless stated) for reasons that are discussed in Section 3.3.4.3 on page 126.

3.3 Results and discussion

3.3.1 Tensile properties of woven flax fabrics

The tensile properties of woven flax fabric tested in both warp and weft directions are presented in Table 3.2.

Table 3.2 Warp and weft tensile properties of plain weaved woven flax fabric.

Test direction	*Mean number of yarns in tensile specimens*	*Mean load at failure (N)*	*Mean tensile elongation at maximum load (mm)*
Warp	38.4 (1.52)	284.5 (55.16)	15.4 (0.54)
Weft	38.4 (0.89)	360.1 (21.44)	5.2 (0.14)

Note: figures in parentheses are standard deviations.

Tensile properties of the woven flax were greatest in the weft direction and specimens tested in this orientation displayed less variance than warp tested specimens. This is likely to be due to the fact that weft yarns lie straighter within the fabric as apposed to the warp yarns that are crimped to a higher degree.

Crimping also explains why warp specimens displayed higher tensile extension values. Figure 3.2 shows the tensile load of two representative specimens from each test direction as a function of tensile extension. In both load-extension curves there is an initial period of extension in which the load increases slowly with tensile elongation. During this extension the yarns are realigning to the direction of the load by attempting to lie straight and parallel to it. As Figure 3.2 graphically shows there is a considerable difference

between warp and weft yarns during this initial period of tensile extension due to crimping.

After the slack of the yarns has been pulled taut and they have realigned, the gradient of the load-extension curve becomes greater. It can be seen from Figure 3.2 that the weft specimen's curve is sharper than the warp specimen. It is believed that the weft yarns in the warp test specimens inhibit the performance because they prevent the warp yarns becoming fully aligned to the load.

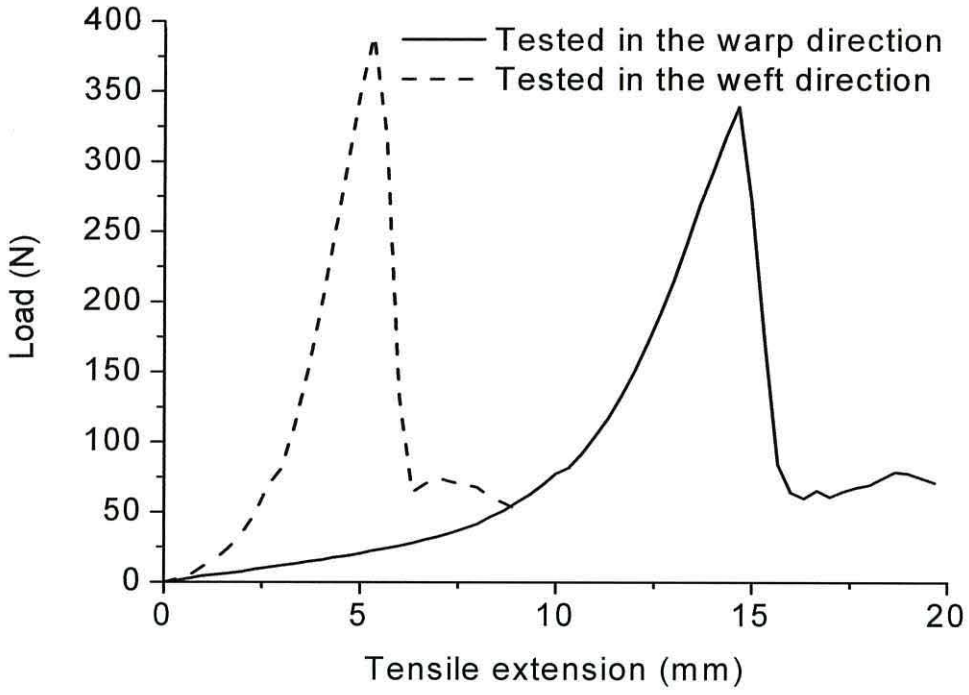


Figure 3.2 The tensile load of two representative woven flax fabric specimens as a function of tensile extension.

3.3.2 Density of woven glass roving

The density of woven glass fibres (as determined by helium pycnometry) was found to be 2582 kg m^{-3}. The standard deviation of the test results was low (2.6 kg m^{-3}) indicating it was a uniform material. The density of E-glass published by Hull and Clyne (1996) and Ivens *et al.*, (1997) is 2560 kg m^{-3}; therefore the value obtained is in agreement with published literature.

3.3.3 Cast polyester properties

The mechanical and physical properties of cast Wresipol 31466 polyester resin are displayed in Table 3.3.

Table 3.3 Physical and mechanical properties of post cured cast resin (Wresipol 31466)

Property	*Number of specimens tested*	*Mean average result*
Density (kg m^{-3})	24	1179.8 (11.93)
Flexural properties		
Flexural stress (MPa)	8	68.6 (22.27)
Flexural modulus (MPa)	8	4108.8 (211.86)
Strain to failure (%)	8	1.67 (0.49)
Tensile properties		
Tensile stress (MPa)	6	31.3 (4.37)
Tensile Young's modulus (GPa)	6	4.7 (0.13)
Strain to failure (%)	6	0.68 (0.12)
Impact properties		
Charpy impact strength (kJ m^{-2})	10	4.9 (1.87)

Note: figures in parentheses are standard deviations.

The cast resin density value shown in Table 3.3 is extremely close to the resin manufacturer's value of 1180 kg m^{-3} given in Table 3.1. Therefore, this value has been used in equations requiring the density of cured resin. The tensile strength and modulus were found to be considerably higher than the manufacturer's reported tensile properties but the strain at break is similar especially when standard deviations are taken into account. A representative stress-stain curve of a tensile tested cast resin specimen can be seen in Figure 3.3. A possible explanation for the differences between the tensile properties may be due to the use of different rates of extension during testing. It is worthwhile mentioning that tensile specimens failed in a brittle manner leaving a clean fractured surface.

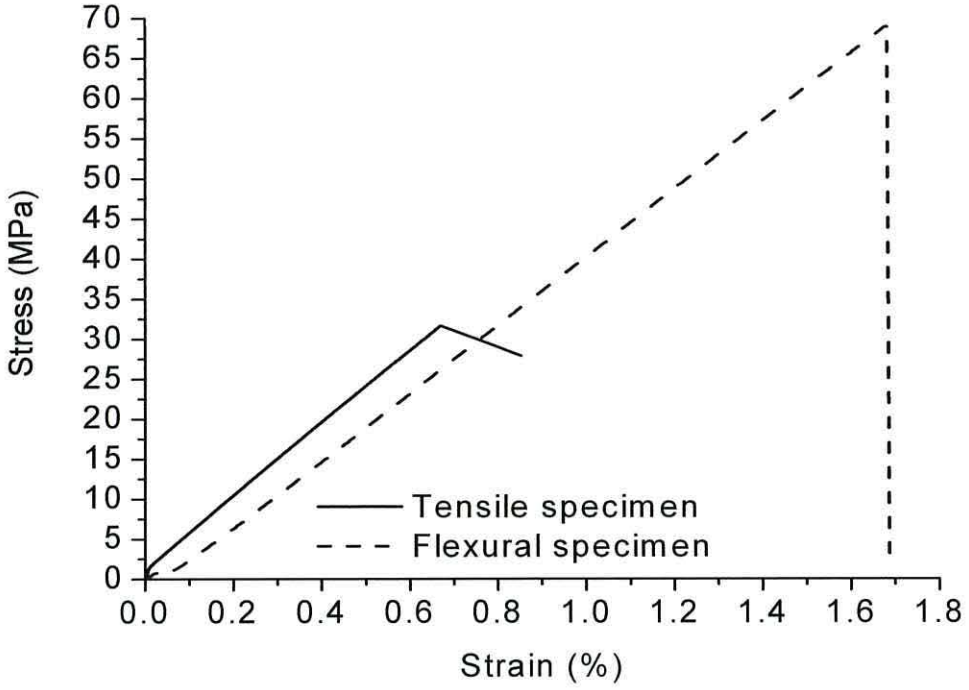


Figure 3.3 Flexural and tensile stress-strain behaviour of representative cast polyester resin specimens.

The crack initiating the catastrophic failure always started on the edge/surface of the specimen and the cause was often a surface defect such as a small indentation. Other properties presented in Table 3.3 seem to be reasonable values and are thought to be representative properties of the cast resin. A representative flexural stress-strain curve is presented in Figure 3.3. Flexural specimens also failed in a brittle manner. All flexural and tensile specimens showed practically linear stress-strain behaviour up to the strain at which they fractured.

3.3.4 Physical properties of the composites

3.3.4.1 Appearance of woven flax composites

Due to the light colour of the woven reinforcement (Plate 3.1 on page 109), the colour of the composites was a light yellowish brown. The surfaces of all the composites were smooth due to the glass sheets on which they were moulded. The surfaces appeared defect free but infrequent small indentations (less then 1 mm diameter) were visible when held at certain angles. It is thought that gaseous by-products released during curing may have created the indentations on the surfaces.

3.3.4.2 Appearance of woven glass composites

Woven glass reinforced polyester composites were all translucent with a slight yellowish colour. The glass fibre reinforcement was visible when composites were held at certain angles. The surfaces were smooth and defect free.

3.3.4.3 Calculation of fibre volume fraction

As previously discussed in Section 2.7.3.1 on page 61, the proportions of the constituents within a composite, expressed as volume fractions, are extremely important factors affecting composite properties. Also reported in Section 2.7.3.1 is the work conducted by Hughes (2000), namely that the fibre volume fractions of non-woven jute and hemp polyester composites calculated with Equation 2.11 $V_f = \frac{M_f}{V_c \rho_f}$ (assuming a bulk fibre density of 1500 kg m^{-3}) were (as expected) lower than fibre volume fractions calculated with Equation 2.12 ($V_f = \frac{V_c - \frac{(M_c - M_f)}{\rho_r}}{V_c}$). Since the differences were less than 2%, Hughes (2000) concluded that void space in the samples he examined was minimal and that the result was within the range of experimental error. Hughes (2000) used Equation 2.11 with a fibre bulk density of 1500 kg m^{-3} as this value is close to the density of plant cell wall material. Ivens *et al.,* (1997) state that a plant fibre's bulk density cannot exceed the density of cellulose (major cell wall constituent), 1540 kg m^{-3}, due to the cell structure and the presence of micro-voids. Bulk densities of plant fibres are thought to lie in the region of 600 to 1200 kg m^{-3} (Bolton, 1994). Assuming that the bulk fibre density is equal to the cell wall material density then a lower limiting fibre volume fraction can be established using Equation 2.11. This condition assumes that the fibres contain no lumen space, or, they are completely filled with resin and that there are no other voids present in the composite structure. Table 3.4 shows fibre volume fractions calculated with Equation 2.11 (column '*A*') which is a lower limiting fibre volume fraction and Equation 2.12, which is a higher estimate for fibre volume fraction (column '*B*') and the percentage differences between values. Equation 2.11 assumes that the fibre bulk density is 1500 kg m^{-3} for flax reinforced composites, a value that is close to the cell wall material density but not as high as the density of cellulose alone stated by Ivens *et al.,* (1997). The density of 2582 kg m^{-3} has been used within Equation 2.11 to calculate fibre volume fractions of glass fibre composites. Equation 2.12 assumes a certain degree of

fibre porosity and allows for the fact that resin may enter the flax fibre lumens during fabrication.

As expected, the fibre volume fractions when calculated with Equation 2.12 were higher than fibre volume fractions calculated with Equation 2.11. The percentage differences between the two methods vary substantially. Generally, composites fabricated with pre-pressed woven flax reinforcement exhibited the highest differences. As the differences are high, it can be assumed that voids are present within the composite system. An estimated void volume fraction (V_{vf}) can be obtained using Equation 3.2.

Equation 3.2

$$V_{vf} = \left[\frac{V_c - \left(\frac{M_c - M_f}{\rho_r} \right) - \left(\frac{M_f}{\rho_f} \right)}{V_c} \right] \times 100$$

Where: M_f The mass of fibre (woven reinforcement).

ρ_f The bulk density of fibre (woven reinforcement).

The void volume fraction obtained with Equation 3.2 produces a high estimate for the flax reinforced composites because of the need to assume a value for the bulk density of the fibre. As densities of fibre or yarns are not available, the upper value of 1500 kg m^{-3} has been substituted and the results are shown in Table 3.4. If a lower value for fibre bulk density is substituted, then it has the effect of reducing the void volume fraction. Figure 3.4 shows a plot of fibre volume fractions calculated using Equation 2.12 against estimated void volume fraction for flax and glass reinforced composites. As fibre volume fraction increases, the calculated amount of voids also increases in a linear fashion for all composite systems. Although it seems likely that the woven flax reinforced polyester composites fabricated contain voids, their location and true quantity remains an unknown.

Table 3.4 Calculated fibre volume fractions and void volume fractions of composites.

No. woven flax ply	*A.* V_f *(%)*	*B.* V_f *(%)*	*% Diff. between A and B*	*Estimated* V_{vf} *(%)*
Flax reinforced				
7	32.1	34.1	6.2	2.04
8	27.2	29.4	8.0	2.18
10*	51.8	68.5	32.2	16.70
10	28.7	31.1	8.3	2.40
12	29.2	30.1	3.0	0.90
12*	34.5	36.8	6.6	2.39
14	34.3	35.8	4.3	1.58
14*	39.1	46.8	19.6	7.68
16*	42.4	46.7	10.1	4.34
Glass reinforced				
4	13.0	13.2	1.5	0.22
6	19.4	20.4	5.1	0.99
10	32.9	35.7	8.5	2.79
12	38.7	44.1	13.9	5.35

*Woven reinforcement pre-pressed before composite fabrication.

It has already been noted that small indentations are visible on the composite's surfaces. However they are not frequent enough to account for the high void volume fraction calculated in some of the composites. The flax reinforced composites exhibiting high void volume fractions are those where the woven flax reinforcement was pre-pressed prior to fabrication. Voids may have occurred throughout these composites because of insufficient wetting of the reinforcement. As described in Section 2.7.2 on page 57, wetting occurs spontaneously when the surface energy of the solid-vapour interface is greater than the liquid-vapour interface. According to Hull and Clyne (1996) the surface

energy of a standard liquid polyester resin (γ_{LV}) is 35 mJ m^{-2}. The surface energies, measured by inverse gas chromatography (IGC) of green and dew retted flax fibres were 50.25 and 43.78 mJ m^{-2} respectively (Zafeiropoulos *et al.*, 2002). Cantero *et al.*, (no date) found the surface energy of untreated retted flax to be 44 mJ m^{-2}. It is likely that the woven flax used in this work would exhibit a different surface energy value than the literature ones cited above. This is because the woven flax has been bleached and contains a size that is hydrophobic which would have the effect of lowering the surface energy. It is probable that the surface energy is still greater than the liquid resin's surface energy but not as high as the published literature values. At this stage it is unknown how well the flax fibres were wetted by the liquid resin. It is worthwhile mentioning however that the surface energy of glass is of the order of 560 mJ m^{-2} (Hull and Clyne, 1996).

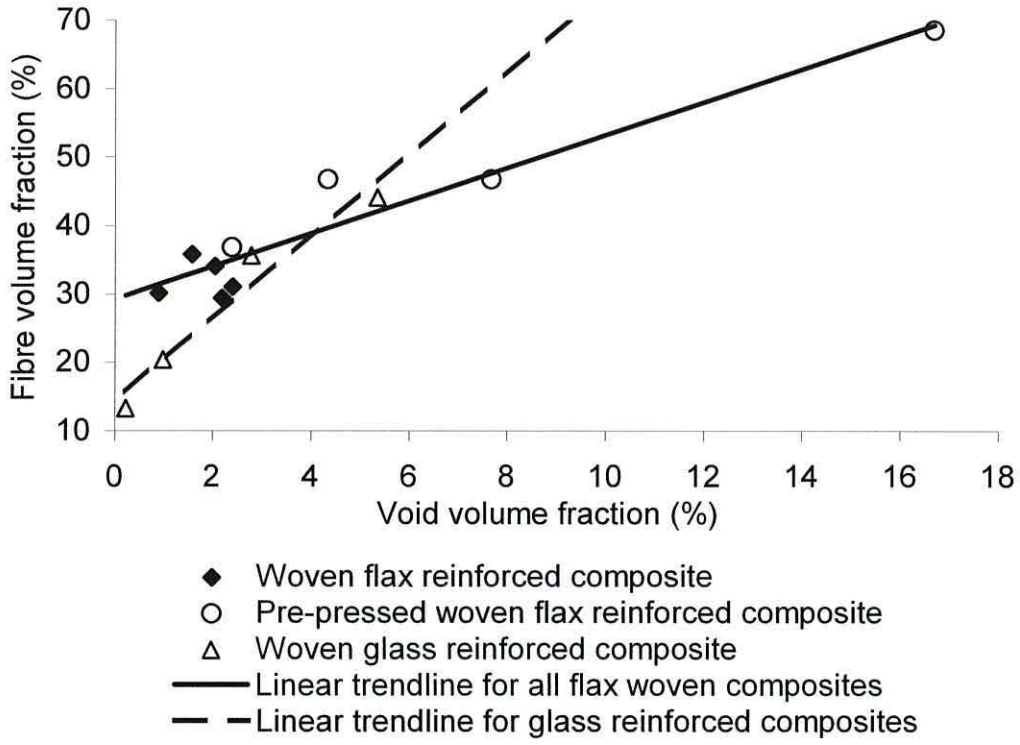


Figure 3.4 Void volume fraction of composites against fibre volume fraction.

If it is assumed that good wetting of the fibre surface occurred, then the high void volume fraction could be explained by lack of resin penetration into the lumen. Resin may penetrate into the lumens of fibres *via* cut ends, pits (natural apertures along cells) or through fractures. The polyester resin used had a low viscosity and it is possible that some resin penetration could occur. The sizing agent present on the fibres surface may have inhibited resin flow into fibres, thus leaving voids within. Also fibres themselves may have physically inhibited the flow of resin throughout the composite structure. The twisted flax fibres producing the yarns are tightly spun, thus not allowing good resin penetration into the middle regions of yarns. As plies of woven reinforcement were laid in the same direction and some were also hot pressed for further consolidation many yarn to yarn contacts would have been created along with resin rich areas such as the space between adjacent warp and weft yarns. Composites fabricated by Hughes (2000) may have had low void contents (less than 2%) because of the open nature of the non-woven reinforcement structure used that allowed for good penetration between fibres. Although woven fabrics appear to be very open structures because of the gaps between yarns, fibres within the yarns may be twisted to the extent they are acting as a barrier to the liquid catalysed resin during fabrication.

It is also worth mentioning that voids within the composites fabricated may have been created during curing. The curing of a polyester resin causes it to undergo a volumetric shrinkage that can be in the order of 4 to 8% (Matthews and Rawlings, 1993). Stresses, arising from the shrinkage may cause the matrix to debond from fibres (providing a weak interface exists), and allow cracks to form within resin rich areas (matrix pockets). It is likely that the yarns and fibres themselves also underwent a volumetric shrinkage due to moisture being driven from them by the elevated post cure temperature (50°C) or from the increase in temperature from the exothermic reaction of curing.

The moisture loss from the woven flax reinforcement will not only cause shrinkage but also some weight loss. A weight loss will have implications regarding Equation 2.12, as it requires M_f to be subtracted from M_c and in Equation 2.11 as it requires M_f to be divided by the volume of the composite multiplied with the density of the cured resin.

Woven flax reinforcement used was weighed before fabrication, and the composite's weight was recorded after the post cure procedure. The mass of the fibre used in the equations will be slightly higher than the actual mass of fibre within the composite after post cure; therefore, the resulting fibre volume fractions from Equation 2.11 and Equation 2.12 are likely to be higher estimates of fibre volume fraction. The moisture content of woven flax fabric before fabrication was 5.53%. The moisture content value is high enough to cause significant differences to the fibre volume fraction of a composite when calculated with Equation 2.11 or Equation 2.12.

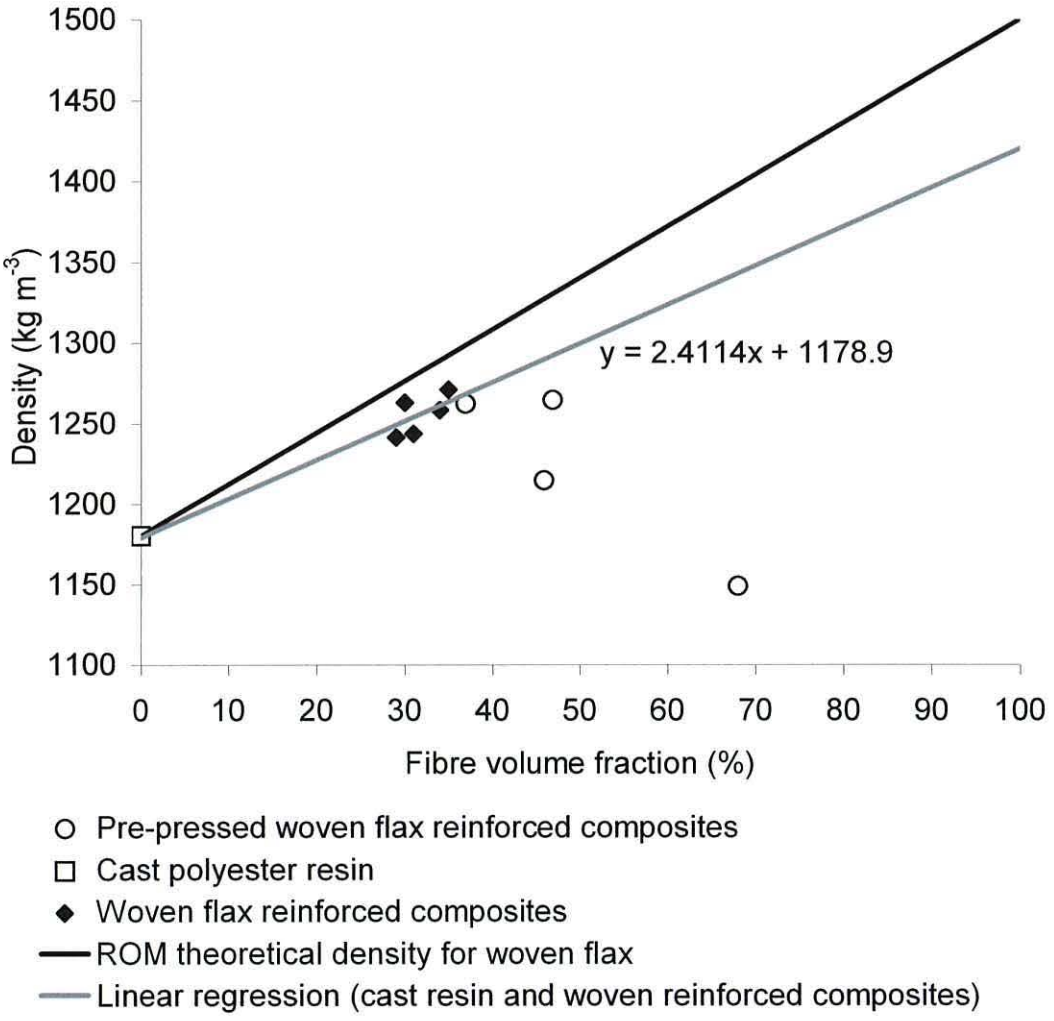


Figure 3.5 Woven flax composite density as a function of fibre volume fraction (Equation 2.11).

The densities of woven flax and pre-pressed woven flax reinforced polyester composites are shown in Figure 3.5 as a function of fibre volume fraction calculated with Equation 2.12. Figure 3.6 presents the densities of glass woven reinforced polyester composites.

Also presented in Figure 3.5 is a theoretical prediction of composite density produced from the ROM relationship using an assumed fibre density of 1500 kg m^{-3} (Equation 2.13). At all fibre volume fractions studied, the determined composite densities using Equation 3.1 are lower than the theoretical prediction from the ROM. Figure 3.5 also shows a linear regression for the data of woven reinforced composites starting from the calculated density of cast resin. Extrapolation of this regression line to the fibre volume fraction of 100% gives a theoretical fibre density of 1420 kg m^{-3}.

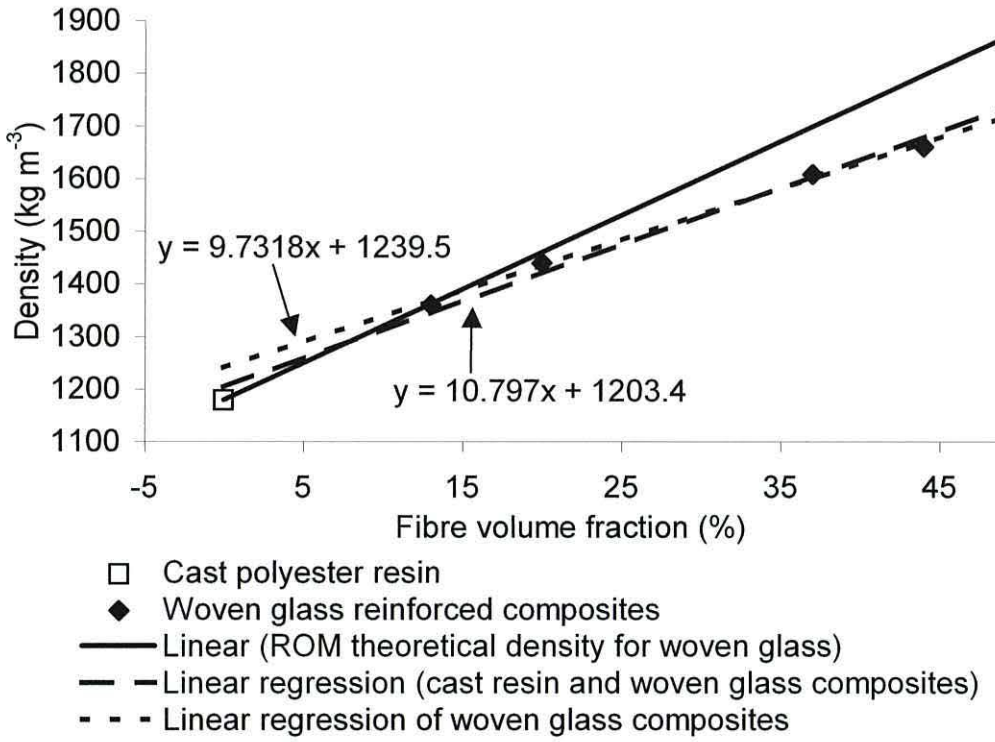


Figure 3.6 Woven glass composite density as a function of fibre volume fraction (Equation 2.12).

The regression equation is shown on Figure 3.5. The theoretical fibre density formed from the extrapolation is a realistic value even though the extrapolated data does not cover a wide range of fibre volume fractions. If the density value of 1420 kg m^{-3} had been utilised in Equation 2.13 then the theoretical prediction of composite density (ROM) would have travelled through the data and shown that there were few voids within the flax woven reinforced composites. This would agree with Figure 3.4 on page 129 that shows that the void content of woven flax composites is less than 2.4%. Hughes (2000) found that non-woven hemp reinforced polyester composites appeared to have lower void contents at higher fibre volume fractions. A possible explanation for the finding was the method used to obtain composites with high fibre volume fractions. Hughes (2000) hot pressed the non-woven reinforcement prior to the composites fabrication, thus possibly collapsing the cell walls of fibres and reducing lumen space. Damage induced to the fibres by pressing may have helped achieve better access for resin to fill any lumens or other voids left.

However this is not the case for the woven flax composites, as the reinforcement was not hot pressed. In review of the findings reported in Figure 3.4 on page 129 earlier in this section, composites that were fabricated with woven reinforcement that have not been subjected to a pre-press treatment displayed estimated void volume fractions considerably lower than composites made with pre-pressed woven reinforcement (Table 3.4 on page 128). Composites fabricated with woven flax may have been wetted well by the resin, thus having smaller void volume fractions than pre-pressed woven flax composites. It is clear from the results shown in Figure 3.5 on page 131 that the densities of pre-pressed woven flax composites do not follow the predicted ROM relationship when based on a density of 1500 kg m^{-3} or a density of 1420 kg m^{-3} for that matter. If a regression line was placed through the data it would show how the density is decreasing as there is an increase in composite fibre volume. As the results in Table 3.4 suggest, this may be due to voids. Figure 3.6 shows that the densities of woven glass reinforced composites increases in a linear fashion with fibre volume fraction. However both extrapolations of data presented in Figure 3.6 whether including the density of cast resin or not, deviate away from the predicted ROM relationship. Since the fibre density is known for the glass

fibre it can be assumed that the void content within the composites is increasing as fibre volume increases as Figure 3.4 on page 129 shows.

Trying to achieve higher volume fractions by consolidating woven plies causes voids, which are likely to be due to poor wetting of fibres within central regions of the yarns. It is believed that poor wetting was not caused just by an incompatibility between flax and polyester resin but by access difficulties for resin caused by changes in the fibre architecture during pre-pressing of the stacked reinforcement.

Using Equation 2.11 with the fibre density (1420 kg m^{-3}), obtained from extrapolating the regression line through the measured densities of woven flax composites and cast resin, other fibre volume fractions of the composites can be established. Table 3.5 shows the flax composite's fibre volume fractions obtained with Equation 2.11 using a fibre density of 1420 kg m^{-3} (column 'C'). Column 'D' in Table 3.5 shows the fibre volume fractions of flax composites when calculated with Equation 2.11 using a density of 1420 kg m^{-3} but also taking account of the maximum fibre weight loss possible due to moisture loss from the reinforcement during curing. Table 3.5 also contains the fibre volume fractions for woven flax composites calculated with Equation 2.12 taking into account the maximum fibre weight loss possible (column 'E') due to moisture loss during curing. Using the more likely fibre density of 1420 kg m^{-3}, other estimates of void volume fraction can also be obtained for the woven flax composites *via* Equation 3.2 (page 127) and are shown in Table 3.5.

True fibre volume fractions of the composites are believed to lie between the values presented in Table 3.5. Although it was mentioned at the beginning of this section that the proportions of the constituents within a composite are important factors as they can determine composite properties, the true volume fractions of the constituents cannot be identified using these techniques. The values presented above are still estimates but are thought to be trustworthy. Figure 3.7 shows the estimated void volume fraction of woven flax reinforced composites calculated with Equation 3.2 using the value of 1420 kg m^{-3}

for the density of the fibre. The void volume fractions of the composites have been plotted against all values of fibre volume fractions obtained from Table 3.5.

Table 3.5 Fibre volume and void fractions of woven flax composites.

No. woven flax ply	*C.* V_f *(%)*	*D.* V_f *(%)*	*E.* V_f *(%)*	*% Diff. between C and E*	*% Diff. between D and E*	*Est.* V_{vf} *(%)*
7	33.9	32.0	31.8	6.6	0.6	0.24
8	28.8	27.2	27.5	4.7	1.1	0.65
10*	54.8	51.7	64.9	18.4	25.5	13.78
10	30.3	28.6	29.1	4.1	1.7	0.79
12	30.9	29.2	28.1	9.9	3.9	-0.75
12*	36.4	34.4	34.4	5.8	0	0.45
14	36.2	34.2	33.4	8.3	2.3	-0.35
14*	41.3	39.0	44.0	6.5	12.8	5.47
16*	44.8	42.3	43.8	2.2	3.5	1.95

*Woven reinforcement pre-pressed before composite fabrication.

As Figure 3.7 shows, the trend of the data remains very similar to Figure 3.4 on page 129 with pre-pressed woven reinforced composites displaying higher void volume fractions than woven reinforced composites, however as expected the void volume fractions of composites have been reduced. Figure 3.7 also graphically illustrates the difference between the fibre volume fractions calculated. The differences are relatively small and do not influence the trends of the data to any great extent at lower fibre volume fractions, but the differences do become greater at higher fibre volume fractions.

Using Equation 2.11 and a density value of 1500 kg m^{-3} a lower limiting fibre volume fraction was found which was believed to be a realistic value (column 'A' in Table 3.4 on

page 128). Equation 2.12 gave higher estimates for the fibre volume fraction of flax composites but the values also are realistic (column 'B' in Table 3.4 on page 128).

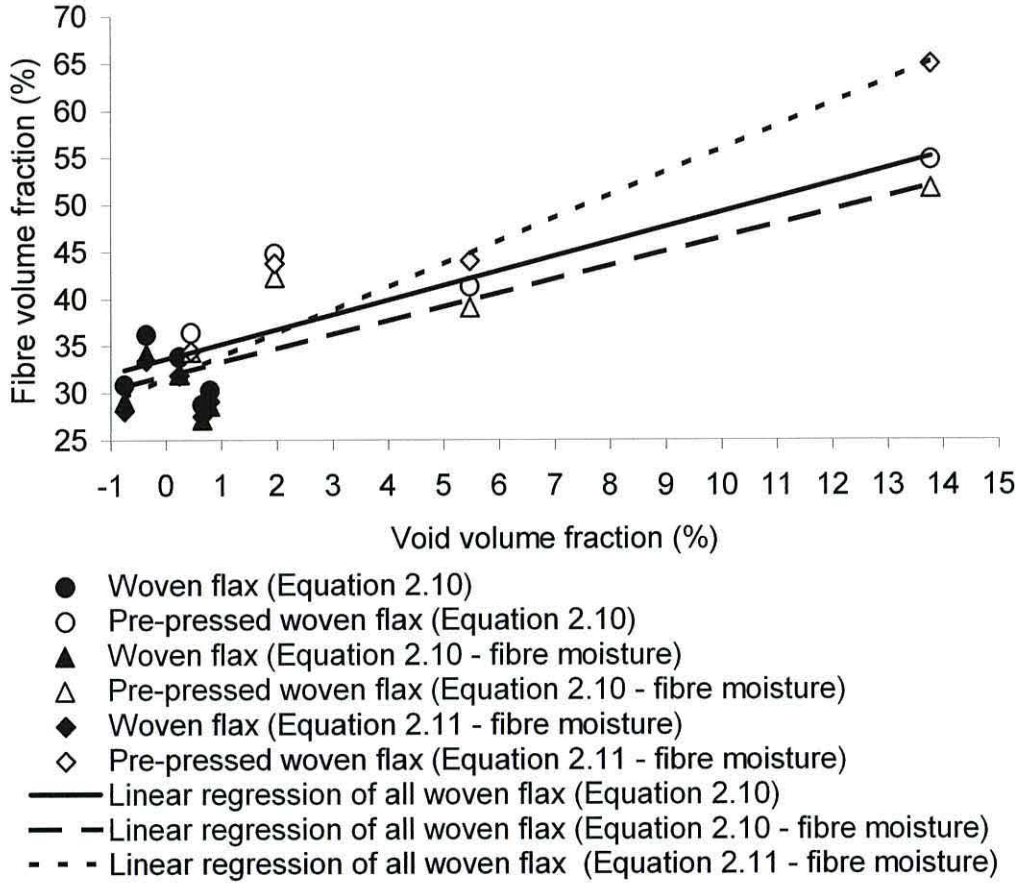


Figure 3.7 Void volume fraction of composites against fibre volume fraction calculated with Equation 2.10 and Equation 2.11.

However using Equation 2.11 with a realistic fibre density (1420 kg m^{-3}) and Equation 2.12 (minus 5.53% from mass of fibre to account for possible loss of moisture) the percentage difference between both methods has been reduced for many of the composites. A further reduction of the percentage differences is seen with most composites when Equation 2.12 (column 'E' in Table 3.5) is compared to Equation 2.11 (column 'D') when both equations are used taking account of the moisture content of the reinforcement. The actual moisture loss from the reinforcement during curing is not

known but the fibre volume fractions are similar to the ones calculated using Equation 2.11 with a fibre density of 1420 kg m^{-3}. The density of plant fibre material is very difficult to measure and is expected to have a high variation due to the non-uniform nature of the fibres, therefore, Equation 2.12 is utilised throughout this chapter for the calculation of fibre volume fraction taking into account the maximum moisture loss possible (Table 3.5, column 'E'). However it is believed that the trends seen within figures and tables presented herein would have not been altered to any great extent if any of the other values of fibre volume fraction were to be used. The fibre volume fraction of glass woven reinforced composites will be calculated with Equation 2.11 using the glass fibre density of 2582 kg m^{-3}. It is thought that Equation 2.11 can be used as the fibre density is known and the fibres are solid and are relatively uniform.

3.3.4.4 Variation in composite fibre volume fractions

The fibre volume fractions from all methods of calculation presented in Table 3.4 on page 128 and Table 3.5 on page 135 show that flax composites containing less plies sometimes have higher fibre volume fractions than composites containing more plies, whether the flax woven reinforcement was pre-pressed or not. This did not occur with the glass woven resin transfer moulded composites where the fibre volume fractions increase with an increase in the number of plies used. It is thought that the fibre volume fractions of woven flax composites do not follow the same trend as the woven glass composites because of variations made during composite fabrication and moulding *i.e.* the fibres are compressible.

During fabrication, the woven reinforcement was saturated with catalysed resin using a vacuum pulled on the interior of a sealed plastic bag. To help impregnation of resin into the stack of woven reinforcement and yarns a hand roller was also used on the exterior of the plastic bag. Resin was moved around inside the plastic tubing and as there was always at least 50% extra resin than was required for total impregnation, excess resin was

pushed off the stack and onto the waste mat (Figure 3.1 on page 108). Resin from within the stack of woven reinforcement was squeezed out by the hand rolling action and the quantity squeezed out onto the waste mat would be dependent on the amount of pressure applied. This action could be responsible for composites containing less plies having higher fibre volume fractions than composites containing more.

Woven flax composites were moulded between two glass sheets that were clamped at each corner with G-clamps. Although the G-clamps were tightened so that there was only a slight pressure placed onto the saturated stack within, the amount of pressure exerted was not measured and may have not been the constant for each composite fabricated. When tightening the G-clamps, excess resin would ooze out from the composite. It is plausible that this moulding technique is, to some extent, responsible for the fibre volume fractions, of composites with fewer plies, being greater than those of composites containing more plies.

3.3.5 Composite tensile properties

As previously mentioned, all composites were tested in the warp direction, e.g. the warp yarns ran parallel to the load for tensile testing. The average tensile load at failure of woven flax fabric (Section 3.3.1 on page 121) was lowest when tested in the warp direction whilst the tensile extension was highest. It is probable that the tensile strengths and moduli for flax composites presented herein are lower than if the composites were tested in the weft direction (preferred yarn orientation). Unfortunately a number of specimens did slip within the testing jaws and computer errors were encountered. As a result of these errors specimens from five woven flax reinforced composites and one woven glass reinforced composite failed in test and thus no data was recorded.

3.3.5.1 Tensile strength

Averages of the warp tensile stress at break (σ_{uc}) of specimens from flax and glass reinforced composite are presented in Figure 3.8 as a function of fibre volume fraction. For woven glass composites an increase in fibre volume fraction resulted in an increase in tensile stress at break. The relationship was proven to be linear by regression analysis that showed a R^2 value of 99%. Although the tensile stress at break of woven flax composites also increased with fibre volume fraction the trend was not as clear as for the woven glass composites and this is shown with the lower R^2 value of 72%. This may be due to the variations in fibre volume fraction previously mentioned in Section 3.3.4.4 on page 137 caused by poor fabrication and moulding procedures. Piggott (1980), states that a linear relationship between tensile stress at break and fibre volume fraction is realistic when composites are well made.

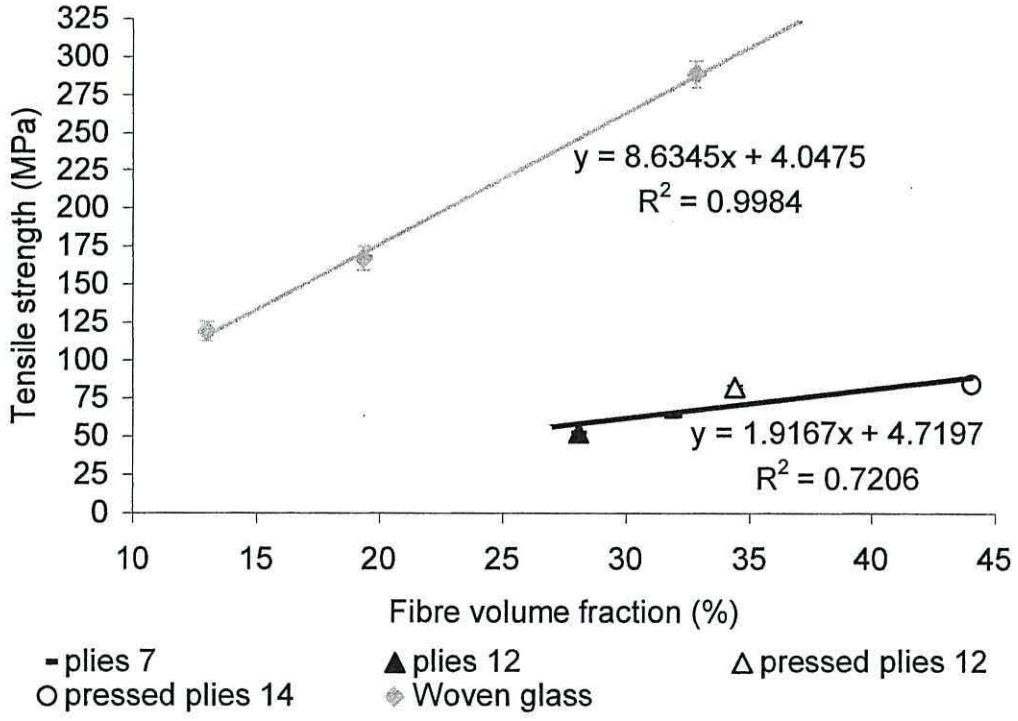


Figure 3.8 Specimens tensile stress at break as a function of fibre volume fraction of woven flax and glass reinforced polyester composites tested in the warp direction.

As expected, composites with high fibre volume fractions displayed higher tensile strengths. Composites that contain non-pressed reinforcement probably have larger resin-rich regions (matrix pockets) within the structure than those that were fabricated with pre-pressed reinforcement, especially between plies. Larger matrix pockets may be susceptible to the stresses that can develop in such regions that can initiate failure. Cracks developing in such resin rich areas may propagate to other highly stressed sections of the matrix. Pre-pressed woven flax reinforced composites may prevent this occurring as easily, because of the closer proximity of the yarns/fibres/plies which provide interfaces. Cracks propagating through rich areas of matrix may then be deflected or even blunted.

The average tensile stress of cast resin determined from this work was 31.3 MPa. All woven flax composites displayed higher tensile strengths than unreinforced cast polyester resin. The woven flax reinforced composite with the lowest fibre volume fraction of 28% had an average tensile strength of 52.7 MPa which is 68.3% higher than the unreinforced polymer. Using the regression equation from Figure 3.8 for the flax composites a theoretical critical fibre volume fraction ($V_{f\,\min}$) can be obtained. At this fibre volume fraction the fibres are not present in sufficient quantity to improve the properties of the cast resin and actually can be detrimental to them (Piggott, 1980). Although the linear regression is not an excellent 'goodness of fits' the minimum fibre volume fraction can be estimated at 14%.

At 44% fibre volume fraction, the woven flax specimens tested exhibited a tensile strength of 69.8 MPa. This value is considerably less than the tensile strength of a woven glass composite at ~13% fibre volume fraction which had an average tensile strength of 119 MPa.

Table 2.8 on page 91 shows the tensile strengths of jute non-woven and woven jute fabric reinforced polyester composites at 35 and 45% fibre volume fraction respectively. Hughes (2000) found the tensile strength of jute non-woven reinforced composites to be 69 MPa. Woven flax reinforced polyester composites at 34% fibre volume fraction had a

tensile strength of 82 MPa which is approximately 19% greater. The tensile strength of woven jute fabric measured by Gowda *et al.,* (1999) was found to be 60 MPa when tested in the preferred orientation. This is considerably less than the tensile strength of a polyester composite with a lower fibre volume fraction containing plain weaved woven flax reinforcement. However Hughes (2000) also tested glass chopped strand mat reinforced polyester composites at approximately 20% fibre volume fraction and found the tensile strength to be 73.4 MPa. Higher tensile strengths can be achieved using woven flax as reinforcement in a polyester matrix but this work has shown that the composite's fibre volume fraction has to be at least 35%.

3.3.5.2 Tensile Young's modulus

The average tensile Young's modulus (E) of woven glass and flax reinforced polyester composites are presented in Figure 3.9 as a function of fibre volume fraction. Increasing fibre volume fraction resulted in an increase of stiffness for woven glass reinforced polyester composites. The overall trend for the woven flax reinforced polyester composites also shows an increase in stiffness with increasing fibre volume fraction. However the regression analysis of the results does show that there is a weak trend. It is believed that this is partially caused by the variation during composite fabrication and moulding methods. Composites fabricated at higher fibre volume fractions likely contain a great deal more voids as detailed in Section 3.3.4.3 on page 126.

As Section 3.3.4.3 discussed, yarns within the composites that were fabricated with pre-pressed reinforcement may not have been fully impregnated with resin during fabrication. Composites containing 12 and 14 pre-pressed woven flax plies only displayed slightly higher moduli than a 12 ply woven flax reinforced composite. The void volume fraction for the 14 ply pre-pressed composite was estimated to be 5.47% (Table 3.5 on page 135). This high void content would have a significant detrimental affect upon the mechanical properties.

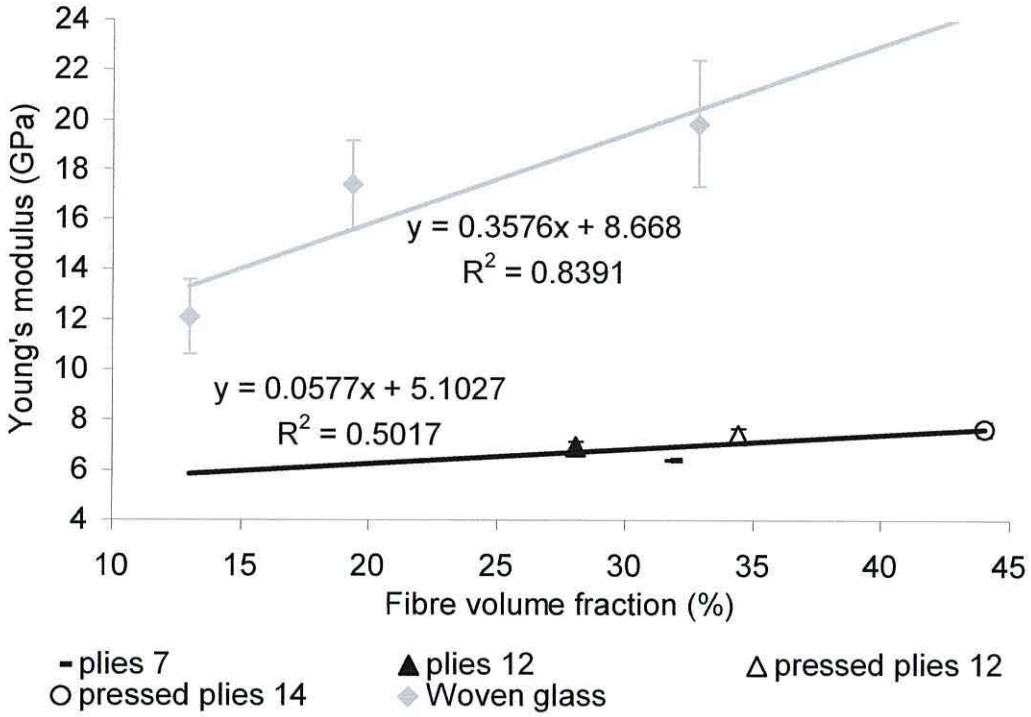


Figure 3.9 Tensile Young's modulus versus fibre volume fraction. Woven glass and flax reinforced polyester composite specimens tested in the warp direction.

The Young's modulus of the unreinforced polyester resin was found to be 4.7 GPa. All flax composites tested showed an improvement to this value, with the composite containing the least fibre giving a 48% increase and the composite with the highest fibre volume fraction giving a 59% enhancement. However, the lowest tensile Young's modulus from a glass reinforced composites is still 161% higher than the Young's modulus from the stiffest flax reinforced composite.

Figure 3.10 shows the specific Young's modulus of both types of composite. The average Young's modulus from each composite has been divided by that composite's density. Even when the lower densities of the woven flax composites are considered the Young's modulus of the stiffest flax composite is 2.7 GPa lower than the glass composite with the lowest flexural modulus.

It is interesting to compare the tensile moduli found from other types of natural fibre and glass reinforced polyester composites with the results found in this experimental. Table 2.8 on page 91 shows that the tensile modulus of non-woven jute mat (35% V_f) reinforced polyester composites is approximately 9 GPa. Woven flax polyester composites at 34% fibre volume fraction were found to have flexural moduli of 7.4 GPa respectively. When fibre volume fraction is considered, the glass composites tensile moduli presented in Table 2.8 are considerable higher than the woven flax reinforced composites.

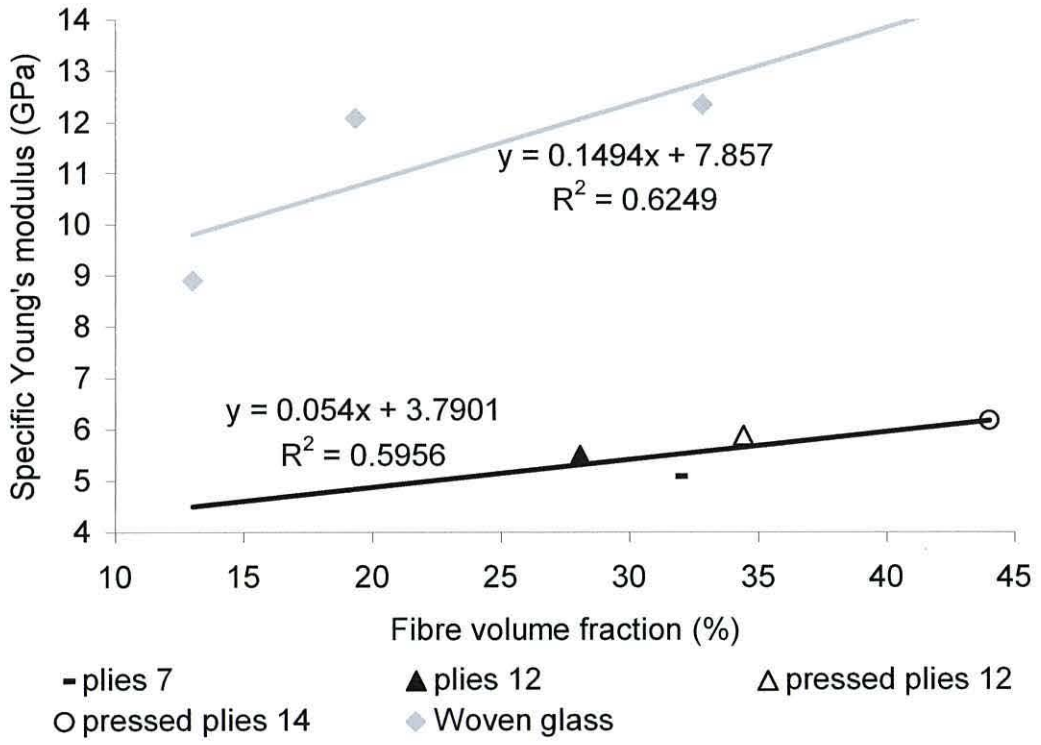


Figure 3.10 Specific Young's modulus of woven glass and flax reinforced composites as a function of fibre volume fraction.

3.3.5.3 Nature of the stress-strain behaviour

A typical tensile stress-strain curve of a specimen from each of the woven flax composites is presented in Figure 3.11, along with a stress-strain curve of an unreinforced polyester resin specimen. A representative stress-tensile elongation curve for each of the woven glass composites is shown in Figure 3.12. The measured strain from the extensometer could not be used to create a stress-strain curve for the woven glass specimens as the extensometer slipped on the surfaces during testing. Glass composites had a much smoother surface than the flax specimens and as a consequence the metal clips on the extensometer slipped.

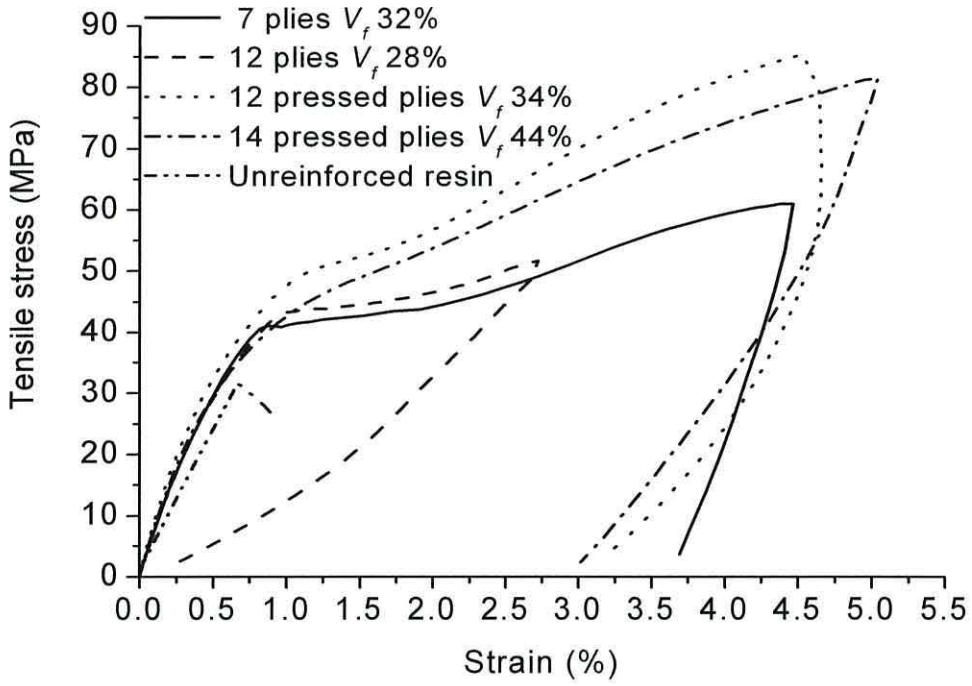


Figure 3.11 Tensile stress-strain curves of representative specimens from woven flax reinforced polyester composites and an unreinforced cast resin specimen.

However the stress-tensile elongation curves presented in Figure 3.12 show the same behaviour of the specimens as would a stress-strain curve, this can be assumed because the gauge length of all specimens tested was the same (100 mm). Tensile elongation can be recorded from the crosshead movement of the universal testing machine.

The stress-strain behaviour of the unreinforced cast resin is essentially linear until fracture which occurred at an average strain of 0.68%. The stress-strain behaviour of the woven flax composites presented in Figure 3.11 appears to be almost linear up to a strain of approximately 0.65%. For all specimens, non-linear behaviour begins at the onset of a 'knee'. The knee can be clearly seen because the slopes of the stress-strain curves undergo an abrupt decrease in gradient. After the initial knee the gradient of all the stress-strain curves begins to decrease for a period of strain before starting to increase once more before failure.

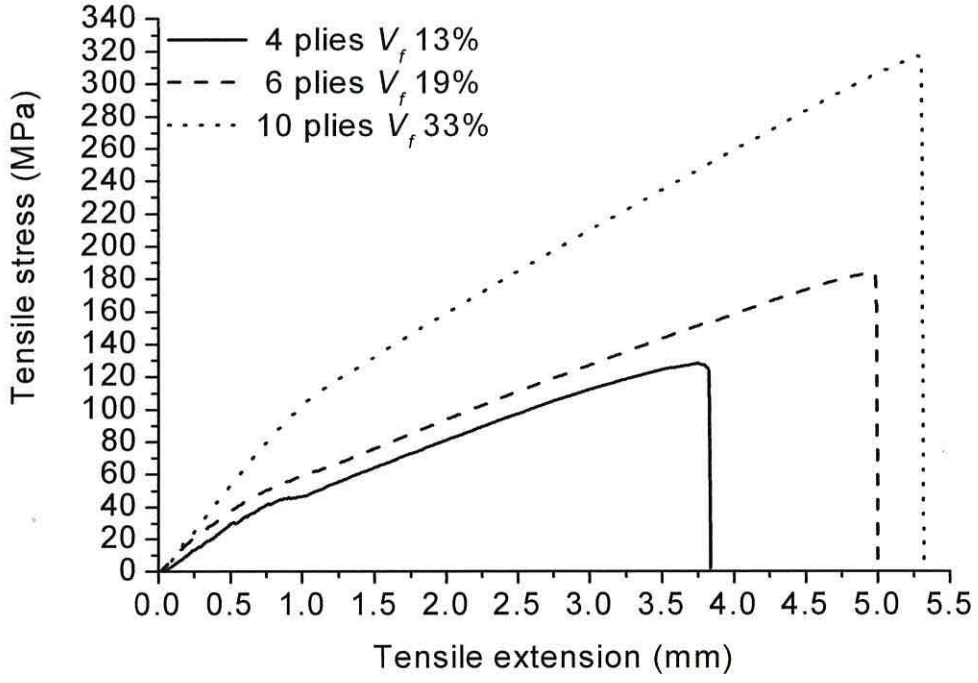


Figure 3.12 Representative stress-tensile elongation curves from specimens for each of the woven glass reinforced polyester composites.

This behaviour was not caused by specimens slipping in the testing jaws and is the recorded behaviour from the extensometer. If the extensometer had slipped then the stress-strain curves obtained would be extremely different, as this has occurred in earlier preliminary work and can be identified.

The stress-tensile extension curves of woven glass specimens shown in Figure 3.12 also show non linear behaviour. It begins between the tensile extensions of approximately 0.73 and 0.89 mm depending on specimen. However apart from the specimens that contained 4 plies of woven glass reinforcement, the curves (6 and 10 ply specimens) remain almost linear until failure after the initial change in slope, unlike the woven flax tensile specimens. Even the curves of woven glass specimens containing 4 plies of reinforcement appear almost linear until failure, but there is a slight dip in gradient after the initial change of slope from the early elastic behaviour.

It is likely that the event causing the initial change in slope for both types of composite is linked to the matrix, because the strains at which it occurs, especially for the woven flax composites, are very close to the average failure strain of the unreinforced polymer. Localised cracking within the matrix possibly may have helped initiate the debonding of yarns within the woven flax specimens. As the yarns are continuous, specimen's failure would not necessarily occur at this point because the reinforcement is able to bridge any cracks that may have formed. If this is causing the dramatic change in slope it may well also explain the following behaviour of the stress-strain curves for the woven flax composites. As matrix cracking occurs, the load borne by the reinforcement would be expected to increase. The bond between the warp yarns and the matrix may break down, thus allowing the crimped warp yarns to straighten as further load is applied. The gradient decrease of the slope after the initial change possibly reflects this process occurring. A higher percentage of the applied load at this point would be carried by the reinforcement as the matrix has partially failed. When the warp yarns reposition to the optimum position to bear the applied load, the gradient of the stress-strain curve starts to increase, as Figure 3.11 shows clearly for the woven flax specimens. The stress-strain

curves gradient increase precedes the failure event, which for the majority of the composites occurred between the strains of 4.47% and 5.05%.

The mode of failure for all the woven flax tensile specimens occurred predominately in a brittle manner that separated specimens into two pieces. Fractured ends from the majority of woven flax specimens were square and not stepped. However the specimens that contained 12 plies of woven flax reinforcement that failed at an average strain of 2.64%, showed fractured surfaces that were not square but were stepped. The step was however small, approximately 5 mm in difference from either side. This suggests that an element of shear failure has taken place. All flax fractured specimens showed some yarn pull-out. The lengths of the yarns that were pulled from the specimens range from 2 to15 mm. Yarns that had been pulled far from the fractured surface appeared to have very little resin within the yarns, and they were not rigid. However resin was visible on the surfaces of some yarns but it did not by any means cover all of the pulled out yarns surface. The yarns ends themselves seemed to have mostly fractured cleanly, leaving a flat surface much like if they had been cut by scissors.

It is probable that the early yielding behaviour seen on the curves in Figure 3.12 is also caused by localised cracking of the matrix. Although it does not coincide with the strain at which the unreinforced resin failed as closely as the woven flax specimen's knees do, it is still believed that a matrix failure causes this knee. It was previously mentioned that the tensile extension can be reported as percentage strain, and the extensions mentioned earlier in this section for the onset of the knee were between 0.73 to 0.89 mm. As this 'strain' is measured from the crosshead movement it is very likely that the true strain occurring within the gauge length is less. Taking this into consideration, it is possible that the knees seen in Figure 3.12 are occurring at roughly the same strains as the knees in Figure 3.11. The knees are thought to be caused by the matrix because the failure strain of glass fibre is approximately 2.6% (Hull and Clyne, 1996). However, the curves in Figure 3.12 are very different to the ones in Figure 3.11 as they remain almost linear after the event that causes the knee. Localised matrix cracking may have unbalanced the share of load between the fibre and matrix by causing the fibre to bear more of it, as is

thought to have been the case with the woven flax specimens but that is where the similarity between the two composite types ends. The bond between the glass fibres and the matrix is thought to have remained intact from the onset of the knee possibly to just before ultimate failure. Although the crimp of the warp fibres is considerably less than the woven flax reinforcement there is nothing occurring along the curves after the initial knee that would suggest that some kind of reinforcement movement is occurring.

The glass composite curves remain almost linear until ultimate failure that occurred in a catastrophic manner. It is thought that there was a combination of failure modes occurring at the same time within the specimens as they broke and released the high amount of energy stored within them. All the specimens remained in one piece but the failure covered more or less the entire region of the specimen's gauge length. It is evident that delamination between plies has occurred and fibre fracture, but this is not localised in one specific place. The surfaces of the fractured specimens contain numerous cracks travelling perpendicular to the test axis.

3.3.6 Flexural properties

3.3.6.1 Flexural strength

The average flexural strength of specimens from both woven flax and glass reinforced composites are presented in Figure 3.13 as a function of fibre volume fraction. As with tensile strength, the flexural strength for both composite types also increases with fibre volume fraction. The linear trend line through the woven flax averaged results shows that the regression R^2 value is low. This is thought to be because of different practices used during fabrication and moulding. The woven glass specimens were subject to less variability in manufacturing and essentially follow a linear trend (with the exception of one composite) and thus have a higher R^2 value than the woven flax specimens.

Compressing woven plies of flax reinforcement has only produced a relatively small improvement in the flexural strength; which can be seen when comparing the composites fabricated with 10, 12, or 14 plies. The flexural strength of the weakest woven glass composite is 33% greater than the highest flexural strength of a woven flax composite. However all woven flax composites had greater flexural strengths than unreinforced cast resin by at least 55%. It is also worth mentioning that the variability of the woven flax specimens tested was always lower that the woven glass specimens. This can be seen in many of the figures presented (error bars). It is believed that the woven glass composites were fabricated in a far more consistent manner than the woven flax composites as they were made within a RTM under the same conditions. The higher degree of variance seen with woven glass specimens from the same composite may be caused by the post-cure regime. The composites were post cured within the heated mould. No investigation was carried out to verify that temperatures were constant throughout the entire mould.

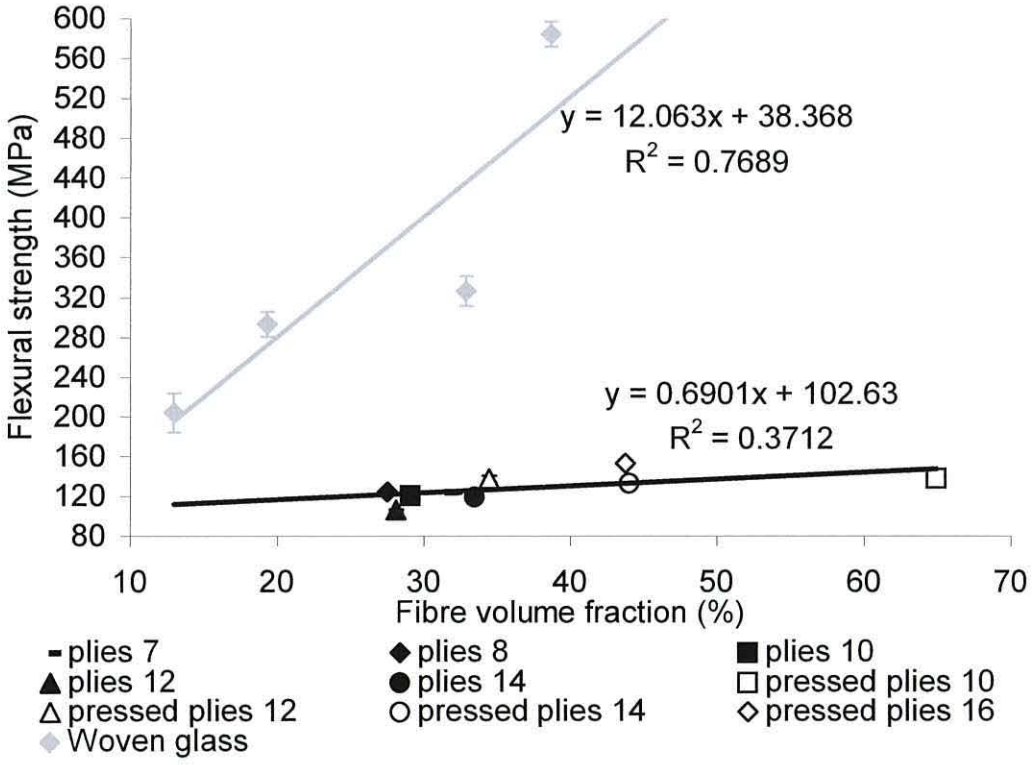


Figure 3.13 Specimens flexural stress at break as a function of fibre volume fraction of woven flax and glass reinforced polyester composites tested in the warp direction.

If the mould did not heat to 50°C evenly over the entire surface then it may explain the higher variance observed, as regions of the composite would then be post-cured at slightly different temperatures.

The flexural strengths of various natural fibres and glass fibre reinforced polyester composites are presented in Table 2.8 on page 91. The flexural strength of woven flax reinforced polyester composite at 29% fibre volume fraction is 22% greater than the flexural strength of a non-woven hemp reinforced polyester composite at approximately 30% fibre volume fraction. Jute non-woven reinforced polyester composites at approximately 35% fibre volume fraction have a flexural strength of 104.4 MPa, whereas results from this experimental work have shown that woven flax reinforced polyester composites have a flexural strength of 137 MPa at a fibre volume fraction of 34%. The flexural strengths from the woven jute fabric reinforced polyester composites (Table 2.8) are also lower than the flexural strength from this experimental work at similar fibre volume fractions. However chopped E-glass and glass chopped strand mat reinforced composites exhibit superior flexural strengths at considerably lower fibre volume fractions than the woven flax reinforced polyester composites fabricated for this investigation.

3.3.6.2 Flexural modulus

Flexural moduli of specimens from both woven flax and glass reinforced composites are presented in Figure 3.14 as a function of fibre volume fraction. As Figure 3.14 shows, the trend for the flexural modulus of woven glass fibre reinforced polyester composites is a linear increase proportional to fibre volume fraction. However, the opposite occurs with woven flax composites as there is an obvious reduction in flexural modulus as fibre volume fraction increases. As mentioned before this may be due to there being a lack of resin within the yarns as the estimated void volume fractions presented in Table 3.5 on page 135 showed. If there was a lack of resin within yarns, only friction and mechanical

locking would have prevented entire warp yarns from slipping after the interfacial bond failed. Another possible explanation may be that as the number of plies increases there is an increase in the number of yarn to yarn contacts within the composite, especially in the pre-pressed composites. In these localised regions stress transfer may be limited.

The decrease in flexural modulus as fibre volume fraction increases was unexpected. However it is important to note that the specimens are not being tested in their preferred orientation and that the fabricating and moulding techniques used may be at fault. The flexural modulus of the least stiff composites was 23% greater than the flexural modulus of cast polyester resin and at a fibre volume fraction of 27%; the flexural modulus is only 436 MPa lower than the lowest woven glass flexural modulus.

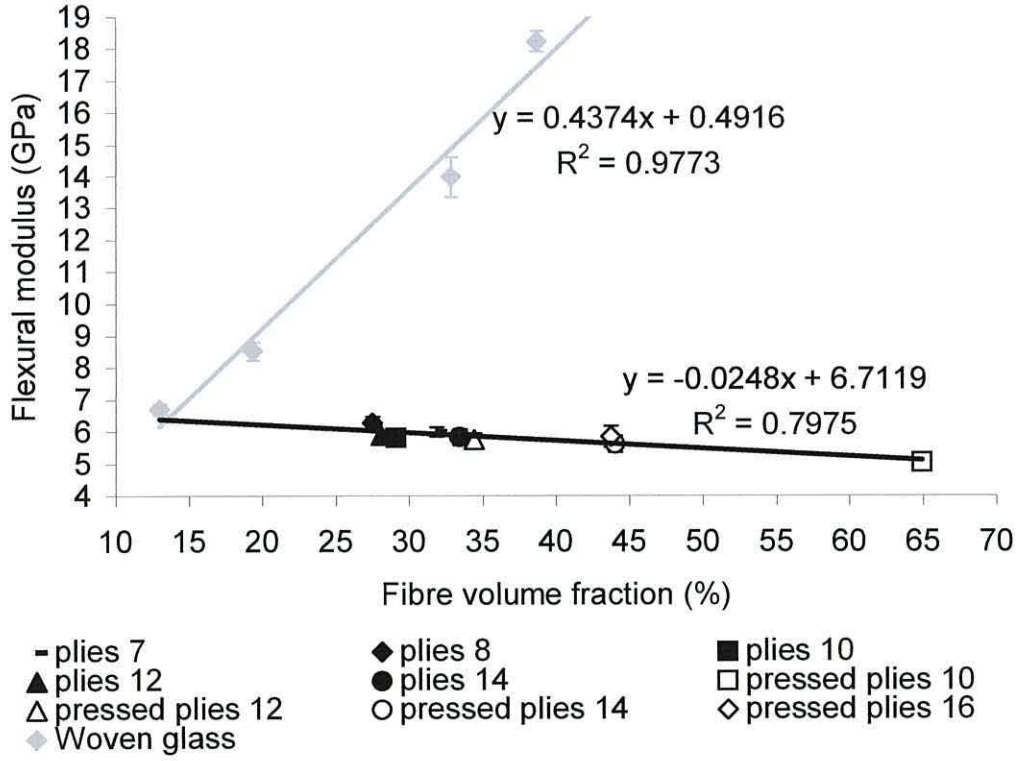


Figure 3.14 Flexural modulus of specimens tested in the warp direction from woven glass and flax reinforced polyester composites as a function of fibre volume fraction.

It is interesting to compare the flexural modulus of the woven flax reinforced polyester composites from this experimental work with the flexural modulus of other types of natural fibre and glass reinforced polyester composites presented in Table 2.8 on page 91. Table 2.8 shows that the flexural modulus of non-woven hemp (~30% V_f) and non-woven jute (35% V_f) reinforced polyester composites is 6 GPa and 7.36 GPa respectively. Woven flax polyester composites at 28 and 34% fibre volume fraction were found to have flexural moduli of 5.8 GPa and 5.7 GPa respectively. Both types of glass reinforced polyester composites presented in Table 2.8 have a far superior flexural modulus than all the other types of composite presented in Table 2.8 when their fibre volume fractions are considered.

3.3.6.3 Nature of flexural stress-strain behaviour

Figure 3.15 shows representative flexural stress-strain curves from different woven flax specimens tested. Data for specimens that contained 7 plies could not be downloaded from the computer and therefore a representative stress-strain curve is not presented. Presented in Figure 3.16 are representative flexural stress-strain curves from each different woven glass composite.

As Figure 3.3 on page 124 shows that the flexural stress-strain curve of unreinforced cast resin is essentially linear to an average strain of 1.67%. Table 3.3 on page 123 shows the average strain at failure for 8 cast resin specimens along with the standard deviation of 0.49%. The stress-strain curves presented in Figure 3.15 show a proportion of the curves to be linear. Using computer software it was concluded that non linear behaviour starts between 0.64 and 1.21% strain, depending on the curve studied. The non-linear behaviour may be due to localised cracking of the matrix and/or the debonding of yarns from the matrix within the specimens. The strains at which non-linear behaviour starts are below the average strain at which failure occurs for the unreinforced cast resin. Even when the high standard deviation is taken into account, the majority of specimens fall

under this value. Stress concentrations (at warp and weft yarn cross over points) caused by the presence of yarns/fibres provide an explanation for the non-linear behaviour starting at a lower strain than unreinforced resin alone, as these may initiate localised cracking of the matrix.

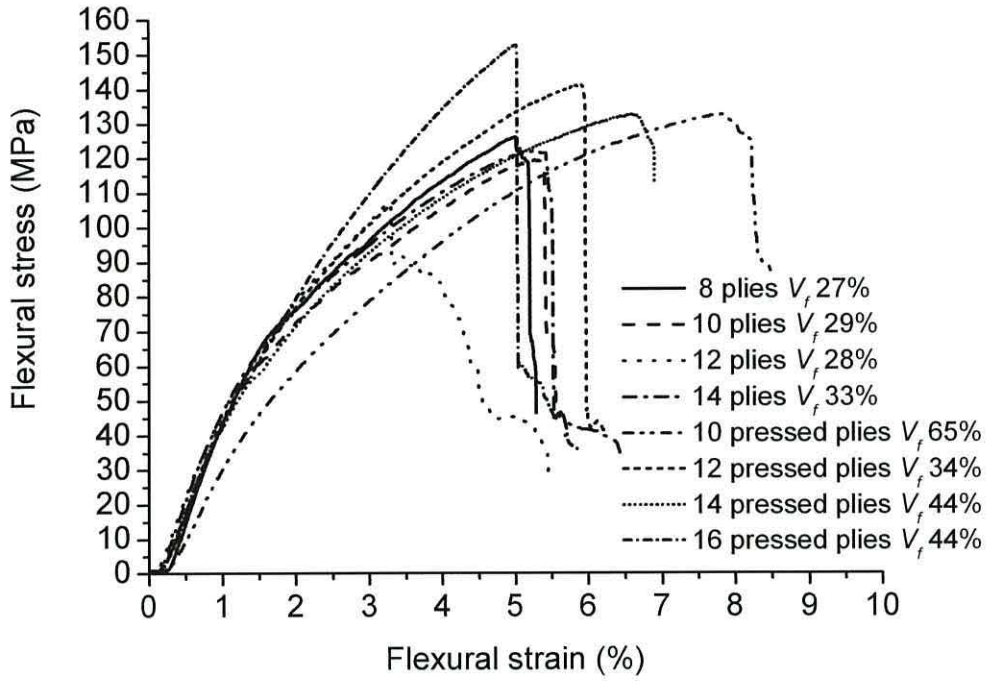


Figure 3.15 Flexural stress-strain curves of representative specimens from woven flax reinforced polyester composites.

Although the stress-strain curves are essentially linear up to almost 1% strain, beyond this point relationship becomes non-linear. Failure of the woven flax specimens was always observed to initiate on the tension surface and then progress towards the neutral axis of the specimen. The failure could not be seen on the compression surface of the specimens, as the depth of the crack spreading across the specimen's width was only approximately half of the specimen's thickness. The failure occurred in a brittle manner.

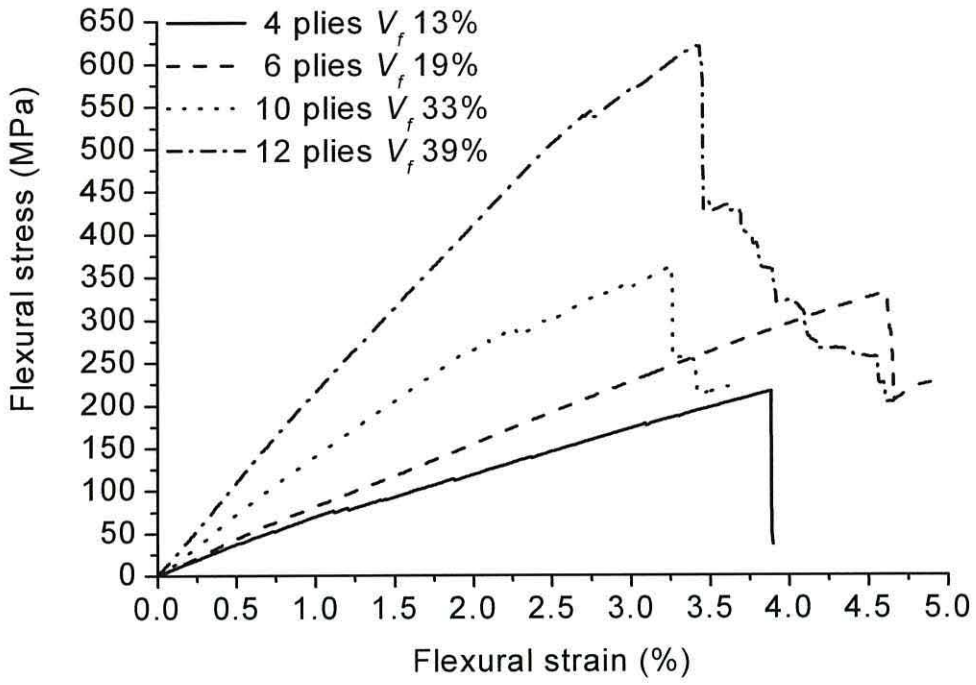


Figure 3.16 Flexural stress-strain curves of representative specimens from woven glass reinforced polyester composites.

Within the crack that led to the catastrophic failure of the specimen, fractured yarns were visible. Although there were yarns that looked as if they had been pulled out, they were in fact weft yarns that had pull-out as they were located at the edge of the fracture. The ends of the fractured warp yarns were very close to the edge of the fracture. However some warp yarns, even the ones closest to the tension face were still intact and bridged the crack that crossed them during failure. This would suggest that some fibres were able to debond from the matrix and slip during the application of load and the subsequent movement of the specimen. Some fibres may not have been bonded anyway because of incomplete resin penetration into the warp yarn.

Stress-strain behaviour of woven glass flexural specimens was similar to the behaviour displayed by the woven flax specimens. Initially, the specimen's strain was proportional

to the applied stress but non-linear behaviour occurred between 0.74 and 1.1% strains depending on specimen. A slight knee can be seen as non linear behaviour occurs but it is not as noticeable on the woven glass specimen's stress-strain curves as it is on the woven flax ones. The stress-strain curves of the specimens that contained the least reinforcement (13% V_f) show deviations along the curve occurring at approximately 1.1% strain. It is probable that as this composite contains the least reinforcement, the deviations seen on the stress-strain curves are a result of localised matrix cracking. The stress-strain curves of woven glass specimens that contained 6 plies of reinforcement showed a relatively constant increase in strain with stress after the initial knee until failure. However the specimens that contained 10 and 12 plies of reinforcement also showed this constant increase in strain with stress but at approximately 1.9 and 2.6% strain respectively the stress-strain curves start to deviate until failure. The deviations are magnified in Figure 3.17.

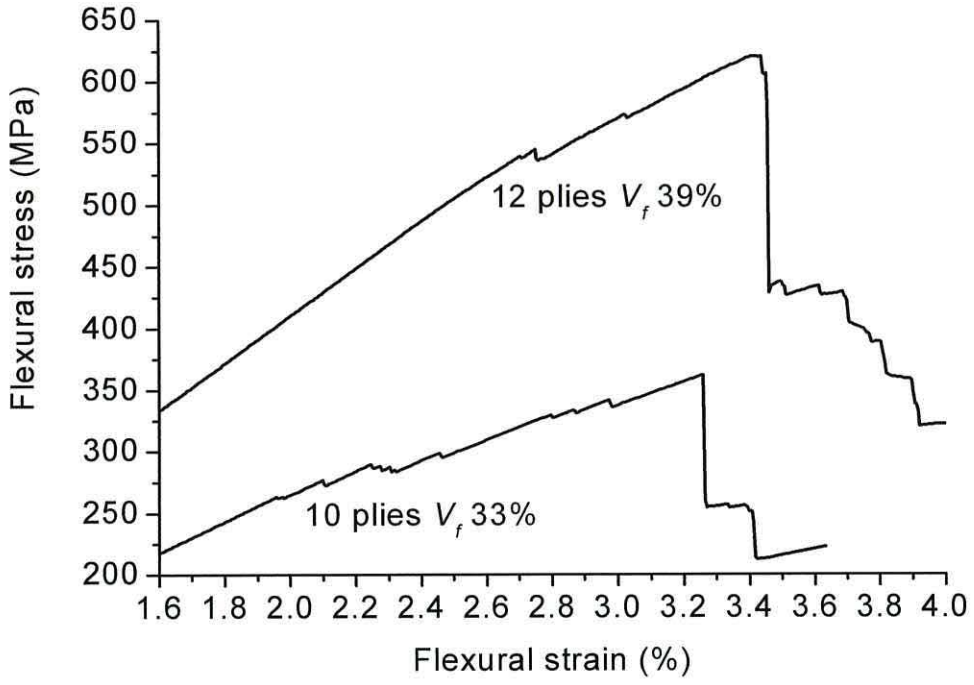


Figure 3.17 Close up of two representative flexural stress-strain curves from woven glass composites that contained 10 and 12 plies of reinforcement.

Specimens that contained 10 or 12 plies of woven glass reinforcement failed differently to the other specimens, and it is believed that the deviations on the stress-strain curves represent this. All specimens show widespread tensile and shear failure occurring on the tension face of the samples. Transverse cracks within the matrix are clearly visible that spread to approximately 2.5 cm either side of the centre of the specimens on the tension side. Fibre fracture was visible especially where the warp fibres were crimped around weft fibres. High stress concentrations were likely to exist at these regions and especially at the centre of the specimens where the load was applied. Failure on the tension surface progressed towards the neutral axis of the specimens. The interior of all specimens remained undamaged; cracks originating on the tension surface stopped. However, flexural specimens that contained 10 or 12 plies of woven glass reinforcement also failed on the compression surface. It is this failure that caused the deviations witnessed on the stress-strain curves. The compressive failure is not as widespread as the failure that occurred on the opposite side of the specimens but fibre buckling occurred, especially where the warp fibres are crimped around weft fibres. Cracks are also visible that propagate though the plane of the specimens, indicating that delamination has also occurred on the compression surface.

The specimens containing 4 and 6 plies of woven glass did not show any compressive failure on the upper surface, probably because reinforcement was not present in this region. As the composites were fabricated *via* a resin transfer machine the upper surface of the composites that contained 4 or 6 plies is extremely resin rich because there was not enough reinforcement to fill the mould completely. The depth of the mould is not supposed to be adjustable but it is thought that it does slightly depend on how tightly it is sealed. This is a concern as it cannot be measured easily. The other composites that contain 10 or 12 plies of reinforcement fill the mould more uniformly and hence have plies very close to the top and bottom surfaces of the composite. Figure 3.18 contains schematic representations of flexural specimens that either consist of 10 or 12 plies of reinforcement ('A') or 4 to 6 plies of reinforcement ('B'). Flexural specimen 'A' is uniform whereas specimen 'B' has a region of solely matrix material on the upper surface as well as a fibre reinforced region. The resin rich area found in specimens containing 4

or 6 plies of woven glass reinforcement can resist the compression stresses better than when fibres are present, thus having different compressive properties. The fibres that are present above the neutral axis in specimens containing 10 or 12 plies may be susceptible to the stresses that arise there and start to buckle and delaminate from the matrix prior to catastrophic tensile failure occurring below the neutral axis. As load is applied, the presence of fibres within the compression region may cause high stress concentrations to develop in localised areas of matrix, therefore allowing cracks to develop in localised regions where fibre buckling may occur.

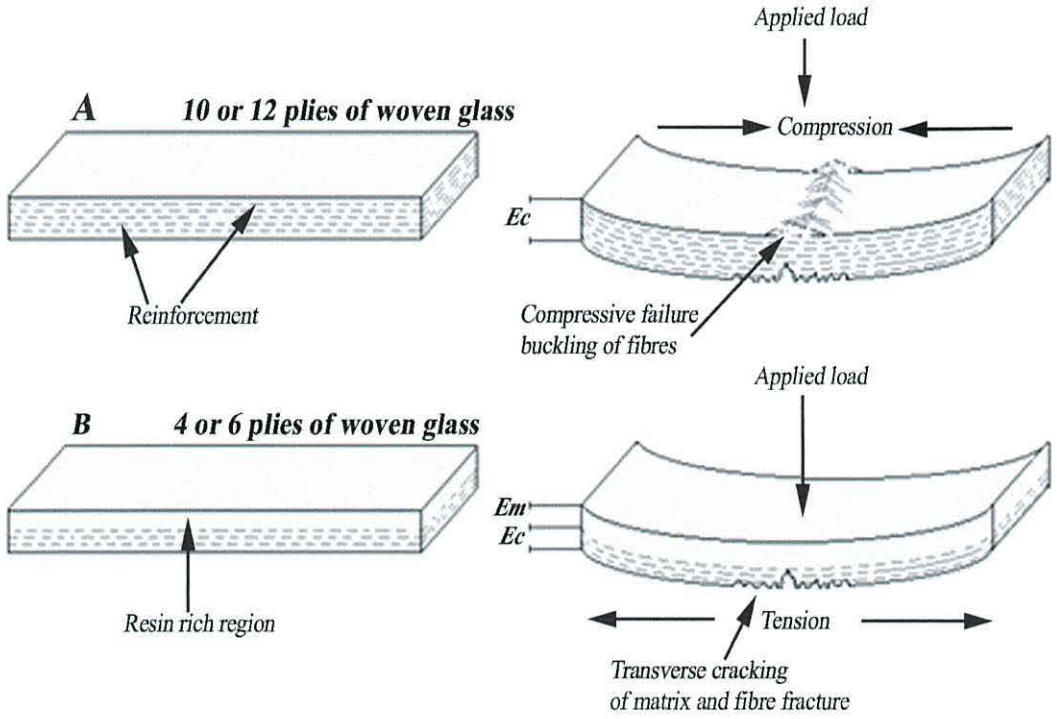


Figure 3.18 Schematic representations of the modes of failure of flexural specimens that contain different numbers of woven glass ply.

3.3.7 Impact properties

A material's Charpy impact strength is a useful measure for evaluating its toughness (the ability of a material to resist the propagation of cracks). Impact strength is defined as the ability of a material to resist fracture when an instantaneous stress is applied. Impact strength or impact resistance is expressed in kilojoules per square metre. As mentioned in Section 2.7.6 on page 73 the work of fracture is a measure of the work required to propagate a crack through a unit area of material and is also expressed in kilojoules per square metre. The material's Charpy impact strength is sometimes also referred to as the material's work of fracture. It is important to distinguish that the work of fracture obtained from impact tests is one that has been measured under dynamic loading conditions. When testing unnotched Charpy impact specimens, it is the work to initiate a fracture as well as the subsequent work of propagating it that is recorded. Due to the many test variables such as velocity and energy of impact, specimen geometry, impactor geometry etc. impact testing is very difficult to categorise and standardise (Sutherland and Guedes Soares, 1999; Sutherland and Guedes Soares, 2004). These factors as well as many others should be taken into account when comparing Charpy impact strengths (work of fracture) from literature sources due to the many variations of the test used. However, the Charpy impact strength calculated from tests performed in an identical manner can be compared successfully but results should not be used for a design criterion.

Figure 3.19 shows the average Charpy impact strengths of woven glass and flax reinforced polyester specimens as a function of fibre volume fraction. No results were obtained for the woven glass specimens at approximately 39% fibre volume fraction, since the 4 J pendulum failed to fracture the specimens and it bounced from the surfaces. Catastrophic failure occurred with all other specimens tested.

The average Charpy impact strength of the un-reinforced polymer was found to be 4.9 kJ m^{-2} (Table 3.3). Norwood (1994) found the Charpy impact strength of fully cured notched polyester resin specimens to be 9 kJ m^{-2}. Unnotched, fully cured general purpose

unsaturated polyester specimens, tested flat wise, exhibit Charpy impact strengths of approximately 9 kJ m^{-2} (Hughes, 2000; Sèbe, 1999). The works of fracture for thermosetting polymers are between 0.1 and 0.3 kJ m^{-2} (Hull and Clyne, 1996; Anderson *et al.,* 1990). The differences between the works of fracture obtained from this experimental study and the works of fracture obtained by Hull and Clyne (1996) and Anderson *et al.,* (1990) are likely to be due to the test methodology. The work of fractures obtained by Hull and Clyne (1996) and Anderson *et al.,* (1990) were from static tests (*e.g.* tension or bending) and by measuring the work (energy) put into a material during its deformation. The work done is the area under the load-extension curve produced from the specimen (provided the energy is permanently absorbed in the specimen). The work of fracture is then obtained by dividing the sectional area of the specimen where failure occurred. Test specimens are usually pre-notched so as to ensure that crack propagation occurs. Unlike the method used in this study herein, cast polyester specimens were unnotched, thus the impact test was measuring the energy required to initiate a crack or cracks and propagate them as mentioned earlier. Static tests on notched specimens measure the work required to propagate a pre-existing crack. The higher work of fracture obtained for the thermosetting polymer in this work may also be attributed to the fact that when the specimens failed they did so by fracturing into many fragments as there were numerous cracks formed on impact. As the specimens shattered on impact and many cracks were formed, this would absorb a higher amount of energy than the propagation of a single pre-existing crack that may fracture a cast polyester resin specimen into two. The fragments of the shattered cast polymer were thrown some distance from the test machine by the energy of the pendulum. It is also worthwhile mentioning that the Charpy impact testing machine will also absorb some energy because of friction.

The Charpy impact strength of both woven flax and E-glass composites is enhanced with increases in the fibre volume fraction (Figure 3.19). There is a positive linear relation between fibre volume fraction and Charpy impact strength and both types of composite display high R^2 values. Woven flax specimens that had fibre volume fractions of 29% and below, completely fractured into two pieces. On the whole, fractured surfaces

showed very little yarn pull-out but some specimens, particularly ones with fibre volume fractions at 29% had warp yarns still connecting the fractured pieces of the specimen. The length of the yarns was never greater than 5 mm. It does indicate that some warp yarns were able to debond and slide though the matrix during fracture, thus absorbing energy. Either side from the fractured surfaces of specimens, there was whitening of the matrix that spread approximately 10 mm from the fracture. It is thought that the flax fabric is debonding from the matrix within this region.

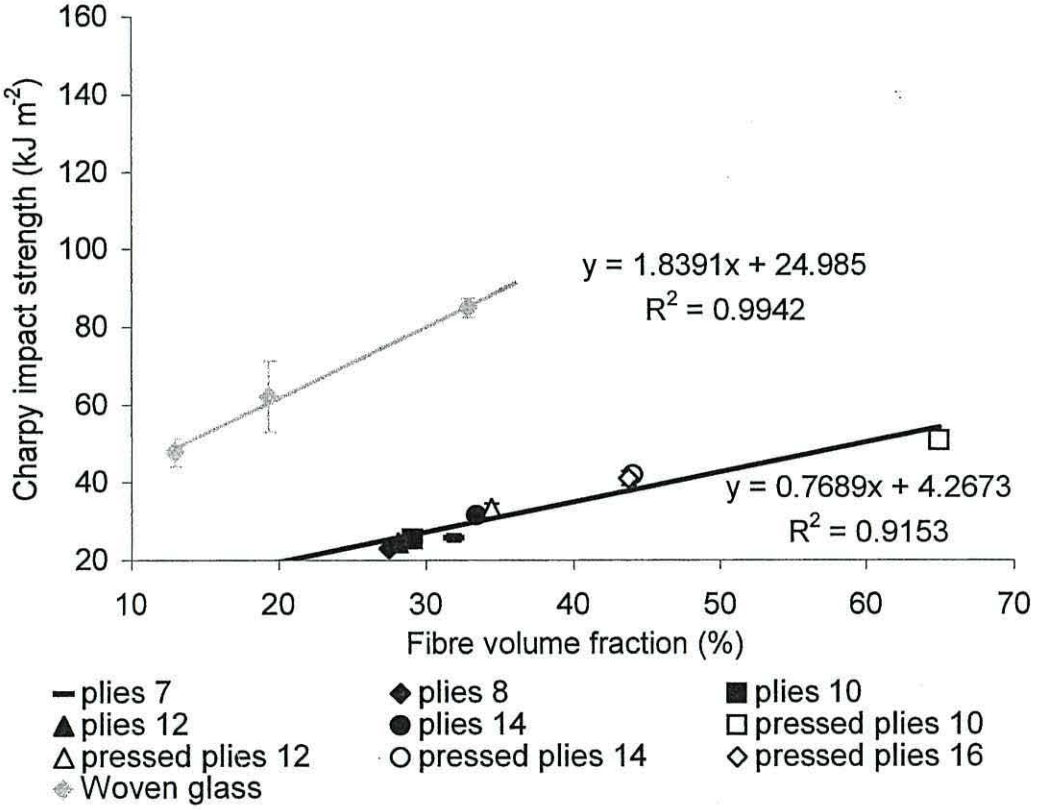


Figure 3.19 Average Charpy impact strength of specimens from woven flax and glass reinforced polyester composites as a function of fibre volume fraction.

Woven flax specimens above 29% fibre volume fraction failed on the tension surface. The fracture was not clean, as the specimen's compression surface was still intact.

Looking at the fractured region on the tension face of specimens, it could be seen that the fracture did not cleanly travel straight through towards the opposite side. The crack propagated though each ply at a slightly different location, but it only deviated laterally by a few mm for each ply. This behaviour possibly indicates that some inter-laminar shear failure is occurring. If inter-laminar shear failure is occurring in specimens with fibre volume fractions greater than 29%, it may partially account for the higher works of fractures compared with specimens at lower fibre volume fractions. Inter-laminar shear failure may absorb a greater amount of energy than a brittle type failure as the surface area of the crack is larger. However the crack may be being constantly deflected transversely when it encounters a ply of fabric which would account for the appearance of the fracture and the higher work of fracture, as more energy is absorbed than in a brittle type failure. Also visible on the surfaces of specimens with higher fibre volume fractions was whitening of the matrix. As with the specimens with a lower fibre volume fractions, it was quite localised, never more than 10 mm away from the fractured edge. A small amount of long yarn pull-out was also noticeable in specimens with higher fibre volume fractions. Some warp yarns, approximately 2-5 mm in length, protruded from the fractured surfaces. Most of the warp yarns within a specimen failed close to the fractured surface some had left sockets in the matrix from the adjacent fractured surface up to 2 mm deep. This small amount of fibre pull-out may absorb a significant quantity of energy during fracture.

Plate 3.3 shows 6 scanning electron micrographs (labelled 'A to F') of the fractured surface of a warp Charpy impact specimen containing 8 plies of woven reinforcement. The fibre volume fraction of the specimen is 27.5%. Micrograph 'A' shows an overview of the fracture; the yarns that have been pull-out during the impact are easily identified. In micrograph 'B' the plies of reinforcement can be seen, cracks that have propagated from one yarn location to the adjacent yarn location are also visible. Micrographs 'C' and 'D' illustrate how most of the yarns failed close to the fractured surface. Micrograph 'E; shows a warp yarn protruding through the matrix material. There does not appear to be a great deal resin situated between the fibres within the twisted yarn.

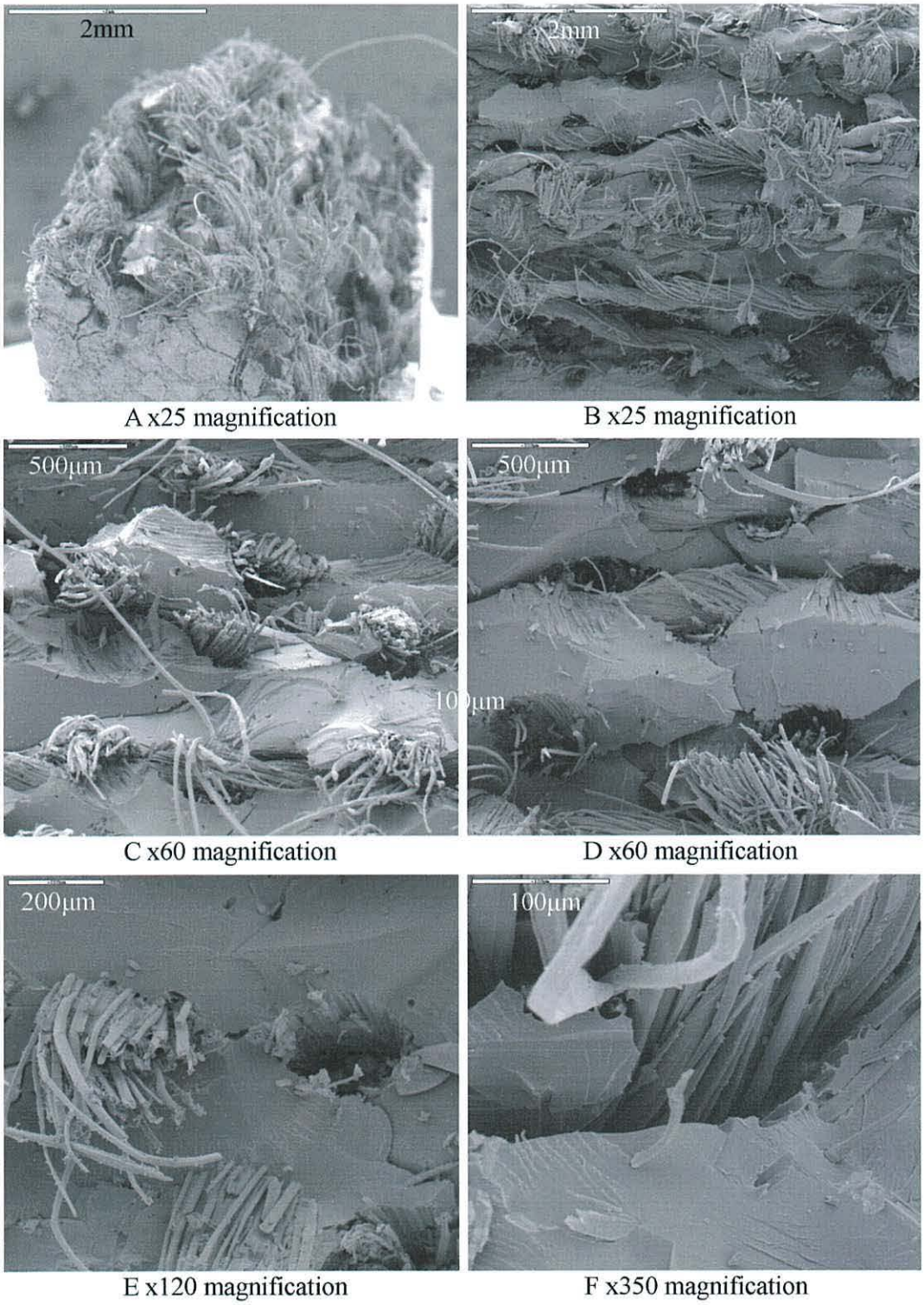


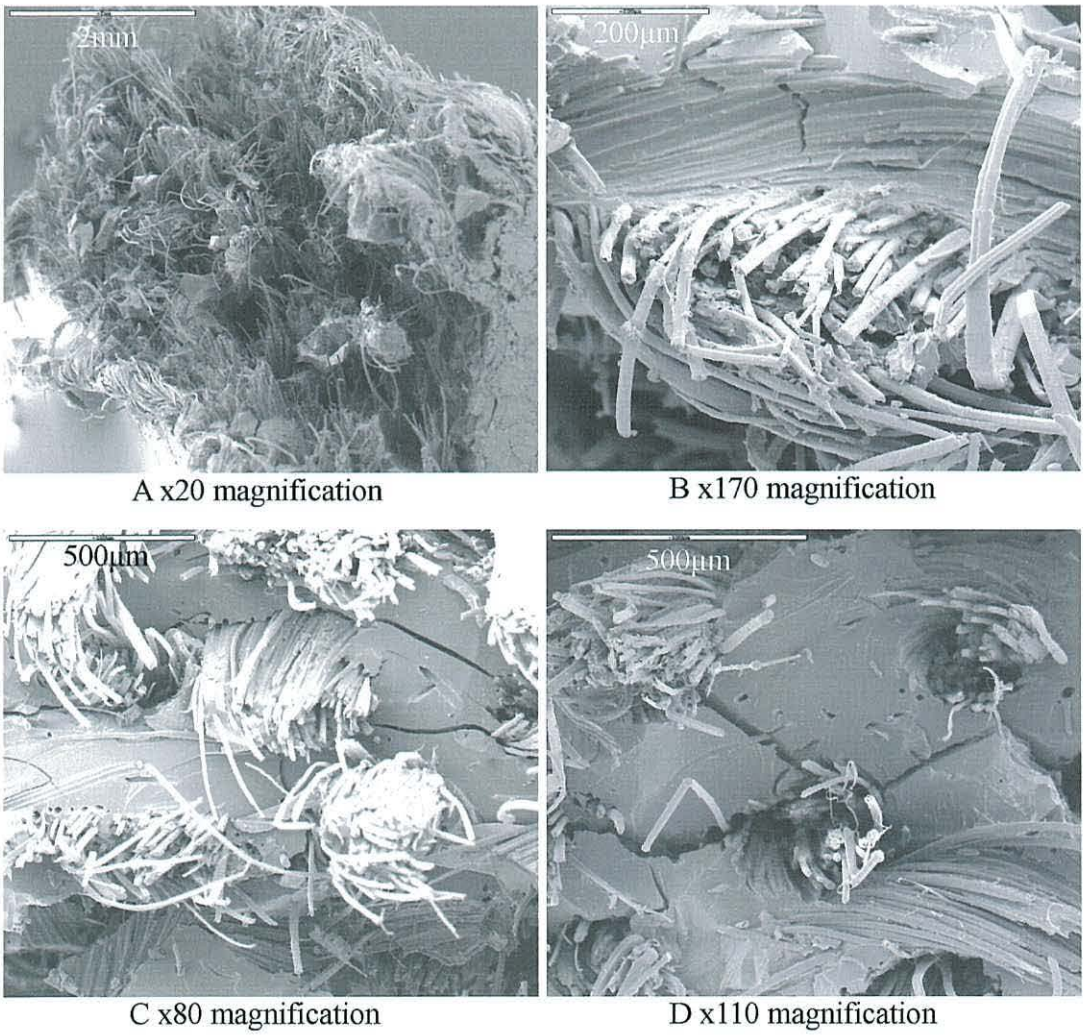
Plate 3.3 Scanning electron micrographs of the fractured surface from a warp Charpy impact specimen. Unsaturated polyester composite reinforced with 8 plies of plain weaved woven flax fabric at a fibre volume fraction of 27.5%.

There is also no visible evidence that fibre fibrillation has occurred which might have suggested that an intimate bond had occurred between the fibres and the matrix. The fibre surfaces appear smooth. Micrograph 'F' shows a warp yarn that has debonded from the matrix. Very few of the fibres from this yarn have remained embedded in the matrix. As this micrograph ('F') illustrates there appears to be very little resin within the yarn structure. Micrographs 'B to F' all show that warp yarns on the fractured surface have debonded from the matrix and their structure has remained relatively intact.

Plate 3.4 shows 4 scanning electron micrographs (labelled 'A to D') of the fractured surface of a warp Charpy impact specimen containing 12 plies of woven reinforcement that underwent pre-pressing prior to composite fabrication. The fibre volume fraction of the specimen is approximately 34%. Micrograph 'A' is an overview of the fractured specimen. In micrograph 'B' there is a protruding warp yarn. Above the warp yarn there is an imprint of a weft yarn remaining in the matrix. The imprint clearly shows that the departed weft yarn was crimping around the warp yarn prior to testing. This micrograph ('B') illustrates that the weft yarns are misaligned by warp yarns. As the reinforcement within this specimen was pre-pressed prior to composite manufacture, it is likely that many of the weft yarns have been misaligned in this manner. Micrographs 'C and D' show warp yarns that have failed near the fracture surface (low amount of yarn pull-out). In both these micrographs cracks are visible. These cracks have propagated through the matrix linking to each of the warp yarn locations. It also appears that the cracks have propagated around the yarns and have then continued to the next nearest stressed region.

Woven glass specimens at approximately 13 and 19% fibre volume fraction failed differently compared to the woven flax specimens. On the tension surface of the specimens there was widespread matrix cracking that covered approximately 80-90% of the area. Warp and weft glass fibres also decoupled from the matrix. As previously mentioned, there was a resin rich area on the upper surface of these composites and on some of the specimen's compression sides the resin rich region fractured completely away. This did not occur where the impactor struck (centre of specimen) but near one end of the specimens. Evidence of significant inter-laminar shear failure was found with

extensive delamination between the woven glass plies within the specimens. The delamination did not extend the entire length of the specimens (80 mm) but started from the region of impact and propagated to one end. Woven glass specimens at 33% fibre volume fraction also had widespread transverse matrix cracking on the tension surface. On the compression surface of the specimens, glass fibres buckled and there was a region of fibre to matrix decoupling. Delamination between plies caused by shear stresses occurred but it was not as extensive as the delamination seen with woven glass specimens at lower fibre volume fractions.



A x20 magnification B x170 magnification

C x80 magnification D x110 magnification

Plate 3.4 Scanning electron micrographs of the fractured surface from a warp Charpy impact specimen. Unsaturated polyester composite reinforced with 12 plies of plain weaved woven flax fabric (pre-pressed prior to fabrication) at a fibre volume fraction of 34.4%.

Comparing the Charpy impact strength of woven glass specimens to woven flax specimens it is obvious that there is a significant difference. Woven glass specimens at approximately 33% fibre volume fraction had a Charpy impact strength that was 157% greater that woven flax specimens at a fibre volume fraction of 34%. The relatively high toughness exhibited by the woven glass specimens is likely to be due to the fact that during failure many energy adsorbing processes occurred, such as the creation of numerous crack surfaces, fibres extensively decoupled from the matrix, the matrix deformed, fibre fracture and the frictional sliding of fibres. Similar differences between natural fibre reinforced thermosetting composites and glass fibre reinforced composites have been reported elsewhere (Hughes *et al.,* 1999; Roe and Ansell, 1985; Sèbe *et al.*, 2000).

Table 2.7 on page 90 shows the works of fracture for various types of composites. As previously mentioned there are many factors that should be taken into account when comparing work of fracture results from one type of test to another, because of the many variables. However it is interesting to compare the difference between the woven flax reinforced polyester composite's Charpy impact strengths recorded from this work to the hemp non-woven reinforced polyester composites from Table 2.7. Sèbe (1999) found that non-woven hemp mat reinforced polyester composites at a fibre volume fraction of approximately 30% had unnotched Charpy impact strengths of 14 kJ m^{-2}. Woven flax reinforced polyester composites at a fibre volume fraction of 29% had an unnotched Charpy impact strength of 25 kJ m^{-2}, which is 78.5% greater. Hughes (2000) found that the Charpy impact strength of jute non-woven reinforced polyester composites was 14.3 kJ m^{-2} at a fibre volume fraction of 36%. Woven flax reinforced polyester composites at a fibre volume fraction of 34% have Charpy impact strengths in the order of 33 kJ m^{-2}, 130% greater. Gowda *et al.,* (1999) found that the unnotched Charpy impact strength of specimens from a plain woven jute fabric reinforced polyester composite with a fibre volume fraction of 45% was 29 kJ m^{-2}. Charpy impact strengths obtained from this experimental work of woven flax composites at 44% fibre volume fraction were found to be 42 kJ m^{-2}. Hughes *et al.,* (1999) found that the unnotched Charpy impact strength of glass chopped strand mat reinforced polyester composites was 80.45 kJ m^{-2} at a fibre

volume fraction of 20%. The work of fracture of the woven flax reinforced composites is considerably greater than hemp and jute non-woven reinforced polyester composites at similar fibre volume fractions and comparable to the work of fractures recorded for unidirectional jute and sunhemp reinforced composites, but the resistance to the propagation of cracks still does not compare favourably with the conventional composites reinforced with man-made fibres such as glass in the form of chopped strand mat or woven roving.

3.4 Summary

It was found that the woven flax fabric exhibited higher tensile strengths and lower extensions to failure when tested in the weft direction. Crimping of the warp yarns around weft yarns caused lower tensile strengths and higher extensions to failure.

By making necessary assumptions it was predicted that the void content increased with fibre volume fraction for unsaturated polyester composites reinforced with plies of plain weaved woven flax and woven glass fibre roving. Furthermore, it is also predicted that composites that contain woven flax reinforcement which underwent pre-pressing prior to fabrication exhibited the highest void contents. Insufficient wetting of the flax fibres by the liquid resin may be partially responsible for the voids present within these composites.

Unexpected variations with the calculated fibre volume fraction of woven flax reinforced composites were thought to be caused by inconsistent methods used during fabrication and moulding. The use of a RTM proved to be useful tool in fabricating woven glass fibre reinforced composites on a laboratory scale as desired fibre volume fractions were easier to achieve and composites had better surface finishes. The observed properties of the woven glass reinforced polyester composites (tensile, flexural and Charpy impact strength) always exhibited an increasing relationship with fibre volume fraction. Poorer relationships between the recorded properties of woven flax reinforced composites and

fibre volume fraction were observed and are thought to have been exacerbated by the errors made during fabrication and moulding.

The tensile strengths and Young's moduli of woven flax composites did not compare well to those of woven glass polyester composites at similar fibre volume fractions. When comparing the specific stiffness of both types of composite the moduli exhibited by the lower density natural fibre reinforced composites were considerable lower than the moduli displayed by the woven glass fibre reinforced material. However, the tensile strength of woven flax reinforced polyester composites do compare well to published values of bast fibre non-woven reinforced polyester composites. The stress-strain behaviour of woven flax reinforced composites studied in this work had a region of linear behaviour but at relatively low values of stress and strain non-linear behaviour was observed. Microstructural damage is thought to occur at the onset of non-linear behaviour and therefore it is postulated that the deformation occurring is of an irreversible form.

Woven glass reinforced composites flexural strengths and moduli are significantly higher than the values observed for woven flax reinforced composites at similar fibre volume fractions. Reported values of the flexural strength and stiffness of glass fibre reinforcement in the form of chopped strand mat encapsulated in a polyester matrix are superior to those obtained from woven flax reinforced composites found in this work. However the flexural strength of woven flax reinforced polyester composites do compare well to the reported values of other natural fibre reinforced composite systems at similar fibre volume fractions. Non-linear behaviour was prevalent in all stress-strain curves examined for woven flax reinforced unsaturated polyester composites.

Charpy impact strengths of woven flax reinforced composites increased in a linear fashion with fibre volume fraction. The toughness of woven flax composites was significantly less than that of woven glass reinforced composites at comparable fibre volume fractions. However, the toughness of woven flax reinforced polyester composites was comparable and often greater than that of other non-woven plant fibre reinforced

composites and some unidirectional composites reinforced with jute and sunhemp fibre that have been reported in the literature.

In summary, it can be concluded that the methods of natural fibre reinforced composite manufacture applied within this work were inconsistent and caused high unexpected fibre volume fraction variations between composites. From the observed tensile properties of the woven flax fabric was found that composite properties vary depending on the direction a stress is applied, either the warp or the weft orientation. Woven flax fibre reinforced un-saturated polyester composites had lower properties than comparable glass fibre reinforced composites and exhibited non-linear behaviour at relatively low vales of stress.

4 WEFT YARN SIZE AND ITS INFLUENCE ON COMPOSITE MECHANICAL PROPERTIES

4.1 Introduction

A factor that may potentially influence the mechanical properties and deformation behaviour of woven flax reinforced polymer composites is the diameter or Tex of the yarns from which the reinforcement fabric is woven. To investigate the effect of yarn Tex, three woven flax fabrics each consisting of the same sized (Tex) warp yarns, but each with different sized weft yarns, were used as three ply reinforcement in epoxy matrix composites. The aim was to fabricate epoxy composites with the three plies of reinforcement in the same direction to find out how weft yarn Tex influences the deformation behaviour and mechanical properties of composites. In addition, extra epoxy composites were reinforced with three plies of the same three woven flax weave types but with the middle ply at 90° to the top and bottom plies to see how the stacking of reinforcement influences the mechanical properties and deformation behaviour.

4.2 Materials and method

4.2.1 Resin

An epoxy resin (Ampreg 20) was obtained from 'Structural Polymer Systems Ltd.' and utilised for the manufacture of all woven flax fabric reinforced composites reported in this chapter and Chapter 5. Ampreg 20 is advertised as a suitable resin for wet lay-up techniques. The characteristics of the resin were suitable for the methods used for this experimental work for fabricating composites, as it has a long gel time (190 min) when mixed with its slow hardener. Table 4.1 details the supplier's physical and mechanical

properties of Ampreg 20 epoxy resin when mixed with a slow hardener. An epoxy resin was used, rather than continuing with the polyester resin used previously as it was readily available and it was easier to work with. Although epoxy resin is usually considerably more expensive than polyester resin the epoxy resin used was obtained free of charge and proved to be a useful matrix material for studying the effects that the reinforcement architecture has upon the properties of the composite.

Table 4.1 Physical and mechanical properties of Ampreg 20 Epoxy Laminating System with slow hardener (*Source*: Structural Polymer Systems Ltd.).

Property	*Ampreg 20*
Liquid resin + hardener properties	
Typical viscosity @ 25°C (poise)	4.4
Fully cured resin properties	
Density (kg m^{-3})	1165
Linear shrinkage (%)	1.7
Tensile strength (MPa)	83.3
Tensile modulus (MPa)	3580
Strain at break (%)	4.2

4.2.2 Woven flax fabrics

Ferguson's Irish Linen (Thomas Ferguson & Co. Ltd.) based in Co. Down, Northern Ireland produced and supplied all woven flax fabrics. Table 4.2 details the three woven flax fabrics used for composite reinforcement to investigate the effect of weft yarn size on the mechanical properties and behaviour of composites, an overview and close-up photo of each fabric and the yarns is presented in Plate 4.1.

Table 4.2 Woven flax fabrics weave type and yarns sizes (Tex) and stacking sequence for composites (0° = warp direction).

Identification	*Weave*	*Warp (Tex)*	*Weft (Tex)*	*Warp fabric count (yarns/inch)*	*Weft fabric count (yarns/inch)*	*Stacking sequence*
1A	$^1/_1$	208.3	208.3	33	21	$[0°]_S$
1B						[0/90/0]
2A	$^1/_1$	208.3	83.3	33	21	$[0°]_S$
2B						[0/90/0]
3A						$[0°]_S$
3B	$^1/_1$	208.3	25.2	33	20	[0/90/0]
3B1						[0/90/0]

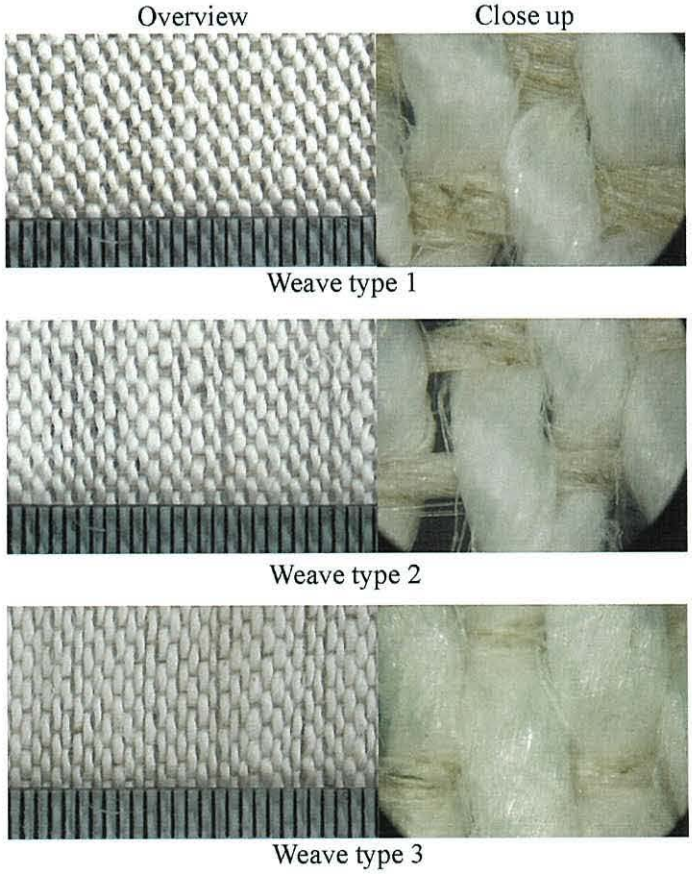


Plate 4.1 Woven flax fabrics with different weft yarn sizes.

All three flax fabrics were woven with a plain weave ($^{1}/_{1}$) and had the same sized warp yarns (~208 Tex). However the weft yarns in each fabric were a different Tex, ranging from the thickest which is approximately 208 Tex to the thinnest weft yarn at approximately 25 Tex. The fabric counts of the warp and weft yarns were essentially the same in all three fabrics as they were very uniform.

Plate 4.1 clearly shows that the warp yarns in these fabrics had been bleached. No information was obtained from Fergusons Irish Linen regarding fibre/yarn treatments apart from confirming that a bleaching process was implemented and that a size has been applied to warp yarns to facilitate weaving.

4.2.2.1 Evaluating the tensile properties of woven flax fabrics

The same method as reported in Section 3.2.2.1 on page 109 was used to evaluate the tensile properties (maximum force and elongation at maximum force) of each woven flax fabric.

4.2.3 Woven flax composite manufacture

From each fabric type 6 plies were cut measuring 260 mm^2. All plies were ironed to avoid creases. For each fabric type, 3 plies were stacked in the same orientation ($[0°]_S$) whereas the other 3 remaining plies were stacked in a simple sequence where the middle ply was 90° to the bottom and top plies ([0/90/0]), thus two composites with different stacking sequences were fabricated from each fabric type as Table 4.2 shows. All stacks of woven flax fabric were weighed on an electronic balance. An extra composite was produced (3B1) to replicate composite '3B'. Composite 3B had surface defects from moulding which was thought might yield poor data. The dimensions of the composites fabricated were 260 mm^2 with average thicknesses ranging from 2.31 to 3.28 mm.

4.2.3.1 Resin preparation

The required amount of slow hardener and Ampreg 20 epoxy resin for each composite was mixed by a ratio of 25:100 (weight) with a mechanical stirrer for 5 minutes. After stirring, the catalysed liquid resin was degassed under vacuum in a desiccator until escaping gasses ceased to raise though the liquid.

4.2.3.2 Resin impregnation into woven reinforcement

All woven flax fabric stacks were impregnated with catalysed resin by the same method as detailed in Section 3.2.4.2 and shown in Figure 3.1 on pages 112 and 113. However, although a hand roller was used over the surface of the polythene tube for all the composites to assist resin flow, very little pressure was exerted on the stack of reinforcement and resin was not squeezed from the stack of woven reinforcement. Previous experimental work fabricating and testing unsaturated polyester composites reinforced with a similar woven flax reinforcement type showed that this practice caused a great deal of variation between the fibre volume fractions of the composites.

4.2.3.3 Moulding and curing

All composites were essentially moulded by the same method as detailed in Section 3.2.4.3 on page 113. However, no G-clamps were used and the only pressure exerted onto the resin saturated stacks of woven flax fabric was the weight of the top glass plate. As the top glass plate was placed onto the stack excess resin would seep from the edges. Composites were left at approximately 20°C for 24 hours before being released from the mould. After release from the mould all composites underwent a post cure in an oven set at 50°C for 16 hours.

4.2.4 Preparation of unreinforced cast epoxy panels

Cast epoxy resin panels were prepared using the same method as cast polyester resin panels (detailed in Section 3.2.5 on page 114). Cast epoxy panels were cured and post cured as detailed in Section 4.2.3.3.

4.2.5 Measurement of composites

After post-cure of the composites, excess resin at the edge of the composites was removed using a fine toothed band saw. Care was taken not to trim any of the composite material during this process. The dimensions and weight of each composite panel and epoxy cast resin panel were measured to the accuracy of one decimal place (±0.1 mm and ±0.1 g) apart from the thickness that was measured to two decimal places (±0.01 mm). The thickness of each specimen cut from a composite or resin panel was recorded and the composite's thickness was taken to be the mean average from these measurements.

4.2.6 Specimen preparation

Tensile, flexural and impact specimens were required from composites and cast epoxy resin panels. In previous experimental work a tile cutter had been used to obtain specimens as this gave an excellent surface finish. However specimens from woven flax fabric reinforced epoxy composites for this experimental were obtained using a fine-toothed band saw. The composites were not exposed to water whilst cutting and the band-saw allowed complicated cutting patterns to be followed accurately and with ease. Approximately 1 cm of composite material was trimmed from every composite edge before cutting of specimens commenced. Cast epoxy resin specimens were obtained using the water lubricated tile cutter.

4.2.6.1 Conditioning

Specimens were conditioned prior to testing at 65% relative humidity at a constant temperature of 20°C for a minimum of one week.

4.2.6.2 Measurement of specimens

The measurement of specimens was conducted as described in Section 3.2.8.2 on page 116.

4.2.7 Testing

Flexural (100 mm by 15 mm), unnotched Charpy impact (80 mm by 10 mm) and tensile specimens (200 mm by 20 mm) were obtained in both warp and weft directions from all composites.

4.2.7.1 Flexural

A minimum of 6 specimens were obtained from each composite. Seven specimens were tested from cast epoxy resin panels. Flexural tests were preformed following the same procedure as detailed in Section 3.2.9.1 on page 117.

4.2.7.2 Tensile

Two tensile specimens were obtained and tested for both warp and weft orientations from all composites. Three cast epoxy resin tensile specimens were tested. Tensile testing was conducted following the same method as the woven flax reinforced polyester tensile specimens described in Section 3.2.9.2 on page 118.

4.2.7.3 Impact

Impact testing was conducted for all specimens following the same method as outlined in Section 3.2.9.3 on page 119. A minimum of 7 warp specimens and 7 weft specimens were tested for each woven flax fabric reinforced epoxy composite. However, generally there were approximately 9 impact specimens obtained for testing in both orientations. Ten cast epoxy resin impact specimens were tested.

4.2.8 Evaluation of physical properties

The densities of all woven flax fabric reinforced epoxy composites and cast resin panels were determined using Equation 3.1 on page 120. The densities of all specimens were also calculated using Equation 3.1. Composite fibre volume fractions were calculated using Equation 2.12 on page 62.

4.3 Results and discussion

4.3.1 Tensile properties of woven flax fabrics

The five warp specimens from each of the three woven flax fabrics contained the same number (mean average) of warp yarns. There were 31 warp yarns on average in a warp fabric specimen. The average number of weft yarns in a weft tested fabric specimen was also the same for all three sets of specimens; the mean average was found to be 19 yarns. Maximum load at failure of warp and weft fabric specimens from weaves 1 to 3 (described in Table 4.2 on page 171) are presented in Figure 4.1 whilst Figure 4.2 shows the warp and weft specimen's tensile extension at maximum load for the three weave types.

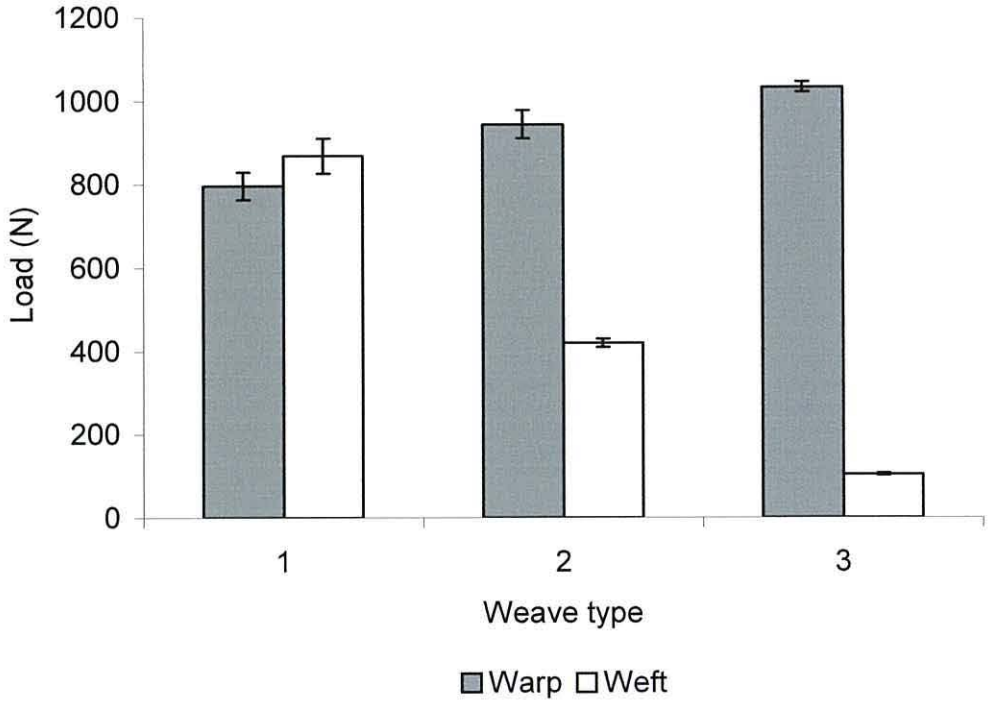


Figure 4.1 The average maximum load at failure of 3 different flax fabrics each with different sized weft yarns tested in warp and weft directions. Weave type corresponds to Table 4.2.

For weave type 1 in Figure 4.1 the warp and weft loads at failure are close. For these three fabrics it is important to take into consideration is made for the fact that on average there are 31 yarns in the warp specimens aligned to the load and only 19 weft yarns in weft orientated specimens. Weave types 2 and 3 show that as the Tex of the weft yarns reduces, there is a relationship between Tex and maximum load at failure. Considering that all warp yarns within the three woven flax fabrics are the same Tex, it is interesting that as the weft yarn Tex reduces, the maximum load at failure for warp yarn tested specimens increases.

Figure 4.2 shows that as weft yarn size in the fabrics reduces, the amount of tensile extension from warp tested specimens also reduces. This is because the crimp is reduced for warp yarns as a consequence of the reducing weft yarn Tex.

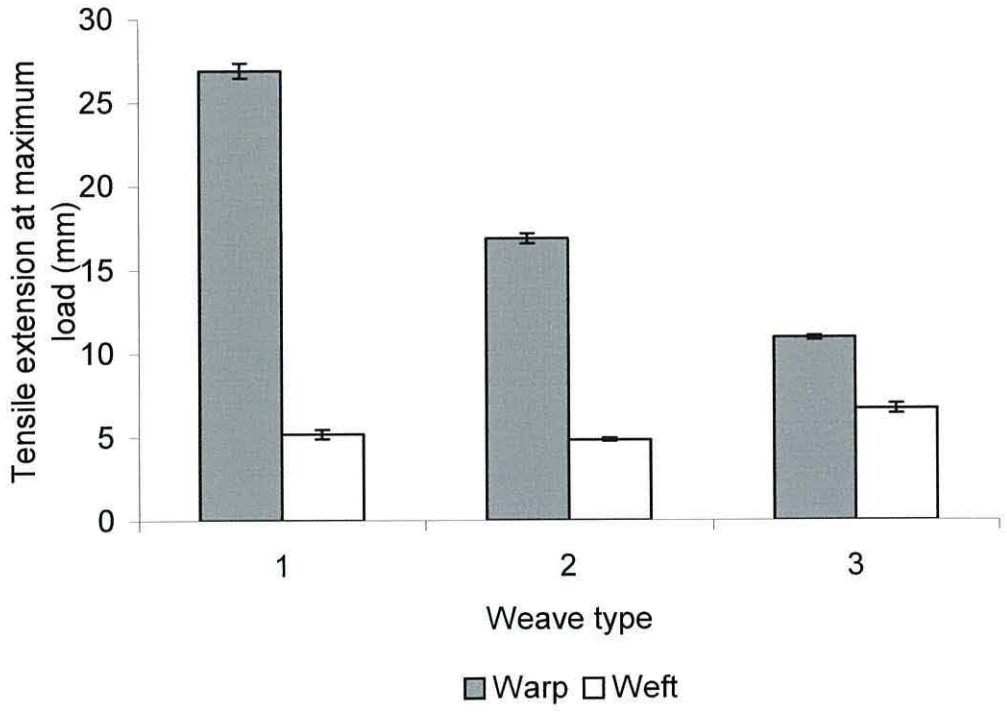


Figure 4.2 The average tensile extension at maximum load of 3 different flax fabrics each with different sized weft yarns tested in warp and weft directions. Weave type corresponds to Table 4.2.

The warp yarns within warp fabric specimens tended to align themselves to the applied load. The differences between warp tested specimens from the three woven flax fabrics are because the length of the warp yarns being tested changes with weave type due to the weft yarn Tex. Bacon (1990) notes that 'keeping fibres as straight as possible is the key to exploiting textiles to the full and engineers designing with straight fibres know that the properties of the composite materials will come close to those predicted from fibre properties'.

4.3.2 Cast epoxy properties

The mean average mechanical and physical properties of cast epoxy resin found from this experiment are presented in Table 4.3.

Table 4.3 Physical and mechanical properties of post cured cast resin (Ampreg 20 Epoxy Laminating System).

Property	*Number of specimens tested*	*Mean average result*
Density (kg m^{-3})	20	1161.6 (5.83)
Flexural properties		
Flexural stress (MPa)	7	134.8 (2.82)
Flexural modulus (MPa)	7	3136.4 (166.2)
Strain to maximum stress (%)	7	5.89 (0.51)
Tensile properties		
Tensile stress (MPa)	3	67.1 (2.08)
Tensile Young's modulus (GPa)	3	3.72 (0.31)
Strain to failure (%)	3	2.02 (0.24)
Impact properties		
Charpy impact strength (kJ m^{-2})	10	43.6 (17.33)

Note: figures in parentheses are standard deviations.

The density of the resin was found to be close to the value reported by the resin manufacturer (Table 4.1 on page 170). A density of 1165 kg m^{-3} has been used in Equation 2.12 for the calculation of composite fibre volume fractions. The tensile strength of the cast resin was found to be lower than the manufacturers value by 16.2 MPa, a difference of 24%, whilst the tensile modulus of the resin was on average higher but within the same range (when its standard deviation is taken into account). Figure 4.3 shows a typical tensile stress-strain curve from a cast epoxy Ampreg 20 tensile specimen.

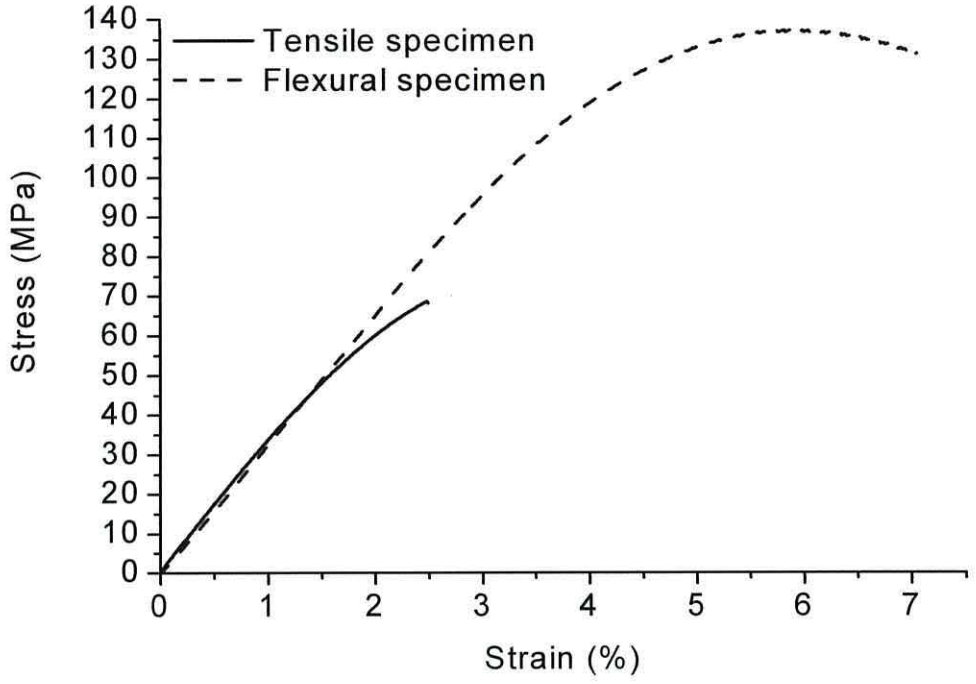


Figure 4.3 Flexural and tensile stress-strain behaviour of representative post cured cast epoxy resin specimens.

The tensile strain to failure of tensile specimen presented in Figure 4.3 is 2.48%. The average strain to failure of tensile tested specimens was found to be 2.02%. The resin manufacturer reports a strain to failure for post-cured cast Ampreg 20 more than twice the average value found from this experiment. An explanation for such differences could

possibly be found if the manufacturer's entire testing procedure was known, especially the method for measuring strain.

In this experiment, non-linear stress-strain behaviour began at approximately 1.1% strain. All tensile specimens failed in a brittle manner producing a clean fractured surface. Other macroscopic cracks within the specimens were not visible although the specimens were transparent.

A representative flexural stress-strain curve is also shown in Figure 4.3. Flexural specimens showed elastic behaviour to approximately 2.5% strain. At this yield point, plastic deformation commenced and the specimen's maximum stress is recorded within this region. The high energy adsorbing plastic deformation continued after the point of maximum stress for all specimens and because of this it was often necessary to stop the test as the distance below the bottom two supports was limited. Specimens that fractured did so in a brittle manner, with a single crack propagating transversely across the width of the specimen.

The average Charpy impact strength of the ten cast epoxy specimens tested was considerably higher than the cast polyester resin reported in Section 3 on page 123. The relatively high toughness is most likely due to the fact that the post-cured epoxy specimens can deform substantially as a load is applied. Not only does the cast resin deform but it does so without cracks forming that may initiate catastrophic failure. When a crack did initiate, impact specimens broke mostly into two larger pieces with few shards. The two larger pieces had stepped fractured surfaces.

4.3.3 Physical properties of composites

4.3.3.1 Fibre volume fraction and density

Presented in Table 4.4 are the calculated fibre volume fractions and densities of the composites. The fibre volume fractions presented in Table 4.4 have a range between approximately 32% and the lowest at 22%. Throughout this chapter the properties of composites are compared to one another without taking into consideration the different fibre volume fractions. However, the proportion of fibre to matrix within a composite obviously influences its properties and therefore it does deserve consideration. A justification for presenting results within this chapter that are not normalised to a fibre volume fraction follows.

An extrapolation technique has been utilised to find the normalised mechanical properties of composites at a specific fibre volume fraction. The fibre volume fraction was arbitrarily set to 27% for the composites presented in Table 4.4.

Table 4.4 Fibre volume fractions and densities of composites reinforced with woven flax fabrics that have different sized weft yarns.

Identification	V_f *(%)*	*Density (kg* m^{-3}*)*
1A	31.9	1219.7
1B	27.3	1225.2
2A	29.5	1204.2
2B	25.3	1218.9
3A	25.8	1211.0
3B	24.8	1203.0
3B1	22.1	1226.7

Figure 4.4 shows the observed and normalised (at 27% fibre volume fraction) warp flexural moduli of composites. Normalising the composites flexural modulus to a mid-value fibre volume fraction does not change the overall trend that can be seen when the observed flexural modulus of composites is presented.

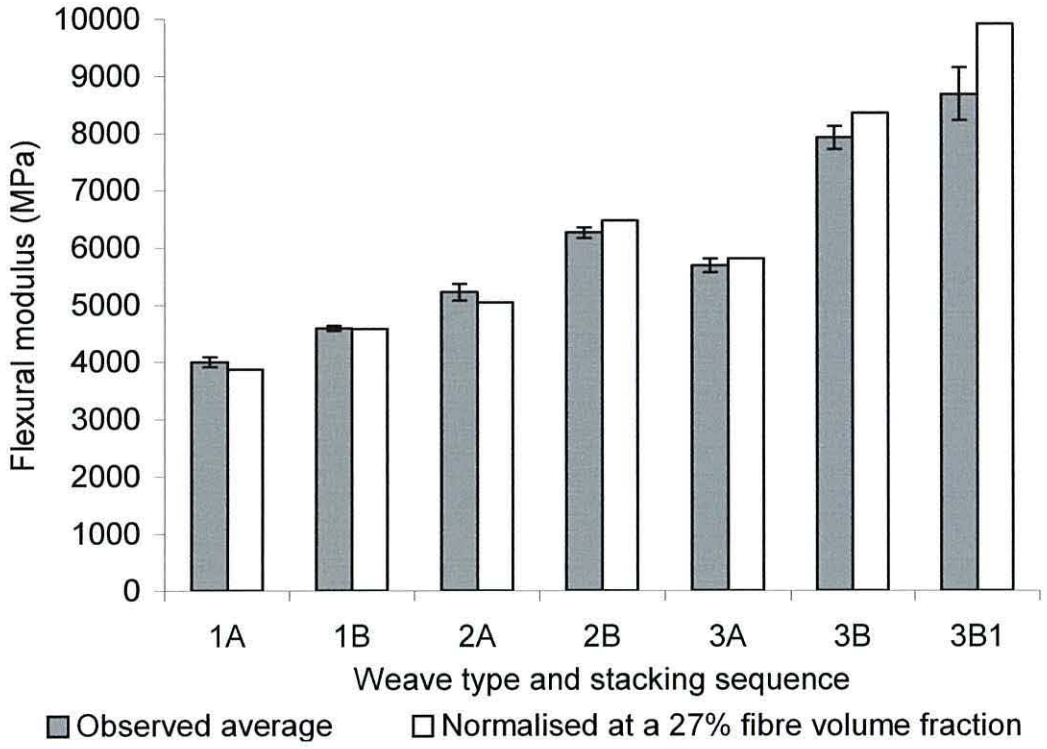


Figure 4.4 The observed and normalised average flexural modulus of warp orientated specimens obtained from the 7 composites containing reinforcement with different weft yarn sizes and that have been stacked into two sequences (A and B). Labelling is given in Table 4.2.

As the trends of the results are not significantly modified by normalising the properties to a fibre volume fraction it seems unreasonable to make assumptions in order to perform the normalisation that are not known to be true for these woven flax reinforced epoxy composites, *i.e.*, the assumption that there is a linear relationship between composite fibre volume fraction and mechanical properties. Sanadi *et al.,* (1986) found that the tensile strength, Young's modulus and work of fracture of polyester composites reinforced with

continuous unidirectional sunhemp had a linear relationship with fibre content. Roe and Ansell, (1985) also found that these three properties also followed the ROM relationship for unidirectional jute reinforced polyester composites. For unidirectional composites containing relatively strong fibres, a linear relationship would be expected, but for more complex fibre architecture it cannot be assumed.

Hill and Shawkataly, (2000) found that the tensile strength of unmodified and modified (acetylated, silane and titanate) coir and oil palm empty fruit bunch fibre reinforced polyester composites had a non-linear relationship with fibre weight fraction. The similarity between the composites that did not follow a ROM relationship was that they all displayed properties at low fibre volume or weight fractions that were less than their unreinforced resins. Hill and Shawkataly, (2000) thought that the presence of voids at the resin-fibre interface may be responsible for tensile strength of the composites being less than the unreinforced polyester at a 15% fibre weight fraction. Beyond a weight fraction of 15% the tensile strength of composites increased proportionally with fibre content until a weight fraction of 55% where a decrease occurred (Hill and Shawkataly, 2000). The decrease in tensile strength observed at high fibre loadings was attributed to an increase in fibre to fibre contact which would be detrimental to the stress transfer between fibres. Although the woven flax epoxy composites fabricated for this experiment had fibre volume fractions below the level where such a decrease in properties is expected, it is thought that the fibre volume fractions are low enough so that voids and matrix rich regions may result in non-linear behaviour between composite properties and fibre volume fraction. For example, the Charpy impact strength of the unreinforced epoxy Ampreg 20 resin is greater than the Charpy impact strength of the fibre reinforced composites. The tensile and flexural strength of some composites from this experiment also were lower than the unreinforced cast epoxy resin, thus it is expected that they would not follow a simple ROM relationship. Although these composites do not have exceptionally high fibre volume fractions, it is likely that there are many fibre to fibre contacts within yarns. Aided by the fact that the fibres are spun tightly together and contain a size, stress-transfer at these points could occur by friction. Results are therefore

presented throughout the following work which show the observed recorded properties from the specimens, rather than normalised.

Composite densities ranged from 1203 to 1226 kg m^{-3}. There was very little difference between these seven composite densities; however as expected, composite 1A had a higher density than composites 2A and 3A. This may be because there is a greater amount of fibre within this composite as the warp and weft yarns are the same Tex and the other composites contain reinforcement that has thinner weft yarns, thus less fibre content. The calculated fibre volume fractions of these composites also reflect this. Composite 1B also had a high density and calculated fibre volume fraction than composites 2B and 3B. However there is an anomaly to this pattern, in that composite 3B1 had the highest density, the lowest calculated fibre volume fraction, and also was the thinnest composite. This suggests that the void content of composite 3B1 may have been lower than the other composites.

4.4 Mechanical properties of composites containing different sized yarns

4.4.1 Flexural properties of composites

4.4.1.1 Flexural strength

Figure 4.5 shows the observed (mean average of flexural specimens) flexural strengths of warp and weft orientated specimens from the woven flax reinforced epoxy composites. Comparing the average flexural strengths of composites 1A, 2A and 3A, it is apparent that the flexural strength of the warp orientated specimens is increasing as the Tex value of the reinforcement decreases. Considering that the warp yarns within composites 1A,

2A and 3A are an identical size it demonstrates that the diameter of the weft yarns has a dramatic effect on the flexural performance; there is a 61.5% difference between the average flexural strength of warp orientated flexural specimens from composite 1A to composite 3A. The warp yarns in composites reinforced with flax fabrics that are woven with weft yarns having a lower Tex are not as severely crimped as they are when they have to pass over and under weft yarns that have a similar or larger Tex than themselves. Within the three woven flax weave types, the warp yarns are crimped approximately the same amount, it is only the nature of the crimp that is changed (angle). The less severe the crimp (lower angle), the straighter the warp yarn lies within the composite, thus acting as a better reinforcement and maybe reducing the severity of the stress-concentrations at warp and weft cross-over points because the fibres are not so misaligned causing them to possibly fail at a reduced strain.

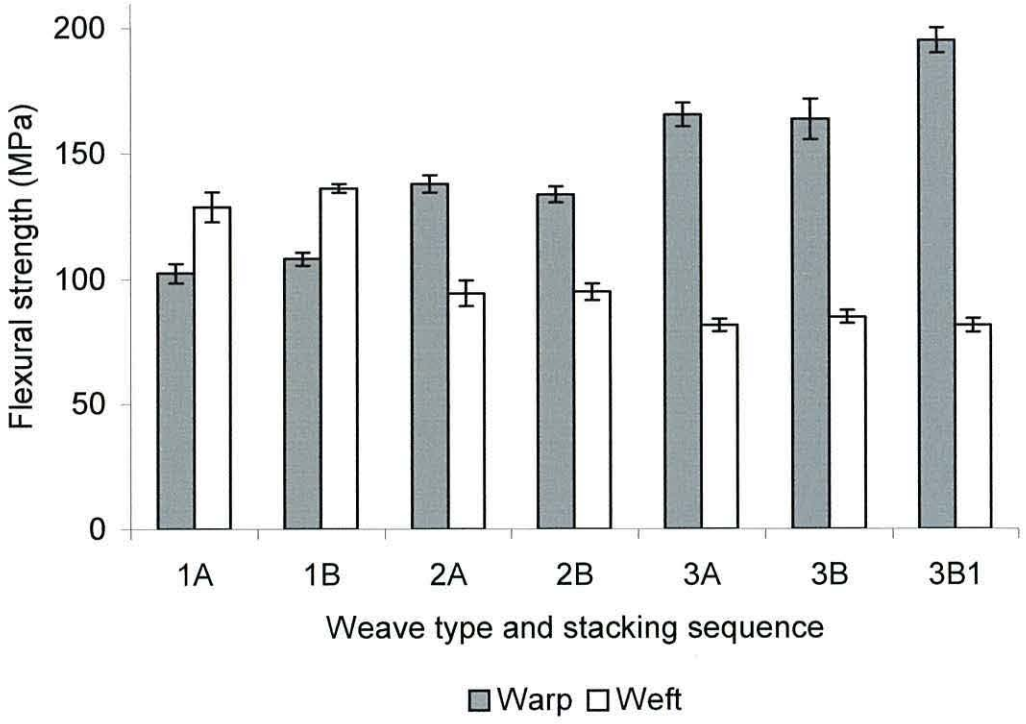


Figure 4.5 The average flexural strength of specimens tested in both warp and weft directions from composites reinforced with woven flax fabrics containing different yarn sizes and stacking sequences. Table 4.2 on page 171 identifies yarn sizes and composite stacking sequence. Flexural strength of unreinforced resin is 134 MPa.

The average flexural strength of weft orientated specimens from composites 1A, 2A and 3A are also all significantly different at a 95% confidence interval. The flexural strength of weft orientated specimens reduces as the Tex of the weft yarns decreases. The average flexural strength of weft orientated specimens from composite 1A is approximately 58% higher than the weft orientated specimens from composite 3A. It is thought that as weft yarn Tex decreases, the number of fibres within a given section of yarn reduces, thus lowering its breaking load. Fergusons Irish Linen did not confirm that the weft yarns in the three woven flax fabrics had the same level of twist. Therefore it has to be assumed that the amount of twist within the different sized weft yarns can vary. Rosiak and Przybyl (2003) state that it is fibre twist which maintains the integrity of yarns and controls their breaking strengths. The amount of twist given to a yarn may also affect other factors such as resin penetration. It is thought that the more twist in a flax yarn the less the epoxy resin will be able to penetrate into the yarn because the fibres themselves form an external barrier. It is also plausible that as weft yarn Tex decreases, the amount of resin penetration into weft yarns may increase as there are fewer fibres to inhibit resin flow, thus the void content within such yarns may decrease as the resin can wet fibres in the interior of the yarns better.

Warp orientated specimens from composite 1B ('B' symbolises the composite stacking sequence [0/90/0]) have a significantly lower average flexural strength than warp orientated specimens from composites 2B, 3B and 3B1 at a 95% confidence interval. Incidentally, composite 3B1 had the highest average warp flexural strength out of all composites, approximately 80% higher than the warp orientated specimens obtained from composite 1B. This increase in warp orientated flexural strength is related to the size of the weft yarns. However, the average flexural strength of weft orientated specimens from composite 3B1 was much lower than the equivalent from composite 1B. As the flax woven reinforcement's weft yarn size reduces, weft flexural strength of the composites also diminishes but the warp flexural strength of the composites increases.

The flexural strengths of all the composites stacked in sequence 'A' are not significantly different at a 95% confidence interval to the flexural strengths of all composites stacked in sequence 'B' in both warp and weft directions, with one exception.

There is however a significant difference between the warp orientated flexural strength from composite 3A and composite 3B1, but no significant difference between the weft orientated specimens flexural strengths between these two composites. Warp orientated specimens from composite 3B are also significantly different to the warp orientated specimens of composite 3B1 at a 95% confidence interval. Composite 3B1 was fabricated because the surfaces of composite 3B had a great deal more surface defects (small indentations) than the other composites. The surface of composite 3B1 was similar to the other surfaces of woven flax composites. However, apart from the composite 3B1, changing the stacking sequence has not significantly changed the flexural strengths achievable from these composites.

4.4.1.2 Flexural modulus

Figure 4.6 shows the average flexural moduli of warp and weft orientated specimens obtained from these composites.

Similar trends exist between the flexural moduli of warp and weft orientated specimens from composites reinforced with woven fabrics stacked in the same orientation ('A') as were noticed with their flexural strengths. The flexural modulus of warp oriented specimens increases for both types of stacked composite ('A' and 'B') as the weft yarns within the three types of woven reinforcement become thinner (lower Tex).

In contrast, the flexural modulus of weft oriented specimens decreases for both types of stacked composite ('A and B') as the weft yarns diameter reduces. Approximately a 42%

improvement in the warp oriented flexural modulus between composite 1A and 3A arises due to the effect the Tex of weft yarns has on the severity of the crimp of the warp yarns.

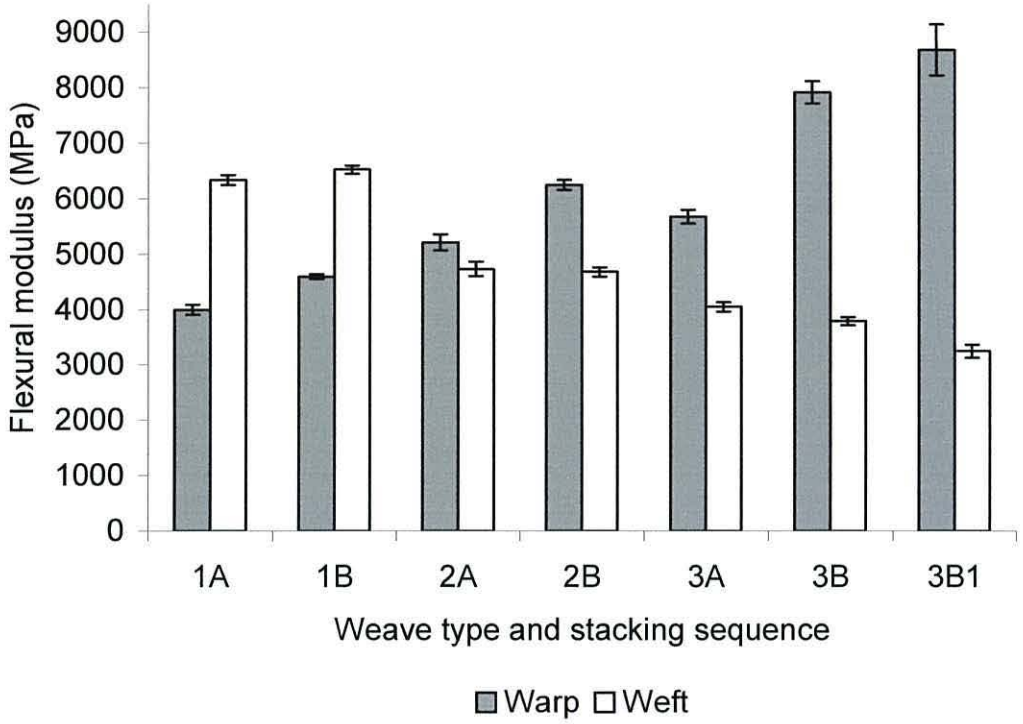


Figure 4.6 The average flexural modulus of specimens tested in both warp and weft directions from composites reinforced with woven flax fabrics containing different yarn sizes and stacking sequences. Table 4.2 on page 171 identifies yarn sizes and composite stacking sequence. Flexural modulus of unreinforced resin is 3136 MPa.

However, the weft oriented flexural modulus of composite 3A is approximately 57% lower than the weft oriented flexural modulus of composite 1A. All the warp and weft flexural moduli are significantly different at a 95% confidence interval. Composite 2A has the least difference between the warp and weft flexural modulus and composite 3B1 has the greatest.

Unlike the trends recorded from the flexural strength of the composites (apart from 3A and 3B1), there is a significant difference at a 95% confidence interval between the warp

orientated flexural moduli of composites reinforced with stacking sequence 'A' and the warp orientated flexural moduli of composites reinforced with stacking sequence 'B'. Stacking the composites with a middle ply at 90° to the top and bottom plies has a positive effect on the flexural modulus properties. The warp orientated flexural modulus of composite 1B is on average 591 MPa higher than the warp flexural modulus of composite 1A. The warp orientated flexural modulus of composite 2B is on average 1035 MPa higher than the warp flexural modulus of composite 2A. This increase in the difference progresses further as the reinforcement's weft yarns become a lower Tex; composite 3B has a warp oriented flexural modulus that is 2240 MPa higher than the warp orientated flexural modulus of composite 3A.

When reinforcing with the same weave type it might be expected that the flexural modulus of composites reinforced with stacking sequence 'B' would be lower or approximately the same as the flexural modulus of composites reinforced with stacking sequence 'A'. This is because the middle ply must be close to the neutral axis and therefore not exposed to the same stress state as the top and bottom plies. The warp orientated specimens obtained from composites reinforced with stacking sequence 'A' have the thicker warp yarns spanning the entire length of the specimen, whereas warp orientated specimens from composites reinforced with stacking sequence 'B', the centre ply of reinforcement consists of thinner weft yarns spanning the length of the specimens. As the weft yarn Tex reduces, the warp flexural modulus of composites containing reinforcement stacked with sequence 'B' is increasing.

It is not known why composites stacked with their reinforcement in the sequence [0/90/0] ('B') have a significantly higher warp flexural modulus than composites that contain reinforcement stacked in the same direction ('A'). However, as the difference between warp and weft orientated specimens from the same composite increases as weft yarn Tex reduces it seems possible that it may be linked to how the three plies of woven reinforcement stack. Table 4.5 shows the thicknesses of the three different weave types when either stacked with all three plies in the same direction ('A') or when the middle ply is at 90° to the top and bottom plies ('B'). Measurements were conducted by using

electronic callipers that exert the same amount of pressure each time they are used to perform a measurement.

Table 4.5 The thickness of three plies of each of the three different woven flax fabrics used in either stacked sequence 'A' or 'B'.

Identification	*Thickness (mm) of stacking sequence A* $([0°]_S)$	*Thickness (mm) of stacking sequence B* *([0/90/0])*	*% difference between A and B* *(%)*
1	2.82	3.13	10.9
2	2.36	2.43	2.9
3	1.90	1.91	0.5

Unsurprisingly, the thickness of the stacked woven flax reinforcement for both stacking sequences reduces as weft yarn Tex reduces. Also presented in Table 4.5 are the percentage differences between the two thicknesses of woven flax fabric stacked in sequence 'A' or 'B'. Weave type 1 (same sized warp and weft yarns) has the largest percentage difference between stacking sequences. As weft yarn Tex reduces (weave type 2 and 3) the percentage difference in thickness between the two stacking sequences also reduces. As the diameter of the weft yarns becomes smaller (lower Tex) the warp yarns are less crimped, thus the fabric plies feel smoother and are not as thick.

The misalignment of yarns within plies by other yarns in neighbouring plies may provide a possible explanation for the observed differences between the flexural modulus of warp orientated specimens from composites reinforced with stacking sequence 'A' and 'B'. Plies of woven flax fabric when stacked with sequence 'A' are, as mentioned, thickest when the weft yarns are the same Tex as the warp yarns. Due to the higher Tex of the weft yarns the crimping in the warp yarns is greater, thus when stacking these plies the peaks of the crimped warp yarns may compress against neighbouring warp yarns causing

them to set within the matrix with further misalignment in addition to that caused by crimping. Stacking three plies with the centre ply at 90° to the top and bottom plies may reduce the misalignment of warp yarns by other yarns in the top and bottom plies. By reducing the weft yarn Tex it may further reduce yarn misalignment especially when plies are stacked in sequence 'B', as the plies are flatter. When applying a load to three-point bend specimens it is the reinforcement near the surfaces that undergoes the most deformation. Thereby any negative influence to the bottom or top plies may have a dramatic effect on the flexural modulus. It is also plausible that the regions of unreinforced matrix between plies that are stacked with the same orientation are larger than when the middle ply is at 90° to the top and bottom plies. The amount of plastic deformation of these resin rich regions during flexural testing may be contributing to the observed differences.

However, at a 95% confidence interval no significant difference exists between the weft flexural modulus of composite 1A and the weft flexural modulus of composite 1B. The same is also true when comparing weft orientated flexural moduli between composites 2A and 2B and also 3A and 3B. The weft orientated flexural modulus of composites is unaffected by the stacking of reinforcement in these particular arrangements. A possible explanation for this may be because the weft yarns do not suffer to the same extent as warp yarns from the misalignment caused by other yarns in neighbouring plies. They are already aligned and held securely by the warp yarns because of the regular weave type ($^{1}/_{1}$).

4.4.1.3 Nature of stress-strain behaviour

Figure 4.7 shows four representative flexural stress-strain curves, two of which are from warp and weft orientated specimens obtained from composite 1A, and the other two are from warp and weft orientated flexural specimens obtained from composite 1B.

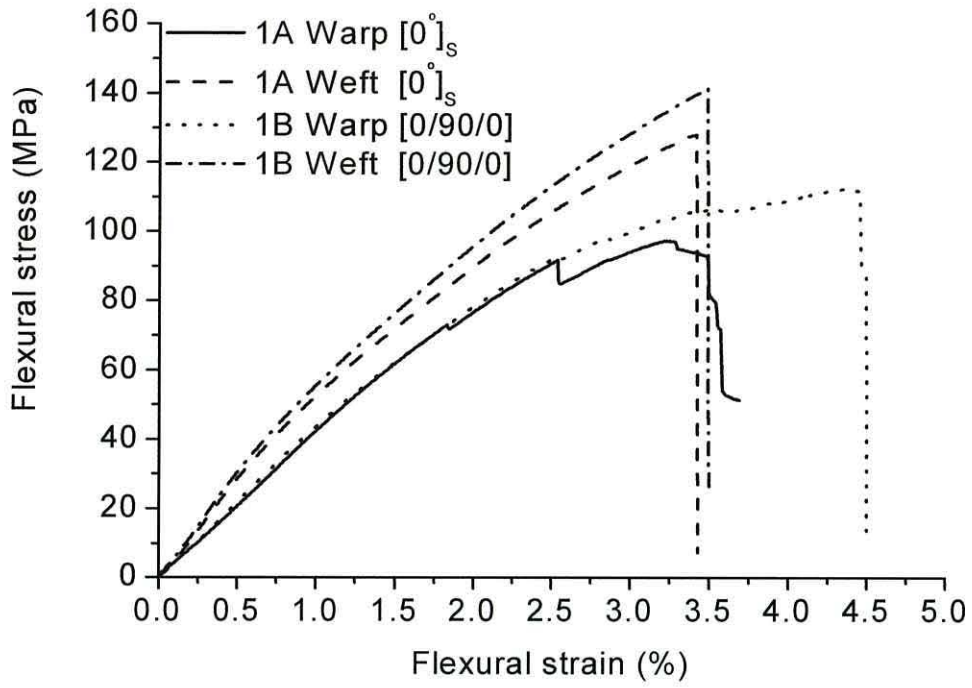


Figure 4.7 Representative flexural stress-strain curves of specimens tested in both warp and weft directions from composites (1A and 1B) that are reinforced with the same woven flax fabric but stacked in two sequences ('A' and 'B').

Figure 4.8 shows representative warp and weft orientated specimen's flexural stress-strain curves obtained from composites 3A and 3B. In both, Figure 4.7 and Figure 4.8 the stress-strain curves of the warp orientated specimens are very similar to each other in terms of overall shape. In Figure 4.7 the stress-strain curves of the warp orientated specimens are clearly seen to have a lower modulus and maximum flexural stress than the weft orientated counterparts. In contrast, the stress-strain curves of the warp orientated specimens presented in Figure 4.8 have a higher modulus and maximum flexural stress than the weft orientated specimens, most likely due to the changes in reinforcement architecture.

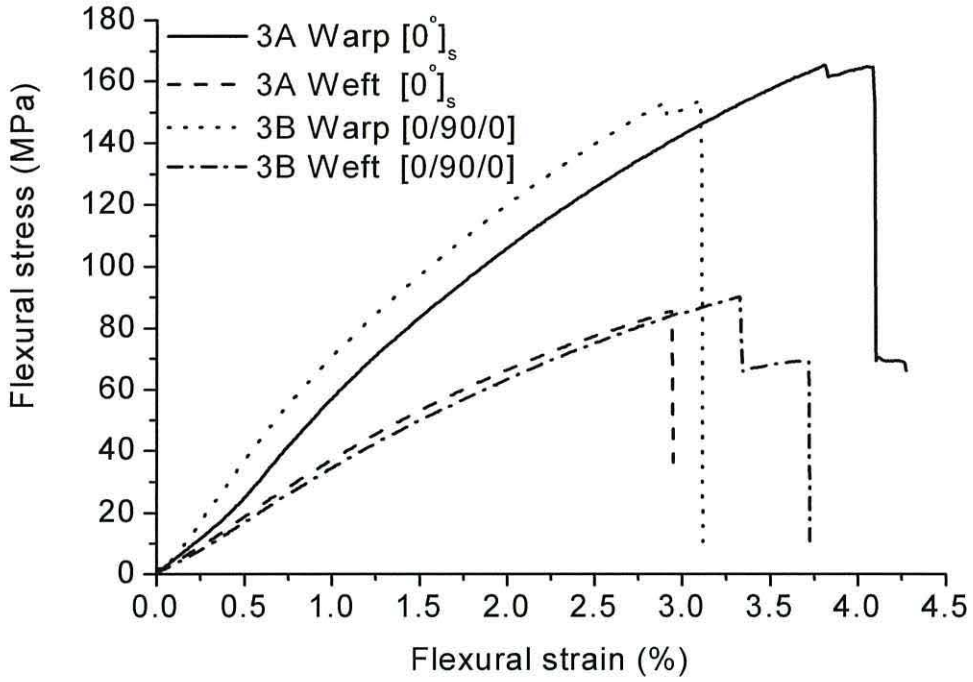


Figure 4.8 Representative flexural stress-strain curves of specimens tested in both warp and weft directions from composites (3A and 3B) that are reinforced with the same woven flax fabric but stacked in two sequences ('A' and 'B').

The warp orientated specimens from composite 1A all exhibited an event occurring on the stress-strain curves at approximately 1.75% strain and again at 2.5% strain. As these deviations are not present in the stress-strain curves from other composites, it is thought that they may be caused by matrix cracking. The plies of reinforcement may not stack effectively because the reinforcement contains weft yarns that are the same Tex as the warp yarns, thus causing severe crimping which in turn prevents plies of reinforcement stacking closely together. Due to the poor stacking, larger regions of unreinforced matrix exist in composite 1A, more than other composites including composite 1B. Matrix cracks have propagated in such regions and caused detrimental effects to the entire specimen's behaviour, however they may not have propagated sufficiently to completely cause catastrophic failure at these strains. If the deviations were caused by warp yarn

fracture, then it would be likely that larger deviations in the stress-strain curve of weft oriented specimen's from composites 2A and 3A would be present. The weft yarns in these composites are considerably thinner (lower Tex) and as Figure 4.1 on page 177 shows, the weft yarns in weft fabric specimens failed at a lower load and tensile extension than the warp yarns in warp fabric specimens from weave type 1 which is the reinforcement in composite 1A.

Some of the flexural stress-strain curves from these composites exhibit an increase in gradient of the stress-strain curve after the initial linear response to the application of load, this modulus increase, as further load is applied, occurs at strains below 1%. Figure 4.9 shows a close view of a warp and weft orientated flexural stress-strain curves from different composites (1A and 3A). It is clear from Figure 4.9 that the stress-strain curve of the weft orientated specimen from composite 1A has an initial linear region ('R1') which then gradually starts to depart from linearity. The region 'R2' of the stress-strain curve does have a lower gradient than the 'R1' region. The warp orientated specimen from composite 3B also has an initial linear region ('R1W'). However, after the 'R1W' region the stress-strain curve starts to increase in gradient ('R2W').

To investigate this behaviour, the modulus of both regions 'R1' and 'R2' (or 'R1W' and 'R2W') were calculated using the data points along the stress-strain curves of all specimens tested. The first data points (0, 0) were never used from the start of the stress-strain curve. Generally, the first set of data points were chosen a slight distance (strain) along the curve, thus allowing for the possibility that the crosshead was not contacting the specimen surface at the start of the test.

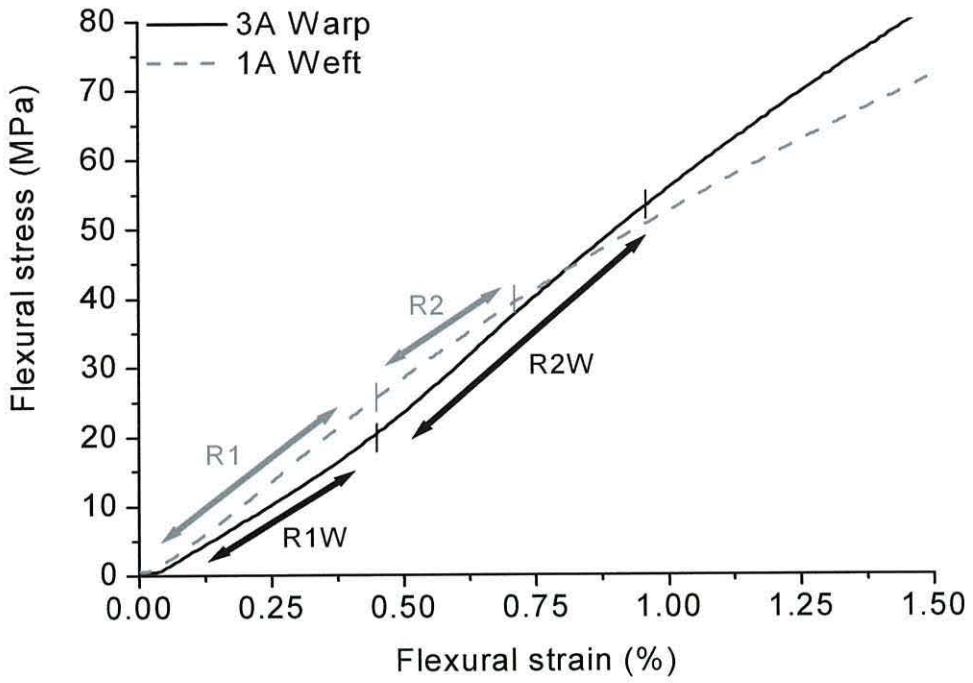


Figure 4.9 Close up of two representative stress-strain curves of warp and weft orientated flexural specimens from two composites reinforced with three plies of woven flax fabric containing different sized weft yarns.

Table 4.6 shows the average manually measured moduli for both regions ('R1' and 'R2') for each type of composite and for both test orientations. The Young's modulus calculated from the computer software attached to the Instron is often different to the calculated initial modulus ('R1'). The computer used the first set of data points (co-ordinates) from the start of the stress-strain curve; this has not been practiced when the modulus is calculated manually. However, if the manually calculated moduli of the initial linear regions of specimens from different composites had been used for the creation of Figure 4.6 on page 189 instead of the automatic Young's modulus calculated by the computer software, the same trends still exist.

Table 4.6 Average modulus of initial and second linear region of flexural stress-strain curves for composites reinforced with different sized weft yarns and stacked in two sequences ('A' and 'B').

Identification	*Modulus of region 1 'R1' (GPa)*	*Modulus of region 2 'R2' (GPa)*	*'R2' > 'R1' = + 'R2' < 'R1' = -*
1A WARP	3.88	4.14	+
1A WEFT	5.85	5.69	-
1B WARP	4.31	4.50	+
1B WEFT	6.08	5.79	-
2A WARP	4.73	5.58	+
2A WEFT	4.44	4.26	-
2B WARP	5.56	5.62	+
2B WEFT	3.82	4.29	+
3A WARP	4.64	6.23	+
3A WEFT	3.31	3.68	+
3B WARP	6.89	7.56	+
3B WEFT	3.14	3.58	+

Table 4.6 shows that all the warp orientated specimens, whether from composites stacked in sequence 'A' or 'B' show an increase in modulus occurring between region 1 and region 2. Weft orientated specimens from composites 1A, 1B and 2A are the only ones that do not show any increases in modulus. It is not known what exactly determines this behaviour. However the reinforcement's architecture is thought to affect the amount of the modulus changes as the average percentage difference between the warp orientated specimens modulus at region 1 and region 2 of composite 1A is 6.72%. In composite 2A the average difference between region 1 and region 2 is 18.02% and between region 1 and region 2 in composite 3A (lowest Tex in weft yarns) the average difference is greater again at 34.27%. The warp yarns within these three composites are progressively becoming straighter as weft yarn Tex reduces. The warp yarns within these composites are approximately 208 Tex. Other factors apart from the Tex of the weft yarns, such as

the amount of resin penetration into the yarns and the wetting of fibres themselves by the liquid resin may also affect this recorded behaviour, yarns may undergo a certain amount of stretching/realignment before resisting the tensile loads near the surface of the specimen that is under tension during flexural testing, this type of behaviour is seen in Figure 3.2 on page 122 where warp yarns undergo large amounts of tensile extension before resisting the application of a tensile load prior to ultimate failure.

Fractured flexural specimens from composites 1A and 1B visually appeared very similar. Catastrophic failure occurred on the tensile face for both warp and weft orientated specimens from both types of stacked composite. A straight crack propagating across the width of the tensile surface of the flexural specimens was visible; although the specimens had not completely broken, the failure appeared to have occurred in a brittle manner. No other matrix cracking was visible on the tensile face of specimens, but on the compression surface of all specimens there was a whitening of the reinforcement under the surface. The weft yarns closest to the tensile face from weft orientated specimens from composite 1A and 1B had all completely fractured. The warp yarns near the bottom of the warp orientated specimens from composite 1B also fractured during failure. However, not all the warp yarns near the tension face of warp orientated specimens from composite 1A had fractured. By completely fracturing the specimens to expose the entire fractured surface, it was apparent that only a few yarns under tension had pulled out and at most by couple of millimetres.

Fractured flexural specimens from composites 2A and 2B were similar to composites 1A and 1B. Warp yarns from warp orientated specimens and weft yarns from weft orientated specimens within the bottom plies near the surface of specimens which is under tension had fractured. Brittle failure had occurred on the tensile face of specimens in the form of a single crack propagating across the width. Specimens were intact and still exhibited a degree of integrity after failing. A certain degree of compression failure had occurred near the compression surfaces as a whitening of the matrix across the specimens was visible. No surface cracks or evidence of yarn buckling was found on any of the compression surfaces of the specimens.

Weft orientated specimens from composite 3A fractured completely. The fractured surfaces were very flat, and weft yarns had fractured flush with the surface in a brittle manner. Slightly crimped warp yarns were found on the fracture surface of the composites, embedded in the resin. All of the fractured specimens from composites 3A and 3B failed on the tensile face in a brittle manner. Not all, but the majority of warp orientated specimens from composites 3A and 3B had other cracking visible on the tensile surface of specimens. Yarn fracture had occurred within the bottom plies in weft and warp orientated specimens from composite 3A and 3B, weft yarn fracture had also occurred in weft orientated specimens from composite 3B.

4.4.2 Tensile properties of composites

4.4.2.1 Tensile strength

Figure 4.10 shows the average tensile strengths from warp and weft orientated specimens from composites containing reinforcement with different sized weft yarns.

The tensile strength of warp orientated specimens that were reinforced with all three woven flax plies stacked in the same direction ('A') increased as weft yarn Tex decreased. Tensile strength of weft orientated specimens also reduced as weft yarn Tex decreased in composites that had all three plies stacked in the same direction. The tensile strengths of warp orientated specimens from composite 1A were lower on average by 86% than the warp orientated specimens from composite 3A. Weft orientated specimens from composite 1A had an average tensile strength 160% higher than the weft orientated specimens from composite 3A.

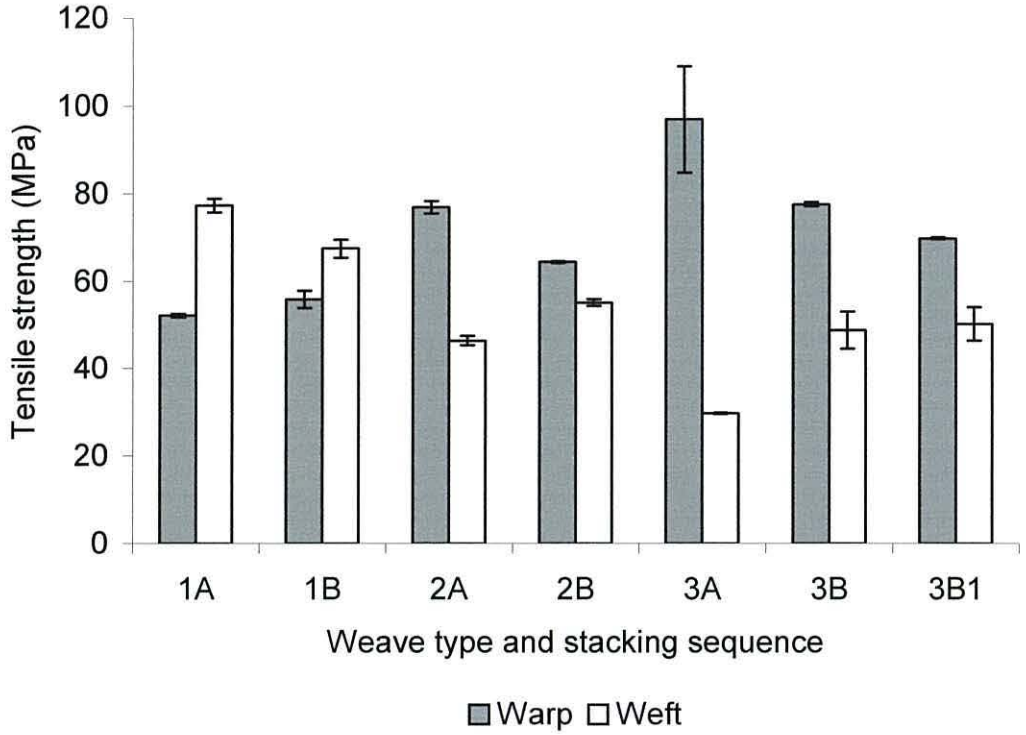


Figure 4.10 The average tensile strength of specimens tested in both warp and weft directions from composites reinforced with woven flax fabrics containing different yarn sizes and stacking sequences. Table 4.2 on page 171 identifies yarn sizes and composite stacking sequence. Tensile strength of unreinforced resin is 67 MPa.

The tensile strengths of warp orientated specimens from composites reinforced with stacking sequences 'B' ([0/90/0]) also increased as weft yarn Tex reduced, although the percentage difference between warp orientated specimens from composite 1B and 3B was 39%. This percentage difference between the averages is smaller than the difference between these specimens when the composites were reinforced with plies in the same direction. The tensile strength of weft orientated specimens reduced as weft yarn Tex decreased in composites reinforced with stacking sequence 'B'. The percentage difference between weft orientated specimens from composite 1B and weft orientated specimens from composite 3B was 38%. The tensile strengths of weft orientated specimens from composite reinforced with woven flax plies stacked in sequence 'B' were higher in composites 2B and 3B than the weft orientated specimens from composites 2A

and 3A. This is obviously due to the central plies of weft orientated specimens from composites 2B and 3B being reinforced with aligned warp yarns that are a higher Tex than the weft yarns that are parallel to the direction of the load. The central ply in composite 3B improved the average weft orientated specimens tensile strength by 64% when compared to weft orientated specimens from composite 3A. The large increase is also attributed to the fact that the warp yarns within the central ply in composite 3B were less crimped and therefore able to act as better reinforcement as the fibres were more aligned to the load from the onset of load application.

Many of the specimens failed at a stress approximately the same or lower than the unreinforced epoxy resin. It is thought that the composite's fibre volume fractions were low and therefore it is possible that failure of the composites initiated in localised regions of the matrix, as the stresses within these composites reach the failure stress of the unreinforced matrix which may even be reduced by the presence of fibres (Hull and Clyne, 1996). As this occurs, cracks propagate though the matrix and flax yarns, causing catastrophic failure. In such failure, the reinforcement has not effectively added to the tensile strength of the composite. Composites that contain low fibre loadings can have a lower tensile strength than the unreinforced matrix because of flaws (Devi *et al.*, 1997). However, some tensile specimens from these composites are greater than the tensile strength of the unreinforced epoxy resin. The failure of these specimens may have occurred as a result of yarn fracture prior to matrix failure as there was a sufficient fibre/yarn loading to carry the applied load.

4.4.2.2 Tensile modulus

The average tensile Young's modulus of specimens tested in both warp and weft directions from these composites are shown in Figure 4.11.

As with tensile strength, tensile Young's modulus of warp orientated specimens from composites that contained plies of woven reinforcement stacked in the same orientation increased as weft yarn Tex decreased. The Young's modulus of warp orientated specimens from composite 3A were on average over double the tensile Young's modulus of warp orientated specimens from composite 1A. Similar to the trends recorded for the tensile strength of composites, the Young's modulus of weft orientated specimens decreased as weft yarn Tex became lower within composites that were reinforced with plies stacked in sequence 'A'.

Weft orientated specimens from composites 1B, 2B and 3B appear to have similar averaged tensile Young's moduli (6.9, 6.4 and 6.63 GPa, respectively). It might be expected that as weft yarn Tex reduces, the tensile Young's modulus of weft orientated specimens from these composites would decrease. The central plies of these composites, as mentioned previously, contain warp yarns that are parallel to the applied load during tensile testing. As weft yarn Tex reduces, the warp yarns in the central and outer plies for that matter are becoming straighter as the crimping is not so severe, thus the warp yarns are progressively becoming a better reinforcement, especially the warp yarns located in the central ply as warp yarns located in the outer plies are transverse to the load. It is thought that the tensile Young's modulus of weft orientated specimens from composites 1B, 2B and 3B were similar and did not decrease in modulus, unlike the weft orientated specimens from composites 1A, 2A and 3A because of the improved warp yarn architecture within central plies.

As the weft yarns within outer plies become thinner and therefore weaker, the warp yarns in the central plies are counteracting these negative effects by becoming straighter and adding to the stiffness of the specimens, thus the average tensile moduli of these composites are similar. It also is possible that as the weft yarn Tex reduces, the amount of resin penetration into the weft yarns is also increasing, thus improving the tensile modulus.

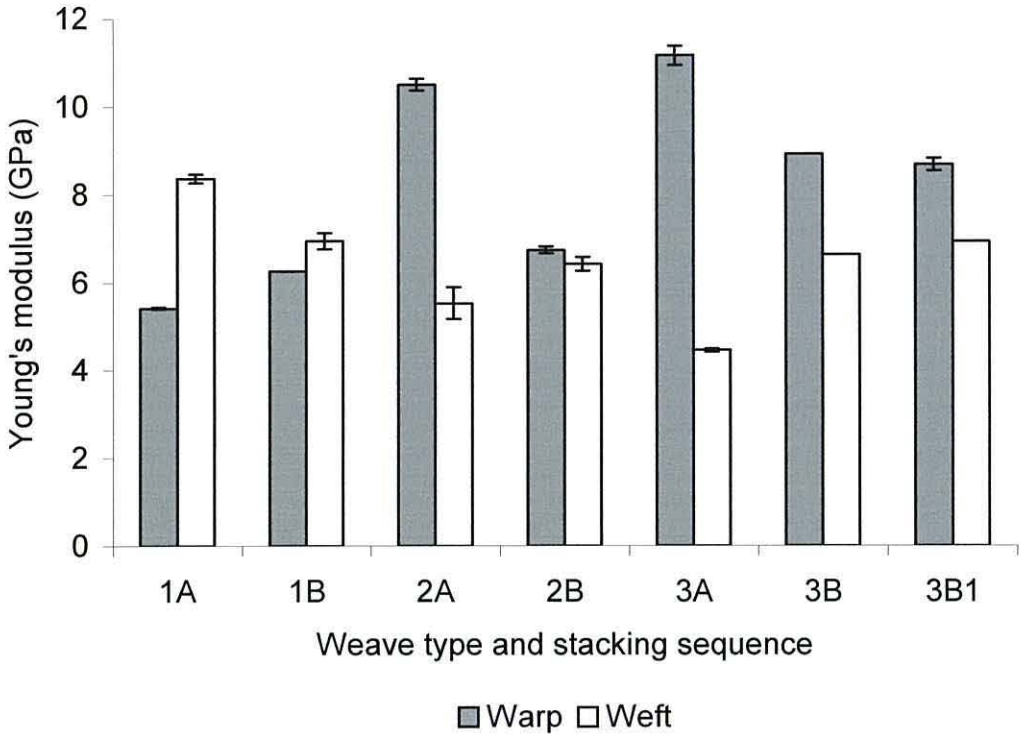


Figure 4.11 The average tensile Young's modulus of specimens tested in both warp and weft directions from composites reinforced with woven flax fabrics containing different yarn sizes and stacking sequences. Table 4.2 on page 171 identifies yarn sizes and composite stacking sequence. Young's modulus of unreinforced resin is 3.72 GPa.

Hepworth *et al.,* (2000) showed that it is possible to increase the tensile modulus of a flax-epoxy unidirectional composite by increasing the 'co-operation' (stress-transfer) between individual flax fibres within a fibre bundle by achieving better epoxy resin penetration which then locks the structure. This was achieved *via* a fibre treatment using urea. Urea may affect the hydrogen bonding between microfibrils. This allowed more water to enter and swell cell walls. Replacing the water with alcohol prevents the cell walls from shrinking. Catalysed epoxy resin (S.P. Systems Ampreg 26) was able to penetrate *via* micro-pores in the cell wall that had been swelled with urea to a sufficient size to accommodate the polymer chains. Hepworth *et al.*, (2000) found that the tensile modulus of the urea fibre treated composites was approximately 30% higher than the

tensile modulus of untreated flax fibre composites. Epoxy resin may be penetrating well into the interior of yarns and encompassing a large quantity of the fibres, however it is the changes to the architecture of the reinforcement that are thought to account for the differences in tensile moduli observed between these composites.

All composites in this study had a tensile Young's modulus which was greater than the tensile Young's modulus calculated from the tensile stress-strain curves of unreinforced epoxy specimens. Weft orientated specimens from composite 3A had the lowest tensile Young's modulus out of all the other woven flax reinforced composites; its tensile modulus was 20% higher than the tensile modulus of the unreinforced epoxy resin.

4.4.2.3 Nature of stress-strain behaviour

Figure 4.12 shows four representative tensile stress-strain curves, two are from warp orientated specimens from composites 1A and 1B and the other two stress-strain curves are from weft orientated specimens obtained from composite 1A and 1B.

Two representative stress-strain curves from warp orientated specimens obtained from composites 3A and 3B are presented in Figure 4.13, also shown are two representative weft orientated specimens stress-strain curves from composites 3A and 3B.

All stress-strain curves presented in Figure 4.12 and Figure 4.13 have a similar shape, although the initial moduli, maximum stress and sometimes strain at failure are different. The curves show relatively small regions of linear behaviour, followed by the onset of non-linear behaviour which extends for a relatively long period of strain until failure. It is interesting to compare the warp tensile stress-strain curves from Figure 4.12 to the warp tensile stress-strain curves of woven flax reinforced polyester composites presented in Figure 3.11 on page 144. The stress-strain curves presented in Figure 3.11 have a much more distinct yielding point than the curves in Figure 4.12. The abrupt yielding

seen in Figure 3.11 was thought to have been caused by matrix cracking and subsequent debonding of yarns.

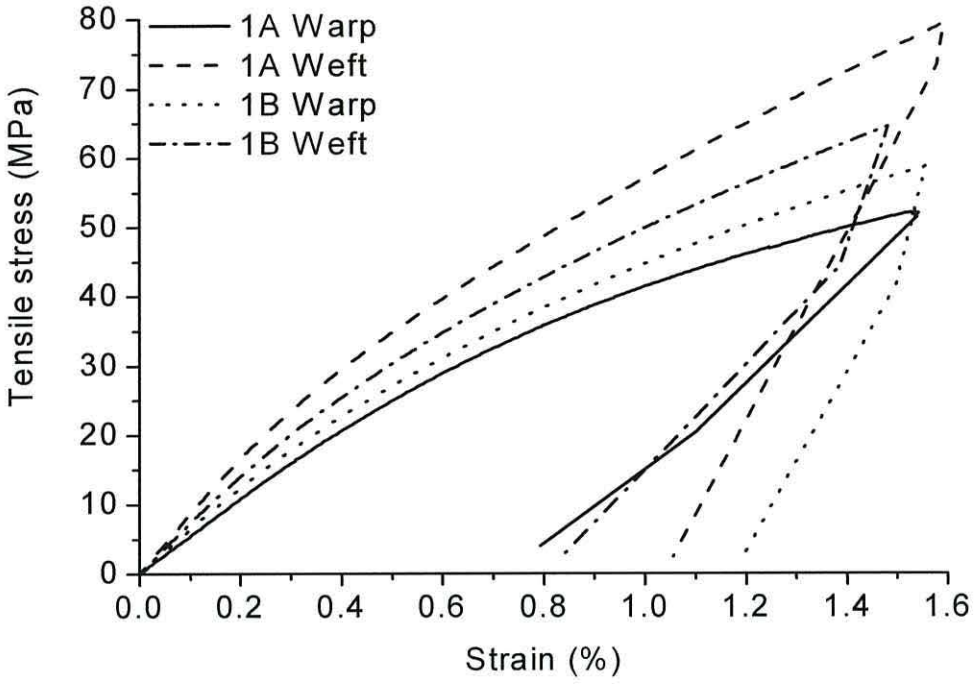


Figure 4.12 Representative tensile stress-strain curves of specimens tested in both warp and weft directions from composites (1A and 1B) that are reinforced with woven flax fabric and stacked in two sequences (A and B).

As the epoxy resin has a much higher strain to failure than the polyester resin used in Chapter 3, it is thought that the yielding seen in Figure 4.12 is mainly caused by the architecture of the reinforcement. As the tensile specimens are strained the matrix is able to deform and the yarns within the matrix are realigning (extending) with it. As Figure 4.12 shows, differences in modulus and maximum tensile stress between warp and weft orientated specimens from composite 1A are greater than they are for warp and weft orientated specimens from composite 1B. However, the strain at which failure occurs is quite similar for all specimens from both types of composite. As previously described,

the matrix may have failed prior to the yarns or *visa versa*. As there are no deviations occurring on the tensile stress-strain curves it indicates that failure was instantaneous and it occurred in a brittle manner, which indeed it was and it did.

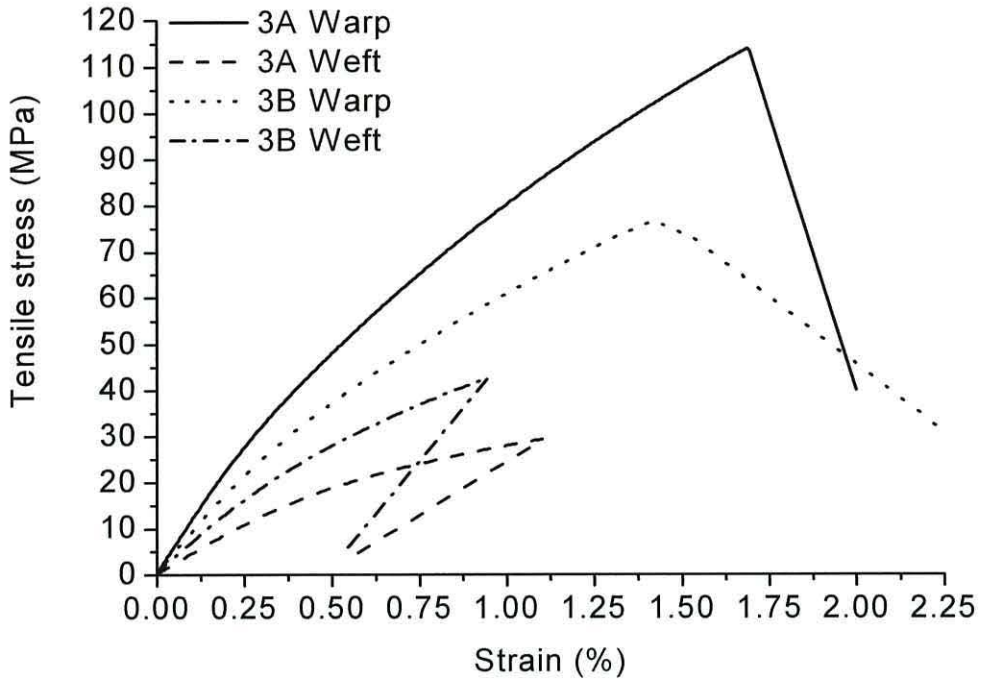


Figure 4.13 Representative tensile stress-strain curves of specimens tested in both warp and weft directions from composites (3A and 3B) that are reinforced with woven flax fabric and stacked in two sequences (A and B).

Figure 4.13, the weft orientated specimens from composites 3A and 3B have failed at lower strains than the warp orientated specimens from composite 3A and tensile weft orientated specimens from composite 1A and 1B. As the weft orientated specimens from composites 3A and 3B contain lower Tex weft yarns which are parallel to the direction of the load it indicates that perhaps yarn failure precedes matrix failure.

All tensile specimens from the seven composites, failed in a brittle manner. The tensile specimens were completely separated in two pieces leaving two square fractured surfaces. No evidence of shear failure was visible and the remainder of the specimens visually appeared to be undamaged. The fractured warp yarns were visible from failed warp orientated specimens. Fractured weft yarns from weft orientated specimens were also visible from composites 1A and 1B. Yarn pull out from these composites was minimal; the fractured yarns barely protruded from the level of the fractured surface. The level of warp yarn pull-out was the same for warp orientated specimens from composites 2A, 2B, 3A and 3B. However, the fractured weft yarns from weft orientated specimens from composites 3A, 3B and 3B1 were flush with the fractured surfaces. The higher Tex warp yarns were clearly seen on the fractured surfaces, spanning the width of the specimens. The transverse warp yarns near/on the fractured surfaces of weft orientated specimens from composite 2A had pulled out to some extent during the fracture. This had also been observed with the warp yarns transverse to the applied load in the outer plies of weft orientated specimens from composite 3B1.

4.4.3 Impact properties

Figure 4.14 shows the average Charpy impact strengths of warp and weft orientated specimens from these composites.

As warp yarn crimping became less severe, because of the reduction in weft yarn Tex, the Charpy impact strength of warp orientated specimens increased; this can be seen when comparing results for composites 1A, 2A and 3A. As previously noticed with other properties, as weft yarn Tex reduces, the weft Charpy impact strength also decreases, this is also demonstrated in composites 1A, 2A and 3A.

Apart from the warp orientated specimens from composite 1A, composites 2A and 3A had slightly higher average Charpy impact strengths than the average Charpy impact strengths of warp orientated specimens from composites containing the same weave type

as reinforcement but stacked in sequence 'B'. The stacking of reinforcement with a middle ply at 90° to the top and bottom plies (sequence 'B') only benefits the weft orientated impact properties, the differences between these composites is only significant when comparing weft orientated specimens from composite 3A to weft orientated specimens from composite 3B and 3B1.

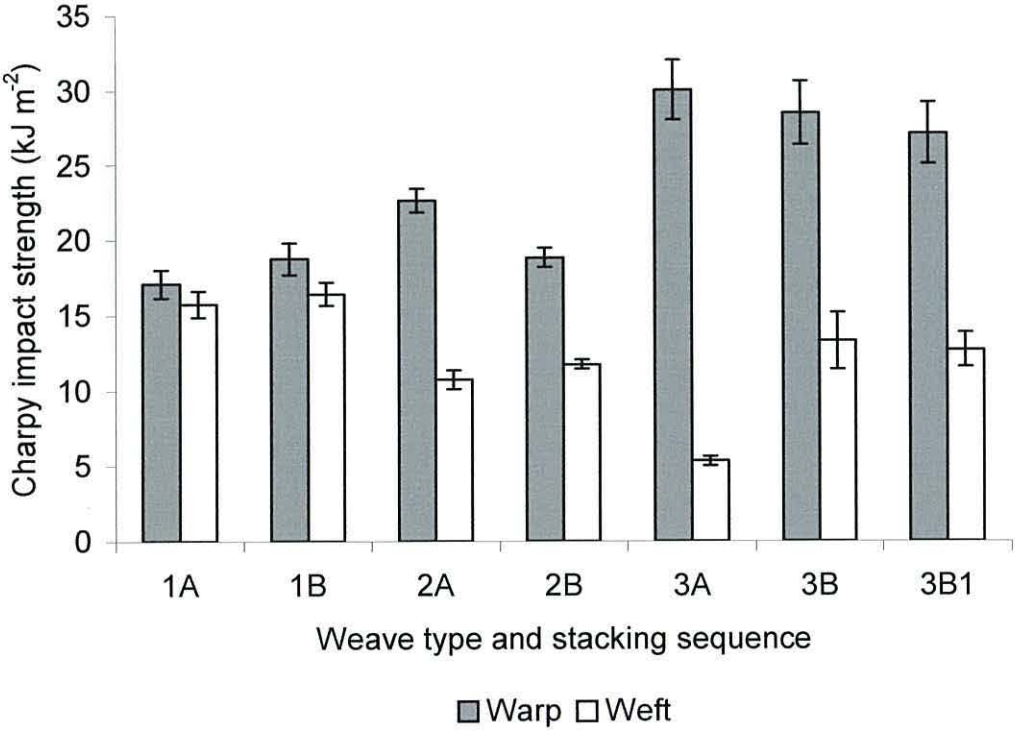


Figure 4.14 Averaged Charpy impact strengths of specimens tested in both warp and weft directions from composites reinforced with woven flax fabrics containing different yarn sizes and stacking sequences. Table 4.2 on page 171 identifies yarn sizes and composite stacking sequence. Charpy impact strength of unreinforced resin is 43.6 kJ m^{-2}.

The Charpy impact strengths of all the specimens are well below the average Charpy impact strength achieved by the unreinforced epoxy resin by at least 45%. It is thought that the fibre loadings within the composites are insufficient to improve upon the cast epoxy resins resistance to the propagation of cracks. The inclusion of fibre to the

polymer has introduced flaws such as voids and created stress-concentrations that cause cracking to occur at lower strains than they may occur in unreinforced matrix material.

The differences in Charpy impact strengths between composites are more likely to be due to a number of factors, one being the size of unreinforced matrix regions between plies of reinforcement and between yarns from the same ply. As Plate 4.1 shows, the empty space between weft yarns from the three different weave types increases as weft yarn Tex reduces, thus the size of the unreinforced resin pockets between weft yarns in composites that are reinforced with weave type 3 have the potential to be larger. The Tex of the yarns also influences the composite's resistance to the propagation of cracks. It is thought that cracks propagating during catastrophic failure are mostly passing though the flax fibre yarns and not being deflected or blunted at the yarn/matrix interface. The amount of energy absorbed for a crack to propagate through a higher Tex yarn (which contains a greater number of fibres) is probably more than the amount of energy absorbed when a crack propagates through a yarn that has a lower Tex and therefore contains less individual flax fibres. Obviously there are differences in the number fibre/matrix interfaces in yarns that are spun to different Tex.

Warp and weft orientated impact specimens from all composites fractured completely through the thickness of the specimen during the impact test. The failure occurred in a brittle manner. The fractured surfaces of the specimens were sometimes square (90° to the top and bottom surfaces of the specimen) or sometimes slightly sloping. There did not seem to be any trend between specimens from different composites with respect to the fractured surface angle. Fractured warp yarns from composites 1A and 1B slightly protruded from the fractured surface; they felt rigid, indicating that they did contain cured resin. Warp orientated specimens from composites 2A, 2B, 3A and 3B also had similar warp yarn fracture, just above the fractured surface of the matrix (minimal yarn pull-out). Fractured weft orientated specimens from composite 1A also had weft yarns that had fractured slightly away from the fracture surface of the matrix. Weft orientated specimens from composite 1B showed both warp and weft yarn fracture on the fractured surface of the specimens. However, it is noticeable that the warp yarns within the middle

ply protruded further from the fractured surface than the weft yarns above and below in the outer plies. The limited amount of fibre/yarn pull-out and the apparently good bond between fibres and matrix are thought to have contributed to the woven flax epoxy composite low toughness.

4.5 Summary

The Tex of weft yarns within woven flax fabrics influences the maximum loads achieved at failure and amounts of tensile extension exhibited by woven flax fabrics when tested in the warp direction. As weft yarn Tex reduces; woven flax fabrics warp orientated maximum load at failure increases whilst the tensile extension decreases. In contrast, the woven flax fabrics weft orientated maximum load at failure decreases as weft yarn Tex is reduced. This trend is shown in epoxy composites reinforced with three plies of woven flax fabric stacked either in the same direction or with a central ply at 90° to the top and bottom plies in the following ways. The warp orientated flexural and tensile properties of composites increase as weft yarn Tex is reduced. This occurs for composites containing woven flax plies stacked in the same direction and composites reinforced with a central ply at 90° to the top and bottom plies. Composite's weft orientated flexural moduli and strength along with tensile strength and moduli decrease as weft yarn Tex in the plies of reinforcement fabrics is reduced. The warp and weft flexural strengths of composites reinforced with a middle ply at 90° to the top and bottom plies were very similar to the strengths observed from equivalent composites containing reinforcement stacked in the same direction. However, a significant difference between the warp flexural moduli of composites containing all three plies of woven flax reinforcement in the same direction and composites reinforced with a central ply at 90° to the top and bottom plies was observed. Composites containing a middle ply at 90° exhibited better warp flexural moduli. In contrast, no significant differences exist between weft flexural moduli and the two types of reinforced composites. The composite that had the most similar warp and weft flexural moduli was one with reinforcement plies consisting of warp yarns that were 208 Tex and weft yarns that were 83 Tex and each ply of reinforcement was stacked in

the same direction. Stress-strain curves of flexural specimens from all composites displayed linear and non-linear behaviour. An increase in modulus was observed to occur for all warp orientated flexural specimens and some weft orientated specimens obtained from both types of reinforced composite as further stress was applied. The increase in modulus was not sustained and stress-strain curves did yield as further stress was applied.

Weft tensile strengths of some composites (reinforced with fabrics than were woven with the weft yarns at a lower Tex than adjacent warp yarns) reinforced with a central ply at 90° were higher than the weft tensile strengths of equivalent composites reinforced with woven flax plies in the same direction. Although, the warp tensile strength of composites reinforced with all three plies of woven flax in the same orientation was higher than the warp tensile strength of composites reinforced with a central ply at 90°. The weft tensile moduli of composites reinforced with a central ply at 90° to the top and bottom plies were similar between the three different types of woven flax reinforcement used. Improved alignment of warp yarns within the central ply of reinforcement as weft yarn Tex reduced is thought to be responsible for the similar moduli observed, as the warp yarns are not as severely crimped and therefore act as better reinforcement, counteracting the weaker weft yarns in the outer plies. The composite reinforced with woven flax plies consisting of warp yarns at 208 Tex and weft yarns at 83 Tex, stacked with a central ply at 90° to the top and bottom plies exhibited the most similar warp and weft tensile moduli and strength. Tensile stress-strain curves, showed relatively small regions of linear behaviour. The gradual onset of non-linear behaviour was observed.

As weft yarn Tex was reduced in woven flax fabric reinforcement, warp Charpy impact strength of epoxy composites increased. This occurred for both types of reinforced composites but the increase was more noticeable in composites containing reinforcement stacked in the same direction. Weft Charpy impact strength decreased as weft yarn Tex was reduced, this occurred for both types of reinforced composite. The warp and weft Charpy impact strength of a composite was most similar when the reinforcing plies were the same Tex and the three plies were stacked in the same direction.

In summary, the warp and weft mechanical properties of woven flax fabric reinforced epoxy composites are influenced by the Tex of the weft yarn reinforcement because of their effect upon warp yarn crimping; in addition, simple stacking sequences of woven flax fabric reinforcement can also influence these effects and alter properties of composites in both a positive or negative manner.

5 WEAVE ARCHITECTURE AND THE INFUENCE ON MECHANICAL PROPERTIES

5.1 Introduction

The following experimental work was designed to explore the influence that the weave type of woven flax reinforcement has upon the mechanical properties of a composite. The aim was to compare and analyse the mechanical properties and deformation behaviour of laboratory fabricated epoxy matrix composites that were reinforced with 12 different woven flax fabrics.

5.2 Materials and method

5.2.1 Resin

An epoxy resin (Ampreg 20) was utilised for the manufacture of all woven flax fabric composites reported in the following chapter. Table 4.1 on page 170 details the supplier's physical and mechanical properties of Ampreg 20 epoxy resin when mixed with a slow hardener. Properties of the epoxy resin were determined for a previous experiment, which are presented in Table 4.3 on page 179.

5.2.2 Woven flax fabrics

Ferguson's Irish Linen (Thomas Ferguson & Co. Ltd.) based in Co. Down, Northern Ireland produced and supplied all woven flax fabrics. Table 5.1 details the 12 woven flax fabrics that were used to reinforce composites (1 to 12).

Table 5.1Woven flax fabrics showing weave types and yarn size (Tex).

Identification (Weave type)	*Weave*	*Warp (Tex)*	*Weft (Tex)*	*Warp fabric count (yarns/inch)*	*Weft fabric count (yarns/inch)*
1	$^{1}/_{1}$	83.3	83.3	32	37
2	$^{1}/_{2}$	83.3	83.3	33	36
3	$^{1}/_{3}$	83.3	83.3	33	34
4	$^{1}/_{4}$	83.3	83.3	30	36
5	$^{1}/_{5}$	83.3	83.3	30	35
6	$^{1}/_{6}$	83.3	83.3	32	33
7	$^{1}/_{7}$	83.3	83.3	31	32
8	$^{1}/_{8}$	83.3	83.3	32	32
9	$^{1}/_{9}$	83.3	83.3	29	32
10	$^{1}/_{10}$	83.3	83.3	30	34
11	$^{1}/_{1}$	66.6	83.3	34	38
12	Honeycomb	83.3	104.1	32	32

As Table 5.1 shows, the first weave type is a plain weave ($^{1}/_{1}$). In this weave type, a warp yarn passes over a weft yarn then below a weft yarn then over the next, and so on. The second type is a $^{1}/_{2}$ weave, the warp yarns pass over 2 weft yarns then under a weft yarn, then over 2 again, and so on. This incremental increase of the warp yarns passing over an extra weft yarn continues until weave type 10 ($^{1}/_{10}$) in which the warp yarns pass

over 10 weft yarns then under a weft yarn, then over 10 again, and so on. This increase in the number of weft yarns that the warp yarns skip over could potentially be greater, but this would be detrimental to the fabric's integrity and stability. Table 5.1 also shows that the warp and weft yarns in fabrics 1 to 10 were the same size at approximately 83 Tex. Weave type 11, a plain weaved ($^{1}/_{1}$) fabric was slightly different to weave type 1 as it contains warp yarns that have a lower Tex (thinner).

Weave type 12 also contained different sized weft yarns and was woven to form a honeycomb weave. This consisted of a 10 warp yarn by 10 weft yarn block repeating pattern. Within this, there were four 5 yarn by 5 yarn sub-blocks, of which 2 diagonally opposite were a $^{1}/_{1}$ weave type and in the other two sub-blocks all the fibres skip over or under for 5 yarns. Within the sub-blocks that were not plain weaved, two warp yarns were on the surface of the fabric, passing over three of the weft yarns which pass over the other three warp yarns, with the other two weft yarns on the base. This means that in these sub-blocks the fabric consisted of 4 layers, thus making the fabric thicker in these regions and giving a 'bumpy' feel as the other sub-blocks were much thinner/smoother because they were essentially plain weaved. An overview and close-up photo of the fabrics and their yarns is presented in Plate 5.1. 'Fabric count' is a measure of the number of yarn ends per inch of fabric. Table 5.1 shows the fabric counts for both warp and weft ends. However, although these counts were made from representative regions of the fabrics they may contain variation that has gone unnoticed as it was only counted once. No information was obtained from Fergusons Irish Linen regarding fibre/yarn treatments apart from confirming that a size has been applied to warp yarns to facilitate weaving.

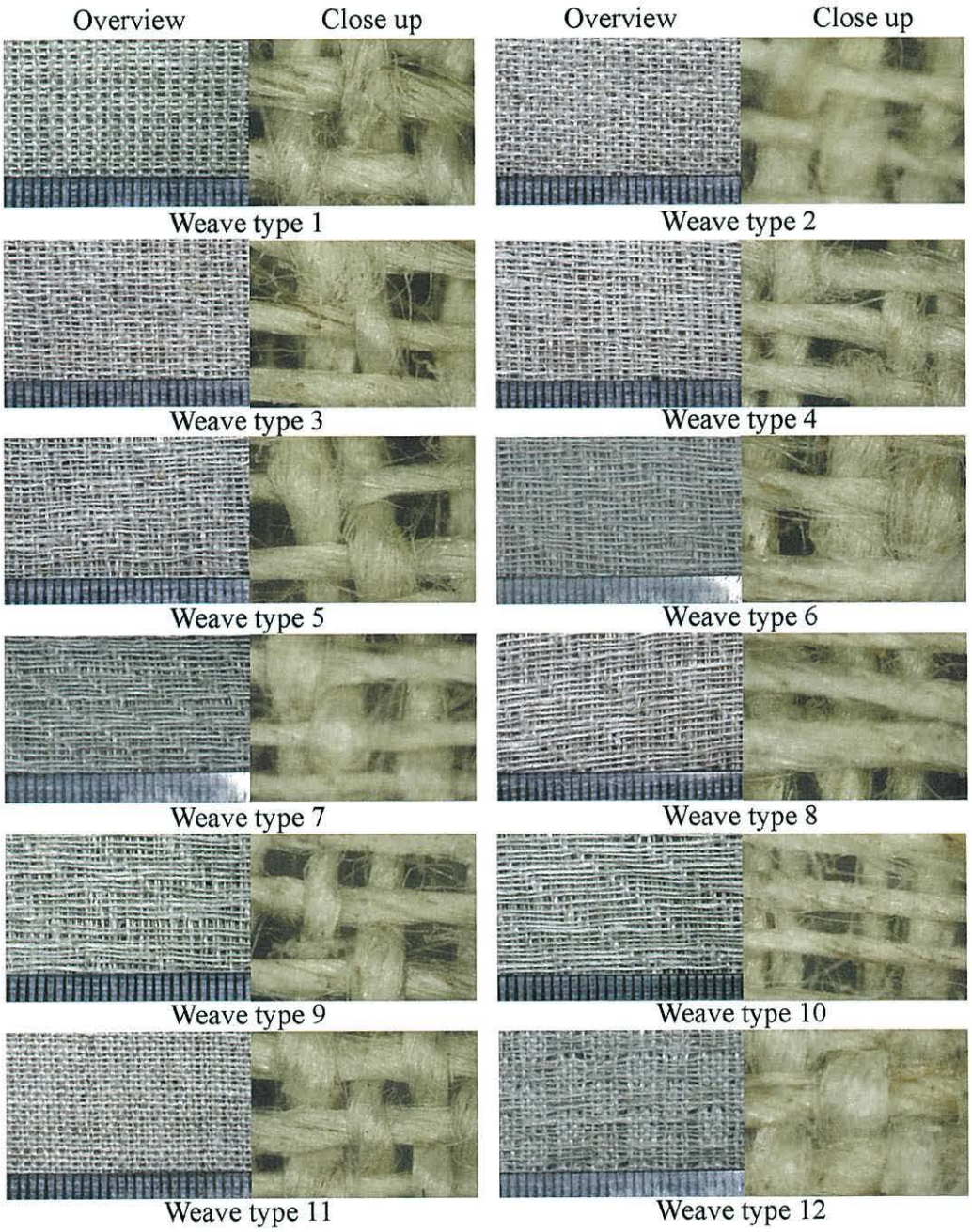


Plate 5.1 Overview and close up photo (approx 3mm width) of the 12 different weave types of woven flax fabric.

5.2.2.1 Evaluating the tensile properties of woven flax fabrics

The same method as reported in Section 3.2.2.1 on page 109 was used to evaluate the tensile properties (maximum force and elongation at maximum force) of each woven flax fabric.

5.2.3 Woven flax composite manufacture

All 12 woven flax fabrics were used to reinforce composites measuring approximately 260 mm in length and 195 mm in width with varying thicknesses, ranging from 2.65 (weave type 11) to 4.09 mm (weave type 12). The thicknesses of composites reinforced with weaves 1 to 10 ranged from 2.79 to 3.36 mm. From each of the 12 woven flax fabrics 5 reinforcement plies were cut, all with the same orientation and measuring 260 mm in length (warp direction) and 195 mm in width (weft direction). The plies were then ironed to remove creases in the fabric. The 5 plies for each composite were then stacked all in the same orientation ($[0°]_S$) and weighed on an electronic balance ready for resin impregnation.

5.2.3.1 Resin impregnation into woven reinforcement

Resin preparation was conducted with the same method as described in Section 4.2.3.1 on page 173. Impregnation of catalysed epoxy resin into the stacks of woven flax fabrics was essentially conducted using the same methods as described in Section 3.2.4.2 on page 112. However, although a hand roller was used over the surface of the polythene tube for all the composites to assist resin flow, very little pressure was exerted on the stack of reinforcement and resin was not squeezed from the stack of woven reinforcement.

5.2.3.2 Moulding and curing

Composites were essentially moulded by the same method as detailed in Section 3.2.4.3 on page 113. However, no G-clamps were used and the only pressure exerted onto the resin saturated stacks of woven flax fabric was the weight of the top glass plate. As the top glass plate was placed onto the stack, excess resin would seep from the edges. Composites were left at approximately 20°C for 24 hours before being released from the mould. After release from the mould all composites underwent a post cure in an oven set at 50°C for 16 hours.

5.2.4 Measurement of composites

The composites were measured using the same approach as described in Section 4.2.5 on page 174.

5.2.5 Specimen preparation

Tensile, flexural and impact specimens were required from composites. Specimens were obtained from the composites using a fine-toothed band saw; approximately 1 cm of composite material was trimmed from every composite edge before cutting of specimens commenced.

5.2.5.1 Conditioning

Specimens were conditioned prior to testing at 65% relative humidity at a constant temperature of 20°C for a minimum of one week.

5.2.5.2 Measurement of specimens

The measurement of specimens was conducted as described in Section 3.2.8.2 on page 116.

5.2.6 Testing

Flexural (100 mm by 15 mm) and unnotched Charpy impact specimens (80 mm by 10 mm) were obtained in both warp and weft directions from all composites. However, only warp tensile specimens could be obtained because of the composite's size. This was considered when fabricating composites, but due to the limited quantity of fabric the size of composites had to be limited.

5.2.6.1 Flexural

A minimum of 5 specimens from both warp and weft orientations were tested from each composite. Flexural tests were preformed following the same procedure as detailed in Section 3.2.9.1 on page 117.

5.2.6.2 Tensile

Two warp tensile specimens were tested from each of the composites (warp yarns are parallel to the length of the specimens). Tensile testing was preformed following the same procedure as detailed in Section 3.2.9.2 on page 118.

5.2.6.3 Impact

A minimum of 7 warp specimens and 7 weft specimens were tested for each woven flax reinforced epoxy composite. However, 9 impact specimens were usually tested in both orientations. Impact testing was conducted with the same method as described in Section 3.2.9.3 on page 119.

5.2.7 Fractography

The modes of failure were examined using a dissecting microscope. Fractured tensile specimens were examined using scanning electron microscopy. Fractured surfaces were cut away from the specimen using a fine toothed band saw. This left the fractured surface with approximately 5 mm of composite material. The flat cut composite material was then secured to aluminium stubs with conducting epoxy adhesive, leaving the fractured surface exposed. The samples were dried in an oven set at 100°C for a few hours before being placed over silica gel for 24 hours. The samples were splutter coated using a Polaron E5000 set to 1.2kV and 10mA. The samples were coated in gold from a pure gold target for 2.5 minutes. A Hitachi S-520 scanning electron microscope (SEM) was set to 12kV and used at various magnifications to record the fractures.

5.2.8 Evaluation of physical properties

The densities of composites were determined using Equation 3.1 on page 120. The densities of all specimens were also calculated using Equation 3.1. Composite fibre volume fractions were calculated using Equation 2.12 on page 62.

5.3 Results and discussion

5.3.1 Tensile properties of woven flax fabrics

The average number of yarns for both warp and weft fabric specimens for weaves 1 to 12 can be found in Table 5.2.

Table 5.2 Average numbers of yarns present in woven flax fabric warp and weft fabric tensile specimens.

Identification	*Average number of warp yarns*	*Average number of weft yarns*
1	31	35
2	33	35
3	33	32
4	32	35
5	29	34
6	31	32
7	28	31
8	27	32
9	28	31
10	28	31
11	33	35
12	30	31

The average maximum loads at failure for weaves 1 to 12 are presented in Figure 5.1 whilst Figure 5.2 shows the tensile extension at maximum load from weaves 1 to 12 for warp and weft specimens.

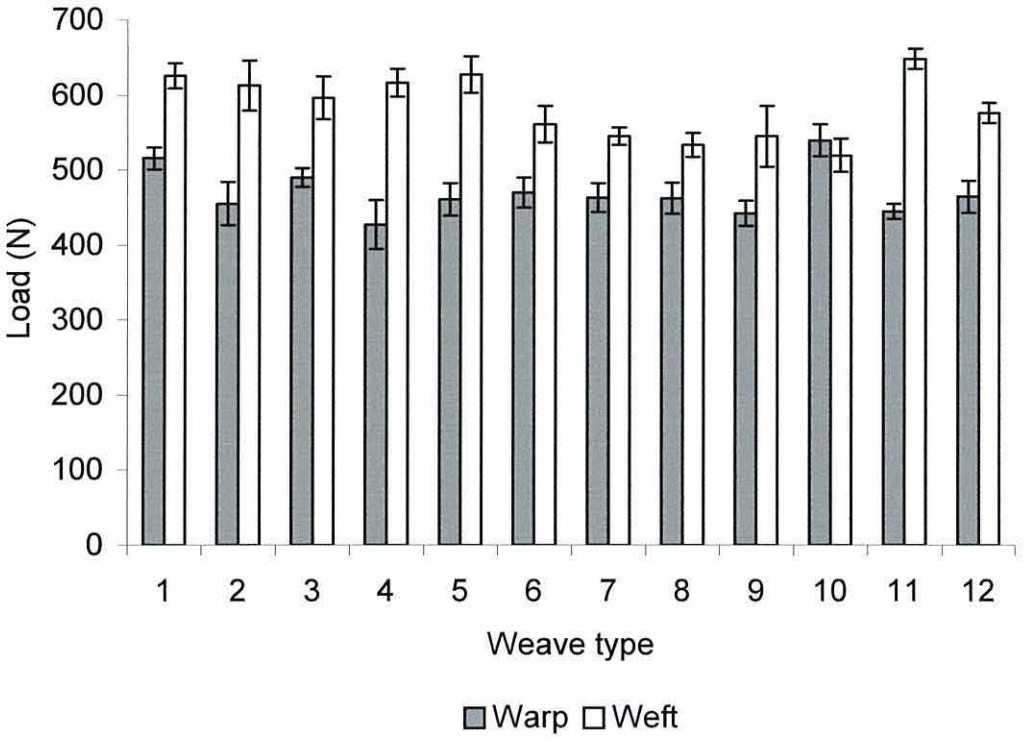


Figure 5.1 The average maximum load at failure of 12 different woven flax fabrics tested in warp and weft directions. Weave type corresponds to Table 5.1.

As with the majority of weave types, types 11 and 12 also exhibited large differences between the failure loads of warp and weft yarns. This is not surprising, as weave type 11 had thinner warp yarns, and thus contained less fibre which were crimped highly because they were woven into a plain weave. Weave type 12 is woven with thicker weft yarns and the warp yarns are crimped highly in certain regions which may also affect the failure load.

Figure 5.2 shows that weave types that contained warp yarns that are highly crimped such as $^{1}/_{1}$ and honeycomb weaves had high tensile extensions at maximum load. As the amount of warp yarn crimping is reduced the extensions of the yarns gradually become similar to the un-crimped weft yarns. It is also interesting to note that the tensile extensions of the weft tested specimens remained relatively similar (weave types 1 to 10).

As with the maximum load at failure of the weft tested fabric specimens, the tensile extension at failure of weft yarns does not seem to be influenced by the amount of warp yarn crimping.

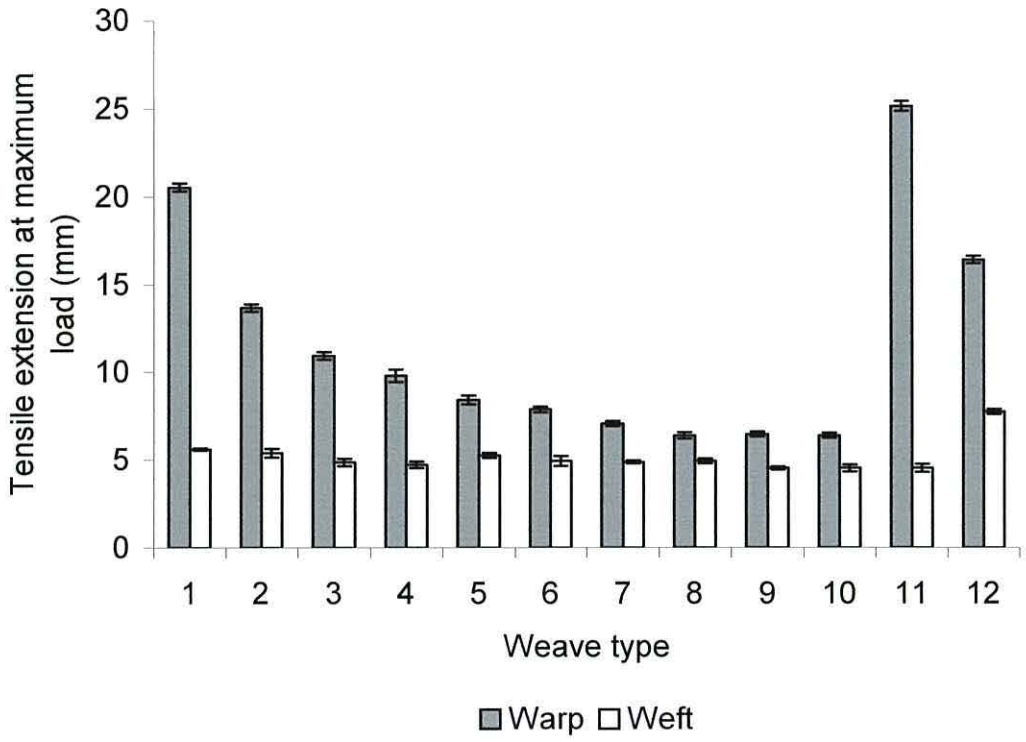


Figure 5.2 The average tensile extension at maximum load of 12 different flax fabrics tested in warp and weft directions. Weave type corresponds to Table 5.1.

Figure 5.1 clearly shows that there is a significant difference between the warp and weft specimens load at failure for the majority of weave types. Generally, weft specimens display higher maximum loads than their warp specimen's counterparts, even though for weave types 1 to 10 their yarns are the same Tex. The difference between the maximum loads at failure for warp and weft specimens does appear to decrease, this can be observed in weaves where the warp yarns become less crimped and therefore pass over more weft yarns. An extreme example of this is weave type 10. The warp specimens from this weave type have a slightly higher average maximum load at failure than the

weft specimens. It could be argued that the differences observed between warp and weft specimens from the same weave types may be because the number of yarns in weft specimens is greater than the number of yarns in the warp specimens. In many cases, this is true and it is possible that this would cause such a result. However, weave type 3 had an average of 33 warp yarns present in warp fabric tensile specimens and weft fabric tensile specimens contained an average of 32 weft yarns. The weft fabric tensile specimens failed at a significantly higher load. Also the difference between the numbers of yarns in either the warp or weft specimens is not large. Using weave type 10 again as an example, the warp specimens contained an average of 28 warp yarns which is three yarns less than the weft tested specimens, the warp specimens failed at a higher load.

There is no significant difference between weave types 1 to 5 when comparing the average maximum load at failure for weft tested specimens. However, comparing the weft properties of these 5 weave types to weave types 6 to 10 there does appear to be a difference between the weft specimen's maximum load at failure. Performing a 't-Test' on the averages it was established that a significant difference does exist between the two groups of weft specimens, the maximum loads at failure of specimens 1 to 5 are significantly higher than specimens 6 to 10 at a 95% confidence interval. However, this is not because the weft test specimens for weaves 1 to 5 contain stronger weft yarns than the others, it is more likely due to the fact that the number of weft yarns in weft tested specimens from weaves 1 to 5 was greater. For specimens 1 to 5 it was on average 34.2 yarns which was greater than it was for weaves 6 to 10, as the average weft yarn count in test specimens was 31.4. In this case, it is thought that the number of weft yarns in specimens is responsible for the differences between weave types 1 to 5 and 6 to 10, as weft yarns are not crimped and theoretically should all be very similar, as care was taken to ensure that all fabric specimens were placed into the grips with aligned yarns. Comparing the warp specimen's maximum load at failure between fabric types 1 to 9, there are not any large differences between weave types, especially when taking into account the average number yarns within the weave type test specimens.

5.3.2 Physical properties of composites

5.3.2.1 Fibre volume fraction and density

Table 5.3 reports the fibre volume fractions and densities of composites fabricated with the woven flax fabrics shown in Plate 5.1 on page 216.

Table 5.3 Fibre volume fractions and densities of composites reinforced with different weaved woven flax fabrics.

Identification	V_f *(%)*	*Density (kg* m^{-3}*)*
1	27.0	1223.7
2	28.4	1209.7
3	26.5	1211.2
4	26.0	1200.8
5	23.3	1204.8
6	22.1	1213.9
7	27.2	1165.8
8	25.1	1204.2
9	27.4	1199.2
10	24.1	1213.4
11	30.1	1246.3
12	23.7	1195.6

The fibre volume fractions presented are not definitive values, as it is extremely difficult to calculate these accurately (as discussed in Section 3.3.4.3 on page 126). As one of the aims of this experiment was to fabricate the sets of composites with similar fibre volume

fractions, the range between composites shown in Table 5.3 is relatively small; the lowest fibre volume fraction is approximately 22% and the highest 30%. Throughout the chapter the observed properties of the composites are presented. However, the fibre volume fraction was arbitrarily set at 26% for all composites as this is approximately the mid value between the ranges of fibre volume fractions. By adjusting the composite's properties accordingly (increasing property if composite has a fibre volume fraction below 26% or decreasing property if composite has a fibre volume fraction above 26%) it was observed that little change occurred and the same trends between composites existed as with the observed results, therefore normalising composite properties to a single fibre volume fraction did not dramatically influence the results. This approach has been previously discussed in Section 4.3.3.1 on page 182 for a similar set of composites and was not utilised due to assumptions that have to be made in order to adjust composite properties to a different fibre volume fraction.

The densities of the woven flax reinforced composites shown in Table 5.3 range from 1165 to 1246 kg m^{-3}. There is a great deal of variation between these densities. These differences may be due to the void content of composites, the size of resin rich regions (matrix pockets) between plies or yarns and the fabric count of the reinforcement. For example, composite 11 had the highest density; it also had the highest calculated fibre volume fraction and the highest fabric count. Although its warp yarns were spun to a slightly lower Tex, (thus containing less fibre than the other warp yarns) weave type 11 did have slightly more yarns per inch of fabric when compared to the other weave types. The smaller warp yarns may have enabled the plies of reinforcement to stack closer together and therefore reduce the size of resin rich areas between them. Composite 11 was the thinnest composite at 2.65 mm. In contrast, composite 7 had the lowest density, it did not have the lowest fibre volume fraction but its fabric count was one of the lowest. As the density of the resin is likely to be lower than the fibre density, it is possible that the lower density of the composite is caused by large resin rich regions between yarns, as there are fewer yarns per inch. Composite 1 had a high fabric count, and its density is considerably above the average of all the composites densities (average of all composites 1207 kg m^{-3}). Composite 12 had the second lowest density and this is thought to be due

to the architecture of the reinforcement, large matrix pockets may be present between plies, as this was the thickest composite at 4.09 mm.

5.4 Mechanical properties of composites containing different weave types

5.4.1 Flexural properties of composites

5.4.1.1 Flexural strength

Figure 5.3 shows the average warp and weft orientated flexural strengths. Figure 5.3 shows that the composites reinforced with weave types 1, 2, 3, 4, 5, 6 and 7 exhibited a significant difference between the flexural strength of warp and weft orientated specimens from the same composite. This anisotropic behaviour between these two test directions gradually reduces (composites 8, 9 and 10).

It is plausible that the differences between warp and weft orientated specimens from the same composite may be due to the amount of twist in the yarns. It is thought that a high degree of twist in a yarn will increase its strength up to a certain limit, beyond which any further increase in will result in a decrease of the yarn strength. However, if the weft yarns in these flax fabrics have a higher yarn twist than the warp yarns it may account for the improved flexural strength in certain composites. Confirmation that the warp and weft yarns in these woven flax fabrics used are twisted to the same degree was obtained from Linda Cantley from Ferguson Irish Linen (*pers.com*).

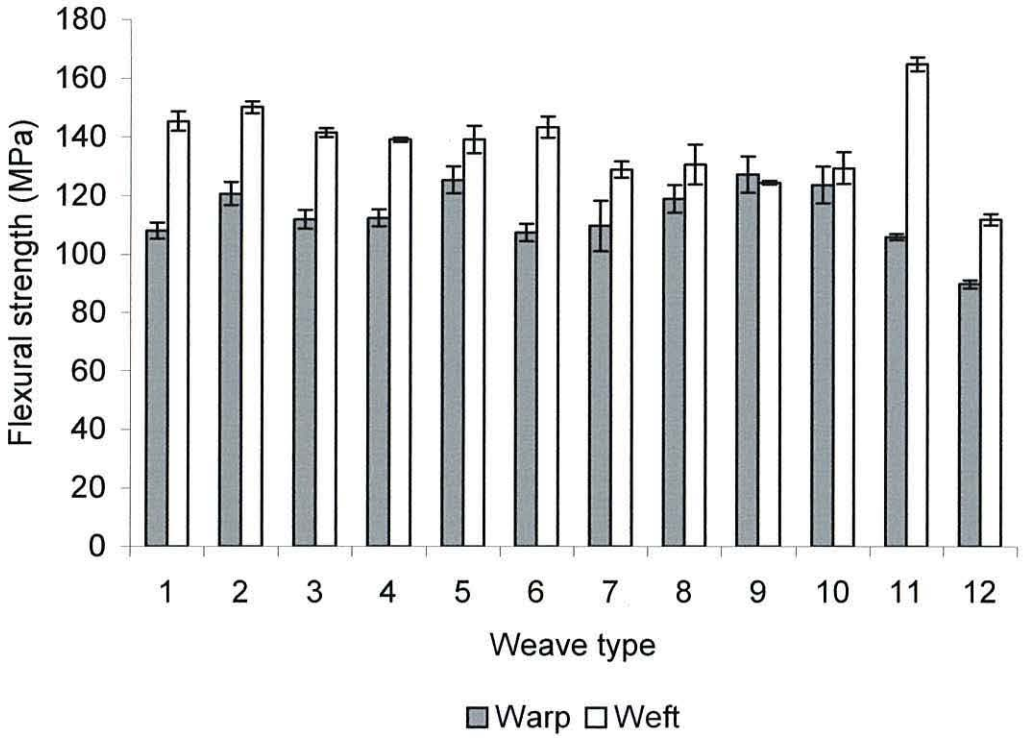


Figure 5.3 The average flexural strength of specimens tested in both warp and weft directions from composites reinforced with 12 different woven flax fabrics. Table 5.1 on page 214 identifies weave type. Flexural strength of unreinforced resin is 134 MPa.

Section 5.3.1 on page 221 mentioned that there was a difference between weave types 1 to 5 and 6 to 10 in terms of the weft maximum loads at failure (Figure 5.1), with weave types 1 to 5 failing at slightly higher loads. It was thought that the differences between these fabrics might be because weave types 1 to 5 had higher weft yarn fabric counts than the following 5 weave types (Table 5.2). A similar result was noticed with composites reinforced with some of these weave types. Composites reinforced with weave types 1 to 6 had higher weft flexural strengths than composites reinforced with weave types 7, 8, 9 and 10. As previously mentioned, the difference between the warp and weft flexural strength in composites 8, 9 and 10 is smaller than other composites. This is likely to be because the warp yarns are crimped less in these fabrics compared to the others (1 to 6)

consequently, the warp yarns are straighter and are a more effective reinforcement, and thus warp oriented specimens have similar properties as the weft orientated specimens. However, it is important to discuss the following possibilities. Weft specimens in composites 7 to 10 are failing at lower stresses than other composites such as 1 to 6, this result could be interpreted in a number of ways. For example, the weft orientated specimens may be failing at a lower stress. This may be because the fabric counts are generally lower or that the amount of warp crimping affects the properties of the fabrics in another manner which is reflected in this composite property.

The integrity of a woven fabric is maintained by the mechanical interlocking of the yarns. A plain weaved fabric ($^{1}/_{1}$) has good stability because the weft yarns are regularly crossed over and under by warp yarns and these hold them securely in position *i.e.* 90° to the warp yarns. The yarns in a woven fabric with a $^{1}/_{10}$ weave are not as interlocked as a $^{1}/_{1}$ woven fabric, therefore the fabric has less stability *i.e.* the yarns may not cross at 90° because they can easily be misaligned. During fabrication of these composites, the woven flax fabrics were ironed and stacked upon each other with care taken to ensure that the warp yarns from each ply were parallel with each other and that the weft yarns were at 90° to the warp yarns in each ply. However, during resin impregnation it is possible that the resin front may have caused some misalignment of yarns within woven fabrics that do not contain a high amount of warp and weft yarn interlacing. If weft yarns are misaligned within the composites because of the liquid resin flow then it would explain the lower flexural strength of weft oriented specimens from composites 7, 8, 9 and 10 as the specimens were cut squarely to the warp axis during specimen preparation. Plate 5.4 on page 249 shows three SEM micrographs of a fractured surface from a tensile specimen reinforced with weave type 10 ($^{1}/_{10}$ weave). Micrograph 'A' on page 249 shows an overview of the surface. Two weft yarns are partially visible within the micrograph as regions of them are still firmly embedded in the epoxy matrix. However it appears that the weft yarns are not well aligned.

The weft orientated specimens of composite 11 (reinforced with a plain weaved woven flax) had the highest average flexural strength. The average flexural strength of

specimens from composite 11 was approximately 10% greater than the weft orientated specimens from composite 2 whose specimens also had the second highest average flexural strength. The weft yarns in composite 11 were the same Tex as the yarns in weave types 1 to 10 (83.3 Tex), but the warp yarns in composite 11 were slightly thinner at 66.6 Tex. The high average flexural strength of weft oriented specimens may be because the weft yarns were not misaligned to the same degree as weft yarns in other composites by warp yarns from neighbouring plies during stacking, impregnation and moulding. The thinner warp yarns from in weave type 11 possibly enable the fabric reinforcement to stack more effectively and thus do not compress the weft yarns and cause them to deviate from the previously straight positions. Plate 5.3 on page 248 shows four SEM micrographs of a fractured surface from a tensile specimen reinforced with weave type 1 ($^{1}/_{1}$ weave). Micrograph 'A' on page 248 clearly shows an indention in the matrix. The indention is from a weft yarn that has debonded during failure. The indentation is not straight as might have been expected. This is because the weft yarn that existed was misaligned because of an adjacent plies warp yarn pressing on it from above. This is speculative as only one SEM micrograph has been obtained, thus no conclusions can be made, however it does illustrate how it is possible that yarns can be misaligned from the yarns from neighbouring plies.

Warp and weft specimens from composite 12 had the lowest flexural strength. This result is quite interesting as the fabric strips from weave type 12 did not have the lowest maximum load at failure in either test direction. However, the warp and weft fabric tensile extensions were relatively high. It is thought that the flexural strengths of this composite are lower than the other composites because the warp yarns are highly crimped, as the weft yarns had a Tex of 104.1. Weft yarns in weave type 12 may be crimped more than other weft yarns in other fabrics because of the honeycomb weave type. Composite 12 was the thickest composite because of the fibre architecture, thus it is thought that it contains large resin rich regions between plies. It is a possibility that matrix cracking may have occurred in these specimens at a lower strain than the other 11 composites, because of stress concentrations that can develop in these regions, and initiate and assist cracking.

Warp orientated specimens from all composites had a lower flexural strength than the unreinforced epoxy resin. Composite 1 had a calculated fibre volume fraction of approximately 27%, warp orientated specimens had a flexural strength that was on average 20% lower than the unreinforced resin (presented in Table 4.3 on page 179). Only weft orientated specimens from composites 1, 2, 3, 4, 5, 6 and 11 had higher flexural strengths than the unreinforced resin. These composite are reinforced with weave types which are relatively stable *i.e.* there warp yarns interlace the weft yarns frequently helping to maintain their integrity.

5.4.1.2 Flexural modulus

Figure 5.4 shows the average flexural moduli of warp and weft orientated specimens from the composites 1 to 12.

The flexural moduli show similar trends to the flexural strengths. Warp and weft orientated specimens from composites reinforced with woven fabrics that contain regularly crimped warp yarns, have large differences between the flexural moduli. The flexural modulus of warp orientated specimens does appear to increase slightly, as the amount of warp yarn crimping reduces in weave types. The average warp orientated flexural modulus of composite 7 appears to be an anomaly to this above mentioned trend. As previously stated in Section 5.3.2.1 on page 225, composite 7 has the lowest density of all 12 composites and a relatively low fabric count. The lower than expected flexural modulus of warp orientated specimens may be caused by the composite having a greater void content or larger matrix pockets than some of the others, or because there are not so many warp yarns per inch of fabric.

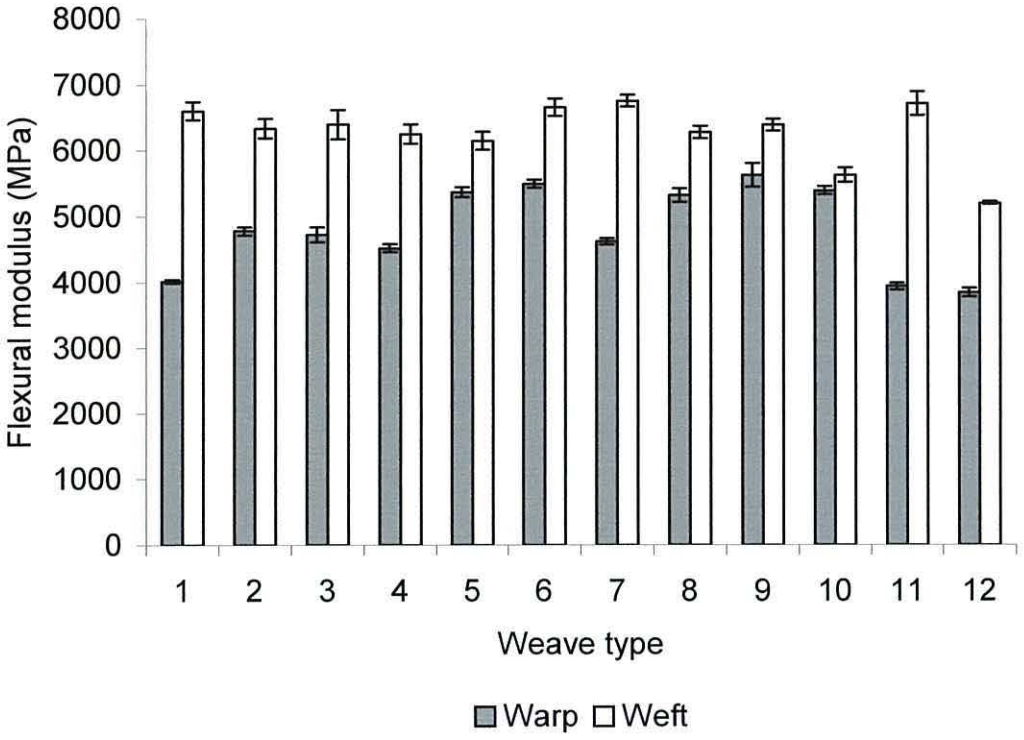


Figure 5.4 The average flexural modulus of specimens tested in both warp and weft directions from composites reinforced with 12 different woven flax fabrics. Table 5.1 on page 214 identifies weave type. Flexural modulus of unreinforced resin is 3136 MPa.

Warp and weft specimens from all composites had a higher flexural modulus than the unreinforced epoxy resin by at least 22%. This increase in modulus is due to the reinforcing effect of the fibres.

5.4.1.3 Nature of stress-strain behaviour

Figure 5.5 shows representative warp and weft orientated flexural stress-strain curves from composites 1 and 10. The moduli of the warp and weft specimens, maximum stress and strain at failure of composite specimens reinforced with weave type 1 are

considerably different. The flexural modulus, maximum stress and strain at failure of warp and weft orientated specimens from composite 10 are however quite similar. The stress-strain curves all exhibit slight elastic behaviour, the gradient then gradually reduces until failure. The stress-strain curves of warp and weft orientated specimens from epoxy composites 2 to 9 were very similar to the data presented in Figure 5.5. The stress-strain curves of warp orientated specimens were different to the warp oriented specimens stress-strain curves of plain weaved woven flax reinforced polyester composites presented in Figure 3.15 on page 153. The warp orientated specimens from these epoxy composites do not exhibit such an abrupt change in gradient in the curve as the woven flax reinforced polyester composite did. This is because the epoxy resin used has a strain to failure that is considerably higher than the Wresipol 31466 polyester resin used throughout Chapter 3 and is not cracking at such low strains. It is also likely that a lower amount of interface decoupling occurs in the woven flax epoxy composites, as the interfacial bond between fibres and matrix is stronger, as indicated by the specimen's brittle failure.

Failure of the warp oriented specimens occurred on the tensile face, a major crack propagating across the width of the specimens in a brittle manner. Warp yarn fracture and matrix cracking did not propagate though the entire thickness of the composites. The reinforcement and matrix near the compression surface appeared to be unchanged (above the neutral axis). Smaller matrix cracks ran across the specimen's width on the tension faces of specimens, these were very close to the major cracks, no more than 5 mm away and were relatively shallow compared to the major crack, they certainly did not propagate though yarns. By completely breaking some of the flexural specimens it was possible to identify that hardly any warp yarn pull out had occurred and warp yarns had fractured nearly flush with the surface of the fracture in a brittle manner. The very few warp yarns that had been pulled out were never greater than 4 mm in length. Weft orientated specimens failed in a similar manner to the warp orientated specimens. No specimens showed evidence of delamination between plies.

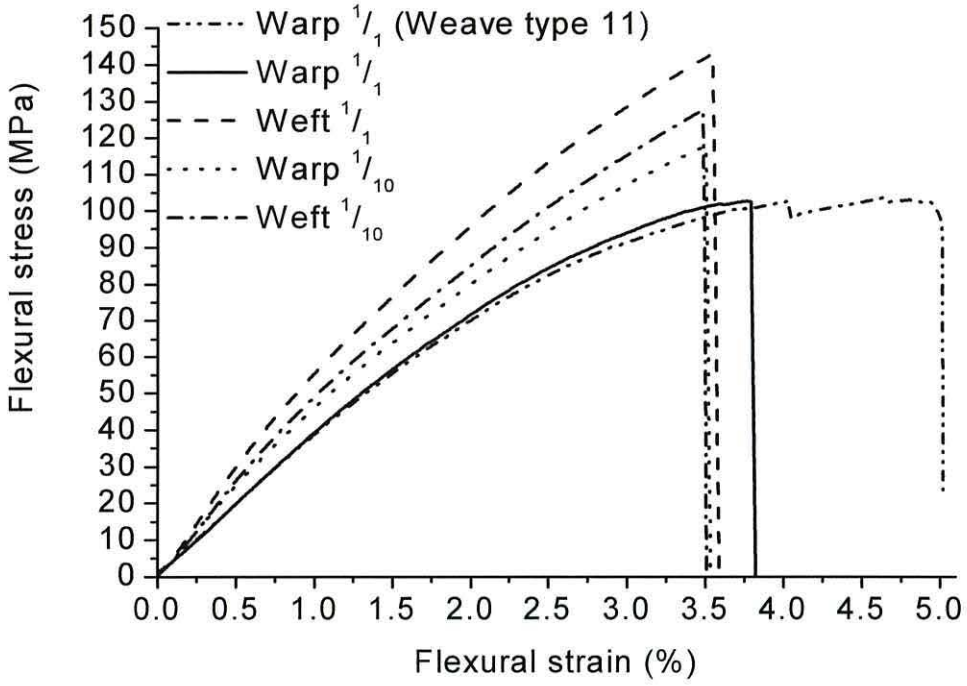


Figure 5.5 Representative flexural stress-strain curves of both warp and weft orientated specimens from two epoxy composites, one reinforced with plies of woven flax that has a $^{1}/_{1}$ weave style (weave type 1) and the second is reinforced with fabric plies which have a $^{1}/_{10}$ weave style (weave type 10). A representative stress-strain curve from a warp oriented specimen from a composite reinforced with weave type 11 woven flax fabric is also presented.

Using computer software (Origin®) the strain at which non-linear behaviour initiated was identified for all composite specimens. Table 5.4 shows the average strain for the onset of non-linear behaviour for warp and weft orientated specimens

It was expected that the elastic region of the stress-strain curves for weft orientated specimens would be greater than the warp oriented specimens because weft yarns are not crimped and are therefore thought to be straighter than warp yarns, thus a better reinforcement. As Table 5.4 shows, for the majority of composites this is not the case, only weft oriented specimens from composites 6, 9 and 10 have an elastic region that

extends to a average strain that is the same or greater than the warp specimen counterparts.

Table 5.4 Average strain at the onset of non-linear behaviour for warp and weft oriented flexural specimens.

Identification	*Average strain at onset of non-linear behaviour*	
	Warp oriented specimens	*Weft oriented specimens*
1	1.15	0.62
2	0.78	0.63
3	0.71	0.57
4	0.69	0.47
5	0.68	0.57
6	0.47	0.55
7	0.58	0.45
8	0.70	0.43
9	0.49	0.49
10	0.53	0.64
11	1.10	0.63
12	0.75	0.44

As mentioned earlier in this section, small cracks were visible on the tensile face of specimens. These smaller cracks are thought to have occurred in the matrix only. The cracks are thought to have occurred at higher strains than for the onset of the non-linear behaviour for specimens; therefore it is thought that matrix cracking has not caused this behaviour, as the unreinforced epoxy cast resin flexural specimens failed at a strain that was in excess of 5.89%, and their fractured specimens had no other cracking visible.

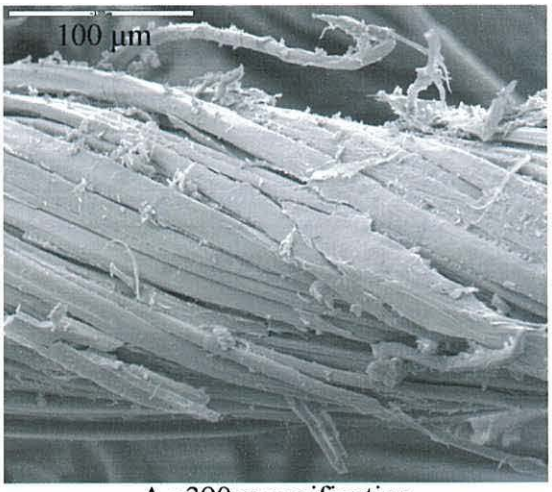
For weft orientated specimens, it can be assumed that the weft yarns are straight within the matrix and are parallel to the tensile and compressive stresses applied. As a load is applied then they immediately start to share the load and strain with the matrix proportionally. However as the stresses build the yarns in tension may debond from the matrix. This irreversible damage would cause inelastic behaviour.

The warp yarns within warp oriented specimens are also parallel to the tensile and compressive stresses applied (the length of the specimen (100 mm) but do not lie straight within the matrix because of crimping. As a load is applied, the warp yarns also share the load with the matrix proportionally, however as the interfacial shear stress increases the yarns may realign as the specimen deforms. Warp yarns may have a higher amount of mechanical keying with the matrix, as they do have a greater surface area for adhesion and therefore this could delay interfacial debonding for a short period of strain, therefore accounting for the difference noticed between types of flexural test specimens. As Table 5.4 shows, some composites that contain reinforcement which are woven so warp yarns are subject to less crimping have similar or the same strains at which non-linear behaviour starts for both warp and weft orientated specimens.

As a load is applied to a warp or weft oriented flexural specimen, the stressed yarns may also try to untwist if in compression or twist further if stressed in tension. Plate 5.4 on page 249 shows SEM micrographs from the fractured surface of a tensile specimen reinforced with weave type 10 ($^{1}/_{10}$). Image 'C' from Plate 5.4 shows a cavity in the matrix created by the departure of a warp yarn as it has been pulled out during failure. Located on the sides of the cavity featured in the micrograph, embedded in matrix, is flax fibre material from the warp yarn that existed there prior to testing, this indicates that a good bond can exist between the two phases. However the rest of the yarn or the interior fibres have been pulled away. If the fibres within central regions of yarns are trying to twist as they are subjected to a tensile load (or the tensile face of a flexural specimen) it may lead to irreversible damage and a weakening of the reinforcement as it is the twisting of the individual fibres that form the continuous yarn and maintains its integrity and controls its breaking strength (Rosiak and Przybyl, 2003). The crimping of the warp

yarns may restrict them from untwisting in the same manner as fibres in the weft yarns until they have become fully extended and as aligned as possible. This may also delay the onset of inelastic behaviour. Warp yarns in all 12 weave types have been coated with a size whereas the weft yarns are not. The size may also help maintain the integrity of the yarns within the composite as this is its purpose during weaving. Although the chemical constituents of the size are unknown it is very likely that this acts as an adhesive between individual fibres (Section 2.5.9 on page 42).

As yarns are strained in tension it is plausible that they may try to contract; sections of individual fibres may be located near or on the surface of yarns but not for their entire length as the fibres are not straight. Plate 5.2 is a micrograph that shows the degree to which the individual flax fibres are twisted in order to form a yarn.



A x390 magnification

Plate 5.2 Scanning electron micrograph of a weft yarn located on the surface of a fractured Charpy impact specimen.

The fibre orientation will mimic a helical structure like a spring. The sections of fibres located near or on the yarn's outer surface may be bonded strongly to the surrounding matrix and may be unaffected as stress is transferred at the interface *via* shear stresses in an elastic manner. However, fibres located within the interior of yarns may have less of

an interaction with the matrix or they may not be bonded at all to the matrix. This would depend upon the penetration of the liquid resin during fabrication and how well it wets the fibres. The fibres in the middle of yarns may be bonded to matrix in small regions along their lengths. Stress is transferred between fibres by friction if polymer is not present. Yarns may contract when stressed in tension if matrix is not present throughout the yarn, fibres within central regions of the yarns that are not wholly bonded to the matrix along their lengths may debond as their aspect ratio is in affect shorter and therefore the fibres do not reach the theoretical maximum axial stress for the applied strain. Stress transfer from the matrix to these decoupled fibres or reduced yarns may occur at their interfaces by friction, however due to the irreversible damage caused from yarn contraction and subsequent debonding of fibres, the stress-strain behaviour of the composite would be inelastic. As the warp and weft yarns are the same Tex (weave type 1 to 10) and have the same degree of twist, it can be assumed that the degree of resin penetration into both types of yarn is similar. However it is possible that the degree of resin penetration into yarns may be influenced by the yarn's orientation within the vacuum bag with respect to the resin flow during fabrication. During the fabrication of these composites the warp fibres were parallel to the resin flow and the weft yarns were perpendicular to it. Resin may have been able to penetrate into one type of yarn better than the other during the time under vacuum. It is also important to remember that the size added to the warp yarns may hinder resin penetration and the wetting of the fibres themselves because of the change in surface characteristics.

Plastic deformation of the matrix may cause the onset of inelastic behaviour. The stress-strain curve of a representative unreinforced cast epoxy flexural specimen is shown in Figure 4.3 on page 180. Non-linear stress-strain behaviour starts at the approximate strain of 2.72% for the cast epoxy flexural specimen presented in Figure 4.3. The average strain at which the cast epoxy flexural specimens stress-strain curves deviate from linearity is 2.69%. The linear behaviour of the woven flax reinforced composites flexural specimens for both test orientations ends at considerably lower strains, therefore it is thought that matrix deformation is not the cause for the onset of inelastic behaviour.

However as the load increases, it is thought that the ability of the resin to deform plays a crucial role in the behaviour of these composites.

Figure 5.5 on page 234 shows a flexural stress-strain curve of a warp orientated specimen from composite 11, which was reinforced with a plain woven flax fabric that consisted of warp yarns that were a lower Tex (thinner) than the weft yarns. Warp orientated specimens from composite 11 all showed similar stress-strain behaviour. As can be seen from Figure 5.5, the strain at which failure occurred for the specimens is higher than the other specimens presented, in fact the warp orientated specimens from composite 11 had the highest average strain to failure of the flax reinforced epoxy composites. Due to the lower Tex of the warp yarns, it is likely that they would be more severely crimped than with the other weave types tested. Stress-strain curves of warp orientated specimens from composite 11 were the only ones that showed a small failure event occurring just prior to final failure. It is likely that this could be due to matrix cracking, as this event occurs at a high strain. It is also possible that the sudden deviation of the stress-strain curve may have been caused by warp yarn failure. The warp yarns in composite 11 are approximately 66 Tex and therefore they contain less flax fibre than the other 11 composite's warp yarns whose Tex was approximately 83.

5.4.2 Impact properties

Figure 5.6 shows the average Charpy impact strengths of warp and weft orientated specimens from the 12 composites.

Apart from composites 4 and 8 there was a significant difference between the Charpy impact strengths of warp and weft orientated specimens from the same composite at a 95% confidence interval. Taking into account all 12 composites, the weft orientated specimens on average exhibit a 27.6% higher Charpy impact strength than the warp orientated specimens. The variation in Charpy impact strength showed no correlation with weave type. For example, the difference between warp and weft orientated

specimens from composites 9 and 10 are greater than the differences between warp and weft specimens from composites 3 and 4.

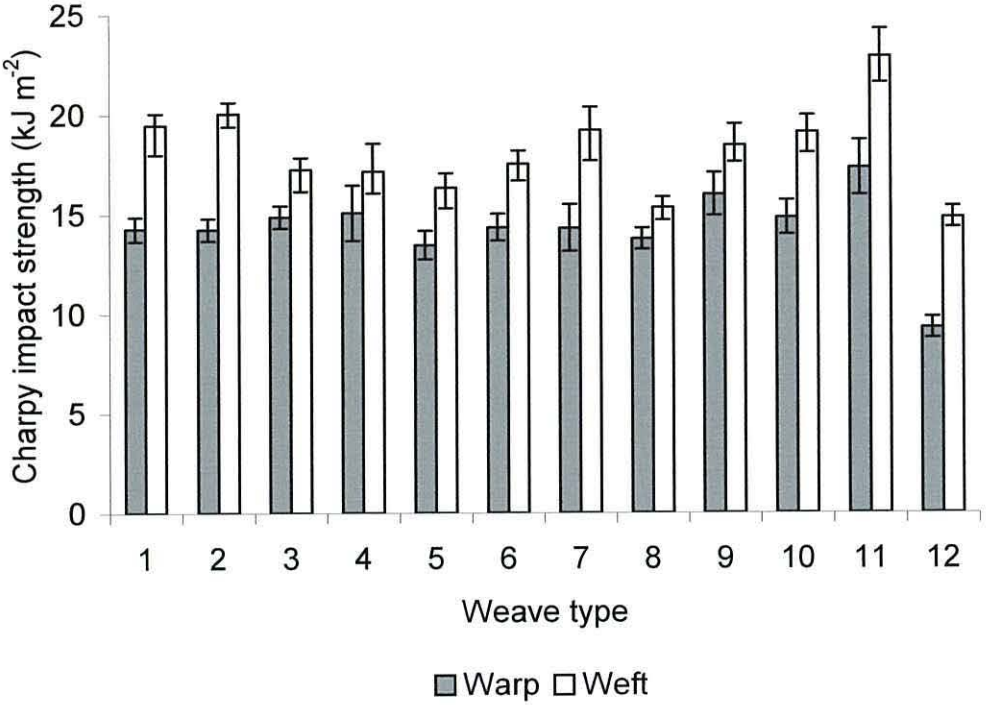


Figure 5.6 The average Charpy impact strengths of specimens tested in both warp and weft directions from composites reinforced with 12 different woven flax fabrics. Table 5.1 on page 214 identifies weave type. Charpy impact strength of unreinforced resin is 43.6 kJ m^{-2}.

Apart from composites 11 and 12, the other 10 composites warp orientated specimens average Charpy impact strengths are quite similar, ranging from 13 to 16 kJ m^{-2}. Although the weft oriented specimens (1 to 10) exhibited slightly more scatter in Charpy impact strength, no significance is attached to this.

Epoxy composites reinforced with weave type's 1 to 10 all have a similar resistance to the propagation of cracks when comparing warp or weft test orientations. As previously mentioned, the thicknesses of these composites are also similar and as there is approximately the same fibre content within these composites as each one contains 5

plies of woven flax, each woven flax ply has the same sized warp and weft yarns, and each has a fabric count of at least 30 in both orientations. It is thought that the behaviour of these composites under dynamic loading conditions is alike and weave type is not such an influencing factor as it is in three point bend flexural testing.

The majority of warp and weft orientated specimens from composites 1 to 10 fractured into two pieces, specimens that did not completely fracture were held together with a few yarns near the compression surface. Fractured surfaces from both types of test specimen contained very few yarns that had pulled out from the opposite fractured piece, as many of the yarns had fractured in a brittle manner very close to the two fractured surfaces. The few yarns that had pulled out were very short, to touch they felt rigid, as if they contained matrix, this indicates that a significant degree of resin penetration into the yarns had occurred. Some specimens had fractured surfaces that were 90° to the composite top and bottom surfaces, fractured surfaces were also sometimes at a slight angle. No trends were identified with these two types of fracture surface with the composite weave type or test orientation. No other visible cracks were identified around the fractured ends of specimens. Composite material close to the fractured surfaces on all impact specimens appeared to be sound, *i.e.* no delamination between plies, evidence of matrix cracking or the decoupling of yarns from the matrix were identifiable.

It is thought that the toughness and the manner in which these 10 composites failed is alike because their reinforcement had bonded reasonably well to the epoxy matrix. As a dynamic load was applied, a crack was initiated, its propagation though the composite material was relatively unhindered as few energy absorbing processes took place such as fibre/yarn pull-out; this resulted in a brittle failure. No other cracks were created during the catastrophic failure event. Unreinforced epoxy cast resin specimens had an average Charpy impact strength of 43.6 kJ m^{-2}. This is approximately 90% greater than the highest Charpy impact strength measured for a woven flax reinforced epoxy composite.

The large difference that exists between the unreinforced epoxy resin and the flax reinforced composites is due to a number of factors. One is due to composite weakening

defects, such as voids, that are included into the matrix when fibres/yarns are added. Unreinforced epoxy resin deformed during flexural testing to a strain of approximately 5.89%. An unreinforced polymer deforming to high strains during the application of stress absorbs a great deal of energy, as much of the deformation is non-elastic. As woven flax fibre is added to the polymer matrix it may cause it to stiffen and therefore partially reduce its ability to deform to the same strains as when unreinforced, thus reducing the amount of energy that it is able to absorb in this manner as a stress is applied. Regions within the composites that initiate cracks that can cause failure such as interfaces and matrix pockets between yarns and plies and general areas of high stress are all created when reinforcement is added. The above effect combined with a good bond between phases and the presence of defects such as voids within the composites and indentations on the surfaces all act as loci for cracks to start.

Composite 11 (thinnest composite) may have had relatively high average weft oriented Charpy impact strength because the fabric count for weave type 11 was the highest for that orientation, therefore there would be a higher number of interfaces within these weft orientated specimens as there are more yarns. Unsurprisingly, composite 12 (the thickest composite) in both test orientations had the lowest Charpy impact strength out of all other epoxy composites tested. This is probably due to the large resin rich regions that exist between plies, because the un-smooth honeycomb weave type. Composite 12 also contained more surface indentations than other composites and had quite a low density indicating that the void content may have been higher than some of the other composites.

5.4.3 Tensile properties of composites

5.4.3.1 Tensile strength

Figure 5.7 shows the average tensile strengths of warp orientated specimens from composites 1 to 12. It is important to consider that only two tensile specimens were tested from each composite, and therefore any trends that exist between different weave types are not significant and possibly exist because of experimental error. Due to the small number of warp oriented tensile specimens the mechanical properties will not be discussed to the same extent as flexural and impact properties have been.

The average warp orientated tensile strengths of composites 1 to 10 do not display any strong trends. However, these results do possibly indicate that the tensile warp orientated specimens strength increases as the amount of warp yarn crimping reduces. Composites 1, 11 and 12 all contain woven reinforcement that has highly crimped warp yarns. These three composites also have the lowest average warp orientated tensile strengths. Composite 9 had the highest tensile strength; other composites with similar weaves to weave type 9 did not follow this trend.

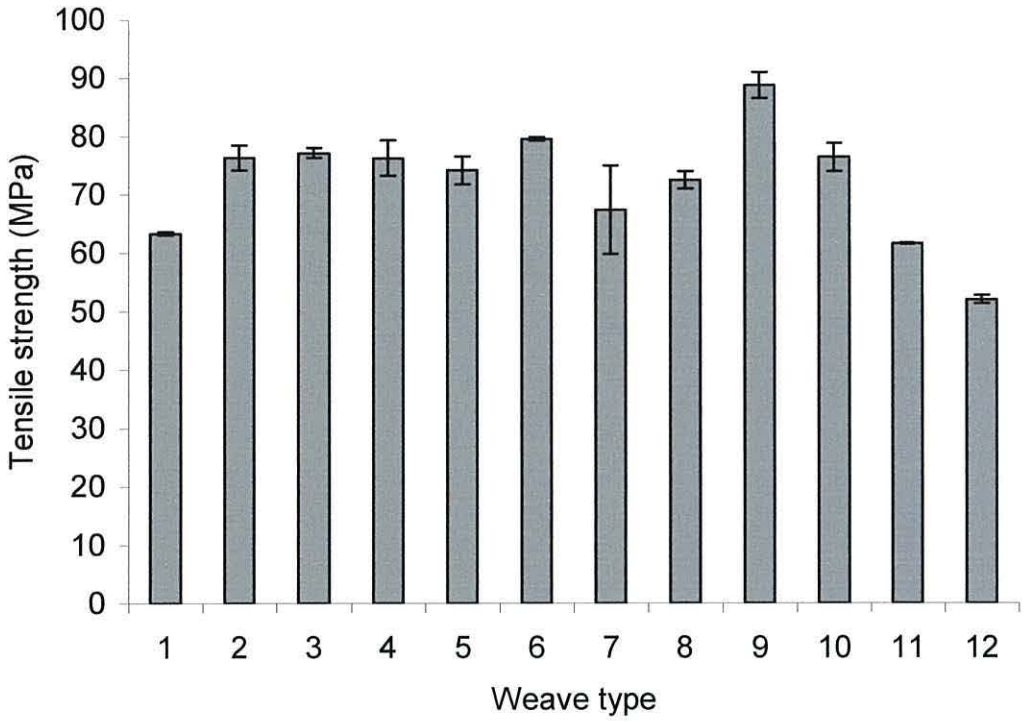


Figure 5.7 The average tensile strength of specimens tested in the warp direction from composites reinforced with 12 different woven flax fabrics. Table 5.1 on page 214 identifies weave type. Tensile strength of unreinforced resin is 67 MPa.

5.4.3.2 Tensile modulus

Figure 5.8 shows the average tensile Young's moduli of warp orientated specimens from composites 1 to 12.

Unlike tensile strength and weave type, the tensile Young's moduli of composites 1 to 10 appear to have a stronger positive relationship. Composites that contain highly crimped warp yarns have a lower modulus than composites that contain straighter warp yarns which are only crimped every few weft yarns. The percentage difference between weave type 1 and 10 is approximately 37.4%. The lowest Young's modulus of woven flax

reinforced epoxy composites is 41% higher than the Young's modulus of unreinforced epoxy resin.

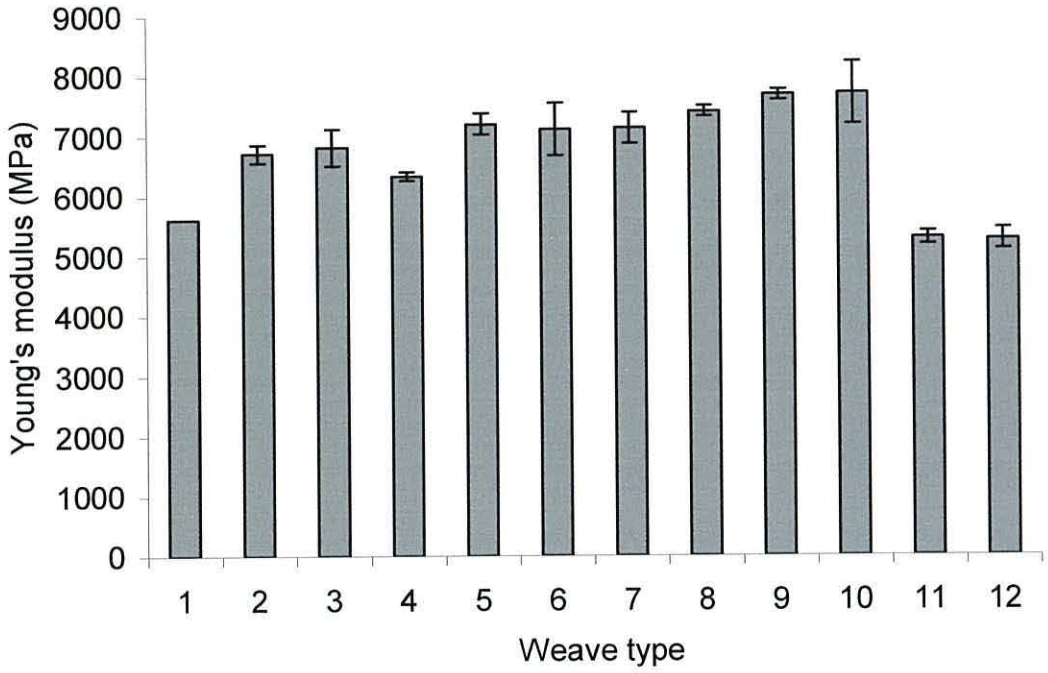


Figure 5.8 The average tensile Young's modulus of specimens tested in the warp direction from composites reinforced with 12 different woven flax fabrics. Table 5.1 on page 214 identifies weave type. Young's modulus of unreinforced resin is 3.72 GPa.

5.4.3.3 Nature of the stress-strain behaviour

Figure 5.9 shows four representative tensile specimen stress-strain curves; each one is from a specimen from a different composite and therefore reinforced with a different weave type.

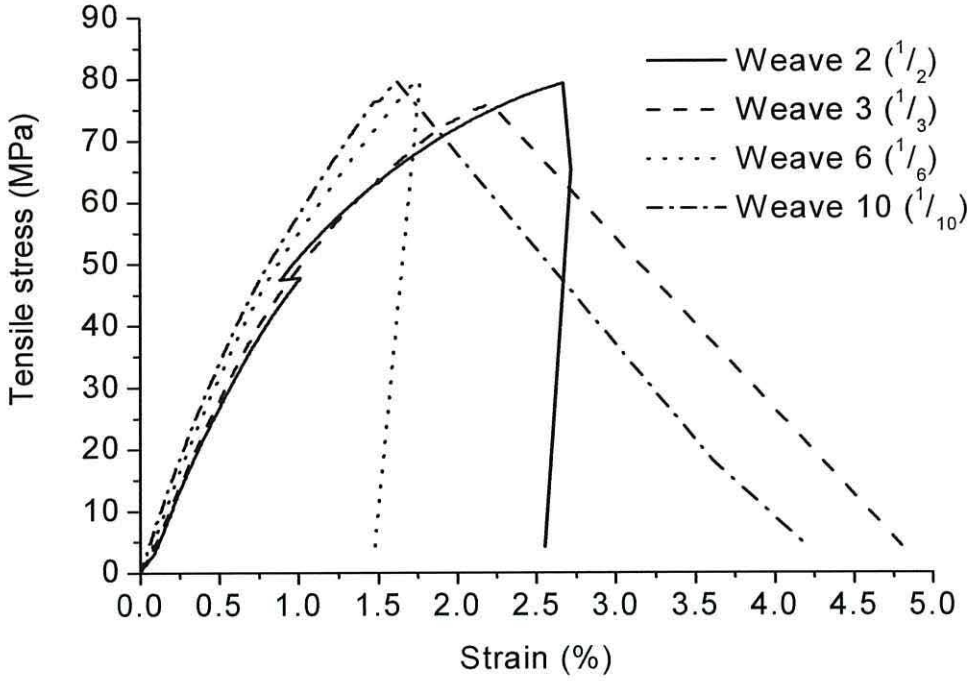


Figure 5.9 Representative tensile stress-strain curves from woven flax reinforced epoxy composites that contain reinforcement with different weave types.

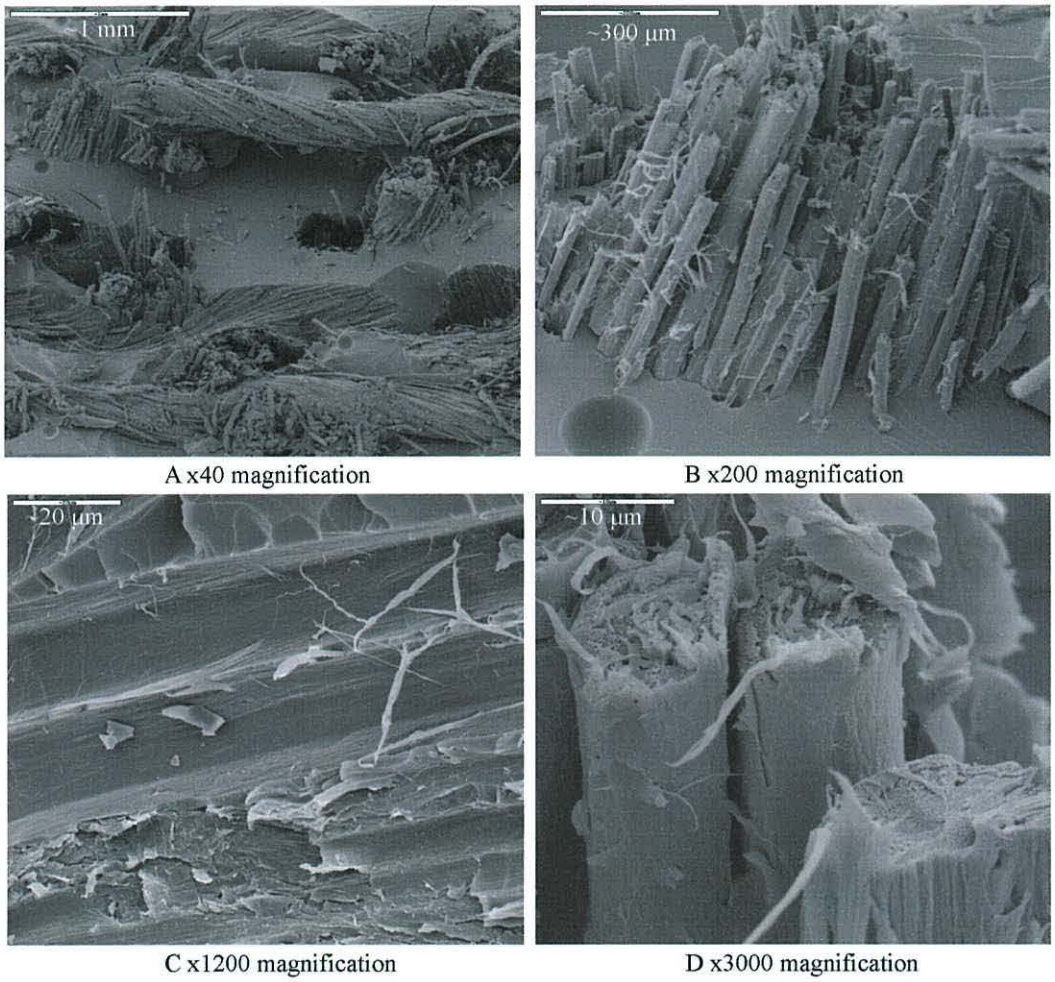
The stress-strain curve representing the tensile specimen reinforced with weave type 2 has a small deviation occurring at approximately 1% strain. This was caused because the extensometer slipped during testing; unfortunately this occurred on both specimens. However, Figure 5.9 clearly shows that the initial gradient of the curves increases (increased modulus) and the strain to failure also slightly reduces as the woven reinforcement used in specimens contains less crimped warp yarns. It is also apparent that the departure of the stress-strain curves from linearity appears to be influenced by the amount of warp crimping. Severe crimping of warp yarns causes the specimens to deform to higher strains, thus, it is thought that there is a greater amount of irreversible damage occurring in composites that contain highly crimped reinforcement. It is thought that the fibres within highly crimped warp yarns are straightening.

Not all the fibres within these yarns are showing this behaviour as some will remain bonded to the matrix; however it is plausible that fibres from the interior of these yarns are able to move as they may not be bonded to the polymer. The size used on warp yarns may have inhibited resin penetration.

All of the tensile specimens failed in a brittle manner. Apart from the two fractured surfaces across the width of the tensile specimens, there were no other visible signs of damage such as delamination or matrix cracking. Very little yarn pull-out occurred, as many of the warp yarns failed close to the fractured surface. Plate 5.3 on page 248 shows four SEM micrographs of a fractured surface from a tensile specimen reinforced with weave type 1.

Micrographs 'A' and 'B' show warp yarns which have broken close to the surface of the fracture. Micrograph 'B' shows that some of the individual fibres within the warp yarn are still embedded in the matrix. The interface between some of these fibres and the matrix appears to be intact. However, small matrix cracks starting from the fibre/matrix interface have propagated though the cast resin to other stressed regions, such as voids. In Micrograph 'D' there are the fractured surfaces of flax fibres from within a warp yarn.

The fibres ends appear very flat, giving them the appearance that they failed in a brittle manner. A good bond was formed between the flax fibres and the epoxy matrix. Micrograph 'C' shows indentations left in the matrix from a weft yarn that has been pulled away. Left embedded within the imprint of the weft yarn is flax fibre material.



A x40 magnification

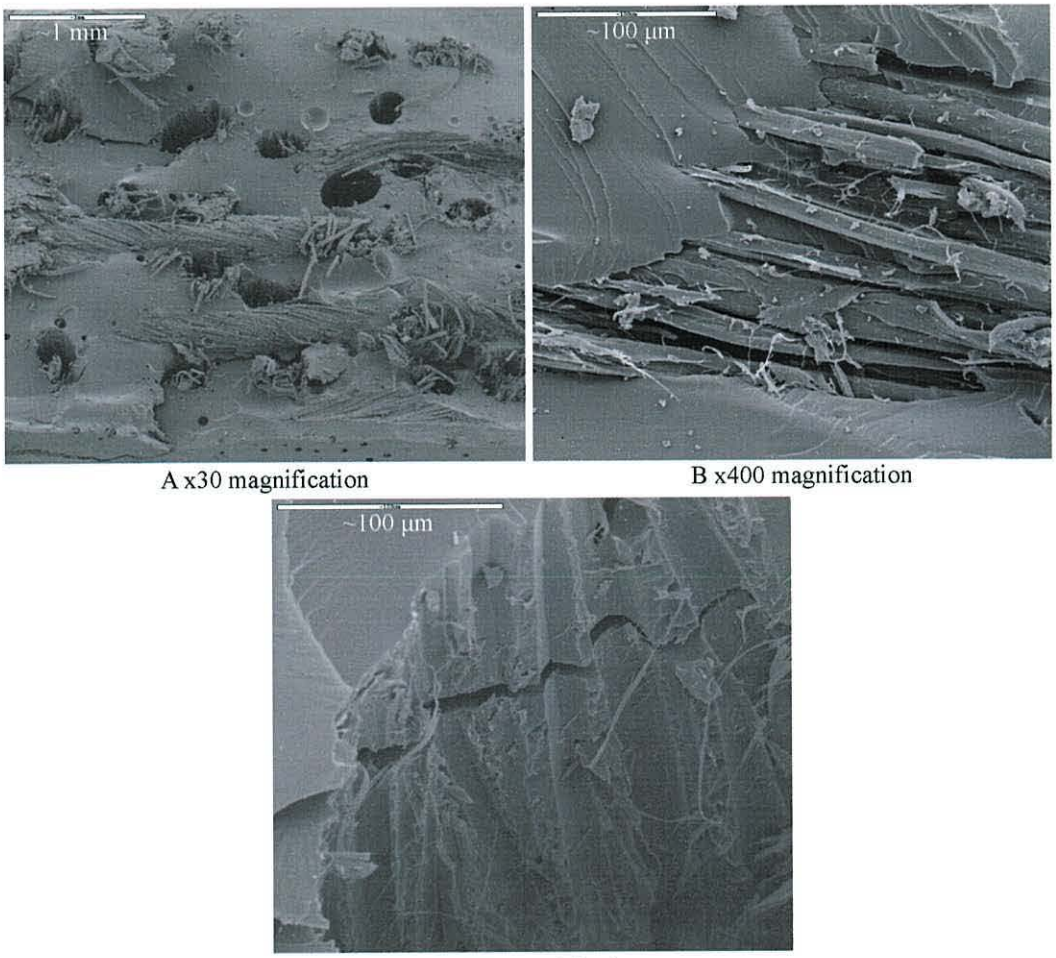
B x200 magnification

C x1200 magnification

D x3000 magnification

Plate 5.3 Scanning electron micrographs of a tensile fracture surface from a composite reinforced with weave type 1 ($^{1}/_{1}$ weave) at various magnifications. A) 'overview of the fractured surface with weft yarns running horizontally' B) 'fractured warp yarn still embedded in resin with evidence of conchoidal fractures (bottom left)' C) 'grooves left by weft fibres with some remains of embedded fibre material' and D) 'brittle fracture of warp fibres'.

Plate 5.4 shows three SEM micrographs of the fractured surface of a failed tensile specimen. This tensile specimen was reinforced with weave type 10 and the composite had an average thickness of 3.30 mm. The thickness of the tensile specimen shown in Plate 5.3 was on average 2.79 mm.



A x30 magnification

B x400 magnification

C x500 magnification

Plate 5.4 Scanning electron micrographs of a tensile fracture surface from a composite reinforced with weave type 10 ($^{1}/_{10}$ weave) at various magnifications. A) 'overview of fractured surface with weft yarns running horizontally and holes left from pulled out warp yarns' B) 'weft yarn torn apart on the fractured surface with conchoidal fractures' C) 'a hole with fibre material embedded in the edges where a warp yarn has been pulled out and there is a crack propagating under the fractured surface'.

Micrograph 'A' from Plate 5.4 is an overview of the fractured surface. Within this micrograph there appear to be large regions of unreinforced matrix containing voids. Two weft yarns running horizontally across the micrograph appear to be firmly embedded into the matrix. As with micrograph 'A' and 'B' in Plate 5.3, the yarns have failed close to the fractured surface. Micrograph 'B' shows a section of a weft yarn. It is

thought that some of this weft yarn has been pulled away during fracture. If this is true, cast resin is seen between fibres that are within the central region of the yarns indicating that the liquid resin was able to penetrate into these yarns. Propagating small cracks from the fibre/matrix interface are also visible; they are conchoidal in appearance.

Higher stress-concentrations are thought to exist at the warp and weft yarn cross over points, as these are the regions where the warp yarns are less aligned to the applied load. As a load is applied in the direction of the warp yarns, the well-bonded weft yarns are preventing them from becoming aligned. It is plausible that failure initiated in these areas of the composite, as the fibres with yarns may have a reduced strain to failure since they are not aligned to the direction of the load.

5.5 Summary

Woven flax fabrics containing highly crimped warp yarns display high warp tensile extensions, but as the frequency of warp yarn crimping reduces, the tensile extension also decreases, becoming closer to the value of the weft tensile extension exhibited at failure. Woven flax fabric's weft tensile extension and maximum load at failure is unaffected by the amount of warp yarn crimping for woven flax fabrics consisting of the same Tex warp and weft yarns. Nearly all the woven flax fabrics tested had significant differences between warp and weft maximum loads achieved at failure, with weft maximum loads at failure being higher. However, warp and weft maximum loads at failure were very similar for a woven flax fabric that consisted of warp yarns that were only crimped every 10 weft yarns passed over, the two types of yarn weaved in this fabric were also the same Tex.

A five ply epoxy composite was successfully fabricated from each of the different woven flax fabrics. It was found that the observed properties from each composite could be compared with other composites without considering differences in fibre volume fractions as the variations in calculated fibre volume fractions were relatively small and

therefore did not alter the observed trends. However, relatively large variations were observed between composites densities. The variation was thought to exist because of differences in composites void contents, sizes of resin rich regions and the number of reinforcing yarns per square inch.

Significant differences were observed between some composites warp and weft flexural strengths, weft flexural strengths were greater. Composites that exhibited significant differences were ones that contained more frequently crimped warp yarns. As warp yarn crimping became less frequent, the observed anisotropic behaviour between both test directions decreases until the warp and weft flexural strengths were more or less the same. Composites reinforced with woven flax fabrics that consist of warp yarns passing over 8, 9 or 10 weft yarns of the same Tex before crimping had similar warp and weft flexural strengths. Composites reinforced with woven flax fabrics that contained frequently crimped warp yarns displayed completely different moduli when tested in either the warp or weft direction, the composites weft flexural moduli was always greater. The differences between warp and weft composite moduli did appear to reduce as warp yarn crimping became less frequent. However, only one composite which was reinforced with a woven flax fabric that consisted of warp yarns that passed over 10 weft yarns before crimping again had truly similar values of warp and weft composite moduli. All stress-strain curves obtained from flexural specimens had an initial linear region. It was observed that a difference existed with the strain at which the onset of non-linear behaviour occurred between warp and weft specimens from the same composite. Non-linear behaviour initiated at a lower strain when tested in the weft direction for the majority of the composites.

Most composites exhibited a significant difference between warp and weft Charpy impact strengths. The weft Charpy impact strength of a composite was always observed to be greater than the same composite's corresponding warp Charpy impact strength. Correlations between composite's Charpy impact strengths and the weave type of the woven flax fabric reinforcement used were not found. It would be incorrect to summarise how a composite's tensile properties are influenced by the weave type of flax

reinforcement because of the low number of tensile specimens tested for each type of woven flax reinforced epoxy composite. However it was observed, as the frequency of warp yarn crimping reduced, the composite's warp orientated tensile Young's modulus of gradually increased.

To summarise, the weave type of woven flax fabric reinforcement used in epoxy composites does influence the composite's flexural properties in the warp direction. Reducing the crimping frequency of warp yarns within woven flax fabric reinforcement results in the warp orientated flexural properties of the composite becoming closer to the weft orientated properties, which are generally greater because of the better yarn alignment, since the weft yarns are not crimped. Different weave types of woven flax reinforcement used within composites did not influence the Charpy impact strengths of composites.

6 MECHANICAL PROPERTIES AND DEFORMATION BEHAVIOUR OF FLAX FIBRE UNIDIRECTIONAL COMPOSITES

6.1 Introduction

The mechanical properties of thermosetting polymer matrix composites reinforced with various natural fibres have been reported in the literature *e.g.* Roe and Ansell (1985); Sanadi *et al.,* (1985); Sanadi *et al.,* (1986); Kumar (1986); O'Dell (1997); Sèbe *et al.,* (1999). Eichhorn and Young, (2003 and 2004) studied stress-transfer at the fibre to matrix interface and Hughes (2000) analysed the effects that microscopic fibre defects have upon the stress-strain field in the matrix surrounding flax fibres, the mechanical properties of composites are partially dependent on the nature of the fibre to matrix interfaces within. As previously mentioned in Section 1.5 on page 12, much interest has been focused on improving natural fibres themselves, an example is the chemical modification of their surfaces to reduce the effects of the fibre's hydrophilic nature and to make their surfaces more compatible with the polymers they are combined with. Other workers have focussed on the influence that fibre parameters have upon the macroscopic properties of the composite (Oksman, 1999; Bos and Van den Oever, 1999).

Few studies have, however, considered the nature of the deformation behaviour of natural fibre thermosetting polymer composite systems and how this relates to the structural application of these materials. Furthermore, whilst there has been a certain amount of work undertaken to characterise the deformation behaviour of model systems, such as individual fibre micro-tensile composites (Eichhorn & Young, 2003 and 2004), this has in general, not been related to the bulk behaviour of the material. If confidence in these materials is to be given to design engineers, it is vital that a full understanding, not only of the macroscopic behaviour, but also the relationship between microstructure and macro-properties, be achieved.

Experimental work by Hughes, (2000) and Hughes *et al.,* (2002) has highlighted that non-linear behaviour in bast fibre reinforced-unsaturated polyester composite systems occurs at low values of stress and strain. Stress-strain curves obtained from experimental work in Chapter's 3, 4 and 5 in this thesis have also shown that non-linear behaviour occurs at low levels of stress and strain.

This present study was initiated with a view to elucidating this behaviour and to attempt to gain a fundamental understanding of the underlying micromechanical processes operative. The mechanical properties (flexural, toughness and tensile) of unidirectional flax fibre reinforced thermosetting polymer composites are recorded at various fibre loadings, the mechanical response of unidirectional flax fibre reinforced thermosetting polymer composite systems under quasi-static monotonic tensile loading was recorded and analysed. The effect of varying interfacial properties upon the deformation and fracture behaviour was assessed through appropriate chemical modification of the fibre prior to lamination.

A unidirectional composite system was used to study the micromechanical processes as it was thought this type of composite system is less complex than a woven reinforced composite system. It is easier to control unidirectional composites fibre volume fractions. Acoustic emissions analysis has also been used during this investigation as a tool to identify acoustic events. Acoustic emissions analysis was not used for woven flax reinforced composites as it was thought that the noise would have been so great it would have swamped the acoustic events and made an analysis more difficult.

6.2 Materials and method

6.2.1 Resin

An unsaturated polyester resin from Resinous Chemical Ltd was obtained and utilised for the fabrication of all composites. The physical and mechanical properties of Wresipol 31466 obtained from the suppliers are shown in Table 3.1 on page 108. The physical and mechanical properties of Wresipol 31466 determined from cast resin panels manufactured and tested during a previous investigation are presented in Table 3.3 on page 123.

6.2.2 Glass fibre

Unidirectional E-glass fibre was obtained from Scott Bader and utilised for the fabrication of unidirectional composite bars.

6.2.3 Flax fibre

High quality linen grade flax fibre in the form of sliver was obtained from SANECO, (Zone Artisanale, 231 Ruelle DUFOUR, 59850 Nieppe, France). A section of the flax sliver is presented in Plate 6.1 .

The fibre was solvent extracted in a soxhlet for 5 hours to remove any waxy substances prior to use. A mixture of toluene, methanol and acetone were used in the proportions 4:1:1 (by volume). This fibre was used in an unmodified form (UnM) and in two modified forms (sees Section 6.2.3.1).

Plate 6.1 Flax fibre in the form of sliver.

6.2.3.1 Flax fibre modification

Fibre was modified by reaction with two reagents: (i) methacrylic anhydride (MeA) and (ii) propionic anhydride (PrA). 50 g samples of Soxhlet extracted fibres were oven dried at 105°C for 16 hours and subsequently weighed to 4 decimal places (±0.001 g) immediately after being taken from the oven. The samples were reacted in 1 Molar solutions of reagent (195 ml of propionic anhydride in 1305 ml pyridine and 223.5 ml of methacrylic anhydride in 1276.5 ml pyridine) in pyridine at 95°C for 7 hours. Pyridine was used as it is a very good cell wall swelling agent and acts as a base catalyst for the reaction. After quenching the reaction, the fibre samples were soxhlet extracted to remove any unreacted reagent with a mixture of toluene, methanol and acetone in the proportions 4:1:1 (by volume) for a further 5 hours, prior to oven drying as before. After reweighing, the weight percentage gain (WPG) was determined.

The rationale for modifying the flax fibre (sliver) in the manner described above was to explore the effect of varying fibre to matrix adhesion upon the mechanical properties, particularly the deformation behaviour, of flax fibre reinforced unidirectional polyester composites. The purpose of pursuing this modification regime was not, therefore, an

attempt to develop new natural fibre treatments for composite applications, rather, to assist an investigation into the understanding of bast fibre reinforced PMC systems.

Activation of the fibre surface for subsequent co-polymerisation with the resin matrix or alteration of the fibre surface chemistry for improved compatibility with the matrix phase, was achieved by reaction with di-functional methacrylic anhydride and propionic anhydride respectively. A scheme showing the reaction between the hydroxyl (-OH) groups and (a) methacrylic anhydride and (b) propionic anhydride is shown in Figure 6.1.

Chemical modification through the introduction of reactive vinylic groups at the fibre surface by esterification of the flax –OH groups with methacrylic anhydride is expected to lead to subsequent radical copolymerisation between these vinylic groups and the unsaturated bonds of the resin during fabrication (Sèbe *et al.*, 2000; Hill and Cetin, 2000; Cetin and Hill, 1998).

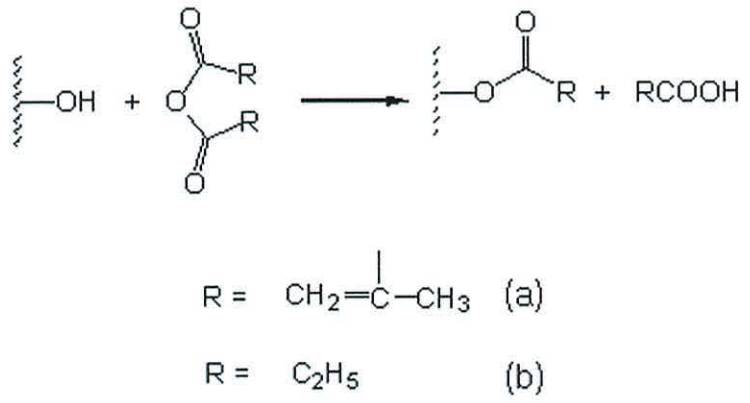


Figure 6.1 The reaction mechanism between flax fibre –OH groups and (a) methacrylic and (b) propionic anhydrides.

Unlike methacrylic anhydride, propionic anhydride modified fibre does not contain a functional site for reaction with the resin matrix, but alteration of the chemistry of the fibre surface, rendering it more hydrophobic, will occur, leading to improved

compatibility with a more hydrophobic polymer (Hill and Cetin, 2000; Cetin and Hill, 1998).

6.2.4 Composite fabrication

Before composite fabrication, both the unmodified and modified flax fibre was allowed to equilibrate under ambient conditions of RH and temperature.

6.2.4.1 Resin preparation

Resin for each unidirectional composite bar was mixed with 1% (by weight) of catalyst (Butanox M50, organic peroxide obtained from Akzo Nobel Chemicals Ltd) with a mechanical stirrer for at least 5 minutes. After stirring, the catalysed liquid resin was then degassed within a desiccator for at least 5 minutes before use.

6.2.4.2 Resin impregnation of reinforcement

The required weight of fibre was vacuum impregnated with degassed catalysed resin to ensure good wet out. A similar method used to impregnate these four types of fibre is described in Section 3.2.4.2 on page 112 and shown in a schematic representation in Figure 3.1 on page 113. All four types of fibre were impregnated using this method (E-glass, UnM, MeA and PrA). All fibre types were kept straight during this process and were not bunched together.

6.2.4.3 Moulding and curing of unidirectional composite bars

Unidirectional composite bars were fabricated in a closed compression mould. Resin impregnated fibre was laid into a mould with the following dimensions 450 mm × 25 mm × 3.5 to 4.7 mm. By hand, fibres were aligned to the long axis of the mould before closure. The fibre volume fractions of the finished unidirectional composite bars were adjusted by varying the weight of fibre used in the initial lay-up within the plastic bag prior to resin impregnation. All composites were cured at room temperature for 12 hours, followed by post curing at 50°C for 45 minutes.

6.2.5 Measurement of composites

The length and weight of each composite bar was measured to an accuracy of ±0.1 mm and ±0.1 g respectively. The thickness and width was measured at ±0.01 mm. The width and thickness were measured five times along the composite's length and the mean average taken.

6.2.6 Specimen preparation

All post cured composites were trimmed to remove the ends (100 mm from either end), leaving 250 mm long tensile specimens for testing. Only one tensile specimen was obtained from each composite bar. Aluminium end tags were glued with Araldite adhesive to tensile specimens. Some of the 250 mm long unidirectional composite bar Sections were cut further to obtain two flexural specimens with nominal dimensions 100 mm × 15 mm × 3.5 to 4.7 mm and two Charpy impact specimens with nominal dimensions 80 mm × 10 mm × 3.5 to 4.7 mm. All cutting was performed with a fine toothed band saw. As will be mentioned in Section 6.2.7.2, notched Charpy impact

specimens were tested. The type B notch was cut with a fine bladed circular saw attached to a lathe for precision and control.

6.2.6.1 Conditioning

All specimens were conditioned at 65% RH and 20°C for at least 48 hours prior to testing at the same environmental conditions.

6.2.6.2 Measurement of specimens

The width and thickness (to an accuracy of ±0.01 mm) were recorded three times along the length of the specimens and the mean average used for any calculations. The length (to an accuracy of ±0.01 mm) was also recorded from each specimen. The specimen's weight was recorded to an accuracy of ±0.01 g.

6.2.7 Testing

6.2.7.1 Flexural

Flexural testing was conducted using the method described in Section 3.2.9.1 on page 117.

6.2.7.2 Impact

The flat-wise impact properties (direction of blow parallel to the thickness of the specimen with impact on the broad longitudinal surface) of type B notched specimens were found using an analogue Zwick 5102 Pendulum impact tester. Notched specimens were tested because it was found that unnotched specimens did not fail and the energy recorded went off the scale on the analogue Zwick Pendulum tester. Figure 6.2 shows a type B notched Charpy impact specimen. Testing was conducted in accordance with BS 2782: Part 3: Method 359: 1984 ISO 179-1982 – Determination of Charpy Impact Strength of Rigid Materials. A testing span of 60 mm was implemented and a 4 J pendulum was used. The direction of the 'blow' was on the opposite side to the type B notch.

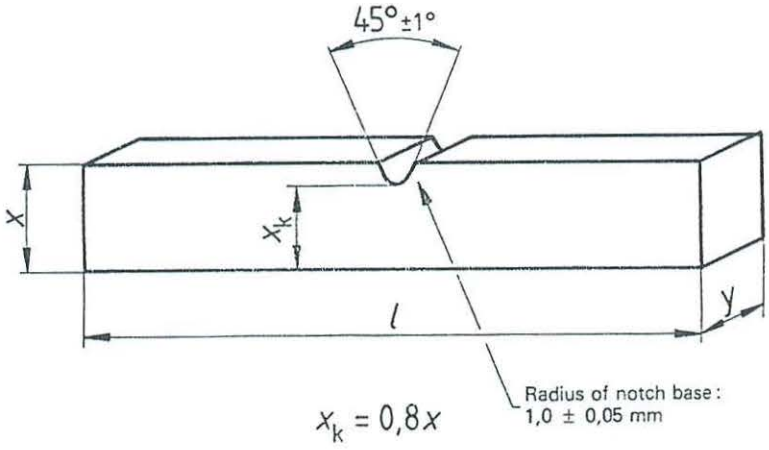


Figure 6.2 Type B Charpy impact specimen.

6.2.7.3 Tensile

Tensile tests were performed on an Instron model 1195 universal testing machine fitted with a 100 kN capacity load cell. Specimens were clamped using Instron self tightening jaws. Load and extension data were acquired digitally and specimen extension was

measured by an Instron extensometer. The extensometer was positioned centrally within the gauge length (150 mm). With a cross head speed of 10 mm min^{-1} testing was conducted in accordance with BS 2782: Part 10: Method 1003:1977 (EN 61). In addition, a number of unmodified flax fibre reinforced unidirectional composite bars were loaded to set points along the load-extension curve and subsequently unloaded. The loading and unloading cycle was recorded.

6.2.7.4 Acoustic emissions analysis

Acoustic emission testing was performed at the University of Bath. UnM flax or E-glass fibre composite specimens were also tested in uniaxial tension as described in Section 6.2.7.3 but at a speed of 1 mm min^{-1}. The lower rate of deformation was adopted to enable the capture of data arising from acoustic events. As specimens were deformed, the acoustic emissions (AE) were detected by a surface mounted piezoelectric transducer and analysed on an MR1004 acoustic emission analyser. AE events were sorted into 25 amplitude levels each 2.4 dB wide. Guild *et al.*, (1985) provide further detail on the AE techniques employed. Plate 6.2 shows an UnM flax tensile specimen in test setup with an extensometer and the piezoelectric transducer attached. The extensometer used throughout the acoustic emissions analysis was not the same type used in other tensile testing.

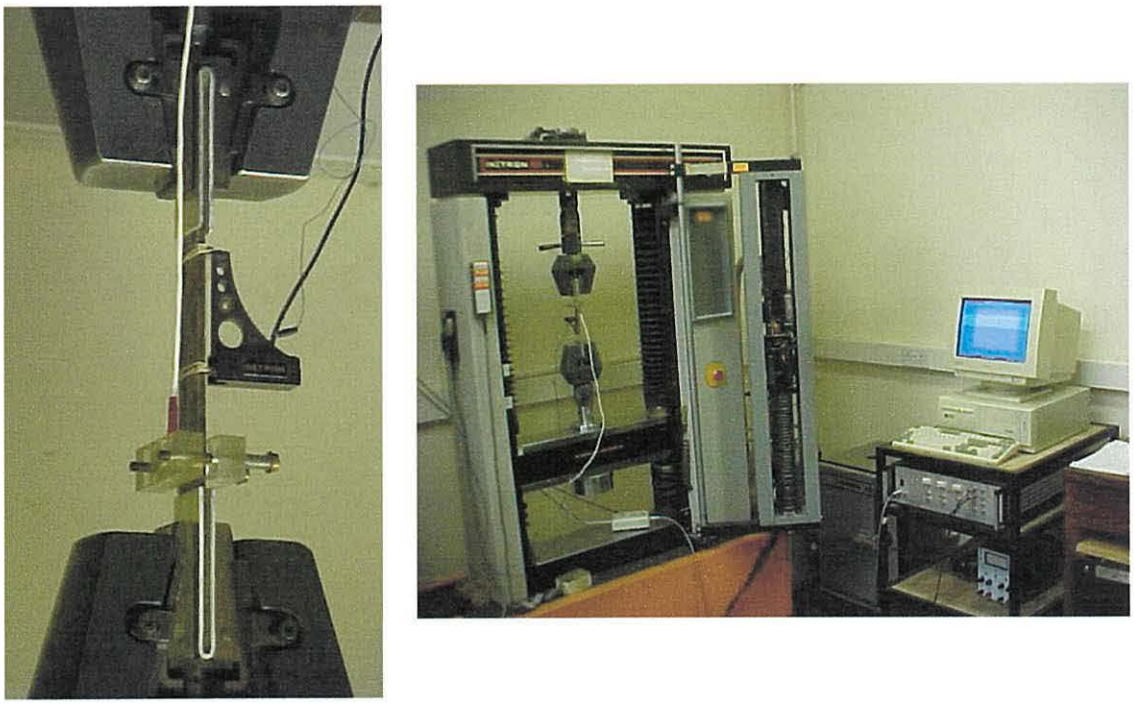


Plate 6.2 Photos of acoustic emissions analysis tensile test set-up.

6.2.8 Fractography

Samples for SEM analysis from flax reinforced composite tensile specimens that exhibited brittle failure were obtained by cutting a section of the fractured surface away using a fine toothed band-saw. SEM samples were obtained from flax unidirectional tensile specimens that exhibited shear failure and delamination by peeling away the sections of delaminated fibres and matrix. The samples were then secured to aluminium stubs with conducting epoxy adhesive, leaving the fractured surface exposed. The samples were dried in an oven set at 100°C for a few hours before being placed over silica gel for 24 hours. The samples were splutter coated using a Polaron E5000 set to 1.2kV and 10mA. The samples were coated in gold from a pure gold target for 2.5 minutes just prior to placement within the SEM. A Hitachi S-520 scanning electron microscope (SEM) was set to 12kV and used at various magnifications to record the fractures.

6.2.9 Evaluations of physical properties

6.2.9.1 Measurement of density

Densities of all unidirectional composites were calculated using Equation 3.1 on page 120. The volume of the composite was calculated using the average measurements of the exterior dimensions.

6.2.9.2 Measurement of fibre volume fraction

Composite constituent's volume fractions were calculated using Equation 2.12 on page 62 because of the inherent variability of natural fibre and the difficulty in obtaining accurate data on fibre density (Roe and Ansell, 1985).

6.3 Results and Discussion

6.3.1 Physical properties of the composites

6.3.1.1 Composites fibre volume fractions and densities

UnM flax fibre reinforced composites were fabricated with fibre volume fractions ranging from 12.5 to 57%. Tensile, flexural and Charpy impact tests were performed on these composites. Figure 6.3 shows the variation of density of 16 unidirectional composites with fibre volume fraction, as fibre content increases the composite density

also increases. However, composites at low fibre volume fractions had densities lower than the density of the cast polyester resin (1180 kg m^{-3}). Unlike the theoretical prediction made by the ROM (Equation 2.13 on page 63) assuming that the fibre density is 1500 kg m^{-3} the relationship between fibre volume fraction and density is not linear, especially when accounting for the density of the cast resin. A linear regression line is presented in Figure 6.3, however, and extrapolation to 100% fibre volume fraction leads to a theoretical fibre density of 1451 kg m^{-3}. Extrapolation of the trendline curve to 100% fibre volume fraction leads to a fibre density of 1516 kg m^{-3}. Both of the above theoretical flax fibre densities are close to those reported in the literature (Oksman *et al.*, 2003; Madsen and Lilholt, 2003; Voorn *et al.*, 2001; Tröger *et al.*, 1998; Ivens *et al.*, 1997).

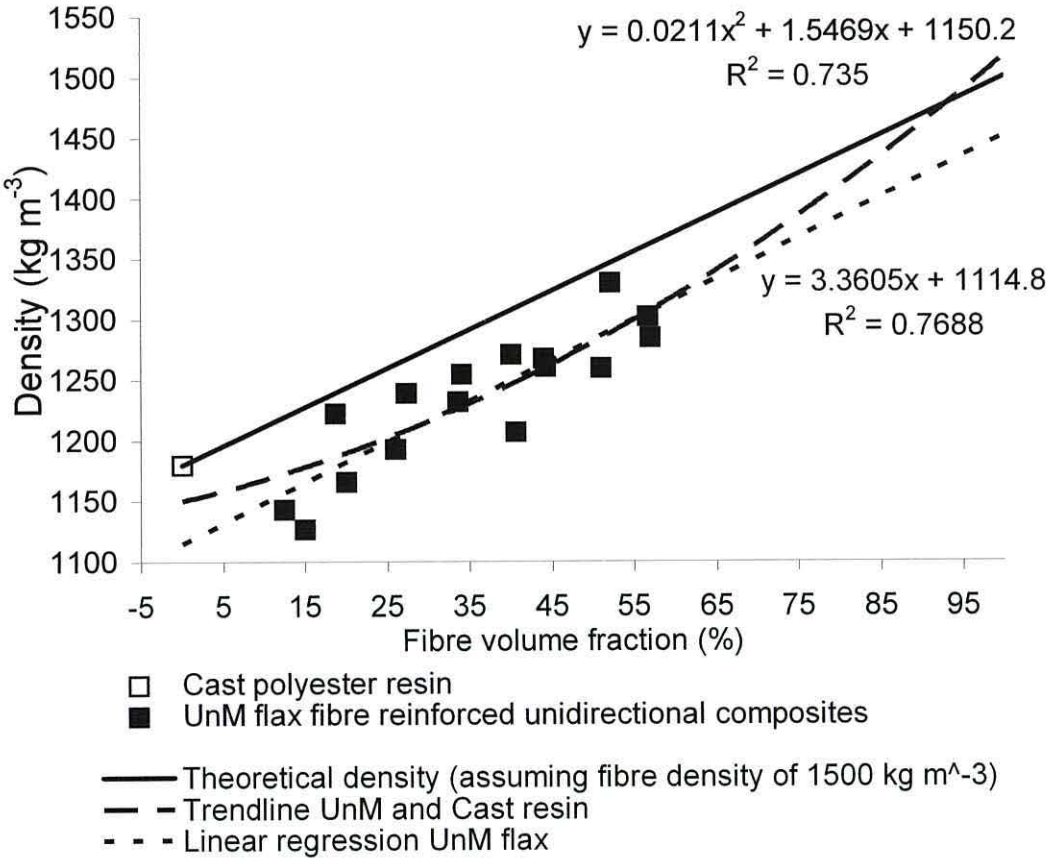


Figure 6.3 Variation of UnM flax fibre reinforced unidirectional composite density against fibre volume fraction.

6.3.2 Unmodified flax composites flexural properties

Figure 6.4 shows the flexural strengths of UnM flax reinforced polyester composites against fibre volume fraction. Figure 6.5 on page 267 shows the UnM flax reinforced composites flexural modulus against fibre volume fraction. As may be observed from Figure 6.4 and Figure 6.5, with increasing fibre volume fraction the composites flexural strength and modulus improve in a linear fashion, to an extent that at nearly 60% fibre volume fraction, a flexural modulus of 20 GPa and a flexural strength of around 200 MPa were achieved. At a fibre volume fraction of 15%, the flexural strength of UnM flax composites is approximately the same as the unreinforced polyester resin and the flexural modulus is approximately 5% lower than that of the matrix material.

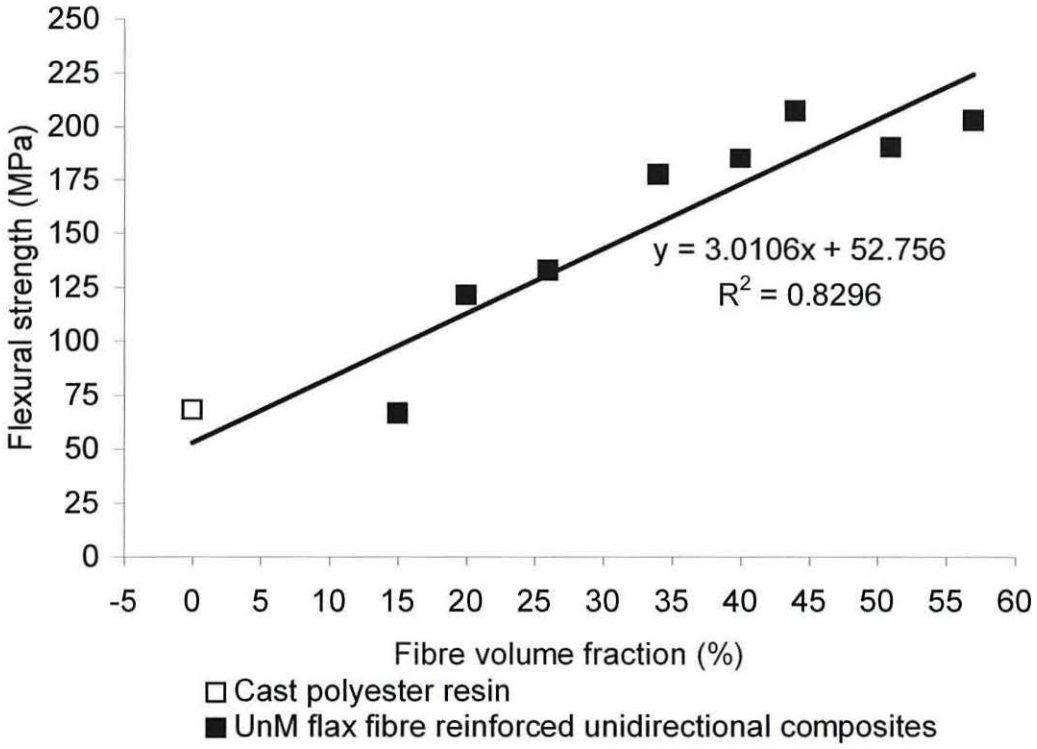


Figure 6.4 Flexural strength of UnM flax reinforced unidirectional polyester composites against fibre volume fraction.

Wang, (1999) observed the flexural modulus and flexural strength of hand laid plain weaved E-glass roving reinforced epoxy composites at a 54% fibre volume fraction (as reinforcement is woven, the V_f of the composite can be considered to be 27%) to be 14.5 GPa and 330 MPa respectively. Although a different resin system, the flexural modulus of UnM flax unidirectional composites compares well to the E-glass fabric roving used as reinforcement by Wang, (1999). The flexural moduli of E-glass fabric reinforced polyester (Wresipol 31466) composites at various fibre volume fractions are presented in Figure 3.14 on page 151. Extrapolation of the linear regression line presented in Figure 3.14 to a fibre volume fraction of 57% yields a predicted flexural modulus of 25.42 GPa. At the same fibre volume fraction the flexural modulus of a UnM flax reinforced unidirectional composite is approximately 27% lower. Hepworth *et al.*, (2000) report epoxy composites reinforced with glass fibre CSM having a flexural modulus and flexural strength of 34 GPa and 527 MPa at a fibre volume fraction of 60%.

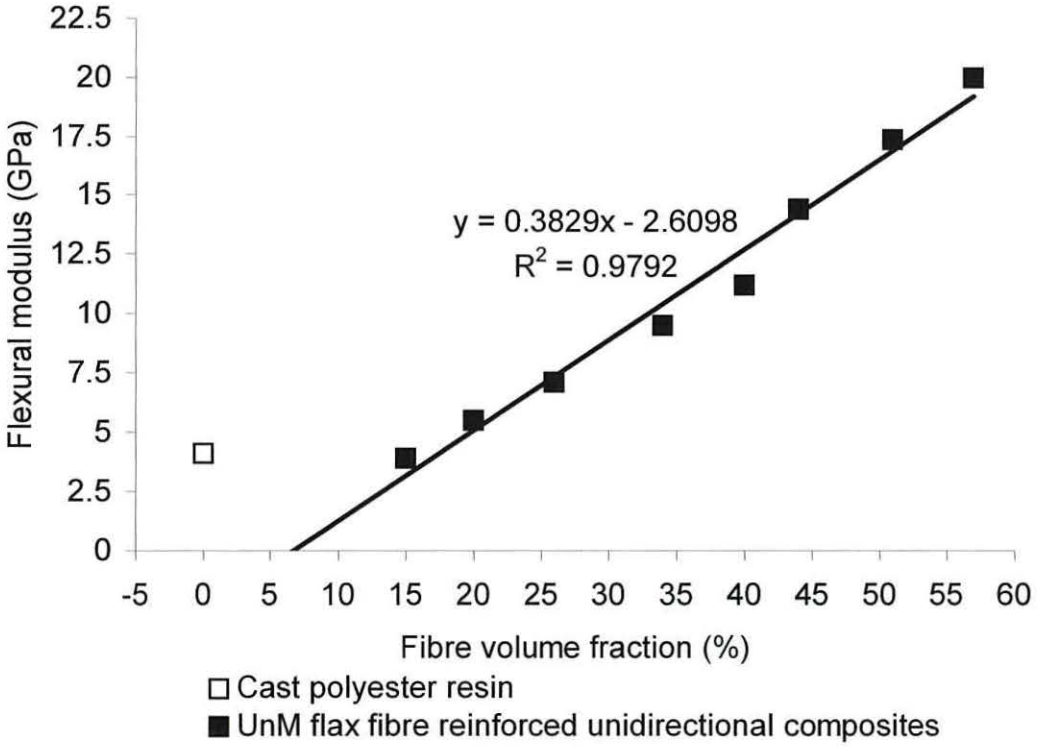


Figure 6.5 Flexural modulus of UnM flax reinforced unidirectional polyester composites against fibre volume fraction.

The specific stiffness (E_c / ρ_f) of the CSM epoxy composite fabricated and tested by Hepworth *et al.*, (2000) was 18.08 GPa, whilst a comparable UnM flax polyester composite in this study has a specific stiffness of 15.62 GPa. Although lower, it does demonstrate that there is potential for flax unidirectional polyester composites to compete with glass fibre reinforced PMC's in terms of flexural stiffness, due to their densities being generally lower.

6.3.2.1 Nature of the flexural stress-strain behaviour

Presented in Figure 6.6 are the flexural stress-strain curves of UnM flax reinforced unidirectional composites.

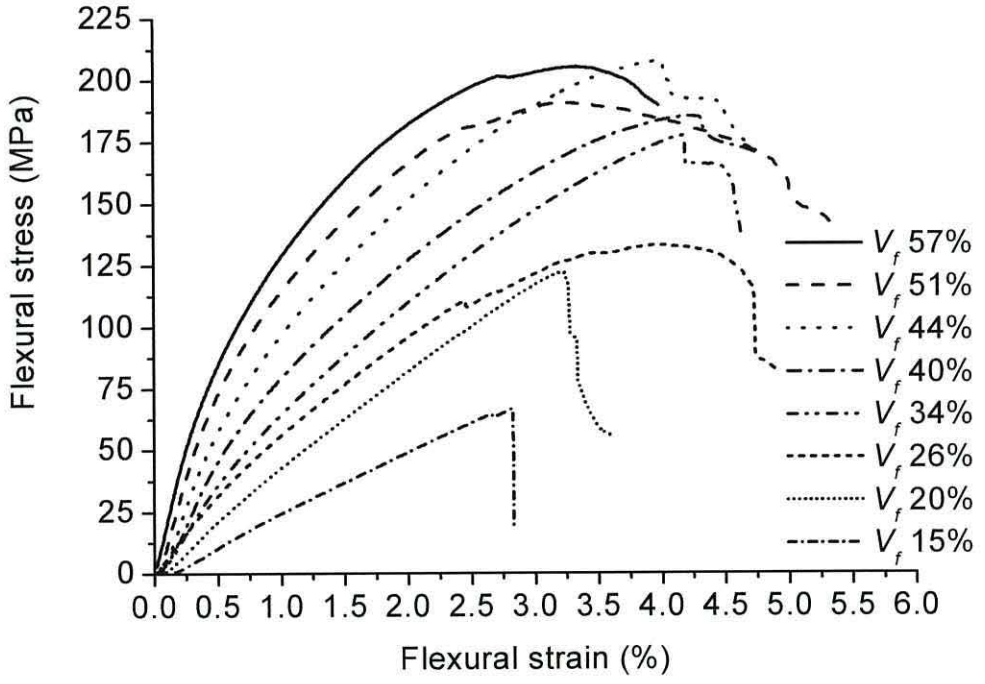


Figure 6.6 Flexural stress-strain curves of UnM flax reinforced unidirectional polyester composites.

Using Origin® software a tangent was placed along the initial linear region of each flexural stress-strain curve. The strain and stress at which the stress-strain curve deviates away from the tangent line were recorded for each specimen and are presented in Table 6.1. As may be observed from Table 6.1, with the exception of one, the stress at which non-linear behaviour starts gradually decreases, and the strain gradually increases as there is a decrease in the composite's fibre volume fraction.

Table 6.1 Stress and strain at which UnM flax reinforced flexural specimens stress-strain curves depart from linear behaviour.

V_f (%)	*Strain (%)*	*Stress (MPa)*
57	0.24	50.50
51	0.28	44.74
44	0.34	40.41
40	0.41	37.84
34	0.39	28.26
26	0.19	13.11
20	0.57	24.49
15	0.82	19.82
0 (resin)	1.67	68.60

Composites with high fibre contents had greater regions of viscoelastic and/or plastic deformation occurring. Composites with fibre volume fractions of 34% and above, remained intact, with failure occurring predominantly in shear. Delamination between fibres close to the tensile face was also noticeable on the edges of flexural specimens. Fibres close to the compression face of specimens from composites with fibre volume fractions of 44% and above had risen from the surface. It was not possible to visually detect if this fibre bucking had caused fibre failure. Flexural specimens with fibre volume fractions of 26% showed clear evidence of shear failure followed by a tensile

failure shearing across the width of the tensile face. Flexural specimens with fibre volume fractions of 20 and 15% remained as a whole, but failed in tension with a crack propagating straight across the width of the tensile face and some distance vertically up through the thickness of the specimen. The regions of elastic behaviour exhibited by the UnM flexural specimens end at relatively low values of strain, indicating that microstructural deformation arising from microstructural events are occurring within or around the reinforcement and/or at the fibre to matrix interface.

6.3.3 Unmodified flax composites impact properties

Figure 6.7 shows the Charpy impact strengths of UnM flax reinforced polyester composites against fibre volume fraction.

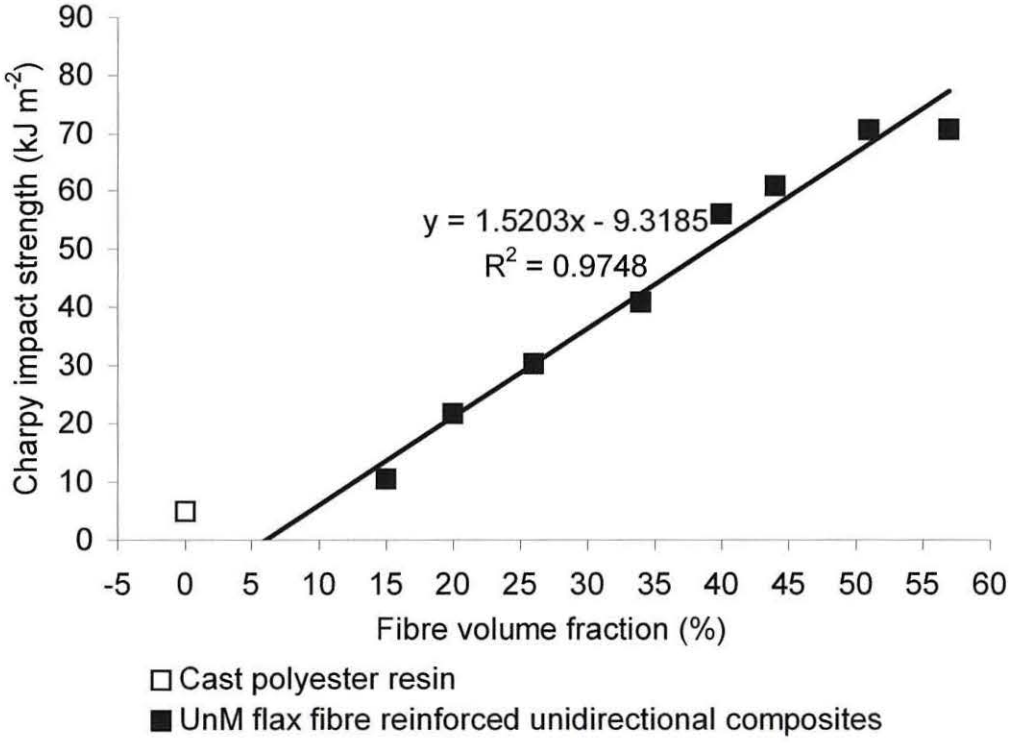


Figure 6.7 Charpy impact strength of UnM flax reinforced unidirectional polyester composites against fibre volume fraction.

As may be observed from Figure 6.7, with increasing fibre volume fraction the Charpy impact strength improves, to an extent that at a fibre volume fraction of 57% a notched Charpy impact strength of 70.8 kJ m^{-2} was achieved. Notched Charpy impact specimens with fibre volume fractions of 40% and above failed on the tensile face with fibre failure and pull-out visible. In addition, multiple shear failures were clearly noticeable on the edges of these specimens and fibre buckling had also occurred on the compression faces. Failure occurred on the tensile face of impact specimens with fibre volume fractions of 34% and 26%. The majority of these specimens remained as a whole; one specimen did fail completely showing no evidence of delamination between fibres. Impact specimens with fibre volume fractions of 15% and 20% failed completely into two sections exhibiting a more brittle mode of failure than the other specimens with higher fibre volume fractions. Table 2.7 on page 90 reports the work of fracture of two unidirectional polyester composite systems reinforced with natural fibre (jute and sunhemp). The work of fracture of composites from this investigation compare very well to those of other unidirectional polyester composites reported in Table 2.7.

6.3.4 Unmodified flax composite tensile properties

Presented in Figure 6.8 are the tensile strengths of UnM flax reinforced polyester composites against fibre volume fraction. The tensile strength of composites increased in an essentially linear fashion with increasing fibre volume fraction, at a fibre volume fraction of 57% a tensile strength of 303 MPa was achieved. Figure 6.9 shows the UnM flax reinforced composites Young's modulus against fibre volume fraction. As may be observed, Young's modulus increased in a practically linear fashion with fibre volume fraction following a ROM relationship. Extrapolation of the linear regression line featured in Figure 6.9, yields a theoretical fibre Young's modulus of some 45 GPa. This value is somewhat lower than that often quoted in the literature. For example, Ivens *et al.*, (1997) quote the Young's modulus of flax to be in the region of 50 to 70 GPa, whilst Bledzki *et al.*, (1996) quote Young's modulus for flax to be 100 GPa. Nevertheless, other workers have quoted much lower values, with Sridhar *et al.*, (1982), finding the

Young's modulus of flax to be 28 GPa. The figure reported by Davies and Bruce, (1998) at 52 GPa and the Young's modulus reported by Baley *et al.*, (2002) of 54 GPa are in broad agreement with the theoretical value predicted in this work.

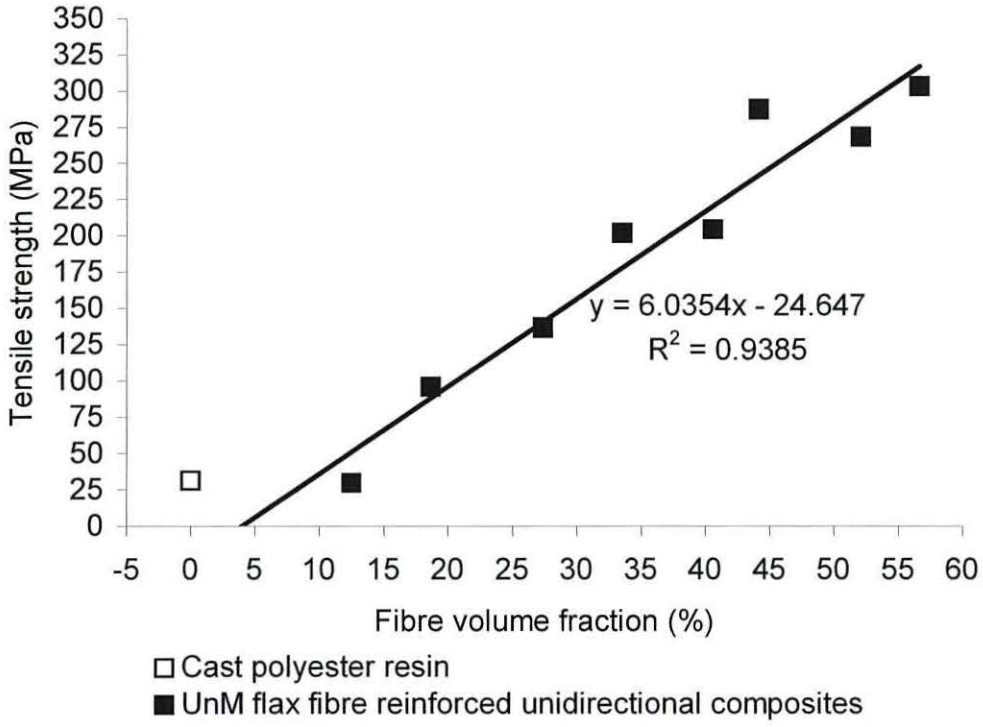


Figure 6.8 Tensile strength of UnM flax reinforced unidirectional polyester composites against fibre volume fraction.

Davies and Bruce, (1998) also found that fibre properties are strongly dependent upon the occurrence of fibre damage. This may well partly explain the wide variation in values for Young's modulus reported in the literature. Section 2.9.1 on page 81 also reports other factors that influence flax fibre properties, these include; fibre structure, test conditions and the method used to measure the dimensions of the fibres themselves.

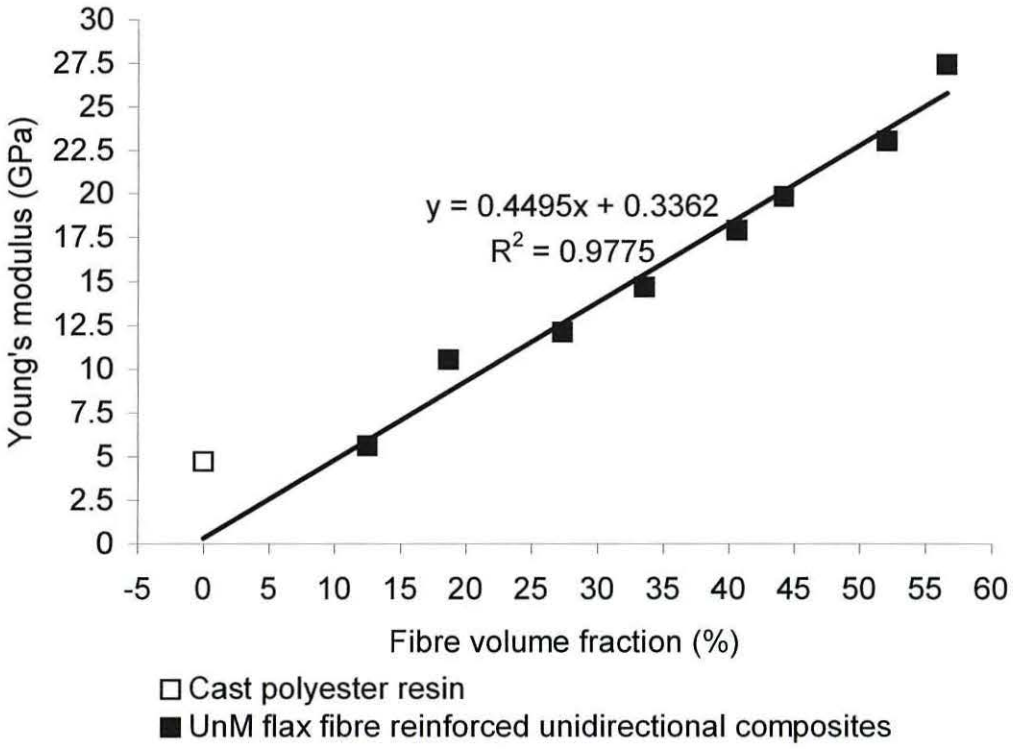


Figure 6.9 Tensile Young's modulus of UnM flax unidirectional polyester composites against fibre volume fraction.

6.3.4.1 Nature of the tensile stress-strain behaviour

Presented in Figure 6.10 are the tensile stress-strain curves of UnM flax composites at various fibre volume fractions. The UnM flax unidirectional composite's tensile stress-strain behaviour is initially linear, but at low strains non-linear behaviour is observed. The point at which non-linear behaviour commences is noticed by the distinct change in the gradient of the stress-strain curves. This 'knee' is more noticeable in composites that contain higher volume fractions of fibre. This deformation has been investigated further and is detailed in the following sections, different UnM flax unidirectional composites have been studied for the deformation investigation than those that have been reported previously in Figure 6.3 to Figure 6.10.

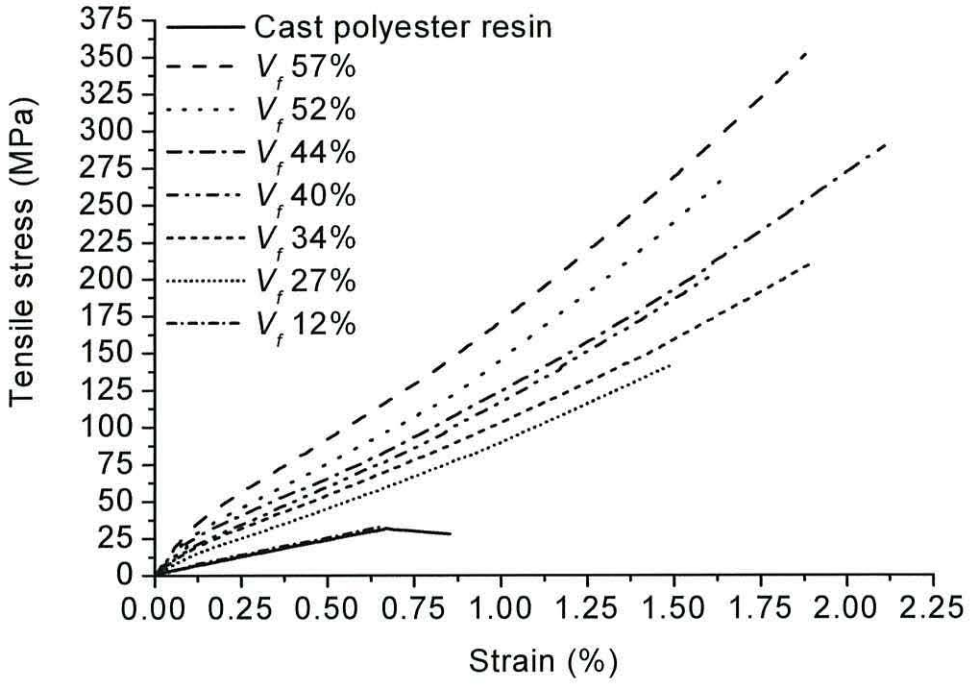


Figure 6.10 Tensile stress-strain curves of UnM flax reinforced unidirectional polyester composites at various fibre volume fractions.

6.3.5 Mechanical properties of unmodified, modified flax and E-glass composites

Extra UnM flax fibre composites were fabricated with fibre volume fractions ranging from 53 to 60%. These composites were subsequently tested in tension to investigate their tensile deformation and fracture behaviour. The average fibre volume fraction and density of unidirectional UnM flax, PrA (propionic anhydride) modified flax, MeA (methacrylic anhydride) modified flax and E-glass fibre reinforced composites are presented in Table 6.2.

Presented in Table 6.3 is a summary of the tensile mechanical properties of UnM flax fibre, PrA modified flax fibre, MeA modified flax fibre and E-glass fibre reinforced unidirectional polyester composites.

Table 6.2 Average fibre volume fraction and density of all four types of reinforced unidirectional polyester composites.

Reinforcement type of unidirectional composite	*Number of composites*	*Average V_f (%)*	*Composite density (kg m^{-3})*
UnM flax	10	57.6 (2.1)	1302 (27)
PrA flax	4	55.2 (2.3)	1287 (31)
MeA flax	4	59.6 (4.3)	1288 (53)
E-glass	4	42.4 (3.5)	1684 (74)

Note: figures in parentheses are standard deviations.

Table 6.3 A summary of the average tensile mechanical properties of UnM flax fibre, PrA modified flax, MeA modified flax and E-glass fibre reinforced unidirectional polyester composites.

Reinforcement type of unidirectional composite	*Average V_f (%)*	*Young's modulus (GPa)*	*Tensile stress at break (MPa)*	*Strain at maximum stress (%)*
UnM flax	57.6 (2.1)	29.9 (1.8)	304 (29)	1.73 (0.10)
PrA flax	55.2 (2.3)	27.8 (2.3)	234 (17)	1.12 (0.05)
MeA flax	59.6 (4.3)	27.8 (3.1)	165 (23)	0.79 (0.15)
E-glass	42.4 (3.5)	30.6 (2.2)	695 (60)	2.37 (0.36)

Note: figures in parentheses are standard deviations.

Presented in Figure 6.11 are representative tensile stress-strain curves of UnM flax, PrA modified flax, MeA modified flax and E-glass fibre reinforced polyester composites. As may be observed, E-glass fibre reinforced composites practically exhibit linear behaviour up until the point of failure, which occurs at a strain of over 2% (for clarity, the stress-strain curve belonging to the E-glass fibre reinforced composite, featured in Figure 6.11 has been truncated).

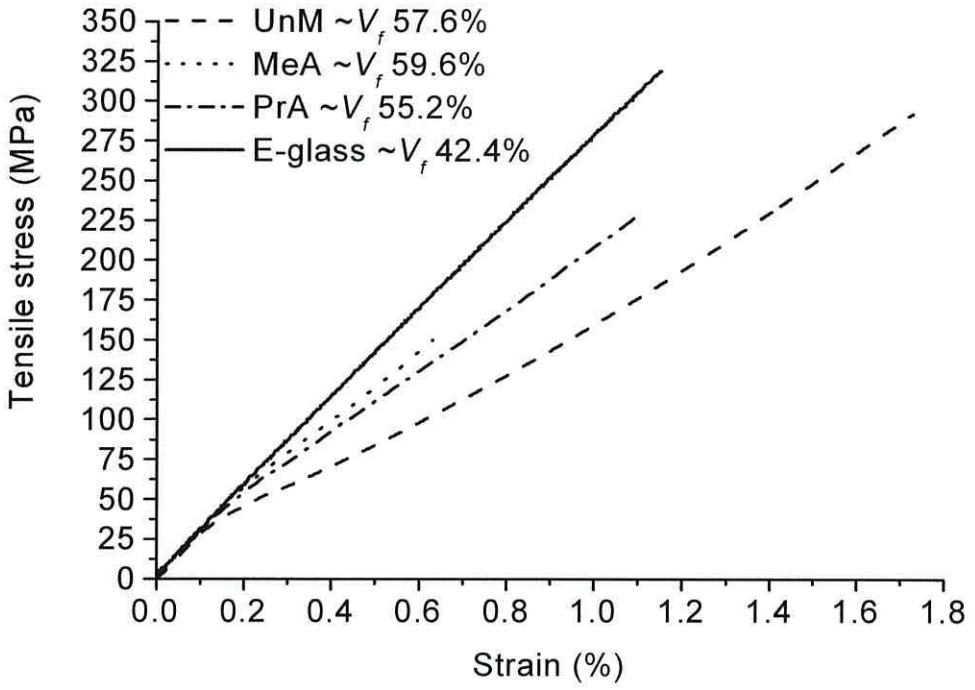


Figure 6.11 Representative tensile stress-strain curves of UnM flax, MeA modified flax, PrA modified flax and E-glass fibre reinforced unidirectional polyester composites.

The stress-strain curves of composites reinforced with UnM flax fibre exhibited initial linear behaviour to approximately 0.1% strain. At this strain a distinct change in the gradient of the stress-strain curve is apparent. Both modified flax reinforced composites also exhibit initial linear behaviour followed by a reduction in the gradient of the stress-strain curve, however, the change in gradient of the modified flax reinforced composites stress-strain curves is not as pronounced as it is for the UnM flax composites.

The average fibre volume fraction of E-glass fibre reinforced unidirectional composites is significantly lower (approximately 26% lower) than the fibre volume fraction of UnM flax reinforced composites but the density of E-glass composites is significantly higher (Table 6.2). When density is taken into account, the specific stiffness (E / ρ) of the UnM

flax fibre reinforced unidirectional composites is approximately 26% higher than that of E-glass fibre reinforced unidirectional polyester composite material. This is significant and could have real benefits in practice.

Comparing the average Young's modulus (Table 6.3) of UnM flax reinforced composites to both the Young's moduli of PrA and MeA modified flax fibre reinforced composites, it may be observed that there is little difference. Any variation observed may well be attributed to differences in the composite's fibre volume fractions. The Young's modulus of composites presented in Table 6.3 was measured in the linear elastic region of stress-strain curves, before the onset of any irreversible events, therefore the relatively low variation between moduli is perhaps not unexpected. These results also suggest that both modifications to flax have not degraded the Young's modulus of the flax fibres themselves.

However, both fibre modifications have affected the tensile strength and strain to failure of unidirectional composite material in a significant manner when compared to the UnM flax reinforced composites. The tensile strength and strain to failure of UnM flax reinforced composites was on average 304 MPa and 1.73% respectively. Unidirectional polyester composites reinforced with flax fibre modified with propionic anhydride had a lower tensile strength (234 MPa) and strain to failure (1.12%) than UnM flax composites. The reduced tensile strength and strain to failure of PrA modified flax composites may be attributed to the modified interfacial properties, thus causing a reduced crack stopping ability of the composite material and an increased tendency for cracks, once initiated, to propagate catastrophically. Unidirectional polyester composites reinforced with methacrylic anhydride modified flax exhibit even lower tensile strengths and strain to failures than PrA modified flax reinforced composites. On average, the tensile strength and strain to failure of MeA modified flax unidirectional composites is 165 MPa and 0.79% respectively. The differences observed between MeA and PrA modified flax composites may be the result of the degree of adhesion between phases, as discussed in Section 6.2.3.1 on page 256.

Evidence for improved bonding with both modified flax fibre reinforcements and the matrix was provided by fractographic examination. Plate 6.3 shows the macroscopic failure observed in UnM flax (A), MeA modified flax (B) and PrA modified flax (C) fibre reinforced unidirectional polyester composites.

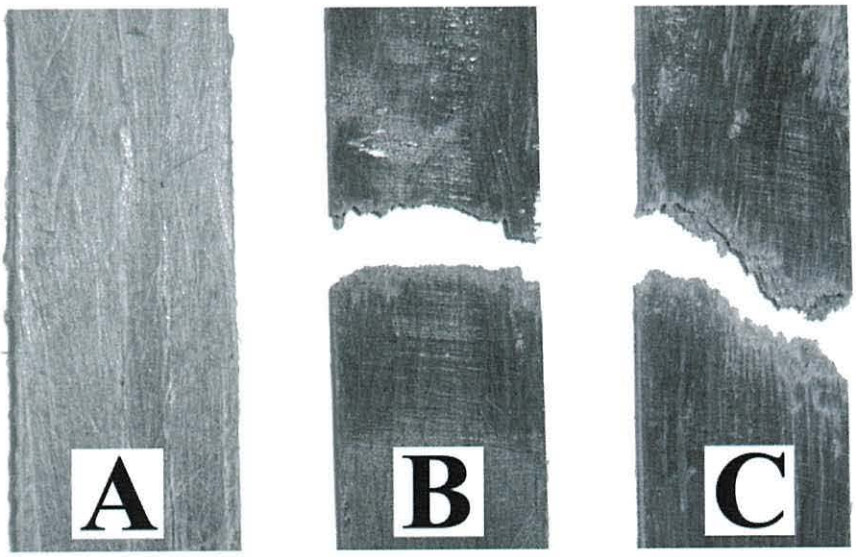
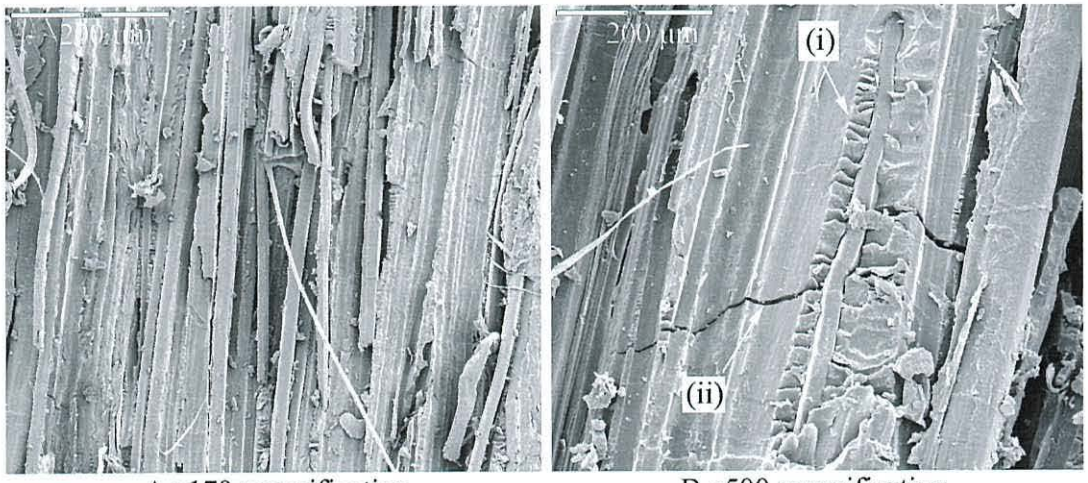


Plate 6.3 Failed tensile specimens: (A) unmodified, (B) methacrylic anhydride and (C) propionic anhydride modified fibre reinforced unidirectional unsaturated polyester composites.

In the case of unmodified fibre reinforced composites, failure occurred through delamination and wide-scale debonding between fibre and matrix (A). With both the MeA and PrA modified flax fibre reinforced polyester composites, the mode of failure was observed to change to one of brittle tensile (B) or a mixture of tensile and shear failure (C). Improved interfacial adhesion between methacrylic anhydride modified hemp fibre and an unsaturated polyester matrix has been reported previously (Sèbe *et al.*, 2000).

Examining the fractured surfaces of UnM flax reinforced composite material through a SEM revealed that there was apparently little adhesion between fibre and matrix. Plate 6.4 shows SEM micrograph evidence of extensive flax fibre separation from the

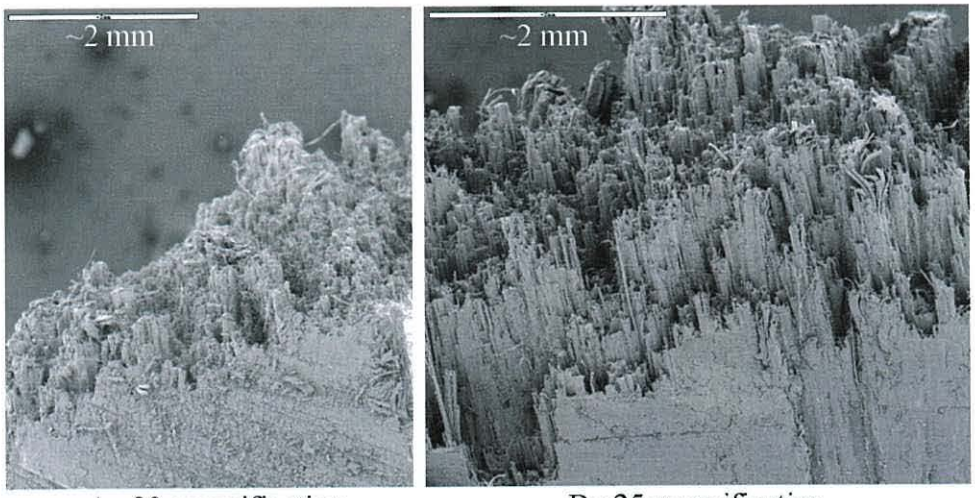
encapsulating polyester matrix (Plate 6.4 A) and river lines (Plate 6.4 B at 'i'), as described by Hull, (1999). Significant transverse cracks (Plate 6.4 B at 'ii') can also be observed that have propagated across the composite a distance of several fibre diameters.



A x170 magnification B x500 magnification

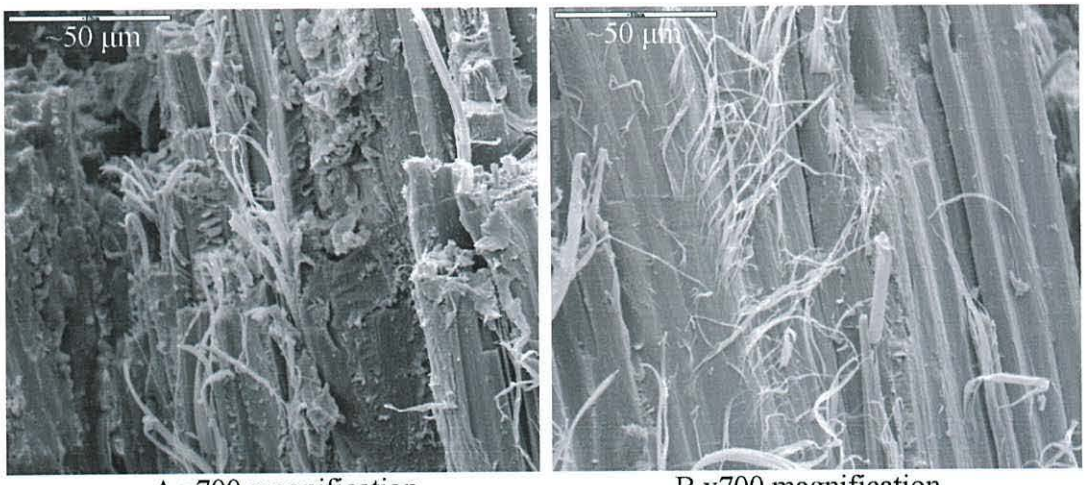
Plate 6.4 Scanning electron microscope micrographs of a failed UnM flax fibre reinforced unidirectional polyester composite. The fracture surface is parallel to the fibre axis.

Presented in Plate 6.5 are SEM micrographs showing the overview of two fractured surfaces; the first is MeA modified flax (A) and the second is PrA (B) modified flax fibre reinforced unidirectional polyester composites. Plate 6.6 shows two SEM micrographs of close up sections of the fibre surfaces from MeA modified flax (A) and PrA modified flax (B) fibre reinforced unidirectional polyester composites. Plate 6.5 and Plate 6.6 show that an intimate bond was formed between reinforcement and matrix with the aid of chemical modification. Both MeA and PrA modified flax fibre reinforced composites have fracture surfaces that display little evidence of fibre pull-out, the surfaces exhibit a distinct 'blocky' appearance (Plate 6.5 A and B). Inspecting the fibre surfaces at higher magnification it was observed that PrA modified flax fibre composites had somewhat greater pull-out and a greater degree of inter laminar shear failure (Plate 6.6 B) than MeA modified flax reinforced composites.



A x20 magnification B x25 magnification

Plate 6.5 Scanning electron microscope micrographs of a failed MeA modified flax (A) and PrA modified flax (B) fibre reinforced unidirectional polyester composite.



A x700 magnification B x700 magnification

Plate 6.6 Scanning electron microscope micrographs of a failed MeA modified flax (A) and PrA modified flax (B) fibre reinforced unidirectional polyester composite. Both micrographs show evidence of fibrillation at the fibre surface.

This observation is consistent with the lower adhesion that might be expected in this modified (PrA) flax fibre reinforced composite system. The presence of what appears to be extensive fibrillation on the fibres surfaces of both types of modified flax reinforced

composites suggests that strong interfacial bonding between the fibre and matrix occurred (Plate 6.6 A and B). Fibrillation was probably caused by regions of the fibre being well bonded to the matrix. As failure occurred, the matrix may have been torn from the underlying layers of the fibre.

Flax sliver modified with propionic anhydride may have interacted better with the matrix allowing for improved wet out of the fibres because of the greater hydrophobicity introduced to them. True chemical bonding is thought to exist between the surface of flax fibre modified with methacrylic anhydride and the matrix (Hill and Cetin, 2000). As Sèbe *et al.*, (2000) states, improving the degree of adhesion between fibre and matrix is detrimental to the material's resistance to the propagation of cracks, because it changes the mode of failure from fibre pull-out to fibre fracture which results in less energy involved in the failure of the composite because there is little frictional sliding occurring. Hughes *et al.*, (1999) suggests that micro-compressive defects lead to fibre failure but they also introduce stress concentrations into the composite structure. It follows that defects such as these coupled with a high degree of adhesion causes a reduction in the tensile strength and strains to failure (Table 6.3). It is not known if modifying flax fibre with methacrylic anhydride reduces the fibre's tensile strength, if this was the case, composites reinforced with MeA modified flax fibre may exhibit lower tensile strengths. This possibility cannot be ruled out in the case of the above MeA flax reinforced composites. With the fractographic evidence obtained, it is very difficult to categorically decide if interfacial bonding is greater in the MeA modified flax fibre reinforced unidirectional composites or not.

6.3.5.1 Deformation behaviour

Presented in Figure 6.12 is a representative tensile stress-strain curve of an UnM flax reinforced unidirectional polyester composite. The stress-strain curve is not linear and as highlighted in Figure 6.12 distinct regions of the curve can be identified. The initial

portion of the curve (A) is essentially linear and remains so up to an average strain of 0.06%. At an average strain of 0.12% (B) there is a distinct knee in the curve denoted by a rapid change in slope. In the following region (C) there is a drop in the modulus initially, but then a gradual increase (D) which precedes failure at E. This behaviour is entirely different from that observed in glass fibre reinforced unidirectional polyester composites, which exhibited predominantly linear behaviour to failure (Figure 6.11 on page 276).

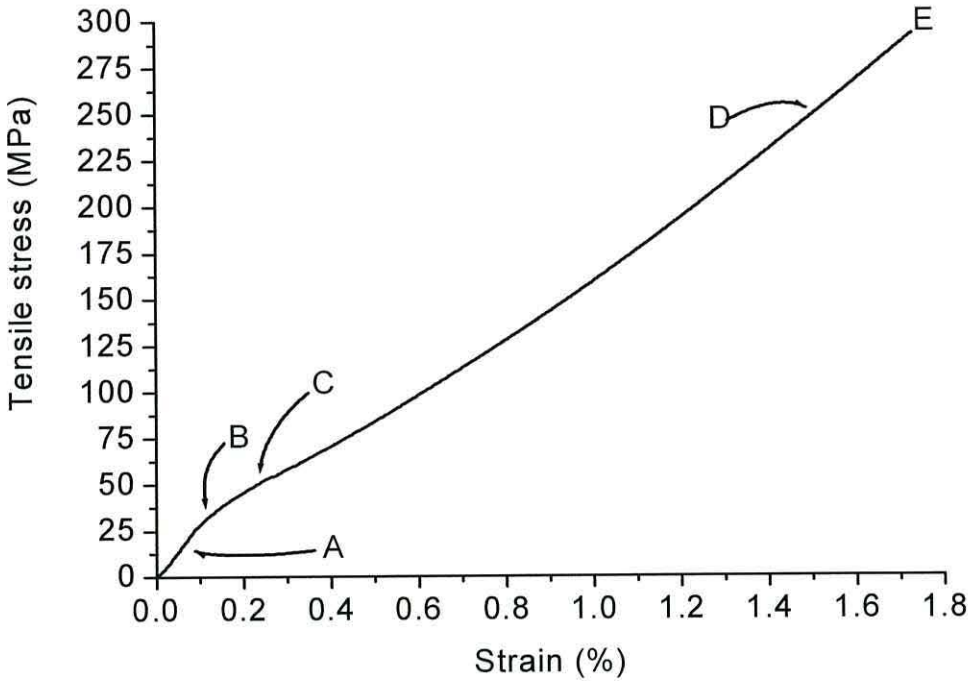


Figure 6.12 Representative tensile stress-strain curve of an UnM flax fibre reinforced unidirectional polyester composite.

To investigate if the processes leading to the occurrence of the knee (B) are reversible or not, UnM flax fibre reinforced unidirectional composites was loaded to various pre-set points along the stress-strain curve prior to, and after, the knee and then unloaded. Presented in Figure 6.13 is a portion from the loading-unloading curve produced from

UnM flax reinforced composite. As may be observed from Figure 6.13, up to point A, in the linear region of the curve before the onset of the knee, the unloading record is effectively overlaying the loading record, indicating that the process is reversible (elastic behaviour).

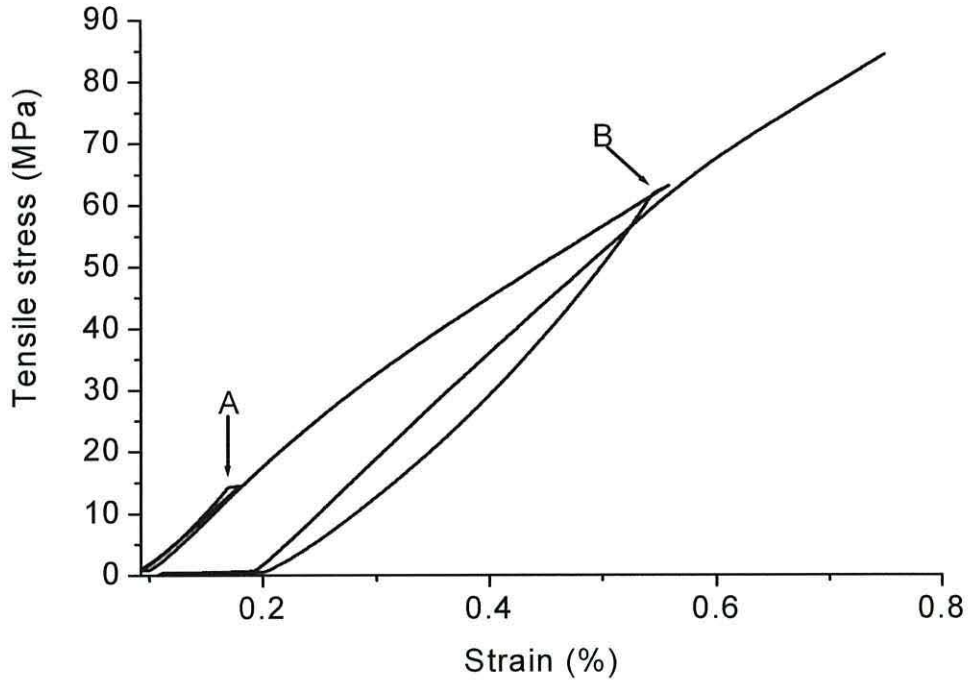


Figure 6.13 Potion of loading-unloading curve (region up to failure not shown) for a UnM flax fibre reinforced unidirectional polyester composite, loaded to a point just below the yield (A) and after the yield point (B).

Loading again to a set point after the knee (point B), followed by unloading, results in hysteresis, indicating that there is a degree of irreversibility in the process arising from microstructural damage, possibly accompanied by some non-linear or viscoelastic behaviour. Once the load is removed entirely after loading to point B, a permanent deformation has been imparted to the composite. Thus, it appears probable that at least parts of the processes associated with the 'knee' are irreversible microstructural events,

leading to what might be termed a yield point. An average yield stress can be associated with this point, having a value of around 36 MPa.

6.3.5.2 Effect of varying the interfacial adhesion

A number of processes such as fibre fracture, matrix yielding, matrix fracture and fibre to matrix debonding could be responsible for the microstructural damage that is causing the yielding of the stress-strain curves of UnM flax reinforced composites. As previously mentioned, the influence that fibre to matrix debonding has upon this behaviour was investigated by varying the degree of interfacial adhesion and by analysing and comparing the stress-strain curves of composites reinforced with unmodified and modified (MeA and PrA) flax fibre. Altering the degree of interfacial adhesion *via* two chemical modifications did modify both the fracture and stress-strain behaviour (see Figure 6.11 on page 276) of these composite systems when subjected to tensile loading as described in Section 6.3.5 on page 274. An analysis of the tensile stress-strain behaviour of UnM flax, MeA modified flax and PrA modified flax fibre reinforced composites is presented in Table 6.4.

Table 6.4 Analysis of the influence of fibre to matrix adhesion upon yielding behaviour.

Reinforcement type	*Modulus*			*Yield onset*		*Yield point*	
	Young's modulus (GPa)	*Tangent modulus (GPa)*	*Difference (%)*	*Onset strain (%)*	*Onset stress (MPa)*	*Yield strain (%)*	*Yield stress (MPa)*
UnM	29.9 (1.8)	13.7 (0.9)	-54	0.06 (0.01)	18.1 (3.9)	0.12 (0.01)	32.3 (2.3)
PrA	27.8 (2.3)	18.6 (2.7)	-33	0.13 (0.02)	38.2 (6.2)	0.18 (0.02)	48.4 (5.7)
MeA	27.8 (3.1)	18.1 (2.9)	-35	0.11 (0.01)	31.1 (2.8)	0.17 (0.04)	46.6 (9.8)

Note: figures in parentheses are standard deviations.

Individual stress-strain curves were analysed as follows. Tangent moduli were constructed using Origin® software, from which values of Young's modulus and the tangent modulus in the region immediately after the yield point (region C in Figure 6.12 on page 282) were computed. The intersection of these two tangents provided a value for yield strain, thus yield stress was taken directly from the stress-strain curve at the yield strain. The initial departure from linearity was used to determine the onset of yielding, and values for the onset stress and strain are presented along with Young's and tangent modulus in Table 6.4. The average Young's modulus of UnM flax and both MeA modified flax and PrA modified flax reinforced composites are very similar (on average a difference of 2.1 GPa separate them). However, following yielding, the reduction in modulus (tangent modulus) is significantly less in the modified flax fibre reinforced composites than it is for UnM flax reinforced composites, indicating that some modification of the microstructural processes has occurred with both modified flax reinforced composites. The tangent modulus of UnM flax fibre reinforced composites was on average 54% lower than the initial Young's modulus, whereas the tangent modulus of both PrA and MeA modified flax fibre reinforced composites was on average 33 and 35% lower than their average initial Young's moduli respectively. Additionally, the values for both yield stress and yield strain are increased for both composite systems reinforced with chemically modified flax fibre. It is interesting to note that there is little difference between PrA and MeA modified flax reinforced composites, in terms of the loss in composite stiffness after yielding or the values of yield stress and strain, indicating that the degree of interfacial adhesion provided by these two modification types has little direct effect upon the micromechanical deformation and failure mechanisms leading to the yielding phenomenon. Whilst, as discussed in Section 6.2.3.1 on page 256, the degree of interfacial adhesion provided by these two modification regimes might be expected to differ, it is possible that some threshold value of interfacial adhesion has been exceeded with both modification methods, beyond which further improvements in adhesion are not manifested in the properties of the composites. This line of reasoning is supported by the observation that although both modifications influence the onset of yielding behaviour by increasing the onset values greater than those recorded for UnM flax reinforced composites; little difference between the two modified reinforced

composite systems is noted. This may again indicate that perhaps other factors play a more dominant role in the yielding behaviour. It will be recalled, however, that a significant difference in the failure stress and strain was observed with the two types of modified fibre reinforced unidirectional polyester composites (Table 6.3 on page 275) indicating that in terms of crack propagation at least, the degree of adhesion between the phases does affect the composite system significantly.

6.3.5.3 Acoustic emissions

To try and understand microstructural failure events contributing to the yielding behaviour of composites reinforced with UnM flax, acoustic emissions analysis was employed.

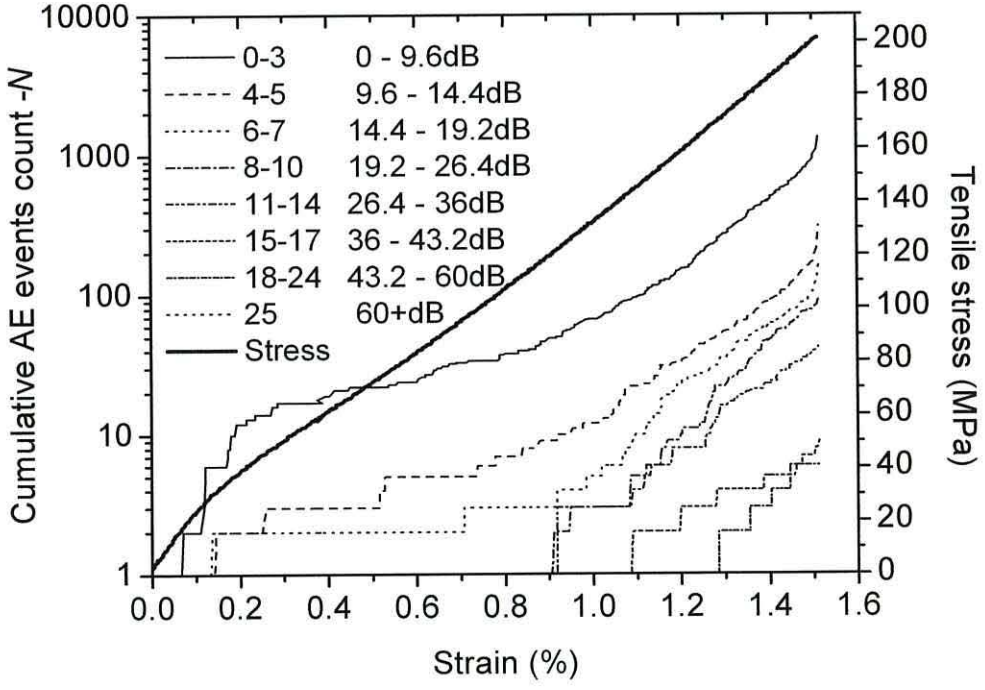


Figure 6.14 Acoustic emissions from an UnM flax reinforced unidirectional polyester composite, showing cumulative acoustic emission events as a function of strain, together with the corresponding stress-strain response of the composite.

Figure 6.14 shows a representative stress-strain curve for UnM flax reinforced composite with accompanying AE emissions, presented as cumulative event counts *N*. The system comprised of 25 channels each 2.4 dB width. To facilitate simpler analysis, the channels were grouped into 8 bands as shown in Figure 6.14. As may be observed, AE events were first recorded at strain values of less than 0.1% for channels 0 to 3 (0 to 9.6 dB, lower energy events) and between 0.1 and 0.2% strain for channels 4 to 7 (9.6 to 19.2 dB). Higher energy events were recorded and are displayed in channels 8 to 25. As high stored strain energy is released from within the composites as major matrix cracking occurs or fibre pull-out the events are recorded within the higher energy channels. Guild *et al.,* (1985) also used a similar system but one that contained 50 channels each 1.2 dB width. Guild *et al.,* (1985) excluded channels 0 to 3 (0 to 4.8 dB) from their analysis, because they found that background noise was present. However, in this work, events occurring within these channels have been included and as may be observed from Figure 6.14, cumulative counts arising from within these channels (0 to 3) rise rapidly between 0.1% and around 0.3% strain, which corresponds with the yield point. This evidence suggests that microstructural failure events do indeed occur in the vicinity of the yield point. From the AE analysis employed in this work, it is not possible to make any firm conclusions about the exact nature of these events. However, it is possible to postulate that the AE emissions that initiate at approximately 0.9% strain originate from a mixture of flax fibre fracture and severe matrix cracking. There are two reasons why these AE emissions are thought to be from these events. The strain to failure of flax has been reported by Davies and Bruce, (1998) to occur at an average strain of 1.33%. Davies and Bruce, (1998) also report that the standard deviation was high at 0.56. Ivens *et al.*, report that the strain to failure of flax occurs between 1.3 and 3.3% strain. Hemp is a similar bast fibre to flax and Eichhorn and Young (2004) found that hemp fibre's strain to failure was on average 0.8%. As may be observed from Figure 6.14, the AE emissions detected at 0.9% strain and onwards originate from channels 8 and above, indicating the occurrence of higher energy events within the composite system. As strain increases, and becomes nearer the reported average strain for flax fibre failure, the number of these higher energy events increases, therefore it is possible that the AE emissions detected are as result of flax fibre fracture. The tensile strain to failure of cast polyester resin was

found to occur at an average strain of 0.68% (Table 3.3 on page 123). It seems reasonable to postulate that the AE events detected in channels 4 and 5 at approximately 0.7% strain are from minor matrix cracks developing in the system. As may be observed from Figure 6.14, within channels 4 and 5 the number of AE events increase with greater frequency at the strain of 0.7%. Some of the AE events occurring within higher channels (8 and above) and at higher strains are also thought to originate from matrix cracking.

6.3.6 Discussion

Within the range of 53 to 60% fibre volume fraction, the Young's modulus and density of UnM flax fibre reinforced unidirectional polyester composites was found to be approximately 30 GPa and 1302 kg m^{-3} respectively. Using the ROM relationship (Equation 2.13 on page 63) and taking the E_m of the matrix to be 4.7 GPa (Table 3.3 on page 123) and assuming values of E_f for the fibre to be 76 GPa (Hull and Clyne, 1996), a comparable glass fibre reinforced composite with a fibre volume fraction of 57.6% might be expected to display a Young's modulus of around 45.7 GPa. If it is assumed that the density of the E-glass fibre is 2560 kg m^{-3} and the matrix density is 1180 kg m^{-3} (Table 3.3) then the hypothetical E-glass reinforced polyester composite density at a fibre volume fraction of 57.6% would be expected to be 1974 kg m^{-3}. Whilst the predicted value of the Young's modulus of the hypothetical E-glass reinforced composite is greater, when the lower density of the UnM flax fibre reinforced composite is taken into account, the specific stiffness values are comparable, the specific stiffness of E-glass fibre and UnM flax fibre reinforced polyester composites at a fibre volume fraction of 57.6% are 23.1 GPa and 22.9 GPa respectively. Thus, where stiffness is the main design criterion, natural fibre reinforced composites offer good possibilities in structural or semi-structural applications. However, whilst the initial Young's modulus of the flax reinforced composites is of value in engineering terms, the existence of a distinct yield point may well have a significant impact in practice, especially because it occurs at relatively low values of stress and strain.

The natural fibre reinforced PMC's studied in this work, revealed a departure from linear behaviour at low values of stress and strain when loaded in tension parallel to the direction of the fibre (Table 6.4 on page 284). The behaviour of similar E-glass fibre reinforced composites is entirely different to that of flax reinforced composites because they exhibit practically linear behaviour to the point of fracture (Figure 6.11 on page 276). The departure of UnM flax reinforced composites from linearity is probably associated with a true 'yield point', since when composites were loaded beyond this point and subsequently unloaded, they were found to have undergone permanent deformation (Figure 6.13 on page 283). The permanent deformation is thought to be the result of some form of microstructural damage and this contention is supported by the AE analysis (Section 6.3.5.3 on page 286) wherein acoustic events (admittedly of low intensity and low frequency) were first detected in the region of the yield point indicating incipient microstructural damage.

To find the origins of this behaviour, it is first necessary to consider the characteristics of the reinforcing fibres themselves as well as the interaction between the fibre and the matrix. Flax fibres are considerably different to E-glass fibres, as they are heterogeneous in structure and do not display linear elastic behaviour to failure. As previously mentioned in Section 2.9.1 on page 81, Hornsby *et al.*, (1997) observed strain hardening in the deformation behaviour of flax fibres when subjected to tensile loading, whilst Baley, (2002) also reports non-linear behaviour of flax fibres. Baley, (2002) states that the plastic deformation observed can be ascribed to the 'extension of defects' and 'reorganisation of the cellulose fibrils in the direction of the fibres axis'. It is known that flax fibres possess microstructural defects that are often referred to as kink bands (Plate 2.1 on page 87) and that the presence of such features results in a reduction in the strength and stiffness of fibres (Davies and Bruce, 1998). In addition, it is known that the defects located along flax fibres directly contribute to the non-linear straining behaviour of flax fibres (Baley, 2002). A manifestation of these defects when the fibres are used as composite reinforcement is that they lead to stress concentrations in the matrix in the proximity of the micro-compressive defects (kink bands) when the composite is loaded parallel to the axis of the fibre (Hughes *et al.*, 2000a; Hughes, 2000). As may be

observed from Plate 2.1 on page 87, kink bands occur with relative frequency along the length of a typical flax fibre, with perhaps fewer than 10 fibre diameters between successive defects (Hughes, 2000). Kink bands can be looked upon as being regions along a flax fibre that have lower stiffness, since when the fibres are strained the micro-compressive defects begin to extend (Baley, 2002). Eichhorn and Young, (2003) found that stress concentrations in a fibre exist at a strain of 0.6% at the site of defects when the fibre is embedded in a single fibre composite. Furthermore, it has been shown that the Young's modulus of flax is dependent upon the ratio of damaged to defect free fibre; a greater proportion of fibre damage leading to a reduced Young's modulus (Davies and Bruce, 1998). Whilst flax fibres are continuous, the presence of micro-compressive defects along the length of the fibre may have the effect of 'segmenting' the fibre, so that the whole fibre can be considered as a series of much shorter fibres of, typically, low aspect ratio, 'joined' by regions of fibre having lower relative stiffness. As such, although the fibres are continuous, they will act, in part, as short fibres, with the interfacial shear stress (τ_i) varying along the length of the fibre in a shear lag manner. The possibility that micro-compressive defects lead to uneven interfacial shear stress distributions is reported in the review paper by Eichhorn *et al.*, (2001).

Fibre defects and the consequent fibre straining behaviour together with manner of stress transfer that occurs in the flax reinforced unidirectional polyester composites studied in this work may help explain the presence of the observed yield point. The flax unidirectional composites behave in an elastic manner at low values of applied stress and strain before the yield point, elastic stress transfer occurs over each of the fibre segments, following a shear-lag type mechanism (Section 2.7.1.2 on page 49). The highest values of interfacial shear stress occur at the ends of each of the fibre segments, in the vicinity of the defects rather than at the actual ends of the fibre. As the composite system is strained further, the interfacial shear stress will gradually increase at the fibre segment ends until their magnitude reaches some threshold value at which the fibre starts to debond from the matrix. High stress concentrations at the fibre segment ends are possibly exacerbated by the morphology of the micro-compressive defects (Eichhorn *et al.*, 2001). Indeed, fibre to matrix debonding may not be the only mechanism responsible for the irreversible

microstructural damage observed in the flax reinforced unidirectional composite systems. Hughes *et al.*, (2000a) observed single hemp fibres embedded in an epoxy matrix loaded in tension parallel to the fibre axis. It was observed that the high stress concentrations surrounding the micro-compressive defects sometimes lead to matrix yielding and cracking. Hughes *et al.*, (2000a) also observed fibre fracture to occur at the locations of micro-compressive defects. Nevertheless, it seems likely that fibre to matrix debonding at least contributes to the yielding phenomenon observed, since modifying the fibres to improve adhesion between fibre and matrix had a significant effect upon the yield point. Both PrA and MeA modified flax fibre reinforced unidirectional polyester composites had average yield stresses of approximately 46 to 48 MPa at corresponding yield strains of about 0.18%, whereas the UnM flax reinforced polyester composites had an average yield stress of 32 MPa and a corresponding yield strain of approximately 0.12% (Table 6.4 on page 284). For both types of modified flax reinforced composites, such an increase would be expected if the critical value of interfacial shear stress (τ_i.) were increased, thereby restraining the onset of fibre to matrix debonding. This would also explain why the decrease in modulus is less for both types of modified flax reinforced composites; the reinforcing efficiency of the fibre segments would be retained since a greater proportion of stress transfer would take place through elastic processes. However, some inelastic processes must be occurring in both types of modified flax reinforced unidirectional polyester composites to give rise to such macroscopic yielding behaviour (Figure 6.11 on page 276). Although variation in the level of adhesion between fibre and matrix might be expected between both types of modified flax fibre, it is interesting to note that there is very little difference in the yield point between both composite systems. It might be thought that this would affect the onset of yielding, but as noted in Table 6.4 on page 284, there is little difference in the onset strain and stress between the two types of modified flax reinforced composites. As will be discussed in more detail later in this section, however, the level of adhesion (as opposed to mechanical interlocking) may well be a significant factor during fracture. Presumably, a threshold value of interfacial shear strength is reached with the PrA modified flax fibre, beyond which no further increase in bonding, as in the case of the MeA modified flax fibre reinforced composites, affects the yield point.

As mentioned in Section 6.3.4 on page 271, a theoretical Young's modulus for flax fibre was calculated by extrapolating the regression line obtained from the relationship between the Young's modulus of UnM flax reinforced composites against a range of fibre volume fractions. The theoretical Young's modulus of the flax was found to be some 45 GPa, which is in broad agreement with values published in the literature.

The post yielding modulus on average was found to be 13.7 GPa for an UnM flax reinforced composite, the initial Young's modulus was on average 29.9 GPa. As the average yield strain for UnM flax reinforced composites occurs at 0.12%, little or no flax fibre failure would be expected at the yield point as the likely strain for flax fibre failure is significantly higher (1-3%). Even if there were to be no further contribution to the composite stiffness from the polyester matrix following yielding, theoretically the stiffness would still be in the region of 26 GPa, from the contribution made by the fibres alone (based on a fibre volume fraction of 57.6%). This is clearly not the case, since the modulus immediately after the yield point (region C in Figure 6.12 on page 282) is on average only 13.7 GPa, which is just over half of that expected, and thus there must be an alternative explanation. As has been outlined above, if it is now assumed that the flax fibre acts as a series of shorter fibres, or rather segments as portrayed in Figure 6.15, with a defined aspect ratio, *s*, (=L/d) joined by regions of damaged fibre (at points A and B) having lower stiffness, it is possible to describe the elastic behaviour of the composite using shear lag theory. Equation 6.1 expresses the theoretical elastic stress-strain relationship before the onset of yielding (Hull and Clyne, 1996).

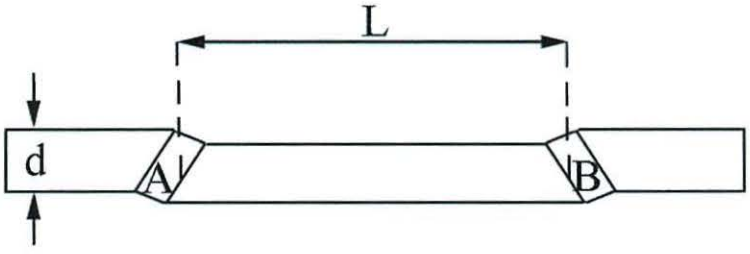


Figure 6.15 Schematic representation of a fibre segment bounded by damage in the form of micro-compressive defects at A and B. The aspect ratio of the segment (*s*) is defined as the segment length (L) divided by the fibre diameter (d).

Equation 6.1

$$\sigma_c = \varepsilon_1 \left[V_f E_f \left(1 - \frac{\tanh(ns)}{ns} \right) + (1 - V_f) E_m \right]$$

Where:

$$n = \left[\frac{2E_m}{E_f (1 + \nu_m) \ln(1/V_f)} \right]^{1/2}$$

σ_c The composites tensile stress.

ε_1 The applied composite strain.

ν_m The matrix Poisson's ratio assumed to be 0.35 (Hull and Clyne, 1996).

As it is assumed that there are no defects in each of the flax fibre segments, the stiffness of the fibre free from defects, or having the defects 'pulled-out' is taken to be 90 GPa as Davies and Bruce (1998) observed, flax ultimates tested at 50% RH with 0% fibre damage to have a modulus between approximately 86 and 93 GPa (values taken from a presented graph). Baley, (2002) using a model assuming small diameter fibres with little defects estimated the Young's modulus of flax to be 87.3 GPa which was found to be very close to the average experimental value of 91.8 GPa. Therefore it is possible to construct theoretical stress-strain curves for the unidirectional composite system using Equation 6.1, assuming that the fibre segment stiffness is 90 GPa, and taking the modulus of the matrix to be 4.7 GPa as found from a previous investigation (Table 3.3 on page 123) and the composite's fibre volume fraction to be 58%. Figure 6.16 shows, for a range of values of s, the theoretical stress-strain curves in the region before the yield point. As the aspect ratio increases, the derived composite modulus increases and reaches a maximum value as $s \rightarrow \infty$. As the aspect ratio decreases to 5, a value that might represent the aspect ratio in real flax fibre segments, a derived theoretical modulus of approximately 28 GPa is obtained. This theoretical modulus is significant as it is very similar to the experimental values obtained in this work.

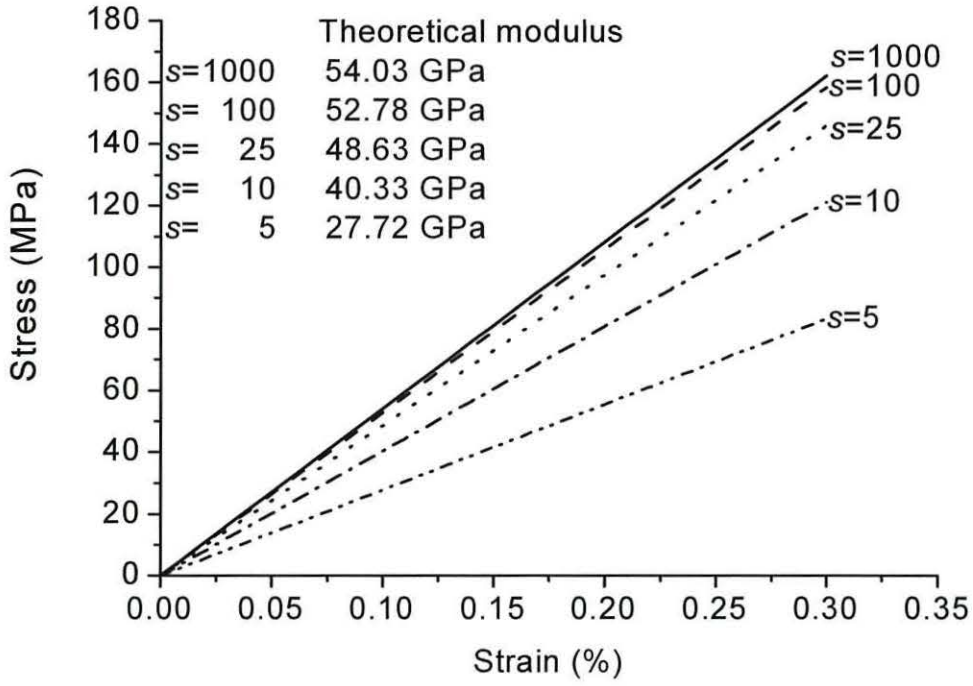


Figure 6.16 Theoretical stress-strain curves in the region before the yield point, for a range of values of *s* (aspect ratio).

Many assumptions have been made in this analysis; in particular it has been assumed that the fibres are discontinuous and the effect on the stress-strain behaviour by the micro-compressive defects (kink bands) themselves has been disregarded. This is clearly an over simplification, but the analysis does serve to show that the model is physically realistic.

Expanding on the same shear lag approach, it is possible to predict the onset of inelastic behaviour. Equation 2.5 on page 54 is an expression that provides predictions of the composite strain ($\varepsilon_{i.}$) at the onset of inelastic behaviour (interfacial sliding), which will occur when some critical value of interfacial shear stress ($\tau_{i.}$) is reached. Assuming that the aspect ratio is 5 and the yield onset occurs at a composite strain of 0.06% (Table 6.4 on page 284), slightly lower than the yield point itself, it can be seen that this corresponds

to a critical value of interfacial shear stress of around 9.7 MPa. Not many critical interfacial shear stress values have been reported in the literature for plant fibre reinforced PMC systems and those that have vary substantially. Hill and Shawkataly, (2000), for example, reported values ranging from 1.48 to 1.98 MPa for the interfacial shear strength of unmodified and modified (acetic anhydride) coir fibre reinforced polyester composites. Using a micro-droplet technique combined with Raman spectroscopy Eichhorn and Young, (2004) found that the maximum interfacial shear stress in single hemp fibre reinforced epoxy composite was 'of the order of the shear yield stress of the resin (40 to 45 GPa). Sanadi *et al.*, (1986) calculated the interfacial frictional shear strength by the Kelly-Cottrell equation for unidirectional sunhemp fibre reinforced polyester composites and found it to be 4.34 MPa. The value of 9.7 MPa obtained by this work is therefore consistent with what might be physically reasonable for such a composite system.

As the critical value of interfacial shear stress increases, the strain at the onset of inelastic behaviour increases proportionally. As noted in Table 6.4 on page 284, there is a difference between the yield onset points between both modified types of flax and unmodified flax reinforced composites. However, as discussed previously there appears to be a threshold value of the critical interfacial shear stress, as neither the onset of yielding nor the yield point itself differ significantly between the MeA and PrA modified flax fibre reinforced composites. Since a difference would be expected from the form of bonding induced between fibre and matrix, this may indicate that other microstructural failure processes, such as matrix yielding or fracture become important as interfacial adhesion is improved.

Beyond the yield point, the initial decrease in the modulus of the composite (region C in Figure 6.12 on page 282) is explained by the occurrence of microstructural damage as discussed above. As the UnM flax reinforced composites are strained further, the modulus appears to increase (region D in Figure 6.12). This phenomenon probably arises because of the non-linear straining behaviour of the flax fibres themselves. Fibre to matrix debonding, matrix yielding, and matrix fracture occurring in the region C in

Figure 6.12, in the vicinity of the yield point may partially free the flax fibre from the encapsulating matrix, allowing them to 'realign' better to the direction of the applied load. This realignment most likely takes the form of an 'extension of the micro-compressive defects' (Baley, 2002). After extension of the micro-compressive defects the modulus of the flax fibres themselves may increase, and this would be manifested in the composite as an increase in modulus. It is likely that during fabrication initially damaged flax fibre would either undergo further damage (formation of new micro-compressive defects) or that existing damage would be compounded. The unsaturated polyester resin (Wresipol 31466) used throughout this investigation is also likely to undergo significant volumetric shrinkage during curing (Matthews and Rawlings, 1993), which might be expected to place the flax fibres encapsulated in it under compression, resulting in the creation of more micro-compressive defects. Cure shrinkage and its effects upon the reinforcing fibre might be expected to exacerbate the effects of the non-linear behaviour observed in these natural fibre reinforced composites.

It is the failure of the composites reinforced with both types of modified flax fibre that shows the influence fibre modification has had on the composite systems as a whole. In the case of modified flax fibre reinforced composites, a significant reduction in the tensile strength is observed. A likely reason for this is that once a microscopic crack with the composite begins to propagate, it does so catastrophically due to suppression of crack stopping mechanisms such as fibre to matrix debonding. It is, perhaps, significant that the failure strain of the MeA modified fibre composites, at an average of 0.79%, is lower than might be expected from the fibre and the same order as that of the resin and smaller than that of the PrA modified flax reinforced composites. It is also interesting to note, if the onset of matrix cracking does initiate catastrophic failure of the MeA composites at a strain close to 0.79%; at approximately the same strain higher energy, AE events were detected in UnM flax reinforced composites, these are likely to be matrix cracking and the onset of fibre fracture (Figure 6.14 on page 286). As the failure strains are different between the two types of modified flax composites, it may indicate that the microstructural failure mechanisms differ between them. Modification may well affect the strength and failure strain of the fibre, this has not been verified in this work, and

therefore it is inappropriate to draw any conclusions regarding the ultimate failure of the composites.

6.4 Summary

High quality linen grade flax fibre reinforced unsaturated polyester composites can compete favourably with their glass fibre equivalents in terms of specific stiffness, they display reasonable toughness and flexural properties when compared to other natural fibre reinforced composite systems. During straining, deformation occurs in a non-linear fashion. Strong evidence has been provided *via* loading and unloading behaviour and acoustic emissions analysis that these composites undergo yielding at comparatively low values of stress and strain. The likely source of the yielding behaviour observed with these composite systems is the non-linear behaviour of the flax reinforcing fibres caused by the existence of micro-compressive defects along their lengths and the effect the defects have upon the stress-transfer.

Necessary assumptions have been made in the theoretical analysis applied to this deformation behaviour; however, the deformation behaviour observed is real and will have practical implications for the use of these materials in structural applications.

7 CONCLUSIONS AND RECOMMENDATIONS FOR FURTHER WORK

7.1 Introduction

The aim of this final chapter is to review the research presented in the thesis and to summarise the main conclusions for each investigation. Not all of the issues that arose during this research programme were investigated. However, this chapter makes suggestions for further research that could be performed as a result of the knowledge gained in this work.

7.2 Summary of the main conclusions

The preliminary investigation reported in Chapter 3 was undertaken to gain an understanding of the physical and mechanical properties of plain weaved flax reinforced unsaturated polyester composites. It was undertaken to compare their macromechanical properties to those exhibited by glass woven roving reinforced composites fabricated with the same matrix system. A further purpose of the investigation was to become familiar with the fabrication methods available for such composites. One of the main findings from this aspect of the investigation proved to be of great importance, as it helped develop the methodology used to fabricate other woven flax reinforced composites in the laboratory that were necessary for future investigations (Chapters 4 and 5).

A lack of experience regarding methods for the impregnation and moulding of woven flax reinforced composites caused unexpected variations in the calculated fibre volume fractions of the manufactured composites. The observed mechanical properties of woven flax reinforced polyester composites had poorer relationships with fibre volume fraction

than the woven glass composites that were fabricated using an RTM technique. The relationship between composite properties and fibre volume fraction is thought to have been affected by the fact that those composites that contained more plies of reinforcement had lower fibre volume fractions than composites which contained less plies of reinforcement. However, it was shown that woven flax reinforced polyester composites with fibre volume fractions ranging from approximately 27 to 65% could be fabricated using simple techniques. By making necessary assumptions about fibre density, it was also predicted that the void content of woven glass and flax composites increased with fibre volume fraction. Woven flax composites that contained plies which had been pre-pressed prior to resin impregnation had the highest predicted void contents. Although flax fibres contain natural voids (lumens), it is possible that insufficient wetting of the flax fibres within yarns by the liquid resin is responsible for the voids present.

The issue of resin penetration into flax fibre yarns was not investigated within this thesis. Further studies are deemed necessary to establish the effects that yarn Tex, yarn twist, yarn to yarn contact and the size used on the yarns during weaving have upon the penetration of resin within yarns and the wetting of flax fibre by resin if woven flax composites are to compete against conventional composites in certain applications.

Tensile, flexural and Charpy impact properties of woven flax reinforced polyester composites were found to be considerably less than the values exhibited by woven glass roving reinforced polyester composites. Glass fibres are more compatible with the matrix system used (they are truly continuous, homogeneous, and they are not twisted) which will help resin penetration throughout the composite structure. Also, the crimping of warp yarns within glass fibre rovings is not as severe as it is with warp yarn crimping in the particular woven flax fabric used as reinforcement for the studies reported in Chapter 3. The specific stiffness of natural fibre reinforced composites is often comparable against the specific stiffness of equivalent glass fibre composites, but this was found not to be the case for the woven flax composites in this investigation (when tested in the warp direction).

The tensile and flexural strengths of woven flax reinforced polyester composites did compare well to published values of non-woven bast fibre reinforced polyester composites. The Charpy impact strength also was found to be greater than natural fibre non-woven reinforced composites. As lack of toughness has been reported as being one of the main limiting factors of natural fibre reinforced thermosetting matrix composites, it is necessary that further investigations are undertaken. Further development of woven flax composites may produce methods to increase their toughness considerably. In addition, combining non-woven and woven natural fibre reinforcement together in a composite system may increase their suitability for replacing conventional composites in some applications. A hybrid composite containing both non-woven and woven reinforcement may have an acceptable level of toughness because of the woven reinforcement. It also may have a higher modulus that cannot be achieved presently by a composite reinforced with 100% woven flax fabric because of the addition of a non-woven layer.

Investigations reported in Chapter 4 and 5 were undertaken to examine the effect that reinforcement architecture has upon the mechanical properties of woven flax fabric reinforced epoxy composites.

The research conducted for Chapter 4 investigated the influence that weft yarn Tex and the stacking of plies of woven flax fabric have upon the warp and weft properties of composites. It was observed that weft yarn Tex did influence the warp orientated flexural and tensile properties of composites. This was found for composites containing plies of woven flax reinforcement stacked in the same direction, or with a central ply at 90° to the top and bottom plies. A composite containing woven flax reinforcement that had been woven with lower Tex weft yarns than the warp yarns exhibited better warp orientated properties than a composite consisting of reinforcement that had higher Tex weft yarns or the same Tex weft yarns as the warp yarns. As weft yarn Tex reduces in woven reinforcement, the weft flexural and tensile properties of the composites it reinforces become lower. It was also found that the warp and weft flexural strengths of composites containing reinforcement with a middle ply at 90° to the top and bottom plies were

comparable to the flexural strengths of composites reinforced with the same woven flax fabric, but stacked with all three plies in the same direction. This trend was not observed between the two types of reinforced composite's flexural moduli. A significant difference was found between the warp flexural moduli of composites containing reinforcement stacked with a central ply at 90° to the top and bottom plies and equivalent composites containing the same reinforcement but stacked in the same direction. Composites reinforced with a middle ply at 90° were stiffer in the warp direction than composites reinforced with all three plies stacked in the same direction. When comparing the weft flexural modulus of composites reinforced with the same woven flax fabric but stacked with the two geometries, no significant differences were observed.

The Charpy impact strength of the composites in the warp direction increased as the woven flax reinforcement contained lower Tex weft yarns than warp yarns. The weft Charpy impact strength decreased as the weft yarn Tex of reinforcement became lower than the Tex of warp yarns. These two above mentioned trends occurred for both types of reinforced composite, composites containing plies stacked in the same direction or composites reinforced with a centre ply at 90° to the top and bottom plies.

The Tex of the weft yarns within woven flax fabrics used as reinforcement in epoxy composites did influence the warp and weft mechanical properties of the material. The stacking of woven flax fabric plies in a simple sequence to act as reinforcement also influenced the warp and weft properties of the composites. By reducing weft yarn Tex it allows the warp yarns to become straighter within the woven flax fabric because they do not have be crimped to such an extent in order to be able to pass over and under the weft yarns. As observed from tensile tests performed on the woven flax fabric, the warp extension to failure became lower as weft yarn Tex reduced. It is possible that the improvement in properties as weft yarn Tex reduced is because the warp yarns are acting as a better reinforcement. This is because load transfer from the matrix occurs immediately a stress is applied, rather than the yarns having to re-orientate before load transfer can occur.

Chapter 5 describes the results gained from the experimental work aimed at studying the influence that the weave type of the woven flax reinforcement has upon the mechanical properties of the composite it reinforces. The majority of the woven flax fabrics used consisted of the same Tex warp and weft yarns.

Results gained from testing the tensile properties of the woven flax fabrics in both the warp and weft directions enabled the following trends to be identified.

- Woven flax fabric weft tensile extension was always lower than the fabric's warp tensile extension.
- The more frequently crimped the warp yarns within woven flax fabrics were, the higher was the warp tensile extension.
- As the frequency of warp yarn crimping reduced, the tensile extension also reduced.
- The weft tensile extension of the woven flax fabric and maximum load at failure was unaffected by the frequency of warp yarn crimping, provided the warp and weft yarns were the same Tex.
- Woven flax fabrics consisting warp and weft yarns of the same Tex usually exhibited a significant difference between the warp and weft maximum loads at failure, with the weft direction showing the higher value.
- The warp and weft maximum load at failure was similar when the warp yarns were only crimped every 10^{th} weft yarn.

The epoxy composites fabricated from the 12 different woven flax fabrics had similar fibre volume fractions and thus the observed properties could be compared equally without the need to normalise the results to an arbitrary fibre volume fraction. High variations existed between the densities of the composites. These were thought to have been caused by the composites having different void contents, dimensions of resin rich regions and the number of reinforcing yarns per unit volume.

The flexural and Charpy impact strengths of the epoxy composites were influenced by the weave type of the woven flax fabric in the following ways:

- Composites that contained woven flax fabric that consisted of frequently crimped warp yarns, exhibited significant differences between warp and weft flexural strength. The weft flexural strength was greater in these types of reinforced composite.
- As composites contain reinforcement (same Tex warp and weft yarns) that is weaved with less frequent warp yarn crimping, the differences between warp and weft flexural strength reduce. The warp and weft flexural strength of some composites can be similar and it is possible that the warp flexural strength can be slightly higher.
- Composites that contained woven flax fabric that consisted of frequently crimped warp yarns exhibited significant differences between warp and weft flexural moduli, the weft flexural moduli was always greater in these types of reinforced composite.
- Differences between the warp and weft flexural moduli reduced as warp yarn crimping became less frequent, but only one composite that was reinforced with the least crimped warp yarn exhibited a warp composite stiffness similar to the corresponding weft stiffness.
- No correlations existed between the weave type of the woven flax fabric reinforcement and the warp or weft Charpy impact strengths of composites.
- The majority of composites showed a significant difference between the warp and weft Charpy impact strengths.
- The weft Charpy impact strength was always greater than the warp Charpy impact strength of the same composite.

A limitation of the investigation was the size of the composites fabricated. This was obviously realized at the time of planning the investigation but because of the limited supply of woven flax fabric available, very little could be done to correct the situation. If the composites could have been fabricated to a larger size, extra tensile specimens could

have been tested. As a result, only two tensile specimens (both in the warp direction) were tested from the composites fabricated for studies described in Chapter 5. The same issue also arose for the composites described in Chapter 4; only two tensile specimens from both the warp and weft direction were tested. The small number of tensile specimens may not have portrayed the real tensile properties of the composites. However, the warp tensile specimens described in Chapter 5 did show that as the warp yarn crimping became less frequent, the composite's tensile Young's modulus gradually increased. This observed trend does have some similarities with the composites warp flexural modulus.

In conclusion, the composite's warp flexural properties are influenced by the frequency the warp yarns within the woven flax fabric reinforcement are crimped. Reducing the frequency of warp yarn crimp has the effect of reducing the anisotropic behaviour of the warp and weft flexural properties of epoxy composites they may reinforce. However, there is an unknown limit to how much the frequency of warp yarn crimping can be reduced before the woven flax fabric losses its integrity and the benefits of having a bi-directional reinforcement ply for composites is lost. As there are many other weave types available that flax fibre could be woven to form, it is essential that these are investigated. Altering the Tex of the yarns, the twist of the yarns, fabric count of the fabric and the weave type used as reinforcement should be investigated further to try and find the optimal architecture for the reinforcement of thermosetting polymer matrix composites.

All stress-strain curves obtained from investigations described in Chapters 3, 4 and 5, have shown that non-linear behaviour is present and is often initiated at low values of stress and strain for unsaturated polyester or epoxy composites reinforced with woven flax fabric. This phenomenon was not fully investigated within this thesis for woven flax fabric reinforced composites but was investigated with a more fundamental form of composite (*i.e.*, unidirectional). The work presented and discussed in Chapter 6 was focused on the nature of the deformation behaviour of unidirectional unsaturated polyester composites reinforced with flax fibre. The investigation aimed to not only record the mechanical properties of such composites but also to gain an understanding of

the micromechanical processes that cause the onset of non-linear behaviour at low values of stress and strain.

The following conclusions can be made regarding the physical, mechanical and deformation properties of high quality grade flax sliver fibre reinforced unsaturated polyester composites:

- As fibre content of composites increased up to a 60% fibre volume fraction, the density of flax fibre reinforced unidirectional composites also increased.
- As fibre content of the composites increased up to approximately 60% fibre volume fraction, the composites flexural strength and modulus also increased in a linear manner.
- At nearly 60% fibre volume fraction, the flexural strength and modulus was 200 MPa and 20 GPa respectively.
- The flexural specific stiffness of the flax fibre unidirectional composites was comparable to the specific stiffness exhibited by glass fibre chopped strand mat reinforced composites.
- As fibre content of composites increased up to approximately 60% fibre volume fraction, the composite's Charpy impact strength increased in a linear manner.
- Flax fibre reinforced unidirectional unsaturated polyester composites had Charpy impact strengths of up to 70 kJ m^{-2}.
- As fibre content of composites increased up to approximately 60% fibre volume fraction, the composite's tensile strength and Young's modulus increased in a linear manner.
- At nearly 60% fibre volume fraction the composite's tensile strength and Young's modulus was 303 MPa and 30 GPa respectively.
- Flax fibre reinforced unidirectional unsaturated polyester composites can compete favourably with their glass fibre equivalents in terms of tensile specific stiffness.
- Flax fibre reinforced unidirectional unsaturated polyester composites exhibited flexural properties and Charpy impact strengths that allowed them to compare favourably with other natural fibre reinforced thermosetting composite systems.

- A theoretical flax fibre Young's modulus of 45 GPa was found by extrapolation of tensile Young's modulus data.
- During tensile straining of flax fibre reinforced unidirectional unsaturated polyester composites deformation occurred in a non-linear fashion.
- It is extremely likely that flax fibre reinforced unidirectional unsaturated polyester composites undergo yielding at comparatively low values of stress and strain.
- It is likely that the source of the yielding behaviour observed with these composite systems is the non-linear behaviour of the flax reinforcing fibres caused by the existence of micro-compressive defects along their lengths and the effect the defects have upon the stress-transfer.

In conclusion, the mechanical properties of the flax fibre unidirectional unsaturated polyester composites are comparable with glass fibre equivalents and other natural fibre reinforced thermosetting composite systems. However they exhibit yielding behaviour when exposed to low values of stress and strain which is unlike conventional composites. This behaviour would have implications for the use of natural fibre reinforced composites in structural applications. Further research, investigating the nature of the fibre to matrix interface is necessary. This could be investigated using single fibre composites to improve understanding of the fibre to matrix interface and the effects of micro-compressive defects. These negative aspects may be reduced through chemical modifications of the fibre to matrix interface to allow the flax fibres to act as a continuous reinforcement fibre rather than a fragmented fibre.

Natural fibre reinforced thermosetting composites still appear to be of particular interest to the research community and industrial companies wishing to produce materials that have a lower impact on the environment. However, as some of the above work has shown, not all of the mechanical properties of natural fibre reinforced composites are at present able to fully compete with the synthetic reinforced composites that are currently used. It is believed that with time and further development, natural fibre reinforced thermosetting polymer matrix composites will find a niche in the composite market and will grow in popularity.

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APPENDIX 1

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