Journal of
Composites Science

MDPI

Article

Manufacture of Hybrid Natural/Synthetic Fiber Woven Textiles for Use in Technical Biocomposites with Maximum Biobased Content

Madina Shamsuyeva *, Jana Winkelmann and Hans-Josef Endres**

Fraunhofer Institute for Wood Research WKI, 30453 Hanover, Germany;
jana.winkelmann@wki.fraunhofer.de (J.W.); hans-josef.endres@wki.fraunhofer.de (H.-J.E.)
* Correspondence: madinashamsuyeva@hotmail.com; Tel.: +49-511-9296-2291

Received: 27 February 2019; Accepted: 18 April 2019; Published: 1 May 2019

check for updates

Abstract: This feasibility study investigates the flexural properties of biocomposites containing woven flax textiles (plain, twill, satin) and woven twill patterned hybrid textiles containing flax-/glass or flax-/carbon mixture for lightweight applications. Synthetic fibers are integrated as weft and flax fibers are integrated as warp yarns using a double-rapier weaving machine with a Jacquard attachment. The corresponding biocomposites are manufactured via vacuum infusion process using a biobased epoxy resin as a matrix. The manufactured biocomposites are analyzed with regard to their density and flexural properties. The results show that the use of hybrid textiles offers a promising solution for the manufacture of biocomposites with a higher biobased content and significantly improved flexural properties. Furthermore, the introduction of high-performance synthetic fibers in textiles enables the manufacture of biocomposites with an isotropic mechanical performance.

Keywords: weaving technique; natural fibers; biocomposites; hybrid textiles; hybrid biocomposites

1. Introduction

Due to growing environmental awareness, various industry sectors already use or attempt to use materials based on renewable resources in their production. In this context, numerous lightweight application sectors, like automotive or sport and leisure, have already validated the beneficial use of biobased composites containing natural fibers [1,2]. In comparison with synthetic fiber composites, natural fiber composites offer potentially more effective recycling possibilities, mainly due to their biobased feedstock and the possibility to generate an additional energy output in the case of incineration. Both life cycle-analysis of numerous natural fiber composites and various recycling approaches are described in literature [3]. Besides the ecological advantages that natural fibers like flax or hemp offer in comparison with synthetic high-performance fibers like glass or carbon, natural fibers also offer numerous technical benefits, like low density [4,5] or good sound and vibration damping properties [6]. The dynamically growing use of natural fibers in composite applications has been extensively described in literature [1,2,7,8]. Currently, natural fibers are mainly used as a single type of reinforcement in one composite. Several studies reported successful use of a mixture of two or more natural fiber types in one composite [9] or a mixture of natural fibers with glass fibers in injection molding [10,11], a sheet molding compound process [12] or as textile composites [11]. The main advantage of the hybridization of natural and synthetic fibers is the possibility to produce biobased composites with a significantly enhanced mechanical performance. Nevertheless, it is important to consider that despite the biobased feedstock of natural fibers, currently there are no satisfactory solutions for the recycling of hybrid composites consisting of natural and synthetic fibers.

However, in the case of hybrid textile biocomposites, the reported approaches concentrate on the use of sandwich structures, where the natural fiber mats are sandwiched between synthetic fiber mats, like e.g., Palmyra fibers sandwiched between glass fiber mats [11,13] or flax fiber plies sandwiched between carbon plies [14]. This approach is represented schematically in Figure 1.

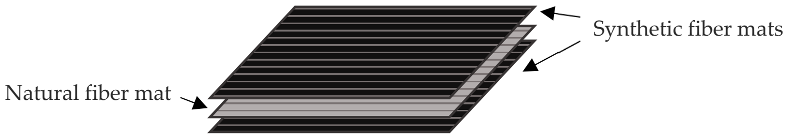

Figure 1. Schematic representation of natural fiber mats sandwiched between synthetic fiber mats.

Although this approach offers a simple manufacture of biobased hybrid composites with significantly improved mechanical properties, it is not possible to adjust the mechanical isotropy of the resulting composite material. Consequently, hybridization of natural and synthetic fibers directly in one textile would enable the manufacture of tailored biobased hybrid composites with isotropic mechanical properties. Therefore, the approach of this study represents an alternative solution for the manufacture of hybrid natural/synthetic fiber composites. Instead of stacking the textiles in a certain sequence to sandwiches, the new approach combines synthetic and natural fibers using a weaving technique directly in one textile. The aim of this study is to manufacture hybrid woven textiles containing flax fibers as a warp yarn and carbon fibers or glass fibers as a weft yarn and to evaluate the flexural properties of the resulting hybrid biocomposites as a function of yarn direction and compared with the flax biocomposites.

Woven textiles for composite applications: Woven textile composites containing a polymer matrix and a textile reinforcement provide an attractive solution for industry, since they are easy to handle and enable simple manufacturing of flat construction parts. The woven textiles for composite applications are available in various weave patterns. The basic weave pattern types used in this study, namely, plain, twill, and satin, are represented in Figure 2.

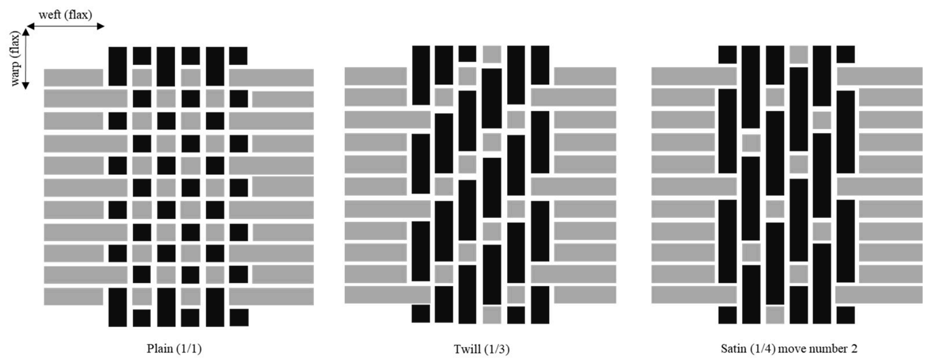

Figure 2. Schematic representation of the basic weave patterns: plain (1/1) meaning one warp yarn down and one warp yarn up, twill (1/3) meaning one warp yarn down and three warp yarns up and the satin (1/4) move number 2 meaning one warp yarn down and four warp yarns up with the displacement to the new line by step two.

The selected pattern has a significant effect on the properties of the manufactured textile and the corresponding composite material. Mass per area of textile and crimp of the yarns in the textile are among the most important properties affected by the weave pattern. The mass per area (or textile density) depends on the number of yarns required to realize a certain pattern. The crimp refers to the relation of the length of a strip of the woven textile and the length of the yarn, used to weave this textile [15,16]. Lower crimp means that the yarns in a woven textile are more stretched and the corresponding composites possess higher mechanical properties, e.g., satin compared with plain weave. At the same time, the textile composites with tighter crimp, e.g., plain weave, offer higher structural elongation than the corresponding composites with twill or satin weave [15]. The crimp of warp and weft yarns affect one another, and the desired balance can be adjusted using the settings of the corresponding weaving machine. It is important to consider, that in composites, the crimp is also responsible for weak points, because adequate wetting of fibers by matrix beneath the crossover points of the weft and warp yarns is challenging [17]. Furthermore, since the yarns are not straightened in the textile, the polymer matrix permeability is further inhibited. An overview of the advantages of the basic patterns at identical warp and weft density and identical yarn material for composite applications is presented in Table 1 [15].

Table 1. Overview of properties of different weave patterns used composite applications [15].

Property	Plain	Twill	Satin
Structural deformation	+++	++	+
Slippage resistance	+++	++	+
Single yarn pull-out strength	+++	++	+
Bending strength	+++	++	+
Permeability	+	++	+++
Drapability	+	++	+++
Handling	+++	++	+
Mechanical properties in the composite	+	++	+++

Note: "+"—low, "++"—average and "+++"—high.

In the cases where the number of warp and weft yarns per cm, i.e., yarn count, and the corresponding spacing between the yarns is equivalent, the fabric is called "balanced woven fabric". The balanced woven fabrics result in uniform tensile properties in both longitudinal and transverse directions [18]. Variation of the yarn counts can be used as an approach to adjust the isotropy of mechanical properties. Another possibility to optimize the mechanical performance of textile composites is to use at least two different yarn materials in one woven fabric. This method results in a so called "hybrid textile". In the last few years, high potential of the hybrid textiles made of synthetic fibers for composite applications gained the attention of industry and researchers. Numerous studies reported advantageous mechanical performance of these hybrid composites, e.g., aramid-/glass fabric polyester [19], glass-/aramid and glass-/polyethylene fabric epoxy [20]. An extensive review of the hybrid composites with glass, carbon and aramid fibers hybridized at different levels from ply to yarn level is described in literature [21].

However, the approaches for the manufacture of biobased hybrid woven textiles containing natural and synthetic fibers for composite applications are hardly known. Consequently, this feasibility study aims to demonstrate, that the manufacture of the twill-weaved biobased hybrid textiles for composite manufacture offers not only ecological benefits but also application-oriented advantages regarding bending properties.

2. Materials and Methods

2.1. Materials

In order to manufacture hybrid textile biocomposites, commercially available materials are used in this study. The properties of yarns used for the manufacture of the hybrid fabrics are listed in Table 2. The composites with the thermoset resin are fabricated using a partially biobased epoxy resin from Sicomin (France) with the trademark Greenpoxy 56 and the corresponding hardener SD 8822. The weight ratio of the resin and hardener is set according to the technical data sheet as 100:31. The density of resin is 1.198 g/cm^3 and the density of the hardener is 0.935 g/cm^3.

Table 2. Yarn types used for the manufacture of the hybrid textiles.

Yarn Type	Manufacturer/Supplier	Fineness (Tex)	Density (g/cm^3)
Flax	Franz Holstein GmbH	200	1.0 [5]—1.57 [22]
Glass *	P-D Glasseiden GmbH Oschatz	300	2.56 [5]
Carbon	Toho Tenax Europe GmbH	400 (6 K)	1.77

* E-glass.

2.2. Textile Fabrication

All of the textiles are manufactured using a double-rapier weaving machine with Jacquard attachment from the company Van de Wiele. The synthetic fibers are integrated as a weft yarn. The mass fraction of the synthetic fibers in hybrid textiles is set as 50%. In order to achieve this mass fraction, the weft yarn count and the corresponding distance between the weft yarns, x, is adjusted with respect to the fineness and the absolute mass of the used synthetic fibers. Consequently, the distance between flax fiber yarns in flax-/carbon textile is larger than the distance between glass fibers in flax-/glass textile or flax fibers within the flax textile:

$$x_{FLA} < x_{CF/FLA} < x_{GF/FLA}$$

The schematic representation of the pursued approach is demonstrated in Figure 3. The corresponding yarn counts in weft and in warp directions are summarized in Table 3.

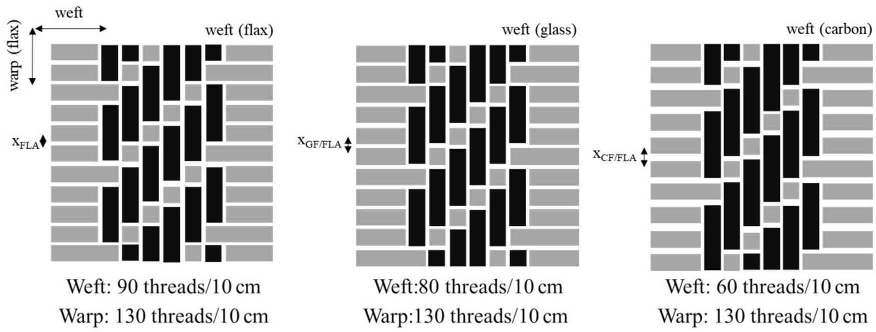

Figure 3. Schematic representation of the twill patterned hybrid textiles (from left to right): flax, flax-/glass, and flax-/carbon.

Table 3. Overview of the parameters used in this study during textile and composite manufacture as well as testing.

Material Form	Parameter	Flax Composites	Hybrid Composites
Textile	Weave art	Plain, satin, twill	Twill
	Warp yarn/weft yarn	FLA/FLA	FLA/GF FLA/CF
	Weft yarn counts (/10 cm)	Plain: 90 T Twill: 90 T Satin: 90 T	FLA/GF: 80 T FLA/CF: 60 T
	Warp counts (/10 cm)	Plain: 60 T Twill: 130 T Satin: 145 T	FLA/GF: 130 T FLA/CF: 130 T
	Areal weight of the textiles (g/cm^2)	Plain: 322 Twill: 483 Satin: 488	FLA/GF: 504 FLA/CF: 497
Composite	Number of plies	8	8
	Stacking sequence	0°/0°	0°/0°
	Manufacture method	Vacuum infusion	Vacuum infusion
	Direction of the flexural testing	Warp Weft	Warp Weft
	Digital microscope imaging of cross-section	Weft	Warp Weft

Note: T—Thread.

2.3. Composite Fabrication

Prior to the composite manufacture, all of the textiles are dried in oven at 80 °C for at least 24 h. Each composite contains 8 textile plies. The fibers are oriented in such a way that the warp yarns of all plies are aligned in the same direction (0°/0°). The composites are fabricated using vacuum infusion process. During this process, the dried textiles are firstly placed on a straight waxed steel mold. Afterwards the textiles are covered with a multifunctional fleece from the company Hacotech with the trademark Hacopeel, which combines three functions: it promotes resin flow and acts simultaneously as a perforated foil and a peel ply. The perimeter of the mold is sealed with a vacuum sealing tape. Two pieces of a spiral tubing are lied on the opposite sides of the textiles. At the opposite corners, on the outer side of the vacuum sealing tape, each piece of the spiral tubing is connected to a pump hose. One of the pump hoses is connected to a vacuum pump and the other is dipped into the epoxy system. At the beginning, the latter piece of the pump hose is closed with a clamp. The textiles are evacuated until a vacuum of 5 mbar is reached. Afterwards the system is closed off from the vacuum pump and left for at least a couple of hours to make sure that it is vacuum-tight. Before infusion, the epoxy resin is evacuated for 15 min in a desiccator connected to a vacuum pump. Schematic representation of the vacuum infusion process is presented in Figure 4. The rate of the infusion is controlled manually using the clamp on the pump hose. The vacuum infusion process takes place at room temperature, namely at 23 °C. The infused textiles are cured in an oven under a pressure mold at 80 °C for 30 min and cooled slowly overnight in the mold. The most important paramaters of the composite fabrication are represented in Table 3.

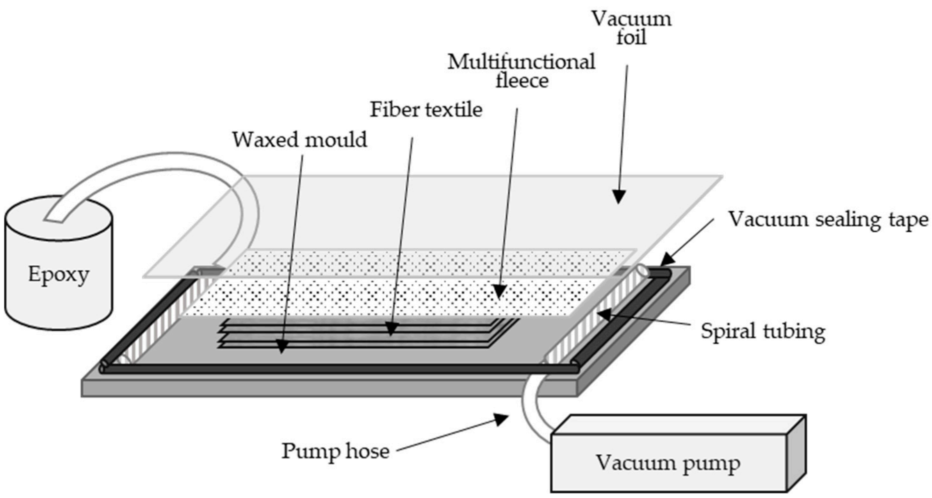

Figure 4. Schematic representation of the vacuum infusion process.

2.4. Flexural Properties

Each sample is tested in longitudinal and transverse directions, using five specimens. The specimens with a length of 100 mm and a width of 15 mm are prepared using a diamond saw from the composite samples with a length of 250 mm and a width of a 200 mm. The thickness of the specimens varies depending on the weave pattern. Therefore, it is presented and discussed as a part of results. Prior to the testing all of the specimens are conditioned for 88 h at 23 °C and 50% relative humidity. The testing is performed using Zwick/Roell Z2, 5 KN TN with a speed of 1 mm/min. The flexural properties of the manufactured composites are tested following the ISO 14125 using three-point bending testing at the Hanover University of Applied Sciences and Arts, Institute for Bioplastics and Biocomposites. In the ISO 14125, the preferential specimen thickness of woven textile composites is 4 mm. In the case of a deviating specimen thickness this standard advices to adjust the ratio of span length-to-thickness for the woven textile composites as 16 [23]. However, since the thickness of the manufactured composites varies from 5.10 to 7.36 mm, the advised span length, namely 81.6 to 117.76, mm could not be realized due to the limitations of the machine settings. Consequently, the span length for every tested composite, L_c, is adjusted individually and equals to ten times the specimen thickness, h_c, (Equation (1)). The composite thickness and span length values are represented and discussed as a part of results.

$$L_c = 10 \times h_c \tag{1}$$

Moreover, in order to evaluate these relative results of the modified testing, both mean values of flexural strength and modulus as well as the corresponding stress-strain curves are presented. The most important testing parameters are summarized in Table 3.

2.5. Digital Microscope Imaging

Digital microscope imaging system is used in order to examine qualitatively the void content in the manufactured composites. The analysis of the cross-sections of the tested specimens is carried out using a digital microscope VHX 5000 Keyence with a magnification of 30×. Prior to the testing, the surface of the specimens is cleaned with compressed air. The cross-section of flax composites is tested parallel to the weft yarn and the hybrid composites in both directions. The most important testing parameters are summarized in Table 3.

2.6. Density Measurement

The density of the manufactured specimens is measured via the buoyancy method (Archimedes' principle), which is also used in literature for the determination of the density of plant composites [24,25]. During the measurement, ethanol is used as a displacement liquid. The density of composites is calculated according to the Equation (2).

$$\rho = \frac{W_{air}}{W_{air} - W_{ethanol}} \times \rho_{ethanol}, \tag{2}$$

where: ρ is the measured density of the composite, $W_{ethanol}$ is the weight of the composite in ethanol, W_{air} is the weight of the composite in air, and $\rho_{ethanol}$ is the density of ethanol, respectively. Three specimens are measured for each composite.

3. Results and Discussion

In the following section, the manufactured composites are characterized according to their physical and mechanical properties.

3.1. Density

The generally used approach to determine the density of the fiber composites containing only synthetic fibers is based on volume fractions of the composite constituents with known densities [26]. Furthermore, the information regarding density of the composite components and the corresponding volume fractions can be effectively used for the estimation and evaluation of the void content and the mechanical performance of the synthetic fiber composites. However, in contrast with the synthetic fibers, the measurement of the density of natural fibers is challenging due to the geometry of these fibers and the presence of lumen. Moreover, according to literature, the measured density of flax depends on the measuring method and the origin of the fibers [27,28]. As an example, the density of the same flax measured via weighing in air and in a liquid (Archimedes' principle) and using a helium pycnometer differs by up to 12% [28]. Besides the measuring method, factors like origin and retting process as well as the moisture content affect the density of the flax fibers. Consequently, the density values of flax available in literature vary from 1.0 g/cm^3 [5] to 1.57 g/cm^3 [22].

Therefore, in this study the density of the produced composites is measured experimentally using the buoyancy method based on Archimedes' principle. One drawback of this method is that the liquid can enter the porosity network of the composites through the surface-connected pores [5]. Consequently, the results presented in this study merely demonstrate a relative comparison. In order to analyze the void content, the cross-sections of the composites are examined.

The density values of the flax and the hybrid composites are represented in Table 4, respectively. In the case of flax composites, it is observed that all of the composites possess nearly the same density. These results show a good correlation with the literature values of flax fiber-reinforced epoxy composites manufactured via vacuum infusion [25], namely, 1.28 g/cm^3 and 1.26 g/cm^3. Two explanations are possible for this behavior. Firstly, it is possible that the density of flax and epoxy resin is very similar, and therefore change in mass fractions is not reflected in the overall composite density. Secondly, since the density is the ratio of mass to volume, the comparable density values show that the mass and the volume of the composite specimens increase proportionally. However, the proportional increase of the ratio can be induced by different sources, because the wetting degree of textiles has not been analyzed yet. In the latter case, the slightly lower mean value of the flax composite with twill weaved textile can be induced by higher void content. Furthermore, the density value of this composite compared with the density values of other two composites has higher deviation, showing the inhomogeneity of the composite. In the next step, the density values of flax composites are correlated with the fiber mass fraction and images of the cross-sections of the specimens.

Table 4. Density values of composites.

Sample	Density (g/cm^3)
Plain_FLA	1.29 ± 0.01
Satin_FLA	1.29 ± 0.01
Twill_FLA	1.28 ± 0.02
FLA/CF	1.34 ± 0.01
FLA/GF	1.45 ± 0.01

A clear density difference is observed in the case of the hybrid composites. Namely, the density of the flax-/carbon composite is 5% and density of the flax-/glass composite is 13% higher than that of the flax biocomposite. It is important to consider that except the specimen thickness, other parameters of the hybrid composites, like e.g., the areal weight of the hybrid textiles, the absolute mass of the hybrid composites as well as the mass fractions of flax and synthetic fiber yarns are same (Table 3). Consequently, this difference in density is definitely induced by the smaller volume of the hybrid composites, i.e., the thickness of the flax-/glass composite (5.89 mm) is lower in comparison with the thickness of the flax-/carbon composite (6.44 mm). The mass fraction of synthetic fibers in flax-/carbon and in flax-/glass composites is same, namely, 50%. The density difference can be further increased by void content or material-specific properties of synthetic fibers, which promote or inhibit their compatibility with flax fibers, like e.g., sizing, rigidity, etc.

To sum up, differences in fiber volume of the synthetic fibers used for the manufacture of hybrid textiles, void content and possibly differences in sizing or rigidity of glass and carbon fibers may lead to the differing density values. Therefore, similarly, as in the case of the flax composites, the determination of the fiber mass content in the composites and optical examination of the cross-section are conducted in order to better understand this behavior.

3.2. Fiber Mass Fraction

The values of the specimen thickness, span length, absolute mass of the composite, and fiber mass fraction are summarized in Table 5. The results show that both weave pattern and used yarn type have a significant influence on the fiber mass fraction in the biocomposites and the thickness of the specimens. As already mentioned above, both areal weight as well as the height of the resulting composites are dependent on the number of yarns employed in the realization of a certain pattern.

Table 5. The height and the corresponding span during the testing of the flexural properties.

Composite	Composite Thickness (mm)	Span Length Offlexural Testing (mm)	Absolute Mass of Composites (g)	Mass Fraction of Textile in Composite (%)
Plain_FLA	5.10 ± 0.09	51.0	580	37
Satin_FLA	7.36 ± 0.09	73.4	839	41
Twill_FLA	6.53 ± 0.07	65.1	832	35
Twill_FLA/CF	6.44 ± 0.10	65.0	753	47
Twill_FLA/GF	5.89 ± 0.10	58.3	754	47

In this study, the number of warp counts used for the manufacture of plain, twill, and satin patterned flax textiles is 60 T/10 cm, 130 T/10 cm and 145 T/10 cm, respectively (Table 3). According to the results, the thickness of the flax composites shows good correlation with the areal weight of the textiles presented in Table 3 and the absolute composite mass presented in Table 5. The lowest height belongs to the composite with plain weaved fabric, namely, 5.10 mm (322 g/cm^2 and 580 g), followed by twill 6.53 mm (483 g/cm^2 and 832 g) and satin 7.36 mm (488 g/cm^2 and 839 g). The information about the specimen thickness is important for the testing and it plays a significant role in a product manufacturing, since it affects the wall thickness of the construction part. Furthermore, the change in the thickness of the specimens leads to the change in volume and thus, possibly, the density of the specimens. During vacuum infusion process, only one metal mold is used and the thickness of the resulting specimen depends significantly on the textile type and the areal weight.

Regarding the fiber mass fraction, the flax composite with satin weave results in the highest fiber mass fraction (41%), followed by the composite with plain weaved textile (37%) and twill weaved textile (35%), respectively. General comparison with other studies on other natural fiber composites shows that these fiber mass fractions are reasonable. As an example, manually manufactured plain, twill and satin weaved kenaf textiles integrated into unsaturated polyester resin via vacuum infusion result in mass fractions of 37.9%, 40.0% and 38.0%, respectively [29]. A further example is a unidirectional (UD) and multidirectional (MD) flax fiber-reinforced epoxy composite manufactured using vacuum infusion [25]. In this case, based on the mass fraction of the flax and its density taken as 1.540 g/cm^3, the authors report the fiber volume fractions. Recalculation of the reported fiber volume fractions to the fiber mass fraction results in 44.5% for UD composite and 37.9% for MD composite, respectively.

Comparison of the fiber mass fractions of the plain, twill and satin patterned flax composites shows that the highest fiber mass fraction (41%) is achieved in the case of the satin textile with the highest areal weight (488 g/m^2). This leads to the assumption that the satin weave including the largest number of fiber yarns may have the lowest epoxy resin permeability and fiber wetting. The mass fraction of flax fibers in the composites with plain and twill weaves is comparable, namely, 37% and 35%, although the number of the used yarns and the areal weight of the plain and twill fabrics differ significantly.

In the case of the hybrid textile composites, the height of the flax-/glass composite is 9% lower than that of the flax-/carbon composite. At the same time, both of the composites have the same overall fiber mass fraction (47%). Hence, the lower thickness and correspondingly lower volume of the flax-/glass composite is induced by the difference of the glass fiber density in comparison with the carbon fiber density. As such the same mass of glass fibers possesses lower volume results in a thinner composite.

3.3. Digital Microscope Images

Depending on the manufacturing method and matrix type, there are several types of voids, which may be present in composite materials [5]. Particularly, large cavities can be induced by gross defects during manufacturing and small voids adjacent to fibers can be caused by incomplete infiltration. There are two commonly used approaches for the determination of the void content: examination of a composite cross-section or measurement of the composite density [5]. Both of these methods have advantages and disadvantages during application on natural fiber composites. Namely, examination of the cross-section provides information about the distribution and a relative size of the voids. However, during the sample preparation, such as during polishing, the fiber fragments may be lost. Furthermore, this approach represents void content merely in the analyzed specimen area. Consequently, if the voids are not homogeneously distributed within the specimen, the results of the analysis can be misrepresentative. During the measurement of the density using Archimedes' principle, this problem is avoided. However, in order to use this approach for the evaluation of void content, the density values of all constituents have to be determined. As already mentioned above, an accurate measurement of flax density is challenging. Thus, in the scope of this study the relative void content in the composites is determined qualitatively combining the results of the both methods.

The digital microscope images of the composite cross-sections are represented in Figure 5. These images show clearly that the composites include different void content. Particularly, the flax composite with plain weaved textile, which contains the lowest number of yarns (Table 3) and has the lowest thickness shows nearly no voids. On the other hand, the flax composite with twill weaved textile has many large voids. The flax composite containing satin weaved fabric includes also some voids, but these voids are significantly smaller, than in the case of the composite with the twill weaved textile. At the same time, both of the latter composites contain a comparable number of yarns (Table 3). Furthermore, the comparison of the images representing the cross-sections of the twill and satin weaved flax composites shows that, the voids are present at the points where the weft and warp yarns are crossing each other, hence representing bad infiltration [5]. The presence of these large voids explains the lower density value of the flax composite with twill patterned textile and its high deviation. However, it does not explain the lower fiber mass fraction of the twill patterned flax composite in comparison with the flax containing plain patterned textile.

In the case of hybrid textiles, the void content in the composite with flax-/carbon textile is significantly higher than in the case of the composite with flax-/glass textile. This behavior further explains the lower density of the flax-/carbon composite in comparison with the flax-/glass composite. Thus, showing that there are mainly two factors leading to the lower density of the flax-/carbon composite, namely, lower volume of the glass fibers required to reach the mass fraction of 50% and the higher void content in flax-/carbon composite.

Regarding the source of the voids in hybrid composites, as mentioned above, there are two major reasons: manufacturing defects or incomplete infiltration. Since all of the composites are manufactured using the same technique and no visual defects are notified during or after the fabrication, most probably the higher void content in flax-/carbon composite is induced by the incomplete infiltration. One of the possible explanations could be better compatibility of glass and flax fibers than that of carbon and flax fibers. This can be induced by the chemistry of the glass fiber surface. Both of these fibers are polar due to the hydrophilic nature caused by the presence of hydroxyl groups on the fiber surface. Furthermore, the sizing of the glass fibers can promote the compatibility. As an example, silane-based coupling agents, which are commonly used in order to improve fiber-matrix-adhesion in glass fiber composites are also effectively used for the same aim in natural fiber composites [30]. On the other hand, the inert surface of carbon fibers is firstly oxidized and afterwards coated with a polymer finish, promoting fiber-matrix-adhesion [31]. Commonly, this polymer coating is adjusted for a special matrix type. Consequently, this may inhibit the compatibility between carbon and flax fibers. However, in order to prove this assumption, a separate study focused on the compatibility of synthetic and natural fibers should be conducted.

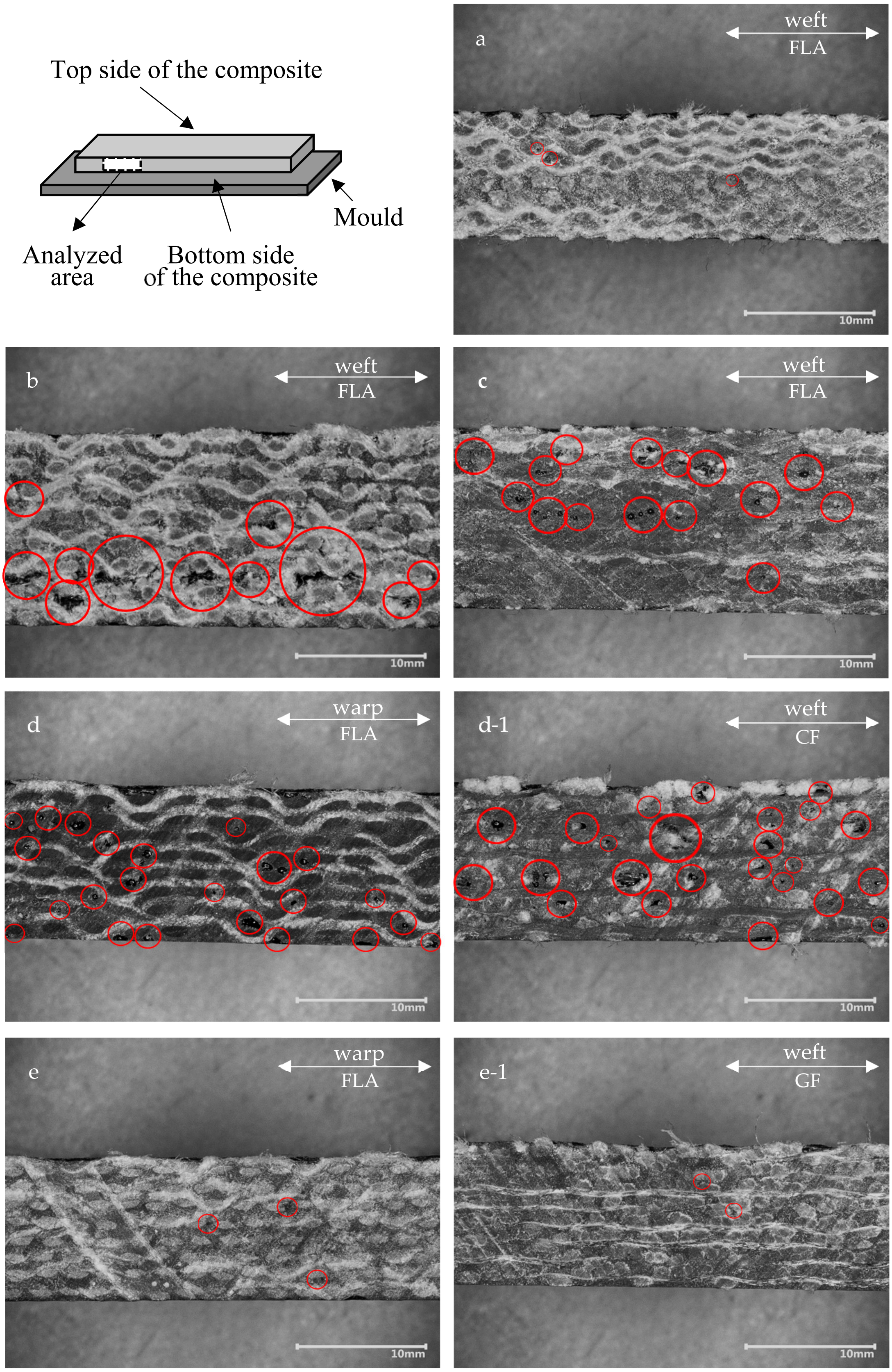

Figure 5. Digital microscope images of the cross sections of the flax composites with plain (**a**), twill (**b**) and satin (**c**) weaved textiles and hybrid textiles composing of carbon and flax (**d**) and (**d-1**) as well as glass and flax (**e**) and (**e-1**).

3.4. Flexural Properties of Flax Composites

Due to the modification of the span length, the achieved results represent merely a relative comparison of the flexural properties of the composites. According to literature, the span length significantly affects the results of the synthetic fiber-reinforced composites. As an example, the values of flexural strength and modulus of a unidirectional glass fiber-reinforced thermoset polymer analyzed with a span length varied from 10 mm to 20 mm, result in 540 MP and 9.7 GPa and 792 MPa and 20.3 GPa, respectively [32]. Whether the varied span length affects the flexural properties of natural fiber composites and hybrid textile composites as significantly as in the case of the synthetic fiber composites has not been studied yet. However, it is important to consider that under accurate adjustment of the span length according to testing standard, the mean values of the composite properties can vary.

The flexural modulus values of the flax composites are presented in Figure 6. The composites with plain and twill weaves show similar tendency regarding isotropy, i.e., the modulus in the weft direction is higher than that in the warp direction. The composite with satin patterned fabric shows an inverse behavior. In the warp direction, the composite with the satin weave shows the highest result (6690 MPa). This is followed by the composite with the twill patterned fabric (5364 MPa) and the composite with plain weave has the lowest modulus (4480 MPa). This tendency is expected based on the overview described in Table 1 and is induced by the increasing number of yarn counts, namely 60, 130 and 145 T/10 cm for plain, twill and satin, respectively (Table 3).

In the weft direction, the mean value of the composite with twill weaved fabric is the highest, followed by slightly lower modulus of the composite with plain patterned textile and the composite with satin weave has the lowest value. Correlation of these data with the fiber mass fraction represented in Table 5, shows that the decrease of the flexural modulus in weft direction is inversely proportional to the decrease of the fiber mass fraction of the biocomposites. Namely, twill fabric composite with 6806 MPa and 35%, plain fabric composite with 6481 MPa and 37% and twill fabric composite with 5729 MPa and 41%.

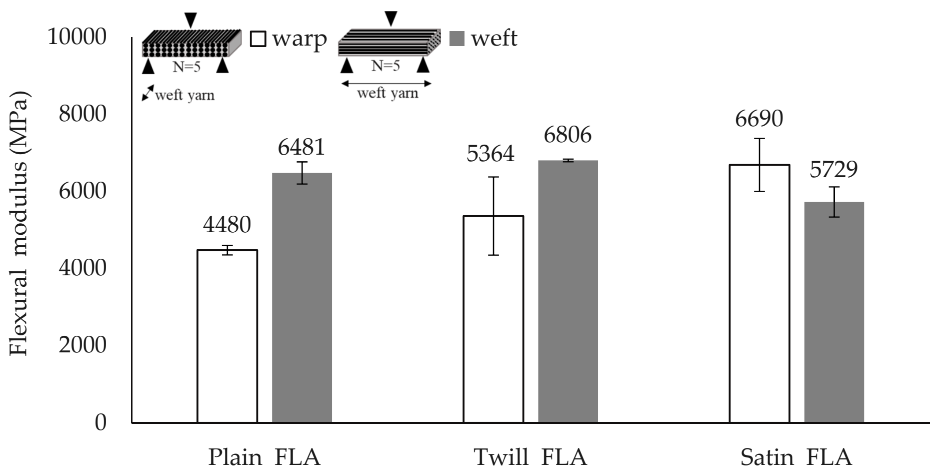

Figure 6. Flexural modulus of flax composites.

The flexural strength of the flax composites is represented in Figure 7. Similarly, as in the case of the flexural modulus, the strength of composites with plain and twill weaved fabrics in weft direction is higher than in warp direction. The composite with the satin weaved fabric shows an inverse trend and its strength in warp direction is 16% higher than in the weft direction. In the warp direction, the composite with plain weaved fabric shows the lowest strength (115 MPa) and the composite with the satin weaved fabric shows the highest strength (156 MPa). Consequently, the flexural strength of the composite with twill weaved fabric in warp direction is 23% higher than that of the composite with plain weaved fabric. These results are in agreement with the increasing number of yarns in this direction (Table 3).

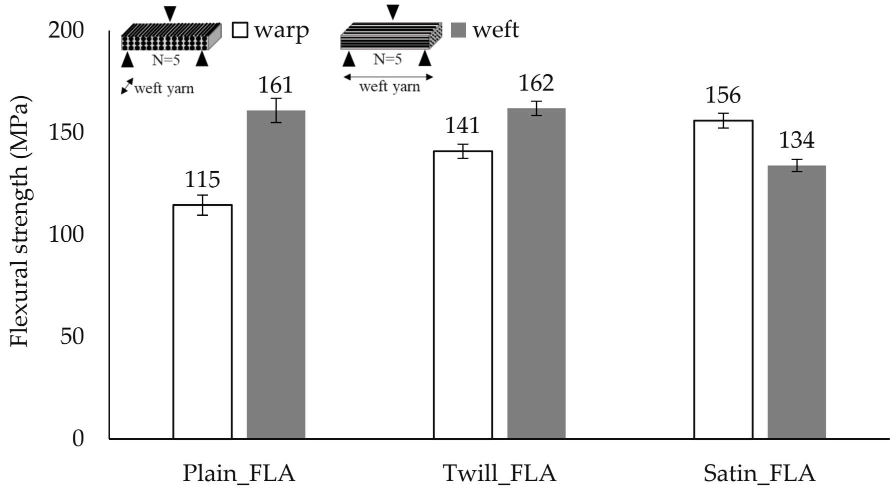

Figure 7. Flexural strength of flax composites.

In the weft direction, the strength of the composites with plain and twill patterned fabrics is nearly equal, 161 MPa for plain and 162 MPa for twill, respectively. The strength of the composite with satin patterned fabric is approximately 17% lower. This tendency shows that the strength is not related to the number of yarns in weft direction (90 T/10 cm). However, similarly as in the case of flexural modulus, this tendency is inversely proportional to the fiber mass fraction. At the same time, the fact that the plain weaved textile reinforcement leads to a nearly the same strength as the twill weaved textile makes it more advantageous for application. The plain weaved textile has a significantly lower areal weight than twill fabric and results in a lighter and thinner composite. The areal weight of plain fabric is 322 g/cm^2 and of twill 483 g/cm^2 (Table 3). The stress-strain curves of the flax composites represented in Figure 8 shows that all of the composites show brittle behavior.

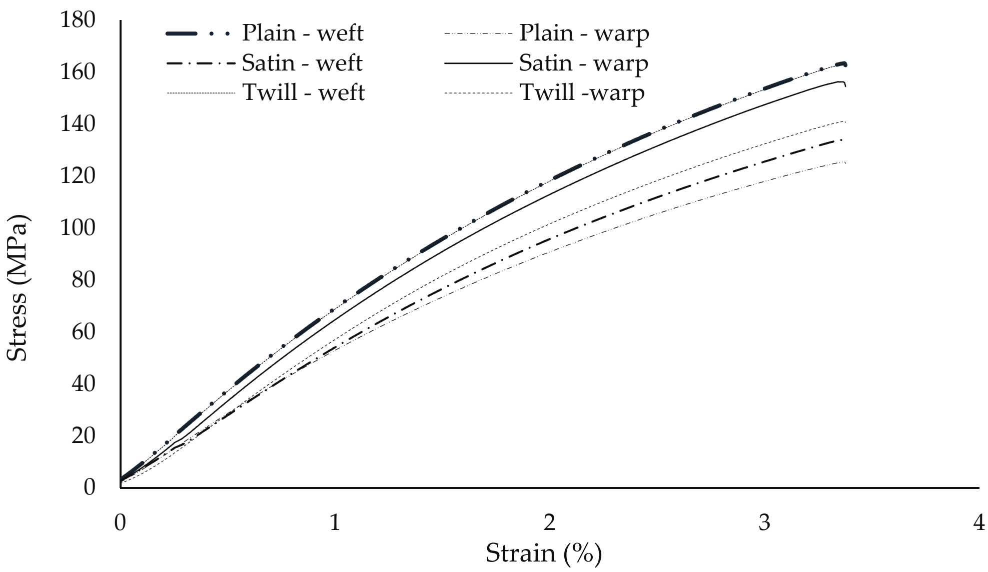

Figure 8. Stress-strain curve of flax composites.

In textile composites, there are a lot of sources, which affect the mechanical performance of the composites, e.g., fiber type, yarn type, twisting, fineness, etc. Furthermore, the natural instability of natural fibers plays an important role. Thus, the properties of the same fiber type are strongly dependent on the source, retting process, etc. and may significantly differ. Therefore, an accurate comparison of the achieved results with literature is hardly possible. However, a general overview of the literature shows that the achieved results are reasonable. As an example, the analysis of the flexural properties and further mechanical characteristics (tension, Charpy) of a woven flax fabric-reinforced epoxy in weft direction are higher, than in the warp [33]. This tendency seems to be independent of the polymer matrix or surface treatment of flax fibers. As an example, the investigation of the plain weaved flax fiber-reinforced soy protein concentrate composites with regard to the flexural properties in the weft and warp directions shows that independently of the surface modifications of the composites via additives the flexural properties in weft direction are higher than that in the warp [18]. A study on the effect of plain and 2/2 twill weave patterns with varied yarns counts in weft and warp directions on the flexural properties of composites in warp direction shows that, the flexural strength of the composites with twill pattern is only slightly higher than that of the composite with the plain patterned fabric [34]. This short review shows that although numerous parameters are varied among the different studies (yarn counts, type of twill pattern and material origin) there are certain common tendencies, if natural fibers are used as a textile reinforcement in composites. As an example, if the mechanical properties of plain and twill patterned textile composites are higher in the weft direction than in the warp direction, the plain and twill textile composites will result in comparable flexural properties.

At the same time, it is important to consider that the elimination of the voids determined via digital microscope imaging and density measurement, would further improve the flexural properties of the flax composites with twill and satin patterned fabrics.

3.5. Flexural Properties of Hybrid Composites

The values of the flexural modulus of the hybrid textile composites are presented in Figure 9. As expected, the highest flexural modulus value (13,465 MPa) belongs to the flax-/carbon composite in weft direction. The lowest modulus value of 5364 MPa belongs to the flax composite in warp direction. It is interesting to note, that the flax-/glass composite in weft direction (7134 MPa) is only approx. 5% higher than that of the flax composite (6806 MPa). In warp, i.e., flax fiber, direction the modulus increases from 5364 MPa (flax composite) to 6474 MPa after introduction of the carbon fibers and to 7329 MPa in the case of the flax-/glass biocomposite.

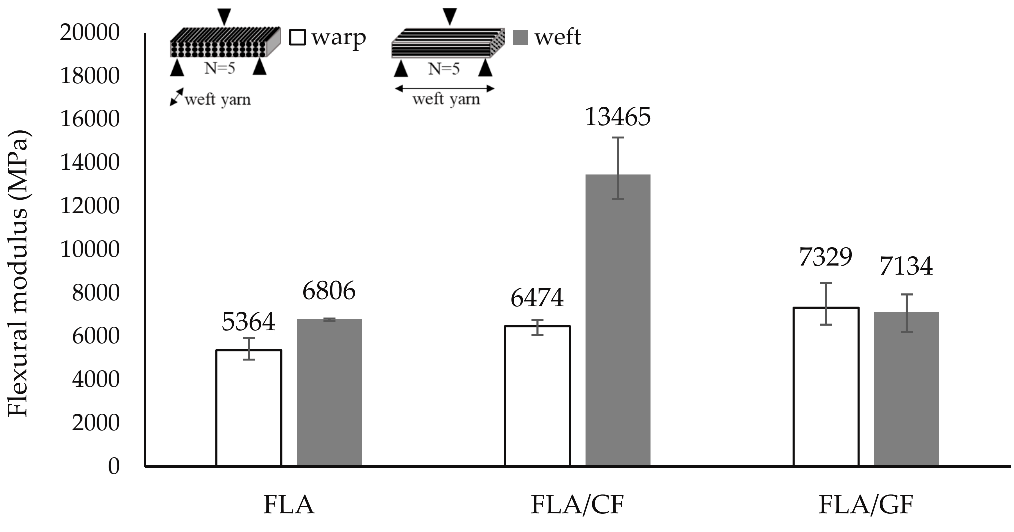

Figure 9. Flexural modulus of hybrid composites.

The flexural strength of the hybrid composites is demonstrated in Figure 10. The highest value of 332 MPa is achieved, as expected, in the weft direction of the flax-/carbon composite. The flax-/glass composite has a flexural strength in glass fiber (weft) direction, nearly 14% lower than the composite with carbon fibers, namely, 286 MPa. The introduction of synthetic fibers into the flax textile has a controversial influence on the flexural strength of the composite in warp direction (flax fibers). In the case of flax-/carbon composite, the flexural strength in warp direction is decreased by 5% and in the case of flax-/glass composite the value is increased by 13%. This is possibly induced by the changed alignment of the synthetic fibers within the composites, namely in the case of flax-/glass composite the flax fibers are more squeezed than in the case of flax-/carbon composite (Figure 5). However, since there are several sources affecting influencing this behavior, namely, type of synthetic yarns, weft counts, void content, it is not possible to make a clear statement on this behavior.

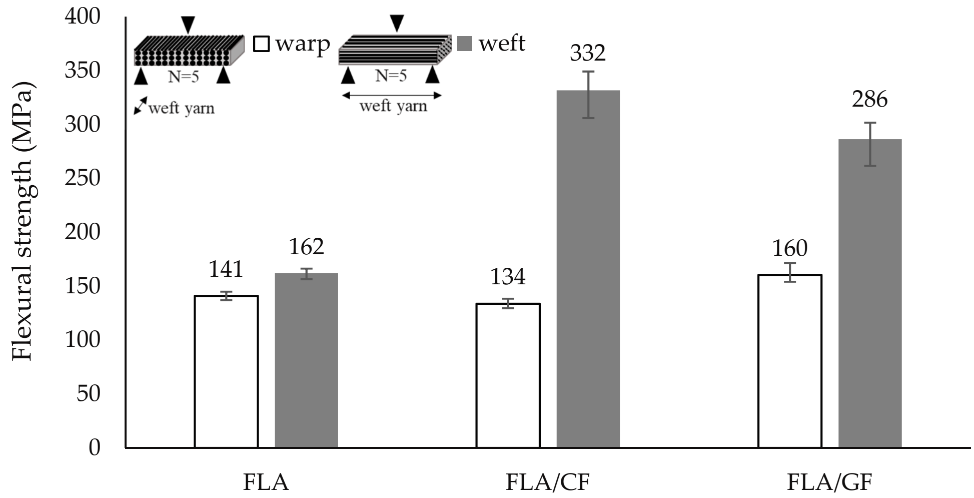

Figure 10. Flexural strength of hybrid composites.

The stress-strain curves of the twill weaved flax and hybrid composites, which are represented in Figure 11 show that in the warp direction all of the composites show brittle behavior. However, in the weft direction of the hybrid composites, the failure takes place in several stages. This is the direction, where synthetic fibers undergo flexural testing. This is probably the effect of the shear behavior, which is caused by the decreased span length during the testing.

Similarly, as in the case of the flax composite, the elimination of voids in the case of carbon-/flax composites would further improve the flexural properties.

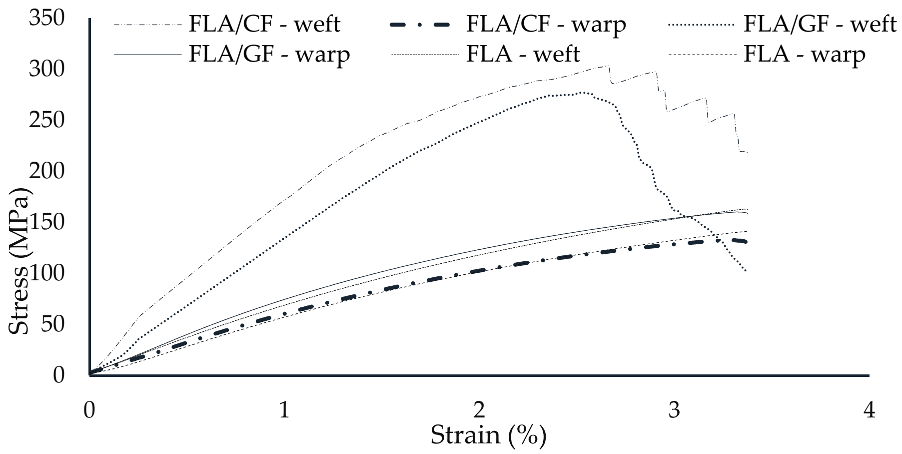

Figure 11. Stress-strain curves of the flax composite with twill weaved textile and hybrid textiles composing of flax and carbon as well as flax and glass fibers.

4. Conclusions

The aim of this study is to analyze the flexural properties of epoxy resin composites reinforced with textiles containing only flax fibers and hybrid textiles containing flax-/carbon and flax-/glass fibers. The textiles are integrated into biobased epoxy resin and analyzed with regard to the flexural properties in weft and warp directions. This study shows that the partial substitution of natural fibers with high-performance synthetic fibers enables manufacture of biocomposites with superior isotropic mechanical performance. The main findings show that by using a weaving technique, it is possible to manufacture hybrid textiles from natural and synthetic fibers with accurately adjusted mass fractions of fibers. Substitution of 50 wt %. of flax fibers with carbon or glass fibers leads to a significant improvement of flexural strength and modulus of the biocomposites. At the same time, further extensive research should be conducted in order to understand the process-technical behavior of the hybrid textiles during the composite manufacture. In particular, improvement of the infiltration and elimination of voids in composites would further improve flexural properties. Furthermore, several factors affect the mechanical properties of the analyzed composites simultaneously, namely, yarn type and fineness, yarn counts, number of plies in composite, and fiber volume fraction. Consequently, in order to investigate the trade-off between these parameters and the mechanical properties of the composites, a statistical approach should be pursued, such as a Design of Experiments or response surface methodology.

Author Contributions: Investigation, methodology, conceptualization and data curation (composite manufacture, mechanical testing, density measurement), visualization, writing—original draft preparation, review and editing, visualization, project administration, M.S.; investigation, methodology, conceptualization and data curation (weaving technique), writing—review and editing, visualization, J.W.; conceptualization of the project, funding acquisition, writing—review and editing, H.-J.E.

Funding: This study was implemented in the scope of the project "Functionally-integrated, three-dimensional variable production of bio-hybrid components with maximum bio-based content (ProBio)" funded by the Ministry for Science and Culture of the State of Lower Saxony (MWK).

Acknowledgments: The authors appreciate the assistance of Ricardo Wege from the Fraunhofer WKI, Application Center HOFZET for the fabrication of the textiles and Tim Federer for the conduction of the density measurements.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Faruk, O.; Bledzki, A.K.; Fink, H.-P.; Sain, M. Progress Report on Natural Fiber Reinforced Composites. *Macromol. Mater. Eng.* **2014**, *299*, 9–26. [CrossRef]
2. Mohammed, L.; Ansari, M.N.M.; Pua, G.; Jawaid, M.; Islam, M.S. A Review on Natural Fiber Reinforced Polymer Composite and Its Applications. *Int. J. Polym. Sci.* **2015**, *2015*, 1–15. [CrossRef]
3. Campilho, R.D.S.G. (Ed.) *Natural Fiber Composites*; Crc Press Taylor & Francis Group: Boca Raton, FL, USA; London, UK; New York, NY, USA, 2016; ISBN 978-1-4822-3900-3.
4. Dicker, M.P.M.; Duckworth, P.F.; Baker, A.B.; Francois, G.; Hazzard, M.K.; Weaver, P.M. Green Composites: A Review of Material Attributes and Complementary Applications. *Compos. Part A Appl. Sci. Manuf.* **2014**, *56*, 280–289. [CrossRef]
5. Hull, D.; Clyne, T.W. *An Introduction to Composite Materials*, 2nd ed.; Cambridge Solid State Science Series; Cambridge University Press: Cambridge, UK, 2001; ISBN 978-0-521-38855-9.
6. Prabhakaran, S.; Krishnaraj, V.; Kumar, M.S.; Zitoune, R. Sound and Vibration Damping Properties of Flax Fiber Reinforced Composites. *Procedia Eng.* **2014**, *97*, 573–581. [CrossRef]
7. Al-Oqla, F.M.; Sapuan, S.M. Natural Fiber Reinforced Polymer Composites in Industrial Applications: Feasibility of Date Palm Fibers for Sustainable Automotive Industry. *J. Clean. Prod.* **2014**, *66*, 347–354. [CrossRef]
8. Sanjay, M.R.; Arpitha, G.R.; Naik, L.L.; Gopalakrishna, K.; Yogesha, B. Applications of Natural Fibers and Its Composites: An Overview. *Nat. Resour.* **2016**, *7*, 108–114. [CrossRef]
9. Nunna, S.; Chandra, P.R.; Shrivastava, S.; Jalan, A.K. A Review on Mechanical Behavior of Natural Fiber Based Hybrid Composites. *J. Reinf. Plast. Compos.* **2012**, *31*, 759–769. [CrossRef]
10. Kc, B.; Faruk, O.; Agnelli, J.A.M.; Leao, A.L.; Tjong, J.; Sain, M. Sisal-Glass Fiber Hybrid Biocomposite: Optimization of Injection Molding Parameters Using Taguchi Method for Reducing Shrinkage. *Compos. Part A Appl. Sci. Manuf.* **2016**, *83*, 152–159. [CrossRef]

11. Dong, C. Review of Natural Fibre-Reinforced Hybrid Composites. *J. Reinf. Plast. Compos.* **2017**, *37*, 331–348. [CrossRef]
12. Davoodi, M.M.; Sapuan, S.M.; Ahmad, D.; Ali, A.; Khalina, A.; Jonoobi, M. Mechanical Properties of Hybrid Kenaf/Glass Reinforced Epoxy Composite for Passenger Car Bumper Beam. *Mater. Des.* **2010**, *31*, 4927–4932. [CrossRef]
13. Velmurugan, R.; Manikandan, V. Mechanical Properties of Palmyra/Glass Fiber Hybrid Composites. *Compos. Part A Appl. Sci. Manuf.* **2007**, *38*, 2216–2226. [CrossRef]
14. Flynn, J.; Amiri, A.; Ulven, C. Hybridized Carbon and Flax Fiber Composites for Tailored Performance. *Mater. Des.* **2016**, *102*, 21–29. [CrossRef]
15. Cherif, C. (Ed.) *Textile Material for Lightweight Constructions: Technologies—Methods—Materials—Properties*; Springer: Berlin/Heidelberg, Germany, 2016; ISBN 9783662463406.
16. Denninger, F. (Ed.) *Lexikon Technische Textilien*; Deutscher Fachverlag: Frankfurt am Main, Germany, 2009; ISBN 9783866410930.
17. Rattan, R.; Bijwe, J. Carbon Fabric Reinforced Polyetherimide Composites: Influence of Weave of Fabric and Processing Parameters on Performance Properties and Erosive Wear. *Mater. Sci. Eng. A* **2006**, *420*, 342–350. [CrossRef]
18. Huang, X.; Netravali, A. Characterization of Flax Fiber Reinforced Soy Protein Resin Based Green Composites Modified With Nano-Clay Particles. *Compos. Sci. Technol.* **2007**, *67*, 2005–2014. [CrossRef]
19. Felipe, R.C.T.D.S.; Felipe, R.N.B.; Batista, A.C.D.M.C.; Aquino, E.M.F. Polymer Composites Reinforced with Hybrid Fiber Fabrics. *Mater. Res.* **2017**, *20*, 555–567. [CrossRef]
20. Hufenbach, W.; Gude, M.; Ebert, C. Hybrid 3d-Textile Reinforced Composites with Tailored Property Profiles for Crash and Impact Applications. *Compos. Sci. Technol.* **2009**, *69*, 1422–1426. [CrossRef]
21. Priyanka, P.; Dixit, A.; Mali, H.S. High-Strength Hybrid Textile Composites with Carbon, Kevlar, and E-Glass Fibers for Impact-Resistant Structures. A Review. *Mech. Compos. Mater.* **2017**, *53*, 685–704. [CrossRef]
22. Shah, D.U. Characterisation and Optimisation of the Mechanical Performance of Plant Fibre Composites for Structural Applications. Ph.D. Thesis, University of Nottingham, Nottingham, UK, 2013.
23. *ISO 14125:1998: DIN EN ISO 14125 Fibre-Reinforced Plastic Composites—Determination of Flexural Properties German Version*; Beuth: Berlin, Germany, 2011.
24. Madsen, B.; Lilholt, H. Physical and Mechanical Properties of Unidirectional Plant Fibre Composites—An Evaluation of the Influence of Porosity. *Compos. Sci. Technol.* **2003**, *63*, 1265–1272. [CrossRef]
25. Koh, R.; Madsen, B. Strength Failure Criteria Analysis for a Flax Fibre Reinforced Composite. *Mech. Mater.* **2018**, *124*, 26–32. [CrossRef]
26. Altenbach, H.; Altenbach, J.; Kissing, W. *Mechanics of Composite Structural Elements*; Springer (Engineering Online Library): Berlin, Germany, 2004; ISBN 3-540-40865-7.
27. Amiri, A.; Triplett, Z.; Moreira, A.; Brezinka, N.; Alcock, M.; Ulven, C.A. Standard Density Measurement Method Development for Flax Fiber. *Ind. Crop. Prod.* **2017**, *96*, 196–202. [CrossRef]
28. Le Gall, M.; Davies, P.; Martin, N.; Baley, C. Recommended Flax Fibre Density Values for Composite Property Predictions. *Ind. Crop. Prod.* **2018**, *114*, 52–58. [CrossRef]
29. Yuhazri, M.Y.; Amirhafizan, M.H.; Abdullah, A.; Sihombing, H.; Saarah, A.B.; Fadzol, O.M. The Effect of Various Weave Designs on Mechanical Behavior of Lamina Intraply Composite Made from Kenaf Fiber Yarn. *Mater. Sci. Eng.* **2016**, *160*, 12021. [CrossRef]
30. Xie, Y.; Hill, C.A.S.; Xiao, Z.; Militz, H.; Mai, C. Silane Coupling Agents Used For Natural Fiber/Polymer Composites: A Review. *Compos. Part A Appl. Sci. Manuf.* **2010**, *41*, 806–819. [CrossRef]
31. Sharma, M.; Gao, S.; Mäder, E.; Sharma, H.; Wei, L.Y.; Bijwe, J. Carbon Fiber Surfaces and Composite Interphases. *Compos. Sci. Technol.* **2014**, *102*, 35–50. [CrossRef]
32. Alander, P.; Lassila, L.V.J.; Vallittu, P.K. The Span Length and Cross-Sectional Design Affect Values of Strength. *Dent. Mater. Off. Publ. Acad. Dent. Mater.* **2005**, *21*, 347–353. [CrossRef] [PubMed]

33. Cerbu, C. Mechanical Characterization of the Flax/Epoxy Composite Material. *Procedia Technol.* **2015**, *19*, 268–275. [CrossRef]
34. Pothan, L.A.; Mai, Y.W.; Thomas, S.; Li, R.K.Y. Tensile and Flexural Behavior of Sisal Fabric/Polyester Textile Composites Prepared By Resin Transfer Molding Technique. *J. Reinf. Plast. Compos.* **2008**, *27*, 1847–1866. [CrossRef]