HIGH PERFORMANCE APPLICATIONS OF PLANT FIBRES IN AEROSPACE AND RELATED INDUSTRIES

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ABSTRACT

Originally coming from aerospace technology, fibre reinforced plastics (FRP) are successfully used for various applications, today because of their excellent specific properties, e.g. high strength and stiffness, low weight and the potential of optimisation by orientating (esp. continuous) fibres along the load paths.

In order to successfully meet the environmental problems of these classic composites, the DLR Institute of Structural Mechanics developed an innovative idea in 1989:

By embedding natural and near natural reinforcing fibres e.g. flax, hemp, ramie, cellulose etc. into a biopolymeric matrix from cellulose, starch or lactic acid derivatives etc. (thermoplastics as well as thermosets), new fibre reinforced materials, called biocomposites, were created and are still being developed. In terms of mechanical properties being comparable to glass fibre reinforced plastics (GFRP), latest developments on new fibre/matrix combinations and environmentally compatible flame retardants enable biocomposites to replace GFRP in most cases. Biocomposites are designed to meet the processing requirements for commonly used manufacturing techniques, e.g. pressing, injection moulding, filament winding, BMC, SMC etc.

Apart from anisotropic and specially tailored lightweight structural parts with continuous fibre reinforcements, biocomposites are very well suited for panelling elements in cars, railways and aeroplanes, etc. using different kinds of nonwovens from single fibres (needlefelt nonwovens, fleeces etc.) to be easily adapted to the usually curved shapes of panellings, fairings etc.

When modifying the resin systems more or less extensively, biocomposites can be designed for different applications either to be stable or biodegradable. Apart from re-use or recycling, this offers additional options of a convenient removal after the end of the lifetime, i.e. combustion of any kind of biocomposites now being carbon dioxide neutral and completely slag-free, or biodegradation or composting of the biodegradable kinds of biocomposites. Thus they are fully integrated into natural cycles and can also meet the steadily increasing environmental demands of legislative authorities.

INTRODUCTION

Aerospace technology is the original range of application for fibre reinforced polymers (FRP). In the meantime, however, these construction materials are also used for numerous technical applications, especially if high strength and stiffness at low weight is required. The good specific, i.e. weight related properties are due to the low densities of the applied matrix systems (unsaturated polyester, polyurethane, phenolic or epoxy resins) and the embedded fibres giving high strength and stiffness (glass, aramid and carbon fibres). Furthermore, during production the option of tailoring a composite part to the special demands is made use of by orientating the reinforcing fibres into the load directions. Thus, the compound, i.e. the material itself, results directly from the manufacturing of the structure. Therefore different technologies have been developed.

With the classic fibre reinforced polymers, however, there are often considerable problems with respect to re-use or recycling after the end of the life time, mainly due to the compound of miscellaneous and usually very stable fibres and matrices. A simple landfill disposal is more and more excluded when regarding the increasing environmental sensitivity. Therefore environmentally compatible alternatives are looked for and examined, e.g. recovery of raw materials, CO_2-neutral thermal utilisation, or biodegradation in certain circumstances. An interesting option may be given by construction materials from renewable resources consisting of natural fibres, embedded into so-called biopolymers as well as economically and ecologically acceptable manufacturing technologies, all of them being objects of current research works at the DLR Institute of Structural Mechanics.

NATURAL FIBRES AS REINFORCEMENTS

In a fibre reinforced polymer the fibres serve as a reinforcement and therefore have to show a high tensile strength and stiffness, whereas the tasks of the matrix are to hold the fibres together, to transmit the shear forces, and to work as a coating. The materials behaviour of usually applied matrices is characterised by a functional relationship of time and temperature, a considerably lower tensile strength and a comparatively higher elongation. Therefore, the mechanical properties of the fibres determine the stiffness and tensile strength of the composite decisively. Generally, very thin fibres showing a large surface to volume ratio are used for a good adhesion of the fibres and the matrix.

According to the orientation of the fibres, the materials behaviour of composites can be e.g. quasi-isotropic (all (short) fibres randomly orientated, no privileged direction of mechanical properties), anisotropic (all fibres orientated in one or more directions with corresponding mechanical properties), or orthotropic (fibres orientated in mainly two directions rectangular to each other showing corresponding materials behaviour).

The selection of suitable fibres is determined by the required values of stiffness and tensile strength of a composite (Moser, 1992). Further criteria for the choice of suitable reinforcing fibres are e.g.

- elongation at failure,
- thermal stability,
- adhesion of fibres and matrix,
- dynamic behaviour,
- long time behaviour,
- price and processing costs.

Natural fibres can be subdivided into vegetable, animal, and mineral fibres. Mineral fibres are no longer or only in very small amounts applied in new technical developments because of their carcinogenic effect.

All vegetable fibres (e.g. cotton, flax, hemp, jute, etc.) are built up from cellulose, whereas fibres of animal origin consist of proteins (e.g. hair, silk, wool). Vegetable fibres can be generally classified as bast, leaf, or seed-hair fibres, depending on their origin. In the plant, the bast and leaf fibres lend mechanical support to the plant’s stem or leaf, respectively, e.g. flax hemp, jute, or ramie. In contrast, seed-hair fibres, such as cotton and milkweed, are attached to the plant’s seeds and aid in wind dispersal (Wagner, 1961; Flemming et al., 1995; N.N., 1955; Koch, 1994; Haudek and Viti, 1994).

Many natural fibres have a hollow space, the so-called lumen, and in irregular distances, there are nodes dividing the fibre into individual cells. The surface of natural fibres is rough and uneven giving good adhesion to the matrix in a composite structure. As mentioned above, the specific mechanical properties of natural fibres are of great importance for their use in composites. For comparison, the breaking length or tenacity and elongation at failure of both natural and synthetic fibres are presented in figure 1, making clear that especially hemp, flax and ramie fibres can compete with some frequently used synthetic engineering fibres. The breaking length is a term for tenacity, a specific measure known from textile industry and given in meters. It marks the length of a fibre fixed at one end, at which it breaks due to its own weight.

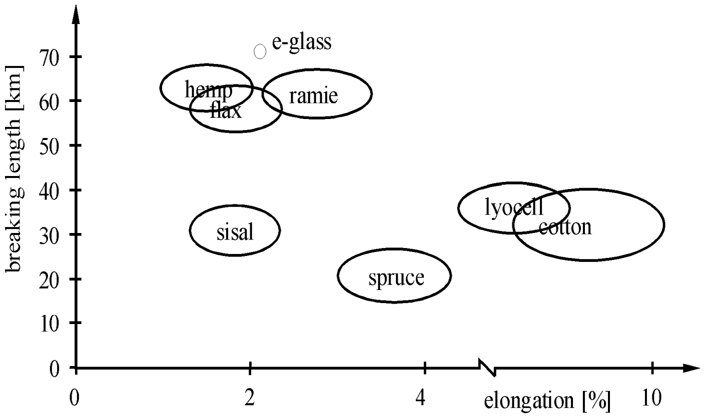

Figure 1: Properties of different natural reinforcing fibres

The diagram in Figure 1 reveals that flax, hemp, and ramie fibres show highest values of breaking lengths compared with e. g. sisal, spruce, and cotton. Due to the significantly higher elongation at failure up to 10 %, cotton fibres are unsuitable as a reinforcement in biocomposites. The corresponding values of E-glass fibres, e.g. Al-B-silicate glass (Ruge, 1989), serve as a reference because of their great importance in composite technology.

Influence of the preparation on the properties of natural fibres

The influence of the preparation on the fibre qualities is represented at the example of flax. When the flax has reached maturity, the plants are pulled and may be left on the field for the so-called retting process, a controlled decay and separation at which the cementing substances connecting the bast with other plant parts are decomposed by microbes. This process is ended after three to five weeks depending on weather conditions so that the mechanical processing can be started. Another option is to exploit green flax, where the so-called field retting is omitted, and after drying the plants are directly submitted to the mechanical preparation termed flax scutching. In spite of the biological and mechanical preparation, dust and other impurities e.g. pectin etc. still stick to the fibres. These accompanying substances, however, can be removed by washing in a special procedure.

Biocomposites of BIOCETA and differently treated flax fibres, i.e. green flax, retted flax, and flax from which the pectin had been removed, were made by pressing and tested in order to evaluate the influence of the fibre preparation on the composite properties.

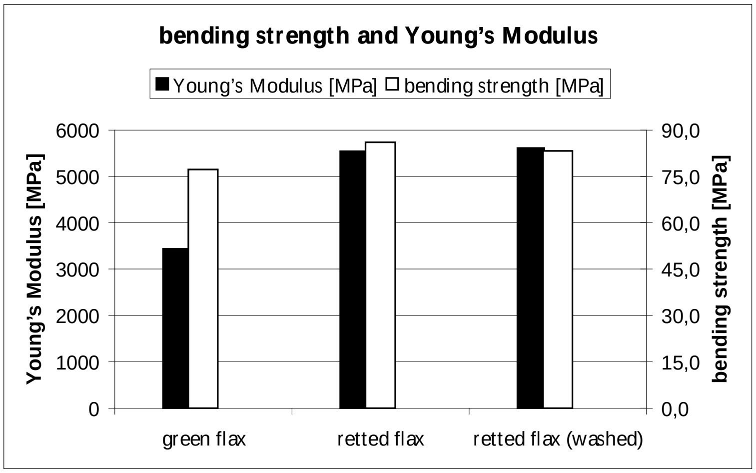

Figure 2: Mechanical properties from bending tests of biocomposites made from differently prepared flax fibres

As shown in Figure 2, needlefelt nonwovens from retted flax generally give a better reinforcement than fleeces from green flax (different states of retting were not examined), since a retting process leads to finer fibres and therefore to a larger surface (= contact surface) and a better adhesion of fibres and matrices. Removing the pectin (watery cleaning) improves the reinforcing effect only insignificantly.

BIOPOLYMERS AS MATRIX SYSTEMS

The already described natural fibres are embedded in a biopolymeric matrix system, the task of which is to hold the fibres together, thus giving and stabilising the shape of the composite structure, to transmit the shear forces between the mechanically high quality fibres, and to protect them against radiation and aggressive media. Usually, polymers are subdivided into thermosets and thermoplastics, both of them suitable as matrix systems for construction materials from biocomposites.

In the following, polymers with the basic parts predominantly consisting of renewable resources are termed biopolymers. In addition, the basic parts can be formed either by the main chain, or by the side chain(s) or even by monomers as basic elements of a polymer. Numerous variations for optional structures of biopolymers result from this fact.

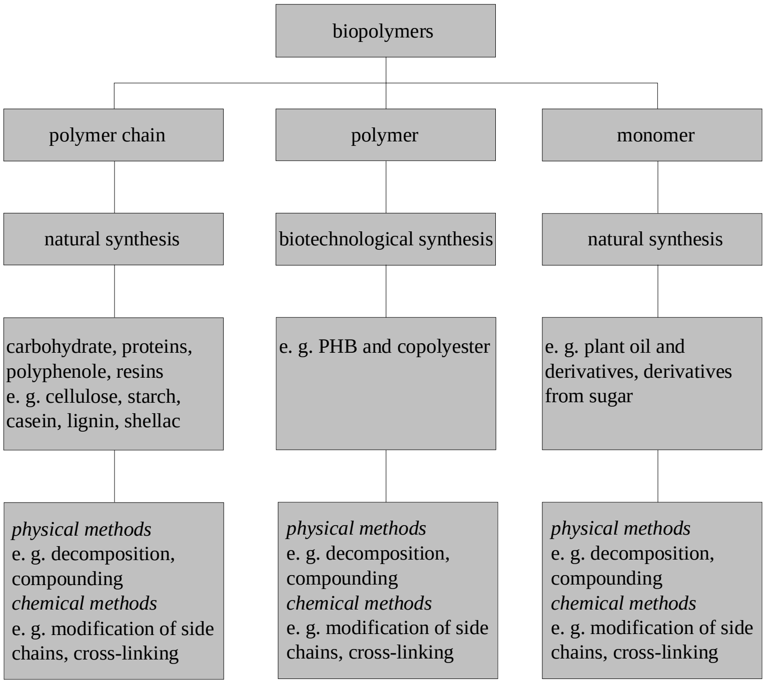

Figure 3: Classification of biopolymers

Polymers of natural origin, e.g. starch and cellulose, have to be modified physically or chemically to be suitable for the processing as thermoplastic resins, e.g. the structure of starch can be made thermoplastic using adjuvants, e.g. glycerol and water (Fritz, 1997). A frequently used option to improve the properties is to add copolymers which can even be of petrochemical origin (MATER-BI is one of the products) (Bastioli, 1998). But also by partial or complete esterification of the hydroxyl groups in the side chains with short-chained organic acids, e.g. acetic acid, and perhaps by adding plasticizers, this effect can be obtained (e.g. the product SCONACELL A) (Rapthel and Kakuschke 1997). In addition, other physical, chemical, mechanical and thermal properties of the biopolymers are influenced as well by these modifications. The esterification of the hydroxyl groups at the side chain is preferred for making cellulose a thermoplastic material while keeping its cellulose chain structure (e.g. the product BIOCETA) (N.N., 1997; Kuhne, 1998).

Applying the methods of biotechnological synthesis, in many cases by fermentation, polymers in particular built up by micro-organisms are obtained, using such polymers as a storage for energy. Compared with plants, here the task of storing energy is done by the starch. Polyhydroxybutyric acid (PHB) and its copolyesters are specified here as the most important example for fermented biopolymers (e.g. BIOPOL) (Schack, 1998).

In general, polymers are made by chemical synthesis from mainly small components, the so-called monomers. Such monomers are either synthetisized in a completely natural manner, e.g. lactic acid, or only slightly modified in their chemical structures, e.g. various epoxidised sunflower, rape, or soybean oils.

Up to now the last mentioned basic components are still cross-linked with hardeners from petrochemical origin (e.g. ELASTOFLEX) (Scherzer, 1997). But also the other natural raw materials, like cellulose, shellac, and lignin etc. which offer corresponding functionalities can be cross-linked, and further raw materials can be added.

There are various options to modify the available matrices (Klein et al., 1997; Herrmann, Nickel and Riedel, 1998), thus, the material selection has to be adapted to the given requirements. Criteria for selecting a suitable matrix system for high performance construction materials are the temperature in use, mechanical loading, manufacturing technology, etc. An important demand for the matrix is also an adequately low viscosity for a good impregnation of the reinforcing fibres. Additional basic qualities, e.g. the elongation at failure, which should match the corresponding values of natural fibres, and a good adhesion to the natural fibres must be given, too. Apart from further matrix qualities, the above-mentioned criteria are essential for optimum fibre reinforced composites.

At the DLR-Institute of Structural Mechanics, the available biopolymers are tested with respect to their suitability as matrices for biocomposites. Apart from the performance of various biopolymers, their potential to be applied with new manufacturing techniques is being examined (e.g. new resin injection procedures, i.e. the differential pressure resin transfer moulding-(DP-RTM)-technology, which was developed at the DLR-Institute of Structural Mechanics).

Technological requirements for biopolymers

Frequently, polymers from renewable resources do not sufficiently fulfil the requirements to be used as matrices in biocomposites. This deficit is based on the historical development, since these polymers have originally been designed for the packaging sector. In particular, they show either too high values of elongation at failure, or their rheological behaviour is a strong restriction for the application in biocomposites.

An essential requirement for a good fibre matrix adhesion is an optimised impregnation of the reinforcing system. In order to evaluate the degree or quality of impregnation of composite plates, the well-established ultrasonic test method is used. As the specimens are exposed to water during testing, and some biopolymers are critical to moisture, this may cause a greater rate of swelling compared with petrochemical matrix systems of classic composites.

In case of a petrochemical polymer combined with flax fibre nonwovens the limiting value of viscosity was found to be 100 Pas, i.e. the viscosity should not exceed this value for a good impregnation of the selected reinforcing material when processed in the film-stacking technology.

The manufacture of structural parts can more easily be realised when using the so-called comingled technology, since apart from the technological advantages because of fewer steps to make semiproducts, there are minor requirements for the matrix, too. It is common knowledge in composite technology that the viscosity of polypropylene at processing temperature should not exceed 1000 Pas for being combined with flax fibre nonwovens. The film-stacking procedure is especially of great importance for those polymers coming more frequently as films, but not as fibres. Knowing that the viscosity of a matrix should not exceed a limiting value of

1000 Pas (co-mingled procedure) or 100 Pas (film-stacking procedure), depending on the technology, the viscosity of various thermoplastic biopolymers was examined and the polymers were evaluated with respect to their suitability for biocomposites.

Among the thermoplastic matrix systems especially Polylactid, the SCONACELL A types, the BIOPOL types, and HPCL II are well suited as matrix systems for biocomposites considering the viscosity at processing conditions.

The values of elongation at failure reveal HPCL II as very brittle compared with other polymers so that fibres from HPCL II could not be realised until today. When producing fibre reinforced polymers from BIOPOL, an extremely bad adhesion of fibres and matrix appeared, resulting in delaminations at a low level of loading so that BIOPOL was excluded from further tests.

Manufacture of structural components

The manufacture of composite parts by pressing shall be presented in the following. This technology is comparable to the well-known pressing process of glass mat reinforced thermoplastics (GMT). Here, thermally and mechanically pre-compacted semiproducts from the co-mingled, film-stacking procedure and powder impregnation are used, as well as prepregs from the wet impregnation (figure 4).

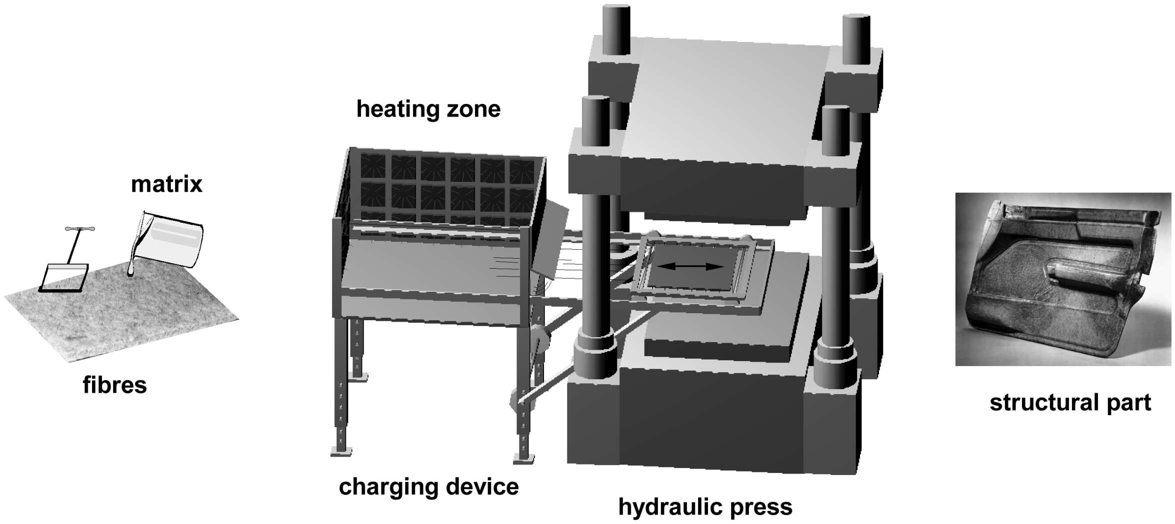

Figure 4: Thermal moulding process for the manufacture of components

Prepregs with thermoplastic matrix systems are heated and pre-compacted under slight pressure. In a corresponding mould the softened or plasticized semiproducts are pressed to a structural part which is cooled down in the mould in order to fix its shape.

On the other hand, prepregs with thermosetting plastic resins which are flexible at room temperature are cross-linked during the moulding at raised temperature. Subsequently, the component hardens during the pressing process so that no further moulding is possible.

In figure 5, selected biocomposites are compared with glass fibre reinforced polymers (GFRP), regarding the tensile properties. This comparison shows that the properties of GFRP can almost be achieved at the same fibre content. With respect to the density of natural fibres of approximately 1500 kg/m ^{3} (far below approx. 2500 kg/m^3 of the glass fibres) a higher fibre content at the same structural weight can be achieved with biocomposites, resulting in a higher reinforcement.

Based on commonly used procedures in composite technology, manufacturing techniques for the production of biocomposites are developed, analysed, and optimised. These are especially the press technique, hand lay-up, filament winding technique, and pultrusion, which are tested with only slight modifications for the manufacture of components. To take advantage of the anisotropy of the fibre reinforced composites, unidirectional (UD) fibre reinforced laminates or non crimp fabrics have to be applied. For minor requirements in terms of mechanical properties, nonwovens are used as reinforcements.

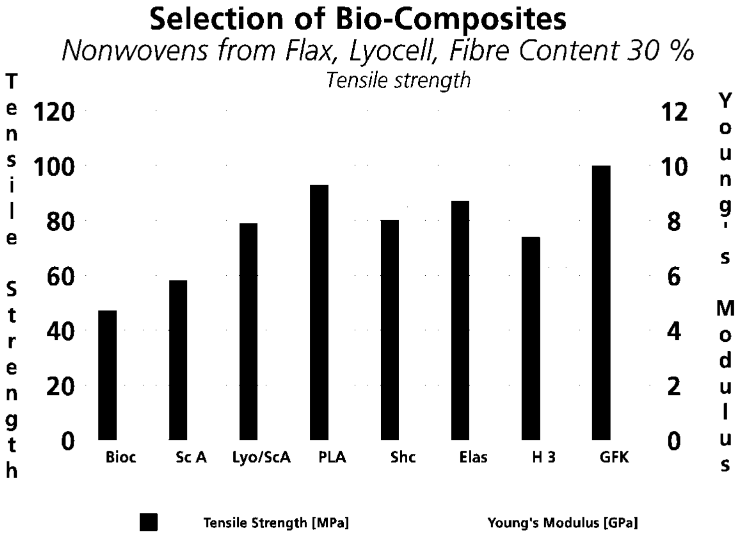

Figure 5: Tensile properties of selected biocomposites with nonwovens reinforcement from natural fibres

Figure 6 shows the results of the tensile tests of unidirectional fibre reinforced biocomposites in comparison with GFRP.

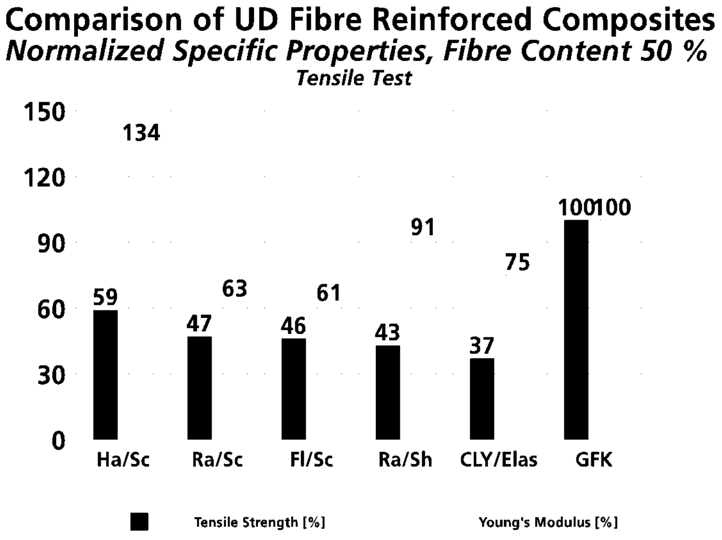

Figure 6: Tensile properties of selected unidirectional fibre reinforced biocomposites (standardised, specific data)

ANALYSIS OF BIOCOMPOSITES

Fibre-matrix adhesion

A good fibre matrix adhesion is of essential importance for optimum composite properties. A statement about this bases e.g. on the examination of the interlaminar shear strength and optical tests of the fractured surface using an electron scanning microscope.

As the examined biopolymers have a very high elongation at failure, the interlaminar shear strength (ILS) could not be proceeded by the usually applied short bending test. Therefore, the tensile shear test according to DIN 65 148 was applied. The DIN standard is valid for carbon fibre and glass fibre reinforced composites. It had to be adapted to biocomposites as the specimens failed in the area of the notches. This effect results from the fact that the shear forces exceeded the tensile strength in the residual thickness.

For the ramie yarn reinforced SCONACELL A composites the ILS was found to be 18 MPa.

As for the manufacture of yarns from natural fibres, the application of finishings avoids a fanning out of the fibres, thus, a suited, low-damaging twisting of the yarn is guaranteed, negative influence on the interlaminar shear strength is expected.

Soaps, oils, and also potato starch are frequently used as finishings for natural fibre processing, but the recipes are subject to concealment. The mentioned finishings can be removed by washing and cleaning procedures. Specimen were taken off a reel and washed with different solvents, then the surface of the yarn was analysed in ESM studies. Here, no remarkable modification of the surface of the ramie yarn could be observed.

A bleaching agent was applied as a further means for removing the finishings. In this case a bleaching and a decrease of the yarn strength from 563,99 ± 32,48 MPa down to 473,22 ± 36,67 MPa (approx. 16 %) could be seen after 10 minutes. Both phenomena point to a removal of the finishing.

Figure 7 shows the fibre, enclosed almost completely by a substance which gives a smooth surface to it. Furthermore, several small particles can be seen which either consist of the same substance or are due to impurities. In comparison, figure 8 shows no residues on the fibre.

The washed ramie yarn was applied as UD reinforcement in a SCONACELL A composite which was then tested regarding the interlaminar shear strength and analysed in ESM studies. The composite test resulted in an interlaminar shear strength of 31 MPa of the treated ramie yarn, which was 72 % higher than that of the untreated ramie yarn.

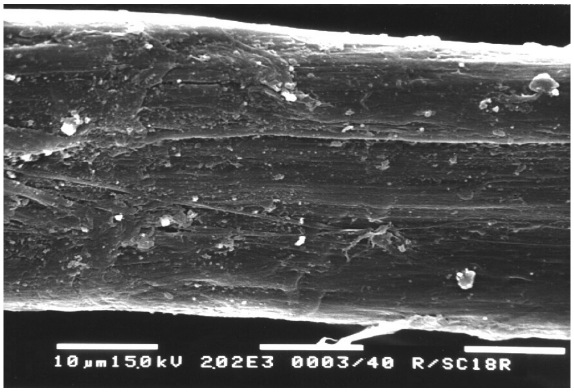

Figure 7: ESM photo of the untreated yarn

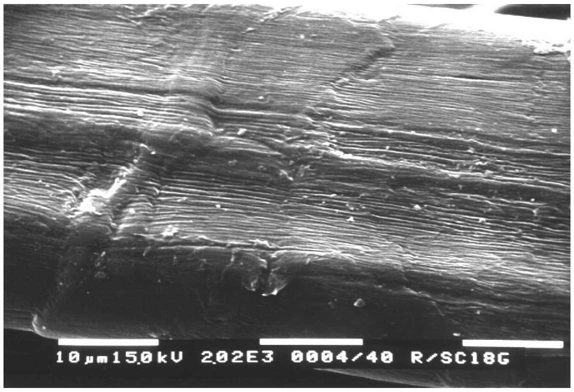

Figure 8: ESM photo of the cleaned yarn

Biocomposites with flame retardants

In contrast to the components for the interior design of automobiles successfully realised so far, the requirements concerning flame resistance in other branches of traffic are considerably higher. Therefore, studies aiming at the modification of biocomposites with flame retardants were carried out, in order to meet the railway specifications.

For the selection of flame retardants, it was difficult to keep the concept of „biocomposites“ which demands an exclusive application of renewable ressources or at least non-polluting materials. Consequently, flame retardants of mineral origin were applied, such as aluminium, magnesium hydroxide, and ammonium polyphosphate. It was pointed out that biocomposites modified in this way are well-suited to be applied for covering elements in the interior of railways. Here, the specifications S4, SR2, and ST2 were achieved, according to DIN 5510 part 2.

SUMMARY

All these studies show the excellent capability of biocomposites to be processed to structural parts. The weight-related properties also allow to aim at applications which are today dominated by glass fibre reinforced plastics. Nevertheless, there a restrictions with respect to extreme environmental conditions.

An essential branch of applications is to be seen e.g. in covering elements with structural tasks in automobile and railway design, in furniture industry, and in the field of leisure industry. Figure 9 shows a pipe and an even sandwich plate manufactured from biocomposites. These components have a high stiffness and strength, thus, they can be applied in any technological area.

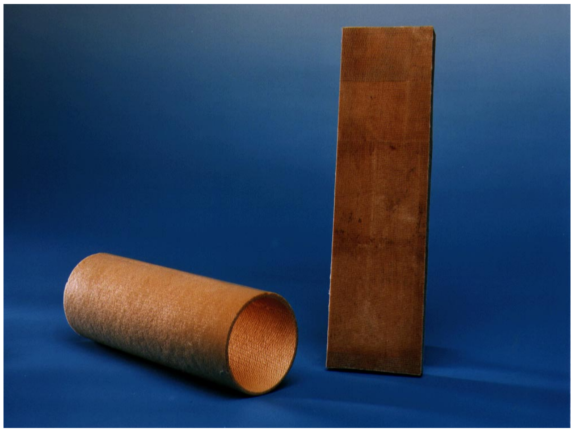

Figure 9: Pipe and sandwich plate from biocomposites, manufactured at the DLR

Figure 10: Car door interior panelling from biocomposites, manufactured at the DLR

In figure 10 a door interior panelling from biocomposites is shown which is used in automobile design (research and development project with Johnson Controls Interiors Ltd., promoted by the Lower Saxonian ministry for nutrition, agriculture, and forestry).

Furthermore, the DLR Institute of Structural Mechanics, Braunschweig, in co-operation with its partners, is able to work at further projects for industry from the idea up to maturity phase.

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