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PROCESSING, APPLICATIONS AND NEW DEVELOPMENTS OF NATURAL FIBER

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Abstract

Composite materials with thermoplastic matrices and a reinforcement of natural fibers are increasingly regarded as an alternative to glass fiber reinforced composites. The substitution of the traditionally used reinforcing glass fibers by natural fibers such as flax, kenaf or cotton can lead to a reduction of the component's weight and furthermore to a significant improvement of specific properties like impact strength, crash behavior or sound absorption. One of the major field of application for such materials can be found in structural components for the automotive industry. Product examples are door trim panels, headliners or back panels. At present the processing of such materials to structural parts usually takes place by thermal compression molding. As semi-products so-called hybrid fleeces are employed which are generated by carding or airlaid processes and subsequent mechanical bonding. The paper presents a survey and latest developments on the material, processing technologies and fields of applications. Furthermore, acoustical investigations on cotton based composite materials are presented.

Introduction

Because of excellent characteristics such as low weight or high strength and stiffness fiber reinforced composites matrices have found a large dispersal in several areas of technical applications. However, because of increasing environmental consciousness and demands of legislative authorities, the manufacture, use and removal of traditional composite structures, commonly using synthetic fiber materials such as glass, carbon or aramid, is considered more critically.

For that reason the substitution of the traditionally used synthetic fiber materials by natural fibers has become an emerging area of interest and development in polymer science and industrial applications. Despite of ecological gains like less environmental impact of the later product within the formation, usage and disposal period further technical and economical benefits result from this strategy.

The variety of natural vegetable plant fibers is abundant as shown in Fig. 1. Natural fibers can be subdivided referring to their origin. They may be extracted from plants, animals or minerals. For a reinforcement of composites, generally plant or vegetable fibers are utilized. Plant fibers can be further distinguished into hairs such as cotton, fiber-sheafs of dicotylic plants or vessel sheafs of monocotylic plants, i.e. bast fibers such as flax, jute or hemp and hard fibers such as sisal [1].

The use of natural fibers in polymer composites is growing rapidly. One of the major area of application can be found in the automotive industry. In addition to reduced weight and cost and improved safety, natural fiber reinforced polymers offer increased recycling capabilities over conventional polymers used in the automotive industry. Natural plant fibers are incorporated into door panel trims, package trays, trunk trims, and other interior parts. Referring to the polymer both thermoplastic and thermoset materials are utilized [3].

Motivation for the Utilization of Natural Fibers as Reinforcing Components in Composite Materials

Some of the major reasons for the interest in natural fibers can be found in ecological considerations as illustrated in Fig. 2. The approach of using plant fibers as a reinforcing component follows the ambition to reduce the green-house effect by establishing a nearly closed carbon dioxide cycle. Contrary to synthetic materials natural fibers offer the possibility of carbon dioxide neutrality by waste incineration with thermal dissipation. It can only be released the same amount of carbon dioxide the plants assimilated from the atmosphere during their growth period. In addition, the use of the renewable material supports the preservation of limited fossil resources and opens up new sources of income for the agribusiness. Furthermore, the energy demand required for the fabrication of a nonwoven fabric from natural fibers comprising cultivation, harvest and fiber dissociation only amounts to approximately 30 to 40 percent required for the production of a glass fiber mat [4].

Apart from these ecological considerations also several technical aspects promote the replacement of glass fibers by plant fibers. Considerably in the first place are the good mechanical properties of most plant fibers coupled with a low density. On account of their initial assignment to stabilize the stem of the plant, natural fibers offer a high mechanical potential, which, however, partly depends on the applied fiber pulping process. Most natural fibers have a hollow space, the so-called lumen (see Fig. 3) determining an up to 40 percent lower density compared to glass fibers. Therefore, plant fibers offer a high potential for an outstanding reinforcement in lightweight structures.

Automotive interiors with a reinforcement of natural fibers are considered to be physiological safer than glass fiber parts as no sharp-edged fracture surfaces occur in case of a crash. Further advantages result from their air conditioning effect and particularly their high absorbency. With respect to industrial safety, natural fibers do not cause allergic reactions or skin irritations. And finally a positive image and product marketing, related to the utilization of a renewable material, should be taken into consideration. Problems and limitations for product applications may occur from the high absorption of natural fibers for moisture as shown in Fig. 4. Furthermore, their properties decrease strongly under the influence of high temperatures.

Manufacturing Technologies for Natural Fiber Thermoplastic Composites

The processing of natural fibers into structural parts requires several sub processes which can be synthesized to a process chain as shown in Fig. 5. Due to it's specific characteristics and features each sub process has an individual impact on the fiber properties and therefore on the quality of the later composite part. To enable a successful substitution of glass fibers the previously described mechanical potential of natural fibers has to be preserved throughout the process chain and finally be implemented in the finished interior part.

The manufacture of thermoplastic structural parts reinforced by natural fibers in current industrial scales is mainly achieved by compression molding of textile nonwovens. The required semi-products are produced in a textile carding or airlaid process. At first, polymeric and natural fibers are blended to ensure a homogenous compounding. Following, this fiber blend transfers a carding or airlaid process to generate a fibrous web. Applying mechanical carding technology results in oriented fleece properties with higher strength in machine direction whereas aerodynamic carding generates semi-products with homogenous properties. Subsequent bonding usually is carried out by mechanical needling (Fig. 6). Such semi-products may also be named as hybrid fleeces and can be described by a maximum of homogeneity and easy handling.

For the manufacture of composite parts by compression molding cut nonwoven blanks are automatically taken from a magazine and traverse a preheating device (Fig. 7). Here, the prepregs are heated up to the required processing temperature. The processing temperature has to be chosen high enough to melt and plasticize the thermo-plastic binder and depends strongly on the utilized polymer. For polypropylene (PP) temperatures between 200 and 250°C are required to achieve a sufficient low viscosity. Subsequently, in a corresponding mold the plasticized prepregs are finally pressed to the final shape. Simultaneously the material is cooled down, leading to a curing and hardening of the binder component.

Properties of Natural Fiber Materials and Potential of Associated Composites

Table 1 displays major characteristics of the most important natural fiber materials in comparison to the synthetic E-glass fiber.

Even though it is apparent that the absolute mechanical data of natural fibers is inferior, the characteristic values of the specific properties of plant fibers may reach levels that are well comparable to the widely used E-glass fiber. Fig. 8 compares the specific tensile strength and stiffness of the different fiber materials. They are also known as the breaking length and breaking elongation, two specific measures frequently used in the textile industry. Evidently, especially fibers like ramie, hemp or flax are well suited to reinforce polymers from this point of view. Because of it's mechanical properties and high availability flax is the most commonly used natural fiber in technical applications in Europe.

Despite of these encouraging features of the separate natural fibers, a possible application as a reinforcing component in an actual product is determined by the mechanical data of the complete composite material. To enable the aimed substitution of synthetic fiber materials at least equal properties to the commonly used E-glass reinforced composites have to be achieved. The generation of rigid and solid composite materials requires reinforcing fibers with high strength and low elongation (Fig. 9). Referring to this approach and with regard to Table 1, flax, hemp, kenaf and ramie are expected to offer the highest reinforcing properties by displaying the highest stress and lowest strain values of all termed plant fibers.

To verify this assumption, Fig. 10 compares the tensile strength data for composite materials basing on PP and different fiber materials.

Regardless of the employed type, the incorporation of a reinforcing component results in an increase of the composite's tensile strength compared to the pure PP reference sample. Furthermore it is obvious that composite materials with a reinforcement of natural fibers offer the same dimension of tensile strength as E-glass reinforced composites. The composites basing on flax and hemp even show superior tensile strength values. Even though the fiber tensile strength values were basically comparable for all tested natural fibers with approximately 1.000 MPa (for comparison: cotton 585 MPa, Table 1), composites basing on flax fibers show higher tensile strength values than those basing on hemp or kenaf fibers. From that it can be concluded that flax offers higher reinforcing properties than hemp and kenaf. A reason for this can be seen in the fiber morphology. Flax fibers show a higher fineness and also more unique fiber diameter distribution compared to hemp or kenaf. Especially the latter usually can be characterized by wide ranging diameter distributions and comparatively large diameters. A high fiber fineness should result in better fiber embedment during compression molding and therefore higher mechanical composite properties. Fig. 10 also shows the influence of the share of natural fiber material in the composite. Obviously, a maximum in tensile strength is reached with a share of 60% of flax fibers whereas higher or lower shares result in a decrease in tensile strength.

Similar results can be attained for the stiffness of the composite materials, i.e. the modulus of elasticity in tension, as shown in Fig. 11. Again, with a range from about 45 to 50 GPa, the modulus values for the separate fibers were basically on the same level for all tested materials (cotton approximately 12 GPa).

The impact strength of composite materials reinforced by natural fibers is often considered to be inferior to a reinforcement with glass fibers. Fig. 12 displays that basing on an optimized manufacture referring to thermal processing conditions as described in [13] even the impact strength can reach values well comparable to those of glass fiber composites. For high impact properties it has to be noted, that a slightly weaker adhesion between fiber and polymer should result in a higher degradation of impact energy, supporting the so-called fiber-pull-out. Good adhesion on the contrary results in abrupt fiber fracture with a minor energy degradation.

As it also can be seen from the preceding Fig. 10 to 12, the volume portion of fibers in the composite almost needs to be doubled in order to achieve equal mechanical data when substituting glass fibers. This may be explained by the superior mechanical data of the glass fiber material as it was shown in Table 1. Other additional causes may be found in the fineness of the fiber, the wetting of the fibers with the polymer component or the processing conditions. Especially the fiber fineness has a strong impact on the composite's mechanical data. A higher fineness results in an improved fiber length to diameter relation and an increased contact surface between fiber and matrix. Furthermore it leads to an increased amount of fibers in the composite and a decrease in stress concentration at the end of the fibers and finally to an increased energy loss in combination with higher fiber-pull-out.

With regard to these aspects especially cotton fibers appear to be an interesting fiber material for certain future applications. As it can be seen from Table 1, cotton fibers show the highest fineness of all natural fibers. Even though the tensile properties are relatively low and at the same time the elongation is higher compared to other natural fibers, Muessig found remarkable high mechanical data for composites basing on an epoxy resin and a reinforcement of cotton fibers [14]. Compared to composites reinforced with different types of hemp fibers the composites basing on cotton showed a significant improvement referring to impact strength. Also the values for tensile strength and Young's modulus were higher than for the hemp fiber composite samples. However, with regard to possible future applications of cotton fibers in the area of composite materials a reduction of the actual relatively high price level would be an essential requirement.

Technical Applications of Natural Fiber Reinforced Composites

One of the major field of application of natural fiber reinforced composites can be found in the automotive industry. Amounting to a total of only 4000 to 5000 tons in 1996, the use of natural fibers in the European automotive industry has quintuplicated with more than 28.000 tons in the year 2000. In medium terms, a total use between 50.000 and 100.000 tons can be expected in this area [15, 16]. At present time, almost all European automotive producers employ interior parts reinforced by natural plant fibers. Table 2 shows the growth in the utilization of natural fibers in the automotive industry for Germany and Austria. It is evident that flax, hemp and kenaf are the most applied types of fibers.

Furthermore, approximately 50.000 to 60.000 tons of cotton shoddy and another 50.000 tons of wood fibers, mostly in combination with phenol resin, are utilized per year. Currently, an average of 5 to 10 kg of natural fibers is incorporated in every European passenger car with interiors parts such as door trim panels or trunk liner as the main fields of application (Fig. 13). Recently, also automotive exterior applications basing on natural fiber reinforced thermoplastics have successfully proved their suitability for serial utilizations. It may be assumed that such products will be employed in standard automotives shortly.

Currently, a growing number of non-automotive applications and products is being presented for natural fiber composite materials. Examples for such applications are floor coverings, employing the outstanding acoustical properties of the fibrous material, security helmets for construction areas or monitor housings for the computer industry (Fig. 15).

Current Research Work on Cotton based Composite Materials

Particularly in the automotive industry and with respect to driving comfort the acoustical properties of structural parts are considered to be increasingly important. In this context composites and layered compounds basing on natural fibers integrated in a thermoplastic polymer may represent a suitable alternative for a lot of applications. Earlier investigations have proved a strong impact of fiber fineness on the absorption capabilities of composite materials and therefore on the acoustical behavior [19]. From that point of view it appeared to be promising to analyze the acoustical properties of composite materials with a reinforcement of cotton fibers.

Applied Materials

The subsequently described investigations were carried out employing different composites with the following compounding:

- Phenol resin / cotton fibers 20/80, fleece weight 1200 g/m^2
- Phenol resin / cotton fibers 35/65, fleece weight 1200 g/m^2
- Epoxy binder / cotton fibers, fleece weight 1200 g/m^2

Phenol resin may be considered as a thermoset. It has to be pointed out that the epoxy binder contained additives of polyester, causing a thermoplastic character of the polymer.

The materials were obtained as fleeces already containing the binder component. The fleeces had a thickness of approximately 30 mm. They were processed to flat samples by means of a compression molding process. The thickness of the compressed samples was varied between 5, 10 and 20 mm.

Acoustical Investigations

Two different means of measurement were applied to investigate the absorbance characteristics of the materials. Absorption coefficients can be determined by the means of impedance tubes and the so-called Alpha cabin. Impedance tubes primarily measure material properties by perpendicular sound while the Alpha cabin determines geometrical and surface influences by applying a diffuse sound field. The schematic of an impedance tube is shown in Fig. 16. It can be used for measurements covering the frequency range from 50 Hz to 6,4 kHz. To cover this entire range different tubes with varying diameters are required. Fig. 17 presents the schematic and an inside view of an Alpha cabin. The sound field is generated by means of three acoustic sources.

The Alpha cabin can be used for test specimen ranging from 0,6 to 2,4 m^2 with a standard size of 1,2 m^2. The absorption coefficient of a standard specimen can be determined by the following equation:

$$\overline{\alpha} = 0{,}805 \left(\frac{1}{t} - \frac{1}{t^*}\right)$$

$\overline{\alpha}$ = *Absorption coefficient*
t = *Time of reverberation without specimen*
t^* = *Time of reverberation with specimen*

For specimen deviating from this standard a so-called equivalent absorption coefficient can be determined:

$$\overline{\alpha}^* = S \cdot \overline{\alpha}$$

$\overline{\alpha}$ = *Absorption coefficient*
$\overline{\alpha}^*$ = *Equivalent absorption [m^2]*
S = *Surface of specimen*

Results and Discussion

The investigations focused on a general analysis of the acoustic properties of cotton based composite materials and the impact of different binder components.

Fig. 18 shows the absorption properties of the uncompressed fleeces for a diffuse sound field.

Hybrid fleeces with a phenol resin binder show slightly higher absorption than the fleece with the epoxy binder. Comparing the two types of phenol binder fleeces with a higher share of phenol binder leads to marginal higher absorption in the higher frequency range above approximately 2 kHz.

Also after compressing the hybrid fleeces to composite samples this finding remains generally unchanged. Fig. 19 shows the absorption of compressed samples with a thickness of 10 mm.

As obvious, compression generally leads to lower absorption capabilities especially in the lower frequency range. The composite with a binder of phenol resin and a share of 35% shows the best absorption over the entire viewed frequency range. The epoxy binder based composite offers the lowest absorption. As expected the absorption is declined when increasing the degree of compression as shown in Fig. 20 for composite samples with a thickness of 5 mm.

Similar results can be obtained applying perpendicular sound by means of the impedance tube. Fig. 21 shows the absorption coefficients for composite samples with a thickness of 5 mm received by this measurement device.

As already pointed out the absorption capabilities are strongly influenced by the degree of compression. Fig. 22 shows the diversification in absorption when varying the sample thickness.

With regard to all displayed results, the investigated cotton based composites showed remarkable good acoustical properties. A reason for this fact can be found in the rather fibrous structure which uncloses by microscopically views of the sample's surface and cross sections. Fig. 23 shows such views for a composite basing on cotton fibers and an epoxy binder. The sample was compressed to a thickness of 5 mm. Considering the emphasized acoustical potential it has to be noted that this fibrous character results in lower stiffness and stability of the composite.

Summary

The paper gave a survey and presented new developments on the current fields of application and research of natural fiber reinforced composites. Furthermore, investigations on the acoustical behavior of such composites basing on cotton fibers and both thermoset and thermoplastic matrices were described. Among other parameters the investigations covered the influence of such different binder materials and the share of binder component in the hybrid fleece and composite. It was shown that a matrix of phenol resin offers slightly higher absorption capabilities than an epoxy binder. As expected higher compression degrees lead to a decline in absorption. Generally, the utilized composite materials basing on cotton fibers showed a remarkable acoustical behavior. However, it has to be pointed out that this enormous potential was gained at the expense of mechanical strength and stiffness.

Acknowledgements

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Table 1. Properties of natural and synthetic fibers (mechanical pulping) [1, 7, 10, 11].

		Cotton	Flax	Hemp	Jute	Kenaf	Ramie	Sisal	E-Glass
Fineness	[dtex]	1-4	1-7	2-6	2-3	5-6	5-13	-	-
Diameter	[µm]	-	11-33	15-50	200	200	40-80	50-200	5-25
Length	[mm]	10-60	10-40	15-28	1-5	2-6	60-260	1-5	-
Max. stress	[cN/tex]	25-50	30-62	35-70	30-34	34,5-50	40-70	30-45	-
Tensile strength	[MPa]	330-585	345-1035	690-1000	393-773	930	400-1050	511-635	1800
Young's modulus	[GPa]	4,5-12,6	27,6-45,0	50,0	26,5	53,0	61,5	9,4-15,8	69,0-73,0
Density	[gcm^{-3}]	1,5-1,54	1,43-1,52	1,47-1,50	1,44-1,50	1,5	1,5-1,6	1,16-1,5	2,5
Max. strain	[%]	7,0-8,0	2,7-3,2	1,0-1,6	1,5-1,8	1,6	3,6-3,8	2,0-2,5	2,5-3,0
Specific tensile strength	[km]	39,2	73,8	69,3	52,5	63,2	71,4	43,2	73,4
Specific stiffness	[km]	0,85	3,21	3,47	1,80	3,60	4,18	1,07	2,98

Table 2. Usage of plant fibers for composite materials in Germany and Austria [15, 16].

Natural fiber	1996	1999	2000	2005 (estimated)	2010 (estimated)
Flax	2.100	15.900	20.000	-	-
Hemp	0	1.700	3.500	-	-
Jute	1.100	2.100	1.700	-	-
Sisal	1.100	500	100	-	-
Kenaf	0	1.100	2.000	-	-
Coir	0	0	1.000	-	-
Total	4.300	21.300	28.300	50.000-70.000	> 100.000

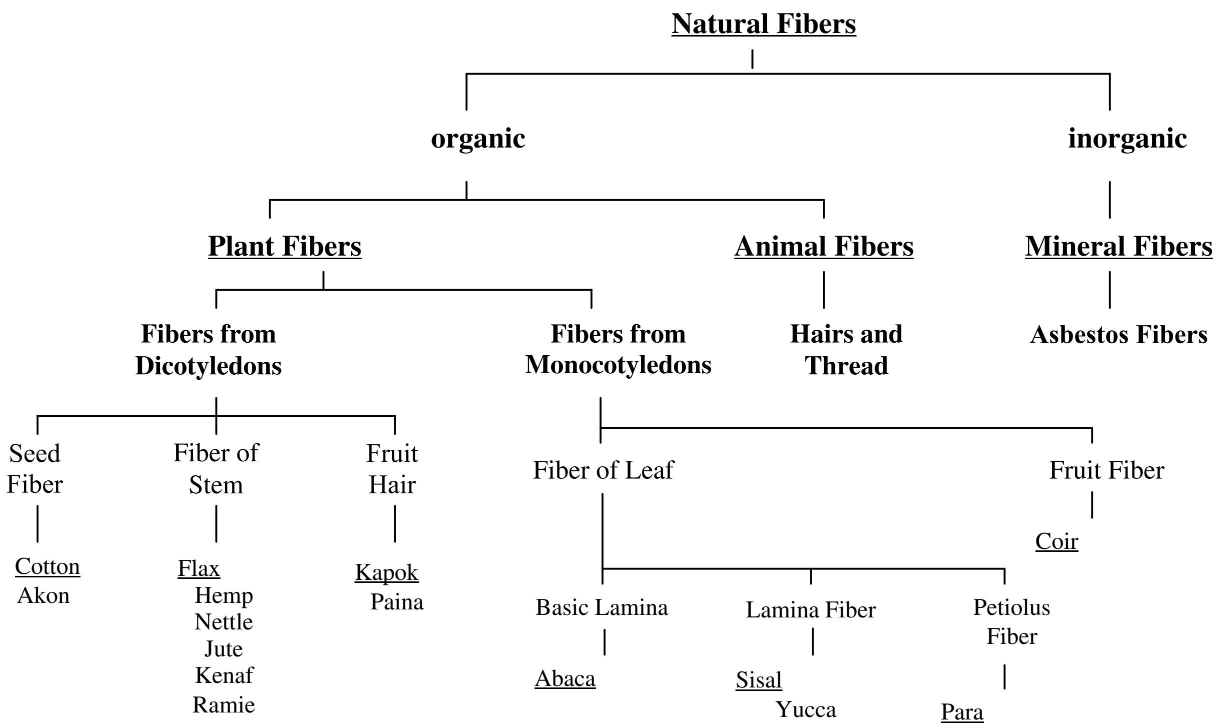

Figure 1. Classifications for natural fibers [2].

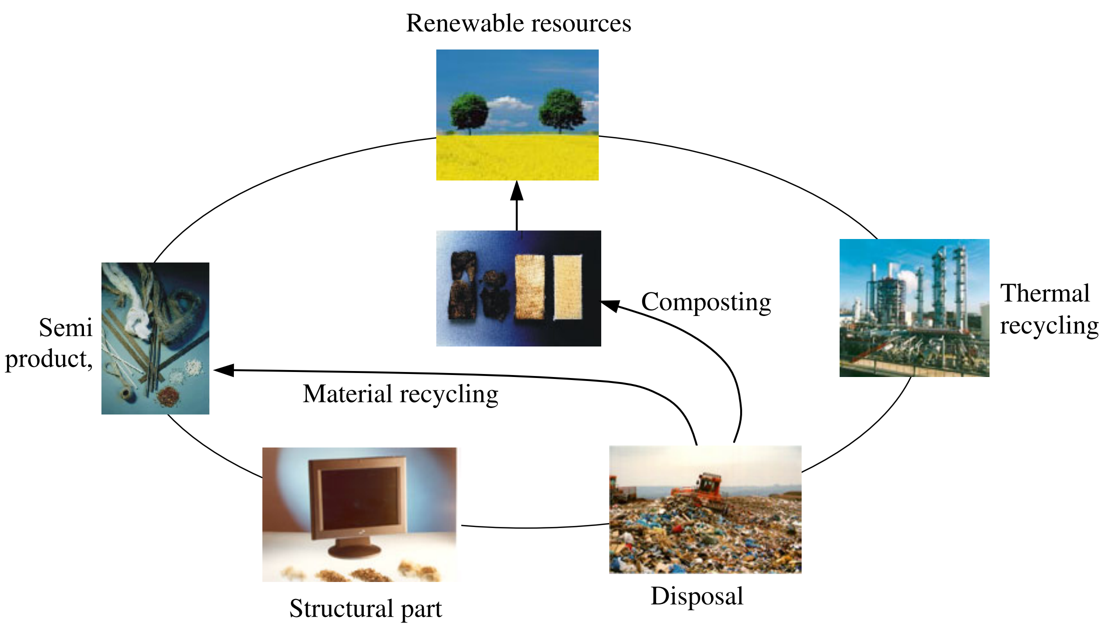

Figure 2. Natural cycle of renewable materials [5, modified illustration].

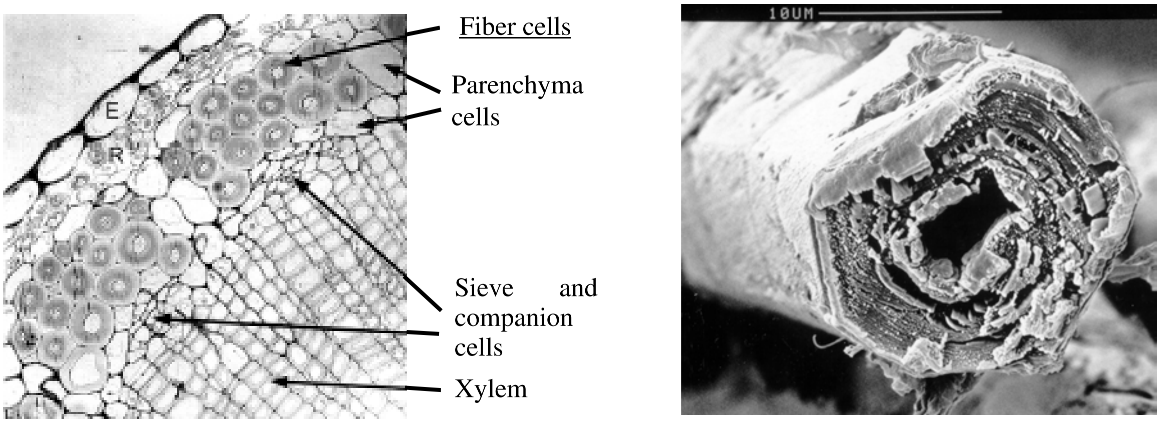

Figure 3. Structure and fracture surface of a flax fiber, noticeable are center lumen and cellular walls [6].

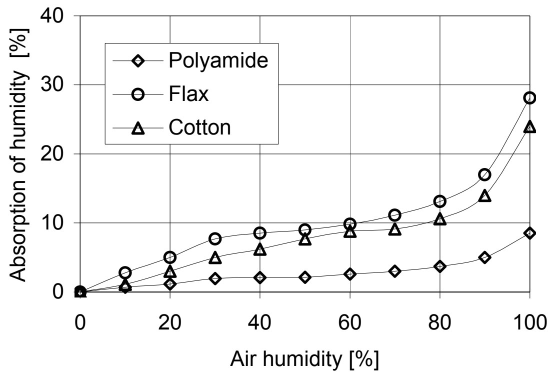

Figure 4. Absorption of humidity for different fiber materials [7].

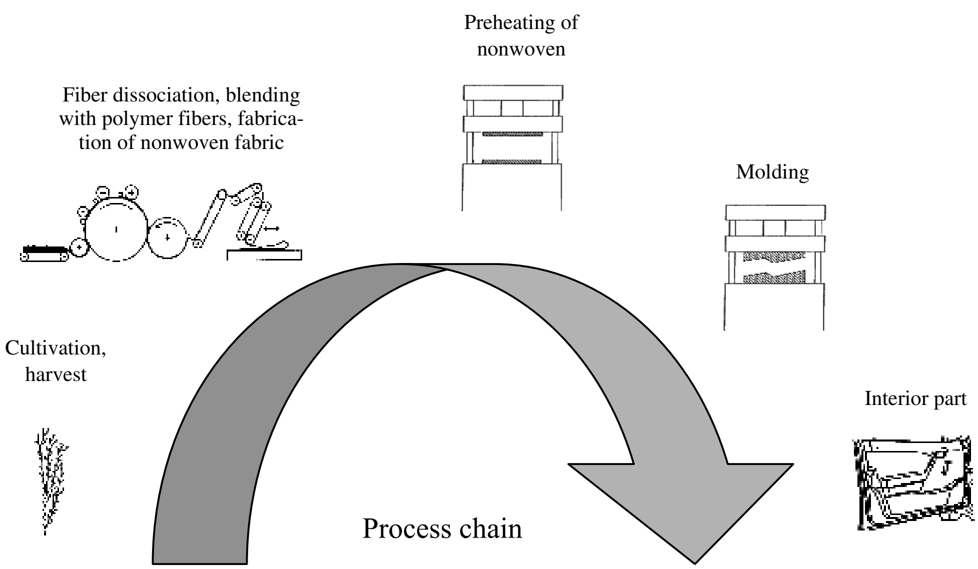

Figure 5. Process chain for the production of interior parts with a reinforcement of natural fibers [8, modified and extended illustration].

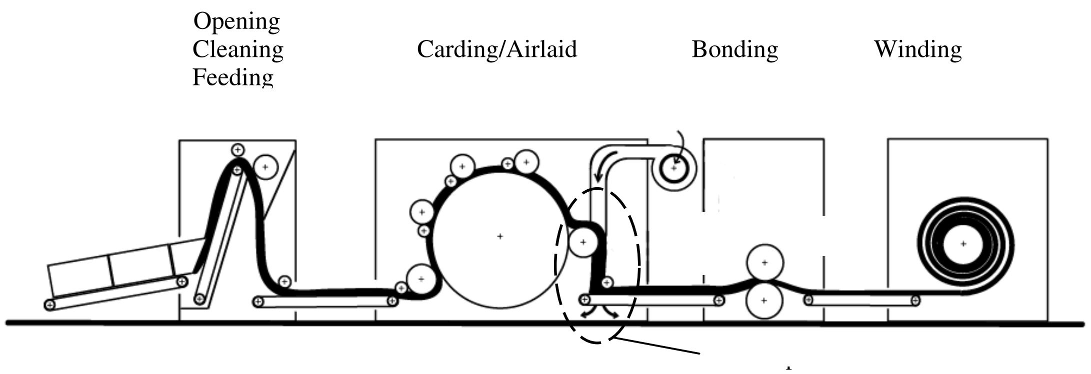

Figure 6. Generation of hybrid fiber fleeces by airlaid [9, modified illustration].

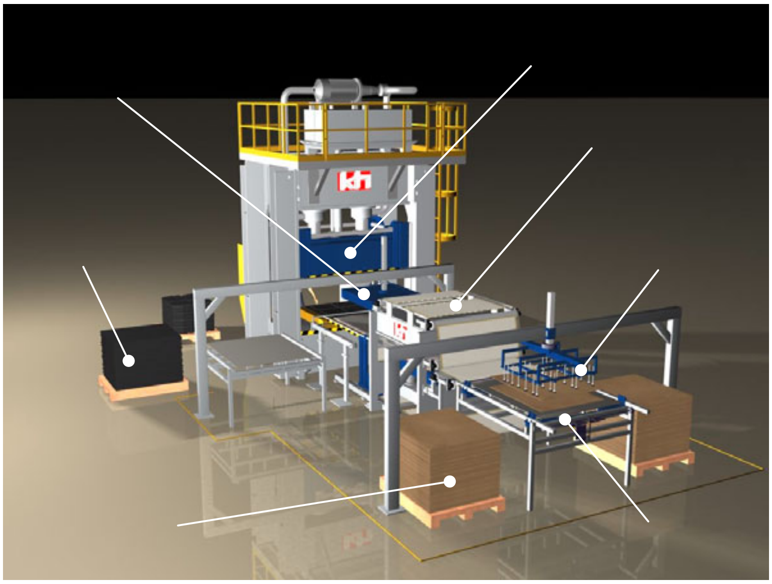

Figure 7. Compression molding plant for the processing of natural fiber reinforced thermoplastics [Origin: Heidel GmbH, Germany].

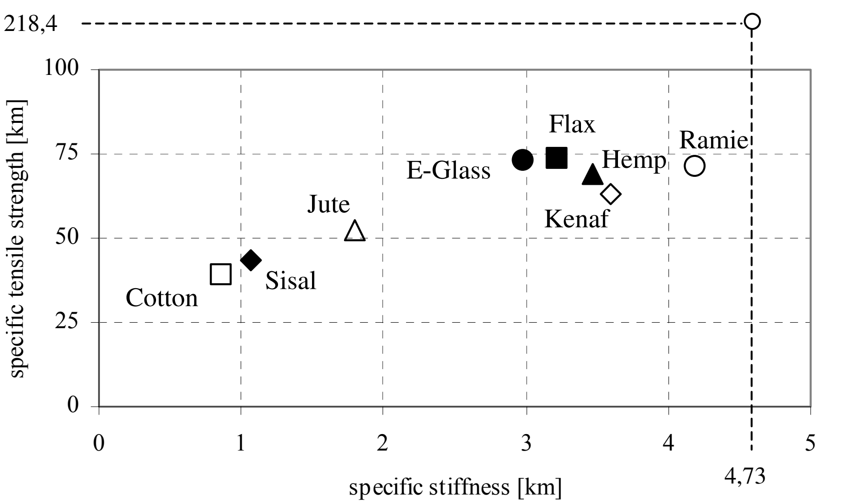

Figure 8. Specific stiffness and specific tensile strength for different plant fibers compared to synthetic fibers.

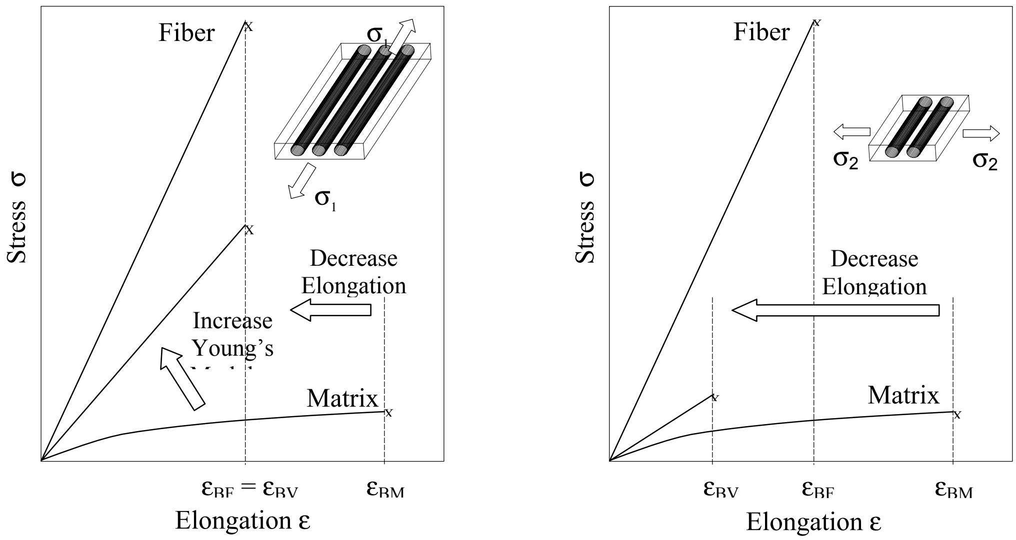

Figure 9. Stress vs. elongation for fiber reinforced composites [12, modified illustration].

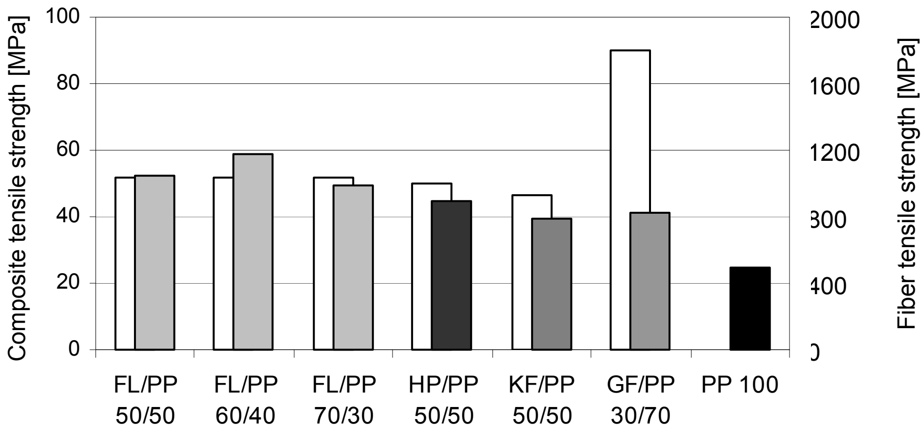

Figure 10. Tensile strength for composites basing on different fiber materials and pure PP in comparison to tensile strength for separate fibers (white bars).

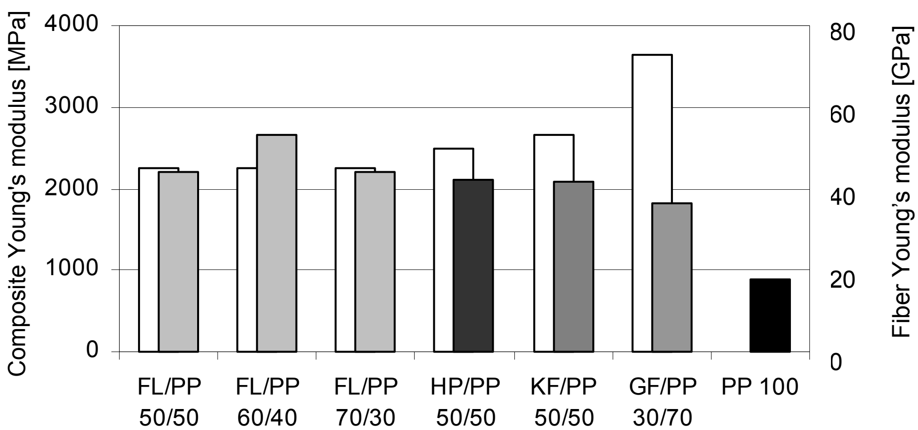

Figure 11. Young' modulus for composites basing on different fiber materials and pure PP in comparison to tensile strength for separate fibers (white bars).

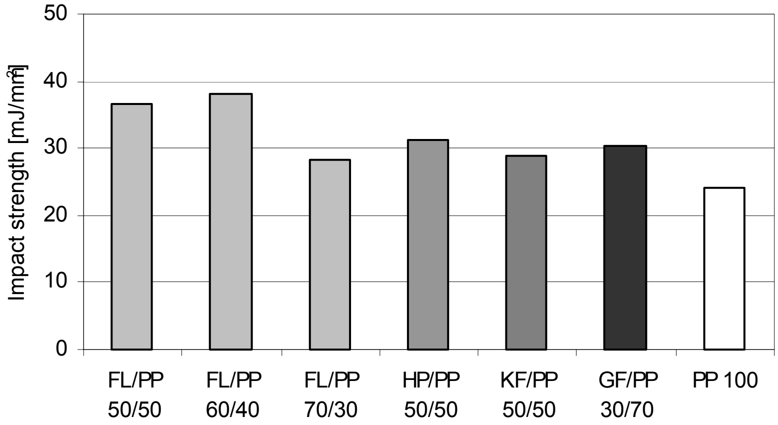

Figure 12. Impact strength for composites basing on different fiber materials and pure PP.

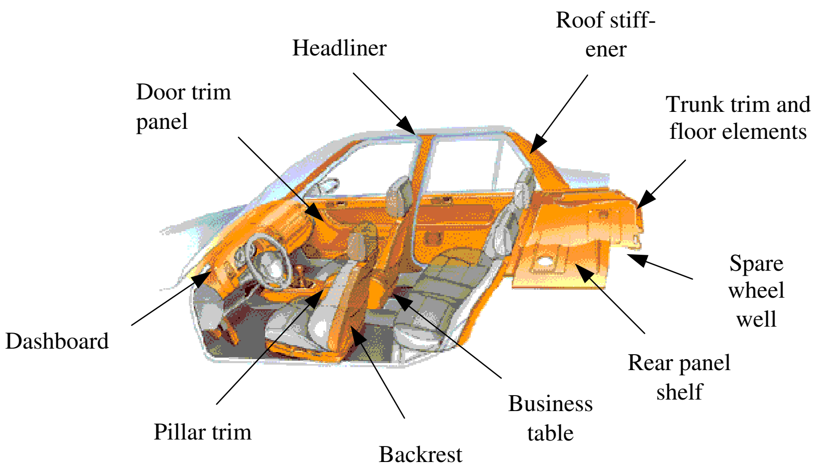

Figure 13. Applications of natural fiber composites as car interiors.

Figure 14. Concepts of pre-serial products for exterior automotive parts made of natural fiber reinforced composites [17].

Figure 15. Possible non-automotive applications for natural fiber reinforced composites [5, 18].

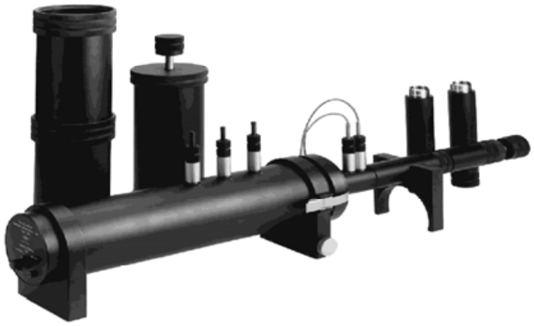

Figure 16. Impedance tube (Origin: B & K, Denmark).

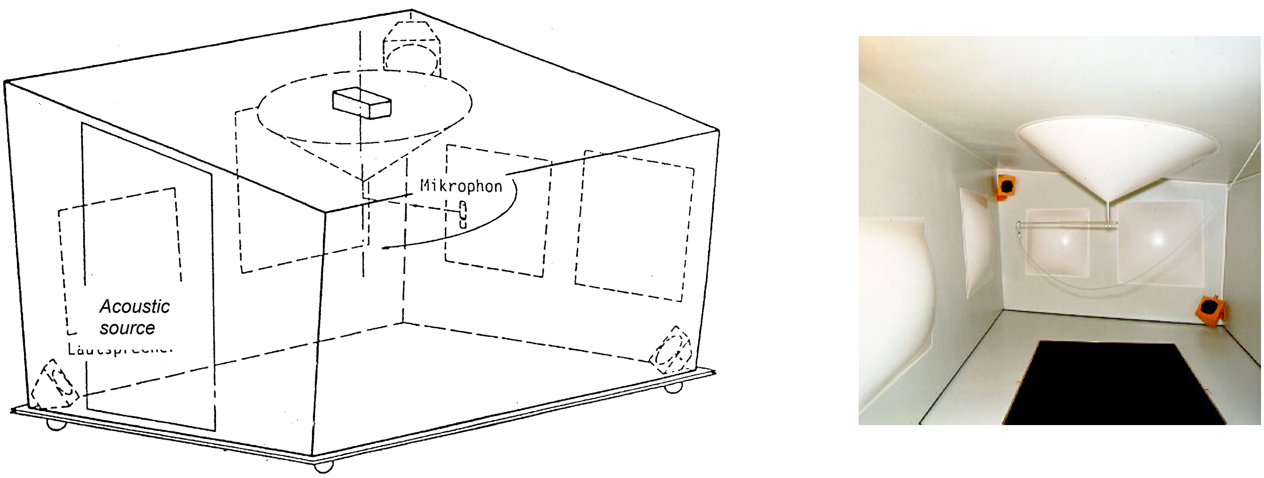

Figure 17. Schematic and an inside view of an Alpha cabin (Origin: BIK).

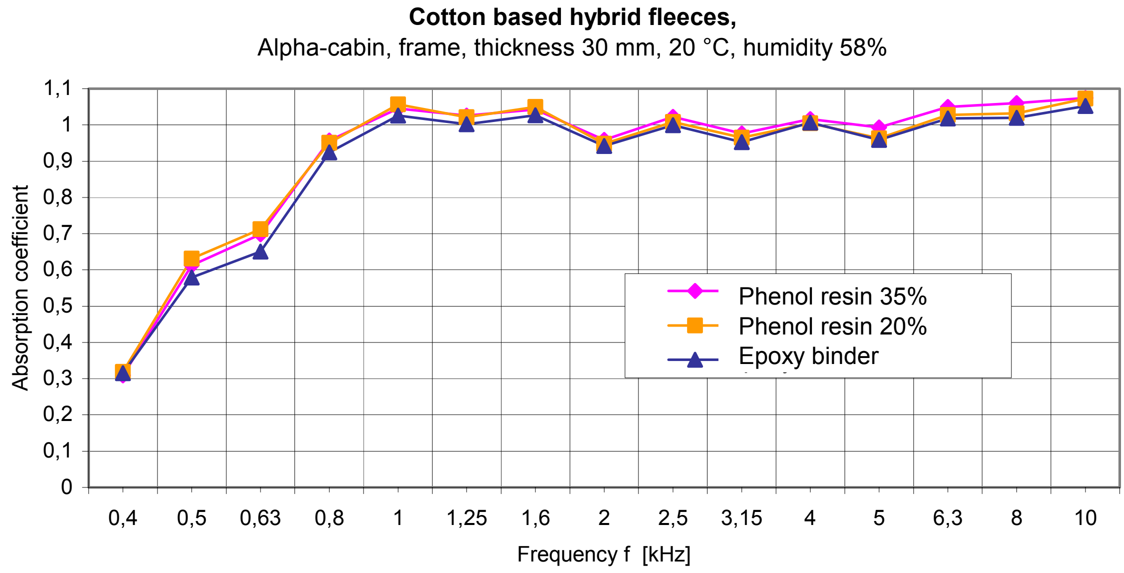

Figure 18. Absorption coefficient of uncompressed cotton based hybrid fleeces with different binder materials, Alpha- cabin.

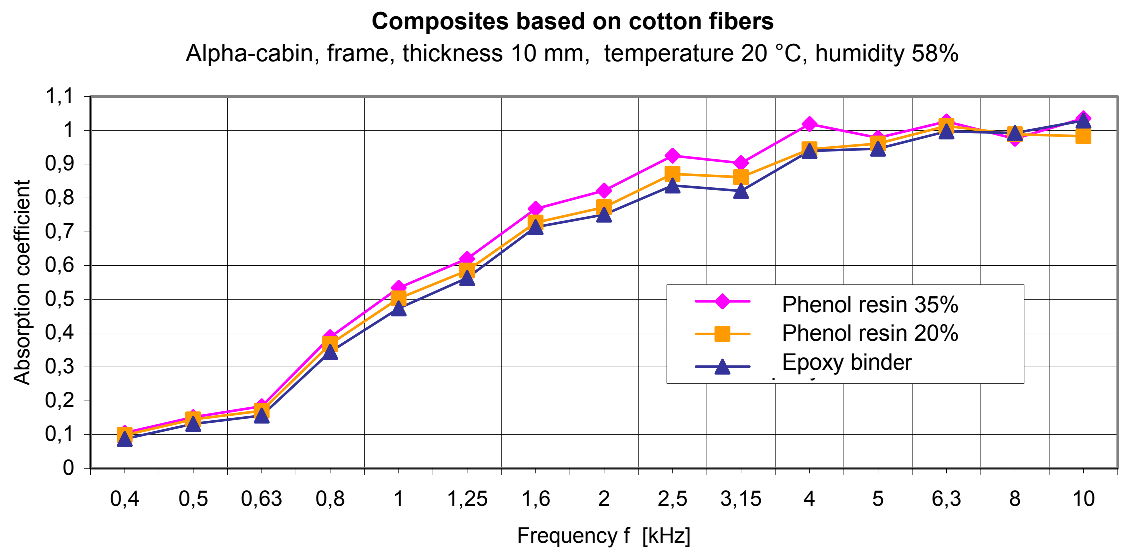

Figure 19. Absorption coefficient of cotton based composites with different binder materials, sample thickness 10 mm, Alpha cabin.

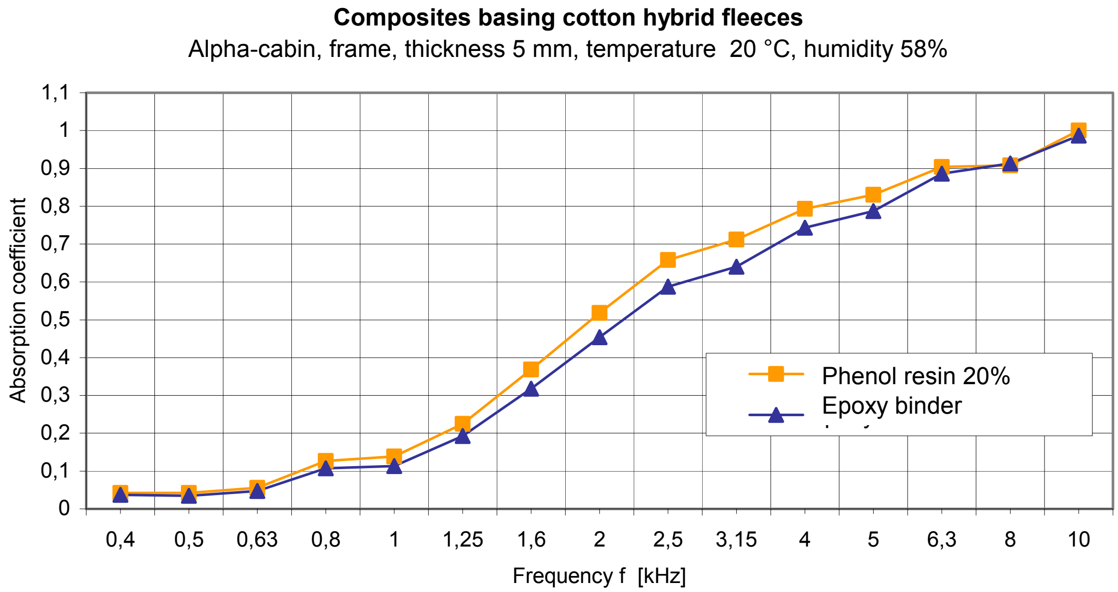

Figure 20. Absorption coefficient of cotton based composites with different binder materials, sample thickness 5 mm, Alpha cabin.

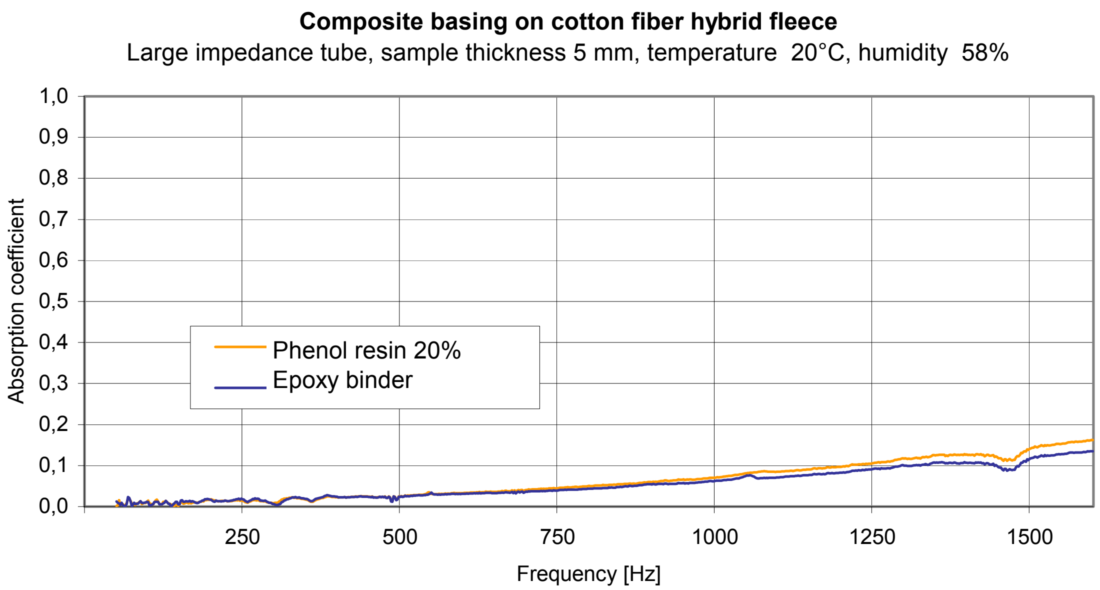

Figure 21. Absorption coefficient of cotton based composites with different binder materials, sample thickness 5 mm, large impedance tube.

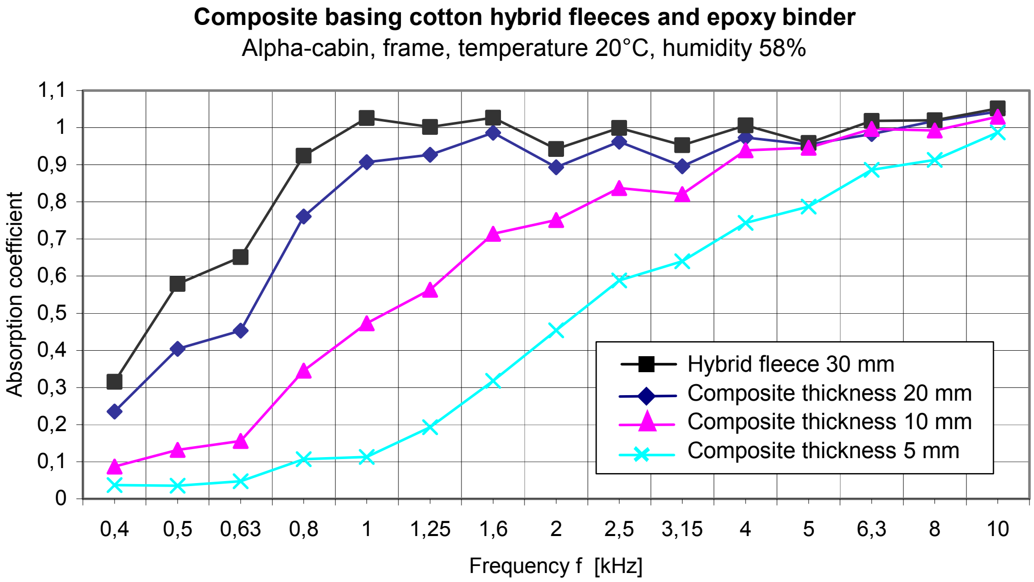

Figure 22. Absorption coefficient of cotton based composites with epoxy binder, Alpha cabin.

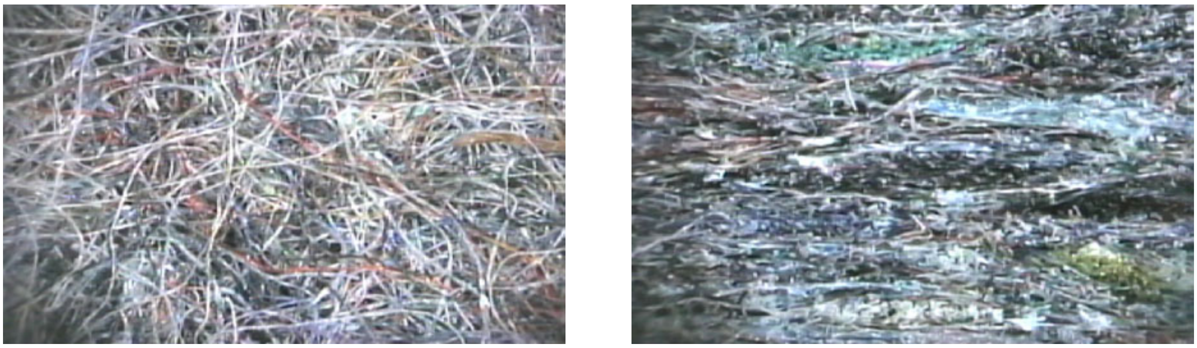

Figure 23. Surface (left) and cross section (right) of a cotton-epoxy composite, sample thickness 5 mm, magnification 100 x.s.