

Effect of flax fibers addition on the mechanical properties and biodegradability of biocomposites based on thermoplastic starch

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Abstract: The research was intended to develop a biocomposite as an alternative biodegradable material, for the production of, e.g., disposable utensils. The author's tested thermoplastic maize starch, both without additives and with the addition of crumbled flax fiber in the share of 10, 20 and 30 wt%. The plasticizer added was technical glycerin and the samples were produced by a single-screw extruder. The mechanical strength tests were performed, including the impact tensile test and three-point bending flexural test. Afterwards, the samples were tested for biodegradability under anaerobic conditions. The methane fermentation process was carried in a laboratory bioreactor under thermophilic conditions with constant mixing of the batch. All samples proved to be highly susceptible to biodegradation during the experiment, regardless of the flax fiber share. The biogas potential was about 600 ml·g⁻¹, and the methane concentration in biogas ranged from 66.8 to 69.6%. It was found, that the biocomposites can be almost completely utilized in bioreactors during the biodegradation process. The energy recovery in the decomposition process with the generation of significant amount of methane constitutes an additional benefit.

Introduction

New regulations related to the processing and marketing of the plastics products manufactured with modified natural polymers, or the plastics manufactured from bio-based, fossil or synthetic starting substance have been introduced in the European Union (Directive EU 2019/904). From 2021 onward, the marketing of single-use products such as cutlery, plates, drinking straws, stirrers for drinks or sticks made of plastic materials will not be allowed. Until now, these products have been manufactured from degradation-resistant artificial polymers, which break down in the natural environment for a very long time. In the near future, the composites of non-renewable and non-degradable polymers may be replaced with the composites of bio-based polymers reinforced with natural fibers of plant materials (Ibrahim et al. 2013).

Presently, many global companies are introducing the single-use disposable products from various types of biodegradable materials. Particular attention was paid to the study on a group of natural materials produced from starch (Bootklad and Kaewtatip 2013). This compound is perceived as a low cost substrate having the potential for replacing the oil-

based polymers on a large scale (Moad 2011). The biopolymer is obtained after mixing the starch with a plasticizer (often glycerine) and heating the material to a temperature lower than the starch degradation temperature, ranging from 70 to 80°C (Mitrus et al. 2016). The starch after plasticization is called thermoplastic starch (Xie et al. 2013, Oniszcuk et al. 2019).

Products with the share of thermoplastic starch have already started reaching our homes, while manufacturers are looking for newer and better technologies for their production. These starches are fully biodegradable, which solves the problem connected with the disposal of used products (Nafchi et al. 2013). They can be subjected to the composting process and used for agricultural purposes (Feuilloley et al. 2005) or decomposed under anaerobic conditions in order to obtain methane used as fuel for the heating purposes (Pagliano et al. 2017) or fuel for motor and generator drive (Ueno et al. 2007). Methane is a powerful greenhouse gas emitted by human activities such as leakage from natural gas systems and the raising of livestock, as well as by natural sources such as wetlands. It has a direct influence on climate, but also a number of indirect effects on human health, crop yields and the quality and productivity of vegetation through its role

as an important precursor to the formation of tropospheric ozone (Reay et al. 2007).

In order to improve the physical properties of biocomposites, and sometimes to reduce the price of the finished product, various types of plant fiber fillers are added to starch material. The most commonly used ones are fibers from flax, hemp, jute, lignocellulose, coir, cotton and oriental plants (Ma et al. 2005, Kaushik et al. 2010, Gurunathan et al. 2015).

This work presents the method of producing biocomposites and the procedures of their testing for use as an alternative material for disposable products. The aim of the study was to assess the strength properties and biodegradability under anaerobic conditions of the products obtained as a result of extrusion-cooking of thermoplastic maize starch with the addition of flax fiber. The additional solution was to check the possibility of utilization of these products for energy recovery in the form of methane.

Materials and methods

Raw materials

The investigated material was maize starch of MERIZET 100 (Segezha, Ireland) with the chemical composition formula $C_6H_{10}O_5$ and 16% moisture content. The plasticizer was technical glycerin with a purity of 99%, added to the starch in an amount of 20 wt%. The samples were prepared with plant additives in the form of flax fiber waste with the chemical formula of $C_{23}H_{38}O_{17}N$, according to Haug (1993). Several plant species (sisal, jute, palm tree etc.) were examined but flax attracted the greatest interest (Romhány et al. 2003). Compared with other plants, flax is cheap, grown in Poland, and has numerous beneficial properties. Single flax fibers have a density of $1.5 \text{ g}\cdot\text{cm}^{-3}$, an elongation to break of 2.4%, a tensile strength of 1100 MPa, and a Young's modulus of 100 GPa. The approximate chemical composition of flax fiber in wt% is: cellulose 64.1, hemicellulose 16.7, pectin 1.8, lignin 2.0, water soluble compounds 3.9, fat and waxes 1.5 (Foulok et al. 2006).

In the study the authors used flax (*Linum usitatissimum* L.) of the Szafir variety, obtained from a Polish agricultural producer – Mokranoy Nowe Agricultural Farm. The flax fibers were crushed using a beater chopper type H-111/3 on sieves with 4 mm mesh size for an average particle size of 1.2 mm. After mixing with the plasticizer using a laboratory

ribbon mixer, the material was placed in tightly sealed bags, but immediately before the extrusion it was mixed again. Just after extrusion the samples were stored for about two hours in an air-dryer at air humidity of 45% and temperature of 24°C. Then, the samples were cured for at least 72 hours under standard conditions (25°C at 1000 hPa). The share of flax fibers in the prepared samples was 10, 20 and 30 wt% as starch replacement. The samples were marked as follows (Figure 1): S0 – pure plasticized starch without plant additives, S1 – starch with the addition of 10 wt% shredded flax fiber, S2 – starch with the addition of 20 wt% shredded flax fiber, and S3 – starch with the addition of 30 wt% shredded flax fiber.

Samples preparation

The blends of materials were put into the feeding system of a TS-45 type modified single-screw extruder (ZMCh Metalchem, Gliwice, Poland). Generally, twin-screw extruders are used to obtain better mixing and distribution of the fillers (Battagazzore et al. 2016), but the construction modification designed as a Polish Design Patent (Juško et al. 2009) was adequate for using in a single-screw extruder for successful extrusion (Figure 2). The extrusion channel was divided into three sections: feeding zone, transition or kneading zone, and cooking or metering zone. The starting material was fed from a hopper directly into the feed section, which has deeper flights or flights of greater pitch. This geometry enables the feed material to fall easily into the screw for conveying along the barrel (Wójtowicz 2008). The pitch and helix angle determine the throughput at a constant rotation speed of the screws. The material was transported as a solid plug to the transition zone where was mixed, compressed, melted and plasticized. Compression developed by decreasing the thread pitch but maintaining a constant flight depth result in an increased pressure as the material moves along the barrel. The melt moves by circulation in a helical path by means of transverse flow, drag flow, pressure flow and leakage. The helix-screw was equipped with an additional mixing section, and owing to this modification a preferable composite compound can be obtained. The extruder working section consisted of two heating zones and a cooling section, where the following temperatures were used: 1st section – 80°C, 2nd section – 100°C, 3rd section – 60°C. The extruder was equipped with a material dispenser, a plasticizing system connected to the drive, an electric cylinder

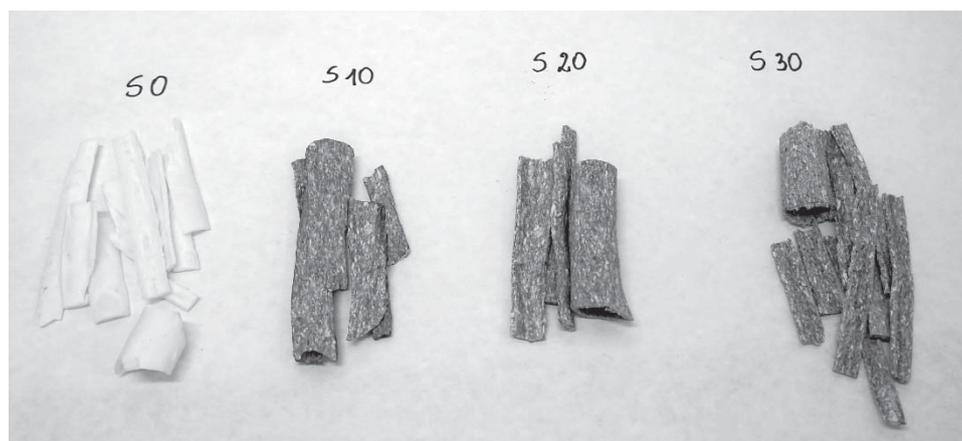


Fig. 1. The view of the samples (Photo by G. Borowski)

heating system and a cooling section. The applied plasticizing system was equipped with a helix-screw of length (L) and diameter (D) in the ratio $L/D = 18/1$ (Figure 3), and with a steel die mold. The rotational screw speed was set at 60 rpm. The screw speed was controlled using the DM-223AR electronic tachometer. The temperature was measured by means of the AZ8852 instrument installed in the extruder cylinder. The use of intensive cooling in the last working section of the extruder influenced the stabilization of the biocomposite and limited the degree of sample expansion. The visual extruded product had a homogeneous structure, without bubbles or cracks.

In tests the samples of cuboid shape were used, in sizes $4 \times 10 \times 80$ mm. They were cut from the wall of tube shaped product, with a diameter of 17 mm.

Mechanical properties

The examination of mechanical properties included two strength tests: (1) impact tensile test, and (2) three-point bending *flexural* test. The impact tensile test consisted of tearing the sample through a one-off impact using a QC 639F Mechanical Impact Tester, Charpy type (Figure 4a). The impact energy was 2 J, the pendulum impact speed by the sample was $2.9 \text{ m}\cdot\text{s}^{-1}$. The methodology of sample preparation and determination of impact tensile strength was in accordance with the PN-EN ISO 8256:2006 standard (Plastics – determination of impact tensile strength). The sample after the impact tensile test is shown in Figure 4b.

The three-point bending *flexural* test was performed to determine the rigidity of the biocomposite material, which is a brittle material characterized by small values of relative elongation, most often up to 1–5%. Figure 5 shows the trend of four samples variations of the strain curves depending on the axial force and displacement of the device head. These curves revealed higher values of the bending force of the S1 and S2 samples, compared to the S0 and S3 samples, which turned out to be more susceptible to deflection. The tests were carried out with a Zwick Z010TN (Germany) testing machine. The *grip distance* was 50 mm, a load cell was 10 kN, the crosshead speed was set to 50 mm/min. The samples for mechanical tests, in addition to the application of the load and the location of the support points were resolved in accordance with the PN-EN ISO 14125:2001 standard (Fiber-reinforced plastic composites – determination of bending properties), for fiber-reinforced plastics and polymer composites.

The value of flexural strength was calculated according to the relationship:

$$\sigma_M = Mg/Wx \text{ (Pa)} \quad (1)$$

where: Mg – bending moment ($\text{N}\cdot\text{m}$), Wx – flexural indicator (m^3).

The measurements of Young's modulus, flexural strength, total deformation and the vertical *deflection* were repeated

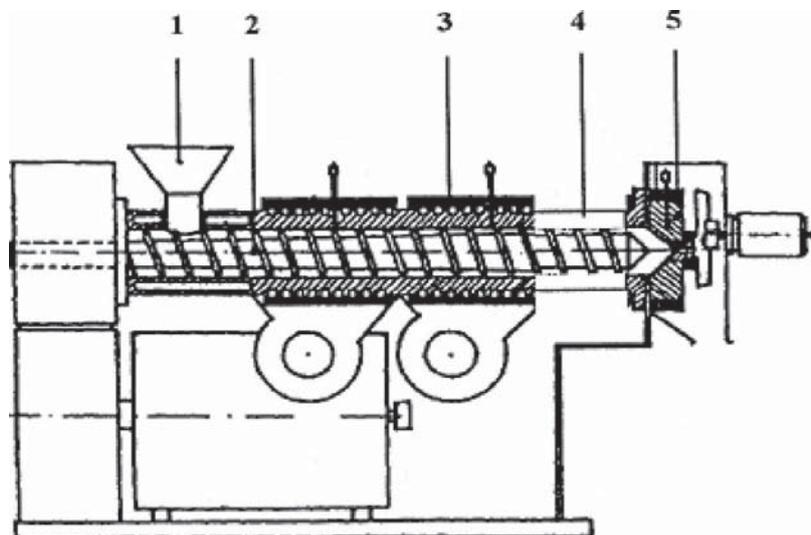


Fig. 2. Schematic diagram of single screw extrusion-cooker TS-45 [Wójtowicz 2008]:

1 – dumping hopper, 2 – screw, 3 – heating zone, 4 – modified section including screw and barrel extension and cooling section, 5 – head with shaping die

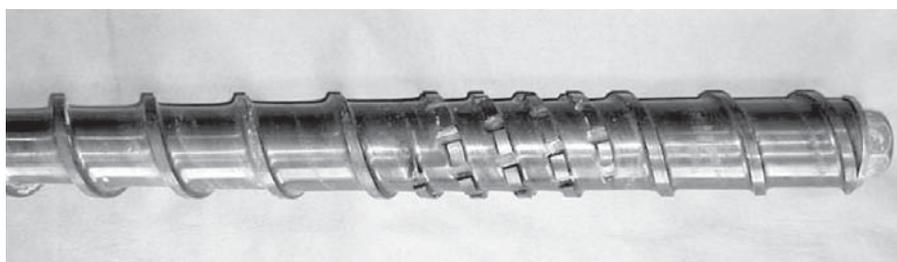


Fig. 3. The view of extruder screw (Photo by T. Oniszczyk)

fivefold for each composite, and then the average values were calculated. But for statistical analysis at least six replicates of each test are recommended [Hietala et al. 2013].

Biodegradability tests

The biodegradability tests of the samples were carried out under laboratory conditions, in a batch arrangement, in the bioreactors with a total volume of 2 L (BioReactor Simulator BRS, Bioprocess Control, Sweden) with an automatically

controlled system of mixing the feedstock and measuring the amount of the produced biogas (Figure 6). A digestate from an agricultural biogas plant was used as an inoculum. The inoculum to substrate ratio was 3:1 on dry organic matter basis. The basic parameters of the examined biocomposites are presented in Table 1. These tests were carried out with the thermogravimetric analyzer (TGA 4000 by Perkin Elmer), at a rate of 10°C/min from ambient to 650°C, under a nitrogen atmosphere with a 20 ml/min flow rate.

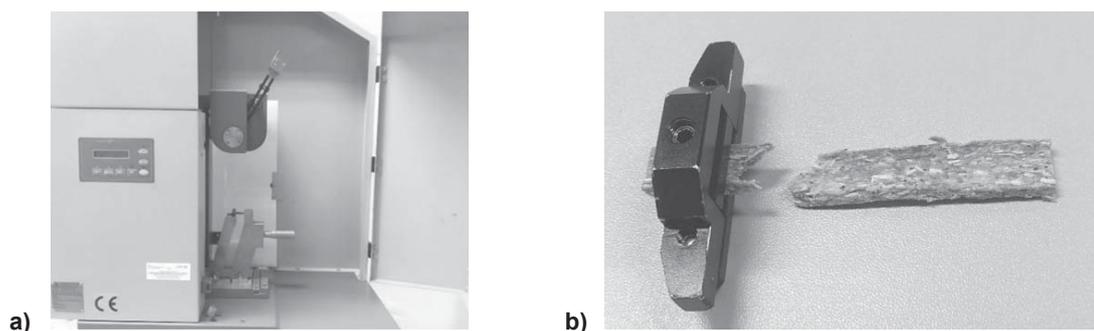


Fig. 4. Stand with Charpy QC 639F hammer (a) and the sample after the impact tensile test (b) (Photo by T. Klepka)

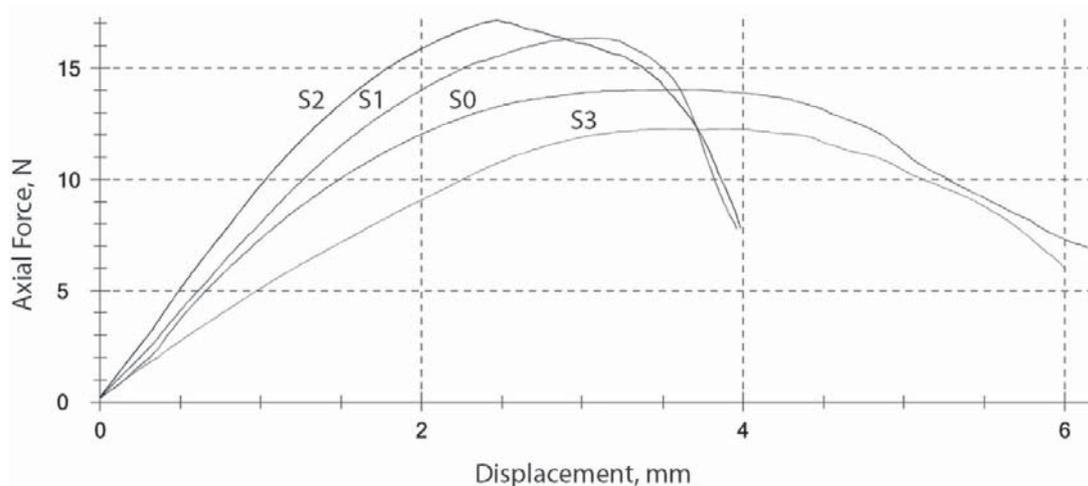


Fig. 5. The trend of the strain curves in the three-point flexural test for samples: S0, S1, S2, and S3

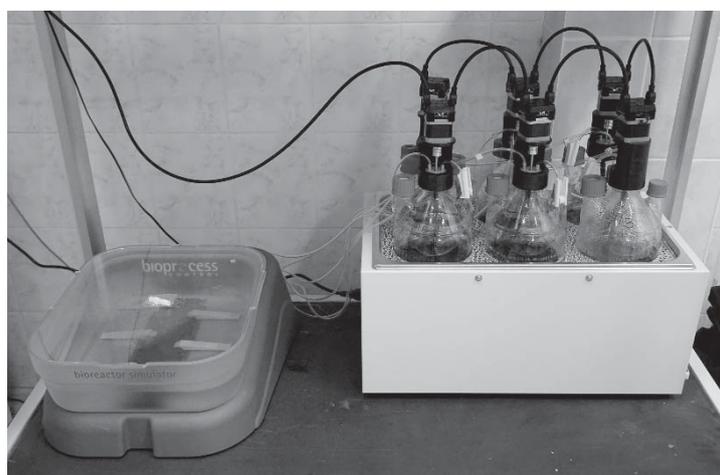


Fig. 6. The simulator of bioreactor set with water bath (Photo by G. Borowski)

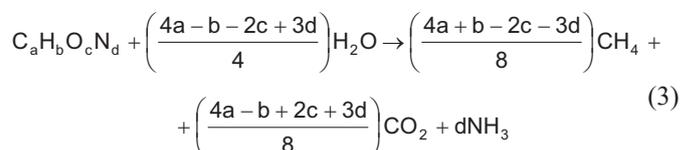
After placing the biocomposites samples (10 g) in the reactors and mixing with the inoculum (1200 ml of digestate from biogas plant) and sealing them, the headspace above the feedstock was flushed with nitrogen to create anaerobic conditions. The anaerobic digestion process was carried out under thermophilic conditions (at a temperature of $55 \pm 1^\circ\text{C}$) with constant mixing of the batch. The experiment was conducted for four weeks. After its completion, the following parameters were determined:

- Biogas potential (GP) of materials, which was expressed as gas volume (under normal conditions) with reference to the dry matter unit of material introduced into the bioreactor;
- Concentration of methane and carbon dioxide in biogas in the final phase of the experiment using a Trace GC Ultra gas chromatograph (Thermo Fisher Scientific, Italy) with FID detector. The volumes of these gases were expressed in relation to normal conditions (temperature 273.15 K and pressure 1013.25 hPa = 1 atm);
- Biodegradability of the material (f_d), which was calculated according to formula (Win et al. 2018):

$$f_d = \frac{GP}{Bu} \quad (-) \quad (2)$$

where: GP – biogas potential determined experimentally ($\text{ml} \cdot \text{g}^{-1}$), Bu – theoretical biogas potential ($\text{ml} \cdot \text{g}^{-1}$).

The value of theoretical biogas potential (Bu) was calculated from the Buswell and Neave equation (Eq. 2), that shows a scheme of the anaerobic decomposition of the organic matter, expressed by a summary formula $\text{C}_a\text{H}_b\text{O}_c\text{N}_d$ (Win et al. 2018):



where symbols a , b , c , and d mean the number of atoms of particular elements in the molecule of an organic compound.

The biodegradability tests were carried out in triplicate and then average values with standard deviations were calculated. Due to the small number of repetitions, the wider statistical analysis of the results was not made.

Results and discussion

Mechanical properties

The results from impact tensile tests of biocomposite samples with the Charpy hammer are presented in Table 2. In turn, the results from flexural tests of the considered samples on the strength machine are presented in Table 3.

On the basis of the results of the mechanical tests it was found that the S3 sample has the lowest impact value, so

Table 1. The content of dry mass and organic dry matter in the studied biocomposites

Sample type	The share of flax fiber [wt%]	Dry mass [% by weight]	Organic dry matter [% by dry matter]
S0	0	97.14 ± 0.02	99.82 ± 0.02
S1	10	96.36 ± 0.15	97.65 ± 0.05
S2	20	96.53 ± 0.42	98.88 ± 0.05
S3	30	96.65 ± 0.19	98.73 ± 0.04

Table 2. The average values of impact tensile tests of biocomposites

Sample type	The share of flax fiber [wt%]	Hammer deflection angle [°]	Work [J]	Impact strength [$\text{kJ} \cdot \text{m}^{-2}$]
S0	0	128.60 ± 1.00	0.26 ± 0.02	8.36 ± 0.45
S1	10	132.57 ± 1.13	0.20 ± 0.01	9.02 ± 0.55
S2	20	126.83 ± 0.97	0.29 ± 0.03	9.39 ± 0.58
S3	30	127.09 ± 1.01	0.28 ± 0.02	8.21 ± 0.42

Table 3. Average values of flexural test results of biocomposites

Sample type	The share of flax fiber [wt%]	E [MPa]	σ_M [MPa]	ϵ [%]	h [mm]
S0	0	150 ± 5.3	5.48 ± 0.41	7.30 ± 0.14	3.72 ± 0.05
S1	10	255 ± 8.7	7.72 ± 0.49	2.60 ± 0.06	3.48 ± 0.06
S2	20	435 ± 10.1	8.35 ± 0.55	4.40 ± 0.09	3.42 ± 0.05
S3	30	404 ± 11.4	6.41 ± 0.45	5.00 ± 0.11	3.97 ± 0.07

Note: E – Young's modulus, σ_M – flexural strength, ϵ – total deformation, h – vertical deflection.

this sample is the most susceptible to destruction (fracture) due to the highest amount of flax fiber, which may be responsible for weakening the structure of the extruded products (Migneault 2009). While studying the results from Table 3, it can be observed that the highest flexural strength with the least total deformations was obtained for the S1 and S2 samples.

Saiah et al. (2009) investigated wheat starch composite with flax fiber from 0 to 20 wt% and glycerol (22 wt%). They found that the flexural strength and tensile modulus increase along with the fiber content. For the composite with 20 wt% flax fiber, the values of flexural strength reached 8.9 MPa and Young's modulus was 465 MPa; these values were very comparable to the results obtained in this study – 8.35 MPa and 435 MPa, respectively (Table 2).

It was found that testing composite of flax fiber in the share of 30 wt% (sample S3) led to obtaining lower flexural strength, comparing to the S2 sample. The values of flexural strength of the S3 sample and control sample without plant additives (S0) was similar. Therefore this sample was not included for the further biodegradability and biogas tests.

Thakur and Singha (2010) conducted the mechanical tests of the composite of lignocellulosic material filled with Pine needles. They showed that the mechanical properties of a particle reinforced composite was more effective comparing to short and long fibers. It goes against the general laws of composite mechanics, because usually long fibers contribute to higher performance of materials. Muthuraj et al. (2016) reported that good mechanical properties can be obtained from miscanthus fiber composites. The composites were prepared in a lab-scale extrusion and injection molding process. The values of flexural strength of miscanthus fiber composites resulted in approx. 35 MPa, which was four times greater than our result. In this case, the dimension of the tested specimen was not comparable, as for example the one tested by Averous and Boquillon (2004).

Biodegradability properties

The values of theoretical biogas potential of pure maize starch and plant biomass calculated on the basis of Buswell equation (Eq. 2) were 742.9 ml·g⁻¹ (including methanogenic potential,

371.9 ml·g⁻¹) and 750.7 ml·g⁻¹ (including methanogenic potential, 368.7 ml·g⁻¹), respectively.

The graphical results of cumulated biogas production during the experiment are shown in Figure 7. The highest total biogas production was obtained from the S0 control sample (thermoplastic maize starch) in comparison to the biocomposites with the addition of 10 and 20 wt% of flax fiber (S1 and S2, respectively). Russo et al. (2009) described that the higher amount of starch in the blend with plastics caused an increase in biogas production and methane concentration in the biogas, although starch biodegradation was inhibited by plastic addition.

The biogas potential of the S0 sample of 721 ml·g⁻¹ was about 20% higher than the S1 and S2 samples, which were characterized by quite similar values of this potential, being 613.0 and 590.3 ml·g⁻¹ respectively. The methane concentration in the produced biogas in the final phase of the experiment ranged from 66.78 to 69.64% (Table 4). For the produced biogas, the methane concentration was quite similar for all samples, and the differences were not really relevant, seeing that they did not exceed 3%. For all tested samples, the highest concentration of biogas produced was observed on the second day of the experiment, and then it decreased to almost zero on the fourth day (Figure 7). However, in the following days, the biogas production gradually increased to reach a local peak on the ninth day. Further local peaks were observed at intervals of 2–3 days. Therefore, it is confirmed that the biogas production is affected by the chemical composition of the material which was determined by the share of flax fibers.

The methane production in two-stage anaerobic digestion test resulted in much higher efficiency compared to one-stage anaerobic digestion (Lavagnolo et al. 2018). Similarly to our results, the composition of biogas changed during the anaerobic digestion, and the carbon dioxide concentration predominated over methane in the initial stage of the process. The individual measurements of the biogas composition do not allow calculating the methanogenic potential of the plant materials, because the value of cumulative methane production is necessary for this purpose (Montag and Schink 2016).

The course of the organic matter biodegradation and biogas production was similar for all the samples tested (Figure 8). Rapid material decomposition to gaseous products was observed

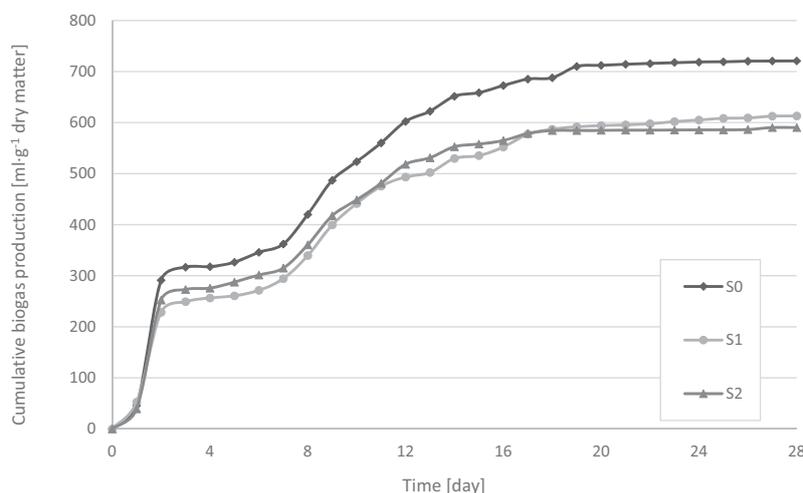


Fig. 7. Cumulative biogas production obtained during anaerobic digestion of biocomposites

during the first three days of the experiment (the highest value of biogas production was obtained in the second day), followed by the almost complete stop of biogas production, afterwards by a gradual increase the biogas production (with the local peaks on the 8th–9th days of the experiment). Banks et al. (2011) found that the highest total biogas production from food waste allows obtaining the methane content of around 62%, with was lower value compared to our results, up to about 69% (Table 4).

The rate of particular biocomposites biodegradation fluctuated during the course of the experiment, which is shown by the local peaks presented in Figure 8. This indicates a heterogeneous chemical composition of the compounds building the tested materials. A high number of local peaks were observed in all the materials. In the case of S1 and S2 (with the addition of 10 and 20 wt% of flax fibers, respectively) it can be explained by the complex composition of these materials, which are the mixture of the plasticized starch and plant tissues, composed of lignocellulose, proteins and other substances. For the sample containing only the maize starch (S0), the lower number of peaks of biogas production was expected, compared to the plant fiber biocomposites. In that

case, the observed time-dependent variations in the course of decomposition of S0 sample were caused by the thermal processing of material. Within 20 days of test, the majority of the biodegradable components were decomposed. Then, in the next days of the experiment, almost no biogas emerged (Figure 8).

The values of the theoretical biogas potential, calculated on the basis of formula (Eq. 3) and the experimentally obtained ones, as well as the biodegradability of the biocomposites, calculated on the basis of formula (Eq. 2) are presented in Table 5.

Contrary to the experimental results, the data shows that the theoretical biogas potential was the highest for the S2 sample and the smallest one for the S0 sample. The increase in theoretical biogas potential relates to the flax biomass addition, because it is characterized by a higher biogas potential (of 750.7 ml·g⁻¹) than pure maize starch (742.9 ml·g⁻¹).

The results presented in the Table 5 indicate that the difference between the values of the experimentally determined biogas potential and the theoretical potential increases along with the addition of flax fiber to the biocomposite. The maximal

Table 4. Concentration of methane and carbon dioxide in biogas measured in the final phase of the experiment

Sample type	The share of flax fiber [wt%]	Gas concentration [% vol.]	
		Methane	Carbon dioxide
S0	0	69.64 ± 0.01	27.26 ± 0.06
S1	10	69.62 ± 0.05	27.10 ± 0.06
S2	20	66.78 ± 0.02	27.21 ± 0.04

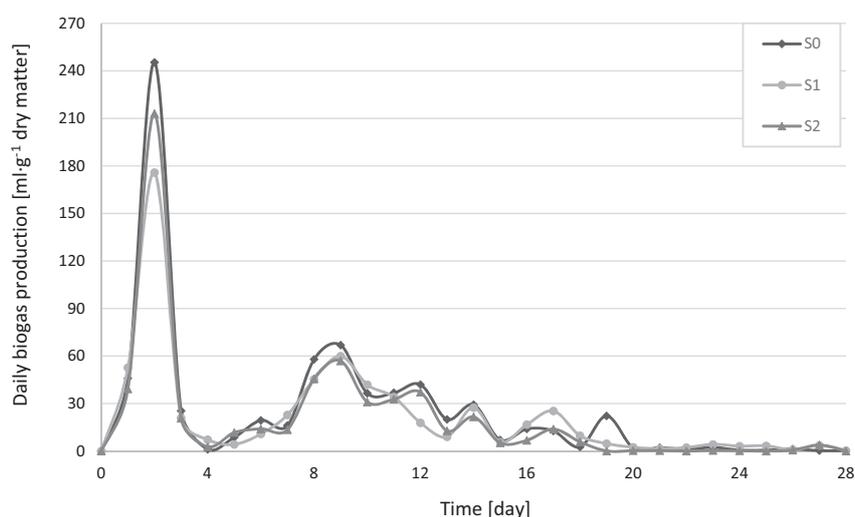


Fig. 8. The daily biogas production during the test

Table 5. The biogas potential and biodegradability of tested biocomposites

Sample type	The share of flax fiber [wt%]	Biogas potential [ml·g ⁻¹]		Biodegradability [-]
		Theoretical	Experimental	
S0	0	742.9	720.8	0.97
S1	10	743.6	613.0	0.82
S2	20	744.4	590.3	0.79

biodegradability under anaerobic condition turned out to be almost equal to the theoretical value calculated for free-addition biopolymer. The biodegradability of the biocomposite decreases slightly with the flax fiber share. It was noted that regardless of the share of plant additives, the biodegradation process of all biocomposites types finished within the period of 20 days. For the assessment of biogas production from various kinds of substrates, the period of 21 days is referred to as standard test in Europe (Lebiocka 2019).

Similarity to our results, very high biodegradability of biopolymers produced on the basis of polysaccharides was obtained by Ryan et al. (2018) who studied the composites produced with cellulose. Their test was conducted under anaerobic conditions in continuously stirred bioreactor, at a temperature of $37\pm 2^\circ\text{C}$. Complete biodegradation calculated on the basis of the mass loss of bioplastics was observed on the 28th day of the experiment. Comparing these results to our experiment, we observed a faster decomposition of the tested composites, owing to the higher temperature of decomposition process and the less complex bio-material used.

Conclusions

The studies on the mechanical properties shown that flax fiber mixed with the maize starch can be used successfully as a basis of biodegradable composites. The most favorable results of the impact tensile test, and three-point bending *flexural* test were obtained for a biocomposite with flax fiber of 20 wt%. Under anaerobic conditions of 55°C , the biodegradation process was finished within the period of 20 days, which fulfilled the requirements of European standardized tests. A high methane concentration (approx. 67% vol.) in the biogas was observed in the experiment. Nevertheless, most biogas was produced in the initial stage of the biodegradation process. A smaller share of plant additives increased the biogas potential and biodegradability of the tested biocomposites, however, their mechanical properties were much worse than expected. Nevertheless, it was confirmed that the flax fiber biocomposites formed by the extrusion process were proven to be a fully biodegradable material and can be successfully used for energy recovery through biogas production.

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