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Thermal and Mechanical Properties of Flax-reinforced Polycaprolactone Composites

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Abstract: Thermomechanical and mechanical properties of polycaprolactone/flax composites were investigated. The composites were prepared by melt mixing with three different filler loadings. Except from heating (to eliminate the moisture) no other procedures were applied to the raw materials, neither compatibilizer agent was used. The tensile and impact properties were evaluated and dynamic mechanical analysis was performed. Additionally the materials have been characterized by means of DSC technique. The resulting material reveal enhanced mechanical properties due to reinforcement caused by fibers and by high interfacial adhesion.

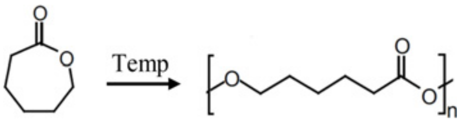
Keywords: Polycaprolactone, Flax, Biodegradation, Crystallinity

Introduction

The today's world cannot stand without plastics. These materials use to be widely applied in many industrial branches and in numerous consumer applications. Plastics are lightweight, do not stain and exhibit considerable mechanical strength. Formability into a desired shape is almost unrestricted, moreover there exist a huge number of polymers which fulfill rigorous demands of their applications at different temperatures and under various chemical environments. The possibility to produce composites with fillers of different shape, origin and filling degree resulted in a vast number of engineering materials with different properties (mechanical, barrier, surface, sorption etc.). These enumerated features are undoubtedly placed on a bright side of the plastics industry. However, for all synthetic plastics, there exists a problem of waste management after end of their life. Although the mechanical or chemical recycling and incineration are known for decades as the favorable plastic waste management options, still the after-use disposal of post-consumer waste constitutes almost 50 % [1]. Lack of a free space at landfills for a steadily increasing amount of waste and long-termed (if, at all) degradation, constitute the weak points of the petrochemically originated plastics after completion of their life time.

Mechanical recycling allows to recover substantial amounts of discarded waste plastics, but after several cycles, even the recyclates finally have to be landfilled or incinerated. One of the way to solve the problems arising from the storage of plastic at landfills, is to replace them with biodegradable analogues. An increasing public awareness in the area of plastic waste and searching for sustainable resources, opens up new horizons for developing biodegradable polymers and composites based on them [2,3]. According to EN 13432 and ISO 472 biodegradable materials undergo significant changes in their chemical structure at specific environmental conditions, thus loosing their initial chemical and physical properties. Biodegradation should eventually result in a transformation of plastics into the biomass, carbon dioxide and water [4,5] with the lowest energy expenditure, comparing to known waste management techniques [6].

Polycaprolactone (PCL) is an alyphatic polyester, that is usually produced from ε-caprolactone by a ring opening polymerization [7], according to a following reaction scheme:



PCL is biodegradable due to the presence of ester bonds in the macromolecule. In particular, the place where enzymes start their attack is a carbonyl group [8]. PCL exhibits a low melting temperature (58-60 ^{o}C), is stable in the aqueous environment, biodegradable [9-12] and tough [13]. Due to biodegradability, PCL was adopted in many applications, like medical implants [14], time-retarded medicines [15], slow released pesticides or fertilizers [16,17], or packaging [18]. Comparing to polylactic acid (PLA), biodegradation of PCL is much slower and takes up to two years for its complete dissolution. For that reason PCL is used in a cardiac surgery and urology as a drug release carrier [15]. On the other hand, a high price and low melting point of PCL makes the material not attractive for popular consumer applications.

However, the price reduction is possible if combine polycaprolactone with cheap fillers, thus manufacturing composite materials. Physical properties of polymer composites depend on the physico-chemical properties of the polymer matrix and the filler, on the interfacial adhesion and on the filler loading. For selection of proper composite constituents also other aspects should be considered, like availability and price of the components, wearing of the filler on a surface of

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*Corresponding author: karol.leluk@gmail.com the processing machinery and a final application of the materials. For example in the tissue engineering the materials used for a bone repair have to match properties of the tissue and exhibit high enough mechanical properties. An established position, as the bone substitutes, have composites of PCL with hydroxyapatite, which is the main mineral component of bone and biocompatible filler [19,20]. Other fillers used for enhancement of the stiffness and hardness of PCL are bioactive glass nanofibers [21], glass microspheres [22], or TiO_2 nanoparticles [23]. Recently, as sustainability is gaining high importance, also cellulose have been implemented in polymer composites [24,25]. Wood and natural fibers are used as fillers in polymer composites applied in the automotive and construction industry due to their low density and non-abrasive performance during processing, despite their chemical inhomogenity, low thermal stability and incompatibility with hydrophobic polymers [26-37].

The flax fibers are naturally occurring, sustainable products originating from the plant's bast. Flax fibers are known as a reinforcement in polymer composites [38], however their strength, stiffness and surface properties may vary according to growth conditions or type of the plant.

To our knowledge, there are only a few papers describing properties of PCL composites reinforced with flax fibers. One of the possible explanation is a relatively low softening temperature of PCL (around 60 °C). Moreover, although the polymer is biodegradable, it is manufactured from the petrochemical raw materials.

The aim of this paper is to describe the characteristics of PCL filled with flax fibers and compounded at the lowest possible temperature, in order to avoid degradation of the polymer matrix. Authors supposed that due to the presence of polar, ester bonds in PCL and similar chemical character of flax fibers (polar cellulose) no adhesion promoters are required to achieve a considerble reinforcing effect. The materials have been characterized in terms of their thermal and mechanical properties in relation to their morphology.

Experimental

Materials

PCL grade CAPA®6400 was acquired from Perstorp, UK (Solvay). According to the manufacturer a mean molecular weight of the polymer was estimated to 37,000 g/mol. The product was supplied in a form of pellets diameter of 3 mm. Some physical properties of the investigated polymer have been listed in Table 1.

Table 1. PCL physical properties (following manufacturer's note)

Parameter	Value
Melting temperature, °C	58-60
Water content, %	<1.0
Elongation at break, %	660
MFR (2.16 kg, 160 °C), g/10 min	40
OH value, mg KOH/g	4

Ready to use flax fibers (length of 4 mm) were delivered by EKOTEX, Poland. The plants were harvested in Poland, Lower Silesia region. The fibers, besides necessary treatment (dew retting), were not otherwise processed.

Sample Preparation

Four materials were prepared for testing, namely the matrix polymer, and PCL composites with fiber loadings of 10, 20 and 30 wt.%. Polymer pellets, as well as flax fibers, were vacuum dried before processing. Drying conditions were 50 °C, 30 min. (PCL) and 105 °C, 120 min. (flax fibers).

The composites were prepared by compounding the components using POLYLAB QC mixer, volume of 50 ccm. The processing temperature was 60 °C and rotational speed of rotors constituted 60 rpm. The overall process took up to 8 minutes (counting from the time-point when the momentum has reached a constant value). Small portions of flax fibers were added step-wise, to ensure their regular distribution in the polymer matrix.

After mixing, the composites were grinded and samples for mechanical testing were prepared with the micro-injection moulding machine (PROMA) to the bone-shape specimens dimensions of 4×2×30 mm for tensile testing, and bars 10×4 ×100 mm for bending.

Moreover, the plates for impact testing (100×100×1 mm) were manufactured using a hydraulic press (LabTech LP-20B). Polymer pellets were heated up in a mold to 70 °C and kept molten for 3 min. Next a pressure was applied for 2 minutes, then a plate was cooled with water.

Microscope Investigation

Two microscope techniques were applied - for the crystal growth observations the optical microscope (BIOLAR) equipped with a heating stage was used. For observation of the fracture surfaces the scanning electron microscope (EVO LS15) was used.

Thermal analysis

Differential Scanning Calorimeter Q20 (TA Instruments) was employed for the thermal properties characterization. The samples were cooled down to −80 °C and heated up to +70 °C, well below and above the material's glass transition and melting temperature. Heating and cooling rate was set to 10 °C/min. Empty crucible was used as a reference material. Glass transition temperature, melting point and crystallinity were the parameters acquired from the DSC investigations. All measurements were performed under nitrogen.

Thermomechanical Tests

Dynamic mechanical analysis was performed with DMA 2980 apparatus (TA Instruments). The measurements were executed in air, temperature range from −75 °C up to +75 °C with a heating rate of 3 °C/min at a frequency of 1 Hz (single cantilever deformation mode). The samples dimensions were 17,5×10,2×4,0 mm.

Mechanical Tests

The tensile and bending measurements were carried out using Lloyd LR10K tensile tester according to ISO 527 standard. The tensile speed was 10 mm/min and the stress-strain dependency was recorded until destruction of the sample. During the tests following parameters were evaluated: Young's modulus (at tensile and bending), elongation at break (tensile), strain, stress and sample deflection at break (bending). In each test (tensile, bending), 5 samples were investigated.

Impact resistance of the samples was investigated with the falling dart impact DartTester, CEAST. The drop height was 1000 mm, dart speed 4,43 m/s. The test was repeated five times for each material.

Results and Discussion

Dynamic Scanning Calorimetry

The melting point was acquired as the temperature responding to the endothermic peak, whereas the glass transition temperature was estimated from the deflection point on a heat flow curve. Endothermic signals related to melting were sharp and symmetric for all curves, indicating that only one thermal process occurred (Figure 1). The measured T_g and T_m values of PCL sample stay in a good agreement with the manufacturer's data and that of other researchers [39].

Melting occurred at 58.5 °C for PCL and for the composites T_m was only slightly higher (Table 2).

The glass transition temperature was hardly readable (Figure 2), what was also reported by other authors [40].

T_g was estimated to −61.1 °C for PCL and increased a little with the filler content, thus reflecting a reduced mobility of PCL macromolecules caused by the flax fibers.

As the fusion enthalpy of fully crystalline PCL is known (139,6 J/g [41]) the crystallinity degree was calculated with the formula (1) [42]. Designated parameters and PCL crystallinity have been presented in Table 2.

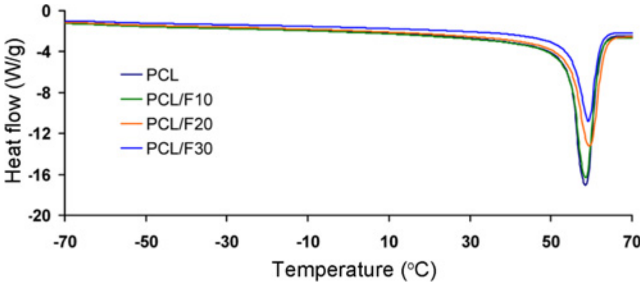

Figure 1. DSC curves of PCL and its composites. Signal related to the melting is clearly visible.

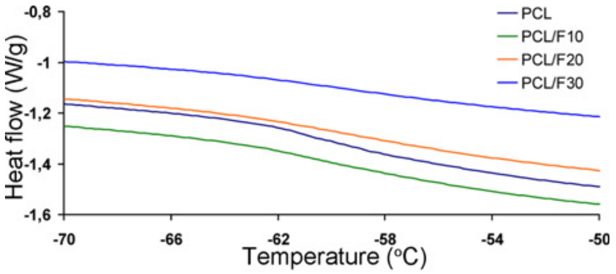

Figure 2. Enlarged region of DSC curves (presented on Figure 1) showing the glass transition region of each material.

$$X_c = \frac{\Delta H}{\Delta H^0 \cdot (1 - X_{filler})} \cdot 100\ \% \tag{1}$$

X_c: crystallinity degree (%)
ΔH: measured heat of fusion
ΔH^0: heat of fusion for hypothetical 100 % crystalline PCL
X_{filler}: mass fraction of flax fibers

Crystallinity degree estimated for PCL revealed that CAPA®6400 is a semi-crystalline polymer, which may be also deduced from a shape of the storage modulus run in the low temperature region [43].

Crystallinity degree of PCL composites may be influenced by at least two opposite effects. Assuming that X_c of a polymer in the composite remains unchanged, addition of a filler should cause a decrease in the crystallinity, as a total volume of the polymer reduces. However, in the fiber reinforced polymer composites, frequently a nucleation of crystals induced by fibers was reported, which resulted in a higher crystallinity [44,45]. Moreover, in the highly loaded composites the free volume available for polymer chains arrangements is markedly constrained, which inhibits the crystal growth, thus decreasing the crystallinity. In the investigated samples, as it can be concluded from Table 2, all these effects occured. Even though a mass fraction of semi-crystalline PCL decreased in composites with increasing filler amount, the crystallinity remained constant except of the highly loaded PCL-flax composite, which exhibited slightly lower crystallinity index.

Since the polymer mass fraction in composites is known

Table 2. Glass transition temperature, melting point and crystallinity degree calculated from DSC measurements

	T_g (°C)	T_m (°C)	Crystallinity degree (%) (calculated from DSC runs)	Crystallinity degree (%) (estimated from the filler/polymer ratio)
PCL	-61.1	58.5	41	-
PCL/F10	-60.8	58.6	42	38
PCL/F20	-59.5	59.5	41	34
PCL/F30	-59.2	59.3	37	30

(0.9, 0.8 and 0.7), it was possible to calculate the theoretical crystallinity degree. Firstly, the volume fraction of a polymer in the composites must be determined (which is 0.91, 0.83 and 0,74 respectively), then it should be multiplied by the crystallinity index of PCL (41 %). According to the last column of Table 2, these values are lower than these calculated from DSC measurements. That suggests that in the composites a nucleation process has occurred.

Optical Microscope Investigation

The crystallite growth has been observed using the optical microscope with a heating stage. After the polymer was melted, the heating was switched off and the cooling process started. The temperature, fixed at $40\,^{o}$C, remained unchanged during rest of the experiment.

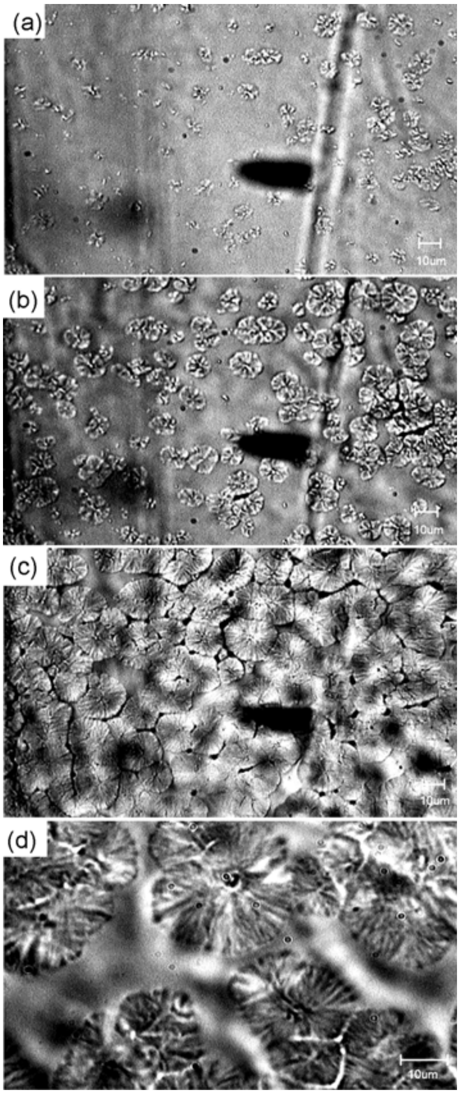

Figure 3. Snapshots of PCL crystallites while growing process: (a) initial phase, (b) after 1 min., (c) after 3 min. The digital magnification of (c) is presented on the last image (d).

The sequence of pictures (snapshots) presented in Figure 3 shows the crystallite growth in PCL.

The final crystal size equals to 25-30 μm, which is in a range reported by Liu *et al.* [46].

The crystallite growth took around three minutes. Figure 3(d) is an enlarged version of the shot taken 3 min. after the first crystallites appeared to show a structure of the crystallites.

Dynamic Mechanical Analysis

From the dynamic mechanical analysis the storage modulus, loss modulus and tangent delta were obtained. Substantial enhancement of the storage modulus with an increased filler content has been reported (Figure 4).

The change in modulus was not linear, especially that measured below the glass temperature (Figure 5) and may be related to a mode of the fibers distribution and free volume restrictions.

With an increasing fiber content they start overlapping and distribute non-uniformly in PCL matrix, thus decreasing the ability to an effective transfer of mechanical loadings trough the composite. That was evidenced clearly for the materials below the glass temperature of PCL. Low free volume and lack of freedom for the fibers re-orientation caused that at 30 % filler content the modulus was lower than expected and equal to that measured for PCL/flax composite with 20 % fibers loading.

Higher free volume and possibility of the conformational mobility of macromolecules above the glass temperature of a polymer matrix brought about a more pronounced increase in the composite modulus with a filler content, observed also

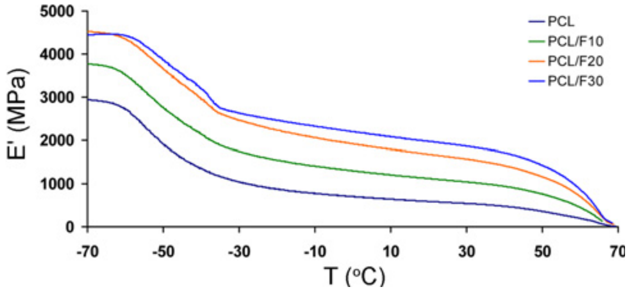

Figure 4. Temperature dependence of storage modulus for PCL and PCL/flax composites.

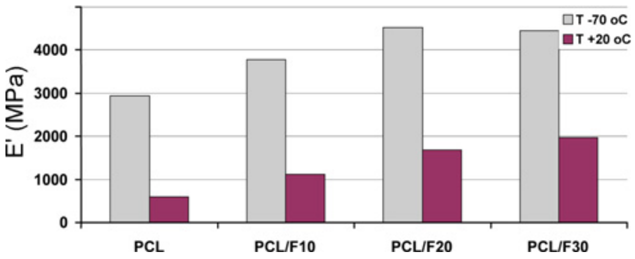

Figure 5. The values of storage modulus for PCL and PCL/flax composites for selected temperatures (below and above T_g).

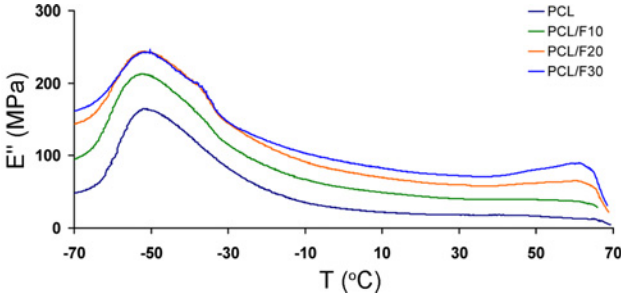

Figure 6. Temperature dependency of loss modulus for PCL and PCL/flax composites.

Table 3. Storage modulus for investigated samples measured at −70 °C (glassy state) and +20 °C (rubbery state)

Temperature	PCL	PCL/F10	PCL/F20	PCL/F30
−70 °C	2938	3772	4524	4445
+20 °C	590.2	1120	1680	1975
C coefficient	-	0,68	0,54	0,45

Table 4. Average differences in E' and E'' moduli observed between investigated samples

Difference between samples	PCL-PCL/F10	PCL/F10-PCL/F20	PCL/F20-PCL/F30
E' modulus (MPa)	620	622	205
E'' modulus (MPa)	37	30	10

for the highest loading (30 %). What is interesting, the modulus of PCL composite with just 10 % of flax fibers is in a range of that for polypropylene, being known for numerous application in several industrial branches.

To estimate the efficiency of a filler incorporated into the PCL matrix on the E' modulus, the coefficient C was calculated [47,48] according to the formula (2):

$$C = \frac{E_g^{\prime comp}/E_r^{\prime comp}}{E_g^{\prime PCL}/E_r^{\prime PCL}} \tag{2}$$

$E_g^{\prime comp}$ and $E_r^{\prime comp}$ are the storage moduli for the composite in the glassy and rubbery state (respectively), whereas $E_g^{\prime PCL}$ and $E_r^{\prime PCL}$ are the storage moduli obtained for the polymer in its glassy and rubbery state (respectively). For this purpose two temperatures (at which the polymer and composite samples were in different states) were selected (Table 3).

According to Table 3 the coefficient decreased with an increased fiber content from 0.68 (PCL/F10) to 0.45 (PCL/F30). The lower the coefficient value, the higher is the filler efficiency. Flax fibers exhibited the highest influence on E' in PCL/F30 sample, but more interesting are unequal differences between the subsequent samples. Namely, the efficiency of flax fibers increased to its highest in the sample PCL/F30, but the increase rate is lower (0.09) comparing to that calculated for PCL/F20 sample (0.14). Similar conclusion can be formulated after analyzing the E' and E'' temperature dependency presented on Figure 4 and Figure 6. An increase in the storage and loss moduli was observed in the entire temperature range after addition of different amounts of a filler (Table 4).

However, the curves representing the storage and loss moduli dependence on temperature for composite with 30 % loading are almost overlapping with that obtained for the composite with 20 % fiber content. This reflects the above presented calculations: a further increase of the filler content in a composite does not proportionally reflect the efficiency coefficient and in turn affects some properties.

The loss modulus temperature dependence can be also divided into two groups: first including PCL and 10 wt.% loaded composite, and the second with composites filled with 20 wt.% and 30 wt.% of fax fiber. For the second group plots, two maxima were observed, that correspond to the glass transition and melting point. In contrary, for the polymer matrix and PCL/F10 composite only one signal occurred at the glass transition region. An increase in the storage modulus visible on the Figure 4. occured close to the materials melting point. Such behavior was reported for the materials, that release stress when heated [49]. This phenomenon may be explained in two ways: by simple releasing a mobility of the polymer chain (subsequent release of freedom degrees with the temperature). Of course, in highly loaded composite samples (PCL/F20 and PCL/F30) the presence of a filler and its influence on the loss module has to be taken under consideration. It seems that in these materials, the stress release may also originate from the shear stress concentration on the ends of fibers embedded in the polymer matrix [47].

Another possible explanation is the filler non-uniform distribution within the polymer matrix. It has to be noted that during processing of PCL composites loaded with 20 wt.% and 30 wt.% of flax fibers, the temperature in a processing chamber has risen up to 82 °C and 95 °C, respectively. The temperature increase is a measure of the heat generated due to a friction during processing. In composites with 20 wt.% and 30 wt.% filler loading, the fibers are overlapping more likely, or may be distributed differently in various parts of the sample. Alternating the orientation degree may result in regions with higher fiber content and hence in locally increased stress level. Of course, the regions with under-average fiber concentration could be also formed, so statistically the filler distribution in a whole sample is even. That is why, while performing the tests revealing the composite macroscopic parameters (like: tensile or impact properties) the local fluctuations do not play a substantial role. By implementing analytical techniques (like DMA, or microscopy) the situation become different. Although the filler distribution is uniform in the major part of the sample,

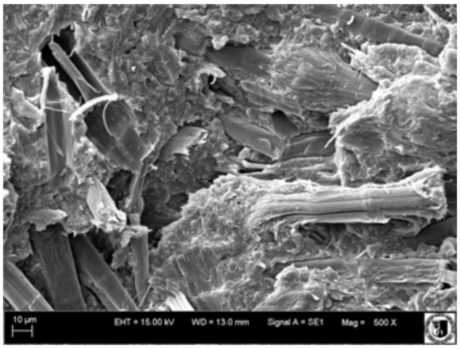

Figure 7. SEM picture of 30 wt% PCL-flax composite (magnification 500×).

the input originating from the uneven regions should be pronounced on the corresponding curves, what has been shown on Figure 6 (loss moduli).

Electron Microscopy Investigation

The picture presented in Figure 7 shows the scanning electron micrograph for 30 % loaded composite, illustrating the above considerations. Basing on the fiber traces left on the polymer surface as well as other fibers embedded within the matrix, it may be concluded that the interfacial interactions are of an adhesive nature. The adhesion between the polymer matrix and a filler may increase, what was described in [38].

The differences in the storage and loss moduli between the samples of subsequent filling degree deserve for comments. As it has been emphasized in Table 4, *E′* increased regularly (for 600 MPa steps) starting from the neat polymer up to 20 wt.% loaded composite. Similar observation concerned the loss moduli, with the increasing step of 34 MPa. The exception is the composite filled with 30 wt.% of flax fibers, for which the. increase in both moduli was about 3 times lower (200 MPa and 10 MPa, respectively). The possible explanation may deliver “C” coefficient, reflecting the fiber efficiency. According to Table 3 the difference in “C” coefficient between PCL/F20 and PCL/F30 sample is lower of about 35 % comparing to PCL/F20 and PCL/F10 sample.

Mechanical Tests

The highest impact resistance was obtained for neat PCL (Figure 8). Addition of 10 wt.% of a filler caused a tremendous decrease (13×) of a measured parameter. The changes occurring between the composites with a higher filler content were much lower, but the decreasing tendency was still observed. Such behaviour is similar to the findings reported by Sahari *et al.* [50].

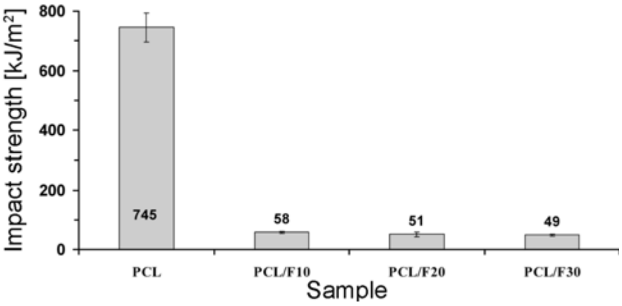

Figure 8. Impact strength results for investigated samples.

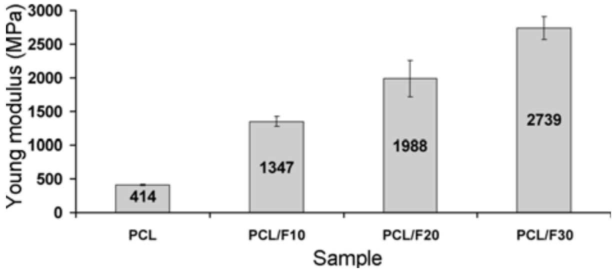

Figure 9. Tensile Young modulus for PCL and PCL/flax composites.

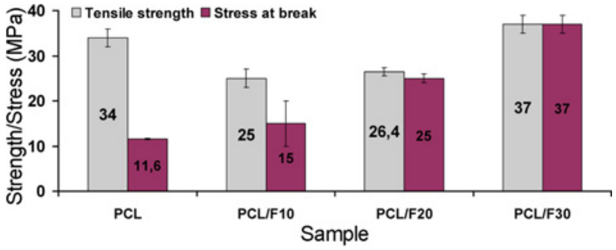

Figure 10. Tensile strength (gray bars) and stress at break (red bars) for PCL and its composites.

Tensile test results have been presented on Figures 9 and 10. Addition of flax fibers caused an increase in the composite stiffness. Comparing to PCL matrix the Young modulus of composites increased for 3, 5 and 7 times for the samples with 10, 20 and 30 wt.% fiber loading, respectively (Figure 9). That increase was proportional to the fiber contect in composites. Assuming the Young modulus for PCL/F10 sample as a reference (equal “1”), and calculating the Young modulus relative increase (RI) in the PCL/F20 and PCL/F30 samples, the change occurred to be 1,7 and 2,5 times higher (respectively) relating to PCL/F10 sample. The following calculation proves that the investigated system complies the mixing rule. Similar observations were reported by other authors according to their investigations of fiber reinforced polymer composites [51,52].

$$RI = \frac{Y_{Cx} - Y_{PCL}}{Y_{C1} - Y_{PCL}} \tag{3}$$

where:

RI: relative increase of Young modulus
Y_{Cx}: Young modulus value observed for composite C_x ($x = 1, 2, 3$)
Y_{PCL}: Young modulus measured for unloaded polymer

In the composite with 10 wt.% of a filler the tensile strength has decreased of about 30 % comparing to PCL.

Table 5. Elongation at break (in brackets-calculated standard deviation)

Sample	Elongation at break (%)
PCL	923 (65)
PCL/F10	7,7 (0,7)
PCL/F20	5,1 (0,7)
PCL/F30	4,1 (0,3)

After increasing the filling degree to 20 wt.% a slight increase of that parameter was observed (Figure 10), whereas for PCL/F30 sample the tensile strength was higher than that of the polymer matrix. As for the stress at break, the observed differences between the samples were of the same range-each subsequent sample indicated about 50 % increase as compared to the previous one. Starting from the 20 wt.% fiber loading, the investigated material cracked rapidly (brittle) what is indicated clearly on the Figure 10.

Grey bars (Figure 10) show the tensile strength of the composites. Its decrease may be interpreted as a lack of adhesion between the flax fibers and PCL matrix. That was discussed also in [53].

The changes are typical for the fiber filled polymer composites [54,55] - the addition of 10 wt.% of fibers dramatically decreased the elongation at break from more than 900 % to about 8 % (Table 5).

The values of Young modulus measured in tensile test are comparable to those obtained at the bending test (compare Figure 9 and Figure 11).

Like for the tensile tests, also for bending experiments the RI factor was calculated. The relative increase was 2,4 and 3,7 times higher in the composites PCL/F20 and PCL/F30, respectively. Following the rule of mixture, these values are a bit higher than expected, especially for the composite with 30 wt.% filler content.

Figure 12 exibits the deflection and maximum stress of the samples in the bending tests. For all samples the deflection decreased gradually with a filler content. The largest difference (about 9 mm) was observed for the composites filled with 10 wt.% and 20 wt.%, whereas in other cases it was two times lower. The stress at maximum deflection (Figure 12-purple bars) increased with the filler content.

Arbelaiz *et al.* [53] have investigated the PCL-flax composites compounded at 150 °C at 40 rpm. The composites were manufactured using maleic anhydride grafted PCL matrix (PCL-g-MA) with various anhydride contribution (from 1 to 40 wt %) as well as totally grafted PCL-g-MA matrix. The flax fiber loading varied from 0 % to 60 %. The authors came into conclusion that the mechanical performance of the composite does not depend linearly on the coupling agent content, but results from several factors (sometimes acting in an opposite manner), for example: the matrix crystallinity. the sample morphology and molecular weight of the coupling agent.

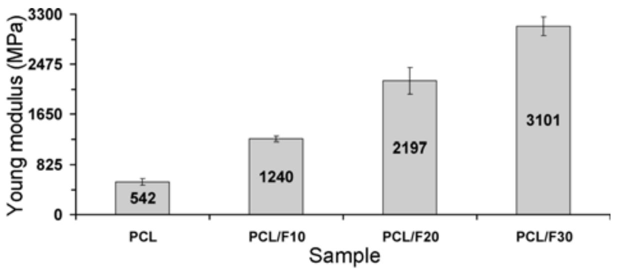

Figure 11. Bending test results (Young modulus) for PCL and PCL/flax composites.

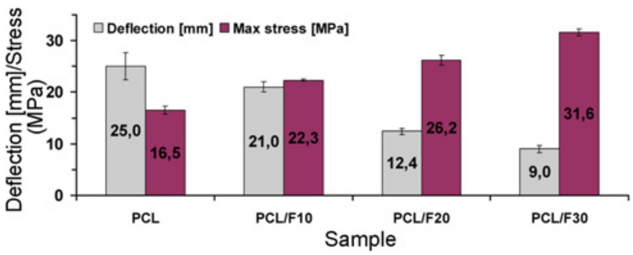

Figure 12. Deflection and max stress of PCL and PCL/flax composites (bending test).

Asokan *et al.* have compared mechanical properties of biodegradable composites based on polylactic acid, polyhydroxyalkanoates and PCL [56]. PCL filled with flax fibres was reported as the material of the lowest modulus and strength both at tensile and bending.

The results presented above reflect the reinforcing effect of short flax fibers. The microscopic investigation revealed a sufficient adhesion at the polymer/fiber interface which was reflected in the tensile measurements. Although the reinforcing effect (Young modulus, stress at break, deflection, etc) seem to be proportional to the fiber loading, a detailed calculation based on DMA measurements shade a different light on the obtained results. The reinforcing efficiency of the filler in the subsequent composite samples increased with the fiber content, but the increase is consequently lower, thus revealing the non-linear character of that dependency. This shows that incorporating more than 20 % of fibers constrains their performance, most likely due to reduction of the space in the matrix, available for a proper fiber arrangement. The last conclusion was also proven by DSC measurements.

Conclusion

Polycaprolactone-flax fiber reinforced composites were investigated in terms of the filler/polymer ratio. Most of the mechanical parameters, like Young modulus, stress at break (tensile and bending test), loss and storage moduli, have followed the rule of mixing. The exception was the highest loaded composite (30 wt. %). Although an increase was observed, it did not gain the theoretically expected level. The reasons may be of the chemical (no compatibilising agent was used) or physical nature (i.e. sample morphology). SEM has shown that in the highly loaded composite the fibers were distributed non-uniformly.

Summarizing, the composites with 10 wt. % and 20 wt. % fiber loadings follow the rule of mixing. Random fiber orientation, their non-uniform distribution, and moderate interfacial adhesion may be the factors responsible for the surprisingly low (comparing to that expected from the rule of mixing) tensile parameters.

Acknowledgments

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