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ORIGINAL PAPER

Accelerated Fatigue Testing of Biodegradable Composites with Flax Fibers

Aneta Liber-Kneć[1] · **Paulina Kuźniar**[2] · **Stanisław Kuciel**[2]

Abstract The objective of the study was to analyze and compare fatigue properties of composites based on two biodegradable polymers: thermoplastic starch and polylactide blend filled with 10 wt% short flax fibers. Accelerated fatigue tests with increasing amplitude were done on neat polymers and their composites. Dissipated energy, strain and the temperature on the sample surface were the three characteristic values measured or calculated during the tests to evaluate fatigue stress of the materials with the use of Lehr's method. The increase in temperature was not observed. Addition of flax fibers resulted in improvement of fatigue strength of polylactide-based composite but thermoplastic starch filled with flax fibers showed lower fatigue strength than neat polymer, shortened fatigue life and higher susceptibility to cyclic creep.

Keywords Fatigue strength · Natural fiber · Biopolymer · Biocomposite · Cyclic loading

Introduction

In the past years, a number of studies were carried out on experimental analysis of responses of polymers and polymer composites subjected to cyclic loading. The researchers focused on synthetic polymers reinforced with glass or carbon fibers for structural applications, which are often exposed to fatigue loads [1–6]. However, research on mechanical properties of biopolymers and their composites filled with natural fibers is usually carried out under static conditions and their fatigue properties are neglected. Meanwhile, some of these biocomposites can be used in long-term applications prone to vibrations and fatigue loads, like for example in automotive industry. It is also expected that thermoplastic composites with flexible viscoelastic lignocellulosic fillers may exhibit different mechanisms of energy dissipation and fatigue fracture than those filled with rigid brittle glass of carbon fibers.

Like in all engineering materials, failure often appears in plastics as a consequence of the initiation and propagation of cracks. However, it should be remembered that polymeric materials, especially thermoplastics, exhibit viscoelastic behavior with characteristic effects like creep, relaxation, energy dissipation. Those effects determine their dynamic properties and may influence failure mechanisms and fatigue life, especially if the material is subjected to stress-induced fatigue load mode at low cyclic frequency or if the material is prone to hysteretic heating [6]. In the case of composite materials under cyclic loading, failure is generally associated with the initiation and propagation of cracks in the matrix and the loss of adhesion at the polymer/matrix interface. This aspect of the interface quality between polymer matrix and natural or synthetic fibers have been already investigated [7–12] and thus will not be closely discussed in the experimental part of this study. The brittleness of fibers and their cracking, the differences in fiber and matrix mechanical properties are the further factors that influence dynamic and fatigue behavior of a material. Fatigue life of a polymeric composite is also affected by a number of test parameters. These include:

✉ Paulina Kuźniar
pkuzniar@pk.edu.pl

[1] Institute of Applied Mechanics, Tadeusz Kościuszko Cracow University of Technology, ul. Warszawska 24, 31-155 Kraków, Poland

[2] Institute of Materials Engineering, Tadeusz Kościuszko Cracow University of Technology, ul. Warszawska 24, 31-155 Kraków, Poland

stress or strain amplitude of the loading cycle; mean stress of the cycle; frequency, temperature and environment of the test [6, 13–15].

To assess fatigue life of polymers, Wohler curve (S–N curve) is usually used and there is a lot of publications presenting such results also for short fiber thermoplastic composites. An S–N curve provides information about the amount of cycles at which the material will fail at different maximum stress levels and allows for determining fatigue limit if such value exists for the tested material. To plot the S–N curve, several long-lasting tests on similar specimens with different levels of stress must be carried out: as a consequence, considerable time is needed [16–18].

For the purpose of approximate determination of fatigue strength, simplified time-saving Lehr's method can be applied, instead of more expensive and laborious Wohler's method. Lehr's method [19–21] was first created for metal testing and is based on the observation that fatigue failure is a consequence of local plastic strains which start to appear after exceeding fatigue strength in individual grains of metal before exceeding yield strain of a whole specimen. This process of failure is accompanied with sudden increase of dissipated energy, temperature and extension or other characteristic parameters. The essence of the method is to record or calculate these parameters in the accelerated fatigue test under conditions of cyclic loading of gradually increasing amplitude and then plot them as a function of maximum stress. This plot is then used for fatigue strength determination as it is graphically shown in Fig. 1. This method was implemented for polymers in Cracow University of Technology. It allowed for determination of the fatigue properties of polyamide filled with mineral tuff filler [22] and composites based on polyhydroxybutyrate filled with natural fibers [23].

The literature on the topic of fatigue properties of polymer composites with natural fibers covers some aspects of fatigue behavior of materials based on narrow group of traditional petrochemical thermoset and thermoplastic matrices tested in long-term or accelerated testing modes. An example is an extensive research of Gassan on composites of epoxy resin, polyester resin and polypropylene with flax fiber and jute fiber yearn or textile [8, 9, 24, 25]. There the fatigue behavior was presented in the terms of one characteristic value, namely, specific damping capacity (dissipation energy/strain energy). With the results of accelerated fatigue testing the author provided information about the influence of fiber treatment, fiber mechanical properties and fiber fraction on fatigue life, fatigue strength and damping of the composites. Composite damping was reduced with increasing fiber fraction, with comparable rates for damage propagation. It was shown that a strong interface is connected with higher dynamic modulus and reduction in stiffness degradation with increasing load cycles and applied maximum stresses and the specific damping capacity had higher values for the composites with poor bonded fibers. In another research, thermosets filled with sisal fibers were tested by Towo et al. [11, 12] and here also the quality of fiber–matrix adhesion was the main topic of interest when discussing fatigue and dynamical properties. It was found that the effect of chemical treatment on the fatigue life is significantly positive for polyester matrix composites but has much less influence on the fatigue life of epoxy matrix composites. Thwe et al. [26] studied fatigue resistance of hybrid composites of polypropylene/bamboo fibers/glass fibers compared to neat PP and its composite with bamboo fibers. The authors found that unreinforced PP had longer fatigue life than the composites at the specified cyclic load levels, and that hybrid composite showed better fatigue resistance than bamboo fiber filled PP. In this research, S–N curves were used for the assessment of the materials properties. More recently, Tong et al. [27] studied fatigue properties of nonwoven hemp fiber mat reinforced polyester also with the analyze of the S–N curves.

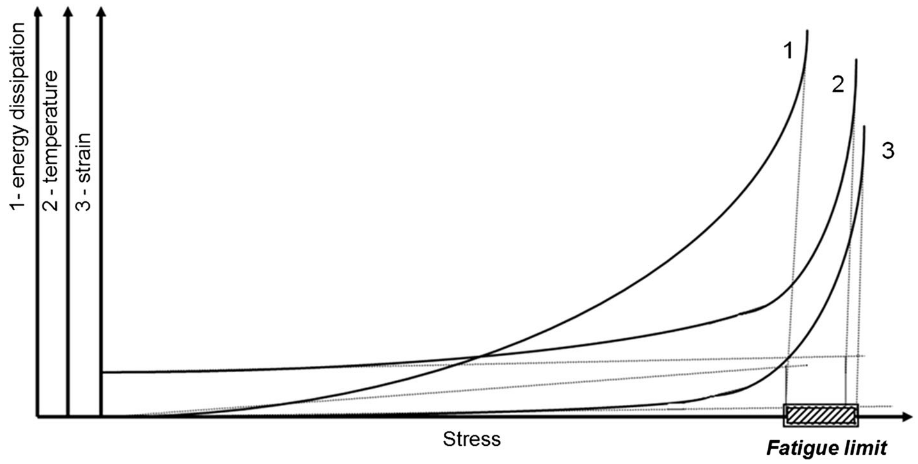

Fig. 1 A scheme of Lehr's method for time-saving fatigue strength determination

Present paper is concerned with experimental investigations of fatigue behavior of two biodegradable polymers and their composites filled with short flax fibers, tested in tension-tension accelerated fatigue test with the use of Lehr's method to compare fatigue strength of the materials.

Experimental

Materials

Two different commercially available biodegradable polymers were selected for testing: thermoplastic starch (TPS) BioCeres BC-LBI08 from FuturaMat, France and polylactide (PLA) blend Bio-Flex® F 6510 from FKuR, Germany. The materials were filled with 10 wt% chopped flax fibers. The fibers with diameter of 15–40 μm were provided by Experimental Station LENKON of Institute of Natural Fibers and Medicinal Plants in Poznan (Poland).

Standard dumbbell-type specimens were produced in Grupa Azoty SA in Tarnow, Poland, by injection molding using Engel ES 200/40 HSL. Granulates of composites with flax fibers were first prepared in Zakłady Azotowe by compounding on extrusion line with two-screw extruder MARIS TM 30VI with a gravimetric screw feeder.

The parameters of the injection process for BC-LBI08 thermoplastic starch were the following: injection temperature: 150 °C, mold temperature: 50 °C, injection pressure: 70 MPa, cycle time: 40 s (cooling time 20 s). For Bio-Flex® PLA blend parameters of the process were: injection temperature: 180 °C, mold temperature: 60 °C, injection pressure: 90 MPa, cycle time: 50 s (cooling time 20 s).

Methods

To determine basic mechanical properties of the materials tensile tests were conducted according to PN-EN ISO 527 at 21 °C and 65 % RH on a universal testing machine (Instron type 4465). Using a constant crosshead speed of 10 mm/min, five specimens were tested for each composite.

Stress-controlled tension-tension fatigue tests were performed at 23 °C using a hydraulic tensile test machine Instron 8511.20. The tests were done at constant minimum and maximum stress for a defined number of cycles (5000 cycles) and afterwards the maximum stress was increased to continue the fatigue test for the next 5000 cycles. This was repeated until fracture occurred. Thus, during the tests an amplitude was changed every 5000 cycle by an increase of a maximum stress level of 5 % of the material tensile strength (σ_M) starting from 30 % σ_M for each material. To remove margin of the testing machine, minimum stress level was set constant at 2 MPa. All fatigue tests were conducted at 5 Hz cyclic frequency with sinusoidal waveform. The specimens surface temperature was measured with pyrometer. Hysteresis loops were registered and then maximum strain and energy dissipated were calculated for every level of the maximum stress. To determine fatigue strength on the base of strain–stress curves or dissipated energy–stress curves, tangent lines to the curves were drawn. The average of the stress values at the tangent intersection for those two curves was the value of the fatigue strength in the modified Lehr's method used.

After these experiments, SEM images were made on the gold-sputtered fatigue-test fracture surfaces of the composites using JEOL JSN5510LV scanning electron microscope.

Results and Discussion

The biocomposites with PLA blend or TPS matrices reinforced with flax fibers were tested in static and cyclic conditions. Static tensile tests were carried out in order to determine basic mechanical properties of the materials and to gain data needed for further fatigue tests. Tensile properties (tensile strength σ_M, tensile modulus E_t and elongation at break ε_B) determined for the tested materials are shown in Table 1. Addition of short flax fibers caused an improvement in strength and stiffness, however the effect was not significant. This can be justified by the low content of the lignocellulosic filler (10 wt%) and the absence of chemical treatment of the fibers. An increase in modulus of elasticity in the case of the composite of PLA blend comparing to neat polymer was higher than for starch based composite. The experimental values of tensile strength of the biopolymers and biocomposites allowed for determination of the stress levels established for the accelerated fatigue tests, which results are presented in Figs. 2, 3, 4, 5, 6, 7, 8, 9, 10 and 11 and Table 2.

It can be seen in Table 2 that a maximum cyclic stresses carried by the materials for given testing mode were from 60 to 65 % of the tensile strength. Flax fibers addition to the starch-based matrix resulted in a decrease of maximum cyclic stress level from 65 to 60 % σ_M at lower number of cycles to failure, comparing to the neat TPS. In the case of

Table 1 Basic mechanical properties of tested materials

Materials	E_t (MPa)	σ_M (MPa)	ε_B (%)
TPS (Bioceres BC-LBI08)	2372 ± 116	15.2 ± 0.9	19.0 ± 3.5
TPS/10F	2976 ± 45	17.8 ± 0.6	2.9 ± 0.2
PLA (Bio-Flex F6510)	2238 ± 13	44.1 ± 0.2	127 ± 39
PLA/10F	3214 ± 74	45.8 ± 0.4	3.9 ± 0.7

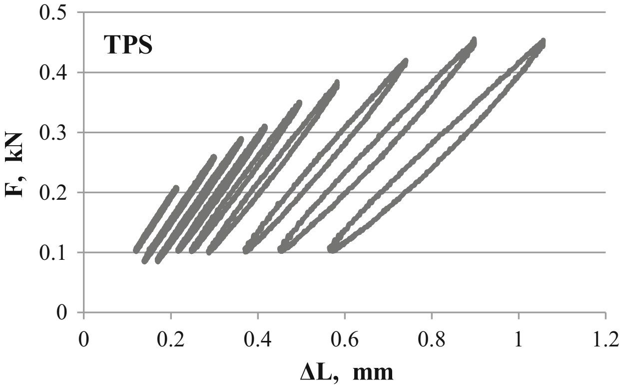

Fig. 2 Hysteresis loops for increasing values of stress for neat thermoplastic starch

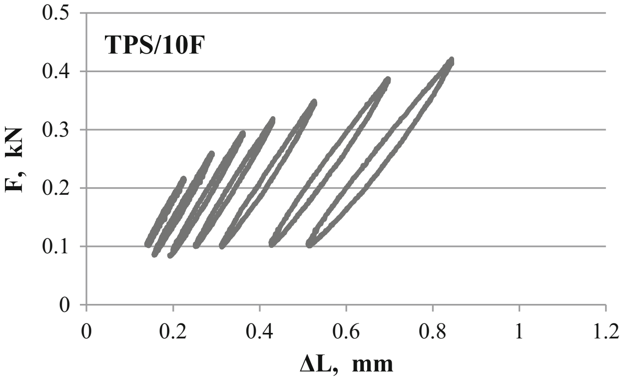

Fig. 3 Hysteresis loops for increasing values of stress for TPS filled with flax fibers

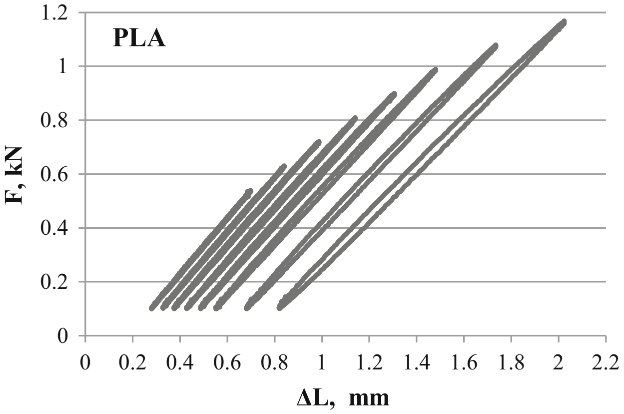

Fig. 4 Hysteresis loops for increasing values of stress for neat PLA blend

polylactide-based composite, this negative effect was not observed.

In Figs. 2, 3, 4 and 5, exemplary hysteresis loops for the tested composites and neat biopolymers are compared. To make the graphs more legible, only one loop was shown for each level of stress. Analyzing hysteresis loops recorded during the fatigue test, an increase of loops surface and mean strain as well as stability of the slopes of hysteresis

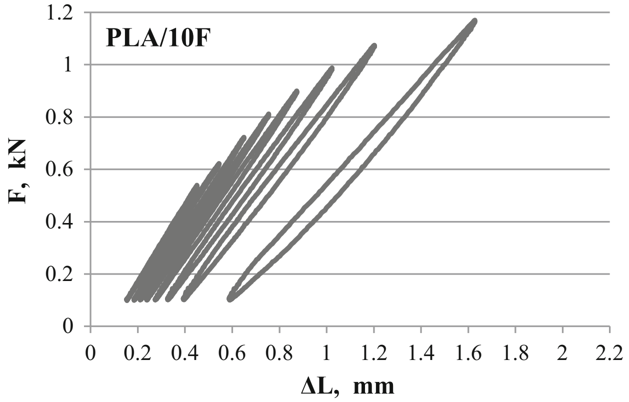

Fig. 5 Hysteresis loops for increasing values of stress for PLA blend filled with flax fibers

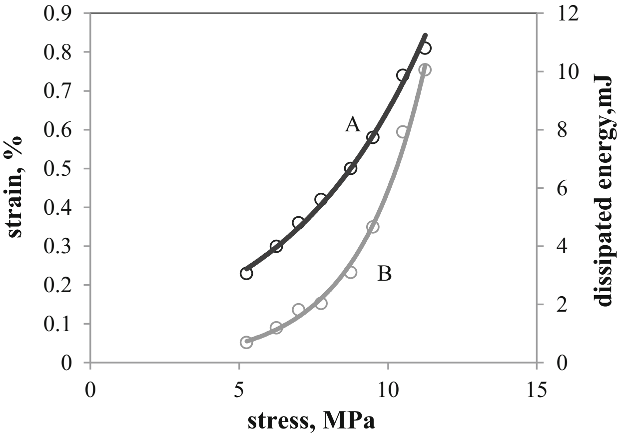

Fig. 6 Curves of strain (*A*) and dissipated energy (*B*) versus stress for TPS

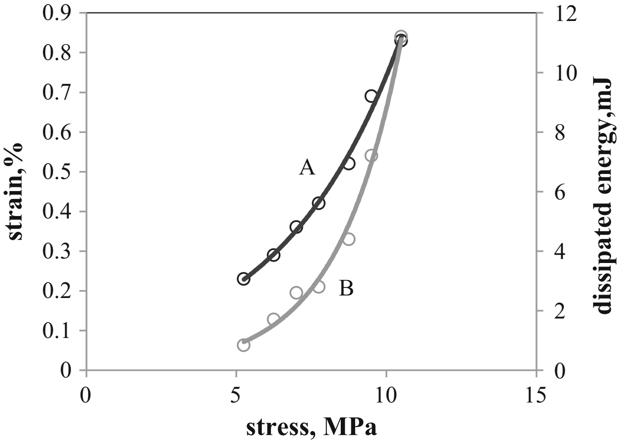

Fig. 7 Curves of strain (*A*) and dissipated energy (*B*) versus stress for TPS/10F

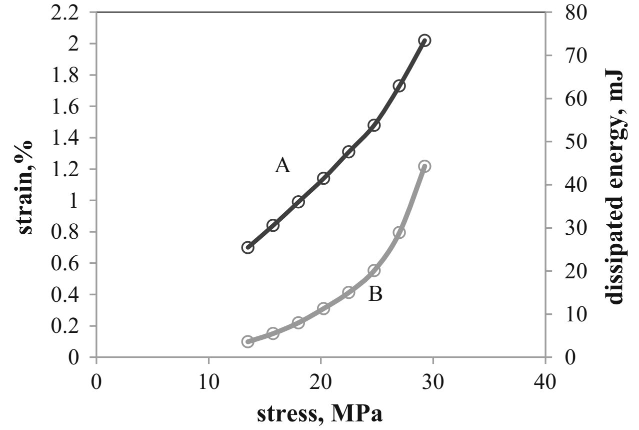

Fig. 8 Curves of strain (*A*) and dissipated energy (*B*) versus stress for PLA

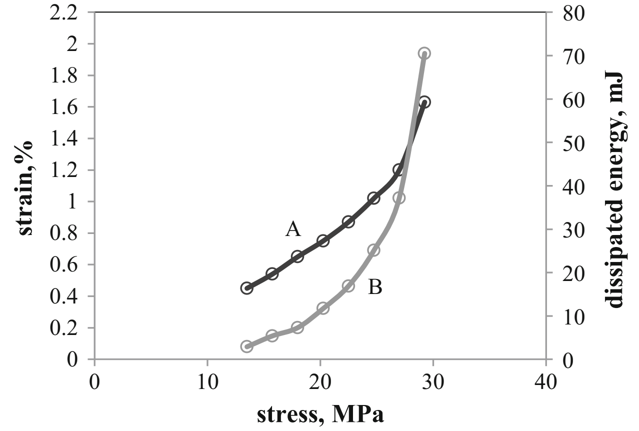

Fig. 9 Curves of strain (*A*) and dissipated energy (*B*) versus stress for PLA/10F

loops under increasing cycles number were observed for all of the tested biopolymers and biocomposites. The surface of hysteresis loop is a measure of an energy dissipated during deformation, and thus informs us about damping properties of the material an its stabilization/destabilization. Generally, damping properties of all tested composites were found to be similar to that of the neat biopolymers. The increase in elongation is a tribute to creep effects which add to the factors influencing fatigue life of the materials. However, it is important to note that the temperature measured on the specimens during the tests remained at a steady level so the effects of hysteretic heating were not observed. This may be also confirmed by a constancy of the hysteresis slopes mentioned above [28]. The same effect was observed by the authors previously for polyhydroxyalkanotes and their composites [23]. The failure of the materials was thus not induced by the significant decrease in mechanical properties due to local heat generation but with initiation of cracks and fatigue crack growth accompanied with creep.

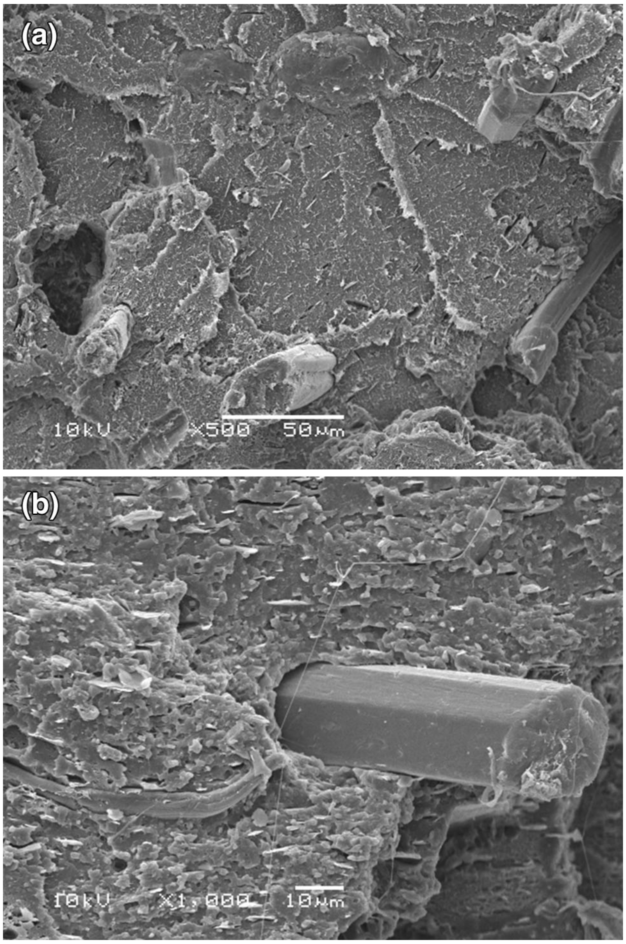

Fig. 10 SEM image of fatigue test fracture surface of composite on the base of PLA filled with 10 % flax fibers, **a** ×500, **b** ×1000

On the base of recorded hysteresis loops for every level of maximum stress, mean strain and energy of dissipation were calculated (as average values of the test results for all tested materials) and plotted as a function of stress as it is shown in Figs. 6, 7, 8 and 9. This graphical representation facilitates the analysis of the fatigue tests results. Adding flax fibers to PLA blend caused a decrease in mean strain for the same level of stress comparing to the neat biopolymer (Figs. 8, 9). The cyclic creep effect was reduced while the amount of dissipated energy significantly increased in the last phase of the tests before failure, most probably because of intensive destabilization of structure caused by fibers debonding and cracking. The opposite effect can be observed for TPS and its composite with flax where the addition of fibers resulted in slight increase in mean strain and caused no statistically important change in dissipated energy. It suggests that in this case fatigue behavior of the composite is not affected with the small

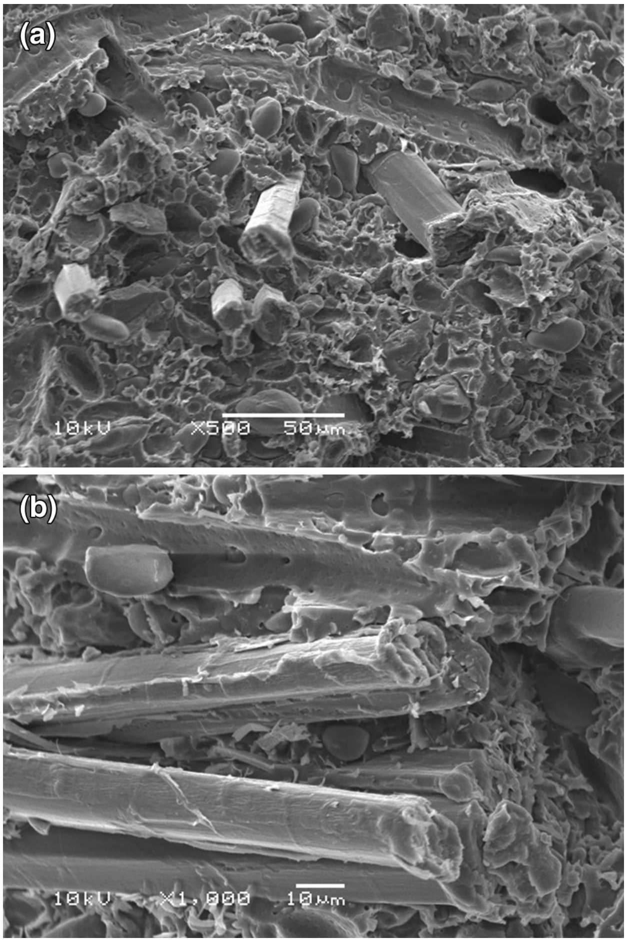

Fig. 11 SEM image of fatigue test fracture surface of composite on the base of filled with 10 % flax fibers, **a** ×500, **b** ×1000

addition of flax fibers and is almost entirely dependent on the matrix. The difference between TPS and PLA based composites hinders drawing general conclusions on the influence of short natural fibers on biopolymers or other thermoplastics. The database of test results obtained for different kinds of such natural fiber composites should be built up for more in depth analysis.

The plots in Figs. 6, 7, 8 and 9 allowed us also to use Lehr's method for determination of fatigue strength. The values of fatigue strength calculated in modified Lehr's method for TPS was 8.8 ± 0.5 MPa and for its composite it was 8.2 ± 0.3 MPa. For PLA blend the value of fatigue strength was more than two times higher than for TPS (22.2 ± 0.5 MPa) and for PLA/flax composite the value was even enhanced (25.5 ± 0.7 MPa).

PLA-based composite under conditions of cyclic loading exhibited higher ability to dissipate energy and higher fatigue strength compared to unmodified polymer. In the case of tested materials, modification with flax fibers was more advantageous in case of PLA blend than for TPS, both in static and dynamic loading conditions. It can be preliminarily assumed that on the contrary to the tested TPS and its biocomposite, the short flax fiber reinforced PLA blend may be applied as a structural material for low-loaded elements of, for example, car interior panels, small household goods or even rehabilitation equipment.

In Figs. 10 and 11, SEM images of fractured surfaces after fatigue tests are compared. It can be seen that some fibers are pulled out especially in the case of TPS based composite and discontinuities between fibers and matrices can be noticed. Taking mechanical test results into consideration, it seems that interactions between fibers and matrix are stronger for PLA-based composite than for TPS-based one where the fibers were more easily pulled out from the matrix and they did not significantly affected fatigue failure mechanisms, however probably they were responsible for faster fatigue crack initiation and propagation comparing to neat TPS. In case of both biocomposites, the effect of intensive deformation near the fibers and wide voids at the interface are not observed in Figs. 10 and 11 which confirms the lack of hysteretic

Table 2 Maximum loads set in the tests and the number of cycles recorded for the levels of stress from 0.3 σ_M to failure

The level of σ_M	0.3	0.35	0.40	0.45	0.5	0.55	0.6	0.65
TPS								
Maximum load (kN)	0.21	0.25	0.28	0.31	0.35	0.38	0.42	0.45
Number of cycles (increasing)	5000	10,000	15,000	20,000	25,000	30,000	35,000	**39,998 ± 1400**
TPS/10F								
Number of cycles (increasing)	5000	10,000	15,000	20,000	25,000	30,000	**30,657 ± 920**	–
PLA								
Maximum load (kN)	0.54	0.63	0.72	0.81	0.9	0.99	1.08	1.17
Number of cycles (increasing)	5000	10,000	15,000	20,000	25,000	30,000	35,000	**35,837 ± 837**
PLA/10F								
Number of cycles (increasing)	5000	10,000	15,000	20,000	25,000	30,000	35,000	**38,140 ± 1000**

Bold values indicate the numbers of the last cycle recorded during the tests (average value and standard deviation) heating effect that usually leads to high local plastic deformation.

Conclusions

Tests results confirmed the possibility of using time-saving Lehr's method for tested polymer composites based on biodegradable polymers. Under cyclic loading, two of three of the characteristic values measured, namely, strain and dissipated energy, increased significantly in the function of number of cycles. There was no important increase in the temperature measured on the samples surfaces and the effect of hysteretic heating was not observed. However the results allowed for the use of Lehr's method for the determination of fatigue properties of tested materials. The method was found useful for the preliminary comparison of the composites and neat matrices. Maximum cyclic stress carried by the tested materials was at the level of 60–65 % of their tensile strength. And average fatigue strength acc. Lehr's method was from 46 (TPS/10F) to 56 % (for TPS) of the materials tensile strength. The influence of flax fibers addition (10 wt%) was not significant, minor improvement was observed for PLA/flax composite. In the case of tested materials, modification with flax fibers was more advantageous in case of PLA blend than for TPS both in static and dynamic properties. Fracture process in tension-tension cyclic loading for the tested materials was mainly attributed to the matrix features. Shortened fatigue life of TPS/flax composite may outcome from lack of adhesion between the matrix and fibers leading to more intense crack initiation and propagation. This statement also finds confirmation in SEM analysis of TPS/flax fatigue facture surfaces showing agglomerates of pulled-out fibers.

There is still a need for research on thermoplastic polymer composites with natural fibers, including bio-based composites. Accelerated fatigue testing is a useful tool to recognize basics of fatigue behavior of those important materials.

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