Original Research

Environmental Degradation of Ramie Fibre Reinforced Biocomposites

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Abstract

The estimation of environmental degradability of different ramie fibre reinforced biocomposites in Baltic Sea water and in compost with activated sludge under natural conditions is the subject of this paper. The characteristic parameters of environments were monitored during incubation time and their influence on the rate of degradation of biocomposites was discussed.

The degradation processes of two kinds of biocomposites, ramie fibre/Ecoflex® and ramie fibre/cellulose nanofibre reinforced corn starch resin, were studied by macro- and microscopic observations of polymer surfaces, changes of weight, and tensile strength during incubation under natural conditions. The macro and microscopic observations and weight changes confirmed the decrease of the tensile strength both of the composites after incubation in compost and in sea water.

It was demonstrated that the biocomposites with natural fibre of ramie were degraded in compost faster than in sea water, due to different microbial communities. The ramie/Ecoflex® biocomposite was degraded more distinctly than ramie/cellulose nanofibre reinforced corn starch resin biocomposite in both natural environments.

Keywords: environmental degradation, sea water, compost, ramie fibre

Introduction

With the large amount of litter contaminating our planet today, environmental concern has recently promoted research toward new environmentally friendly materials for a wide range of applications.

Natural fibre reinforced biocomposites are interesting alternatives to synthetic oil-based fibre enhanced polymer composites, because natural fibres are derived from a renewable resource, do not have a large energy requirement to process, and are light weight, have high specific modulus, and are biodegradable. At the end of their service life, they can be completely degraded in living environment or, alternatively, they can be incinerated for energy recovery in some cases [1].

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Natural fibres may be classified in two categories: non-wood (such as flax, hemp, jute, ramie) and wood fibres [2]. Ramie is a perennial herbaceous plant of the nettle family (*Urticaceae*) native to some Asian countries. Ramie fibres have been used for centuries because of their excellent fibre properties. Fibres from ramie are fine and silk-like, naturally white and lustrous. Ramie is characterized by its long length and great strength – much greater than that of cotton and silk. They are used for clothing fabrics, industrial packaging, twines, cordages, canvas, car interiors, etc. [3, 4].

Natural fibres, in composites, are usually coupled with a natural (such as starch, cellulose, polylactic acid, soybeans) or synthetic (such as polypropylene, polyester, polyurethane) matrix [5, 6].

The main tasks of the embedding matrix are to hold the fibres together (giving and stabilizing the shape of the structure), to transmit the shear forces between the fibres and to work as a coating to prevent the fibres from being damaged by environmental influence [5].

The aim of the present study was an examination of the environmental degradability of ramie/Ecoflex® biocomposite and ramie/cellulose nanofibre reinforced corn starch resin biocomposite in compost containing activated sludge and in Baltic Sea water.

According to standards, Ecoflex® as an aliphatic-aromatic copolyester is biodegradable, atoxic and useful in the composting process [7, 8]. The results of our earlier studies also revealed that the aliphatic-aromatic copolyester Ecoflex® was degraded in compost under natural weather conditions [9].

There were two reasons to undertake the present study:
• more and more biocomposites with natural fibres (like ramie) are used in different areas. Now it is time to answer how they behave under natural conditions.
• according to the literature, ramie fibres are resistant to bacteria, mildew, and insect attack [10], but on the second hand organisms recognize the cellulose and hemicelluloses in the cell wall and can hydrolyze them into digestible units using specific enzyme systems [5].

Thus it was very important to estimate the resistance of ramie fibres under natural conditions.

Experimental Procedures

Materials

Ramie fibre (TOSCO Co. No. 25, 44 warps per inch and 46 wefts per inch) used in this work was symmetrical, with good stability and reasonable porosity.

Chemical composition of the ramie was: cellulose (68.6-76.2 wt.%), lignin (0.6-0.7 wt.%), hemicellulose (13.1-16.7 wt. %), pectin (1.9 wt.%), wax (0.3 wt.%), and moisture content (8 wt.%).

The commercial aliphatic-aromatic copolyester Ecoflex® (E, supplied by BASF) used in this work was made from 1,4-butandiol with adipic and terephtalic acids.

Corn starch resin (S) was obtained from Randy CP-300, Miyoshi Oil and Fat Co.

Cellulose nanofibres (C) were supplied by Diacell Co.Ltd.

The two kinds of biocomposites with ramie natural fibres were environmentally degraded:
1) Biocomposite of the ramie fibre/aliphatic-aromatic polyester Ecoflex® (R/E, 43/57%wt.). Ecoflex® neat used for the composite preparation was fabricated by compression molding of Ecoflex® granules at 120ºC in Hydraulic Press for 15 minutes and cooled to room temperature. The ramie mat was sandwiched between two Ecoflex® sheets by compression molding again at 120ºC.
2) Biocomposite of the ramie fibre/cellulose nanofibre reinforced corn starch resin (R/C/S, 31/5/64 %wt.). Cellulose nanofibres used for the composite preparation were 1 μm long and 20-30 nm diameter and contained 100% cellulose. At first, an aqueous solution of cellulose nanofibres was thoroughly mixed for 5 hrs. with liquid corn starch via ball mill mixing using ceramic balls. The biocomposite was developed by reinforcing with two layers of ramie fibre in corn starch resin. The cellulose nanofibre reinforced corn starch resin was inside the biocomposite. Hand-lay-up method followed by compression molding was used for the composite preparation.

Environments of Degradation

The environmental degradation of biocomposites with ramie was carried out in two kinds of environment: in compost with activated sludge and in sea water under natural weather, depending on conditions.

Degradation in Compost with Activated Sludge

The compost used in this work was formed with activated sludge taken from the municipal waste treatment plant in Gdynia. The compost pile was prepared under the natural conditions of a sewage farm. It consisted of the activated sludge, burnt lime, and straw. Burnt lime (0.45 kgCaO/1kg dry mass of compost) was added to ravage phatogenic bacterium and eggs parasites, to deacidificate activated sludge, and to convert activated sludge to compost. The straw was added to maintain the higher temperature of the compost pile and to loosen the structure of the compost pile. The compost pile prepared under natural conditions was not adequately aerated, so it was expected that a combination of conditions from aerobic at the upper part of the pile, microaerophilic in the middle part, and facultative anaerobic at the bottom of the pile could occur to microorganizm growth.

The characteristic parameters of the compost, such as temperature, pH, moisture content, and activity of dehydrogenases, were measured during the degradation process (Table 1).

Degradation in the Baltic Sea Water

The incubation of biocomposites took place in Baltic Sea water, in Gdynia Harbour.

Table 1. The characteristic parameters of compost (average data from two years).

Month/Year	Parameter			
	T (ºC)	pH	moisture content (%)	activity of dehydrogenases (mol mg^{-1} d.m.)
February	0	6.3	51	0.0177
March	3	6.0	58	0.0331
May	14	6.1	55	0.0297
July	20	5.8	60	0.0387
August	25	5.6	59	0.0471
November	8	5.9	60	0.0318
December	1	6.1	55	0.0192

Table 2. The characteristic parameters of Baltic Sea water (average data from two years).

Month/Year	Parameter			
	T (ºC)	Salinity [ppt]	pH	O$_2$ content [cm^3/dm^3]
February	3	7.2	8.0	9.8
March	3	6.3	8.2	10.6
May	11	6.5	9.0	9.0
June	13	6.1	9.0	8.2
August	18	6.8	8.7	6.8
September	19	6.7	8.4	7.0
November	10	6.8	8.1	8.4

The samples were located in a special perforated basket at a depth of 2 meters in the sea.

The characteristic parameters of the Baltic Sea water, such as temperature, pH, salinity, and oxygen content were monitored by the Institute of Meteorology and Water Management Maritime Branch in Gdynia, Poland (Table 2).

Methods

Characterization of Compost

- The moisture content of compost was determined by drying a sample at 105ºC until constant weight was obtained.
- The activity of the dehydrogenases was measured by a spectrophotometric method using triphenyltetrazolium chloride (TTC), which is a method for the estimation of biochemical activity of microorganisms in an active sludge using the oxidation process. The method is based on the dehydrogenation of glucose added to the compost with a subsequent transfer of the hydrogen to the

colourless biologically active compound of TTC, which undergoes a reduction to TTCH$_2$ (red compound) [11]. Glucose (1.5 g) and TTC (0.2 g) were added to distilled water (100 ml) and 1 ml of that solution was added to 5 ml of a compost solution (50 g compost homogenized with 100 ml of distilled water). The final mixture was incubated at 37ºC for 15 min. The dehydrogenation reaction was stopped by adding ethanol and the intensity of colour in the filtered liquid was measured using a Specol colorimeter at 490 nm.
- The pH of the compost was determined with a Teleko N 5172 pH-meter. The determinations were done on 50 g samples in 100 ml distilled water after homogenization for 30 min and incubation for 1 h at room temperature.

Investigation of Polymeric Materials

After incubation the samples were taken out from both environments and washed with distilled water and dried at room temperature to a constant weight.

The weight, surface morphology and mechanical properties of biocomposites were tested before and after degradation in natural environments.
- The weight of polymer samples was estimated using a Gibertini E 42s electronic balance. The percentage weight changes [%] were calculated from the weight data.
- The surface of the of polymer samples was observed in a macro (naked eyes) and micro scale (microscopic observation). Microscopic observations were carried out in two ways. For optical reflected microscopy the samples were covered with a thin layer of gold, while for electron microscopy a two-step replica procedure [12, 13] was accomplished.
- Maximum tensile strength (MPa) was measured at room temperature using a Tensile Testing Machine Type Fu 1000e made by VEB Thuringer Industriewerk Rauenstein, according to PN-EN ISO 527-1, 2, 3: 1998 Standard.

Results and Discussion

Characterization of Environments

Taking under consideration the fact that the biotic and abiotic parameters of compost (temperature, pH, moisture content, and activity of dehydrogenases) and sea water (temperature, pH, salinity, and oxygen content) have a significant influence on the development of living microorganisms in natural environments, the parameters were monitored during incubation time and their influence on the rate of degradation of biocomposites was discussed.

The characteristic parameters of compost are presented in Table 1.

The characteristic parameters of compost shown in Table 1 indicate that temperature had been fluctuating a lot during the experiment and depended on weather conditions.

Only the average temperature of compost during summer months was on the level preferred for enzymatic degradation [14].

It is known that a wide population of micro- and macroorganisms can exist in compost and the most abundant are bacteria, actinomycetes, and fungi [15]. But the fluctuating temperature (from 0 to 25ºC in compost) and slightly acid pH of compost (~6) probably cause psychrotrophic acidofilic microorganisms (fungi) to play the main role in the degradation of polymers in that composting environment. It is known that fungi usually grow on a solid surface.

Due to natural weather conditions the moisture content in the compost had been fluctuating. It could be expected this influenced the development of living microorganisms and the process of environmental degradation of the biocomposites. With decreasing moisture content a lower absolute value of activity of dehydrogenases, depending on the degree of microorganism growth, was observed.

It is also known that sea water is a very specific environment where abiotic parameters have a significant influence on the development of living organisms. The characteristic parameters of the Baltic Sea water are presented in Table 2.

The characteristic parameters of the Baltic Sea water indicate that the temperature had been fluctuating a lot during the experiment (3-19ºC) and depended on the weather conditions. The temperature of the Baltic Sea water during the incubation time was lower than that preferred for enzymatic degradation (20-60ºC) [14]. The low temperature and the alkaline pH (8-9) of the Baltic Sea caused the psychrotrophic bacteria to play the main role in the degradation in this environment.

During the winter months we could observe the very low temperature and the highest oxygen content. It could be expected that these conditions had an influence on the development of aerobic epilithic bacteria. The metabolism of these microorganisms probably caused a decrease of oxygen content in the summer months and changed the concentration of carbon dioxide in sea water.

Considering the characteristic parameters of compost and sea water presented in Tables 1-2, and the different microbial communities (fungi in compost and bacteria in sea water), we could expect a different rate of degradation of biocomposites in those two environments.

Evaluation of the Changes in Polymeric Materials during Environmental Degradation

The Macroscopic Changes of Surfaces

Fig. 1 represents the surface view at a macro scale of all investigated polymers before and after degradation in sea water (Fig. 1a) and in compost with activated sludge (Fig. 1b).

At the beginning of incubation (4 months) a very slow microbiological attack was noticed in both investigated biocomposites (R/E and R/S/C). Some of them partly changed colour or lost glossiness. The most susceptible for environmental degradation was Ecoflex® (E). Distinctly visible changes of surface and fragmentation were observed after 3 months incubation in compost.

A little bit different behaviour was observed for corn starch resin (S). That resin showed very low vulnerability to the composting process (visually the samples were unchanged after degradation).
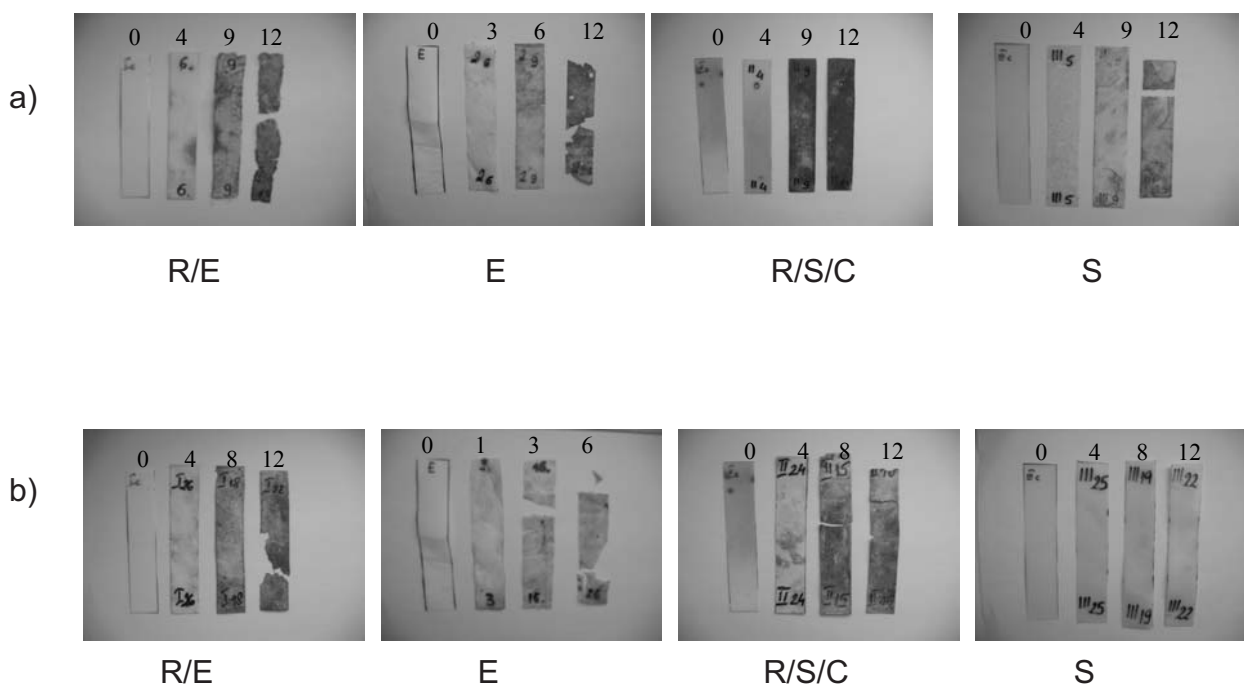

Fig. 1. Macrographs of polymer samples: R/E, E, R/S/C and S before (0) and after (1-12 months) incubation in sea water (a) and in compost (b).

But after incubation of the corn starch resin in sea water the visible macroscopic changes were observed. The black areas on the surface and fragmentation at the end of incubation suggested that corn starch resin was degraded in sea water.

After the next months of incubation in both environments (4-9 months) the matrix of both biocomposites R/E and R/S/C was partially removed and the ramie fibres were clearly visible on the top of the surface.

The distinct black areas on the surfaces of almost all biocomposite samples were observed at the end (12 months) of degradation in compost and sea water (Fig. 1).

The Microscopic Changes of Surfaces

Only the most important results of microscopic observations underlying differences of various samples incubated in compost and sea water are shown.

Fig. 2 presents the surface view of investigated samples before incubation (reference surface view) observed under an optical microscope.

The reference surface of biocomposite R/E was partly rough under optical microscope and was similar to reference surface of E, which was fully rough (Fig. 2). That observation could explain the fact that the quickest changes on the surface are shown in Fig. 1 of those samples during incubation. The least rough surface was observed for sample S, and that sample was almost unchanged in macroscale, especially in compost.

Fig. 3 shows the different behaviour of biocomposite R/S/C incubated in sea water and compost.

After incubation in compost there were clearly visible erosions on the surface (Fig. 3), while after incubation in sea water the surface still resembled the reference sample (Fig. 2b).
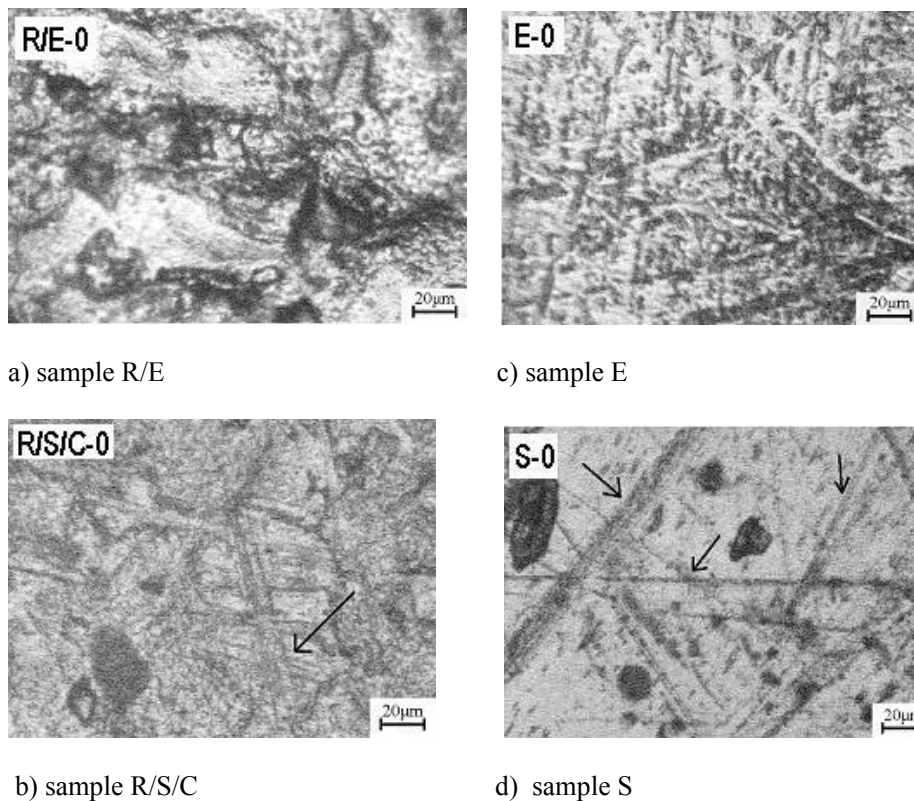

a) sample R/E

c) sample E

b) sample R/S/C

d) sample S

Fig. 2. The reference surface views of the polymer samples before degradation observed under optical microscope.

a) R/S/C incubated 8 months in compost

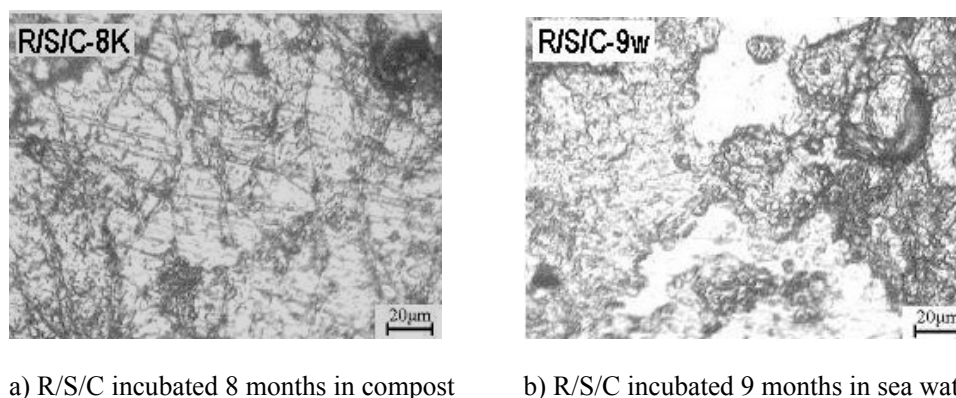b) R/S/C incubated 9 months in sea water

Fig. 3. The surface views of the biocomposite R/S/C after degradation observed under optical microscope.

Fig. 4 presents the surfaces of polymer samples observed under electron microscope (Fig. 4a, b) and optical microscope (Figs. 4c, d) after incubation in natural environments. The strong attachment of microorganisms to the sample surfaces is clearly visible.

Fig. 5 shows the surface of the corn starch resin sample (S) under an electron microscope.

From Fig. 5 it was clear that the degradation had just started after 12 months of incubation in compost. There was no attachment of microorganisms visible on the surface, but the traces of processing lines were much less visible than in the reference surface of the sample (compare the image to Fig. 2d). Moreover, there were present very fine holes (100-500 nm in size) representative of the erosion process; even under macroscopic observations samples were unchanged (Fig. 1b).
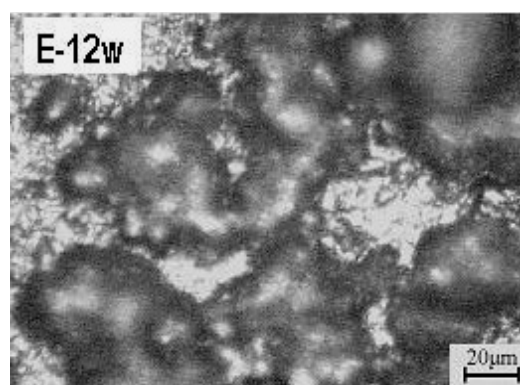
The Changes of Weight

The results of weight changes of investigated biocomposites and, for comparison, of corn starch resin and commercial aliphatic-aromatic copolyester Ecoflex® are presented in Table 3.

The data shown in Table 3 indicate that pure aliphatic-aromatic copolyester Ecoflex® was disintegrated after 6 months in compost and after 15 months in sea water. Then the degradation further proceeds for 18 months.
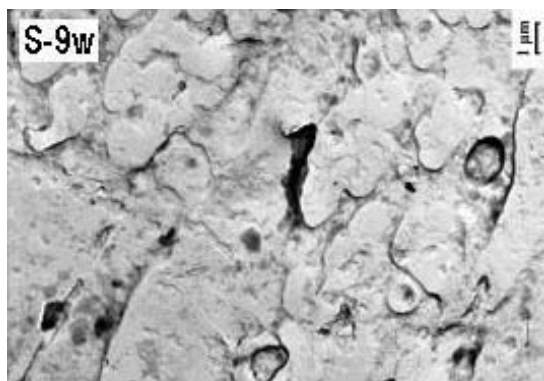
It is known that the aliphatic-aromatic copolyester Ecoflex® is classified as fully biodegradable (at least 60% of the total organic carbon of the polymeric material has been converted into carbon dioxide during a test interval of no longer than 180 days) [16]. According to our studies, degradation of pure Ecoflex® samples in compost and sea
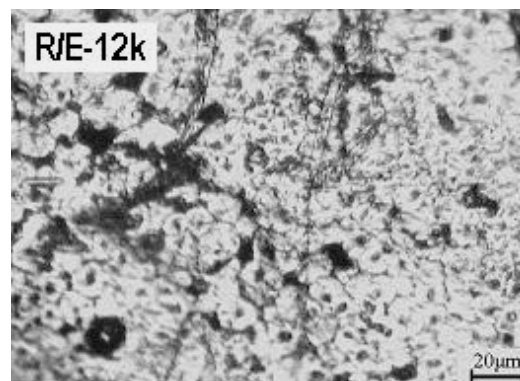

a) E incubated 3 months in sea water
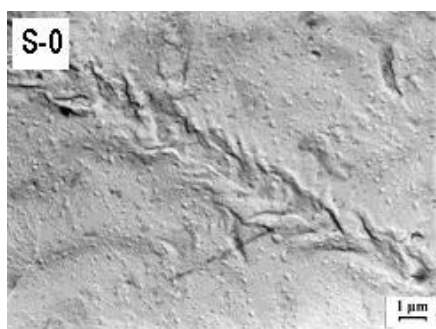

c) E incubated 12 months in sea water

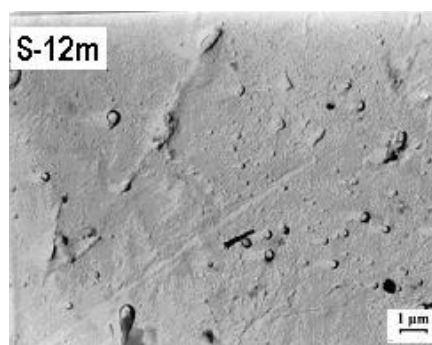b) S incubated 9 months in sea water

d) R/E incubated 12 months in compost

Fig. 4. The surface views of polymer samples after degradation observed under electron (a, b) and optical (c, d) microscope.

a) before degradation

b) after 12 months incubation in compost

Fig. 5. A comparison of surface views of corn starch resin (sample S) before and after degradation observed under electron microscope.

Table 3. Weight changes [%] of polymer samples after incubation in natural environments.

Polymer samples		Weight changes of polymer samples[%]/incubation time [months]									
		1	3	4	6	8	9	12	15	18	24
R/E	Sea water	+0.4	-	-4.6	-5.5	-	-16.2	-40.4	-92.4	disintegration	
	compost	+1.9	-1.9	-2.8	-	-33.9	-	-54.1	-87.5	disintegration	
E	Sea water	-1.5	-2.2	-	-29.0	-	-	-48.5	-62.1	disintegration	
	compost	-1.6	-10.3	-	-57.4	-	-	disintegration			
R/S/C	Sea water	+1.0	-1.1	-1.0	-	-	-12.9	-28.5	-45.4	-55.0	-76.4
	compost	+2.8	-2.9	-4.0	-	-49.0	-	-66.7	-78.1	disintegration	
S	Sea water	-0.5	-2.0	-2.5	-	-	-19.6	-15.8	-19.6	-25.9	-27.8
	compost	+0.5	-0.9	-1.1	-	-1.3	-	-1.4	-1.6	-1.9	-2.3

water under natural weather conditions was longer than in laboratory standard tests [16].

The obtained results indicated the very slow environmental degradation process of corn starch resin in compost and sea water. Generally, the degradation rate of corn starch resin was higher in sea water, because starch is a very hydrophilic and a very poor water barrier material. Our earlier results also revealed that the modified starch had a good enough resistance in sea water under natural conditions [17].

The observed water uptake appearing after 1 month incubation of the biocomposites samples (R/E, R/C/S) in both environments was an important factor in enzymatic degradation as water led to swelling of the polymer and thus facilitated an enzymatic attack [18]. It is known that the absorbed water molecules reduce the intermolecular hydrogen bonding between cellulose molecules in fibre, which may cause the reduction of interfacial adhesion between fibre and matrix and also weight loss [19].

After the next months of incubation in compost and sea water the percentage weight loss values of biocomposites R/E and R/C/S had been increased with degradation time.

But the process of degradation of biocomposites R/E and R/C/S in compost was more intensive than in sea water. The observed differences between the degradation process in compost and sea water were caused by dissimilarity of environmental conditions. The monitored characteristic parameters of the environments indicated that the conditions of compost were more preferable for growth of fungi, while in sea water for bacteria. According to the literature, natural fibres are degraded biologically because organisms recognize the cellulose and hemicelluloses in the cell wall and can hydrolyze them into digestible units using specific enzyme systems [5]. Among all microorganisms involved in the degradation of cellulose fibres, the most attractive are fungi [20].

The rate of degradation of biocomposites (R/E, R/C/S) sped up after 6 months of incubation. That was the period of summer months and the environmental conditions were more preferred for enzymatic degradation than during the winter months. After 15 months of incubation the weight losses of biocomposite samples (R/E, R/C/S) were approximately 45-92%, depending on the environment. Generally, R/E biocomposite had been degraded faster than R/C/S bio-

Table 4. The maximum tensile strength [MPa] of polymer samples before and after incubation in natural environments.

Polymer samples		The maximum tensile strength [MPa]/ incubation time [months]								
		0	1	3	4	9	12	15	18	24
R/E	Sea water	86.9	73.9	-	30.0	10.0	torn up into the pieces			
	compost		43.8	36.1	36.6	torn up into the pieces				
E	Sea water	10.0	9.4	6.7	torn up into the pieces					
	compost		8.9	torn up into the pieces						
R/S/C	Sea water	89.5	70.6	76,1	63,8	24.7	9.8	torn up into the pieces		
	compost		74.9	58.6	42.1	torn up into the pieces				
S	Sea water	18.4	21.4	21.1	19.2	6.2	6.0	torn up into the pieces		
	compost		21.6	21.4	20.6	-	16.3	15.8	15.2	14.6

composite in both natural environments and after 18 months of incubation the disintegration of R/E biocomposite in sea water and compost could be observed. Meanwhile, the weight losses of R/C/S biocomposite were approximately 55% after 18 months and 76% after 24 months of incubation in sea water. But for R/C/S samples incubated in compost the disintegration was already noticed after 18 months in spite of corn starch resin resistant to degradation in this environment.

Changes in Tensile Strength

Tensile strength of investigated polymers before and after degradation in both environments is presented in Table 4.

The data shown in Table 4 show that the maximum tensile strength of pure aliphatic-aromatic copolyester-Ecoflex® decreased during the incubation time in sea water and compost. At the end of the experiment (already after 3-4 months of incubation) microorganisms caused the breakdown of polymer samples, resulting in fragmentation.

The tensile strength of pure corn starch resin decreased much faster during incubation in sea water than in compost. The samples of pure corn starch resin S were torn into pieces after 15 months of degradation in sea water, whereas in compost the maximum tensile strength had been decreased only from 18.4 MPa to 15.8 MPa.

The addition of ramie fibre to Ecoflex® caused a significant increase of tensile strength of the samples. The maximum tensile strength of Ecoflex® was 10 MPa, while of biocomposites R/E – was 87 MPa. The tensile strength of R/S/C biocomposite, in comparison to pure corn starch resin, was much higher. The maximum tensile strength of biocomposite R/S/C was 89 MPa and of pure corn starch resin S – was 18 MPa.

During the incubation of both biocomposites in compost, the tensile strength of the samples had also been decreasing continuously with the degradation time and at the end of experiment microorganisms caused the breaking of polymer chains in the matrix, resulting in the fragmentation of biocomposites.

After 9 months of degradation of R/E biocomposite in compost, the samples of composite were already torn into pieces, whereas in sea water the mechanical properties could be still estimated.

During the first months of the incubation in sea water the tensile strength of biocomposite R/S/C was much higher than in compost. According to the literature the increase of the strength of wet ramie fibre was observed in comparison to dry fibre [21]. The water uptake into the structure of biocomposites, which were incubated in sea water, caused the reduction of interfacial adhesion between the fibre and the matrix and a decrease of tensile strength of composites [19]. The growing up of microorganisms (distinctly visible after 9-12 months incubation, Fig. 1a) caused the deterioration and the braking of chains in polymer matrix, resulting in the decrease of tensile strength.

Conclusions

The results of our study revealed that biocomposites with ramie fibre were degraded in compost with activated sludge and sea water under natural conditions. The rate of the degradation process depended on the nature of the environment. Generally, biocomposites with natural ramie fibre were degraded in compost faster than in sea water, because the conditions of compost were more favourable for growth of fungi.

The composition of biocomposites influenced the rate of degradation process in both natural environments. The ramie/Ecoflex® biocomposite was degraded more distinctly in both environments than ramie/cellulose nanofibre reinforced corn starch resin biocomposite.

The macro and microscopic observations and weight changes confirmed the decrease of the tensile strength of the composites after incubation in compost and in sea water.

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