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Bio-Based Composites for Sound Absorption

Eulalia Gliścińska, Izabella Krucińska, Marina Michalak, Michał Puchalski, Danuta Ciechańska, Janusz Kazimierczak and Arkadiusz Bloda

Additional information is available at the end of the chapter

http://dx.doi.org/10.5772/65360

Abstract

The acoustic thermoplastic composites and a method for their production with the participation of the bio-components were presented. To form composite matrix polylactide fibres (PLA) were used. Natural fibres (flax (LI) and cotton (CO)), straw and cellulose ultra-short/ultra-fine fibres obtained from biomass were used as a reinforcement. Cellulose ultra-short/ultra-fine fibres were obtained from the flax fibres or straw by enzymatic treatment and optionally modified by silane. The tensile stress at maximum load of composites with the sub-microfibres obtained from waste flax fibres after silane modification is twice higher than that of the composite with the sub-microfibres without the silane modification.

The effect of different kinds of natural materials on the acoustic composites was studied. The addition of the straw increases the values of the sound absorption coefficient are higher because of the additional voids caused by the particles of straw. If as a reinforcement the CO fibres and cellulose sub-microfibres are used, the sound absorption of the composite is higher than for composite with only CO fibres. In the case of sub-microfibres obtained from the waste flax fibres the highest sound absorption and tensile stress of the composites gives the modification by solution of silane in ethanol and water.

Keywords: biomass, composite, sound absorption, fibre
1. Introduction

Composites are more and more popular products present in our lives. An extensive use of composites results from the diversity of their functional properties, what is connected with the combination of different components, diversified in terms of materials and forms, and with diversity of the final structures. Recently, the composites have been produced from renewable and sustainable materials. Renewable materials are natural, environmentally friendly and usually cheap. The composites produced from such materials are called ‘green composites’, but this concept is much broader and should concern their production and usage. Now the filling components in composites should be waste and the matrix material should be recyclable such as, for example, thermoplastics. Sustainability concerns three aspects: environmental, economic and social. The use of biomass, paper, fibres, wood as a waste filling material and bio-based thermoplastic polymers as a matrix can be the optimal material’s solution [1].

The production of environmentally friendly composites on the basis of materials obtained from renewable resources is important for the economy and the environment. Recently, new materials on the basis of different plants are more and more popular and used in many industrial and life fields. Materials on the basis of straw, reeds, cattails and bent grass stalks are used in the ecological building sector [2]. The natural fibres can be used as a reinforcement or as a filling in composites, but also can give some new functions to them. Moreover, there is observed a growing trend towards replacing high-modulus reinforcing fibres with natural fibres [3–5]. Many technologies of composites based on different plants, natural fibres and even fibres isolated from plants are developed. The cellulose micro- or nanofibres, or micro-fibrils can be obtained through the chemical, mechanical, ultrasonic and enzymatic treatment of plants, such as jute [6, 7], soya bean source [8], wheat straw [9], soy hulls [10], rice straw [11], regenerated wood fibres [12] and canola straw [13].

Due to the great variety of plants, the properties of composites made of natural components are much diversified what favors different applications. Natural fibre-reinforced composites are used not only for construction but also for new uses, e.g., attenuation of sounds. Sound-absorbing composites can show a high degree of sound absorption, especially at high frequencies [14–19].

The commercially available porous sound-absorbing materials are usually fibrous. The fibres used are mostly synthetic, but recently, natural fibres are more and more popular as a raw material of the sound-absorbing products. Natural fibres are biodegradable and safer for human health than most mineral or polymer synthetic fibres. Introduction of the cellulose ultra-short/ultra-fine fibres prepared from different kinds of biomasses into functional composite structures causes increase in their sound absorption. Conversion of biomass to ultrashort/ultra-fine fibres perfectly fits into the current trends of cellulose nanostructures receiving by a top-down method. Depending on the size of the structures the obtained cellulose is suitably named, for example: microcrystalline cellulose, nanocrystalline cellulose or nanowhiskers.
Ultra-short/ultra-fine fibres, i.e., fibres not only extremely thin, but also extremely short, can be obtained by the enzymatic treatment of flax fibres and different kinds of straw. This form of fibres provides a greater surface area but causes application problems different from the case of electrospun filament fibres. A dust form requires direct screening of fibres onto the substrate. The use of ultra-short/ultra-fine fibres means larger area than in the case of longer fibres, in particular standard fibres. This form of fibres is advantageous from sound absorption point of view. The energy of the sound wave propagating in the material is reduced, and the internal energy of the material increases. Sound waves cause vibration of the fibres in the material and as a result of friction the created energy is converted into heat. A larger fibre surface promotes greater energy loss of the sound wave [16].

The virgin straw can also be used in sound-absorbing composites as an absorption enhancer. Independent of straw type, the values of the sound absorption coefficient increase because of the additional voids caused by the particles of straw and internal channels [20].

The aim of this work is to develop acoustic thermoplastic composites and the method for their production with the participation of the above-mentioned bio-based components. The effect of different kinds of natural materials on the acoustic composites was studied. Both waste natural fibres, straw and cellulose ultra-short/ultra-fine fibres, obtained from biomass can be used as an acoustic component increasing sound absorption of the composites.

In this work, the thermoplastic composites were obtained on the basis of textiles, i.e., nonwovens and above-mentioned natural materials in a thermal pressing process.

2. Materials

In order to thermally connect the composite constituents, one of them, forming the composite matrix, should be thermoplastic. The other ones are reinforcing/filling constituents.

To have a broader view of the acoustic properties of the composites the different raw materials and different combinations of reinforcing materials were used.

As a matrix material, the polylactide (PLA) fibres (6.7 dtex/64 mm) with a melting point in the range of 165–170°C have been selected. These fibres, Ingeo Fiber type SLN2660D, finished with polylactide resin without any hazardous substances, were delivered by Far Eastern Textile Ltd., Taipei, Taiwan.

As a reinforcement the following materials were used:

(a) waste natural fibres, Figure 1:
- flax fibres delivered by Safilin Ltd., Milakowo, Poland,
- Tadzhikistan cotton fibres 1.7 dtex/11 mm in the form of noils, delivered by Alto Ltd. (Gorzów Śląski, Poland), quality/degree of maturity is III/1.66 in a scale from 0 to 5.
(b) straws:

Three kinds of straw: grinded sunflower straw (SS), grinded corn straw (CS) and random cut wheat straw (WS) were cultivated and prepared by University of Agriculture in Cracow, Poland. The reinforcing materials are presented in Figure 2.

![Figure 1. Photographs of the waste natural fibres reinforcement.](image)

![Figure 2. Photographs of the straw reinforcement.](image)

Each kind of reinforcing material is characterized by diversified length and width. The length results from the random cutting process. The width is dependent on the native fibre or straw stem. Moreover, in the case of sunflower straw, two kinds of the particles can be distinguished, oblong particles coming from a hard envelope and more rounded particles coming from the soft inner parts. The corn straw particles are similar to sunflower ones but with smaller part of soft almost round particles. The wheat straw particles have the hollow channels.

(c) cellulose ultra-short/ultra-fine fibres obtained from:
- flax fibres 5.2 dtex/102 mm in the form of bleached roving, delivered by Safilin Ltd., Milakowo, Poland,

- straw of retted fibre flax (SRFF), straw of oil flax (SOF) and hemp straw (HS) delivered by University of Agriculture in Cracow, Poland,

as a result of different processes depending on raw materials.

In order to obtain cellulose ultra-short/ultra-fine fibres, the flax fibres or straw were cut to the pieces of several centimetres and treated by enzyme preparation. The enzymatic treatment causes the decrease in length and fineness of starting materials. The fibres obtained, called as sub-microfibres, were dried at an ambient temperature and ground in a disk mill and then treated or not by 3-aminopropyltriethoxysilane STRUKTOL® SCA 1100 (Struktol Company of America, USA). That modification usually is used to increase adhesion of fibres to a hydrophobic matrix. In this case, silane modification was used also to prevent fibres connection in the polymer matrix. In the case of sub-microfibres obtained from waste flax fibres, two kinds of silane modifications were used, i.e., by 10 wt% solution of 3-aminopropyltriethoxysilane in ethanol or by 10 wt% solution of 3-aminopropyltriethoxysilane in mixture of ethanol and water to obtain additional activation. The surface of sub-microfibres obtained from straw was modified by 10 wt% solution of 3-aminopropyltriethoxysilane in acetone. That modification causes an agglomeration of sub-microfibres.

After silane modification, the cellulose sub-microfibres were ground in a disk mill. The sub-microfibres obtained from waste flax fibres and straw are presented in Figures 3 and 4, respectively.

![Figure 3. SEM images of cellulose sub-microfibres from waste flax fibres.](Image)

Characteristics of all cellulose sub-microfibres, with and without silane modification, are presented in Table 1.

The silane modification of the cellulose sub-microfibres obtained from waste flax fibres has not influence their morphological properties. The fineness of the sub-microfibres without modification is 11.36 μm and the length is 146.55 μm. The fineness of the sub-microfibres after the silane modification in solution of silane in ethanol is 12.28 μm and the fineness of the sub-microfibres after modification in solution of silane in mixture of ethanol and water is 13.46 μm. The length of the sub-microfibres after modification is 117.21 μm and 138.83 μm, corre-
spondingly. Because of high values of variation coefficients of these parameters, the above differences are non-essential. The silane modification results in increase in the value of degree of crystallinity of cellulose sub-microfibres by about 6% apart from the conditions of silane modification. The degree of crystallinity of sub-microfibres without the silane modification is 79% and after the silane modification takes the value above 85%.

Figure 4. SEM images of cellulose sub-microfibres from straw (straw of retted fibre flax, SRFF, straw of oil flax, SOF, hemp straw, HS).

<table>
<thead>
<tr>
<th>Symbol of sub-microfibres</th>
<th>Silane modification of sub-microfibres</th>
<th>Fineness</th>
<th>Length</th>
<th>Degree of crystallinity</th>
</tr>
</thead>
<tbody>
<tr>
<td>LI-1</td>
<td>Without</td>
<td>11.36</td>
<td>81.0</td>
<td>146.55</td>
</tr>
<tr>
<td>LI-2</td>
<td>Silane in ethanol</td>
<td>12.28</td>
<td>37.5</td>
<td>117.21</td>
</tr>
<tr>
<td>LI-3</td>
<td>Silane in ethanol+water</td>
<td>13.46</td>
<td>35.7</td>
<td>138.83</td>
</tr>
<tr>
<td>SRFF-1</td>
<td>Without</td>
<td>5.62</td>
<td>48.40</td>
<td>70.00</td>
</tr>
<tr>
<td>SRFF-2</td>
<td>Silane in acetone</td>
<td>1.64</td>
<td>56.7</td>
<td>1.61</td>
</tr>
<tr>
<td>SOF-1</td>
<td>Without</td>
<td>11.09</td>
<td>46.80</td>
<td>89.07</td>
</tr>
<tr>
<td>SOF-2</td>
<td>Silane in acetone</td>
<td>8.94</td>
<td>55.06</td>
<td>46.35</td>
</tr>
<tr>
<td>HS-1</td>
<td>Without</td>
<td>6.33</td>
<td>40.13</td>
<td>55.24</td>
</tr>
<tr>
<td>HS-2</td>
<td>Silane in acetone</td>
<td>2.43</td>
<td>36.67</td>
<td>19.64</td>
</tr>
</tbody>
</table>

Table 1. Characteristics of cellulose sub-microfibres [21].
The silane modification of the cellulose sub-microfibres obtained from different kinds of straw causes the reduction of fibre dimensions. The fineness of the sub-microfibres from the SRFF straw without silane modification is 5.62 μm, from the SOF straw it is 11.09 μm and from HS it is 6.33 μm. The fineness of the sub-microfibres from SRFF straw after silane modification is 1.64 μm, from SOF straw it is 8.94 μm and from HS it is 2.43 μm. The length of the sub-microfibres after the enzymatic treatment is 70.00 μm for SRFF, 89.07 μm for SOF, and 55.24 μm for SOF straw. The length of the sub-microfibres from straw after silane modification is 16.1 μm for SRFF straw, 46.35 μm for SOF straw and 19.64 μm for HS straw. The values of variation coefficients of these parameters are very high. The silane modification has no significant influence on the degree of crystallinity of the cellulose sub-microfibres obtained from SOF and HS straws. For these sub-microfibres, the values of the degree of crystallinity change from 63.2 to 61.3% for SOF straw and from 65.8 to 65.3% for HS straw. In the case of sub-microfibres obtained from SRFF straw, the values of the degree of crystallinity are lower by 9% after silane modification.

The dimensions of the cellulose sub-microfibres obtained from flax fibres can be further reduced during the mechanical treatment in an aqueous suspension by means of a homogenizer. The length decreases to about 6 μm and diameter to about 300 nm. These fibres, Figure 5, referred to as sub-micro/nanofibres, were not dried to prevent their agglomeration.

![SEM image of the sub-micro/nanofibres from flax fibres](image)

**Figure 5.** SEM image of the sub-micro/nanofibres from flax fibres [16].

### 3. Manufacturing of composites

Thermoplastic composites were obtained from the multilayer structures in a hydraulic press machine using a water-cooling system from Hydromega (Poland). The press conditions for all multilayer structures were the same, i.e., temperature 170–175°C, time 5 min and pressure 0.275
MPa. The multilayer structure was wrapped with Teflon foil to prevent molten polymer propagation during the pressing process.

It was assumed to obtain composite samples with similar thicknesses to allow for assessment of the effect of the reinforcement type. Each multilayer structure was composed of several layers of nonwovens either separated alternately or not by layers of grinded straw or cellulose ultra-short/ultra-fine fibres. The appropriate mass of straw/cellulose ultra-short/ultra-fine fibres was divided in portions, which were put uniformly onto consecutive layers of nonwoven and the top of multilayer structure was covered by nonwoven.

The needle-punched nonwovens were manufactured from matrix fibres ‘PLA (100%)’ or from a blend consisting of matrix fibres ‘PLA’ and reinforcing waste flax fibres ‘LI’ or waste cotton fibres ‘CO’.

In order to manufacture needle-punched nonwovens the fleeces with a cross-system of fibre arrangement were obtained on the roller card. Needle punching of the fleece layers was carried out on an Asselin needle punching machine (France), with the following technological parameters: type of needles – 15 × 18 × 40 × 3 1/2 RB (Groz-Beckert®), number of needle punches – 40/cm², depth of needle punching – 12 mm [16].

4. Methods

The thickness of composites was determined according to the standard procedure ISO 9073-2:1995 and their apparent density was determined as the ratio of mass to volume of samples. The mechanical properties of composites were estimated in unidirectional tensile test by means of testing machine type 3119-410 (Instron, UK) according to the standard ISO 527-4:1997. The sound absorption coefficient was determined according to the standard procedure ISO 10534-2:1998 within the frequency range of 500–6400 Hz. A small size impedance tube (Kundt tube) type 4206 (Brüel & Kjaer, Denmark) was used (Figure 6). The diameter of the investigated samples was equal to 29 mm.

Figure 6. The Kund device for acoustic measurements.
5. Results

The values of mass per square meter were similar for all nonwovens, i.e., in the range of 115–120 g/m². The composites obtained were rigid with a small thickness of a few millimetres, ‘green’, and designed among others from cheap components coming from renewable resources. Each composite was manufactured from the system consisting of 80 wt% of nonwoven (PLA (100%) or PLA/LI (80/20%) or PLA/CO (90/10%)) and 20 wt% of straw or cellulose ultra-short/ultra-fine fibres.

5.1. Composite structure: organoleptic assessment

The most uniform and homogeneous structure is observed for matrix materials. In the case of the composites, the structure depends on the reinforcement type. The photographs in Figure 7 present the structure of the selected composites.

![Figure 7. Photographs of the composites.](http://dx.doi.org/10.5772/65360)

If the fibres are used as a reinforcement, the composite structure is more uniform. It results from the uniform structure of nonwoven composed of reinforcing and matrix fibres and from...
their high mixing level. The external and internal layers of nonwoven are pressed giving uniform, homogeneous porous structure. On the composite surface, the reinforcing fibres, matrixes and voids are visible. If the fibres and straw are used as a reinforcement, the composite structure is more diversified. The addition of straw particles leads to the formation of larger voids inside the composite structure. If only the straw is used as a reinforcement, the layers of matrix and straw can be visible. Inside the composite structure, the straw particles are partially bound with the matrix, and the voids exist. The composite surfaces are formed from the matrix without visible voids. Generally, the straw gives larger voids than the fibres and the larger straw particles give larger voids. In the case of composite, on the basis of the wheat straw, the additional voids are inside the straw particles. In the case of composites with cellulose ultra-short/ultra-fine fibres, the variants with standard fibres give more uniform structures than without, because standard fibres prevent their agglomeration.

5.2. Physical and mechanical properties

The physical properties as a thickness and apparent density and mechanical properties as a tensile stress at the maximum load are presented in Table 2. The thickness and density of the material are the most important factors determining its acoustic properties [21]. In this work, it was assumed to obtain composites of very low thickness and low density. A small thickness of millimetres is important when the composite material is combined with other materials. The low density is important to produce light materials.

5.2.1. Composites on the basis of flax fibres and straw

The thickness of the composites is in the range from 3.52 to 4.71 mm. The thickness of the PLA/straw composites is higher on average by 26.90% than thickness of the PLA/LI/straw composites, and because of that the density is correspondingly lower. The tensile stress at maximum load of the composites depends on a reinforcement type. The highest value of tensile stress, i.e., 2.69 MPa, is determined for the PLA/LI composite characterized by the medium value of apparent density. If the matrix material is used in the form of fibres, their mixing with reinforcing fibres can be excellent, especially in the needle-punched nonwoven. It results in the higher tensile stress of the composite. The lower mixing level of the thermoplastic fibres with straw particles, or with straw particles and reinforcing fibres, gives lower tensile stress of the composite. It means that flax fibres are better reinforcing materials than straw and the level of mixing of reinforcing and matrix materials in nonwoven is high what leads to suitable consolidation. This is confirmed by the PLA/LI/straw composites, for which the values of tensile stress at maximum load are higher than for the PLA/straw composites and lower than PLA/LI composites. The differences in apparent density of the composites have no significant effect on the tensile stress [20].

5.2.2. Composites on the basis of cotton fibres and cellulose ultra-short/ultra-fine fibres

The composites obtained (Table 2) were characterized by similar, very low thickness on the level of about 4.7–5.5 mm. These differences in the thickness of composites are insignificant for their acoustical properties [16].
<table>
<thead>
<tr>
<th>Variant of the composite</th>
<th>Thickness (mm)</th>
<th>Apparent density (kg/m³)</th>
<th>Tensile stress at maximum load (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PLA</td>
<td>3.80</td>
<td>1321.00</td>
<td>–</td>
</tr>
<tr>
<td>PLA/Li (80/20%)</td>
<td>3.92</td>
<td>313.43</td>
<td>2.69</td>
</tr>
<tr>
<td>PLA/CO (90/10%)</td>
<td>4.5</td>
<td>264.32</td>
<td>0.79</td>
</tr>
<tr>
<td>PLA + straw</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>PLA/SS</td>
<td>4.65</td>
<td>271.76</td>
<td>1.12</td>
</tr>
<tr>
<td>PLA/CS</td>
<td>4.65</td>
<td>276.20</td>
<td>1.22</td>
</tr>
<tr>
<td>PLA/WS</td>
<td>4.71</td>
<td>258.63</td>
<td>1.54</td>
</tr>
<tr>
<td>PLA + Li + straw</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>PLA/Li/SS</td>
<td>3.77</td>
<td>393.60</td>
<td>1.98</td>
</tr>
<tr>
<td>PLA/Li/CS</td>
<td>3.52</td>
<td>341.22</td>
<td>1.75</td>
</tr>
<tr>
<td>PLA/Li/WS</td>
<td>3.75</td>
<td>337.75</td>
<td>1.60</td>
</tr>
<tr>
<td>PLA + CO + sub-microfibres from Li</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Li-1</td>
<td>5.3</td>
<td>260.61</td>
<td>1.20</td>
</tr>
<tr>
<td>Li-2 silane in ethanol</td>
<td>4.9</td>
<td>287.57</td>
<td>2.27</td>
</tr>
<tr>
<td>Li-3 silane in ethanol and water</td>
<td>4.8</td>
<td>299.87</td>
<td>2.27</td>
</tr>
<tr>
<td>PLA + CO + sub-microfibres from straw</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SRFF-1</td>
<td>5.4</td>
<td>267.30</td>
<td>1.29</td>
</tr>
<tr>
<td>SRFF-2 silane in acetone</td>
<td>4.8</td>
<td>299.87</td>
<td>1.79</td>
</tr>
<tr>
<td>SOF-1</td>
<td>4.9</td>
<td>242.42</td>
<td>1.04</td>
</tr>
<tr>
<td>SOF-2 silane in acetone</td>
<td>4.7</td>
<td>273.39</td>
<td>1.13</td>
</tr>
<tr>
<td>HS-1</td>
<td>5.5</td>
<td>236.01</td>
<td>1.19</td>
</tr>
<tr>
<td>HS-2 silane in acetone</td>
<td>4.8</td>
<td>287.17</td>
<td>1.54</td>
</tr>
</tbody>
</table>

SS, grinded sunflower straw; CS, grinded corn straw; WS, random cut wheat straw.

Table 2. Characteristics of the composites [20, 21].

For studied composites, the 20% addition of cellulose sub-microfibres results in increase in their mechanical properties. The composite manufactured under the same technological conditions but without the addition of sub-microfibres (PLA/CO) has the lowest tensile stress at maximum load equals to 0.79 MPa. The results show the relationship between the mechanical properties of the composites with cellulose sub-microfibres and the method of the sub-microfibres preparation.

For the composites with sub-microfibres obtained from waste flax fibres, the tensile stress at maximum load is diversified depending on the sub-microfibres preparation. The tensile stress at maximum load for the composite manufactured with sub-microfibres obtained from waste
flax fibres gains the value of 1.20 MPa and for the composites with sub-microfibres additionally modified by silane (LI-2, LI-3), the value of 2.27 MPa is twice higher apart from the conditions of silane modification. These results correspond to the degree of crystallinity of the cellulose sub-microfibres. The same relationship is observed for the composites manufactured with sub-microfibres from straws, after silane modification the cellulose sub-microfibres give better reinforcement than without silane modification. The values of tensile stress at maximum load of the composites with sub-microfibres obtained from each kind of straw after silane treatment are from 9.05 to 38.76% higher than the values of tensile stress of composites with corresponding sub-microfibres obtained without silane modification. The best results of tensile stress are observed for composites with sub-microfibres obtained from SRFF straw.

The tensile stress at maximum load for the composites manufactured with sub-microfibres obtained from the straw of retted fibre flax is 1.29 MPa and for the composites with sub-microfibres additionally modified by silane (SRFF-2) reaches the value of 1.79 MPa. It means that silane modification gives favorable conditions for adhesion between the cellulose sub-microfibres and the PLA matrix. It results advantageously in tensile strength of composites. This effect is more distinct for the composites with sub-microfibres from flax fibres than with sub-microfibres from straws. The tensile strength is the highest for composites with sub-microfibres obtained from waste flax fibres and modified by silane (LI-2, LI-3). Both the silane modification favoring good adhesion to polymer and a high degree of crystallinity of sub-microfibres resulted in the highest values of tensile stress at maximum load for these composites.

In the case of apparent density, which is a very important factor deciding about sound absorption, the differences are more considerable. Density of the composites with cellulose sub-microfibres obtained from waste flax fibres and modified by silane with additional water activation (LI-3) is higher by 4% than the density of the composites with the same cellulose sub-microfibres but modified by silane without water activation (LI-2), and by 15% higher than the density of the composites with the sub-microfibres not modified by silane (LI-1).

Density of the composites with cellulose sub-microfibres obtained from straws and modified by solution of silane in acetone is higher by 12.1% for SRFF straw, 12.8% for SOF straw, and 21.6% for HS straw than the density of the corresponding composites with sub-microfibres not modified by silane [22]. For the studied composites, these differences in material density are not significant from their sound absorption point of view [16].

5.3. Acoustic properties

Acoustic properties were characterized by sound absorption coefficient, which values can be in the range from 0 (total reflection) to 1.00 (total absorption). According to EN ISO 11654, low absorption is from 0.15, absorption from 0.30, high absorption from 0.60 and very high absorption from 0.80.

For studied materials not only the value of sound absorption coefficient is important, but also the sound frequency range of high absorption.
5.3.1. Composites on the basis of flax fibres and straw

The biomass is very diversified and gives a very diversified effect from the acoustical point of view. Figures 8–10 present the most characteristic dependences of the sound absorption coefficient-sound frequency for PLA materials, PLA/LI fibres, PLA/straw and PLA/LI fibres/straw composites. Among studied composites, the highest values of the sound absorption coefficient, even in the range of 0.8–0.9, are determined for the PLA/straw composites [20].

Figure 8. Sound absorption coefficient of the composite-waste flax fibres and sunflower straw as a reinforcement.

Figure 9. Sound absorption coefficient of the composites-waste flax fibres and corn straw as a reinforcement.
The sound absorption coefficient of the PLA matrix material is the lowest and has the value of about 0.1 at the low and mid-frequencies, at high frequencies it increases to 0.38 at 6400 Hz. This material cannot be used as a sound absorber of frequencies lower than 6000 Hz.

If nonwoven from blend of PLA matrix fibres and diversified waste flax fibres is used, the sound absorption of the composite is growing with frequency. The sound absorption coefficient of the composite from the ‘PLA/LI (80/20%)’ nonwoven is higher and depends linearly on a sound frequency arriving maximum value of 0.40 at a maximum studied frequency of 6400 Hz. This almost linear dependence is typical for fibre-based composites [19]. Even addition of particles thicker than fibres gives similar dependence, but with higher sound absorption. If the 20% of ‘PLA/LI (80/20%)’ nonwoven is replaced by straw, the sound absorption is more higher in the range from 3000 to 6400 Hz. It probably results from the fact that beside of small voids in composite, the bigger voids are formed. The sound waves of such frequencies can be reverberated repeatedly inside the diversified voids until soundproofing. The upward trend is evident and similar for all three kinds of composites with straw, as shown in Table 3. For the composite from a multilayer structure consisting of 80% of PLA/LI nonwoven and 20% of corn straw, the sound absorption coefficient increases linearly with sound frequency, and at 6400 Hz it is equal to 0.76. For the composites with sunflower or wheat straw as a reinforcement, the maximum value is 0.71 and 0.68 at 6400 Hz, respectively.

If PLA nonwoven and straw are used, the acoustic characteristic of the composite is totally different. The curve of the sound absorption coefficient-sound frequency dependence has another course. For each PLA/straw composite this course is slight different, because the kind of particle of each straw is slight different. Generally, the maximum values of sound absorption coefficient occur at medium (for PLA/SS and PLA/CS composites) and high (for PLA/WS composite) values of the studied frequency range. It means that the PLA/straw composites can be used as a material absorbing the sound of medium and high frequencies. The maximum values of the sound absorption coefficient of the PLA/straw composites are higher than those of PLA/LI or PLA/LI/straw composites. The sound absorption coefficient reaches the values of
For the PLA/straw composites, the frequency range of the maximum values of the sound absorption coefficient is wider than for PLA/LI or PLA/LI/straw composites.

<table>
<thead>
<tr>
<th>Variant of the composite</th>
<th>Sound absorption coefficient</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Frequency (Hz)</td>
</tr>
<tr>
<td></td>
<td>1000</td>
</tr>
<tr>
<td>PLA/LI</td>
<td>0.061</td>
</tr>
<tr>
<td>PLA/LI/SS</td>
<td>0.067</td>
</tr>
<tr>
<td>PLA/LI/CS</td>
<td>0.071</td>
</tr>
<tr>
<td>PLA/LI/WS</td>
<td>0.059</td>
</tr>
</tbody>
</table>

Table 3. The sound absorption coefficients of the PLA/LI/straw composites.

All of the obtained composites of such small thickness and apparel density show the sound absorption, wherein the values of the sound absorption coefficient depend on the reinforcement type and sound frequency.

5.3.2. Composites on the basis of cotton fibres and cellulose ultra-short/ultra-fine fibres

The CO fibres used as a reinforcement give better effect on sound absorption of the PLA composites than cellulose sub-microfibres. If as a reinforcement the CO fibres and cellulose sub-microfibres are used the sound absorption is more better. Increase in sound absorption over the whole range of investigated frequency is observed for 10% and optimal for 20% by ieswt. sub-microfibre content. The addition of sub-microfibres changes the character of the curve of the sound absorption coefficient in a function of sound frequency (Figure 11).

Figure 11. Sound absorption coefficient of the composites with/without sub-microfibres.
Among composites with sub-microfibres obtained from flax fibres, the composites with fibres without silane (LI-3) are characterized by the highest sound absorption, Figure 12, and the highest tensile stress at maximum load, Table 2 [21]. Among studied composites with cellulose sub-microfibres obtained from straws, the best results, similar to composites LI-1, are observed for composites with sub-microfibres from SOF straw (SOF-1), Figure 13.

![Figure 12](image1.png)

**Figure 12.** The sound absorption coefficient in the function of sound frequency for the composites: 1, LI-1 (without silane); 2, LI-2 (silane in ethanol); and 3, LI-3 (silane in ethanol and water).

![Figure 13](image2.png)

**Figure 13.** The sound absorption coefficient in the function of sound frequency for the composites: 1, SRFF-1; 2, SOF-1; 3, HS-1.

For the composites with sub-microfibres obtained from straws and then modified by solution of silane in acetone, the best results of the sound absorption are observed for the composites with sub-microfibres obtained from SRFF straw (SRFF-2), Figure 14. The silane modification
of sub-microfibres obtained from SOF and HS straws does not change the sound absorption coefficient of the composites with them. In the case of SRFF, the difference between the sound absorption of composites with sub-microfibres modified by silane and without modified by silane is the highest, Figures 15–17. The composites with sub-microfibres obtained from SRFF straw modified with silane (SRFF-2) are characterized by the best results in sound absorption and tensile stress among the composites with sub-microfibres obtained from straws.

Figure 14. The sound absorption coefficient in the function of sound frequency for the composites: 1, SRFF-2; 2, SOF-2; 3, HS-2.

Figure 15. The sound absorption coefficient in the function of sound frequency for the composites with SRFF sub-microfibres: 1, SRFF-1; 2, SRFF-2.
Figure 16. The sound absorption coefficient in the function of sound frequency for the composites with SOF sub-microfibres: 1, SOF-1; 2, SOF-2.

Figure 17. The sound absorption coefficient in the function of sound frequency for the composites with HS sub-microfibres: 1, HS-1; 2, HS-2.

The silane modification of sub-microfibres obtained from straws gives the best effect for SRFF straw because of the highest increase in tensile stress and in sound absorption coefficient. The sound absorption coefficient of the SRFF-1 composite was the lowest among all composites with sub-microfibres without silane modification but absorption coefficient of SRFF-2 composite is the highest in the whole range of investigated frequencies. The increase in stress at maximum load of the SRFF-2 composite is the highest among other composites with sub-microfibres from straws and is 38.8% higher than that of SRFF-1 composite.
For composites with sub-microfibres obtained both from waste flax fibres and from straws, the silane modification of sub-microfibres gives probably higher arrangement of fibres and their better adhesion to the polymer matrix, what is observed in high sound absorption and high tensile stress. In the case of sub-microfibres obtained from waste flax fibres, the highest sound absorption and the highest tensile stress of the composites give the modification by solution of silane in ethanol and water.

In the case of the composites with cotton fibres and sub-micro/nanofibres, Figure 18, the sound absorption increases rapidly to a value of 0.8 in the range of 500–4000 Hz and in the range of 4000–6400 Hz slightly increases. The absorption coefficient at a frequency of 6400 Hz is equal to 0.95. If the content of CO fibres was increased to 50%, the sound absorption coefficient increases in the range of 500–2500 Hz to approximately 0.8, and in the range of 4000–6000 Hz it is almost constant.

Figure 18. Sound absorption coefficient of the composites: 1, 80%PLA/CO (90/10) + 20% cel. Sub-micro/nano; 2, 80%PLA/CO (50/50) + 20% cel. Sub-micro/nano [16].

Results show that sound absorption of the composite depends on the content of reinforcing fibres. For higher percentage of these fibres, the higher sound absorption is observed. The sound wave causes the fibres vibration and as a result of friction the created energy of sound wave is converted to heat conversion. Larger total fibre surface leads to greater interaction of sound wave with the fibres. This effect of interaction between sound wave and fibre surface is stronger if the standard fibres are replaced by ultra-short/ultra-fine fibres [16].

6. Conclusions

The study included not only development of a method for manufacturing sound-absorbing composites, but also determination of the requirements for the characteristics of reinforcing
materials, optimal from the point of view of sound absorption. A comparison was made to the raw material used, and its treatment, with respect to the sound absorption of the composite as well as its strength. Silane modification does not significantly affect change fibre dimensions, and the manner of its implementation and the origin of the fibres show no effect on the sound absorption of the composite. The use of fibres coated by silane preparation is preferred from the viewpoint of mechanical properties. Tensile strength of composites containing fibres coated with the silane preparation is about twice higher than that of the composites with the unmodified fibres.

Composites could be prepared from the layers of needled nonwoven formed from the matrix fibres or a mixture of matrix fibres and reinforcing standard fibres and layers of straw or ultra-short/ultra-fine fibres alternately arranged. In the case of composite reinforced by straw, the presence of the standard fibres is inadvisable. The addition of the straw, independent of straw type, increases the values of the sound absorption coefficient because of the additional voids caused by the particles of straw. Both grinded sunflower straw, grinded corn straw and random cut wheat straw reduce the tensile stress at maximum load of the composites, but increase the sound absorption even to 0.8–0.9 in some frequency ranges.

Studies have shown that the composite structure produced by compressing the fibrous layer system is most advantageous from the point of view of its sound absorption when the standard fibres are as a reinforcement with ultra-short/ultra-fine fibres. The addition of sub-microfibres in an amount of 10–20% by weight of this system gives the significant effect on the composite sound absorption in the entire tested range of sound frequencies, i.e., up to 6400 Hz. The rapid increase of the absorption coefficient to the value of approx. 0.8 is in the range of 500–4000 Hz, followed by a further but slower increase is observed. If, however, as ultra-short/ultra-fine fibres will be used sub-micro/nanofibres, obtained by additional mechanical treatment of sub-microfibres, giving further fragmentation, then this relationship may adopt a different character. For a composite containing approx. 50% of the standard fibres and 20% of sub-micro/nanofibres sound absorption coefficient reaches a high value of 0.75–0.80 even at lower frequencies, i.e., from 2500 Hz and maintains at such a high level at higher frequencies.

Studies have shown that the best effect on the sound absorption of the composite, i.e., high sound absorption coefficient over a wide frequency range, may be obtained using next the standard reinforcing fibres the ultra-short/ultra-thin fibres with a minimum dimensions, because the larger the surface of the fibres, the greater the impact of the sound wave with fibres, and therefore greater its attenuation.

The developed composites are thin (several mm thick), lightweight, rigid, washable, high sound-absorbing of middle and high frequencies, which can be produced from waste fibres and straw, and may be designed to eliminate sound from various sources, for example in devices, vehicles and other means of transport, and for sound absorption in the rooms.

These ecological composites have characteristics in line with European trends and can be used as a stand-alone screen or to fill sound-absorbing panels, as a sound-absorbing contribution under the plaster in the construction of walls and ceilings suspended or in silenced car door, hatch, wheel arches, headliner and trunk.
Acknowledgements

This work has been supported by the Project ‘Utilization of biomass for the preparation of environmentally friendly polymer materials’ (Biomass)—POIG.01.01.02-10-123/09.

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