PROPERTIES OF LONG FLAX FIBER REINFORCED NONWOVEN AND COMPOSITE MATERIALS

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Abstract. Nowadays, especially in the car industry, the demand of light construction usage increases. Combining natural fibres with polymer fibres like polylactide (PLA) as textile material the content makes it also appropriate for composite material production increasing the field of textile material usage. Conducting the present research long flax fibres (40 wt %) and polylactide (PLA) (60 wt %) fibres nonwoven material (NWM) samples of the so-called sandwich type have been developed. The objective of manufacture of NWM samples is their further conversion into a biocomposite material, where flax fibres perform the function of reinforcement and PLA fibres - of composite matrix. The NWM structure is formed by five fibre web layers. Preparation and formation of fibre web layers are done by the air-laid method, and for the manufacture of NWM the mechanical needle punching method is used. Composite samples have been manufactured by inserting a part of the prepared NWM strips into the thermal press. Manufacture and testing of the samples obtained during the research has been done at the RWTH Aachen University Institute of Textile Technology (RWTH ITA). Conducting the research NWM has been obtained with surface density between $1066-1261 \text{ g} \cdot \text{m}^{-2}$, the tensile strength of which varies from 0.03 to 0.05 MPa at the material elongation of 40-59 %. The surface density of the composite obtained from NWM varies in the range from 1035 to 1235 g m⁻², ultimate tensile strength 16.79 to 24.18 MPa at the material elongation 3.56 to 4.31 %. Upon 3-points bending the strength limit varies from 0.92 to 1.11 MPa at material elongation 0.04 %.

Keywords: flax, nonwovens, composites, polylactide.

Introduction

Demand for the use of lightweight structures in various industry sectors, among them also in automotive, is growing. By substituting the commonly used glass fibre with natural fibres in composites, it is possible to reduce the material mass. Combining natural fibres with polymer fibres (particularly those, which are obtained from renewable resources, e.g., PLA), in one material, textiles suitable for manufacture of composite materials can be made. The NWM and composite samples studied under the project have been produced and their physical and mechanical properties have been tested during Erasmus internship program at the RWTH Aachen University Institute of Textile Technology (RWTH ITA). The long flax fibres (40 wt %) and PLA fibres (60 wt %) at the disposal of ITA have been used for the manufacture of samples. The proportion of flax fibres and polymer fibres has been chosen based on the ITA (RWTH University) experience gathered previously in the manufacture of composites reinforced with natural fibres.

Materials and methods

Flax. Common flax or flaxseed (*Linum usitatissimum* L.) is a renewable resource and its crop can be harvested every year. In the Republic of Latvia flax is an ancient agrotechnical crop, grown by Latvians for production of fibre. In Latvia flax is more often grown traditionally in Latgale region as its soil is more suited for flax than for growing cereal crops.

Flax fibre has a wide range of application. It is used both in thermal insulation materials and composite materials, which are used for automotive, aviation, velo-, moto-, musical instruments and other components. The wide application of flax fibre is facilitated by its high tensile strength and small tensile deformation ensured mainly by cellulose contained in its composition (the formula of cellulose is (C6H10)5)n), which is a natural polymer – macromolecular compound with a high polymerization level (300-3000). The cellulose content in flax fibre depending on different conditions is on average 60-80 % of the total fibre mass. Apart from cellulose flax fibres contain such substances as lignins, pectins, plant wax and fats, and different substances soluble in water and hygroscopic water.

The long flax fibres used for the manufacture of samples were tested at the ITA laboratory where the fibre denier was determined -3.87 dtex and the average fibre length -139.23 mm.

Polylactide fibres. PLA is an artificial biopolymer [1] formed by linear macromolecules. Their chain contains at least 85 wt % of lactic acid ester, which is produced from naturally occurring sugars.

The melting temperature is at least 135 °C [2; 3]. PLA is compostable, biologically compatible, as well as can be processed using standard equipment. PLA polymer degradation rate can be controlled – this property should be considered when producing different materials with different service lives.

The PLA fibres used for production of samples were delivered from the company Ingeo [4]. The information distributed by the company shows [5] that PLA is produced from corn. The technical parameters of the fibres used conducting the research are type SLN 2660D, specification: 6.0 dx 64 mm x FB, denier: 6.5 ± 0.5 den, fibre length: 64 ± 4 mm.

Nonwovens manufacturing. NWM allow for obtaining textile material with a wide range of functional characteristics: the porous structure of the material provides for heat and sound insulation and absorption characteristics. These properties are of particular importance for textile materials used for sound damping and regulation of heat transfer, e.g., in car parts.

Ratio of natural fibres and polymer was selected inspired by the experience of other researchers [6]. One of the selected examples were PLA fibre composites reinforced with hemp fibre (similar to flax fibre according to several characteristics) in different proportions. In the conclusions the composite with hemp fibre proportion 30 % of mass (PLA/Hemp30) [6] has been found to be the most effectual. The material demonstrated tensile strength as high as 65.92 ± 5.10 MPa at 1.51 ± 0.21 % material elongation. With the aim to increase the amount of flax fibres, in this research NWM has been developed where long flax fibres 40 wt % of mas are mixed with PLA fibres 60 wt % of mass.

The designed NWM structure consists of several fibre layers of which PLA fibre layers, without admixture of other fibres, are located at the outer sides. The experimental NWM is formed of five fibre web layers (Fig. 1). To manufacture one sample 771 g of pre-prepared fibres were used, out of which 330 g account for flax fibre and 441 g – PLA fibre.



Fig. 1. Schematic structure of nonwoven material

NWM manufacturing process consists of the following steps: fibre preparation (loosening), weighting of an appropriate fibre amount, fibre mixing using the air-laid process, preparation of fibre web layers, preparation of fibre web layers for bonding and layer bonding, arrangement of the bonded fibre web layers according to the desired NWM structure and manufacture of NWM.

NMW manufacturing process was determined by the possibilities of available techniques and their specifics: to obtain a homogenous fibre web, it is practical to divide the total fibre amount into several equal parts, which allows for variation of matrix and flax fibre proportions.

The equipment TRÜTZSCHLER CVT3 1200 was used for fibre loosening, mixing of both the types of fibre and to form fibre web layers. In the equipment measured amounts of fibre are delivered from the fibre feeding point to the vertical vacuum cylinder, which is attached to the equipment. In the equipment the fibres are mixed using the so called air-laid method. In the fibre web layer, which is obtained in the cylinder, the fibres are orientated in different random directions.

For obtaining NWM a mechanical method – needle-punching was chosen. In the process of NWM manufacture needle-punching was done in two stages, of which during the first needle-punching the fibre web layers were bonded and after the second needle-punching the ready material was produced. After the first needle-punching three bonded fibre web layers are obtained – the first fibre web layer (PLA fibre web layer) is needle-punched together with the second fibre web layer (web layer, in which plant and PLA fibres are mixed), the third fibre web layer is needle-punched separately, the fourth

fibre web layer (a layer in which plant and PLA fibres are mixed) is needle-punched together with the fifth fibre web layer (PLA fibre web layer). Before the second needle-punching the three previously bonded fibre web layers are to be arranged and composed according to the structure of the NWM. In both stages of needle-punching the equipment DILO LBM 6 was used. It has one needle board, which is moving vertically from top downwards. The fibre web layers, consisting of two bonded fibre web layers, are inserted into the needle-punching machine with the PLA fibre web layer on top, closer to the needle board. Needles 15X18X25X3 ½ R333 G 3007 produced by the company GROZ-BECKERT were used for the needle-punching process.

Carrying out the research three samples were manufactured, and six 50×300 mm specimen strips were cut from each sample. Of the six specimen strips four were cut in the machine direction and two in the cross-machine direction.

Composite manufacture. On the outer sides of NWM structure fibre layers consisting solely of PLA fibres are placed. Pure PLA fibre layers are made with the aim to form a protective layer of material when composites are manufactured. The structural composition of the produced composite material is similar to laminate. Traditionally laminate is considered to be a kind of composites made by mutually glued layers of material. Another well known explanation of the principle of producing laminate is related to its functionality – making sheet materials more rigid and protecting them from environmental effect. In the material composition developed during the research PLA polymer serves as an adhesive gluing together flax fibre layers.

Of all the cut NWM sample strips two strips of each sample were used for manufacture of composite – two strips cut in the machine direction and one strip in the cross-machine direction. NWM composites were produced using the thermal press technique. Hot pressing was performed by C press available at the RWTH ITA laboratory and consisting of two horizontal surfaces positioned parallel to each other and having integrated heating elements. It is possible to set the desired temperature for each heating surface. Apart from temperature it is also possible to set the pressure, which is applied for pressing both surfaces. Within the framework of the research the operating parameters of the equipment were determined experimentally taking as basis the previous RWTH ITA experience in production of composites and making experiments: the temperature of 162 °C was set for both heating surfaces, pressure used for pressing together both surfaces was 3.75 bar, all the samples were held in the thermal press for 15 min. The operating temperature of the heating surfaces was determined by the melting temperature of the polymer used in the manufacture of the composite.

Results and discussion

In total three NWM samples (A, B and C) were manufactured, initially six NWM sample strips were cut from each sample, four of which were cut in the machine direction and two strips in the cross-machine direction. All the sample strips were cut of equal width and length: 50×300 mm. Initially all the sample strips were marked with a Latin letter of a particular sample supplemented by a number, where the NWM sample strips cut in the machine direction are marked with No.1; 2; 3 and 4, and the strips cut in the cross-machine direction are marked with No.5 and 6.

Using NWM tensile tests A, B and C sample strips marked with No. 1, 3 and 5 were tested. In Fig. 2 strips of sample B are shown. Average NWM thickness varies between 12.62 and 15.54 mm.

PLA dominates on the surface (outer sides) of the NWM samples; on the material surfaces in the needle-punching direction trail left by needles can be seen clearly as imprints of regular shape spaced at regular intervals. Surface density of NWM used in automotive industry depending on the use of materials ranges from 100 to $1400 \text{ g} \cdot \text{m}^{-2}$ [7]. The surface density of NWM developed during the research varies in the range from 1066 to $1261 \text{ g} \cdot \text{m}^{-2}$. Table 1 shows the tensile test results of the tested NWM: ultimate tensile strength of the NWM samples ranges from 0.03 to 0.05 MPa, material elongation ranges from 40 to 59 per cent. Relationship between the NWM strip specimen material elongation during the tensile test and the applied load depicted in diagrams is shown in Fig. 3. In the diagrams one can see the tendency of the specimen strips cut in cross-machine direction to reach the material breaking point at a smaller load, which is indicative of varied mechanical strength depending on the direction of the applied load.



Fig. 2. **NWM and composite sample B strips:** on the left hand side NWM strips AA05-B-1 and AA05-B-3 are cut in the machine direction, strip AA05-B-5 is cut in the cross-machine direction; on the right-hand side composite strips AA05-B-2 and AA05-B-4 are cut in the machine direction, strip AA05-B-6 is cut in cross-machine direction

Table 1

Flax and PLA samples	Strip code	Sample width, mm	Sample length, mm	Sample average thickness, mm	Sample section area, mm ²	Surface density (mA), g·m ⁻²	Ultimate tensile strength (F_{max}) , MPa	Breaking load (F_{bruch}) , MPa	Tensile strength elongation $(e \ F_{max}), \ \%$	Elongation after breaking load (e Bruch), %	Sample after testing
MWM	A1	50	300	14.58	728.78	1260.51	0.03	0.01	56.24	68.54	Not failed
	A3	50	300	12,62	630.88	1232.53	0.04	0.01	55.54	67.61	Not failed
	A5	50	300	13.53	434.70	1132.70	0.05	0.01	39.88	47.02	Not failed
	B 1	50	300	13.15	657.48	1221.27	0.03	0.01	56,50	67.74	Not failed
	B3	50	300	14.68	733,98	1222.01	0.03	0.01	58.96	67.10	Not failed
	B5	50	300	13.96	698.10	1065.74	0.03	0.01	42.40	52.08	Not failed
	C1	50	300	15.54	776.88	1180.77	0.04	0.01	48.99	59.55	Not failed
	C3	50	300	14.35	717.48	1228.77	0.04	0.01	55.79	66.59	Not failed
	C5	50	300	14.72	736.15	1138.87	0.03	0.01	46.59	55.35	Not failed
Composite	A2	50	240	1.86	92.83	1190.25	21.79	21.79	3.99	3.99	Not failed
	A4	50	242	1.99	99.40	1234.52	20.71	20.71	3.59	3.61	Not failed
	B2	50	233	1.78	89.15	1215.21	24.18	24.18	3,56	3.56	Not failed
	B4	50	235	2.29	114.50	1208,09	16.79	16.79	4.31	4.31	Not failed
	C2	50	240	1.80	89.80	1052.32	22.73	22.73	4.13	4.13	Not failed
	C4	50	236	1.95	97.38	1189.27	21.51	21.51	3.86	3.86	Not failed

NWM and composites tensile test results

To produce composite samples the NWM sample A, B and C strips marked No. 2, 4 and 6 are used. Three strips of each NWM sample have been used for the production of composite. Sample B strips, cut in machine direction and in cross-machine direction can be seen in Fig. 2. Composite sample width is 50 mm, length ranges from 232 to 244 mm and thickness – from 1.38 to 2.29 mm. The length of thermo press horizontal surfaces working area (it was shorter than the length of the NWM

samples) and heating trends by operating parameters explain the length wide range of the composite samples.

When NWM strips are placed in the thermal press, the white PLA fibre melts and becomes transparent; therefore the ready composite samples acquire the colour of flax fibre. The flax fibres, which can be seen on the surface of the composite material, are oriented in multiple directions.



Fig. 3. Diagrams showing relationship between NWM and composite material strip elongation and the applied load: 1 – NWM sample A; 2 – composite sample A; 3 – NWM sample B; 4 – composite sample B; 5 – NWM sample C; 6 – composite sample C

Surface density of composite materials varies in the range from 1035 to $1235 \text{ g} \cdot \text{m}^{-2}$. During the tensile test composites cut in the machine direction were tested; in Fig. 3 composite materials elongation depending on the applied load can be seen – the curves demonstrate similarity of the sample strip characteristics. The results obtained during tensile testing are depicted in Table 1. Sample (ultimate) tensile strength ranges from 16.79 to 24.18 MPa and specific elongation – from 3.56 % to 4.31 %. Difference of the composite samples ultimate tensile strength between the lowest and the

highest values amounts to 7.39 MPa while maintaining a uniformly stable material elongation – the difference in the material elongation is within 1 %.

Table 2

Strip code	Composites									
	Specimen section area mm ²	Surface density (mA), g·m ⁻²	Bending strength (F _{max}), MPa	Bending resistance (s _f), MPa	Bending modulus (<i>E_f</i>), GPa	Bending strength elongation (<i>e</i>), %				
A6	40.49	1059.24	0.92	62.02	2.88	0.04				
B6	42.39	1152.40	1.11	66.53	1.99	0.04				
C6	33.94	1034.73	1.10	86.24	4.20	0.04				

Composites 3-point bending test results

Composite samples were tested in 3-point bending test, the test results are shown in Table 2. Their bending strength varies in the range from 0.92 to 1.11 MPa at material elongation of 0.04 %. Bending modulus of the materials ranges from 1.99 to 4.20 GPa.

Conclusions

- 1. Conducting the research the NWM structure of flax and PLA fibres has been developed, for the formation of which five fibre web layers were used. Both raw materials of the material are obtained from renewable resources. One of the above flax fibre can be obtained in the territory of Latvia, where conditions for growing it are suitable.
- 2. The average surface density of the developed NWM ranges from 1066 to 1261 $g \cdot m^{-2}$, which is close to the density upper limit 1400 $g \cdot m^{-2}$ of NWM used in automotive industry [7]. The results prove that the material obtained is suitable for automotive use in some textiles position. The developed composite samples have a similar surface density: from 1035 $g \cdot m^{-2}$ to 1235 $g \cdot m^{-2}$.
- 3. The structure of the developed material is suitable for NWM and composite manufacture. By supplementing the process of NWM manufacture with thermal press processing, a new material can be obtained a composite of biological origin. The mechanical strength of the obtained composite is multiple times higher than that of NWM: the tensile strength is increased and the material elongation is decreased.
- 4. Tensile stress breaking point for NWM specimens, which are made from the strips cut out in the machine direction, can be reached by applying a lighter load than for the other NWM. The material mechanical strength differs depending on the load direction (in the machine direction or in the cross-machine direction). This factor should be taken into account when choosing the position of textile material in the car design.

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