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Formation and Properties of Textile Biocomposites with PLA Matrix Reinforced with Flax and Flax/PLA Weft Knitted Fabrics

Abstract
Eco-friendly textile biocomposites with PLA matrix reinforced with flax and flax/PLA weft knitted fabrics without any additional chemical treatment were prepared in this study. Five different kinds of samples were manufactured and investigated: (i) pure PLA sheet and (ii-v) PLA matrix reinforced with weft knits (ii) single jersey flax knit, (iii) single jersey flax/PLA knit, (iv) double single jersey flax knit, (v) double single jersey flax/PLA knit. Tensile and flexural properties of PLA biocomposites with weft knitted fabrics were higher than pure PLA sheet. The highest properties exhibit PLA biocomposites reinforced with single jersey flax knitted fabric. Differential scanning calorimetry (DSC) showed that crystallization rate of PLA matrix is enhanced with the presence of reinforced material - flax knit. It is possible to do an assumption that flax fibre can act as nucleating agents during the crystallization process.

Key words: *poly(lactic acid) (PLA), flax, biocomposite, tensile properties, flexural properties.*

Introduction

Due to increasing environmental awareness nowadays, biocomposites are becoming one of the most important materials. The development of high-performance eco-friendly engineering products made from natural recourses is increasing worldwide [1].

Natural fibres have been used to reinforce thermoplastics due to their advantages, such as: low cost, low weight, acceptable specific strength, good thermal insulation properties, biodegradability, sequestration of carbon dioxide (reducing the "greenhouse effect"), and most important renewability. Such eco-friendly composites may be used for automotive, mobile phone, and biomedical applications [2 - 4].

Polylactic acid (PLA) among thermoplastic biopolymers is one of the most promising candidates for wide range applications due its benefits, such as: biodegradability, high degree of transparency, and prominent mechanical properties. PLA basic monomer is lactic acid which is derived from starch by fermentation. Lactic acid is then polymerized to poly(lactic acid), either by gradual polycondensation or by ring opening polymerization [3, 5, 6]. Some natural fibre reinforced PLA products are already in the market: Jacob Winter (Satzung Germany) biodegradable urns from flax and PLA by compression moulding [6].

PLA biocomposites have been reinforced mostly by the following fibres: flax, ramie, cotton, hemp, kenaf, lyocell, bamboo, vetiver grass fibre, coconut, banana, jute, coir fibres, pineapple leaf fibres and even silk fibres [3, 5 - 14]. There are some researches where woven bamboo, flax, jute fabrics [2, 15, 16], nonwoven flax fabric [17], flax weft knitted fabric [18] were used as reinforced material for PLA biocomposites. As M.J.A. van den Oever with coauthors reported [19] that comparing to performance data presented in literature, it must be noted that the PLA family exhibits a wide range of properties. These properties depend on the (co) polymer structure, molar mass, and crystallinity.

Mechanical properties of fibre reinforced PLA composites depend on the amount of natural fibres in biocomposite, fibre-matrix adhesion which may also be directly related to processing conditions and reinforced material surface treatment [9, 12, 17, 18]. It was estimated [8] that tensile modulus of PLA composites reinforced by bamboo fibre, vetiver grass fibre and coconut fibre increased by the addition of untreated natural fibres, however tensile strength of biocomposites with vetiver grass fibre and coconut fibre were lower than pure PLA. Tensile properties of PALF (pineapple leaf fibre)/PLA composites were significantly improved by increasing PALF content. PALF reinforced PLA composite containing 40% fibre showed optimum tensile properties [14]. W. Xiao-Yun and et.al [11] reported that PLA composites with flax volume fraction of 35% had the best strength, also arrangement of the flax fibre direction played an important role for mechanical properties of biocomposites. The results that mechanical properties of biocomposites (PLA matrix) increase with the increasing flax fibre amount till 60% wt. (using flax nonwoven material) are presented in S. Alimuzzaman with co-authors study [17].

D.Y. Liu with co-authors [20] had analyzed PLA films with flax cellulose nanofibres. In order to prepare flax cellulose nanofibres of 20 nm in diameter and 300 nm long first of all bleached flax yarns were boiled in distilled water and dried. Dried yarns were soaked in aqueous NaOH at 80 °C and washed in running water. An aqueous suspension of nanocellulose fibres was prepared by acid hydrolysis. After complementation of hydrolysis the flask was cooled in ice-cold water. The aqueous suspension of fibres was diluted, washed and neutralized with aqueous NaOH. The suspension was fronzen and freeze dried. In was estimated that the tensile strength of PLA/flax cellulose nanofibres composites increased with increased amount of flax cellulose (from 25 to 59% respectively than flax cellulose amount in composite 2.5 and 5 wt%) [20].

An impact involves relatively high contact forced acting on a small area over a period of short duration. In general, composites absorb energy through fracture mechanism, such as: indentation, matrix and fibre cracks, and delamination [4]. M.J.A. van den Oever with co-authors [19] had estimated that impact behavior biocomposites decrease with fibre content increasing (to 50%).

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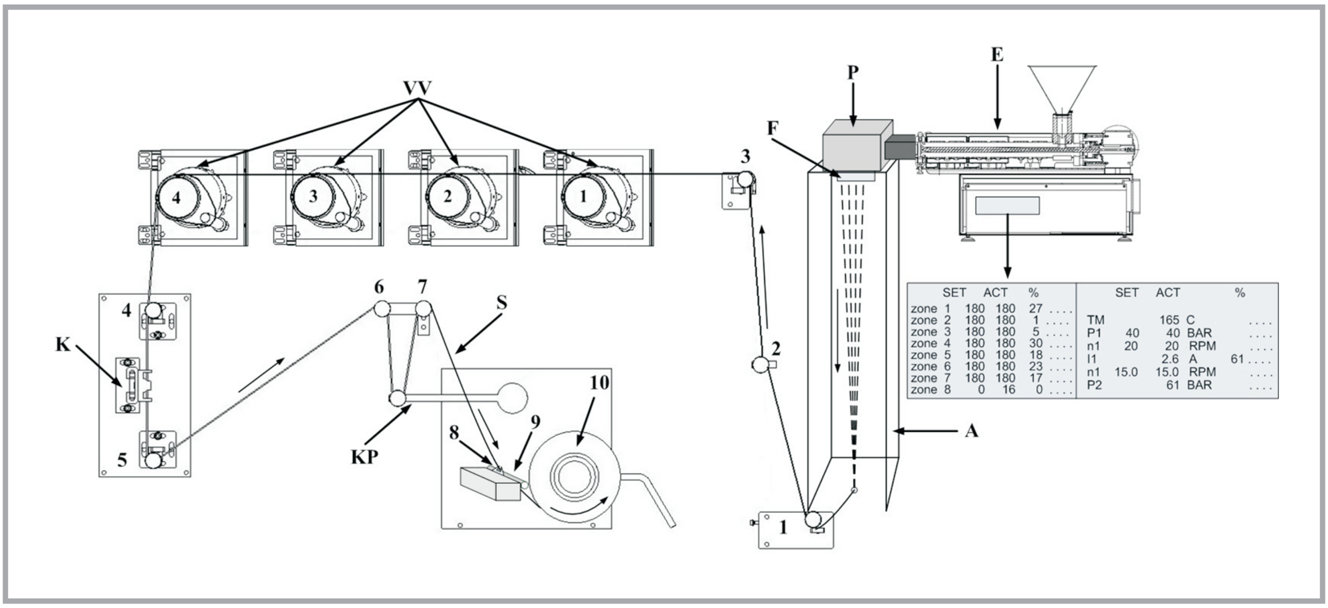

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   SET  ACT  %                    SET   ACT        %
zone 1  180  180  27 ....
zone 2  180  180   1 ....    TM          165 C        ....
zone 3  180  180   5 ....    P1   40      40 BAR      ....
zone 4  180  180  30 ....    n1   20      20 RPM      ....
zone 5  180  180  18 ....    I1          2.6 A     61 ....
zone 6  180  180  23 ....    n1  15.0   15.0 RPM      ....
zone 7  180  180  17 ....    P2           61 BAR      ....
zone 8    0   16   0 ....
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Figure 1. *Principal scheme of the spinning equipment COLLIN® CMF 100 (Dr.Collin Gmbh, Germany): E - extruder, P - melting pump, SP - spinneret, A - air quench cabinet, S - stretching godets, W - whirling unit, Y - multifibres from microfibres yarn [22].*

Differential scanning calorimetry (DSC) is a technique for measuring the energy, necessary to establish a nearly zero temperature difference between a substance and an inert reference material [4]. D.Y. Liu with co-authors [20] had analysed PLA/cellulose nanofibres (cellulose nanofibres were prepared by acid hydrolysis of flax yarns) composite thermal behaviour. During this study it was estimated that flax cellulose nanofibres can induce crystal nucleation of the PLA polymer, which implies that this flax cellulose material can probably be used as a nucleating agent for PLA [4, 20]. Also Y. Dong and et. al. [7] concluded that coir fibres played and effective nucleating role to accelerate the PLA crystallisation process and thus improve the crystal growth rate. M.S. Huda and et. al. [21] had studied PLA and recycled cellulose fibres (from newsprint). Analysing thermal properties of ‘green’/bio based composites, it was concluded that cellulose fibre does not affect significantly the crystallization behaviour of PLA matrix up to 30% cellulose content [21].

According literature review, PLA biocomposites mostly are reinforced by natural fibres. The aim of this study is to develop and characterize eco-friendly (without any additional chemical treatment) polylactic acid (PLA) laminated biocomposite reinforced with single/double layer weft knitted fabrics from flax and assembled flax/PLA yarns.

Experimental

Materials

PLA pellets (Ingeo^{TM} 6202D) obtained from Nature Works LLC (USA) were used for biocomposites matrix. Flax fibre yarns of 34 tex linear density were supplied from JSC “Lietlinen”, Lithuania (breaking force of flax yarns manufactured by wet spinning was 11.3 ± 0.9 N, elongation at break was 10 ± 0.8%, twist of flax yarns 190 t.p.m.).

Multifibre yarns from PLA polymer (Ingeo^{TM} 6201D NatureWorks LLC, USA) were manufactured by COLLIN® CMF 100 (Dr. Collin Gmbh, Germany) single screw extruder equipment in Kaunas University of Technology, Faculty of Mechanical Engineering and Design. The single screw extruder (L/D = 25 : 1) has seven heating zones where the temperature during experiments was seated 205 °C (***Figure 1***). The average speed of extruder was set up at 20 r.p.m. The circular spinnerets (***Figure 1*** indicated by SP) with 24 holes (diameter 0.45 mm) were used during these experiments. Cooling of filaments (***Figure 1*** indicated by A) was achieved with cross-flow air quenching at the temperature of 12 °C. Filaments were drawn in four stages: S1= 200 r.p.m, S2=400 r.p.m., S3=460 r.p.m., S4=506 r.p.m (Figure 1), draw ratio was 2.5. Linear density of manufactured assembled PLA multifibres yarn was 20 tex, diameter of single microfibre 30±1.1 μm (breaking force was 2.1±0.4 N, elongation at break was 45±2.5 %).

Table 1. *Structure parameters of reinforced knits.*

Reinforced weft knit fabric	Wale density Ph, cm^{-1}	Course density Pv, cm^{-1}	Area density Q, g/m^2	Area density after presing at 160 °C temperature, Q_T, g/m^2
Single jersey flax knit	6.0	8	114 ± 14	104 ± 12
Single jersey flax/PLA knit	6.5	10	248 ± 20	294 ± 14
Double single jersey flax knit	6.0	8	216 ± 20	210 ± 20
Double single jersey flax/PLA knit	6.5	10	604 ± 14	634 ± 31

Reinforced single jersey knitted fabrics from flax and assembled flax/PLA yarns were manufactured by flatbed knitted machine Shima Seiki SES 122-S 5E at JSC ”Gija” in Lithuania.

Preparation of biocomposites

Biocomposites reinforced plain jersey knits were manufactured from flax yarns and assembled flax/PLA yarns. The structure parameters of manufactured knits are presented in ***Table 1***.

PLA pellets were melted in a laminating machine NOVA 45 (Reliant Machinery, UK) at 190 °C temperature, at 50 kPa pressure for 30 s. PLA sheets three times were driven at the same forming conditions in order to get uniform PLA sheet. Reinforced weft knit fabric was attached

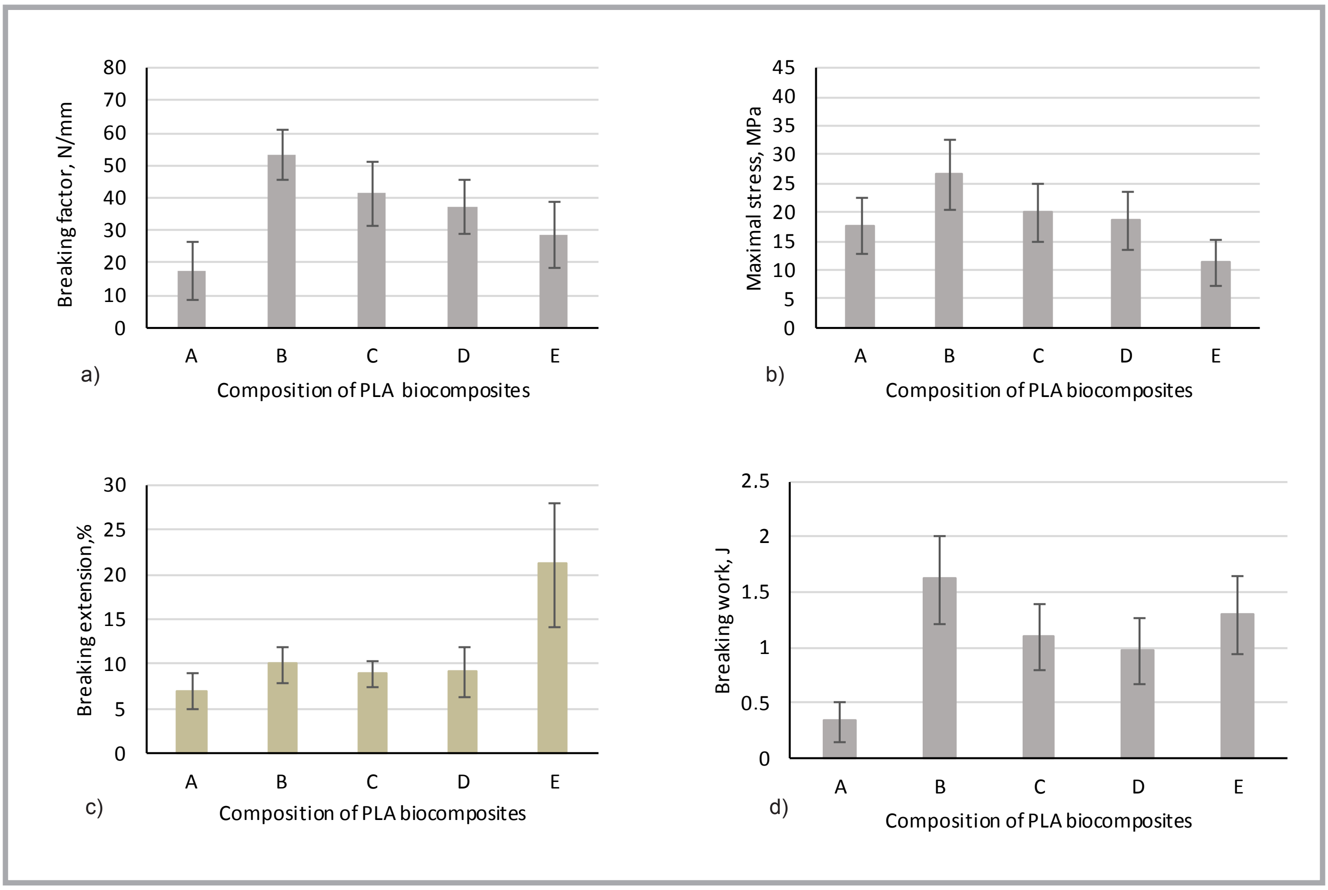

***Figure 2.** Tensile characteristics of PLA biocomposites. The dependence of (a) breaking factor N/mm, (b) maximal stress, MPa (c) breaking extension,%, (d) breaking work, J on composition PLA biocomposites: A- PLA sheet, PLA matrix reinforced with weft knits: B- single jersey flax knit, C- single jersey flax/PLA knit, D- double single jersey flax knit, E- double single jersey flax/PLA knit*

and placed between two PLA sheets and laminated at 160 °C, at 50 kPa pressure for 30 s. Biocomposites were driven at the same forming conditions for three times. Five different kind of samples: (i) pure PLA sheet and PLA biocomposite reinforced with weft knitted fabrics (ii) single jersey flax knit, (iii) single jersey flax/PLA knit, (iv) double single jersey flax knit, and (v) double single jersey flax/PLA knit were prepared and investigated in this study.

Characterization techniques

Tensile test of pure PLA sheet and PLA biocomposites with flax, flax/PLA weft knitted fabrics were carried out using testing machine Tinius Olsen H10KT (England) at room temperature. Specimens of PLA biocomposites reinforced with weft knitted fabrics were tested in knits course direction; the tensile speed was 5 mm/min, gauge length was 55 mm. Eight specimens for each set of specimens were tested and mean values with standard deviations were calculated. Flexural properties of PLA sheet and PLA biocomposites with weft knitted fabrics were determined according LST EN ISO 178:2010 by three-point bending test using a universal testing machine Tinius Olsen, Redhill (England). The width of specimens was 25 ± 0.5 mm, length was 60 ± 2 mm, test speed was 2 mm/min, a load cell of 5kN. Specimens were tested in knits course direction. Six test specimens were tested for each set of specimens and a mean value was calculated.

Differential scanning calorimetry (DSC) was performed on a TA Instruments Q10 (USA)equipped with a refrigerated cooling system and under nitrogen atmosphere at a flow rate of 20 ml/min. To evaluate the thermal behaviour of specimens, i.e. the degree of crystallinity of PLA in the biocomposite, the glass transition (T_g), melting (T_m), and cold crystallization (T_{cc}) temperatures and energy storage capacities that are based on the endothermic and exothermic peaks were also determined. Test specimens were heated and cooled during DSC analyses, within a certain temperature interval ranging from 0 to 200 °C at 10 °C min^{-1} and isothermed for 2 min at the end of each cycle. The DSC was calibrated with indium before the experiments.

The degree of crystallinity of pure PLA sheet and PLA biocomposites reinforced with flax, flax/PLA weft knits was determined from the second heat curve using the following equation [23, 24]:

$$X_c = ((\Delta H_f - \Delta H_{cc}) / \Delta \mathrm{H}_{f0})/\omega \text{ in \% } (1)$$

where, X_c is the weight fraction extent of crystallinity, ΔH_f – is the enthalpy of fusion measures at the melting point, ΔHcc is the enthalpy for the cold crystallization and ΔH_{f0} is the enthalpy of fusion of totally crystalline polymer measured at the equilibrium melting point, ω is the weight fraction of PLA in the composite. Fusion enthalpy of pure crystalline PLA material is 93.7 J/g [25]. The degree of crystallinity is related to many main properties shown by a semi-crystalline polymer including brittleness, toughness, stiffness and etc. [26].

Results and discussion

The properties of composite materials are determined by the characteristics of the polymer matrices, by the content and properties of the reinforcements and by the fibre-matrix adhesion [21]. One of

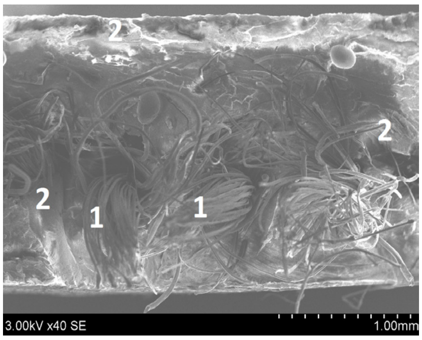

Figure 3. SEM images of PLA biocomposite reinforced with single jersey flax knitted fabric cross-section (after tensile test) 1-flax yarns, 2-PLA matrix.

the main disadvantage of natural fibres in composites is their poor compatibility with matrix and relatively high moisture absorption. Chemical treatment (alkali, silane, acetylation, benzoylation, acrylation, isocyanides, maleic anhydride) is required to modify their surface properties [8, 31]. Any chemical treatments for flax yarns or weft knits during this study for preparation of eco-friendly biocomposites reinforced with weft knitted fabrics were not used. PLA multifibre yarns have been manufactured by melt spinning extruder and reinforced single jersey knitted fabric from assembled flax/PLA yarns were manufactured in order to investigate the influence of PLA melt spun yarns in reinforced fabric on the properties of PLA biocomposites. Data in Table 1 show that single jersey knitted fabric from assembled flax/PLA fibres has higher wale and course density and higher area density (up 50%) than single jersey knitted fabric from flax yarns.

In ***Figure 2*** are presented tensile characteristics of analysed PLA biocomposites reinforced with weft flax, flax/PLA knitted fabrics. Presented data obviously show, that higher mechanical characteristics (breaking factor in N/mm, maximal stress in MPa) have PLA biocomposites with reinforced single jersey flax knit (sample code B) because of better PLA sheet and flax knit adhesion (***Figure 3***).

The breaking factor in N/mm of PLA biocomposites reinforced with analyzed weft knitted fabrics are about 38 - 66% higher than pure PLA sheets (***Figure 2.a***). Biocomposites reinforced with flax/PLA knitted (C and E) fabrics have 20% lower breaking factor than biocomposites reinforced with flax knitted (B and D) fabrics. Amount of reinforced material in composite has influence on composite tensile properties [11, 14]. Reinforced single jersey flax/PLA knit has the higher course and wale density than single jersey knit from flax yarns (***Table 1***); it may be one of the reasons that biocomposites reinforced with flax/PLA knit have lower breaking factor in N/mm. The same tendency can be noticed during the analysis of maximal stress (MPa) (***Figure 2.b***) characteristic of PLA biocomposites. The highest (30 - 40% higher than pure PLA sheet (A) or PLA biocomposite reinforced with double flax/PLA single jersey fabric (E)) maximal stress exhibits PLA biocomposites reinforced with single jersey knitted fabric (B). The lowest breaking factor in N/mm and maximal stress in MPa are characterized by PLA biocomposites with the highest content of reinforcement material (double flax/PLA single jersey knitted fabric (E)). Biocomposite reinforced with double flax/PLA single jersey knit had delaminated during tensile test. This agree with the researchers from the literature [11] that too high flax fibres amount in composite induces defective bonding between the fibre and matrix.

The highest breaking work in J (***Figure 2.d***), which should be reach in order to get breaking force of test, has PLA biocomposites with single jersey flax knit fabric (B). PLA biocomposites reinforced with double flax/PLA weft knit (E) exhibit 15% higher breaking work (J) than PLA biocomposites reinforced with single flax/PLA weft knit (C) due higher breaking extension in % (***Figure 2.c***) which was caused by extension of delaminated reinforced double single jersey knit.

Characteristics of flexural properties of PLA biocomposites reinforced with flax, flax/PLA knitted fabrics are presented in ***Figure 4.a*** and in ***Figure 4.b***. From presented data it is obviously seen that PLA biocomposites reinforced with flax weft knit (B, D) exhibit higher flexural modulus in MPa and flexural stress at maximum in MPa than PLA biocomposites reinforced flax/PLA weft knitted fabrics (C, E). PLA biocomposites reinforced with flax weft knitted fabrics have almost 8 times higher flexural modulus and 2.5 times higher flexural stress at maximum than of pure PLA sheet (A). Flexural modulus PLA biocomposite reinforced with single jersey flax/PLA knit (C) is about 20% lower, respectively flexural stress at maximum in MPa is 27% lower than PLA biocomposite reinforced with single jersey flax knit (B). Flexural modulus or flexural stress at maximum reduce (about 15%) when double layer single jersey flax knit reinforced material

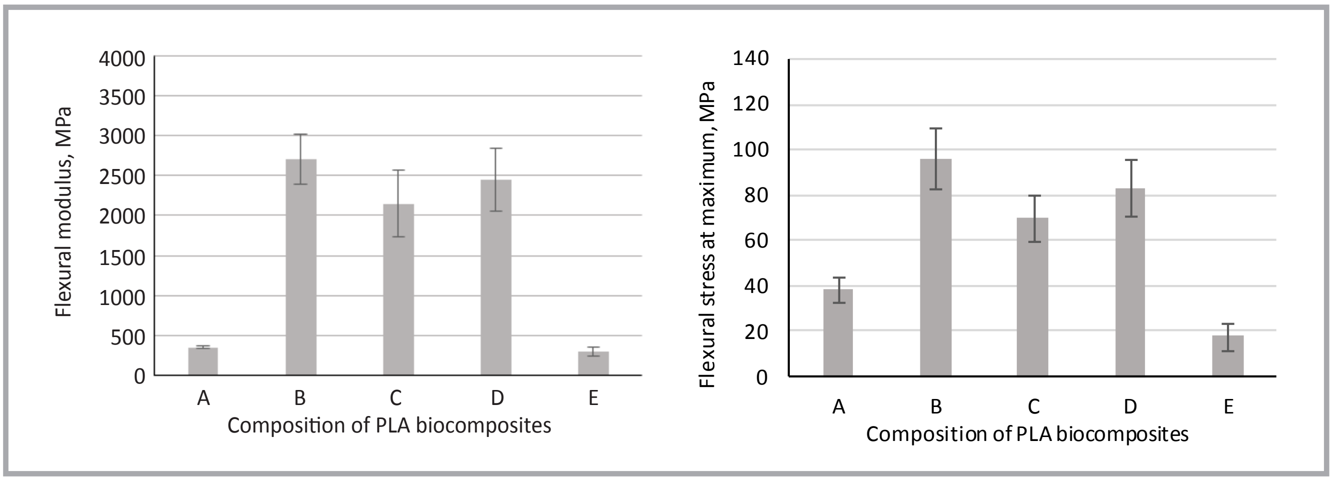

Figure 4. Flexural properties of PLA biocomposites. The dependence of a) flexural modulus MPa, b) flexural stress at maximum on composition PLA biocomposites: A- PLA sheet, PLA matrix reinforced with weft knits: B- single jersey flax knit, C- single jersey flax/PLA knit, D- double single jersey flax knit, E- double single jersey flax/PLA knit.

***Table 2.** Thermal characteristics of PLA sheet and PLA biocomposites reinforced with flax, flax/PLA weft knitted fabrics.*

Code of samples	Sample name	*Tg, °C*	*Tcc, °C*	*ΔHcc, J/g*	*Tm, °C*	*ΔHm, J/g*	*Xc, %*
A	PLA sheet	60.24	115.96	19.77	166.05	26.09	6.74
B	PLA biocomposite reinforced with single jersey flax knitted fabric	61.93	113.63	22.14	167.03	30.78	15.37
C	PLA biocomposite reinforced with single jersey flax/PLA knitted fabric	61.47	112.40	23.41	167.67	34.49	17.91
D	PLA biocomposite reinforced with double single jersey flax knitted fabric	61.87	111.45	23.01	168.21	31.95	23.85
E	PLA biocomposite reinforced with double single jersey flax/PLA knitted fabric	61.17	111.92	23.82	166.46	35.16	22.00

instead of single layer is used. Due quick delamination process PLA biocomposites reinforced with double flax/PLA single jersey knits (E) showed the lowest impact properties (flexural modulus is 90%, flexural stress at maximum is 80% lower than PLA biocomposites reinforced with flax single jersey knit (B)).

Cellulosic fibres have low thermal stability resulting in the exclusion of some manufacturing processes, and also limiting the use of the composites to low temperature applications. The low thermal stability increases the possibility of cellulosic degradation and the possibility of emissions of volatile materials that could adversely affect the composite properties. Processing temperatures are thus limited to about 200 °C, although it is possible to use higher temperature for short periods of time. However, cellulosic fibres are the key materials for eco-friendly biocomposites with natural organic fillers, i.e. fillers coming from renewable sources and biodegradable.

The thermal properties, such as: the glass transition temperature (T_g), cold crystallization temperature (T_{cc}), melting temperature (Tm), crystallization enthalpy (ΔH_c) and melting enthalpy (ΔH_m) obtained from DSC studies of the biocomposites are summarized in ***Table 2*** and graphically showed in ***Figure 5***. The degree of crystallinity (X_c in %) of PLA in the biocomposites was calculated using the ***Equation 1***.

It is observed that each sample exhibits an endothermic melting peak during the thermal scan. It is known that PLA samples exhibit crystallinity, however, no exothermic peak of crystallization was found; it could be explained that the samples were produced from low molecular mass PLA polymer [27, 28]. In place of it, the samples crystallize during the heating when the cold crystallization is observed.

The addition of flax knits does not significantly affect the glass transition and is visible at about 61 °C. In addition to the glass transition temperature, a cold crystallization temperature was also observed for the biocomposites. A slightly decrease of the cold crystallization temperature (T_{cc}) of composites can be observed with the increasing of reinforcing material content, though the melting enthalpy (ΔH_m), crystallization enthalpy (ΔHcc), and crystallinity (Xc) increases. Moreover, the crystallization peak of PLA sheet shifts to higher temperature with the incorporation of reinforcing knit, signifying that the crystallization rate of PLA is enhanced with the presence of flax fibre. The study showed that enthalpy of melting is closed to a third of totally crystalline PLA polymer (i.e., ΔHm approx. equal to 30 J/g). This agrees with the results from the literature [26] that only a small portion of the amorphous material changes state after during the exothermic reaction.

The addition of flax fibre enhanced the crystallinity of PLA (***Table 5***) because flax fibre can act as nucleating agents during the crystallization process. This agrees with the results from the literature that the addition of reinforcing material enhances the crystallinity of PLA [24, 29, 30].

Conclusions

It is possible to prepare eco-friendly composites from natural (without any additional chemical treatment) plain jersey knitted flax yarns and biodegradable PLA polymer.

Mechanical properties of PLA biocomposites reinforced with single jersey knitted fabrics depend on knitted reinforced material composition, and the number of weft knits layers. PLA biocomposites reinforced with single jersey flax knit showed higher tensile (breaking factor N/mm, maximal stress MPa, or breaking work J), and the impact (flexural modulus MPa, flexural stress at maximum, MPa) properties than PLA biocomposites reinforced with double single jersey flax or single/double flax/PLA knits due to the better adhesion between PLA matrix and reinforced flax knitted fabric. Weft knitted reinforced fabric from assembled flax/PLA yarns has higher wale, course

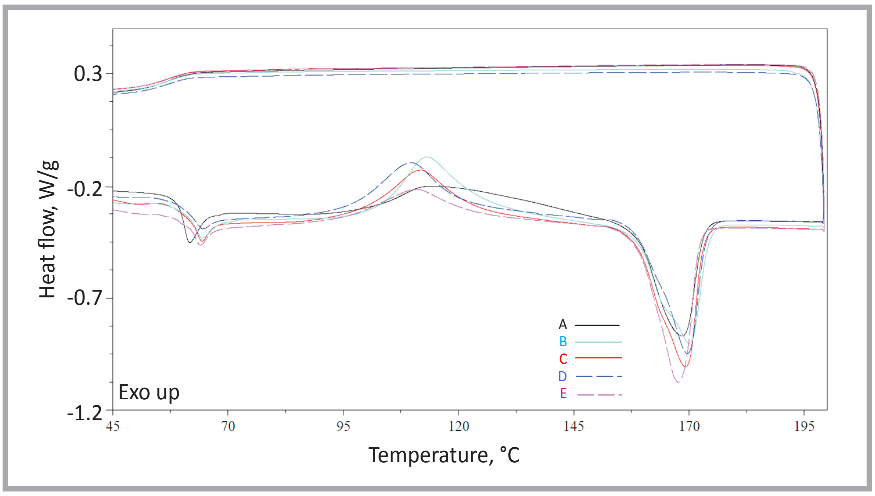

***Figure 5.** DSC thermograms of tested of PLA biocomposites: A- PLA sheet, PLA matrix reinforced with weft knits: B- single jersey flax knit, C- single jersey flax/PLA knit, D- double single jersey flax knit, E- double single jersey flax/PLA knit.*

density and do not improve mechanical properties of analyzing PLA biocomposites. Biocomposites with double flax/PLA knit exhibit the lowest mechanical properties due to high amount of reinforcement material. Too high amount of flax yarn in biocomposite induces defective bonding between the fibre and matrix and causes delamination of biocomposite.

The addition of single jersey flax knit in PLA matrix does not significantly affect the glass transition of biocomposites. Crystallization rate of PLA matrix is enhanced with the presence of reinforced material , i.e. flax knit. It is possible to do an assumption that flax fibre can act as nucleating agents during the crystallization process.

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