# 10

# Bast and leaf fibre composite materials

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## 10.1 Introduction and background

The use of textile fibres for reinforcing composite products is not new but the last decade has seen the development of an entirely new market for bast and leaf fibres in this area; their use as plastic panel reinforcement (composites) for the interior trim of cars. According to Eisele (1994) the automotive industry was using vegetable fibres in car interiors before the 1970s. Examples of the uses of these fibres in older models are:

- jute needle-felts for sound insulation, placed under the carpet
- wadding, based on wool and cotton for seats and door trim panels
- rubberised coir upholstery for seats
- wood fibres for door trim panels.

During the 1970s and 1980s these fibres were partially replaced by petrochemical polymers (e.g. non-reinforced acronynitrile butadiene styrene (ABS) plastic) because of their optimised properties and their faster manufacturing processes. However, in 1999 50,000–70,000 t of wood fibre and 50,000–60,000 t of reclaimed cotton were still used in the manufacture of these products by the German automotive manufacturers alone because they are technically effective and economical.

In the mid-1980s it was thought that the use of natural fibre reinforced composites could offer an interesting alternative to these plastics because of their technical, economic and ecological advantages and social benefits (Harig and Müssig 1999). Several international and national bodies, and in particular the European Union and several of its member countries, provided R & D grants aimed at investigating the technical possibilities of reinforcing these panels with natural fibres and thus improving their performance. These projects were part of wider R & D programmes whose purpose was to develop potential industrial uses for locally produced vegetable fibres and other non-food crops and so help to persuade farmers to decrease those of their products that are subsidised under the European Union's Common Agricultural Policy and whose supply exceeds demand. At about the same time and owing to the completely international nature of the automobile industry similar work on using vegetable fibres to reinforce vehicle trim panels was being carried out in the United States and Japan.

Despite encouraging results from R & D work during this period it produced little concrete follow-up from the automobile industry until the mid-1990s when the first bast fibre reinforced plastic panels were incorporated into a German standard production model. Bast and leaf fibres are now penetrating the market due to the development of new manufacturing techniques. The advantages of these new composites in automobiles compared to those made with wood fibre or reclaimed cotton are:

- lower density of fibres, leading to a reduction in weight of 10% to 30%
- improved mechanical and acoustic insulation properties
- improved processing properties – lower wear of tools
- potential for one-step manufacturing, even when making complex parts
- improved accident performance – high stability, no splintering
- improved eco-balance, both during manufacture and during vehicle use (due to lighter weight)
- improved health benefit in manufacturing in comparison to glass fibres
- no release of noxious/toxic gases
- less condensation of emissions (fogging) compared to phenol-bonded composites
- price and ecological advantages compared to previously used technologies using synthetic or glass fibre
- positive effects on agriculture
- positive effects on the balance of payments in countries with temperate climates under which flax and hemp can be grown
- positive effects on the European Union's Common Agricultural Policy – decrease in excess agricultural production of subsidised crops.

According to a 1996 report by the IFEU (German Institute for Energy and Environmental Research) and nova-Institut, each kilogram of hemp fibre that replaces glass in this end-use saves 1.4 kg of carbon dioxide.

Since then the use of bast, and to some extent leaf fibres for this purpose has developed. Consumption in the European Union reached 18,000 tonnes in 2003 (Table 10.1).

## 10.2 The market, demand and supply

### 10.2.1 Demand

It can be seen from Table 10.1 that there is an almost linear increase in total fibre consumption averaging 10–20% per year from 1996 to 2002. In 2003

*Table 10.1* The use of natural fibres by the German automotive industry 1996–2003 (composites, excluding seat upholstery)

| | 1996 | 1999 | 2000 | 2001 | 2002 | 2003 |
|---|---|---|---|---|---|---|
| Flax | 2,000 | 7,000 | 9,000 | 8,500 | 9,000 | 9,400 |
| Hemp | 0 | 300 | 1,200 | 1,600 | 2,200 | 2,300 |
| Exotic fibres (jute, kenaf, sisal, coir, abaca) | 2,000 | 2,300 | 2,000 | 5,000 | 6,000 | 6,300 |
| Total | 4,000 | 9,600 | 12,200 | 15,100 | 17,200 | 18,000 |

Source: Karus *et al.* 2004.

consumption of these fibres continued to increase, but at a lower rate of about 5%. However, within these totals, different fibres were responsible for the overall growth of the market. Until 2000 this was due to an increase in the use of flax, but the price of flax tow increased considerably in 2001, due to the increased demand for linen. Since its 'rediscovery' in 1996 hemp has shown a continuous increase to about 2,200 tonnes in 2002 but this production started from a very low base and supply was unable to meet demand.

This shortage of hemp, despite a considerable increase in production, and the high price of flax allowed the 'exotic' fibres (jute, kenaf and sisal) to enter the market and partially replace them in 2001 and 2002. Although the use of all these fibres continued to increase in 2003 that of the 'exotics' was somewhat more marked than that of the other fibres. This was probably due to the slightly lower prices of the exotics, which, in its turn, was probably due to the marked decrease in the US$–€ exchange rate.

As stated above, in 2003 some 18,000 tonnes of these natural fibres were used to manufacture composite materials for the automotive industry in Germany and Austria, with a market value of approximately €10 million. This gives an average price of €0.55 to 0.60 per kg.[1] As it is known from previous market research[2] that the consumption of these two countries comes to two-thirds of the total European Union consumption, this is estimated at about 26,000 tonnes for that year.

At present these vegetable fibre composites are used in the manufacture of press-moulded thermoset or thermoplastic panels and 5–10 kg of fibre is used per car. Typical applications include door inserts, baggage racks, pillar covers, and boot linings. From a survey carried out on the ten European manufacturers of composite panels for the automotive industry in 2002, the use of vegetable fibres in the manufacture of these product is expected to increase at an average yearly rate of 14–15%. Table 10.2 lists the models of European cars which, between 1997 and 2001, were trimmed with bast or leaf fibre reinforced press-moulded panels.

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[1] 'Use of natural fibres in the German and Austrian automotive industry', nova-Institut, 2002.

[2] 'Markets and prices for natural fibres in Germany and EU', nova-Institut, 2002.

*Table 10.2* The use of natural fibre composite for series production in the automotive industry (1997–2001)

| Manufacturers/ Customers | Model/application (dependent on model) |
|---|---|
| Audi | TT, A2, A3, A4, A4 Avant (1997), A4 Variant (1997), A6, A8 (1997), Roadster, Coupe<br>Seat back, side and back door panels, parcel tray, boot lining, rear flap lining, rear storage panel, spare tyre lining |
| BMW | 3, 5 and 7 Series and others<br>Door inserts/door panels, headliner panel, boot lining, seat back |
| Citroen | C4 (2001)<br>Door inserts |
| Daimler/Chrysler | A-Klasse, C-Klasse, E-Klasse, S-Klasse<br>Door inserts, windshield/dashboard, business table, column cover |
| Fiat | Punto, Brava, Marea, Alfa Romeo 146, 156, Sportwagon |
| Ford | Mondeo CD 162 (1997), Cougar (1998), Mondeo (2000), Focus<br>Door inserts, B-column cover, parcel tray, in the future also motor protection (cover undershield) |
| MAN | Bus (1997)<br>Headliner panel |
| Mitsubishi | Miscellaneous models (since 1997) |
| Nissan | Miscellaneous models |
| Opel/Vauxhall | Astra, Vectra, Zafira<br>Headliner panel, door inserts, column cover, instrument panel, rear shelf panel |
| Peugeot | New model 406 |
| Renault | Clio, Twingo |
| Rover | Rover 2000 and others<br>insulation, rear storage panel |
| Saab | Coupe (1998)<br>Door inserts |
| SEAT | Door inserts, seat backs |
| Toyota | Miscellaneous models |
| Volkswagen | Golf A4, Golf 4 Variant (1998), Passat Variant, Bora<br>Door inserts, seat backs , rear flap lining, parcel tray |
| Volvo | C70, V70, Coupe (1998)<br>Door inserts, parcel tray |

Source: nova-Institut 2001.

## 10.2.2 The potential market

Approximately 18 million automobiles and lorries are presently manufactured in Europe per year. At 5 kg–10 kg of fibre per unit this would indicate a potential market of between 90,000 amd 100,000 tonnes per year. When the 37 million car and light vans manufactured in the rest of the world are taken into account the global potential market rises to between 250,000 tonnes and 500,000 tonnes.

At present all the bast fibre composite panels are produced using press moulded technology. Should the development of vegetable fibre injection-moulded composites take off, the size of the potential market would increase still further, possibly by as much as one million tonnes per year. However, one must bear in mind that potential markets are one thing, and the rate of penetration of a market by new techniques and products is another. This question is discussed in section 10.5 below.

### 10.2.3 Supply

Table 10.1 above gives details of the consumption of the various fibres used up to now to reinforce plastic car panels in the German automobile industry. At present (early 2004) the preferred fibres are flax, hemp, jute, sisal and kenaf, and Table 10.1 shows that, at the moment, short fibre flax, usually called flax tow (see Chapter 3 sections 3.5 and 3.12) is the principal fibre used. To a certain extent, these fibres are interchangeable and a composite manufacturer will bear in mind certain considerations. The market price for fibres used in the manufacture of these composite panels has settled in the range of €0.50 to €0.60 per kg in Europe.

*Flax*

At the price mentioned above the only grade of flax available is scutched tow. The other important market for this grade is the paper industry where it competes with wood pulp and reclaimed vegetable fibres, mainly cotton. These, in normal circumstances, limit the price obtainable by flax tow for this market to between \$300 to \$350 per tonne. Composites are therefore obviously preferred to paper as a market for these qualities of flax, even if this entails further processing the flax tow to reduce impurities to below the 2% level by weight required by the composite panels manufacturers (see section 10.4).

*Hemp*

All hemp fibre in the EU is produced on ‘total fibre’ lines, which produces fibres for speciality paper pulp (at between €0.40 and €0.45 per kg) or for industrial (as opposed to textile) uses for automobile composites or building insulation (at between €0.50 and €0.60 per kg). The price of hemp is more stable compared to that of flax (see Fig. 10.1).

*Non-European fibres*

The prices quoted for these are generally FOB and it is therefore necessary to add between US\$100 and \$50 to the quoted price for delivery to Europe or North America. When considering different fibres (apart from their delivered prices),

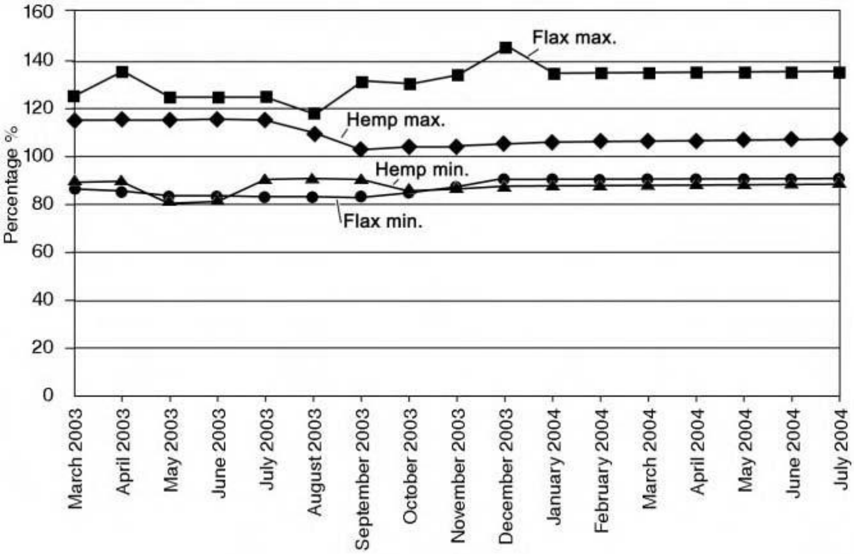

*Figure 10.1* The relative price stability in percentage terms of hemp and flax fibres for use in composite products. Adapted from: European Industrial Hemp Association, Source: Karus *et al.*, 2004 (www.industrial-crops.ed)

composite manufacturers and other suppliers to the automobile industry take the following into consideration:

- Price stability. As Fig. 10.1 shows they can obtain this to a reasonable extent from hemp and, to a less extent, from flax. However, it is true that flax, in this respect, does present a particular problem. This is that the tow fibre used for composites is a by-product of the production of line (long fibre) flax and, when the demand for line is high and its price rises, then the price of tow may also increase by 200% or 300%.. Prices of the other fibres are generally stable but do vary according to supply and demand and also due to exchange rate fluctuations, and especially that of the US dollar.
- The cleanliness of the fibre is important to the satisfactory bonding of the fibres that make up the composite product.
- The fineness of the fibre. Technically it is desirable to use a mixture of finer and coarser fibres (see section 10.4.3).
- There is a strategic requirement to diversify a composite manufacturer's sources of raw materials, thus reducing the risk of any interruption of supply. This diversification applies to the kind of fibre, its countries of origin and security of delivery. Coir (coconut) fibre is not interchangeable with the other fibres because its physical and chemical characteristics are very different (see Chapter 7 and Tables 1.1, 1.6, 1.9 and 1.12 of the Appendix to Chapter 1).

### 10.2.4 Availability

Taking the fibres in the order in which they are listed in Table 10.1.

#### *Flax*

Flax tow is a by-product of the production of line (long fibre) flax (see Chapter 3; section 3.5 and Table 3.11). Total world textile flax production is around 350,000 tonnes, of which it is reasonable to estimate that more than half is tow and the rest line, giving a world annual production of tow of more than 135,000 tonnes. Of this quantity perhaps not more than half, or 85,000 tonnes, is available for use in composites. This is due to several reasons: because of already existing markets for these qualities of flax from other, mostly textile end uses, because of lack of proximity – it is uneconomic to transport a fairly cheap product over long distances; and because not all of these tow fibres are of adequate quality for use in composites. At present levels of demand, therefore, there are sufficient supplies of flax to supply the market, but whether this will be the case in the medium to long term is not so certain and is discussed in section 10.5 below.

#### *Hemp*

Whilst the supplies of hemp are not, at present, large its production within Europe can easily be increased from year to year to meet the rising demand. The main countries producing hemp fibre in the European Union are France, Germany, the UK, and the Netherlands. Spain also produces substantial quantities, but all for the paper industry. The total world hemp fibre production is 50,000–60,000 tonnes. (See Chapter 4, part 2 for a fuller discussion of world, and particularly European hemp production.)

#### *Other fibres*

The global supply of jute, sisal, kenaf and coir is discussed in the chapters covering these fibres and is adequate to cover the present and future quantities required for the manufacture of composites for the automobile industry. If necessary their production can readily be increased. In Europe, jute fibres have been important in the automotive sector for many years. However, due to oily softening finishes, which are often used to improve the processing characteristics of jute fibres, problems are caused in terms of odour and fogging. This led to reduced acceptance in the automotive industry (Harig and Müssig, 1999). It is now clear that the decision to use natural fibres is not only the purchaser's, but that product requirements must be discussed by the entire production chain, if the highest possible and most reliable quality is to be achieved. Demand for kenaf fibres has risen continuously in the automotive industry in the last few years.[3]

[3] M. Karus, private communication, 2000.

## 10.3 The influence of fibre properties and the possibilities of measuring essential fibre characteristics

### 10.3.1 Introduction

In the last few years discussions on topics such as saving fossil resources, recycling, lightweight construction, the ecologically sustainable selection of materials and product-integrated environmental protection have led to a new attitude in the sector of material sciences. As a consequence of this new attitude each step in the flow of materials development, including research, production, processing, distribution, use, and recycling needs not only to meet functional and economic criteria but must also fulfil the ecological and social requirements of sustainability (Müssig, 2003).

In this context, the use in composite materials of natural fibres obtained from plants offers an interesting alternative to industrially created products and in the automotive industry in the last few years research has increased aimed at re-introducing natural fibres into the automobile construction industry. At the present time, all European automotive manufacturers have integrated components with natural fibres in the interior trim of their series production (Table 10.2). Present products range from the cladding of tailgates, columns and boots to covers for baggage compartments and door panels and the first structural elements for external application are being tested.

At the present time (2004), the full potential for natural fibre composites is far from having been achieved, and this is clear if we consider the great variety of possible applications for the interiors of many different types of car and other vehicles (Müssig, 2001). The market survey carried out by nova-Institut for 2003 (Fig. 10.2) shows that compression mouldings using the duroplastic (e.g. polyurethane matrix) method of manufacturing accounted for 35% of total natural fibre composites used by German automotive manufacturers and the

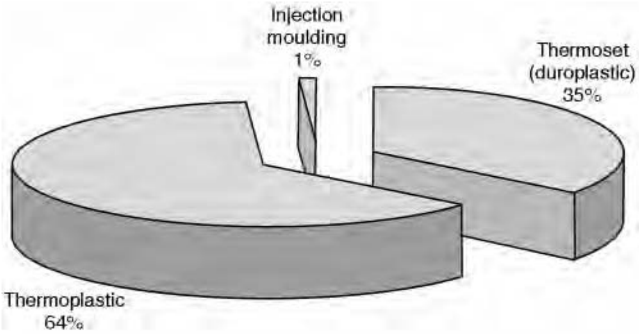

*Figure 10.2* Processing technologies for natural fibre composites in the German automotive industry. Source: Karus *et al.*, 2004

thermoplastic (mostly polypropylene) method for 64%. Injection moulded composites appeared for the first time in these market surveys and although they came to only 1% of total vegetable fibre composite production this is probably a very significant development.

As far as future trends to 2005 are concerned, an analysis of market surveys carried out by the nova-Institut show that 32% of the corporations or institutes questioned expect an increase in the importance of natural-fibre injection moulding (Kaup *et al.*, 2003). Thus this sector may become an important factor of growth in the future application of natural fibres in composites (Müssig *et al.*, 2003). This forecast is also confirmed by the present activities of the research institutes and research departments of industry.

Whilst, in the past, most work has been concerned with the extrusion compounding of short wood fibres, in the last few years the question of polypropylene/natural fibre injection moulding based on annual fibres such as flax or hemp has been the subject of intensive research. (Compare, for instance the work of Michaeli *et al.* (1995), Mieck *et al.* (1999), Snijder (2000), Jakwerth (2002), Reußmann *et al.* (2002), Specht *et al.* (2002) and also Ruch and Fritz (2002)). Nabi Saheb and Jog (1999) point out the importance of the use of adhesion agents in natural fibre/thermoplastic processing and also mention essential work on this subject.)

A particular problem in injection moulding concerns the introduction of the natural fibres into the processing units. Several different manufacturing processes were tested in this respect and also partially applied. In consecutive order these were:

- the trial of the metered addition of cut or pelletised natural fibres in the extruder
- the use of mixing batch techniques as described by Bornemann (2002), Karmaker and Youngquist (1996) for jute/polypropylene and Sanadi *et al.* (1995) for kenaf fibre reinforced compounds)
- the use of card sliver (Ruch and Fritz, 2002)
- the development of long fibre granulate production (Nechwatal *et al.*, 2002).

All of these are quite different concepts.

Beside these aspects of technical implementation, the ecological framework must also be taken into account and in this context the use of whole plants should be mentioned. As stated by von Buttlar *et al.* (2003), the use of natural fibres from whole plants, after they have been thermo-mechanically separated, represents a technically and economically interesting concept for injection-moulding applications with polypropylene. Hornsby *et al.* (1997) describe the possibilities of using a pulping extruder to pulp the stems of linseed flax before the pulp is mixed with polypropylene.

As well as publications and presentations there are many national and international patents, e.g., for the continuous manufacturing by extrusion of composites based on polymers and cellulosic fibres (Snijder *et al.*, 2003). Beckmann (2002) mentions a method for the production of natural fibre material, bonded with a thermoplastic, in a pourable and dispensable form.

There are also important developments in Japan. Kadowaki *et al.* (2003) give a circumstantial overview of the processing techniques published as patents in that country.

- A method in which natural fibre is kneaded to mix it homogeneously in molten thermoplastic resin using a roll kneader. The mixture is then cooled so as to solidify, and the product is crushed to form resin pellets (J-P-A No. 108161/1982).
- A technique in which discontinuous fibre is spun to form spun yarn. This is then processed to form fabrics; woven, nonwovens or mats. The processed product is impregnated with molten polymer in a bath and then cooled to solidify. The cooled product is then cut into a suitable configuration and size to form resin pellets. (J-P-A Nos. 28307/1983, 7307/1991, 30916/1991, and 41280/1997).
- A device in which molten polymer and discontinuous fibre are kneaded using a kneader or a kneading extruder and the molten mixture is then extruded, cooled, and solidified to form a rod, after which the rod is cut into pellets of the required length (J-P-A Nos. 146945/1987, 146947/1987, and 290453/1991).
- A processing technique by which a yarn spun from the reinforcing fibre is twisted with a yarn spun from the thermoplastic fibre. This composite two-ply yarn is then heated, cooled, solidified and cut into pellets of suitable size (J-P-A No. 163002/1992).
- A method in which a reinforcing fibre yarn is impregnated with molten thermoplastic resin, cooled and solidified, and then cut into pellets of a suitable length (for example, J-P-B Nos. 37694/1988, 57407/1994 and J-P-A Nos. 178411/1989, 119807/1992).

As shown above, the activities in the area of natural fibre-injection moulding have become more intense. However, a comparison of fibre and/or matrix properties is sometimes difficult due to differences in the procedural parameters and the different possible combinations of the fibre, matrix and adhesion agent. In view of these difficulties a first series of comparative tests of the different natural fibre-injection moulding procedures of eight leading corporations and institutes were carried out in 2002 (Karus *et al.*, 2002). The evaluation of the results offered a first approach towards improving the comparability of the current combinations of manufacturing, fibre and matrix developments on the basis of reproducible results. For these tests hemp fibres were prepared for processing with PP. It was not possible to use the available fibres as raw material for each of the different processes, which emphasises the difficulties of adding natural fibres to the compounding process.

The samples of composites obtained were tested for their mechanical properties (flexural, tensile and impact strength). Obvious differences between the tested samples were obtained:

- The addition of adhesion agents clearly improved the flexing and tensile properties.
- An increase in impact resistance is achieved by adding fibres with special force-elongation characteristics (Karus *et al.*, 2002).
- Of particular importance is the influence of the fibre properties on the behaviour of the manufactured composites.

In the following section the relationships between fibre and composite material characteristics are addressed and the possibilities of testing fibre properties are presented.

## 10.3.2 Fibre and composite material characteristics

*The influence of fibre length*

In automotive manufacturing, polypropylene is increasingly replacing plastics and metals in components such as dashboards, front-ends or cladding for the underbody. As summarised by Bürkle *et al.* (2003), PP can fulfil the required mechanical characteristics only if the stiffness and impact resistance of the material are increased by reinforcing the final product with fibres. The component production takes place as shown in Fig. 10.3 with the aid of compression or injection moulding techniques. With the aid of GMT very good mechanical characteristics may be achieved due to the possible length and isotropic properties of the fibres. However, due to economic aspects (amongst others) the LFT-procedures and also the IMC-technology represent interesting alternatives. For the processing of natural fibres both the LFT-procedures (compare for instance the work of van den Heuvel (2002)) and the IMC-technology (compare for instance the work of Zimmet (2002) and Ruch *et al.* (2002)) have been studied.

In order to ensure that a fibre that is introduced into a composite shows the highest applicable reinforcement effect the length of the fibre must be greater than the so-called critical fibre length (lc). (The critical fibre length is defined as the minimum length at which a fibre will improve the mechanical characteristics (strength and impact) of the composite concerned.) The polymer may be effectively reinforced by using fibres whose length exceed the critical length in an ideal fibre-matrix-adhesion combination. A connection between fibre length and composite properties is shown by Bürkle *et al.* (2003) as a rough orientation for glass-fibre-reinforced polypropylene (see Fig. 10.4). In natural-fibre reinforced polymers the connection between the length of the fibres and the properties of the composite material is not always as easy to

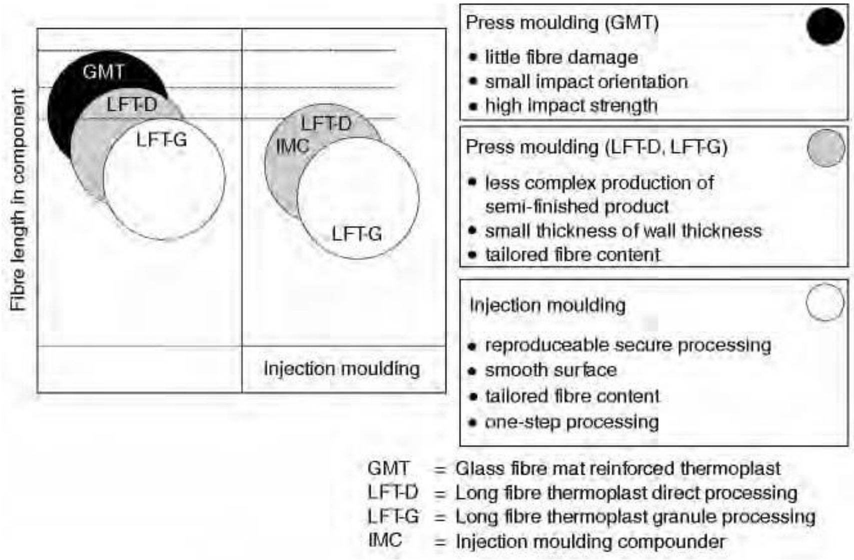

*Figure 10.3* Processing techniques for long fibre reinforced thermoplasts. Source: adapted from Bürkle *et al.*, 2003.

forecast as might be expected from the descriptions of glass-fibre/PP composites.

Ruch and Fritz (2002) report that unlike the reinforcement with extremely resistant glass and carbon fibres, the absolute length of flax fibre-bundles plays a less dominant role with regard to the mechanical properties of a composite. Due to processing with extrusion techniques and the application of combing elements arranged in a screw-type concept, ideally the fibre bundles are afined down to the individual fibres. As shown in Fig. 10.5, the length to diameter ration (l/d)

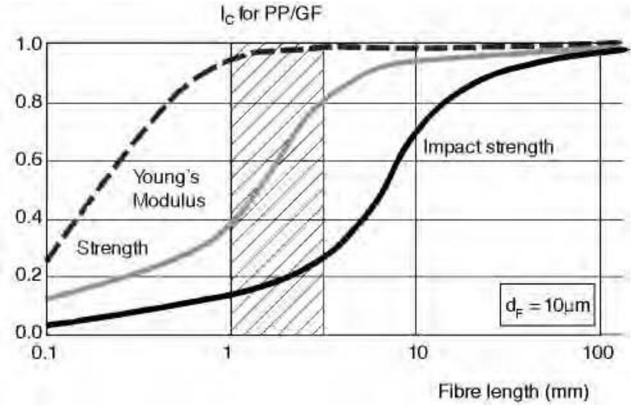

*Figure 10.4* The influence of fibre length on composite material characteristics. Source: adapted from Bürkle *et al.*, 2003.

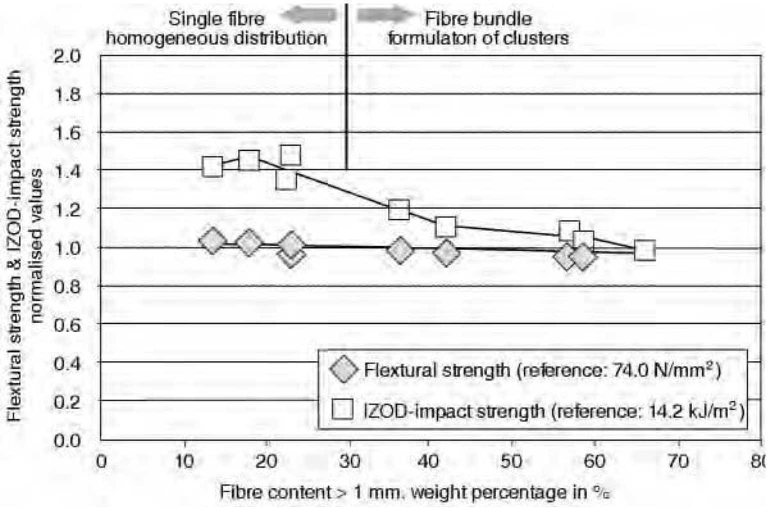

*Figure 10.5* The flexural strength and IZOD impact strength as a function of fibre content. Source: adapted from Ruch and Fritz, 2002.

ratio of the fibre strongly influences the mechanical properties of a composite. Thus excellent mechanical properties may also be achieved by a broader distribution of fibre lengths.

## Recycling

The comparison of recycling characteristics between glass and flax fibre-reinforced thermoplastics is clearly in favour of the natural fibres. As shown in the work of Aurich *et al.* (1998), the repeated re-processing of the brittle glass fibres leads to a considerable reduction of fibre length. For glass-fibre-reinforced thermoplastics this leads to a reduction of 50% of the tensile properties after five reprocessing cycles, whilst the characteristic values of the flax-fibre-reinforced variant remain on the same level.

Apart from fineness and length the orientation of the fibres also plays an important role. Since orientation plays a key role in the improvement of mechanical properties, finer fibres also allow more fibres to be orientated in the 0° direction. This effect has been demonstrated by experiments published in de Albuquerque *et al.* (2000). They determined properties of jute-polyester composite depending on fibre volume fraction and fibre orientation. Their results show that the mechanical properties of samples improve with the number of fibres orientated in the longitudinal direction.

In the use of sisal fibres (*Agave sisalana* P.) for the reinforcement of polyethylene (LDPE), Joseph *et al.* (1993) observe maximum stiffness values for fibre lengths of 6 mm. At lengths of 2 mm and 10 mm, lower mechanical characteristics are determined. At 2 mm, the length of the fibres is insufficient for ideal force transmission, a reorientation of the 10 mm long fibres takes place in processing, so that in this case force transmission is also not ideal. This description stresses the importance of the influence of fibre orientation and that it must also be taken into account for short fibre lengths. In section 10.3.3 the possibilities are mentioned for combining the measurement of fibre orientation with the determination of fibre fineness.

In order to explain the dependence of mechanical properties on the degree of separation of the fibre bundles, the construction of a flax or hemp fibre bundle must be explained and the influence of fibre fineness on the composite behaviour must be pointed out.

### *The influence of fibre fineness*

As shown in Fig. 10.6 in stalk plants such as hemp (*Cannabis sativa* L.) or flax (*Linum usitatissimum* L.), the fibre bundles are embedded in the outermost layer of the plant bark. The fibre cells are glued together to form fibre bundles with adhesive substances of the plant and may be decorticated and separated by physical, physical/chemical, chemical or microbial processes into finer fibre-bundles or into individual fibres.

As mentioned in the work of Ruch and Fritz (2002), in natural fibre flax the mechanical properties of the composites show a strong dependence on the degree of separation of the fibre bundles. As shown in Fig. 10.5, the values for flexural strength are slightly higher when homogeneously distributed individual fibres are present in the polymer. The influence of fibre bundle afinement becomes much clearer when the values for impact strength are considered. Afinement of the flax-fibre bundles into individual fibres and their homogeneous distribution considerably increases the dynamic properties of the composite. Ichazo *et al.* (2000) also report that, in sisal-fibre-reinforced polyolefins the fibre bundles are separated due to their processing in a twin-

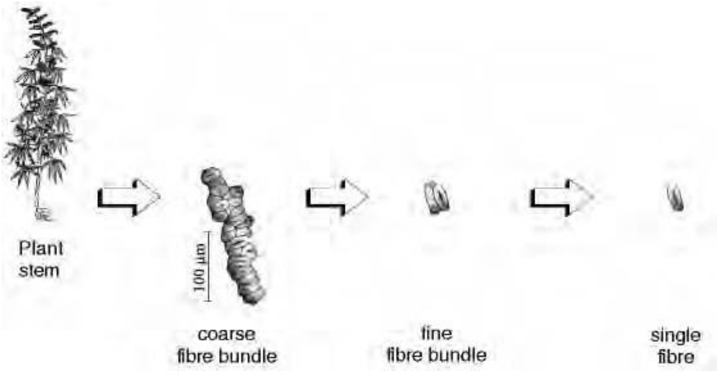

*Figure 10.6* From plant stalk to individual fibre.

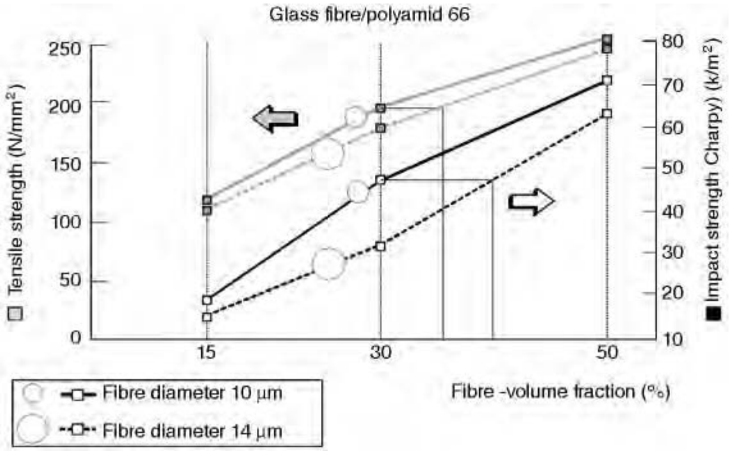

*Figure 10.7* The influence of fibre fineness on composite material characteristics. Source: adapted from Anon., 1995.

screw extruder. Due to this afinement of the fibre bundles, their effective surface area is increased which, on the whole, leads to an increase of the possible contact surface of the polymer and to an improvement in its mechanical properties.

For glass-fibre-reinforced polyamide, similar correlations apply and are represented in the graph of Fig. 10.7. For the combination of glass/PA66 it was shown that impact resistance changes far more than tensile strength when the fibre diameter is reduced. With an increase of the glass fibre diameter from 10 to 14 $\mu$m the impact resistance is reduced by 35% at a fibre content of 30% in the composite material glass/PA66. The values of tensile strength, however, change only by 10% (Anon., 1995).

Our own studies of the influence of fibre bundle refinement on the behaviour of the composite materials show similar tendencies (Müssig, 2001). The application of very fine steam pressure-separated hemp fibre bundles results in improved values for tensile strength compared to coarser separated fibre bundles, but also and above all, in an improvement of the impact strength values. The use of finer fibres provides advantages due to a more favourable length/diameter ratio, a larger contact surface between fibre and polymer, a larger amount of fibres with the effect of reduced tension concentration at the fibre ends and higher energy loss due to stronger fibre pull-out from the polymer. Kohler and Wedler (1995) arrive at the same conclusions in their work on composites of the moulded type flax-epoxide-2D-2D-combination. It thus becomes clear how important fibre fineness is for the behaviour of the composite. In the section 'Measurement of selected fibre properties' an automated image-analysis system is presented, which supplies reliable measurement values across the whole range of fibre finenesses.

*The influence of the force-elongation characteristics of the fibre*

Within the framework conditions of ideal adhesion and above-critical fibre length a fibre – with a corresponding force-elongation curve – may effectively improve its tensile properties for instance. Figure 10.8 shows the principle of reinforcement of a polymer by the introduction of stiff fibres that are orientated in the direction of the force applied.

If plant fibres are to be selected that correspond to the basic construction principles of composite materials according to Fig. 10.8, nature offers a great variety of possibilities. As reported by Herrmann and Hanselka (1995) the fibres of stalk plants such as hemp (*Cannabis sativa* L.), flax (*Linum usitatissimum* L.) or ramie (*Boehmeria nivea* H. et A.) offer high stiffness and are suitable according to Fig. 10.8 for application in composite materials. The influence of the fibre properties, for instance, on flexural properties of composites is, however, not uni-parametric and depends not only on fineness but also on the stiffness and the force-elongation characteristics of the fibre (Müssig, 2002).

If natural-fibre composites are to be optimised in terms of impact resistance values it is necessary to use fibres that provide a special force-elongation characteristic. By the use of strong cotton fibre with simultaneously high elongation values, impact resistance properties may be achieved that are considerably improved over the stiffer hemp fibre (Müssig, 2002). The increase of impact resistance of natural fibre and cellulose fibre composites by the use of fibres with increased energy at breakage is also described by Mieck *et al.* (1999), Reußmann *et al.* (2002) and Weigel *et al.* (2002).

In the area of injection moulding the results of a series of tests (Karus *et al.*, 2002) show that the hemp-fibre-reinforced polypropylene varieties provide

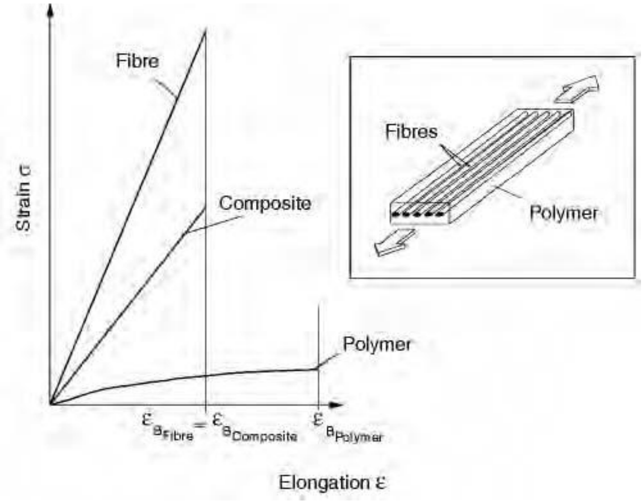

*Figure 10.8* The properties of one-directional composite materials.

relatively low impact strength values. By the addition of PAN-fibres the impact resistance values may be more than doubled compared to the pure hemp/PP variant. The combination of cellulose fibre/PP leads to clearly improved impact resistance values compared to all other trial variations in the test. In summary, it may be stated that compared to all other values of mechanical properties, the impact resistance values show the largest differences within the samples tested.

As shown above, the influence of the fibre force-elongation characteristics is the decisive factor concerning achievable properties of a composite material. However, for the determination of the tensile properties of fibres, reproducible and reliable methods are required which allow the determination of fibre characteristics that may also be used for material models for structural simulations. A method which can meet these requirements is described in the section ‘Determination of the force-elongation characteristics of fibres’.

## 10.3.3 The measurement of selected fibre characteristics

### *Fibre fineness*

The reproducible and quick measurement of fibre fineness is an important precondition for the determination of reliable fibre property values. Until recently, hardly any suitable systems existed for the measurement of the broad spectrum of finenesses of natural fibres that respected the framework conditions mentioned above. In order to solve this problem, a system has been elaborated in the last few years which allows the measurement of all fibre thickness distributions across all types of fibre. The new system is based on a high-performance scanner and image-analysis software that was developed for particle and diamond analysis (Schmid, 1998).

According to Schmid *et al.* (2002), in the development of the new, fully automated image analysis system FIBRESHAPE the following aspects were taken into account:

- development of a fast working method for the determination of thickness distribution of fibres or fibre-bundles
- determination of additional information such as colour, length and orientation
- quick and simple sample preparation
- reliable and reproducible results with little user influence
- no use of natural fibre standards for calibration of the tools.

In the following section the results of the test with the image analysis system FIBRESHAPE are presented. The fibre samples used were stored in a standard climate (20 °C, 65% rel. humidity) for 24 hours prior to preparation. These fibres were: cotton fibres, US-Pima-variety of the type *Gossypium hirsutum* L. (Müssig, 2002); coconut fibres (coir) (*Cocos nucifera* L.) type Omat from Sri Lanka (delivery December 2001); the fibres were provided for the tests by the company Hayleys, Sri Lanka; glass fibres, fibres from an E-glass filament (Nm 15) from a woven material of the company SSB, Bochum; hemp fibres/ mechanically separated, fibre bundles from the medium separation (MS) of hemp stalks (Felina/WE 96) of the unretted variety from the crop-year of 1996 in Oldenburg (Müssig, 2001).

The fibres were laid on a microscope slide with the aid of tweezers and were covered with a second microscope slide. The microscope slides were held together with adhesive tape applied to the short sides. Three microscope slides were prepared per sample. Scanning took place with the instrument Canoscan FS 4000 US and with the scanning software FilmGet FS 1.0 for Windows, and a resolution of 4,000 dpi (half-tone picture/positive) and a monitor with a gamma-value of 1,57 was selected. The exposure and focus were set to automatic. The analysis of the images took place with the image analysis software DIASHAPE Version 4.2.2/Software module FIBRESHAPE. The menu setting AFAS 4000.D00 was used. The results of the test are represented in the graph of Fig. 10.9. As well as the relative frequencies of fibre widths or fibre bundle widths the total frequencies were also noted. While the fibre types that are present as individual fibres (cotton and glass) show an almost normal distribution, the broad deviation to the left is noticeable in the distribution of the hemp fibre bundles.

It is shown that with FIBRESHAPE, on the one hand, a broad spectrum of possible fibre types may be tested, and on the other hand, the distribution of width of different elements may be measured quickly and reliably with little preparation work. As discussed in the section 'Influence of fibre length', the orientation of the fibres has a considerable influence on the properties of the composite. With the image-analysis system presented here it is possible to combine the analysis of width with the determination of fibre orientation. In order to document the feasibility of this option, ramie fibres were cut and prepared without any fibre orientation. Chinese chemically separated ramie (*Boehmeria nivea* H. et A.) – used for card sliver manufacturing (delivery 2000) – was used for this. The fibres were made available by the company Buckmann, Bremen. The prepared fibres were scanned and measured with the aid of FIBRESHAPE (menu setting AOR2400.D00/setup-change 4,000 dpi). In this measurement setting both fibre width and fibre orientation are measured as shown graphically in Fig. 10.10. For the scanned sample the orientation result is shown with a slight prevalence of orientation around 100°.

### *Force-elongation characteristics of fibres*

The influence of fibre stiffness and the force-elongation characteristics on composite behaviour is clear. Reproducible methods are required for the determination of the stated fibre properties that provide measurement values with the best possible usefulness for composite material development.

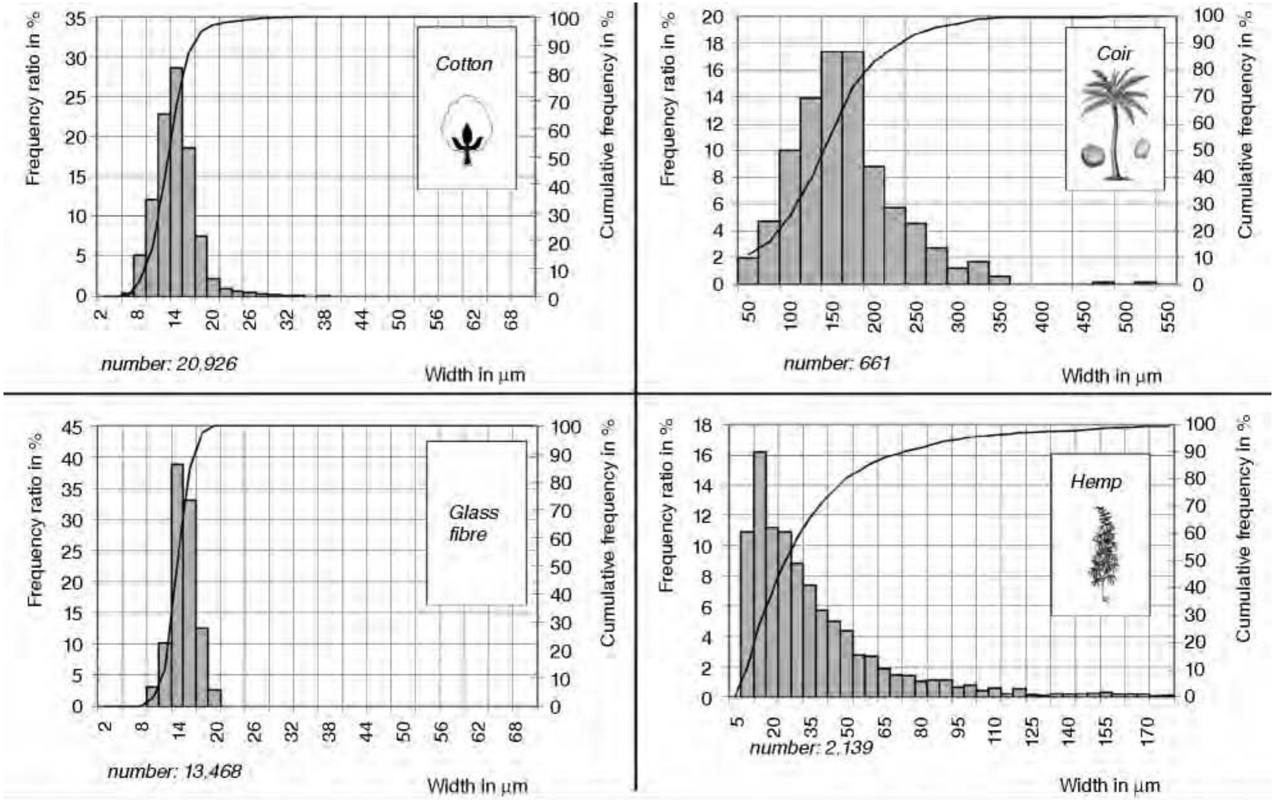

*Figure 10.9* Fibre and fibre bundle width distribution of cotton, coir, glass and hemp fibres. Source: Müssig, 2003.

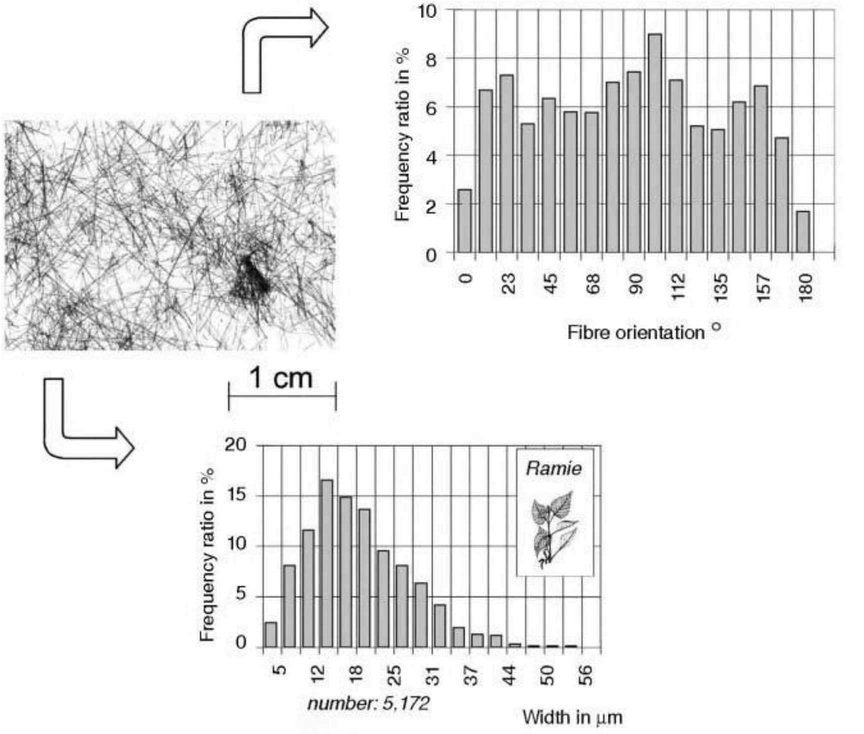

*Figure 10.10* Fibre width and fibre orientation distribution of Ramie. Source: Müssig, 2003.

According to Nechwatal *et al.* (2003) the single element test is of particular importance in the determination of the tensile properties of fibres. Problems in the implementation of such tests lie particularly in:

- the influence of the clamping mechanism and of fibre-slip in the clamp
- various fibre gauge lengths and accounting for this influence
- the determination of the fibre or fibre bundle cross-sectional area
- the calculation of the fibre modulus.

In order to solve the problems mentioned above and to reduce the number of possible influences on the testing result, an appropriate testing instrument was procured after an intensive exchange with the company DIA-STRON Ltd., UK and adapted to requirements.

Using the new method, the individual elements to be tested are no longer clamped but glued in order to reduce the influence of clamping and may be tested at gauge lengths of 30, 20, 10, 5 and 3.2 mm. The compliance of the system, 99% of which is due to the load cell, is corrected directly in the analysis. The compliance of the glue is negligible. Forty-five individual elements may be prepared per auto-sampler which are automatically brought to a laser, one after

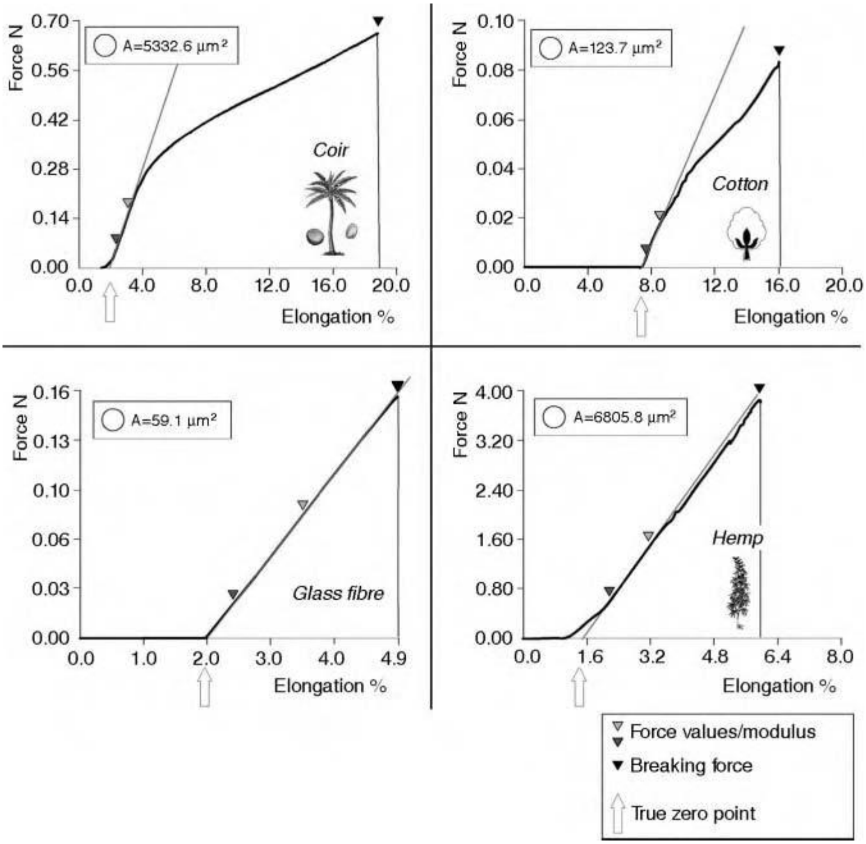

*Figure 10.11* Tensile properties of selected fibres. Source: Müssig, 2003.

another, which measures the cross-sectional area in several places (i.e. with 10 mm gauge lengths in five places). The sample is then automatically brought to the tensile system. After the tensile test with deliberate setting of the desired pre-tension, the sample containers are removed and the next samples are tested automatically. The software allows both the determination of areas (N/mm$^2$) and of fineness related values (cN/tex).

The analysis programme allows an extensive evaluation of the data, such as the determination of the true zero point, elongation correction, extensive possibilities for module determination, determination of breaking energy, and also the energy required for the removal of fibre crimp and the cross-section determination of the elements. In Fig. 10.11 the force-elongation curves from single-element tests (10 mm gauge length/velocity 1 mm/min) are shown as an example. The fibres shown are those that were used for the FIBRESHAPE-measurement in the section on fibre fineness. The tested fibre types show clear differences concerning the force-elongation curves, the maximum force achieved, elongation and stiffness and also in the cross-sectional areas that were determined.

With the system available from the company DIA-STRON and the method adopted, fibre parameters may be determined in a way that is useful for material sciences by responding to the need for a quicker determination of tensile properties of individual fibres and fibre bundles.

### *Natural fibres in compound materials*

Natural fibres from the stalks of plants offer particularly interesting properties for this end-use. Due to their supporting function in the plant they are naturally endowed with very good mechanical properties. Table 10.3 offers an overview of the properties of a few stalk fibres in comparison with glass fibre. These values are mean values whose purpose is to give a rough idea of the comparative characteristics of the fibres. Variation of characteristics due to influences of cultivation, harvest, retting and processing are not taken into account.

According to Herrmann and Hanselka (1994) those natural fibres with characteristics of high stiffness and low density are suitable for use as reinforcement of polymer products. According to Table 10.3, ramie, hemp and flax are particularly suitable. However, jute and kenaf also have interesting properties in composite materials for use as interior trim for automobiles. The possible uses for natural fibres in interior parts are quite comprehensive. One already widely established series component made of natural fibres is the interior door cladding. There are various composite materials with natural fibres and polymers on the market. One widely used combination is that of natural fibre needlefelt and polyurethane resins (Kleinholz *et al.*, 1996, Prömper, 1997 and Müller and Fries, 1998). The application of natural fibres need not be limited purely to interior trim. As Herrmann and Hanselka (1995) note, the potential of natural fibres, particularly hemp, reaches much farther and could allow their use in structural elements that today are typically made of glass-fibre-reinforced plastics.

*Table 10.3* Physical characteristics of selected fibres

| | E-Glass | Jute | Hemp | Flax | Ramie |
|---|---|---|---|---|---|
| Density (g/cm$^3$) | 2.54 | 1.44–1.49 | 1.47 | 1.48 | 1.50–1.55 |
| Tensile modulus (kN/mm$^2$) | 77 | 5–14 | – | – | 5–7 |
| Breaking force (N/tex) | 1.38 | 0.3–0.4 | 0.5–0.6 | 0.5–0.6 | 0.5–0.7 |
| Breaking elongation (%) | 1–4 | 1.3 | 3–4 | 3–4 | 2.4 |
| Tensile strength (kN/mm$^2$) | 3500 | 435–580 | 780–910 | 770–890 | 760–1,060 |

Source: Hermann and Hanselka, 1995.

*Conclusions*

Material development must increasingly not only fulfil functional and economic criteria, but must also meet ecological and social requirements of sustainability. This has led to a change in attitude in the sector of material sciences towards the selection of more environmentally friendly materials. The use of natural plant fibres in composites offers an interesting alternative to industrially created products in this context. The influence of fibre properties on the behaviour of composites is manifold and was shown for the properties of fineness, length, orientation and force-elongation characteristics with various examples and our own results. From this, procedures may be derived which may simplify the creation of material properties. For the determination of tensile properties of individual fibres and of fibre bundles, reproducible and reliable methods are required that allow the determination of fibre properties which can also be used for material models for structural simulations.

With the system presented from the company DIA-STRON, reproducible fibre properties may be determined, which hitherto were determined in time consuming and error-prone ways. The influence of fibre-fineness must particularly be taken into account with natural fibres in a bundle structure. The fibre bundles may be divided into finer bundles or individual fibres with the aid of processing techniques and thus the properties of composites may be improved. For the determination of the width distribution of the fibres or fibre bundles a measurement system was introduced, with which a simple, reliable determination could be carried out. In addition to the large measuring capacity for very different types of fibres the measurement of fibre orientation is also possible. If natural fibre composites can be established as calculable materials with reliable measurement values, the products made from these materials will achieve a broader market introduction and will become an alternative on a lasting basis.

## 10.4 Manufacturing

### 10.4.1 Introduction

At the present time and as stated previously, practically all bast/leaf/fibre composites are manufactured using press-moulded technology, thermoplastic or thermoset. In 2000, these two types of manufacture had approximately equal shares of the market but there is a clearly identifiable trend toward thermoplastic matrix systems using, for example, polypropylene and away from thermoset, usually using polyurethane (see Fig. 10.12). By 2003 only about 34% of natural fibre composites were manufactured via the thermoset route.

The reasons for this change lie in the easier processing and recycling possibilities of thermoplastics as well as certain ‘fogging’ problems that sometimes arise when using duroplastic matrix systems. However, one should

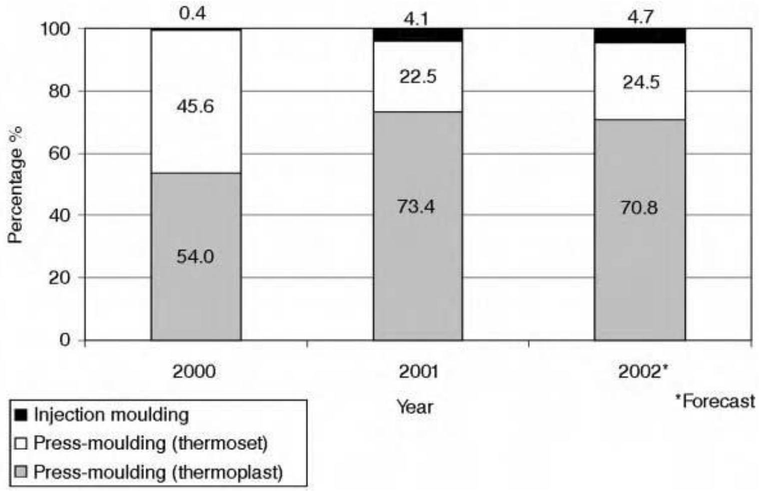

*Figure 10.12* Percentage proportions of various processing techniques of natural fibre composites: 2000–2002. Source: Kaup *et al.*, 2003.

note that the thermoplastic processing of natural fibres produces more odours than thermoset (duroplastic) processing. This trend away from thermoset processing is expected to continue.

It is also interesting to note that 2003 saw the beginning of the production of injection moulded vegetable fibre panels. In general, injection moulding is the preferred route to manufacturing the more complicated shaped panels that are difficult, if not impossible, to make by using the press-moulded, methods; composite panel manufacturers expect that injection moulding will become increasingly important.

## 10.4.2 Thermoplastic manufacturing

In this process either of two methods of blending the vegetable fibres with polypropylene (PP) fibres are used. The fibres are first cut to lengths of between 80 and 120 mm and then either the fibres are blended as uniformly as is practicable before being carded and made into a felt by needle punching, or separate vegetable fibre and PP needle felts are made. These are then built up into as many layers as necessary and are then placed in the moulding presses under heat to achieve the size and shape panel required. So as to improve the PP-natural fibre bonding a small amount of a compatibiliser is sometimes used as an interface between the fibre and the matrix. This is often MAPP, a malic anhydride modification of PP.[4]

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[4] Nabi Saheb and Jog (1999).

### 10.4.3 Thermoset manufacturing

The vegetable fibre needle felts are sprayed with or soaked in synthetic binders such as epoxy resins or polyurethane and then moulded to the desired shape. Concerning the fibres used, it has been found helpful to blend the finer vegetable fibres (flax, jute) with a proportion of the coarser fibres (hemp, sisal, kenaf) as the finer fibres impart stability to the blend but may prevent their complete permeation by the binder if used on their own.

### 10.4.4 General

It is important, whether thermoplastic or thermoset technology is used, that the fibres used be reasonably clean. If not, the adherence of the fibres to each other and to the plastic component of the composite may be diminished, with the consequent possibility of rupture during use. Also, after lamination, the shives appear as blemishes on the surface of the panel. For flax tow, for example, a maximum of 2% impurities is allowed by the composite manufacturers.

In both thermoplastic and thermoset manufacture approximately equal weights of reinforcing fibres and PP or binders are used to create the textile semi-products before thermo-processing. As the vegetable fibres have lower densities than glass fibre the final composites are also lighter, by between 10% and 30%, than those made with glass or wood fibre, or than ABS panels. It should, however, be noted that these vegetable fibres must not be considered, despite their price advantages, as being capable of replacing glass fibre in all composites because of the very different mechanical characteristics of the fibres (see Table 1.1 of the Appendix to Chapter 1). Nonetheless, according to Weigel *et al.* (2002), there is, at present, intensive research into the possibility of replacing some glass fibre used in composites by fibres based on renewable resources.

## 10.5 The future, trends and conclusion

From the information set out in section 10.2 of this chapter we can summarise the present (late 2004) situation as follows:

1. European consumption of natural fibres in composite products for the automobile industry in 2003 was approximately 26,000 tonnes and is expected to grow at around 10% per year, which by 2010 would come to about 50,000 tonnes. Mr Gordon Mackie[5] estimates world consumption at between 110,000 and 120,000 tonnes (Fig. 10.13) but this is not just for

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[5] Mr Gordon Mackie, Textile Consultant, 228 Ballylesson Rd, Drumbo, Lisburn BT27 5TS, UK. Private communication.

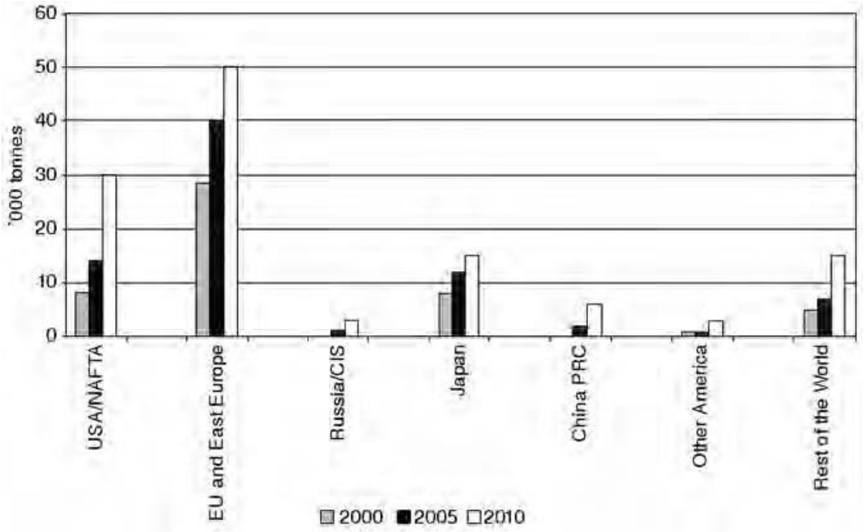

*Figure 10.13* Estimated natural fibre use in reinforced plastics. Source: Gordon Mackie.

automobile panels and includes penetration of the market for composites in other industries held by glass at the moment (Fig. 10.14).

2. When used in cars and light commercial vehicles and using present technology between 5 kg and 10 kg of natural fibres are used per car in Europe.
3. 58,000,000 cars were manufactured in the world in 2002 and this figure is expected to increase to 69,000,000 in 2008, an increase of about 20%.[6]
4. If the fibres are used in simple door panels, vegetable fibre composite panels are more expensive than those made entirely from plastic or reinforced with wood or reclaimed vegetable fibres.

It would be reasonable, therefore, to suggest that the entire market of over 50,000,000 vehicles is not open to vegetable fibre composites at the present time and that they will be used only in the interior trim of top and middle range cars where the door panels are more complex and laminated. Then the 'one-shot' press moulding technique is price competitive. In lower range cars vegetable composites are too expensive because they utilise unlaminated, simple door panels.

We should also take into account, again at present, that only small quantities of vegetable fibres are used in the manufacture of injection moulded composite panels and that this market is still the preserve of glass fibre. It would seem that the factors preventing greater use of vegetable fibres are that certain technical

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[6] Society of Motor Car Manufacturers and Traders, London. Private communication.

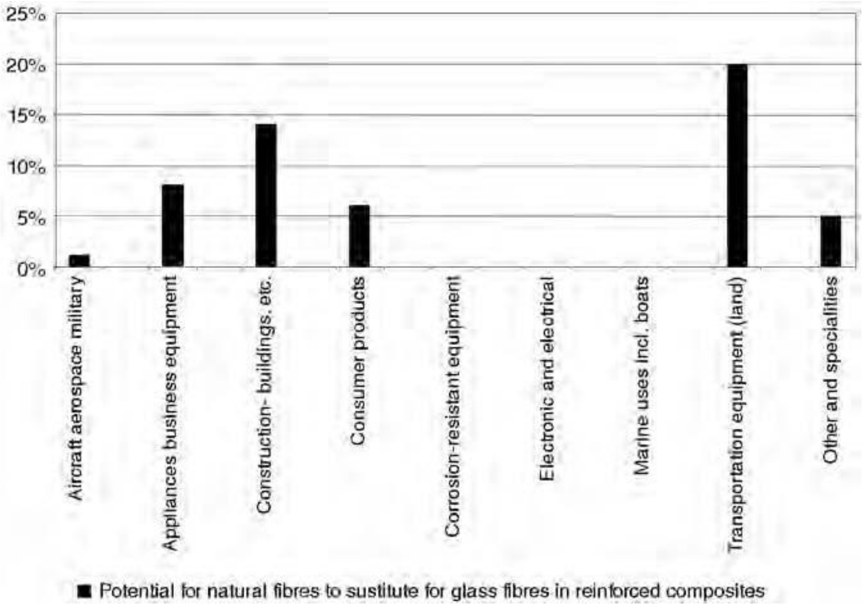

*Figure 10.14* An estimation of substitution/penetration for natural fibres to replace glass fibres in reinforced composites by 2010. Source: Gordon Mackie.

problems still need to be resolved and also that investment in new or modified manufacturing plant will be necessary. It is expected that over the coming few years these technical obstacles will be overcome and that the investment will take place (see Fig. 10.15).

At this point in time the future of bast and leaf fibres in this market looks very encouraging; however, experience shows that just extrapolating into the future the growth rate that a new technical development achieved in its early years can be very misleading. It is therefore appropriate to be cautious when assessing the situation but it would seem reasonable to think that the consumption of vegetable fibres in automobile composites in 2010 would be within the following brackets: for press moulded panels, 40,000–100,000 tonnes; for injection moulded panels, 20,000–50,000 tonnes; total, 60,000–150,000 tonnes. It is, however, difficult to estimate the share taken by individual fibres, but the following market shares, based on present experience, would seem to be reasonable: flax 45%; hemp 15%; 'exotic' 40%.

Looked at from a global point of view, consumption in the Americas and Asia should be added to these quantities. In 2000, Kline & Company (www.klinegroup.com) published the study 'The outlook for natural-fibre composites 2000–2005'. The main conclusion was that the growth in use of natural-fibre plastic composites (including wood fibre applications) had almost reached the ambitious five-year annual rate of 60%.

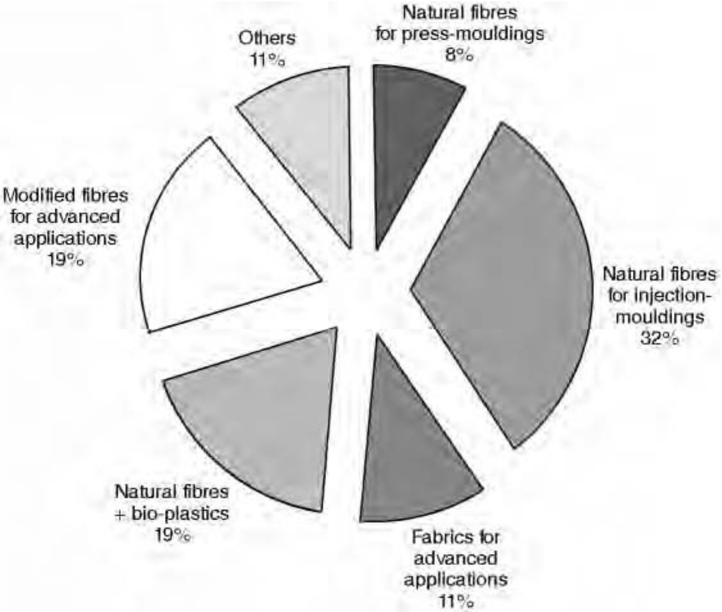

*Figure 10.15* Future trends 2005: which natural fibre technologies for composites will gain in significance? Source: Kaup *et al.*, 2003.

The situation in Japan is also developing rapidly.[7] The present fibre raw materials for car trim are low quality cotton waste and rags, priced at not higher than US$400 per tonne. Various reasonably low cost fibre replacements are available locally, rice straw and Chinese flax tow, for example, but these are both too weak to be technically acceptable. However, as in the rest of the world, considerable research and development effort is taking place, mostly based on jute and kenaf which are available in quantity from several not-too-distant countries. The market has developed to the extent that 4,500 tonnes of these fibres were imported into Japan for the manufacture of composites in 2002 and it expected that the use in Asia generally of bast fibre composite materials for vehicle interior trim will continue to evolve as it is doing in Europe and North America.

A further point should be made concerning the productive potential of China concerning the supply of jute and kenaf, and possibly other fibres for this rapidly developing sector. Chinese agriculture is in a position to supply large volumes of jute and kenaf fibres (FAO, 2001). The decline in plant cultivation since 1985, due to the decrease in demand for ‘gunny bags’ used for the transportation of agricultural products has resulted in fibre processing capacities being unused and available. Delivery reliability, which is so important for the automotive industry is thus assured.

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[7] Y. Akai, Akai Shoton, Kyoto, Japan. Private communication, 2004.

There is a particular need for research in order to determine, implement and supply on an industrial level, fibres with special properties that may be used in the sector of composite materials. However, fibre processing, which at present is strongly geared to the production of yarn, would have to be expanded in the direction of the production of non-wovens. Also Chinese agriculture will become a reliable partner of the automotive industry on a sustainable basis only if quality management starts with the cultivation of the crops.

In the manufacturing of composites from 100% renewable resources, not only natural fibres but also thermosets on the basis of plant oils may be the target of the future. Linseed oil and soy-oil for instance represent interesting raw materials. By the synthesis of plant oils, thermoset systems may be produced that show properties comparable to conventional resin systems. The work of Schönfeld (2000) and Williams and Wool (2000) should be mentioned in this context. With the combination of agriculturally produced plant oils and natural fibres the automotive industry could develop exemplary lightweight constructions on the one hand and on the other, the agricultural structures of China could be strengthened on a sustainable basis.

## 10.6 Acknowledgements

The nova-Institut reports are listed in the references at the end of the chapter.

We are also indebted to Mr Gordon Mackie for his permission to include his graphs on the present and estimated future development of natural fibres in this market.

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