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BIOMIMETIC INTEREST AND POSSIBILITIES FOR REPLACEMENT OF GLASS FIBRES WITH PLANT FIBRES IN COMPOSITE MATERIALS: THE CASE OF IMPACT DAMAGE

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Abstract

In this paper, the sense and importance of a biomimetic approach to materials is discussed, highlighting the most characteristic aspects of this philosophy. In particular, attention is brought then on the possibility for improvement of plant fibres for composite materials reinforcement with the aim of looking at them as a substitute for E-glass fibres, presenting benefits in terms of environmental sustainability and end-of-life scenarios. More specifically, the issue of impact resistance, critical especially in some applications (e.g., production automotive components) is dealt with in the case of jute and flax fibres. The study suggests that, although plant fibres show promising possibilities, allowing a considerable weight reduction to be achieved with some large reduction in impact properties, further work is needed to improve fibre properties via biological treatment.

1. Biomimetics approach to materials

Biomimetics, also known as biomimicry, can be defined as "the abstraction of good design from nature" [1]. Rather than a field of science in itself, it can be intended as a philosophy or a method of inquiry, based on the observation of nature. In slightly more formal terms, biomimetics is "a strategic tool designed for creating an advanced and practical technology or materials of which clues can be obtained from actual biological structures and functions" [2].

The above considerations involve two main issues: how the ideas can be found in nature, and how these are possibly applied to materials engineering.

As Janine Benyus suggests, to be inspired by nature, four successive steps have to be undertaken:

- Quieting (immerse ourselves in nature)
- Listening (interview the flora and fauna of our own planet)
- Echoing (encouraging biologists and engineers to collaborate, using nature as model and measure)
- Stewarding (preserving life's diversity and genius) [3].

From the above considerations, a multidisciplinary approach is increasingly being recognised as necessary to develop *biomimetic materials design*. This can be explained both as the design of new materials using biological tissues, with higher environmental friendliness and a life cycle (from manufacturing to disposal) resulting

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in less resource depletion for the Earth, and as tailoring materials for functions successfully addressed during evolution e.g., "smart packaging" in eggs or shells or "self cleaning surfaces" as in lotus (Nelumbo Nucifera) leaves [4]. This quite obviously places biomimetics at a boundary between biology and engineering, involving also physics, chemistry and materials science concepts. The need to provide students with a broader vision of materials design is increasingly being recognised for teaching of materials engineering in the universities. In the United Kingdom, specific undergraduate courses in biomimetics are offered in Bath and Exeter, while biomimetics concepts, including an introduction to the knowledge of biological systems, are presented in a number of engineering courses in other universities, including e.g., Reading and Plymouth.

In addition, a number of research networks have been established focusing on biomimetics, through the Sustainable Technologies Initiative (STI) of Engineering and Physics Research Council (EPSRC). A specific initiative on natural fibres development as biomimetic materials is SusCompNet, a network open to all parties from all parts of the natural fibres supply chain, including agriculturists, industrialists and academics, with a particular emphasis on manufacturers, designers, specifiers and end-users [5].

In practice, mimicking a natural structure can lead to a number of approaches, from taking inspiration from the simple geometry of a natural object to copying its function. These approaches can be complementary, in the sense that the former is "designed" in nature to fit the latter. In a more complex way, the inspiration from the natural structure can be obtained at different levels, namely super-molecular, molecular and sub-molecular level. Ideas for new materials originating from nature developed in recent years include e.g., lotus leaves for the development of self-cleaning surfaces [6], abalone shells [7] for the development of new ceramics, insect cuticles for new composite structures [8], geckoes [9] and mussels [10] to obtain long duration-highly adaptable adhesives. A part of this research involves the development of new fibres, suitable for the application in biodegradable composite materials, such as artificial production of spider silk (biosilk) [11] and the use of bird feathers [12].

In this field, a possibility would be the extraction and suitable e.g., enzymatic or chemico-physical, modification of plant fibres to serve as reinforcement in advanced composite materials. In the following section of this paper, the opportunities and limitations of this approach on natural fibres are discussed, focusing on their possibilities with respect to impact resistance in automotive components, often referred to as *crashworthiness*. Figure 1, showing the natural fibre composites system, is aimed at describing the complexity of decisions necessary in fibre selection for use.

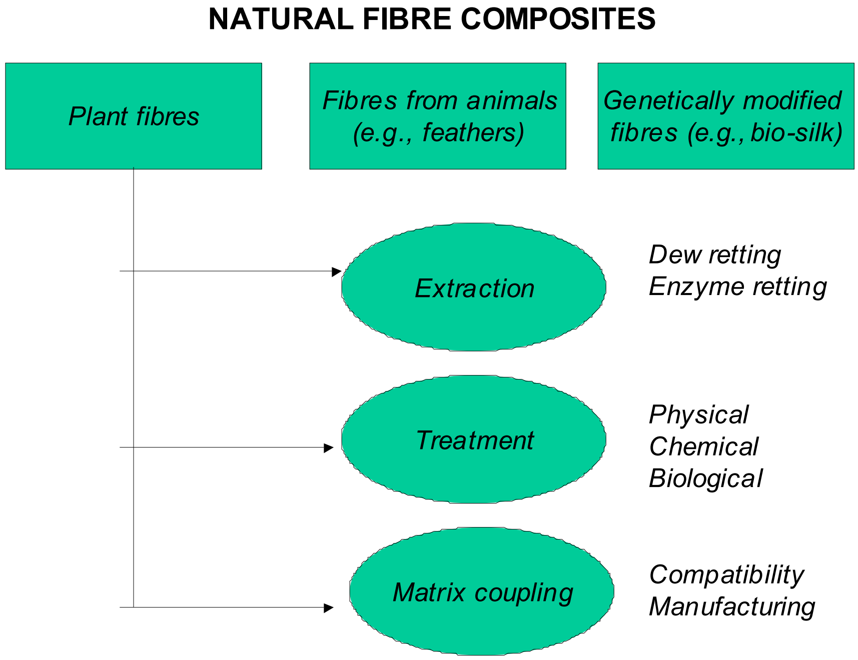

Figure 1 Natural fibre composites system of interest

2. Natural fibres as a biomimetic material

Biological tissues differ especially from man-made materials, in that the former present a hierarchical structure (as an example, the hierarchy present in muscle is shown in Figure 2). This can be explained by thinking of a material assembled with structural elements that have themselves a structure [13]. Designing using a material with hierarchical structure allows less material to be used to achieve a desired strength, although costs associated with fabrication and maintenance can be higher.

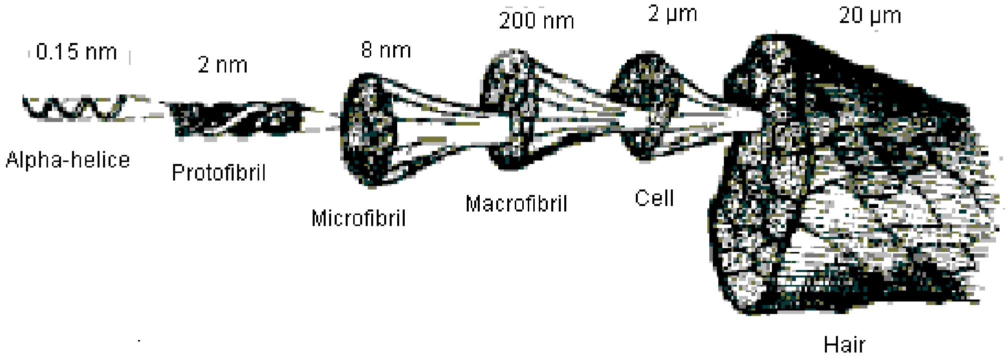

Figure 2 Hierarchical levels of structure for a human hair

When environmental constraints are involved, as is increasingly the case in recent years, biological fibres, including plant-extracted ones, can increasingly come to use, as a replacement for other man-made materials, such as polymer composites. The future of biological fibres would be affected by four factors: *competition of resources* (need for more sensible land use), *inventory, resource selection and sustainability, recyclability and environmental issues, and durability and performance* [14]. In practice, this replacement has environmental interest, in that the end-of-use scenario is more positive than for commonly used plastics. Moreover, it might also present some “added value”, because most natural fibres of interest are cropped in Third World countries, where local expertise exists in manufacturing products with natural fibres (e.g., woven mats, bags, toys, etc.). Also some industrial sectors look with renewed interest to plant fibres to replace glass fibres as reinforcement e.g., in automotive panels and components [15]. In this field, an "easy to disassemble" car is perceived as an essential development during next decade or so, with the idea of feeding back biopolymer/biocomposites components to the materials cycle. However, impact properties of these new conception components need still to be verified in full, although a small number of studies exist [16-18]. In addition, a number of fibres are potentially available for the reinforcement of composites, and this puts a larger emphasis on the creation of a database of impact and fatigue properties: some of the most diffused plant fibres are listed in Table 1.

Plant	**Botanic name**	**Fibres extracted from...**
Jute	Corchorus sp.	Stem
Cotton lint	Gossypium sp.	Seed hair
Flax	Linum usitatissimum	Stem
Hemp	Cannabis sativa	Stem
Bamboo	various species	Stem
Sisal	Agave sisalana	Leaf
Abaca	Musa textiles	Leaf
Coir	Cocos nucifera	Fruit
Ramie	Boehmeria nivea	Stem
Esparto	Lygeum spartum Stipa tenacissima	Stem
Kenaf	Hibiscus cannabinus	Stem
Roselle	Hibiscus sabdariffa	Stem
Banana	Musa sapientum	Leaf
Date palm	Phoenix dactylifora	Leaf
Oli palm	Elaeis guineensis	Fruit
Sunn hemp	Crorolaria juncea	Stem
Kapok	Ceiba pentandra Ceiba occidentalis	Fruit hair

Table 1 Most diffused plant fibres

A limitation envisaged for impact properties are particularly due to damage introduced through retting i.e., plant extraction and breaking up in a decorticator of the fibres. Moreover, plant fibre mats are usually needle-mats, with severely limited impact performance due to poor fibre orientation control and to the presence of a large number of through thickness fibres, the latter being a factor of improvement for impact properties only in small percents (1-2%) and in presence of optimal moulding conditions and accurate fibre orientation control.

A possibility in this regard would be producing hybrids with plant fibre composites used as core interposed between two E-glass fibre composites skins. This would considerably reduce the weight of the composite hopefully resulting in an acceptable decrease of impact properties of the laminate. Plant/glass fibre hybrids can be considerably improved, especially in terms of interlaminar properties, by the use of suitable processing methods, such as Resin Transfer Moulding (RTM) with thermoset matrices, or compression moulding with thermoplastic matrices, acting on a chopped strand mat of natural fibres. Hybrids can be seen as the way forward for the production of fully biodegradable composites, if the problems of integration of a hybrid mat into a biodegradable matrix are solved.

Biodegradable matrices most commonly used so far are originating from starch [19] or from polylactic acid [20]. Another interesting biodegradable matrix is sawdust, if conveniently treated through benzylation, so to acquire some capability of plastic deformation: this showed promising mechanical properties, when reinforced with sisal fibres. In this case, however, the possible manufacturing of mats or fabrics is still under discussion [21].

As a general observation, fibre toughness needs to be improved through different possible methods, including chemical or physical treatment or modification to retting process e.g., involving the use of enzymes.

3. Microstructural improvement of natural fibres

The load-bearing capacity of vegetal fibres in composites can also be enhanced by treating fibres surface to improve fibre/matrix bonding. A number of chemical or physical treatments have been proposed, such as NaOH bleaching [22], acetylation [23], grafting of vynilic monomers into cellulose [24], silane treatment [25] and ultraviolet radiation [26]. Some of the above treatments, especially treatment with alkali, are simple and inexpensive, although their efficacy in improving fibre stiffness and fibre/matrix bond may not result in a similar increase of the static properties of the composite [27]. An alternative method appears to be to increase the resistance of fibre/matrix interface e.g., by allowing penetration of the polymeric resin into plant wall cells, a method that demonstrated to be effective on flax fibre/epoxy resin composites [28].

However, there are currently clear limits in the use and performance of biodegradable composites. The quality of cellulosic fibre used is critical in obtaining composites with satisfactory mechanical performance. A number of factors have an effect on fibre quality and on compatibility with the matrix e.g., fibre maturity, preconditioning and degree of retting. For bast fibres such as hemp and flax, dew retting is the main process currently used for separating the fibres from other plant tissues. The plant stems are left in the field to “weather” until the fibre-matrix bond is broken down, obtaining a fibre mass, which is then broken up in a decorticator, a procedure that results in severe damage to the fibre structure. In particular, micro-compressive defects are induced, which are similar to the kinks formed in synthetic polymeric fibres [29], and reduce both the tensile modulus and the ultimate strength of the fibre [30]. Stress concentrations arise in the matrix in the vicinity of micro-compressive defects, so that they act as preferential locations for fibre fracture [31-32].

An alternative, which proved promising especially on flax fibres, is *enzyme retting* [33]. This would allow improving fibre extraction, while reducing the penalty of mechanical deterioration, limiting chemical treatments and associated costs and developing conversion methods for appropriate semi-finished products such as truly two-dimensional mats with random, bi-directional or uni-directional fibre orientation. Chemical techniques have been developed for sequential removal with minimal disruption for use in cell wall analysis [34]. Subsequent refinements allowed to narrow the classes of materials removed at each extraction stage: in particular phenolic crosslinks conferred enhanced mechanical properties and stabilising tissues against heat induced cell separation, resulting in increased strength of tissues [35].

Developments of enzymatic treatment for fibre extraction suggest that, provided enzyme concentration, temperature, pH, and duration of treatment are controlled, fibre of good quality can be obtained [36]. Their integration in a composite and their effect on impact properties has yet to be studied, although the advantages of enzymatic retting for the environment (no special disposal problems for the solution, possibility of carrying out retting as a local cottage industry thus saving transport costs) suggest the interest of this study.

4. Impact damage in plant fibres vs. E-glass fibres

This part of the paper deals specifically with impact properties of plant fibre composites. From the results obtained from a study on jute/polyester composites, limits and possibilities are highlighted of component manufacturing using plant fibre composites [37]. These are developed in a further evaluation of the properties of flax fibre reinforced composites: flax is a fibre particularly adapted to enzyme treatments, leading to an improvement of their stiffness.

Jute/polyester woven laminates, with thickness variable between 3 and 5 mm., were impacted to energies up to 20 Joules with an impact velocity of 2 m/s using a Rosand IFW5 drop-weight impact tower. The diameter of the hemispherical impactor was 12.7 mm. During dart penetration tests, the jute fibre reinforced laminates appeared slightly curved at energies not exceeding 30% of the measured penetration energy, which was around 125 kJ/m^2, suggesting the presence of internal damage, although not appearing at the surface (Figure 3) [38]. Some more evidence of damage was given by the thermoelastic stress measurement, where delamination was found to be characteristically concentrated on a line along the external boundary of the impacted area (Figure 4). This suggests that resin-rich areas and more in general the presence of manufacturing defects have an influence on impact damage propagation.

In terms of penetration energy, the values yielded for jute fibre composites were in the range of about one third of those offered by glass fibre composites with the same fibre content. However, in the case of Charpy impact tests, the difference is much larger (around 30 kJ/m^2 compared to around 200 kJ/m^2 for glass fibre composites). This was explained by the fact that the material does not undergo penetration until a wide amount of matrix cracking takes place and drives the fibre to be torn off [39], which suggests a higher impact damage tolerance and that this material has a good capacity of sustaining load after damage initiation. Moreover, impact damage characterisation on jute fibre reinforced laminates [40] pointed out that their interlaminar adhesion is sufficient to yield a conical through-the-thickness impact damage pattern typical of stronger composites, often referred to as reversed-pine tree [41] (Figure 5).

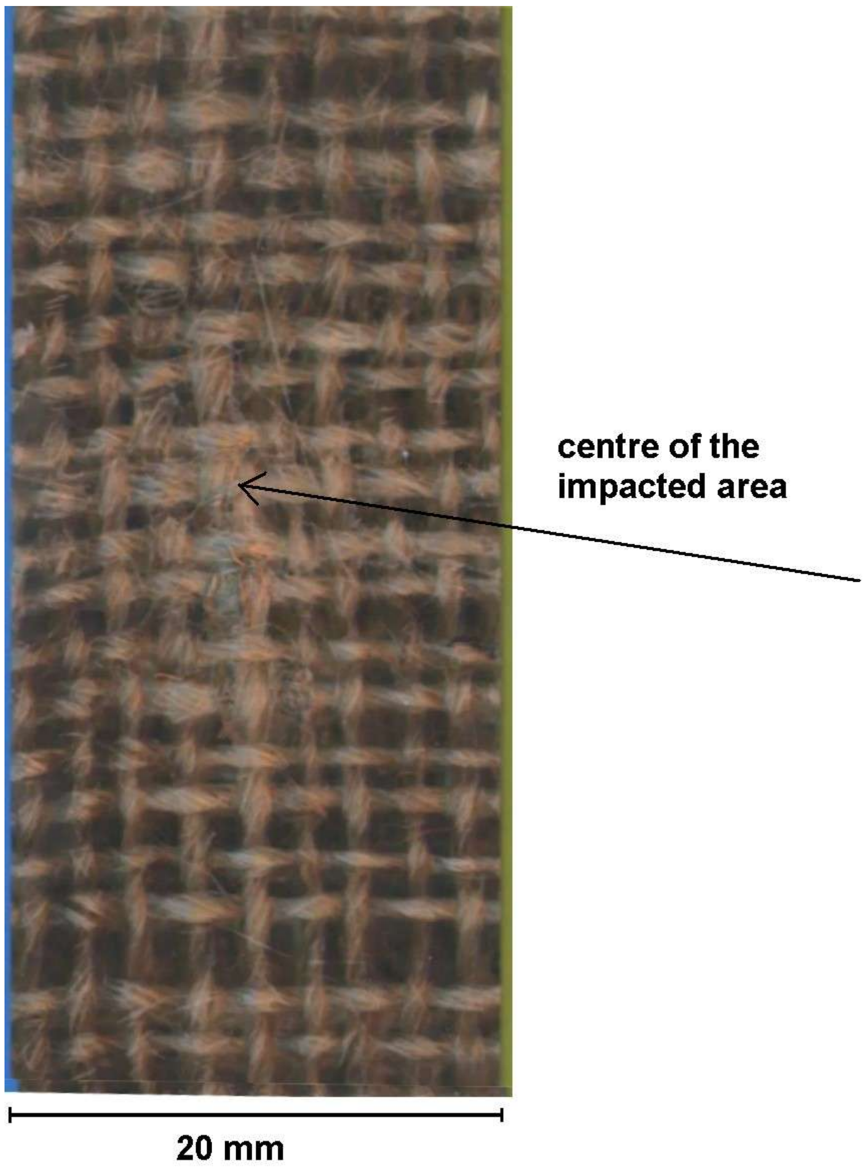

Figure 3 Impact damage on jute/polyester composites

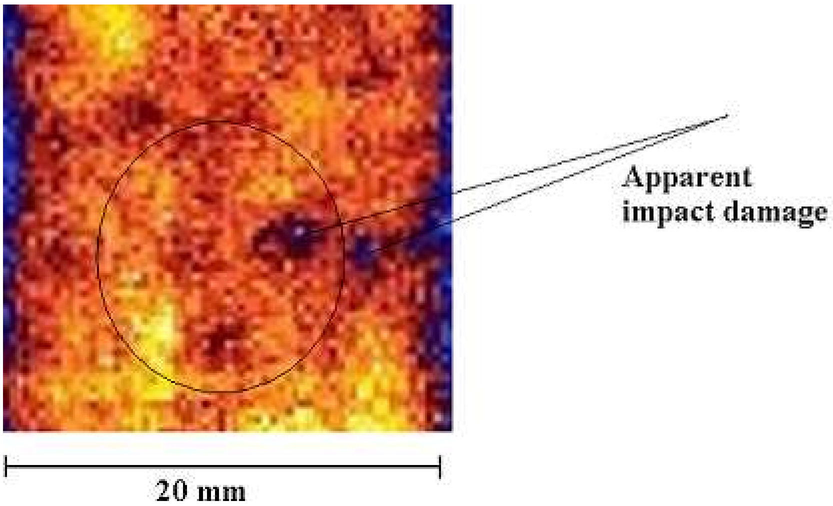

Figure 4 Thermoelastic stress analysis of impacted jute/polyester composites

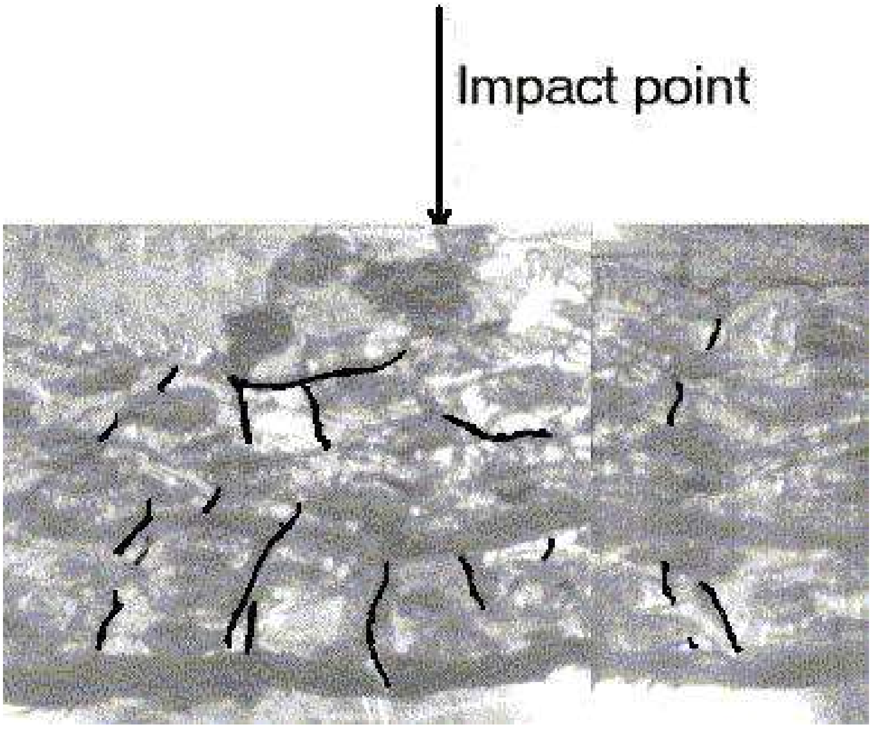

Figure 5 Conical pattern of impact damage in jute-polyester composites

To concentrate on a fibre, which appears more susceptible of improvement through enzymatic treatment, unidirectional flax/epoxy and E-glass/epoxy-flax epoxy hybrid laminates have been subsequently manufactured and tested. Different thread diameters of untreated flax fibres have been used: the one allowing the higher plant fibre content (up to 53±2% by weight) was the 0.8 mm diameter. Laminates were made by hand lay-up in closed matching moulds and tested statically and dynamically (falling weight impact) under three-point flexural loading. In addition, impact damage has been characterised under an optical microscope.

Dynamic results on flax fibre reinforced laminates show an increase of up to 20% in ultimate stress with respect to static ones, which may indicate a relatively small sensitivity of the material to high strain loading, mostly due to viscoelastic effects in the polymer matrix (compare Figure 6 with Figure 11).

Hybrids manufacturing was intended primarily to compare decrease in impact properties with weight reduction in the laminate, due to the introduction of flax fibres. This would allow designers to evaluate the increase in thickness to be applied on a component to obtain the same impact properties for different proportions of glass/flax replacement.

With this aim, hybrids were manufactured using flax/epoxy layers as core sandwiched between E-glass/epoxy skins, in different proportions. As it is shown in Figure 7, introducing some flax fibres (in proportions up to 1/3) results in a moderate reduction of impact properties, but still in a considerable weight gain. In this proportion, flax fibres in the core proved able to protect the non-impacted side from delamination up to impact energies approaching 50 Joules. However, exceeding that amount of flax fibres have more severe effect on impact strength, partially due to the manufacturing process adopted, which does not allow sufficiently high volumes of untreated flax fibres to be introduced, as is the case for E-glass fibres. Optical microscopy indicated also the strong influence of processing on impact properties, in particular since impact damage has been shown to propagate into flax/epoxy laminates mainly staring from surface and sub-surface defects (voids). In addition, also hybrids with flax/epoxy skins and E-glass/epoxy core have been produced, but the results achieved were inferior, since these hybrid laminates underwent delamination at much lower impact energies as in the inverse case.

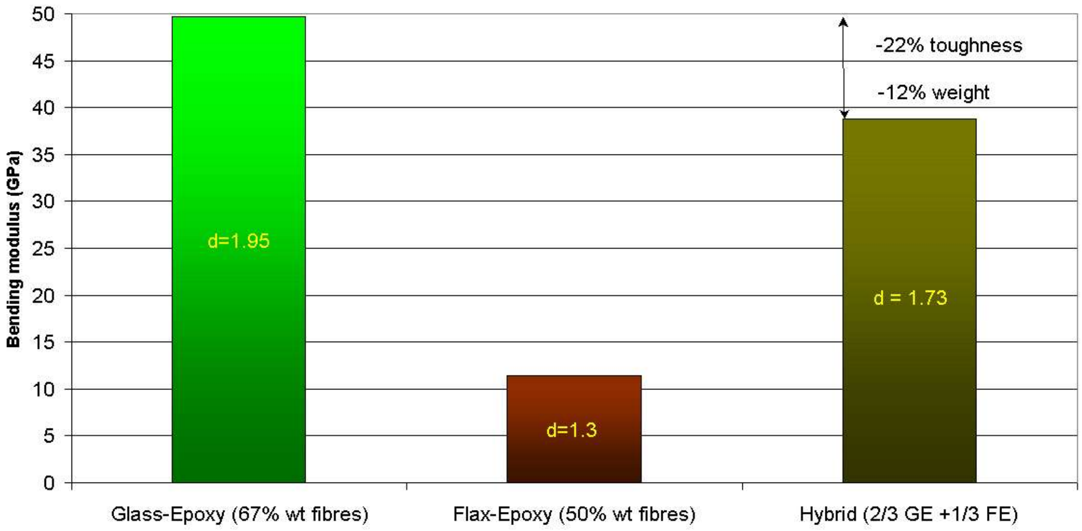

Figure 6 Bending properties of glass fibre, flax fibre composites and glass/flax fibre hybrid composites (0.9 mm. flax thread)

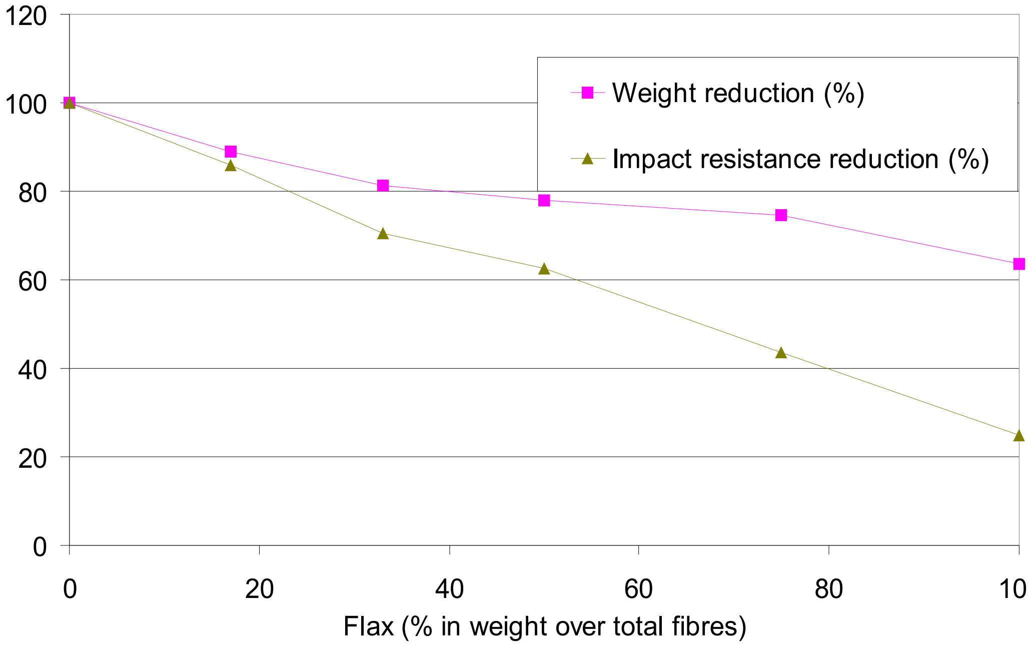

Figure 7 Impact resistance and weight reduction vs. flax content

Main problems observed in the production of flax fibre reinforced composites were the low level and non-uniformity of fibre impregnation, especially noted when using the bigger flax thread (2.3 mm diameter) (Figure 8). This might be solved using other manufacturing procedures, such as RTM (Resin Transfer Moulding). On the other side, the use of the smallest thread (0.2 mm diameter) proved not suitable for containing impact damage, since the fibres tended to be ineffective as a reinforcement and this resulted in extensive cracking of the composite for impact energies exceeding 15 Joules (Figure 9).

The use of an intermediate dimension of flax thread (0.9 mm) allowed better results to be obtained. In particular, a very promising feature concerning interlaminar strength was that in E-glass/flax hybrid composites manufactured with 0.9 mm thread shear strength was not affected by the introduction of flax fibres, until these exceeded 40% of total fibre reinforcement in the hybrid (Figure 10). A global picture of the average impact penetration energy results is given in Figure 11, where hybrids made with 2/3 glass and 1/3 flax fibres are compared to the pure E-glass and flax fibre reinforced composites. It is likely that these results can be improved by adoption of other manufacturing techniques and improvement in fibre extraction via enzyme retting. At the present stage, these composites are prone to defects, which are often initiated in coalesced voids or in resin-rich areas: an example of the latter problem is shown in Figure 12.

Figure 8 Superficial voids, indicating insufficient fibre impregnation on flax-epoxy composite (2.3 mm. thread)

Figure 9 Extensive impact cracking due to fibre tearing off on flax-epoxy composites (0.2 mm. thread)

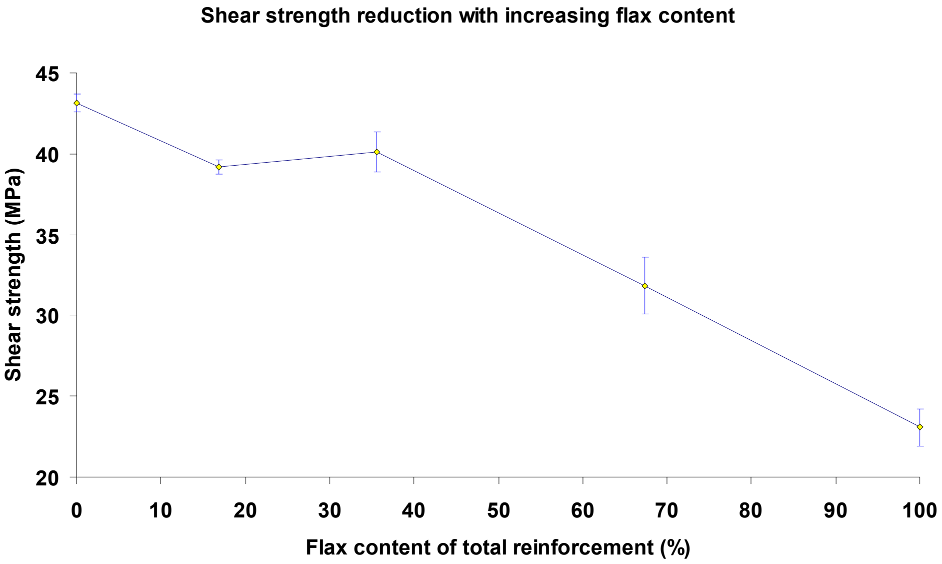

Figure 10 Shear strength vs. flax content in hybrids

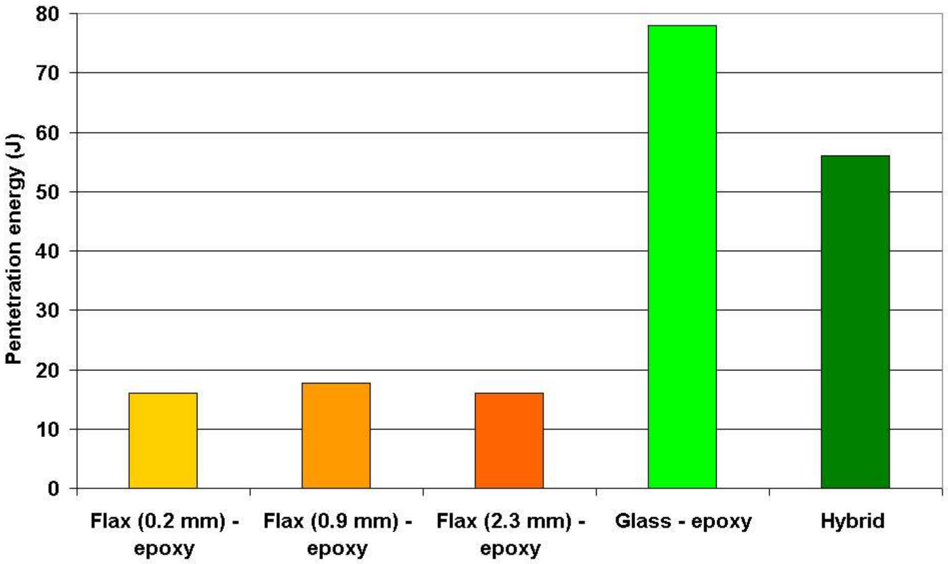

Figure 11 Average penetration energies of flax fibre, E-glass fibre reinforced composites and hybrid (2/3 E-glass + 1/3 flax) reinforced composites

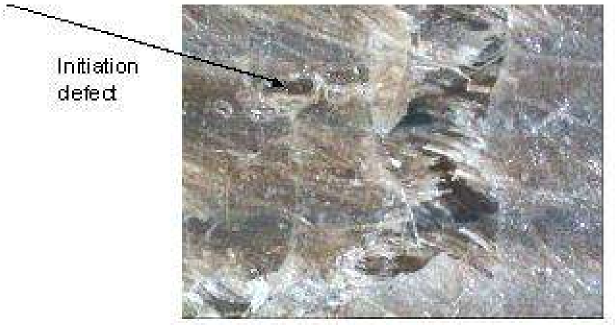

Figure 12 Initiation of impact damage from a manufacturing defect in flax-epoxy composites

5. Conclusions

Reduced weight, coupled with only a lower reduction in static and dynamic properties make plant fibres, such as flax and jute, promising substitutes for E-glass fibres. The rationale for this replacement lies in their easier recyclability and in their possible coupling with biopolymers, with the idea of producing a fully biodegradable composite.

In particular, the success of plant fibre composites for component production is likely to be eased by the possible improvements in fibre extraction, allowed by enzyme retting, and the improvement in mats fabrication, that will have a further positive effect on impact properties.

To pass to the large volume production of plant fibre reinforced components, two conditions have been perceived as necessary. These are an intermediate experimentation stage, involving coupling of different plant fibres with E-glass fibres in manufacturing hybrid laminates, and the building of a database of impact and fatigue properties, including data from the largest possible number of plant fibres/polymers combinations.

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