BIOFIBRES AND BIOCOMPOSITES

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Abstract

This review deals with a study of the literature on the various aspects of biofibres and biocomposites. Biocomposites are finding applications in many fields ranging from construction industry to automotive industry. The pros and cons of using biofibres are enumerated in this review. The classification of biocomposites into green composites, hybrid biocomposites and textile biocomposites are discussed. New developments dealing with cellulose based nanocomposites and electrospinning of nanofibres have also been presented. Recent studies pertaining to the above topics have also been cited. Finally, the applications of biofibre reinforced composites have been highlighted.

Keywords : biocomposite, biofiber, green composite, textile

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1. BIOFIBRES/ LIGNOCELLULOSIC FIBRES/NATURAL FIBRES

Natural fibres are subdivided based on their origins, coming from plants, animals or minerals. All plant fibres are composed of cellulose while animal fibres consist of proteins (hair, silk and wool). Plant fibres include bast (or stem or soft sclerenchyma) fibres, leaf or hard fibres, seed, fruit, wood, cereal straw and other grass fibres. Over the last few years, a number of researchers have been involved in investigating the exploitation of natural fibres as load bearing constituents in composite materials. The use of such materials in composites has increased due to their relative cheapness, their ability to recycle and for the fact that they can compete well in terms of strength per weight of material. Natural fibres can be considered as naturally occurring composites consisting mainly of cellulose fibrils embedded in lignin matrix. The cellulose fibrils are aligned along the length of the fibre, which render maximum tensile and flexural strengths, in addition to providing rigidity. The reinforcing efficiency of natural fibre is related to the nature of cellulose and its crystallinity. The main components of natural fibres are cellulose (α-cellulose), hemicellulose, lignin, pectins and waxes.

Cellulose is a natural polymer consisting of D-anhydroglucose ($C_6H_{11}O_5$) repeating units joined by β-1,4-glycosidic linkages at C_1 & C_4 position[1] . The degree of polymerization (DP) is around 10,000. Each repeating unit contains three hydroxyl groups. These hydroxyl groups and their ability to hydrogen bond play a major role in directing the crystalline packing and also govern the physical properties of cellulose. Solid cellulose forms a microcrystalline structure with regions of high order i.e. crystalline regions and regions of low order i.e. amorphous regions. Cellulose is also formed of slender rod like crystalline microfibrils. The crystal nature (monoclinic sphenodic) of naturally occurring cellulose is known as cellulose I. Cellulose is resistant to strong alkali (17.5 wt%) but is easily hydrolyzed by acid to water-soluble sugars. Cellulose is relatively resistant to oxidizing agents.

Hemicellulose is not a form of cellulose and the name is a misnomer. They comprise a group of polysaccharides composed of a combination of 5- and 6- carbon ring sugars. Hemicellulose differs from cellulose in three aspects. Firstly, they contain several different sugar units whereas cellulose contains only 1,4--β-D-glucopyranose units. Secondly, they exhibit a considerable degree of chain branching containing pendant side groups giving rise to its non crystalline nature, whereas cellulose is a linear polymer. Thirdly, the degree of polymerization of native cellulose is 10-100 times higher than that of hemicellulose. DP of hemicellulose is around 50 to 300. Hemicelluloses form the supportive matrix for cellulose microfibrils. Hemicellulose is very hydrophilic, soluble in alkali and easily hydrolyzed in acids.

Lignin is a complex hydrocarbon polymer with both aliphatic and aromatic constituents. They are totally insoluble in most solvents and cannot be broken down to monomeric units. Lignin is totally amorphous and hydrophobic in nature. It is the compound that gives rigidity to the plants. It is thought to be a complex, three-dimensional copolymer of aliphatic and aromatic constituents with very high molecular weight. Hydroxyl,methoxyl and carbonyl groups have been identified. Lignin has been found to contain five hydroxyl and five methoxyl groups per building unit. It is believed that the structural units of lignin molecule are derivatives of 4-hydroxy-3-methoxy phenylpropane[3]. The main difficulty in lignin chemistry is that no method has been established by which it is possible to isolate lignin in its native state from the fibre. Lignin is considered to be a thermoplastic polymer exhibiting a glass transition temperature of around 90°C and melting temperature of around 170°C[2]. It is not hydrolyzed by acids, but soluble in hot alkali, readily oxidized and easily condensable with phenol[3].

Pectins are a collective name for heteropolysaccarides. They give plants flexibility. Waxes make up the last part of fibres and they consist of different types of alcohols.

Biofibres can be considered to be composites of hollow cellulose fibrils held together by a lignin and hemicellulose matrix[4] . The cell wall in a fibre is not a homogenous membrane (Figure 1). Each fibril has a complex, layered structure consisting of a thin primary wall that is the first layer deposited during cell growth encircling a secondary wall. The secondary wall is made up of three layers and the thick middle layer determines the mechanical properties of the fibre. The middle layer consists of a series of helically wound cellular microfibrils formed from long chain cellulose molecules: the angle between the fibre axis and the microfibrils is called the microfibrillar angle. The characteristic value for this parameter varies from one fibre to another.

Such microfibrils have typically a diameter of about 10-30 nm and are made up of 30 to 100 cellulose molecules in extended chain conformation and provide mechanical strength to the fibre. The amorphous matrix phase in a cell wall is very complex and consists of hemicellulose, lignin and in some cases pectin. The hemicellulose molecules are hydrogen bonded to cellulose and act as cementing matrix between the cellulose microfibrils, forming the cellulose-hemicellulose network, which is thought to be the main structural component of the fibre cell. The hydrophobic lignin network affects the properties of other network in a way that it acts as a coupling agent and increases the stiffness of the cellulose/hemicellulose composite.

The structure, microfibrillar angle, cell dimensions,defects and the chemical composition of fibres are the most important variables that determine the overall properties of the fibres[5]. Generally, tensile strength and Young's modulus of fibres increases with increasing cellulose content. The microfibrillar angle determines the stiffness of the fibres. Plant fibres are more ductile if the microfibrils have a spiral orientation to the fibre axis. If the microfibrils are oriented parallel to the fibre axis, the fibres will be rigid, inflexible and have high tensile strength. Some of the important biofibres are listed in Table 1

1.1 Biofibres: Advantages & Disadvantages

The growing interest in lignocellulosic fibres is mainly due to their economical production with few requirements for equipment and low specific weight, which results in a higher specific strength and stiffness when compared to glass reinforced composites. They also present safer handling and working conditions compared to synthetic reinforcements. Biofibres are nonabrasive to mixing and molding equipment, which can contribute to significant cost reductions. The most interesting aspect about natural fibres is their positive environmental impact. Biofibres are a renewable resource with production requiring little energy. They are carbon dioxide neutral i.e. they do not return excess carbon dioxide into the atmosphere when they are composted or combusted. The processing atmosphere is friendly with better working conditions and therefore there will be reduced dermal and respiratory irritation. Biofibres possess high electrical resistance. Thermal recycling is also possible. The hollow cellular structure provides good acoustic insulating properties. The worldwide availability is an additional factor.

The inherent polar and hydrophilic nature of lignocellulosic fibres and the non-polar characteristics of most thermoplastics results in compounding difficulties leading to non-uniform dispersion of fibres within the matrix which impairs the efficiency of the composite. This is a major disadvantage of natural fibre reinforced composites. Another problem is that the processing temperature of composites is restricted to 200°C as vegetable fibres undergo degradation at higher temperatures; this restricts the choice of matrix material. Another setback is the high moisture absorption of natural fibres leading to swelling and presence of voids at the interface, which results in poor mechanical properties and reduces dimensional stability of composites. Another restriction to the successful exploitation of biofibres for durable composite application is low microbial resistance and susceptibility to rotting. These properties pose serious problems during shipping, storage and composite processing. The non-uniformity and variation of dimensions and of their mechanical properties (even between individual plants in the same cultivation) poses another serious problem.

It is quite clear that the advantages outweigh the disadvantages and most of the shortcomings have remedial measures in the form of chemical treatments. The lignocellulosic fibres have an advantage over synthetic ones since they buckle rather than break during processing and fabrication. In addition, cellulose possesses a flattened oval cross section that enhances stress transfer by presenting an effectively higher aspect ratio.

Researchers[6] at the Institute of Natural Fibres, Poland have developed fire resistant upholstery using fire-retarded flax nonwoven (LinFR) FT. The non-woven used in the composites plays the role of fire barrier that reduces the vulnerability of filling material to the development and spread of fire. The softness of the upholstery system is also enhanced. In another interesting study, researchers[7] investigated the influence of natural and synthetic fibres which covered the forearm muscles on the activity of motor units in these muscles. The electrophysiological parameters of motor units were measured with electromyographical methods. The results indicated that temporary covering of examined muscles in the forearm with the synthetic clothing changed the pattern of motor units activity of muscles. Clothing made of natural fibre did not evoke this kind of effect.

Recently, natural cellulose fibres having properties between cotton and linen and suitable for various industrial applications were extracted from cornhusks[8]. High quality cellulosic fibres from cornhusks mean food, clothing and other major industrial products from the same source without the need for any additional natural resources. Using cornhusks for fibrous applications would save the land and other natural resources required to grow fibre crops and will conserve the non-renewable petroleum sources required to produce synthetic fibres. More than 9 million tons of natural cellulose fibres with a potential sale value of $19 billion and with a value addition of at least $12 billion could be produced from the cornhusks available every year. Utilizing cornhusks as a source for natural cellulose fibres will significantly benefit the agriculture, fibre, food and energy needs of the future and will also benefit the environment. The structure and composition of the natural cellulose fibres obtained from cornstalks were found to be different from that of the common bast fibres such as flax and kenaf. Cornstalk fibres were found to have relatively lower percentage of crystallinity but similar microfibrillar angle as that of the common bast fibres[9].

2. BIOCOMPOSITES

The history of mankind has witnessed several surges in the field of research and development. The rampant use of petroleum products has created a twin dilemma; depletion of petroleum resources and entrapment of plastics in the food chain and environment. The increasing pollution caused by the use of plastics and emissions during incineration is affecting the food we eat, water we drink, air we breathe and threatening the greatest right of human beings, the right to live. The exhaustive use of petroleum based resources has initiated the efforts to develop biodegradable plastics. This is based on renewable biobased plant and agricultural products that can compete in the markets currently dominated by petroleum based products. The production of 100 % biobased materials as substitute for petroleum based products is not an economical solution. A more viable solution would be to combine petroleum and biobased resources to develop a cost-effective product having immense applications. Biopolymers or synthetic polymers reinforced with natural or bio fibres (termed as biocomposites) are a viable alternative to glass fibre composites. Scientists are looking at the various possibilities of combining biofibres such as sisal, flax, hemp, jute, banana, wood and various grasses with polymer matrices from non-renewable and renewable resources to form composite materials to make the biocomposite revolution a reality[10].

Broadly defined, biocomposites are composite materials made from natural/bio fibre and petroleum derived non-biodegradable polymers (PP,PE) or biodegradable polymers (PLA, PHA) . The latter category i.e. biocomposites derived from plant derived fibre (natural / biofibre) and crop / bioderived plastic (biopolymer/bioplastic) are likely to be more eco-friendly and such composites are termed as green composites.

The best known renewable resources capable of making biodegradable plastics are starch and cellulose[11]. Starch is one of the least expensive biodegradable materials available in the world market today. It is a versatile polymer with immense potential for use in non-food industries. Cellulose from trees and cotton plants is a substitute for petroleum feedstocks to make cellulose plastics. Another aspect that has gained global attention is the development of biodegradable plastics from vegetable oils like soybean oil, peanut oil, walnut oil, sesame oil and sunflower oil. Green composites from soy protein based bioplastics and natural fibres show potential for rigid packing and housing and transportation applications[12]. Fish oil based polymers have also attracted the attention of researchers due to their high degree of unsaturation. Fish oil based polymers also possess unique good damping and shape memory properties[13].

The history of fibre reinforced plastics began in 1908 with cellulose fibre in phenolics, later extending to urea and melamine and reaching commodity status with glass fibre reinforced plastics. The fibre-reinforced composites market is now a multibillion-dollar business[14] (Figure 2). Though hailed as a miraculous discovery long back, plastic products now enjoy an ambiguous reputation. Scientists at the BioComposites Centre at the University of Wales, Bangor are developing high-quality packaging of goods, using starch from corn and potatoes to tackle the problem of high cost waste disposal[15]. Researchers[16] are also exploring the aspects of producing plastics from limonene which is extracted from citrus. After decades of obscurity, biofibre reinforced composites are being touted as the material of the millennium.

2.1 Classification of Biocomposites

2.1.1 Green Composites

Research efforts are currently being harnessed in developing a new class of fully biodegradable "green' composites by combining (natural / bio) fibres with biodegradable resins[17]. The major attractions about green composites are that they are environmentally-friendly, fully degradable and sustainable, i.e. they are truly 'green' in every way. At the end of their life they can be easily disposed of or composted without harming the environment.

Fig. 3 shows a classification of biodegradable polymers in four families[18]. Except the fourth family, which is of fossil origin, most polymers (family 1–3) are obtained from renewable resources (biomass). The first family is agro-polymers (e.g. polysaccharides) obtained from biomass by fractionation. The second and third families are polyesters, obtained, respectively by fermentation from biomass or from genetically modified plants (e.g. polyhydroxyalkanoate: PHA) and by synthesis from monomers obtained from biomass (e.g. polylactic acid: PLA). The fourth family are polyesters, totally synthesised by the petrochemical process (e.g. polycaprolactone: PCL, polyesteramide: PEA, aliphatic or aromatic copolyesters). A large number of these biodegradable polymers (biopolymers) are commercially available. They show a large range of properties and they can compete with non-biodegradable polymers in different industrial fields (e.g. packaging).

Another important biocomposites category is based on agro-polymers matrixes, mainly focussed on starchy materials. Plasticized starch, the so-called 'thermoplastic starch' (TPS) is obtained after disruption and plasticization of native starch, with water and plasticizer (e.g. polyol) by applying thermomechanical energy in a continuous extrusion process. Unfortunately, TPS shows some drawbacks such as a strong hydrophilic character (water sensitive), rather poor mechanical properties compared to conventional polymers and an important post-processing variation of the properties. TPS properties reach equilibrium only after several weeks. To improve these material weaknesses, TPS is usually associated with other compounds.

The design and life cycle assessment of green composites have been exclusively dealt with by Baillie.[19] Green composites have been used effectively in many applications such as mass produced consumer products with short life cycles or products intended for one time or short time use before disposal. Green composites may also be used for indoor applications with a useful life of several years. The reinforcement of biofibres in green composites has been highlighted by Bismarck et al.[5] A number of natural and biodegradable matrices that are available for use in such green composites[20] are listed in Table 2. The classification of green composites based upon the different types of biodegradable matrices is given below.

2.1.1.1 BIOFIBER-PLASTIC COMPOSITES

Poly(a-hydroxy acid) such as poly(glycolic acid), PGA, or poly(lactic acid), PLA, are crystalline polymers with relatively high melting point. Recently PLA has been highlighted because of its availability from renewable resources like corn. PLA is a hydrophobic polymer because of the incorporation of the1CH3 side groups when compared to PGA. Poly(b-hydroxyalkanoate)s (PHAs), which are synthesized biochemically by microbial fermentation and which may be produced in the future by transgenic plants, represent natural polyesters. Bacteria came first and are still the only real source of these polyesters; it will still require some more years' research until transgenic plants will be available for production. Poly(b-hydroxybutyrate) (PHB) (commercial name Biopol) is a biotechnologically produced polyester that constitutes a carbon reserve in a wide variety of bacteria and has attracted much attention as a biodegradable thermoplastic polyester. It can be degraded to water and carbon dioxide under environmental conditions by a variety of bacteria and has much potential for applications of environmentally degradable plastics[21].

In an interesting research, Plackett et al.[22] used commercial L-polylactide which was first converted to film and then used in combination with jute fibre mats to generate biodegradable composites by a film stacking technique. Degradation of the polylactide during the process was investigated using size exclusion chromatography. The tensile properties of composites produced at temperatures in the 180–220 °C range were significantly higher than those of polylactide alone. Examination of composite fracture surfaces using electron microscopy showed voids occurring between the jute fibre bundles and the polylactide matrix in some cases. Size exclusion chromatography revealed that only minor changes in the molecular weight distribution of the polylactide occurred during the process.

In an innovative study, novel biodegradable films were fabricated from chitosan and poly(lactic acid) by Sébastien et al[23]. It was found that composite films offered a great advantage in preventing the surface growth of mycotoxinogen strains because of their antifungal activity. However, the physico-chemical properties of such heterogeneous films, dramatically limit their further usage as packaging materials.

Lee and Wang[24] investigated the effects of lysine-based diisocyanate (LDI) as a coupling agent on the properties of biocomposites from poly (lactic acid) (PLA), poly (butylene succinate) (PBS) and bamboo fiber (BF). They observed that the tensile properties, water resistance, and interfacial adhesion of both PLA/BF and PBS/BF composites were improved by the addition of LDI, but thermal flow[25] was hindered due to cross-linking between polymer matrix and BF. Enzymatic biodegradability of PLA/BF and PBS/BF composites was investigated by Proteinase K and Lipase PS, respectively. It was seen that the composites could be quickly decomposed by enzyme and the addition of LDI delayed the degradation. Figure 4 shows the SEM micrographs of PLA or PBS/BF (70/30) composites with LDI degraded for different degradation time. It can be clearly seen that the matrix of both composites became reduced as enzymatic degradation proceeded.

Green composites composed of regenerated cellulose (lyocell) fabric and biodegradable polyesters [poly(3-hydroxybutyrate-*co*-3-hydroxyvarelate) (PHBV), poly(butylene succinate) (PBS), and poly(lactic acid) (PLA)] were prepared by Shibata et al[26] and properties studied. The tensile moduli and strength of all the biodegradable polyester/ lyocell composites were found to increase with increasing fiber content. The biodegradation test revealed that the lyocell fabric in PLA/lyocell composite degraded, as shown in Figure 5 . Pure PLA film was found to be almost unchanged after 120 days.

The thermal stability of biodegradable composite films prepared from blends of poly(vinyl alcohol), cornstarch, and lignocellulosic fibre was investigated by Imam et al.[27]. Thermogravimetric analysis (TGA) indicated the suitability of formulations for melt processing, and for application as mulch films in fields at much higher temperatures. The study also revealed that both starch and lignocellulosic fibre degraded much more rapidly than PVA. The addition of fibre to the formulations was found to enhance the PVA degradation. In another interesting study, the thermal behavior of green composites fabricated from bagasse fibre and polyvinyl alcohol was investigated by Fernandes et al[28]. They observed an increase of thermal stability upon incorporation of bagasse fibre.

In another study biocomposites[29] were fabricated using a non-woven fibre mat (90% hemp fibre with 10% thermoplastic polyester binder) as reinforcement, and unsaturated polyester (UPE) resin as well as blends of UPE and functionalized vegetable oils as the polymer matrix. All composites were made with 30% volume fraction of fibre, which was optimized earlier. The structure–property relationships of this system as well as the thermo-mechanical properties of these composites were measured. The notched Izod impact strength of biocomposites from biobased resin blends of UPE and functionalized vegetable oil and industrial hemp fibre mat were enhanced by 90% as compared to that of the pure UPE-industrial hemp fibre mat composites. The tests also showed an improvement in the tensile properties of the composite as a result of the incorporation of the derivatized vegetable oil.

Researchers have also developed green composites from jute fabric and Biopol composites[30]. Chemical modification of the fabric was carried out to improve interfacial properties. A significant amount of research has been done at the German Aerospace Centre (DLR) in Braunschweg on biodegradable plastics and composites[31]. Starch and modified resins have also been used as matrix to form green composites[32].

Biodegradable composites were developed from poly(propylene carbonate) (PPC) reinforced with granular cornstarch by Ma et al[33]. The authors also investigated the effects of succinic anhydride (SA) on the morphology, thermal properties, as well as mechanical properties of the composites. The authors are of the opinion that SA enhances the properties of PPC/starch composites, which was ascribed to the improvement of the interaction between PPC and granular.

In another study[34] involving poly(propylene carbonate) (PPC) ,starch–g-poly(methyl acrylate) (S–g-PMA) copolymer was used to reinforce PPC. Starch–g-poly(methyl acrylate) (S–g-PMA) copolymer was prepared by ceric ammonium nitrate-initiated polymerization of methyl acrylate onto corn starch (CS). Tensile tests showed that the S–g-PMA incorporation improved the stiffness and tensile strength of composites significantly. TG results indicated that the S–g-PMA addition led to an improvement in the thermal stability of resulting composites.

2.1.1.2 BIOFIBER-SOY BASED COMPOSITES

In US, soybeans provide over 60% of the fats and oils used for food and the majority of the feed protein. Soybeans typically contain about 20% oil and 40% protein. Protein levels as high as 55% have been observed in soybeans. Soybeans consist of discrete groups of proteins (polypeptides) that span a broad range of molecular sizes and are comprised of approximately 38% of non-polar, non-reactive amino acid residues, while 58% are polar and reactive. Modifications that take advantage of water solubility and reactivity are exploited in improving soy protein for use in plastics and other biomaterials[35]. Soy protein plastics of different compositions have been prepared by injection moulding[36]. Compression moulding is also used for soy plastic processing. Dried soy plastics display an extremely high modulus, 50% higher than that of currently used epoxy engineering plastics[37]. So with proper moisture-barrier, soy protein is a potential starting material for engineering plastics.

Researchers recently investigated the effect of stearic acid on tensile and thermal properties of ramie fibre reinforced soy protein isolate (SPI) resin green composites[38]. It was observed that part of the stearic acid crystallized in SPI resin and that the crystallizability was affected by the addition of glycerol as a plasticizer. The fabricated green composite was found to have enormous potential for certain indoor applications.

The interfacial adhesion in the composites was apparent from SEM photomicrographs of the fracture tensile surfaces of the ramie/SPI and ramie/ modified soy protein isolate (MSPI) composites tested in the axial direction is shown in Figure 6 a & b respectively. Some of the fibres at the fracture surface do not show any resin, where as a few other fibres clearly show resin sticking on the surface. The presence of resin on some fibre surfaces for the SPI and MSPI resins clearly indicates good interfacial interaction between the fibre and the resins.

In an interesting study, researchers[39] modified soy flour (SF) by cross-linking it with glutaraldehyde (GA). The cross-linked soy flour (CSF) polymer was characterized for its tensile and thermal properties. The effect of glycerol on the mechanical properties of the soy flour was also characterized and optimized. CSF polymer showed improved tensile properties and thermal stability, compared to unmodified SF resin, for use as a resin to fabricate composites. Unidirectional green composites using flax yarn and CSF resin were fabricated and characterized for their tensile and flexural properties. The composite specimens exhibited fracture stress and Young's modulus of 259.5 MPa and 3.71 GPa, respectively, and flexural strength of 174 MPa, in the longitudinal direction.

Recently green composites were fabricated using pineapple leaf fibre and soy based plastic[40]. The addition of compatabilizer (polyester amide grafted glycidyl methacrylate (PEA-g-GMA) was seen to increase the mechanical properties of composites. In another interesting study involving biocomposites, the effect of alkali treatment on the thermal properties of Indian grass fibre reinforced soy protein green composites was studied by the same group[41].

The characterization of Phytagel® modified soy protein isolate (SPI) resin and unidirectional flax yarn reinforced green composites was undertaken by Lodha and Netravali[42]. The incorporation of Phytagel® in SPI resin led to an overall 10-fold increase in the tensile fracture stress and a nine-fold increase in the Young's moduli of the SPI resin along with a seven-fold decrease in fracture strain. The dynamic mechanical properties such as storage and loss modulus of the modified resin increased and the glass transition temperature also increased by about 56°C.

2.1.1.3 BIOFIBER-NATURAL RUBBER COMPOSITES

The primary effects of bio-fibre reinforcement on the mechanical properties of natural rubber composites include increased modulus, increased strength with good bonding at high fibre concentrations, decreased elongation at failure, greatly improved creep resistance over particulate-filled rubber, increased hardness and a substantial improvement in cut, tear and puncture resistance. Biodegradation of vulcanized rubber material is possible, although it is difficult due to the interlinkages of the poly (*cis*-1, 4-isoprene) chains, which result in reduced water absorption and gas permeability of the material.[43] Certain *Streptomyces* strains were seen to degrade rubber samples after 12 months of incubation.

The reinforcement of coir fibre in natural rubber has been extensively studied[44]. Researchers have also investigated the reinforcement effects of a leaf fibre - sisal fibre -in natural rubber[45]. Thomas and co-workers have also investigated the effect of chemical modification of banana fibre in natural rubber. It was found that modification of banana fibre resulted in superior mechanical properties[46].

Attempts to incorporate oil palm fibre in rubber matrix have also been successful. The effect of fibre concentration on the mechanical properties of oil palm reinforced natural rubber composites was investigated by Ismail et al.[47] They observed the general trend of reduction in tensile and tear strength with increasing fibre concentration.

An interesting report on the reinforcement effect of grass fibre - bagasse - in natural rubber was presented by Nassar et al.[48] Aging experiments revealed tensile strength retention of 97 %. Scientists have also developed composites comprising of kenaf fibre and natural rubber[49]. An increase in rheometric and mechanical properties was observed. Pineapple[50] and jute fibre[51] have also found their way as a potential reinforcement in natural rubber.

In an interesting study, researchers have used a novel fibre –isora fibre-in natural rubber[52]. Isora fibres are present in the bark of the Helicteres isora plant and are separated by retting process. Isora fibre resembles jute in appearance but surpasses it in strength, durability and lustre. The effects of different chemical treatments, including mercerisation, acetylation, benzoylation and treatment with toluene diisocyanate and silane coupling agents, on isora fibre properties and mechanical properties were analyzed. Isora fibre was seen to have immense potential as reinforcement in natural rubber. The variation of tensile strength with chemical modification is given in Figure 7.

Researchers have also designed novel rubber biocomposites by using a combination of leaf and fruit fibre in natural rubber[53]. The incorporation of sisal and coir fibre in NR was seen to increase the dielectric constant of the composites. These hybrid biocomposites were found to have enormous applications as antistatic agents. In another interesting study, the preparation of composites comprising of waste paper in natural rubber along with boron carbide and paraffin wax, for radiation shielding applications, was investigated[54].

In an innovative study, a unique combination of sisal and oil palm fibres in natural rubber has been utilized to design hybrid biocomposites. It was seen that the incorporation of fibers resulted in increased modulus[55]. Chemical modification of both sisal and oil palm fibres was imperative for increased interfacial adhesion and resulted in enhanced properties[56]. The viscoelastic[57], water sorption[58], dielectric[59] and stress relaxation[60] characteristics were also studied.

A major hurdle to the commercialization of green composites is the high cost of biodegradable matrices. Most of the biodegradable resins cost significantly more than the commonly used resins. Inexpensive production of oils for resins through biotechnology would certainly be of help in expediting their commercialization. The manufacturing costs can also be reduced by faster, better and more efficient processing.

Efforts are on the anvil for the development of "advanced green composites" made out of high strength protein fibres (spider silk) and biodegradable matrices. Biotechnology is being used to increase the yield of specific triglycerides and oils in beans for producing resins. These resins will be inexpensive compared to those available today and if suitably modified, could be biodegradable. Research is also being conducted to develop new pathways to synthesize inexpensive biodegradable resins[61] with better mechanical properties and thermal stability using nanotechnology[62].

2.1.1.4. GREEN NANOCOMPOSITES

Biodegradable nanocomposites were successfully fabricated from the thermoplastic cornstarch (TPCS) and activated-montmorillonite (MMT) by melt-intercalation by Huang et al[63]. It was revealed that plasticized thermoplastic corn starch were intercalated into the layers of MMT successfully, and layers of MMT were fully exfoliated and so formed the exfoliated nanocomposites with MMT.

Wibowo et al[64]. successfully developed environmentally benign nanocomposites using cellulose acetate (CA) bioplastic, citrate based plasticizer and organically modified clay nanofillers. The mechanical, morphological, and thermal properties were analyzed and it was found that the intercalated reinforcements resulted in enhancements of the composite tensile strength, tensile modulus, coefficient of thermal expansion (CTE), and heat deflection temperature.

2.1.2 Hybrid Biocomposites

The incorporation of several different types of fibres into a single matrix has led to the development of hybrid biocomposites. The behavior of hybrid composites is a weighed sum of the individual components in which there is a more favorable balance between the inherent advantages and disadvantages. Also, using a hybrid composite that contains two or more types of fibre, the advantages of one type of fibre could complement with what are lacking in the other. As a consequence, a balance in cost and performance could be achieved through proper material design[65]. The properties of a hybrid composite mainly depend upon the fibre content, length of individual fibres, orientation, extent of intermingling of fibres, fibre to matrix bonding and arrangement of both the fibres. The strength of the hybrid composite is also dependent on the failure strain of individual fibres. Maximum hybrid results are obtained when the fibres are highly strain compatible[66].

The properties of the hybrid system consisting of two components can be predicted by the rule of mixtures.

$$P_H = P_1 V_1 + P_2 V_2 \qquad [1.1.1]$$

where P_H is the property to be investigated, P_1 the corresponding property of the first system and P_2 the corresponding property of the second system. V_1 and V_2 are the relative hybrid volume fractions of the first and second system and

$$V_1 + V_2 = 1 \qquad [1.1.2]$$

A positive or negative hybrid effect is defined as a positive or negative deviation of a certain mechanical property from the rule of hybrid mixture.

The term hybrid effect has been used to describe the phenomenon of an apparent synergistic improvement in the properties of a composite containing two or more types of fibre[67]. The selection of the components that make up the hybrid composite is determined by the purpose of hybridization, requirements imposed on the material or the construction being designed. The problem of selecting the type of compatible fibres and the level of their properties is of prime importance when designing and producing hybrid composites. The successful use of hybrid composites is determined by the chemical, mechanical and physical stability of the fibre / matrix system.

Hybrid biocomposites can be designed by the combination of a synthetic fibre and natural fibre (biofibre) in a matrix and a combination of two natural fibre / biofibre in a matrix. Hybridization with glass fibre provides a method to improve the mechanical properties of natural fibre composites and its effect in different modes of stress depends on the design and construction of the composites[68]. The effect of hybridization of glass fibre in thermoset biocomposites has been discussed in detail[69].

The tensile and impact behavior of oil palm fibre-glass fibre-reinforced epoxy resin was investigated by Bakar et al.[70]. The hybridization of oil palm fibres with glass fibres increased the tensile strength, Young's modulus, and elongation at break of the hybrid composites. A negative hybrid effect was observed for the tensile strength and Young's modulus while a positive hybrid effect was observed for the elongation at break of the hybrid composites. The impact strength of the hybrid composites increased with the addition of glass fibres.

Cellular biocomposite cores fabricated from industrial hemp or flax fibres with unsaturated polyester were hybridized with woven jute, chopped glass, and unidirectional carbon fabrics[71]. Material characterization showed improved stiffness, strength, and moisture-absorption stability, while flexural tests on laboratory-scale plates demonstrated enhanced structural behavior. These hybrid cellular biofibre-based composites were found to provide an economic and environmentally friendlier alternative to entry-level synthetic composites.

Hybridization also has a profound effect on the water absorption property of composites. An attempt to study the moisture uptake characteristics of hybrid systems was performed by Mishra et al.[72]. The composite systems chosen were sisal / glass and pineapple /glass fibre reinforced polyester composites. Composites were prepared by varying the concentration of glass fibre and by subjecting the bio-fibres to different chemical treatments. The authors observed that water uptake of hybrid composites were less than that of unhybridized composites.

A comparative study of the water absorption of the glass fibre (7 wt. %) / natural biofibre (13 wt. %) with that of non-hybrid composites is given in Table 3 where a lowering in water absorption is evident[73].

The novel development of a photofabrication process of biofibre composites, based on oil palm empty fruit bunch fibres was recently reported[74]. The process consisted of the following steps: (1) the preparation of nonwoven mat of biofibre, either alone or in combination with glass and nylon; (2) drying the mat; (3) preparation of photocurable resin matrix, consisting of vinyl ester and photoinitiator; (4) impregnation of the mat by photocurable resin; and (5) irradiation of the impregnated mat by UV radiation to effect the cure of the composite. Oil palm fibre, glass, and nylon fibres were mixed in different proportions. A "mixture experimental design" was used to generate experimental compositions of the reinforcing fibres and to model dependency of the response variables on the components through mathematical relationships.

Scientists[75] at the Affordable Composites from Renewable Resources (ACRES) program at the University of Delaware investigated the mechanical properties of glass/flax hybrid composites based on a novel modified soybean oil matrix material. Composites with different glass/flax ratios and different fibre arrangements were made using a modified soybean oil matrix material. The fibre arrangement was varied to make symmetric and unsymmetric composites. The latter were tested in different modes in flexural tests and drop weight impact tests. The mechanical properties of the composites were found to depend upon the glass/flax ratio and the arrangement of fibres in the composite. On proper selection of the arrangement of fibres in the composite, the glass fibres and flax fibres were found to act synergistically resulting in an improved flexural and impact performance.

Another innovative approach to hybrid composites is the incorporation of two natural fibres / biofibre in a matrix system. Recently, the mechanical performance of short randomly oriented banana and sisal hybrid fibre reinforced polyester composites[76] was investigated with reference to the relative volume fraction of the two fibres at a constant total fibre loading of 0.40 volume fraction (V_f), keeping banana as the skin material and sisal as the core material. A positive hybrid effect was observed in the flexural strength (Figure 8) and flexural modulus of the hybrid composites. The tensile strength of the composites showed a positive hybrid effect when the relative volume fraction of the two fibres was varied, and maximum tensile strength was found to be in the hybrid composite having a ratio of banana and sisal 4 : 1.

As a continuation to the above study the dynamic and static mechanical properties of randomly oriented intimately mixed banana and sisal hybrid fibre reinforced polyester composites were reported[77]. Maximum stress transfer between the fibre and matrix was obtained in composites having volume ratio of banana and sisal as 3:1. The storage modulus was found to increase with fibre volume fraction above the glass transition temperature of the composites.

Researchers have also explored the incorporation of a fruit and leaf fibre in natural rubber[78]. The addition of sisal and coir fibres offered good reinforcement and resulted in improvement of properties.

2.1.3 Textile Biocomposites

The development of textile technologies such as weaving, knitting and braiding has resulted in the formation of composites that have superior mechanical properties, as continuous orientation of fibres is not restricted at any point.

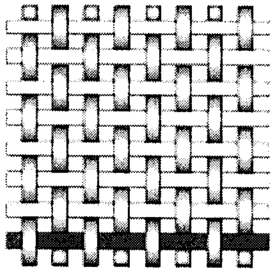
In applications where more than one fibre orientation is required, a fabric combining 0° and 90° fibre orientations is useful. Woven fabrics are produced by the interlacing of warp (0°) fibres and weft (90°) fibres in a regular pattern or weave style. The fabric's integrity is maintained by the mechanical interlocking of the fibres. 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.

2.1.3.1 Common Weave Architectures

The following is a description of some of the more commonly found weave styles:

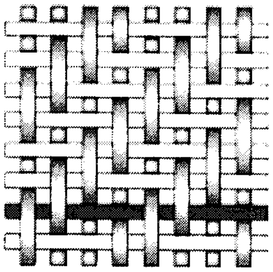
Plain

Each warp fibre passes alternately under and over each weft fibre. The fabric is symmetrical, with good stability and reasonable porosity. However, it is the most difficult of the weaves to drape, and the high level of fibre crimp imparts relatively low mechanical properties compared with the other weave styles. With large fibres (high tex) this weave style gives excessive crimp and therefore it is not generally used for very heavy fabrics.



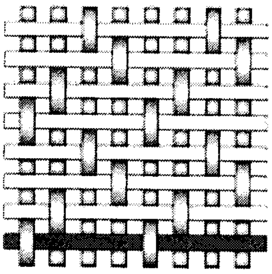
Twill

One or more warp fibres alternately weave over and under two or more weft fibres in a regular repeated manner. This produces the visual effect of a straight or broken diagonal 'rib' to the fabric. Superior wet out and drape is seen in the twill weave over the plain weave with only a small reduction in stability. With reduced crimp, the fabric also has a smoother surface and slightly higher mechanical properties.



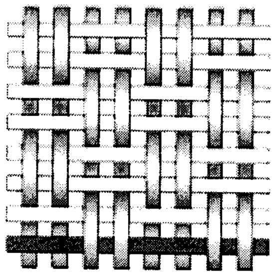
Satin

Satin weaves are fundamentally twill weaves modified to produce fewer intersections of warp and weft. The 'harness' number used in the designation (typically 4, 5 and 8) is the total number of fibres crossed and passed under, before the fibre repeats the pattern. A 'crowsfoot' weave is a form of satin weave with a different stagger in the repeat pattern. Satin weaves are very flat, have good wet out and a high degree of drape. The low crimp gives good mechanical properties. Satin weaves allow fibres to be woven in the closest proximity and can produce fabrics with a close 'tight' weave. However, the style's low stability and asymmetry needs to be considered. The asymmetry causes one face of the fabric to have fibre running predominantly in the warp direction while the other face has fibres running predominantly in the weft direction.



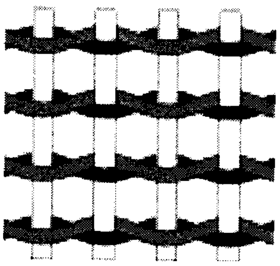
Basket

Basket weave is fundamentally the same as plain weave except that two or more warp fibres alternately interlace with two or more weft fibres. An arrangement of two warps crossing two wefts is designated 2x2 basket, but the arrangement of fibre need not be symmetrical. Therefore it is possible to have 8x2, 5x4, etc. Basket weave is flatter, and, though less crimp, stronger than a plain weave, but less stable. It must be used on heavy weight fabrics made with thick (high tex) fibres to avoid excessive crimping.



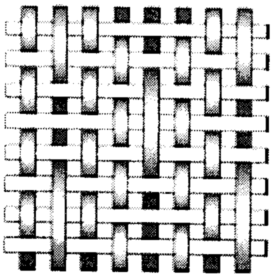
Leno

Leno weave improves the stability in 'open' fabrics which have a low fibre count. A form of plain weave in which adjacent warp fibres are twisted around consecutive weft fibres to form a spiral pair, effectively 'locking' each weft in place. Fabrics in leno weave are normally used in con-junction with other weave styles because if used alone their openness could not produce an effective composite component.



Mock Leno

A version of plain weave in which occasional warp fibres, at regular intervals but usually several fibres apart, deviate from the alternate under-over interlacing and instead interlace every two or more fibres. This happens with similar frequency in the weft direction, and the overall effect is a fabric with increased thickness, rougher surface, and additional porosity.



Researchers have looked into tensile strength of ramie-cotton hybrid fibre reinforced polyester composites[79] .They observed that tensile behaviour was dominated by volume fraction of ramie fibres aligned in the test direction. The fabric and diameter of the thread did not play any role in tensile characteristics. Cotton fabric was found to have minor reinforcement effect due to weak cotton/polyester interface. Similar studies were performed by Mwaikambo and Bisanda[80] on kapok- cotton fibre reinforced polyester composites.

Pothen et al.[81] conducted tensile and impact studies of woven sisal fabric reinforced polyester composites prepared by RTM technique. It was found that the weave architecture was a crucial factor in determining the response of the composites. Researchers have studied the micromechanics of moisture diffusion in woven composites[82]. The weave pattern of the fabric was found to have a profound effect on the water uptake of the composites. They observed that woven composites exhibited quicker diffusion than that of a unidirectional laminate with the same overall fibre volume fraction. The plain weave with a lenticular tow and large waviness was seen to exhibit the quickest diffusion process

Novolac type phenolic composites reinforced with jute/cotton hybrid woven fabrics were fabricated and its properties were investigated as a function of fibre orientation and roving/fabric characteristics[83]. Results showed that the composite properties were strongly influenced by test direction and rovings/fabric characteristics. The anisotropy degree was shown to increase with test angle and to strongly depend on the type of architecture of fabric used, i.e., jute rovings diameter, relative fibre content, etc. The best overall mechanical properties were obtained for the composites tested along the jute rovings direction. Composites tested at 45° and 90° with respect to the jute roving direction exhibited a controlled brittle failure combined with a successive fibre pullout, while those tested in the longitudinal direction (0°) exhibited a catastrophic failure mode. The researchers are of the opinion that jute fibre promotes a higher reinforcing effect and cotton fibre avoids catastrophic failure. Therefore, this combination of natural fibres is suitable to produce composites for lightweight structural applications.

The thermal diffusivity, thermal conductivity and specific heat of jute/cotton, sisal/cotton and ramie/cotton hybrid fabric-reinforced unsaturated polyester composites were investigated by Alsina et al[84]. These properties were measured both parallel and perpendicular to the plane of the fabrics. The results obtained show that higher values were obtained parallel to the plane of the fibres. Sisal/cotton composites showed a particular behavior, with thermal properties very close to those of the resin matrix. The thermal properties of the fabrics, i.e. without any resin, were also evaluated and were used to predict the properties of the composites from the theoretical series and parallel model equations. The effect of fabric pre-drying on the thermal properties of the composites was also evaluated. The results showed that the drying procedure used did not bring any relevant change in the properties evaluated.

Textile biocomposites comprising of woven sisal fabric and natural rubber were fabricated by Jacob et al[85]. The mechanical properties of the textile composites were found to decrease after chemical modification which was contrary to the behaviour in short fiber composites. The viscoelastic[86]and solvent sorption[87] characteristics of the composites were also investigated.

2.1.4 Applications Of Biocomposites

The use of biofibre reinforced composites has extended to almost all fields. Recently three-layer particleboards were produced from a mixture of sunflower stalks and poplar wood at certain ratios utilizing urea-formaldehyde (UF) adhesives. Panels with a density of 0.7 g/cm^3 were manufactured with the ratios of 25, 50, and 75 percent particles from sunflower stalks or poplar. Panels were subjected to various tests for physical properties. Results show that all the panels provide properties required by the normal standards for general purpose-use particleboards[88].

Bio-based composite roof structures were successfully fabricated from soy oil-based resin and cellulose fibres in the form of paper sheets made from recycled cardboard boxes. This recycled paper was previously tested in composite sheets and structural unit beams and was found to give the required stiffness and strength required for roof construction[89].

In a study encompassing many applications, the flame retardancy of biodegradable polymers and biocomposites was investigated[90]. For the comparison, flame retarded lignocellulosic filler reinforced biocomposites were prepared using polypropylene (PP), polyurethane (PUR) and fully biodegradable starch matrices. The compatibility of wood flake with PP was improved by application of an alkoxy silane based reactive surfactant. The silylation improved not only the compatibility but also the thermal stability of the wood flake according to TG measurements. Raman spectroscopic analysis of the silylated product showed that the improved thermal stability is the result of reduced ratio of the amorphous phase of cellulose. The phosphorus additives in flame retarded PUR biocomposites, comprising waste bio fillers and recycled polyol, proved to be very effective because both the matrix and the filler components participate in mechanism of flame retardancy.

Researchers[91] developed a new low dielectric constant material suited to electronic materials applications using hollow keratin fibres and chemically modified soybean oil. The unusual low value of dielectric constant was due to the air present in the hollow microcrystalline keratin fibres and the triglyceride molecules. The authors are of the opinion that the low cost composite made from avian sources and plant oil has the potential to replace the dielectrics in microchips and circuit boards in the ever growing electronics materials field. In an extension of the above study the authors have also observed that the coefficient of thermal expansion (CTE) of the composite was low enough for electronic applications and similar to the value of silicon materials or polyimides used in printed circuit boards[92].

Plastic/wood fibre composites are being used in a large number of applications in decks, docks, window frames and molded panel components[93]. It has been reported that 460 million pounds of plastic/wood fibre composites were produced in 1999[94]. Statistics show that the production of these composites in 2001 has increased to 700 million pounds[95]. Over the last three decades considerable research has been committed to finding an alternative fibre to replace asbestos in fibre cement products. Australian research was centred on natural fibres and ultimately it was a natural fibre, wood pulp fibre, which was responsible for the greatest replacement of asbestos in the beleaguered global fibre cement industry[96]. As these fibres are cheap and readily available the energy required for the processing of these composites is low; also the incorporation of random vegetable fibres in cement matrices requires only a small number of trained personnel in the construction industry. Recently, researchers[97] have explored the use of bamboo fibre as reinforcement in structural concrete elements. Pulp from eucalyptus waste and residual sisal and coir fibres have also been studied as a replacement for asbestos in roofing components[98]. The use of cactus pulp as a stabilising agent to improve the behaviour of the sisal fibre reinforced soil has been studied[99]. Chopped barley straw has also been used as a suitable reinforcement for composite soil[100].

Biocomposites also offer immense opportunities for an increasing role as alternate material, especially as wood substitutes in the furniture market[101]. Yet biofibre as construction material for buildings were known long before. For centuries, mixtures of straw and loam, dried in the sun, were employed as construction composite in Egypt[102]. Pipes, pultruded profiles and panels with polyester matrices are also quite popular. Large projects have been promoted where jute-reinforced polyester resins are used for buildings, e.g. the Madras House and grain elevators. Today, a renaissance in the use of natural fibres as reinforcement in technical applications is taking place, mainly in the automobile and packaging industries (e.g., egg boxes)

In the automotive industry[103], cotton fibres embedded in polyester matrix was used in the body of the East German "Trabant" car. The use of flax fibres in car disk brakes to replace asbestos fibres is also another example. Daimler- Benz has been exploring the idea of replacing glass fibres with natural fibres in automotive components since 1991. A subsidiary of the company, Mercedes Benz pioneered this concept with the "Beleem project" based in Sao Paolo, Brazil. In this case, coconut fibres were used in the commercial vehicles over a 9 year period. Mercedes also used jute-based door panels in its E-class vehicles in 1996. In September 2000, Daimler Chrysler began using natural fibres for their vehicle production. The bast fibres are primarily used in automotive applications because they exhibit greatest strength. The other advantages of using bast fibres in the automotive industry include weight savings of between 10 and 30 % and corresponding cost savings. Recent studies have also indicated that hemp-based natural fibre mat thermoplastics are promising candidates in automotive applications where high specific stiffness is required[104].

Virtually all the major car manufacturers in Germany (Daimler Chrysler, Mercedes, Volkswagen, Audi Group, BMW, Ford and Opel) now use biocomposites in various applications. Interior trim components such as dashboards and door panels using polypropylene and natural fibres are produced by Johnson Controls, Inc for Daimler Chrysler[105]. In 2000 Audi launched the A2 midrange car in which door trim panels were made of polyurethane reinforced with mixed flax/sisal mat. DaimlerChrysler has now increased its research and development in flax reinforced polyester composites for exterior applications[106].

Recent reports by Bledzki et al[107]. point out the importance of biofibers and biocomposites in the automotive industry (Figure 9 a & b). The end of life vehicle (ELV) directive in Europe states that by 2015, vehicles must be constructed of 95 % recyclable materials, with 85 % recoverable through reuse or mechanical recycling and 10 % through energy recovery or thermal recycling[108].This will definitely lead to an increased use of biofibres. The diverse range of products utilizing biofibres and biobased resins derived from soy beans is giving life to a new generation of composites for a number of applications like hurricane-resistant housing and structures[109].

2.2 Designing Biocomposites

Although biofibre reinforced polymer composites are gaining interest, the challenge is to replace conventional glass reinforced plastics with biocomposites that exhibit structural and functional stability during storage and use and yet are susceptible to environmental degradation upon disposal. An interesting approach in fabricating biocomposites of superior and desired properties include efficient and cost effective chemical modification of fibre, matrix modification by functionalizing and blending and efficient processing techniques. (Figure 10)

Another interesting concept is that of "engineered natural fibres" to obtain superior strength biocomposites[110].This concept explores the suitable blending of bast (stem) and leaf fibres. The judicious selection of blends of biofibres is based on the fact that the correct blend achieves optimum balance in mechanical properties for e.g., the combination of bast and leaf fibre is expected to provide a stiffness-toughness balance in the resulting biocomposites.

3. ELECTROSPINNING (ELSP)

Electrospinning has been recognised as one of the most efficient techniques for the fabrication of nanopolymer fibres. When the diameter of polymer fibre is shrunk from micrometer to nanometre range, amazing new properties are exhibited in the nanopolymer fibres. The large surface area of the fibres contributes to various applications ranging from medical prosthesis to composite preparation. In addition, the electrospun fibres find applications in various other fields such as tissue template, protective clothing, cosmetics, drug delivery, conducting nanofibres, sensors, and optical shutters etc.

Electrospinning has recently drawn strong attention in biomedical engineering[111], providing a basis for the fabrication of unique matrices and scaffolds for tissue engineering. ELSP is a spinning method that can produce polymer fibres with diameters ranging from several micros to 100 nm or less under a high-voltage electrostatic field operated between a metallic nozzle of a syringe and a metallic collector in air. The fibres are typically deposited in the form of a non-woven fabric onto a target metallic collector through a random projected jet of polymer solution, the so-called cone or instable jet. The accumulated charges on the polymer solution ejected from the nozzle induce the radial charge repulsion in the electric field, which induces the instable jet. If the random deposition process of fibres in the instability jet is appropriately controlled on the mesoscopic scale, the higher-ordered spatial placement of the nano- and microfibres in the ELSP fabrics will be realized. In addition, the high-speed movement of the collector or nozzle may produce oriented meshes and more higher-ordered macroscopic devices.

Researchers recently developed two novel ELSP techniques: multilayering ELSP and (multicomponent) mixing ELSP, both of which are composed of different nano- and microfibres[112]. In the multilayering ELSP, after electrospinning the first polymer, the second polymer is sequentially electrospun on the same target collector (See Figure 11). Such a sequential spinning process can produce a multilayered fibre mesh, in which a hierarchically ordered structure composed of different kinds of polymer mesh could be obtained. For the mixing ELSP, two different polymers are simultaneously electrospun from different syringes under special conditions. The produced polymer fibres are mixed on the same target collector, resulting in the formation of a mixed fibre mesh.

Tremendous amount of studies are being undertaken in the field of electrospinning. The effect of molecular weight on fibrous PVA prepared by electrospinning has been reported by Koski et al[113]. Zarkoob et al.[114] has reported on the structure and property of electrospun silk fibres. The preparation of PVA/Zirconiumoxychloride composite fibres by using sol-gel process and electrospinning was reported by Shao et al[115]. In the above work, fibres with diameter 50-200 nm could be obtained.

Recently scientists[116] have looked into the governing parameters in the electrospinning of polymer fibres. Researchers have studied the mechanical characterisation of electrospun gelatin nanofibres[117]. Another innovative approach has been the preparation of blend biodegradable nanofibrous non-woven mats via multi jet electro spinning[118] . Recently a leading group[119] at the Chonbuk National University, Republic of Korea has reported on the preparation of nano to submicron fibres of ruthenium doped titanium dioxide /poly (vinyl acetate) hybrid fibres by electrospinning.

Another interesting study was the electrospinning of chitosan nanofibres from aqueous chitosan solution using concentrated acetic acid solution as a solvent. A uniform nanofibrous mat of average fibre diameter of 130 nm was obtained from 7% chitosan solution in aqueous 90% acetic acid solution in an electric field of 4 kV/cm. An aqueous acetic acid concentration higher than 30% was prerequisite for chitosan nanofibre formation, because more concentrated acetic acid in water progressively decreased surface tension of the chitosan solution and concomitantly increased charge density of jet without significant effect on solution viscosity[120].

In an innovative study, ultrafine gelatin (Gt) fibres were successfully produced with the use of the electrospinning technique by by using a fluorinated alcohol of 2,2,2-trifluoroethanol (TFE) as the dissolving solvent[121].

4. CELLULOSE BASED NANOCOMPOSITES

The concept of nanostructured materials design is gaining widespread importance among the scientific community[122]. The strong reinforcement effects at low volume fraction resulted in a tremendous interest from the industry and research circles. With this as an inspiration, the potential of nanoscale cellulose structures as reinforcement in novel composite materials was extremely interesting. The concept of cellulose nanocomposites for load bearing applications is fairly new. Property enhancements are expected due to higher Young's modulus of pure cellulose reinforcement and finely distributed reinforcing microfibrils. A major problem in the commercial use of cellulose microfibrils in structural materials is the disintegration of cellulose from plant cell wall at reasonable cost and without severe degradation. Another major problem is dispersion of cellulose microfibrils in a polymer matrix.

A simple model of the cellulose microfibril structure is presented in Figure12. Cellulose nanocomposites are usually fabricated by utilizing these microfibrils of 10-50 nm on width as reinforcement in a polymer matrix. Although many studies provide detailed knowledge regarding the morphology and crystallography of different types of cellulose, the Young's modulus of microfibrils from different sources and subjected to different types of hydrolysis is seldom discussed.

The research group at CERMAV-CNRS has presented numerous studies based on cellulose whiskers reinforced nanocomposites[123]. The whiskers were disintegrated from an edible tunicate, a sea animal. Another area of interest is that of microfibrillated cellulose (MFC) nanocomposites. MFC based on parenchyma cells (i.e. sugar beet[124], potato tuber) tend to show small microfibril diameter and the fibrillar structures resemble swirled mats of connected, microfibrils rather than discrete rods. A review of the recent research into cellulosic whiskers, their properties and their application in nanocomposite field has been presented by the same group[125].

Recently scientists[126] achieved uniform polypyrrole nanocoating on natural cellulose fibres without disrupting the hierarchical network structures of individual cellulose fibres by means of polymerization-induced adsorption (Figure 13).

High performance composites from low-cost plant primary cell walls were developed by Bruce et al[127]. Purified cell wall fragments and cellulose microfibrils were developed from Swede root to form novel composite materials. The composites fabricated from purified cell wall fragments and acrylic matrix gave the best tensile properties. Researchers[128] have also reinforced natural rubber with nanofibres of sepiolite. The composites also contained silica particles generated in situ by the sol-gel process. The level of reinforcement was assessed from mechanical and orientation behaviour.

Algae are also a potential source of cellulose for use in composite materials. Researchers have concentrated on the morphology of cellulose powder from Caldophora spatza algae from the Baltic sea[129]. The effective stiffness of the algae MFC (which is the combined effect of the microfibril modulus and its aspect ratio) was found to be higher than for the wheat straw cellulose. Bacterial or microbial cellulose (BC) have also found their way as reinforcement in composites. Cellulose can be produced from some bacteria such as A.xylinum, vinegar or acetic acid. Bacterial cellulose is an extracellular product which is excreted into the culture medium. Acetic acid bacteria are not photosynthetic but can convert glucose, sugar, glycerol to pure cellulose. Bacterial cellulose has been frequently studied to clarify the mechanism for biosynthesis[130]. Researchers have looked into the development of cellulose nanocomposites based on bacterial cellulose and cellulose acetate butyrate[131]. Though BC composites have evoked widespread interest, the industrial use of BC composites requires the development of efficient large-scale fermentation technology.

Nanoscale-modifed plant structures such as paper or bast fibres also present a different approach to preparation of nanocomposite structures. An interesting study concentrated on the preserving the microfibril organization of wood veneer in composites[132]. Pine veneer was used as the starting material. The basic idea was to increase the volume fraction of cellulose microfibrils by partially removing the lignin and hemicellulose wood polymers and by compressing the veneer. Phenol formaldehyde resin (PF) was used to preserve the compressed microstructure of the material and to bond microfibrils.

5. CONCLUSIONS

The term biocomposite is now being applied to a staggering range of materials derived wholly or in part from renewable biomass resources. Keeping up with the developments in technology, it is compulsory that we stop developing materials, which are environmentally hazardous. This concept of bio-based materials has now become of key importance due to the need to preserve our environment. Biofibres like sisal, coir, hemp, oil palm are now finding applications in a wide range of industries. The field of bio-fibre research has experienced an explosion of interest, particularly with regard to its comparable properties to glass fibres within composites materials. The main area of increasing usage of these composites materials is the automotive industry, predominantly in interior applications.

Material revolution of this century may be provided by green composite materials. Sustainability, 'cradle-to-grave' design, industrial ecology, eco-efficiency, and green chemistry are not just newly coined buzz words, but form the principles that are guiding the development of a new generation of 'green'materials. Another promising area is that of nanocomposites and electrospinning of nanofibers.

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Cover Letter

Dear Editor

Enclosed please find the review entitled, “Biofibers and Biocomposites” for publication in journal Carbohydrate Polymers. Kindly acknowledge the receipt of the same.

Regards
Maya

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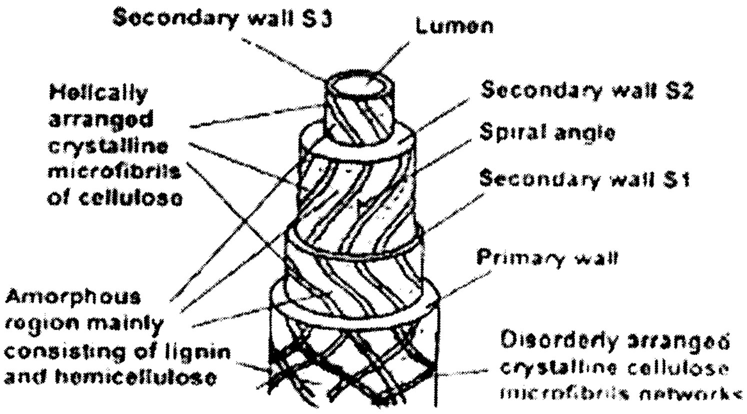


Figure 1. Structure of biofibre

[Reference: Rong M.Z. et al., Comp. Sci. Tech., 61, 1437, 2001]

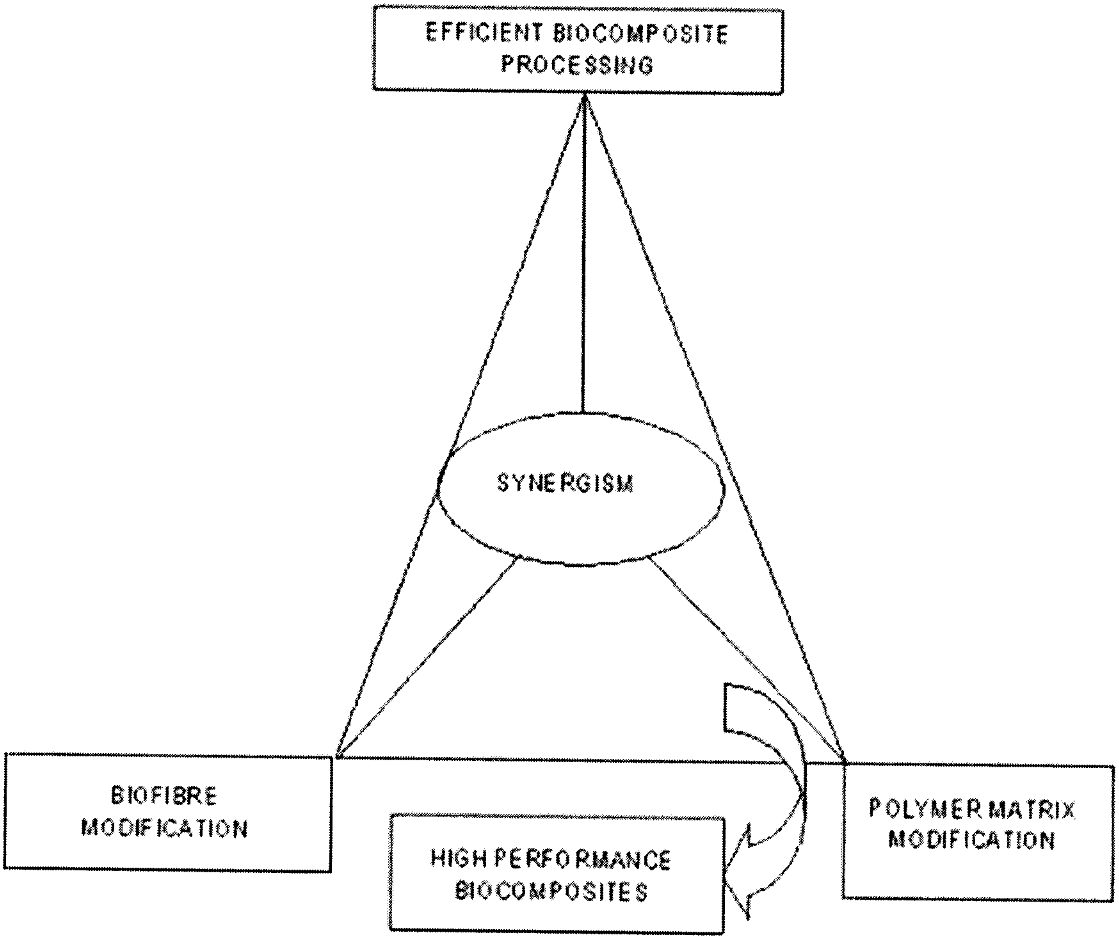


Figure 10 Tricorner approach in designing of high performance biocomposites.

[Reference: Adapted from Natural fibres, Biopolymers and Biocomposites, 37, Edited by Mohanty A.K., Misra M., Drzal L.T., CRC Press, 2005]

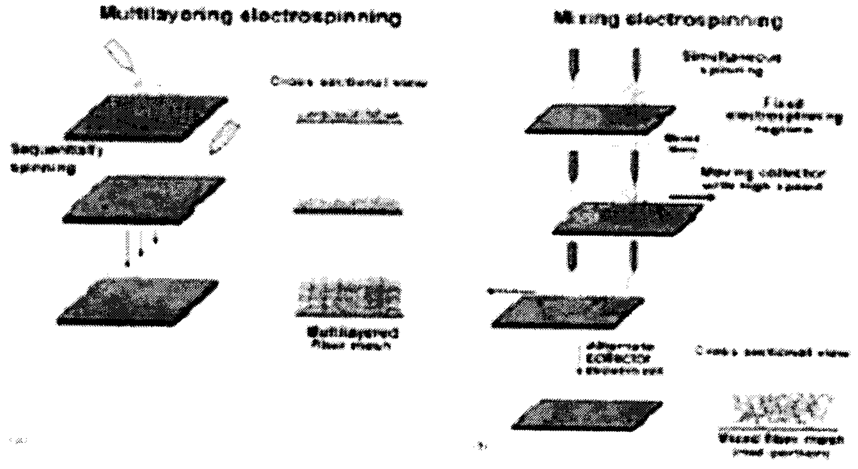


Figure11

[Reference: Kidoaki et al. Biomaterials 26, 37, 2005]

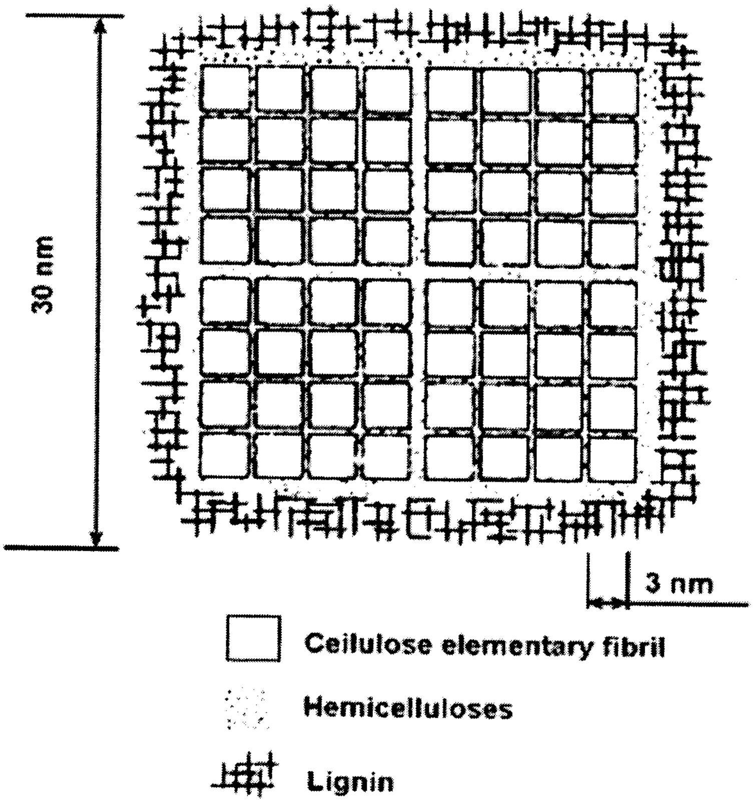
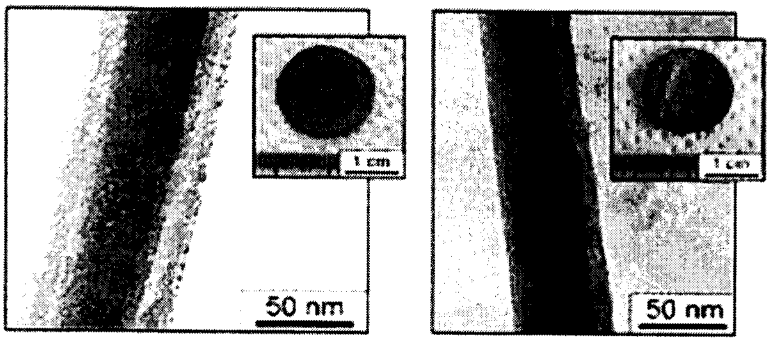


Figure 12 Model of the cellulose microfibril structure [Reference: Berglund L., Natural fibres, Biopolymers and Biocomposites, 808, Edited by Mohanty A.K., Misra M., Drzal L.T., CRC Press, 2005]



Cellulose fiber coated with polypyrrole layer

Cellulose fiber coated with titania and polypyrrole layers

Figure 13

[Reference: Huang et al., Chemical Communications, 13, 1717, 2005]

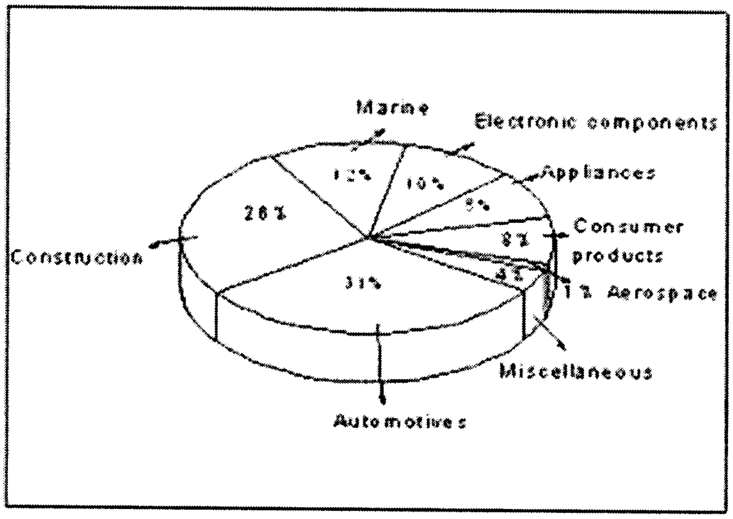


Figure 2 Fibre reinforced plastic composites used in 2002-2.28 X 10^9 lb. [Reference: Adapted from Plast. News. August 26, 2002]

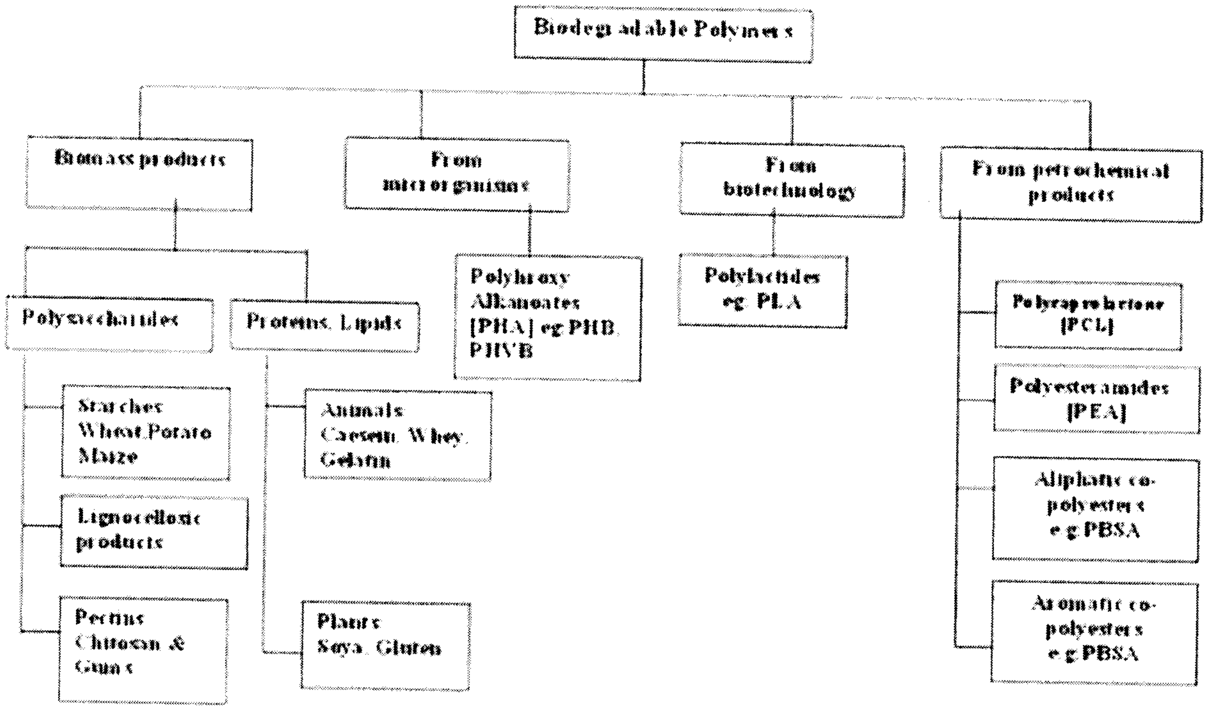


Figure 3 Classification of biodegradable polymers

[Reference: Averous L. and Boquillon N. Carbohydrate Polymers 56, 111–122, 2004]

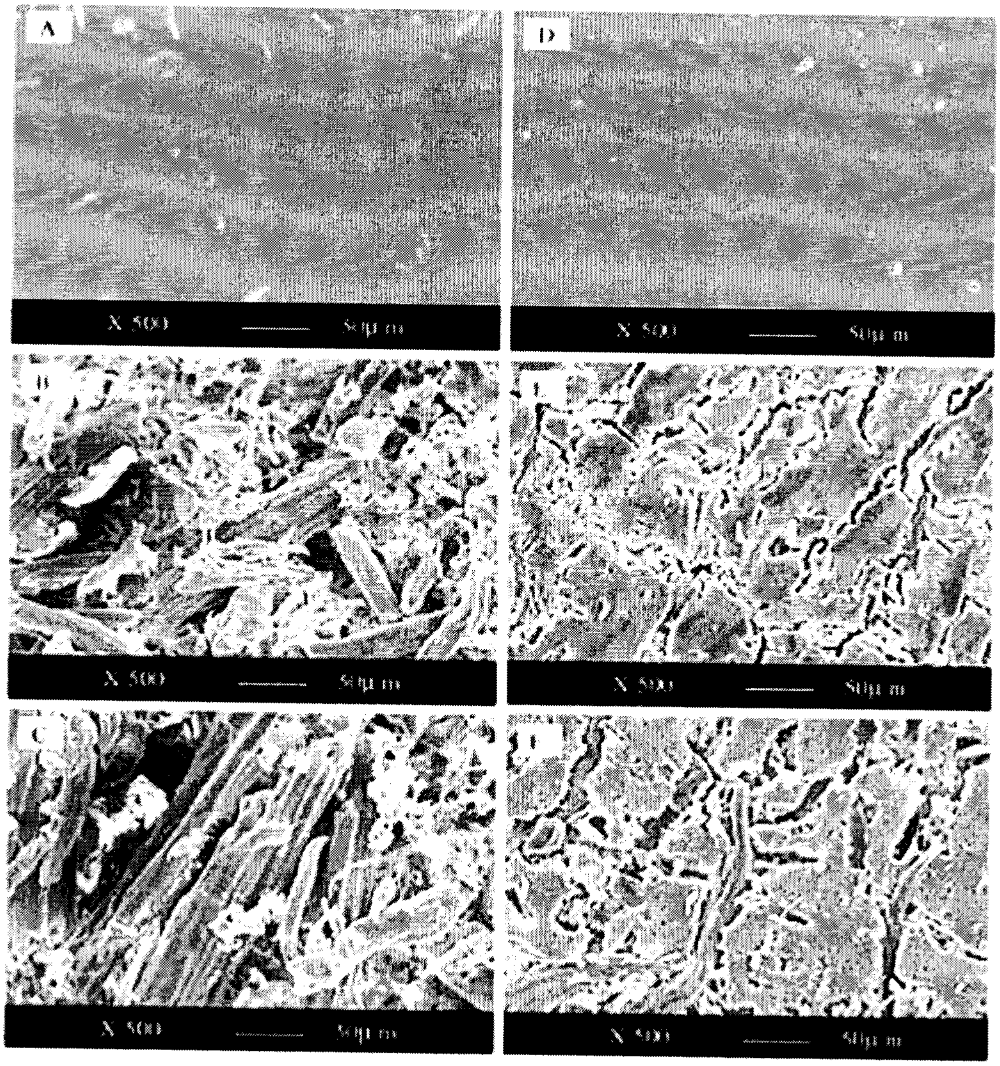


Figure 4. SEM micrographs of PLA or PBS/BF composites with LDI (NCO content, 0.65%) after different enzymatic degradation time. (A) control of PLA/BF composite, (B) after 7 days, (C) 9 days, (D) control of PBS/BF composite, (E) after 2 days, (F) 4 days.

[Reference: Lee and Wang, Composites Part A 37, 80-91,2006]

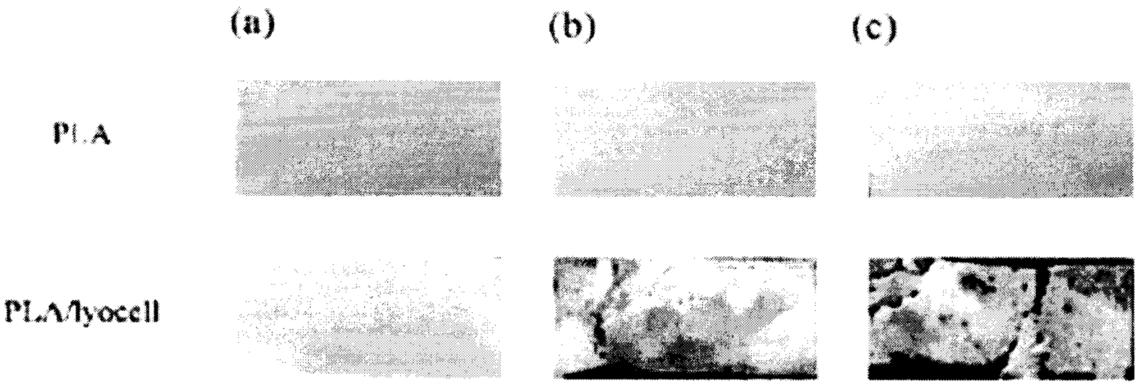
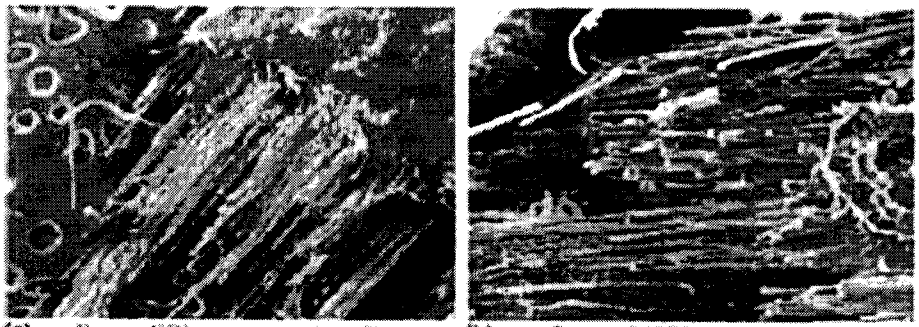


Figure 5 Photographs of PLA (top) and PLA/lyocell composite (bottom) after the soil burial test: (a) before test, (b) at 60 days, and (c) at 120 days.



(a) Ramie SPI composite (axial) (b) Ramie MSPI composite (axial)

Figure 6 (a & b) SEM photomicrographs of the fracture tensile surfaces of untreated and treated composites

[Reference: Lodha and Netravali, Comp. Sci. Tech. 65, 1211, 2005]

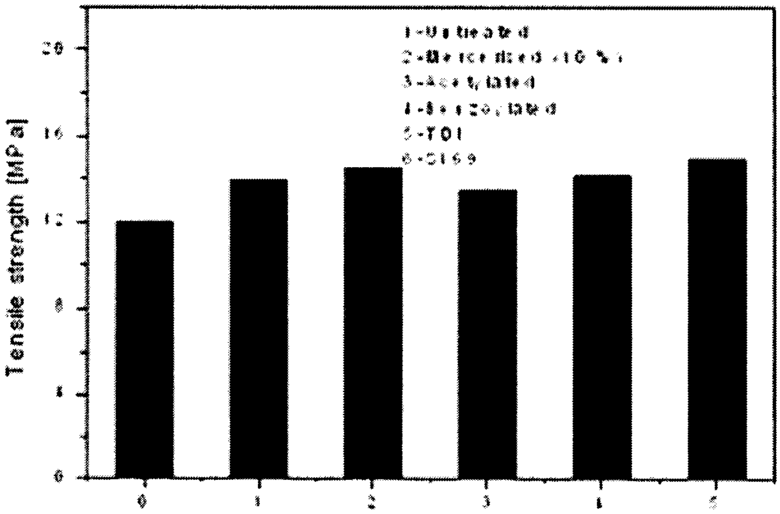


Figure 7 Variation of tensile strength with chemical modification [Reference: Mathew et al., Prog. Rubb. Plast. Recycl. Tech., 20, 4, 337, 2004]

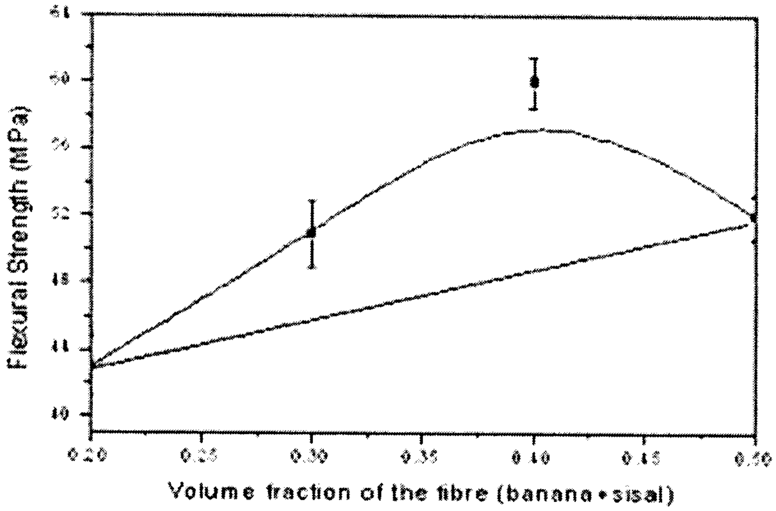


Figure 8 Variation of flexural strength with volume fraction of fibre
[Reference: Idicula M., et al. J. Appl. Polym. Sci. 96, 5, 1699, 2005]

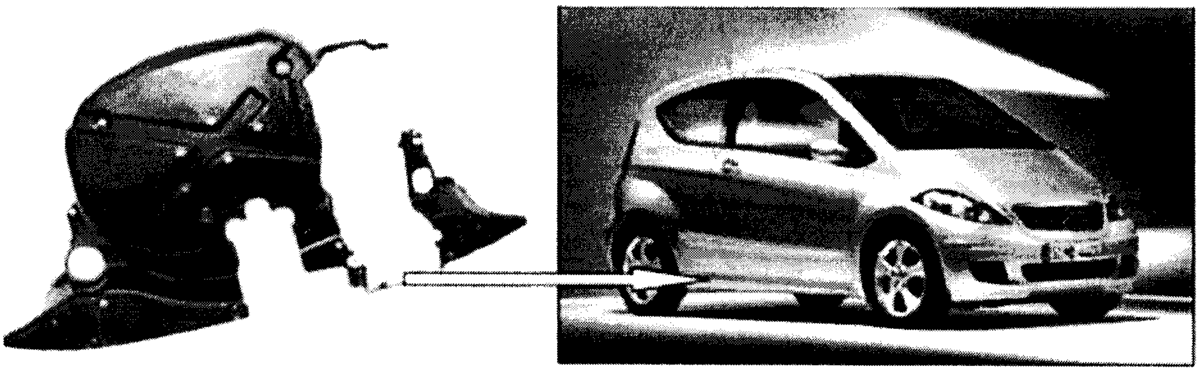


Figure 9 (a) Under floor protection trim of Mercedes A class made from banana fibre reinforced composites.

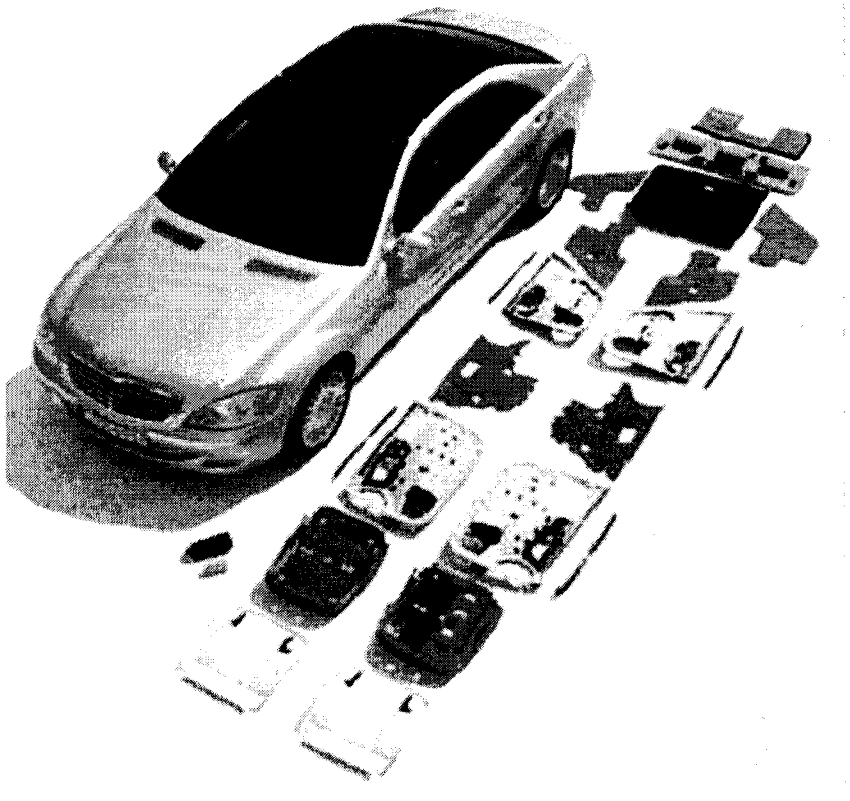


Figure 9 (b) Newest Mercedes S class automotive components made from different bio fibre reinforced composites.

[Reference: Bledzki A.K., Faruk O., Sperber V.E., Macromol. Mater. Eng. 2006, 291, 449–457]

Table(s)

Table 1. List of important biofibres

[Reference: Eichorn et al. J. Mat. Sci., 36, 2107, 2001]

Fibre source	**Species**	**Origin**
Abaca	*Musa textilis*	Leaf
Bagasse	-	Grass
Bamboo	(>1250 species)	Grass
Banana	*Musa indica*	Leaf
Broom root	*Muhlenbergia macroura*	Root
Cantala	*Agave cantala*	Leaf
Caroa	*Neoglaziovia variegata*	Leaf
China jute	*Abutilon theophrasti*	Stem
Coir	*Cocos nucifera*	Fruit
Cotton	*Gossypium* sp.	Seed
Curaua	*Ananas erectifolius*	Leaf
Date palm	*Phoenix Dactylifera*	Leaf
Flax	*Linum usitatissimum*	Stem
Hemp	*Cannabis sativa*	Stem
Henequen	*Agave fourcroydes*	Leaf
Isora	*Helicteres isora*	Stem
Istle	*Samuela carnerosana*	Leaf
Jute	*Corchorus* capsularis	Stem
Kapok	*Ceiba pentranda*	Fruit
Kenaf	*Hibiscus cannabinus*	Stem
Kudzu	*Pueraria thunbergiana*	Stem
Mauritius hemp	*Furcraea gigantea*	Leaf
Nettle	*Urtica dioica*	Stem

Oil palm	*Elaeis guineensis*	Fruit
Piassava	*Attalea funifera*	Leaf
Pineapple	*Ananus comosus*	Leaf
Phormium	*Phormium tenas*	Leaf
Roselle	*Hibiscus sabdariffa*	Stem
Ramie	*Boehmeria nivea*	Stem
Sansevieria (Bowstring hemp)	*Sansevieria*	Leaf
Sisal	*Agave sisilana*	Leaf
Sponge gourd	*Luffa cylinderica*	Fruit
Straw (Cereal)	-	Stalk
Sun hemp	*Crorolaria juncea*	Stem
Cadillo/ Urena	*Urena lobata*	Stem
Wood	(>10,000 species)	Stem

Table 2 Natural and biodegradable matrices

[Reference: Stevens E.S., Green Plastics, Princeton University Press, Princeton 2002]

Biodegradable polymer matrices	
Natural	Synthetic
1. Polysaccharides	1. Poly (amides)
Starch	2. Poly (anhydrides)
Cellulose	3. Poly (amide-enamines)
Chitin	4. Poly(vinyl alcohol)
2. Proteins	5. Poly (vinyl acetate)
Collagen/ Gelatin	6. Polyesters
Casein, Albumin,Fibrogen, Silks	6.1 Poly (glycolic acid)
3. Polyesters	6.2 Poly (lactic acid)
Polyhydroxyalkanoates	6.3 Poly (caprolactone)
4. Other Polymers	6.4 Poly (orthoesters)
Lignin	7. Poly (ethylene oxides)
Lipids	8. Poly (phosphazines)
Shellac	
Natural rubber	

Table 3 Comparative study of the water absorption of the glass fibre / natural biofibre with that of non-hybrid composites

[Reference: Rout J. et al., Composites Science & Technology 61, 1303, 2001]

Sample	**Water Absorption %**	
	Nonhybrid [Coir-Polyester composite] (20 wt. %)	**Hybrid [Coir/Glass - Polyester composite]**
Untreated	8.53	5.186
Alkali- treated (5 %)	4.994	3.147
PMMA grafted (5 %)	3.98	2.663
PAN grafted (10 %)	4.119	2.997
Cyanoethylated	3.6	3.138
Bleached	5.8	3.718