Journal of the Science of Food and Agriculture *J Sci Food Agric* **86**:1781–1789 (2006)

Review

Biocomposites: technology, environmental credentials and market forces

Paul A Fowler,* J Mark Hughes and Robert M Elias
BioComposites Centre, University of Wales, Bangor LL57 2UW, UK

**Abstract: Biocomposites offer a significant non-food market for crop-derived fibres and resins. Considerable growth has been seen in the use of biocomposites in the automotive and decking markets over the past decade or so, but application in other sectors has hitherto been limited. Nevertheless, with suitable development, the potential exists for biocomposites to enter new markets and thus stimulate an increase in demand for non-food crops. This paper reviews some of the technological challenges being faced in bringing these materials to a wider market together with potential solutions, as well as discussing market forces influencing their commercial uptake.
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Keywords: biocomposites; bioresins; biopolymers; non-food crops; wood-plastic composites; interface; matrix; reinforcement

INTRODUCTION

Following an exceptional period of growth[1] in the use of glass fibre reinforced synthetic resin matrices during the past 80 years, in applications as diverse as aerospace and thermal insulation, a resurgence in interest in their traditional forerunners has occurred in the last few years. This trend has been influenced by a number of factors, including increased environmental and health concerns, more sustainable methods of manufacture and reduced energy consumption supporting a desire for lighter-weight structures.[2,3] Materials from renewable resources are being sought to replace not only the reinforcement element[4] but also the matrix phase[5] of composite materials, thereby alleviating some of the sustainability issues associated with using synthetics in composites.

This paper seeks to provide an overview of the science and technology in relation to 'biocomposites', and assess the environmental benefits of the materials as well as the market forces impacting upon them.

DISCUSSION

What is a composite?

Composites consist of two (or more) distinct constituents or *phases*, which when married together result in a material with entirely different properties from those of the individual components. Typically, a man-made composite would consist of a reinforcement phase of stiff, strong material, frequently fibrous in nature, embedded in a continuous matrix phase. The latter is often weaker and more compliant than the former. Two of the main functions of the matrix are to transmit externally applied loads, via shear stresses at the interface, to the reinforcement and to protect the latter from environmental and mechanical damage.[6] The advantage of such a coupling is that the high strength and stiffness of the fibres (which in most practical situations would be unable to transmit loads) may be exploited.

What are biocomposites?

Biocomposites are composite materials comprising one or more phase(s) derived from a biological origin. In terms of the reinforcement, this could include plant fibres such as cotton, flax, hemp and the like, or fibres from recycled wood or waste paper, or even by-products from food crops. Regenerated cellulose fibres (viscose/rayon) are also included in this definition, since ultimately they too come from a renewable resource, as are natural 'nano fibrils' of cellulose and chitin. Matrices may be polymers, ideally derived from renewable resources such as vegetable oils or starches. Alternatively, and more commonly at the present time, synthetic, fossil-derived polymers preponderate and may be either 'virgin' or recycled thermoplastics such as polyethylene, polypropylene, polystyrene and polyvinyl chloride, or virgin thermosets such as unsaturated polyesters, phenol formaldehyde, isocyanates and epoxies.

The reinforcement/filler

Fibres provide strength and stiffness and act as reinforcement in fibre-reinforced composite materials; ultimately the properties of a composite are governed by the inherent properties of these fibres. Natural fibres can be subdivided into vegetable, animal and

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* Correspondence to: Paul A Fowler, BioComposites Centre, University of Wales, Bangor LL57 2UW, UK
E-mail: bss407@bangor.ac.uk

(*Received 14 November 2005; revised version received 6 December 2005; accepted 7 April 2006*)
Published online 17 July 2006; DOI: 10.1002/jsfa.2558

mineral fibres. This review is concerned only with vegetable fibres. Vegetable fibres can themselves be classified as either wood or non-wood. Non-wood fibres may be further subdivided into bast, leaf or seed-hair fibres, depending on their origin[7–10] in the plant, while wood fibre can, for convenience, be subdivided into softwood and hardwood fibre. All vegetable fibres, whether from wood or non-wood origin, are, however, composed of three main cell wall polymers: cellulose, lignin and matrix polysaccharides (such as pectins and hemicelluloses) associated with cellulose and lignin in the cell wall.[11] In addition to these, a number of non-structural components, such as waxes, inorganic salts and nitrogenous substances, broadly referred to as extractives, are also present.[12]

In structure, vegetable fibres can be viewed as miniature composites composed of millions of fibrous units known as microfibrils.[13] In turn, the microfibril can itself also be regarded as a composite material, analogous to man-made fibre composites such as glass reinforced plastic. The fibre constituent in this model is crystalline cellulose, while the matrix consists mainly of lignin, hemicellulose and other polysaccharides. The microfibrils are arranged in lamellae within the cell wall. The bulk of the cell wall, known as the secondary cell wall, is split into three layers – S1, S2 and S3 – with the S2 layer accounting for the greatest proportion of the cell wall material.[14] The microfibrils in the secondary cell wall are arranged in a helical fashion, with the microfibril angle (the angle between the winding angle of the microfibrils and the fibre axis) of the S2 layer influencing to a large extent the mechanical properties of the fibre.[15] Fibres with higher cellulose content, higher degree of polymerisation and a lower S2 layer microfibrillar angle exhibit higher tensile strength and modulus.[16,17]

The selection of suitable fibres is determined by the required values of the stiffness and tensile strength of a composite[18–20] (Table 1). Further criteria for the choice of suitable reinforcing fibres are, for example, elongation at failure, thermal stability, adhesion of fibres and matrix, dynamic and long-term behaviour, price and processing costs.

When considering the potential of natural fibres for composites and comparing the tensile strength, elasticity and elongation at failure with synthetic fibres, it becomes clear that hemp and flax fibres can potentially compete with E-glass fibres, which serve as a reference because of their great importance in composite technology.[9]

Table 1. Mechanical properties of some commercially important natural fibres and comparison with man-made fibres

Fibre type	Young's modulus (GPa)	Ultimate tensile strength (MPa)	Strain to failure (%)	Reference
E-Glass	76	2000	2.6	
HS carbon	230	3400	3.4	21
Kevlar™	130	3000	2.3	
Flax	–	814	–	22
	–	1500[a]	–	23
	103	690	–	24
	85	2000[a]	–	25
	50–70	500–900	1.3–3.3	26
	28	345–1035	2.7–3.2	27
	100	1100	2.4	28
	52	621	1.33	
Hemp	–	690	–	22
	25	895	–	29
	30–60	310–750	2–3	25
	–	690	1.6	27
	57	–	–	24
Jute	–	455	–	22
	8	538	–	29
	10–78	–	–	30
	27.6	393–773	1.7–1.8	26
	13	550	–	27

[a] Denotes fibre ultimates.

The matrix

The matrix in a fibre-reinforced composite holds the fibres together, transfers applied loads to those fibres and protects them from mechanical damage and other environmental factors. The matrix in most common traditional composites comprises either a thermoplastic or thermoset polymer, examples being polyethylene and unsaturated polyester, respectively. The manufacture of true biocomposites demands that the matrix be made predominantly from renewable resources, although the current state of biopolymer technology dictates that synthetic thermoplastics and thermosets dominate commercial biocomposite production.

Thus polyethylene and polypropylene have found widespread use in wood–plastic composites (WPCs), a particular subset of biocomposites that are currently receiving a significant amount of attention and are in widespread commercial production, particularly in North America.

By and large, such thermoplastic biopolymers that have been developed primarily for the packaging industry do not have the material properties to meet the matrix system requirements for other fibre composite materials. In particular, the overly high breaking elongation and high processing viscosity are disadvantageous for this intended usage.

As a result of the limitations of thermoplastics, there is considerable need for development in the area of thermosets from renewable resources.[31–33] Vegetable oils have been used as the building blocks of naturally derived thermosetting resins and typically are modified to form cross-linkable molecules such as epoxides,[34] maleates,[35] aldehydes[36] or isocyanates.[37] Currently, petrochemical reagents are still needed to cross-link these monomers. Among these substances, isocyanates, amines, polyols, phenols and polycarboxylic acids are preferred. The aim, of course, is to maximise the proportion of renewable resources used while retaining acceptable material properties.

Epoxidised vegetable oils form the basis of the nascent thermosetting biopolymer industry. The use of epoxidised vegetable oils and their combination with acrylate,[38] isocyanates[39] and polycarboxylic acid anhydrides[40] have been developed and commercialised for thermosetting and coating applications (see below).

While there is no shortage of potential thermoplastic biopolymers available from nature, for example starch,[41–43] proteins from grains,[44–46] polylactic acid,[47] polyhydroxy alkanoates[48] and natural rubber,[49] the majority remain at the developmental stage with only Cargill's NatureWorks PLA[50] and Novamont's Mater-Bi[51] starch-based thermoplastics in large-scale commercial production. At the time of writing, it is believed that no natural fibre reinforced composite based on these materials is commercially available.

Factors influencing the performance of biocomposites

Many factors combine to affect the properties of a composite material; ultimately, however, the properties of a composite are dictated by the intrinsic properties of the constituents. How these properties are harnessed to reinforce a composite material is largely governed by the same factors that affect other fibre composite materials, namely the fibre architecture and the fibre–matrix interface.

Fibre architecture

Fibre architecture, which encompasses (i) fibre geometry, (ii) fibre orientation, (iii) packing arrangement and (iv) fibre volume fraction, controls many composite properties, particularly mechanical properties.[21] Of these, fibre volume fraction (V_f) is probably the single most important factor,[6] with most mechanical properties increasing with increasing V_f up to a certain point. The maximum V_f achievable is, however, largely governed by the orientation and packing arrangement of the fibres and this is, in turn, dictated by the manufacturing process adopted.

The geometry of vegetable fibres is ultimately controlled by the morphology of the fibre tissue. Fibre geometry can to some extent be influenced by the way in which the fibres are extracted and processed. Softwood fibres, known as tracheids, are generally of the order 1.5–5.0 mm in length with transverse dimensions of between 15 and 80 µm, giving them an aspect ratio of around 100.[52] (Aspect ratio is the ratio of fibre length to diameter; thus fibres with high aspect ratio are long and thin, while fibres with low aspect ratio are shorter in length and broader in the transverse direction.) In certain processes, such as chemical or thermomechanical pulping, much of this aspect ratio can be maintained; however, hammermilling reduces the fibres to a particulate form, with low aspect ratio. The aspect ratio of individual bast fibre cells is somewhat higher and in the region of 1000–1200. Broadly speaking, it is advantageous to retain as much fibre length as possible, since higher aspect ratios give rise to greater reinforcing efficacy. Retaining fibre aspect ratio through processing and in composite manufacture is difficult, to the extent that in compounded and extruded WPCs the 'fibre' frequently serves only as filler and does little to impart true reinforcement to the composite.

At the micro-scale, work is ongoing to separate technical bast fibres into their elementary units so as to capitalise on the higher aspect ratio of these individual bast fibre cells as well as increase the surface area available for bonding to the matrix.[53] This process also serves to eliminate some of the defects inherent in the technical fibres, and strength improvements of up to 50% have been achieved using this method. An approach that is currently receiving attention by the research community is to break the fibres themselves down to form 'cellulose nanofibres', which can then be used to reinforce composites.[54,55] By breaking up the fibre cell, microscopic defects in the fibre, which can lead to localised stress concentrations within the matrix,[56] can be eliminated.

Another way of manipulating fibre architecture to improve performance is to align the reinforcing fibre within the matrix. In doing this, the packing arrangement is also generally improved, leading to higher V_f and hence better performance.

The fibre–matrix interface

The interface between fibre and matrix is also crucial in terms of composite performance. The interface serves to transfer externally applied loads to the reinforcement via shear stresses over the interface. Controlling the 'strength' of the interface is imperative. Clearly, good bonding is essential if stresses are to be adequately transferred to the reinforcement and hence provide a true reinforcing function. Another important mechanical property is toughness, or the ability of an engineering material to resist the propagation of cracks. This occurs in composites by virtue of their heterogeneous structure. It is important that under certain circumstances interfacial adhesion breaks down so as to allow various toughening mechanisms to become operative. These mechanisms include crack blunting as proposed by Cook and Gordon,[57] and various energy absorption processes such as the frictional sliding of debonded fibre fragments within the matrix, fibre fracture and the creation of new crack surfaces.[21]

Owing to the general incompatibility between natural fibres and most matrix polymers, methods of promoting adhesion are frequently needed. Several approaches have been explored, including chemical modification of the fibre prior to composite manufacture and introducing compatibilising agents to the polymer/fibre mix during processing.

Methods to enhance biocomposite performance

As already intimated, the properties of a composite are ultimately dependent upon the properties of the constituents. Thus, careful selection of the reinforcing fibres and matrix polymers, in light of the intended application, is the first step in obtaining a composite with the desired properties. Nevertheless, the properties of a biocomposite may be controlled and indeed enhanced by altering those factors that control composite properties, namely fibre architecture and the fibre–matrix interface. It must, however, be borne in mind that fibre architecture, and to some extent the interface, are affected by the manufacturing technique adopted (see below) and that, depending upon the desired performance characteristics and production volumes, this will control the extent to which performance can be improved.

Generally speaking, improvements in the mechanical properties of a composite can be gained by increasing the fibre volume fraction, although this will reach a maximum value, depending upon the packing arrangement of the fibre. The fibre–matrix interaction may be improved by making chemical or physical modifications to the fibre. All natural fibres are strongly hydrophilic owing to the presence of hydroxyl groups in the cellulose molecules. The hydrophilic nature of biofibres is a potential cause for incompatibility, adhesion and dispersion problems with hydrophobic polymer matrices. Chemical modifications of natural fibres such as acetylation,[58,59] silylation,[60,61] and other treatments reduce their moisture sensitivity. Much remains to be done to change/modify and improve bulk and surface characteristics to improve composite properties.[62]

Manufacture of biocomposites

The techniques used to manufacture biocomposites are based largely on existing techniques for processing plastics or composite materials. These include press moulding, hand lay-up, filament winding, pultrusion, extrusion, injection moulding, compression moulding, resin transfer moulding and sheet moulding compounding, but it is probably fair to say that the majority of current biocomposite materials based on thermoplastic polymers such as polypropylene and polyethylene are processed by compounding and extrusion.

Compounding and extrusion of thermoplastic polymers and natural fibres

During compounding, the thermoplastic polymer is heated, either by an external heat source or as a result of mechanical shearing in the extruder, so that it melts. In this state wood fibre, usually in the form of flour, can be added along with other additives to improve the characteristics of the resultant material. Once the constituents have been thoroughly mixed, the compound can be either extruded directly in the final product (see Fig. 1), or pelletised and packed as a precursor to further extrusion or injection moulding processes. There is great interest in the potential of injection moulding biocomposites and this is seen by some to be the ‘sleeping giant’ in terms of future growth in biocomposites in the industrial sector.[63]

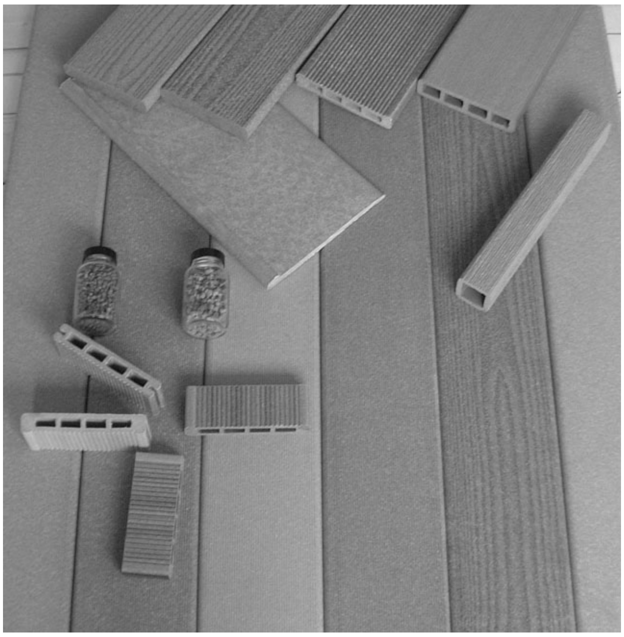

Figure 1. Examples of extruded WPC products (Courtesy of Entek).

One of the current limitations of compounding and extrusion is that only relatively short fibres (which impart limited reinforcement) can be used. If longer fibres are to be included, alternative methods may need to be employed.

Co-mingling of thermoplastic and natural fibres

In the automotive industry, longer fibres from flax, hemp, kenaf and cotton are frequently used.[64] These are generally mingled together with fibres of the thermoplastic polymer being used as reinforcement to form a non-woven ‘fleece’, which is subsequently hot pressed to melt the thermoplastic fibre thereby forming the composite. The advantage of this approach is that longer fibres (with better reinforcement potential) can be used.

Processing of thermosetting polymer matrix composites

Although there is significantly less commercial production of thermosetting polymer matrix biocomposites, interest in this area remains high. Manufacturing techniques broadly mirror those found in the ‘traditional’ composite industry, which include both ‘open mould’ (e.g. hand lay-up and spray-up) and ‘closed mould’ techniques (e.g. resin transfer moulding, vacuum infusion and compression moulding). A current barrier to the uptake of most current thermosetting biopolymers is the high cure temperatures needed. In many cases these exceed 150 °C, thereby limiting the tooling and process options available. Furthermore, most natural fibres cannot withstand processing temperatures higher than 175 °C for prolonged periods, limiting their ability to be used with some high-temperature curing thermosets. This manufacturing route is currently limited by the paucity of suitable renewables-based thermosets in the market.

Commercial applications of biocomposites

Applications of biocomposites are targeted towards the automotive, construction,[65] furniture[66] and packaging industries.[67]

Compared to glass, the hollow tubular structure of natural fibres provides better insulation against noise and heat in automotive applications such as door/ceiling panels and panels separating the engine and passenger compartments.[68] Panels made from long plant fibres such as flax, hemp, jute and kenaf, and from polypropylene or other thermoplastics, are already in use in many vehicles. All major vehicle manufacturers are exploring their use in other interior applications as well. These applications are essentially non-structural and the reinforcement provided to the thermoplastic polymer is good enough.

WPCs, made using wood flour or wood fibre as a reinforcement for polymers such as polypropylene, polyethylene and polyvinyl chloride,[69,70] are currently finding significant commercial application in the construction sector. The products formed from WPCs are commonly used in outdoor decking, window and door frames, automotive panels and furniture. However, their uses are limited to non-structural applications as the mechanical performance is poor; WPCs for decking applications are, for instance, only used in the deck and not the supporting structure. Nevertheless, the market for such composites has grown strongly in the past few years and shows promise of continued growth in the immediate future.[71]

In the USA, Environ Biocomposites LLC uses wheat straw in its Biofiber composites[72] and sunflower hulls in its Dakota Burl[73] composites. Both are made using isocyanate resins and are intended for furniture applications. Another US company, Flexform Technologies LLC, blends kenaf and hemp fibres with polypropylene and polyethylene terephthalate fibres to produce composites for various applications, including automobile door panels and dashboards, acoustic ceiling tiles and wall panels.

Other manufacturers of biocomposites based on triglycerides include Ashland Specialty Chemicals,[74] whose ENVIREZ 5000 product contains only 25% renewable content, while Prof Richard Wool and colleagues at the University of Delaware have collaborated with John Deere & Co. to produce fibre-reinforced composites for use as hay baler doors.[75]

In the packaging industry, starch-based materials are currently being used, as are products based on recycled fibres, which are used for boxes and other rigid packing media.

Phenix Biocomposites has commercialised its decorative composite board, Environ, made using recycled wastepaper and soyflour. Environ can be used for furniture, cabinetry and architectural non-structural applications. Being fully degradable, Environ can also be composted.

Factors affecting growth

Drivers to uptake (economic and environmental)

Factors such as greater environmental awareness, societal concern and the depletion of petrochemical resources together provide an impetus to drive the growth of new materials and products based on natural fibres and biopolymers. Waste disposal is becoming increasingly important with the recognition that landfill is not sustainable and as such costs are increasing, with more responsibility being placed on producers. Such drivers are regulations-based and the governments of a number of countries have established laws to encourage the use of recycled and/or bio-based 'green' products.[76,77] The 'Producer Pays' principle is encouraging manufacturers to take responsibility for their products throughout their whole life cycle.[78,79] Directives such as the End of Life Vehicle (ELV) and the Waste Electrical and Electronic Equipment Directive (WEEE) will not only stimulate improvements in the 'recyclability' of products, but will also create opportunities for the use of biodegradable materials in these products.[80] Components such as the casings for computers, monitors and mobile phone cases could all be produced from biodegradable composite materials.

In this respect, classical fibre reinforced polymers often cause considerable problems in terms of reuse or recycling at the end of their lifetimes. This is primarily because the compounds consist of very stable fibres and matrices. Biocomposite materials provide a competitive advantage for manufacturers over traditional reinforcing fibres like glass and resins such as polyesters as product reuse or recycling at the end of life becomes the norm.[81,82]

Additionally, technical benefits such as low density, high toughness, acceptable specific strength properties, ease of separation, enhanced energy recovery, carbon dioxide sequestration and biodegradability[83] will all act to drive the growth of markets based on biocomposites.[84] In particular, there is the belief that these technical benefits could have a real impact in the built environment, where improved thermal insulation properties and lower density could result in significant energy savings.

Furthermore, the desire to promote the use of crops grown for industrial, non-food purposes is driving the uptake of novel composites. In addition to providing a renewable raw material for a range of industrial applications,[85] the development of a sustainable industrial crops industry will help rejuvenate beleaguered rural communities, particularly if local processing occurs.

Against this backdrop of increasing awareness of sustainability issues and environmental pressures, there are exciting opportunities for 'green' materials. Nevertheless, there are certain technical, commercial and consumer barriers that will need to be addressed if these forms of material are to enter the mainstream market.

Barriers to uptake (technical, economic and environmental)

Technical barriers to the commercial uptake of biocomposite include understanding the current physical limitations of the raw materials and processing restrictions. In particular, the lack of suitable reinforcement textiles is currently hindering the wider-scale use of natural fibre as reinforcement in thermosetting fibre reinforced composites. The reasons for this are both technical and commercial. Current reinforcement textiles are limited to non-woven mats, or woven textiles produced for apparel end uses. Neither is particularly suitable for high-performance composite applications.

Other barriers to the use of natural materials are quality and consistency of the resources and reliability of supply. Particularly within the non-food crops sector, the supply of industrial fibres is currently limited and, allied to the lack of suitable textiles, there is urgent need for more research and development investment to help establish a stable supply infrastructure, including technical textiles' processing.

If biocomposites are to be sold on their environmental credentials, then it is vital that these are substantiated by life cycle assessment (LCA) or other reliable means. The results of LCA can be revealing and it is by no means certain that a natural fibre alternative product will be 'greener'.[86,87] Indeed, it has been demonstrated that the greatest impact in environmental terms often arises from the polymer matrix and it is partly for this reason that there is significant interest being directed towards the development of bio-based thermosetting and thermoplastic resins.

Standards used to assess materials favour existing products, and the cost of accreditation can be a significant barrier to the adoption of new materials. There is, therefore, the need for significant investment in production capability and new processing technologies to overcome some of the technical hurdles and make commercial biocomposites a reality.

FUTURE PROSPECTS

Presently, the main markets for biocomposites are in the construction and automotive sectors. With further developments and improvements in performance, however, new opportunities and applications will likely arise. Significant opportunities are likely to occur in the built environment as this sector is responsible for producing huge volumes of waste at a time when the environmental impact of industries is coming under close scrutiny. For example, new, 'environmentally friendly' materials are needed for off-site construction methods, improved quality and ease of installation and build. However, these opportunities may be hampered by regulations based on existing materials.

A particular area that offers significant potential for growth is in the replacement of preservative-treated wood. The introduction of tighter restrictions on the use of certain preservatives,[88,89] most notably those containing arsenic, presents an opportunity for biocomposites products in applications where there is a high risk of biological attack.

In addition to this, improvements in the mechanical performance of existing biocomposites[90] through, for example, the introduction of new fibre types, processing and additives may well result in an expansion in their use into more diverse, and technically demanding, application areas. An area of note in this respect is the ongoing research into solvent spinning of liquid crystalline cellulose, which looks promising for producing high-strength fibres.[91]

Biotechnology is being used to modify and/or increase the yield of specific triglycerides and oils[92,93] in crops for producing resins.[94,95] These resins will also be inexpensive compared with those available today and, if suitably modified, could be biodegradable. Research is also being conducted at various research laboratories to develop new pathways to synthesise inexpensive biodegradable resins[96,97] with better mechanical properties. Once fully developed, these resins and high-strength fibres hold great promise for replacing many of the synthetic advanced composites currently in use.

There are also opportunities for hybrid materials and products by, for example, using bioresins and bioplastics as adhesives in place of current fossil-based adhesives. There are also good prospects for using reclaimed fibre from products such as MDF (medium-density fibreboard) or other waste streams from the pulp and paper industry to manufacture a range of cost-effective and environmentally effective materials and products. While there is ample opportunity for these products to enter new markets and find new application, it is essential that benefits in terms of cost saving be highlighted and a stronger commercial case for these materials be made. Promoting the use of these materials through more widespread training and education is also needed if they are to be commercially successful.

CONCLUSION

Non-food crops and other bio-renewable resources offer an almost limitless supply of renewable and potentially sustainable raw materials for the production of biocomposites. Although in its infancy, there is a growing market for biocomposite-based products and with further development a whole host of new applications can be envisioned. There is a huge range of potential reinforcing fibres/fillers and an extensive range of processing options to ensure the right fibre at the right price. In parallel, significant developments have been seen in the realm of biopolymers in recent years. These combined ensure that biocomposites are likely to see a period of sustained growth; a note of caution, however – investment is still needed in research and development if a sustainable biocomposites industry is to be established.

This paper has sought to provide a broad overview of the technology associated with biocomposites, outlining some of the limitations of the material and pointing to some of the areas where performance may be enhanced. It has also sought to outline some of the perceived barriers and drives for biocomposites and finally to look at some of the future prospect for these materials.

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