How to Cite: Normurodov, N.F., Berdinazarov, Q.N., Abdurazakov, M., & Ashurov, N.R. (2022) Mechanical and Thermal Properties of Biodegradable Composites Based on graft copolymer LLDPE-g-MA/Gelatin. *Bulletin of the University of Karaganda Chemistry*. https://doi.org/10.31489/2022Ch4/4-22-11

Article Received: 16 June 2022 ǀ Revised: 27 October 2022 ǀ Accepted: 09 November 2022 ǀ Published online: 23 November 2022

UDC 678.742.23+665.931.7 https://doi.org/10.31489/2022Ch4/4-22-11

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Mechanical and Thermal Properties of Biodegradable Composites Based on graft copolymer LLDPE-g-MA/Gelatin

The uncontrolled development of morphology at the stage of formation of biodegradable compositions based on synthetic and natural polymers limits the possibility of achieving satisfactory physical and mechanical and operational characteristics. In the present work, to achieve finely dispersed mixture morphology, an approach was proposed for reactive mixing of functionalized polyethylene with gelatin to form a linear low density polyethylene-grafted-maleic anhydride and gelatin (LLDPE-g-MA/GEL) graft copolymer. Using the selective extraction of the mixture components, we determined amount of graft copolymer LLDPE-g-MA/GEL, free gelatin, mechanical and thermal properties, as well as biodegradability data. It was found that as the amount of maleic groups in the polyethylene macromolecule increased, the amount of graft copolymer increased, and an increase in the content of gelatin in the blend led to a noticeable increase in the elastic modulus, tensile strength, and a decrease in elongation at break. Due to the degradation of gelatin, the thermal stability of the composite (initial temperature) decreased with increasing gelatin content. The maximum rate of destruction of the graft copolymer in the temperature range of 400–500 ºC increased markedly with an increase in the content of gelatin. It was found that the rate of biodegradability would increase with an increase in the content of gelatin in the blend; the maximum level of degradation was observed during the first 10 days and was more than 50 %. It was found that the maximum degree of grafting LLDPE-g-MA and gelatin to each other depended on the amount of maleic anhydride in the graft copolymer. The maximum degree of grafting was observed to be higher with increasing amount of maleic anhydride in the composites.

Keywords: Biodegradation, gelatin, glycerin, polyethylene, maleic anhydride, polymer composite.

Introduction

The packaging industry needs biodegradable materials. Traditional synthetic polymers, in particular polyolefins, despite a good combination of production technology, the possibility of varying physical and mechanical characteristics and prices, pose significant problems for ecology and the environment.

An analysis of the scientific literature indicates that the solution to this problem is seen in the use of biopolymers, or their combination with synthetic polymers. These approaches, of course, have their limitations, such as optimization of physical and mechanical characteristics (with accelerated natural biodegradability) and limited degradability, depending on the biopolymer/synthetic polymer ratio, respectively. The list of promising biopolymers includes proteins (soy and milk proteins, gelatin, etc.) and polysaccharides (chitosan, carboxymethylcellulose, starch, etc.). Gelatin and starch occupy leading positions in the creation of packaging materials based on them due to their low cost [1–4]. The special structure of these biopolymers, that is, the presence of a dense network of hydrogen bonds between macromolecules, requires plasticization to transfer to a thermoplastic state. Many studies carried out in this direction [5–11] have shown the fundamental possibility of forming biodegradable high-quality films with satisfactory physical and mechanical properties. Generally accepted, gelatin is defined as the product obtained by the partial hydrolysis of collagen present in the skin, connective tissues and bones of animals [12, 13]. The structure of gelatin includes a certain sequence of hydrophilic amino acids (glycine, proline, hydroxyproline), when an aqueous solution of gelatin is cooled, a gel is formed with the restoration of the collagen triple helix. The most effective plasticizers for gelatin are glycerol, sorbitol, polyethylene glycols, the variation of the content of which makes it possible to obtain films with a tensile strength in the range of one hundred MPa and more, an elastic modulus of hundreds of MPa with very low relative deformation. At the same time, the high sensitivity of gelatin to water, that is low barrier properties to water vapor (swelling, dissolution) hinder the scale of production and use [14–16].

From this point of view, interest in the creation of blends of gelatin with synthetic polymers continues unabated [17]. At the same time, it was noted that the usual mixing of gelatin with polyolefin in various ratios gave very low biodegradability rates (up to 10 %). Optimization of the blend morphology by transferring gelatin to a thermoplastic state slightly increased this indicator, and record biodegradability rates were achieved when creating conditions for the formation of graft copolymers of polyethylene with gelatin (more than 80 %) [18].

In this work, the formation of such a structure was carried out by means of an additional stage; blends of gelatin with polyethylene were obtained on pre-irradiated polyethylene (^{60}Co source, dose 3.4 kGy/h). Unfortunately, these studies lack the structural data of the polyethylene matrix and their inherent physico-mechanical and thermal characteristics. Previously, we showed [19] the realization of such a possibility on maleated polyethylene.

This approach was implemented via the formation of a graft copolymer throughout the entire volume of the sample, which ultimately resulted in the desired finely dispersed mixture morphology. In mixtures, depending on the content of gelatin and the level of maleated polyethylene, along with the graft copolymer, the presence of free gelatin was detected. The latter, as a rule, aggregated with the gelatin fragment of the graft copolymer. It was of interest to reveal the relationship of structural elements with the physical, mechanical and temperature characteristics of the compositions. The results obtained indicated that mixtures with a finely dispersed morphology increased elastic-strength properties with satisfactory deformability.

Materials and research methods

Materials

Linear low-density polyethylene (LLDPE, Shurtan Gas Chemical Complex of the Republic of Uzbekistan) grade F-0320, d = 0.920 g/cm^3, MFI = 2.5 g/10 min (at a load of 2.16 kg,) and edible gelatin (GEL) grade P-200 (State Standard 11293–2019, JSC “MOGELIT”, Belarus) were used without any purification. Maleic anhydride (MA, $C_4H_2O_3$, analytical grade, colorless rhombic crystals, M_r = 98.06 g/mol) was distilled at T_{bp} = 84.0 °C/14 mm Hg, T_m = 60 °C, ρ^{60} = 1.3140 g/cm^3.

Preparation of thermoplastic gelatin

To dissolve gelatin granules and make them thermoplastic, glycerin was added to distilled water and stirred until the same mixture was obtained. Gelatin was added to the resulting mixture and mixed again, then heated in an oven at 80 ºC for two hours.

Functionalization

Functionalization of LLDPE with maleic anhydride (LLDPE-g-MA) was carried out on a Brabender plastograph (Plasticorder Brabender OHGDUISBURG Germany), with a cam velocity of 98 rpm and at a temperature of 180 ± 5 °C [19]. The concentration of grafted maleic anhydride was varied in the range of 0.5–5.0 wt%.

Polymer blends based on LLDPE-g-MA and gelatin were obtained on a Brabender plastograph, for 30 min, at 50 rpm and 180±5 °C by adding plasticized gelatin to the LLDPE-g-MA melt.

The maximum grafting percentage of LLDPE-g-MA and gelatin was 114 %; the grafting percentage was calculated by extracting unreacted PE and gelatin with water and xylene.

Grafting rate was calculated using the following formula:

$$\text{Grafting}~\% = (W\text{g} - W\text{o}) / W\text{o} \times 100, \tag{1}$$

where Wo and Wg are the masses of the initial composite and after the complete removal of polyethylene and gelatin that did not participate in the reaction, respectively [18].

Bio-degradability test (Soil Degradation)

The polymer compositions were tested for biodegradation in the form of films in a specially prepared soil (with a moisture content of at least 25 %). During the testing period, the soil moisture was maintained at 30 %, and the samples were immersed to a depth of 15 cm. Soil samples were removed every 10 days for biodegradation testing, washed in ethanol, wiped and equilibrated in a desiccator for at least one day before weight was recorded. Sample weight loss (W) in % was determined depending on the number of days as follows:

$$W = (W_0 - W_\text{i}) / W\text{o} \times 100. \tag{2}$$

Weight loss in % after every 10 days (W_{di}):

$$W_{\text{di}} = (W_\text{i} - W_{\text{i-1})} / W_\text{i} \times 100, \tag{3}$$

where W_0 is the initial mass, g; W_i is the final weight every 10 days; $W_{\text{i-1}}$ is the initial weight every 10 days [18, 20].

Physico-mechanical tests

The tensile diagrams of the samples were determined on a SHIMADZU AG-X plus universal testing machine (JAPAN) in the uniaxial tensile mode with a set strain rate in accordance with the requirements established by ASTM D 638-99 “Standard Test Method for the Strength Properties of Plastics”.

For analysis, at least five samples of the studied material were used, made in the form of double-sided blades with a thickness of 2 mm. For measuring tensile module (E), 1 mm/min crosshead velocity was chosen until 0.3 % deformation, after that crosshead velocity was increased immediately to 20 mm/min for further exploring yield stress (σ) and deformation (ε).

DSC and TGA measurements

Thermal analyses DSC and TGA of polymer blends was carried out in a dynamic mode on a LINSEIS THERMAL ANALYSIS PT1600 [thermogravimetric analysis (TGA)] instrument (in air atmosphere) in the range from room temperature to 1000 °C, guided by the requirements established by the ASTM E 1131 standard. The heating rate was 10 °C/min; the sample weight for analysis was from 2 to 100 mg.

Results and Discussion

The uniform distribution of the gelatin phase in the polyethylene matrix and their accessibility to microorganisms determine the biodegradation efficiency of such compositions [20]. The desired uniform distribution is achievable by grafting gelatin onto the polyethylene macromolecules. The possibility of implementing this reaction was shown by us [21] using a maleated linear copolymer of ethylene with butene-1 (MA content was 0.5–5 wt %) and in [18] on pre-irradiated polyethylene [22]. It should be noted that before obtaining blends of polyethylene with gelatin, the latter must be transferred to a thermoplastic state by plasticizing the optimal ratio of glycerol with water [23–25].

To determine the blends compositions at the selected polyethylene/gelatin ratios, the extraction experiments were carried out using xylene and water to selectively dissolve the components, respectively. Due to the gelatin degradation under conditions of LLDPE dissolution in xylene at a temperature of 120 ºC, we analyzed only the extraction data in an aqueous medium. Taking into account that unreacted gelatin was extracted under these conditions, we calculated the amount of polyethylene-gelatin graft copolymer (Table 1).

Table 1

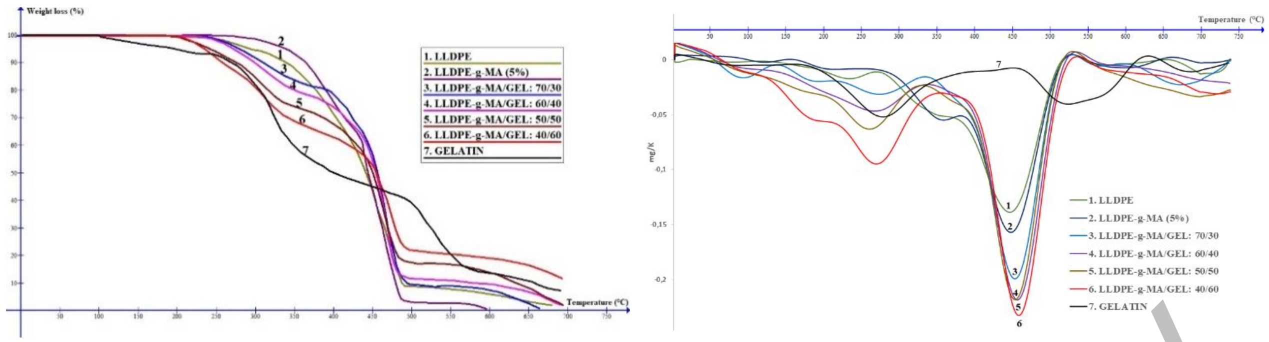
Amount of grafted LLDPE-g-MA/gelatin and unreacted gelatin

Samples	70/30	60/40	50/50	40/60
Extraction Products	%	%	%	%
LLDPE-g-MA(0.5 %)/GEL				
Pure gelatin %	7.6	10.8	20.5	39.5
LLDPE-g-MA-g-GEL %	39.6	59.5	79.6	90.8
LLDPE-g-MA(5 %)/GEL				
Pure gelatin %	1.8	8.5	13.7	24
LLDPE-g-MA-g-GEL %	42	61	86.3	114

As can be seen from the table, the amount of free gelatin varied between 1.8-24 % by weight when its content in the composition of the initial blend was 30–60 % wt., respectively, in relation to the content of PE. Based on these data, the grafting degree of gelatin to polyethylene was determined. Naturally, as the gelatin increased, the degree of grafting increased in the range of 42–114 % by weight. Blends formed in the presence of thermoplastic gelatin with the formation of a graft copolymer were homogeneous formations with a uniform distribution of components [19]. Increasing the gelatin concentration above equimolar with respect to the concentration of maleic groups in polyethylene resulted in the formation of an additional gelatin phase, which agglomerated with the gelatin end of the graft copolymer in the form of a dispersed phase.

As expected, blends of gelatin with polyethylene containing 0.5 % wt. of maleic anhydride contained a markedly higher amount of unreacted gelatin, 7.6-39.5 % wt. On that case, the grafting degree for the same initial formulations was 39.6–90.8 % by weight.

TGA and DSC measurements



a *b*

Figure 1. Thermogravimetric curves (*a*) and the first derivative of the TGA curves (*b*) of the film sample

In mixtures, depending on the content of gelatin and the level of polyethylene maleization, along with the graft copolymer, the presence of free gelatin was detected. The latter, as a rule, aggregated with the gelatin fragment of the graft copolymer.

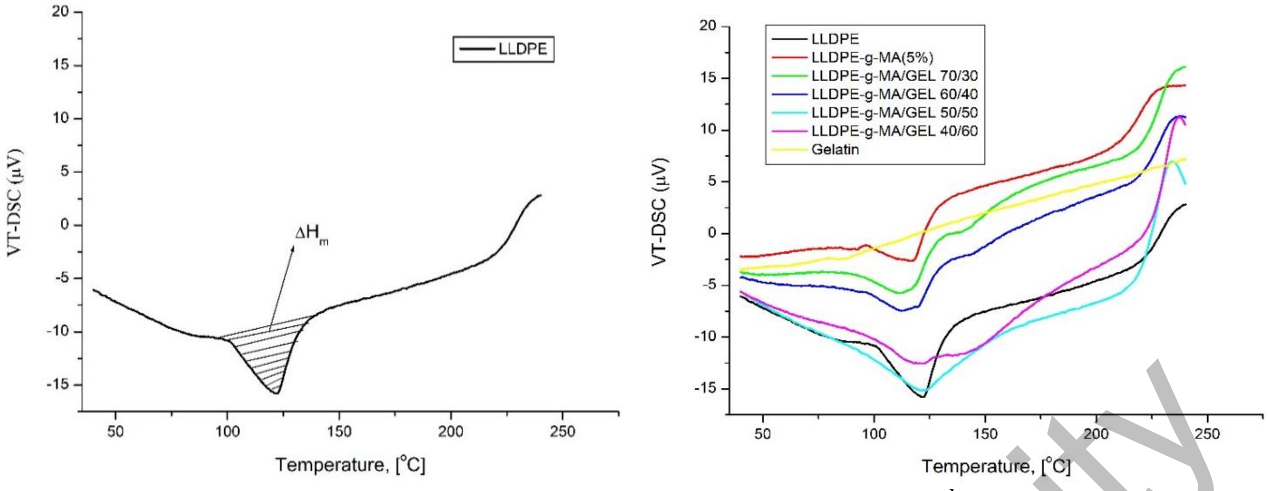
The presence of these reaction products was noticeably manifested in the thermogravimetric characteristics (Fig. 1). The initial degradation temperature (T_i) and the temperature of the maximum degradation rate (T_{max}) for various compositions of mixtures and initial components are given in Table 2.

Table 2

Thermal characteristics of LLDPE-g-MA/gelatin blends

No.	Sample name and component ratio (wt%)	The temperature corresponding to the loss of mass is ºC				Degree of crystallinity, %
		Initial degradation temperature T_i, ºC	Maximum thermal decomposition rates (T_{max}), ºC			
			first stage	second stage	third stage	
1	LLDPE	186	100–200 ºC m(mg)≤10 %	275–400 ºC 10≤m(mg)≤50 %	400–500 ºC m(mg)≥50 %	19.85
2	LLDPE-g-MA (5 %)	242				18.15
3	LLDPE-g-MA/GEL 70/30	233				14.85
4	LLDPE-g-MA/GEL 60/40	223				13.32
5	LLDPE-g-MA/GEL 50/50	190				12.02
6	LLDPE-g-MA/GEL 40/60	201				10.4
7	GELATIN	97				-

The thermal decomposition of the LLDPE-g-MA/gelatin mixture on TGA curves consisted of three sequential stages. The first stage in the temperature range of 100–200 ºC was due to the loss of absorbed and bound water molecules (weight loss less than 10 % weight). The second stage, observed in the region of 275–400 ºC (weight loss in the range of 10–50 wt%), corresponded to the breaking of peptide bonds in the gelatin macrochain [26]. Temperatures, depending on the expected rate of thermal decomposition of the gelatin phase, were found in the range of 250–280 ºC. A partial contribution to the consumption of the composition in this area included the beginning of LLDPE-g-MA dehydration, an increased rate of thermal decomposition (third stage), which felt at a temperature of 450 ºC. The position of this peak was practically independent of the content of gelatin in the composition; as the content of gelatin increased, it increased significantly at high temperatures (up to 7 ºC). Summarizing the above TGA data, it can be noted that the formation of a graft copolymer LLDPE-g-MA/gelatin led to the decrease in thermal stability in terms of T_i by 10–40 ºC, depending on the content of gelatin 30–60 wt%, respectively. While the maximum rate of thermal decomposition of the graft copolymer (third stage) increase in the biopolymer content increased significantly (from –0.14 to –0.23 mg/K) due to degradation of the grafted gelatin.



a *b*

Figure 2. A fragment of the DSC curve of PE (*a*) Fragments of the DSC curves of the studied compositions (*b*)

Figure 2 *a*) shows a fragment of the DSC curve for a PE sample in the melting region. The endothermic peak is highlighted here (shaded part). The melting enthalpy ΔH_m PE was determined, with the help of which it was possible to calculate the crystallinity degree of PE χ_c (%) as a percentage according to equation (4):

$$\chi_c\ (\%) = \Delta H_m / \Delta H_{m0} \times 100\ \%, \qquad (4)$$

where ΔH_{m0} is the value of the melting enthalpy of a hypothetical fully crystalline PE, which is equal to 293 J/g [27].

The values of χ_c (%) for the rest of the studied samples were calculated in a similar way. For this, fragments of DSC curves were used, shown in Figure 2 b). This figure shows that the gelatin sample does not have an endothermic peak corresponding to the melting process, i.e. it does not crystallize. The calculations results of the crystallinity degree of all samples are presented in Table 2.

Mechanical properties

Interfacial adhesion is the determining parameter of the deformation process for the systems under consideration with an elastic matrix and rigid inclusions (gelatinous phase). It should be noted that the studied LLDPE-g-MA/gelatin blends differ from classical two-phase blends [28–34] in the form of clearly separated continuous and dispersed phases. In our case, the dispersed phase consists of segregated domains of grafted gelatin macrochains with free unreacted gelatin.

The general picture of the deformation process for blends proposed in [35] includes three stages, elastic (stage I — up to 1 % deformation), then slight delamination of the matrix from the surface of a rigid inclusion until the appearance of dumbbell-shaped microvoids (stage II — reaching the yield point) and the last stage (III stage) can manifest itself in two forms, as the destruction of the sample due to the coalescence of microvoids, or the ordering of macromolecules in the direction of the applied stress (called orientational crystallization, recrystallization, cold — drawing). Exactly on that part, interfacial adhesion is determined, the strengthening of which is accompanied by an increase in the stress at the yield point and the level of deformation.

Let's consider the features of the curves σ-ε of the compositions studied (Fig. 3).

The introduction of maleic groups into the polyethylene macromolecule did not introduce significant changes in the shape of the σ-ε curves; they contained all three stages of the deformation process. At the same time, the strengthening of intermolecular interactions, due to the presence of maleic functional groups (0.5–5 wt%.), resulted in a noticeable increase in the yield stress (from 8.5 MPa to 13.5 MPa), tensile strength at break (from 11 MPa to 14.5 MPa), modulus of elasticity (from 130 MPa to 220 MPa).

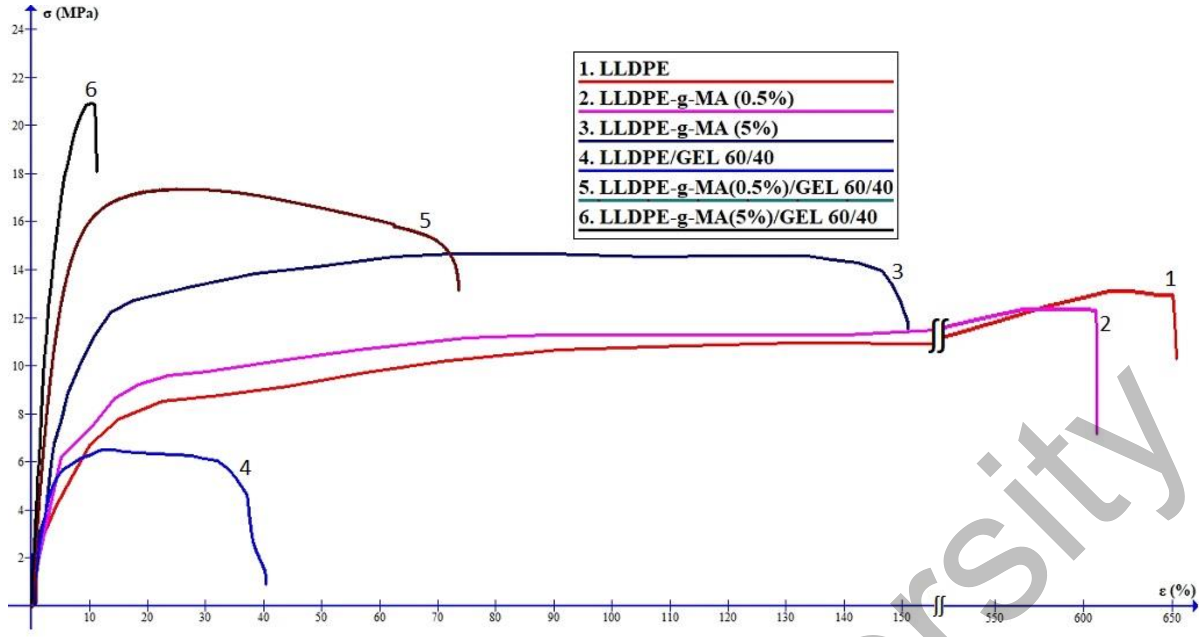


Figure 3. Mechanical properties of samples of different composition

There are curves of deformation of the composition of polyethylene with gelatin (ratio 60/40 % wt.) based on polyethylene with a content of 0 %, 0.5 %, 5 % wt. MA groups were undergone significant changes. On the σ-ε curves, the length of the orientational ordering stage noticeably decreased, that was, after reaching the yield stress, the colescence of microvoids around inhomogeneities led to a sequence of fibrillation and rapid destruction processes. This was most pronounced for conventional blends of polyethylene with gelatin and polyethylene with a content of 5 % by weight, maleic groups, the relative elongation at break of which was 48 % and 11.5 %, respectively. Accounting for 80 % deformability, maleinization of polyethylene at the level of 0.5 % wt. was also significantly inferior to the original polyethylene (600 %). Let's consider the mechanical properties of the PE/gelatin composition by varying the content of gelatin based on polyethylene with different content of maleic groups (Table 3).

Table 3

Physical and mechanical properties of composites

No.	Content of composites	Tensile strength, MPa	Elongation at break, %	Young modulus, MPa
		LLDPE/GEL		
1	LLDPE	11±0.22	655±0.5	129.9±3.6
2	70/30	7.03±0.31	125.6±11.3	237.79±10.12
3	60/40	6.52±0.28	48±10.4	253.19±31.96
4	50/50	3.8±0.26	26.8±6.6	179.8±31.12
5	40/60	4.4±0.21	15.6±5.3	293.62±44.78
		LLDPE-g-MA(0.5 %)/GEL		
6	LLDPE-g-MA(0.5 %)	10.92±0.5	610.1±18.03	119.25±10.34
7	70/30	16.20±0.33	122.71±10.58	286.46±3.64
8	60/40	17.48±0.15	78.65±5.24	338.31±10.07
9	50/50	17.68±0.91	54.88±2.96	278.41±12.33
10	40/60	19.84±1.02	26.21±2.15	335.99±34.66
		LLDPE-g-MA(5 %)/GEL		
11	LLDPE-g-MA(5 %)	14.65±0.28	146.8±7,32	218.1±5.8
12	70/30	17.41±1.9	12.40±1.84	481±10.98
13	60/40	20.63±1.74	11.43±1.41	550±27.77
14	50/50	24.02±3.24	10.21±1.29	676±8.07
15	40/60	27.56±2.42	6.73±1.06	736.53±49.84

In conventional compositions (the absence of graft copolymers), due to the lack of interfacial adhesion, and hence the weak stress transfer at the PE/gelatin interface, as the gelatin content increased, a noticeable drop in σ_y, σ_{break}, and ε_{break} by more than 40 % was observed.

On the contrary, the presence of graft copolymers segregated with free gelatin enhanced σ_y and σ_{break} by 20-80 % (0.5 % wt. MA) and more than 100 % in polyethylene compositions with gelatin at content of 5 % wt. MA on a polyethylene macromolecule.

At the same time, the relative elongation at break for the composition based on graft copolymers, due to the presence of inhomogeneities at the phase boundary, as well as for conventional PE/gelatin compositions, was significantly reduced. As can be seen from Table 3, for all three groups of the composition, as the gelatin content increased, despite the insignificant degree of crystallinity of the polyethylene phase (Table 2), it increased in comparison with the original polyethylene and maleated polyethylenes in the range of 130 %, 180 %, and 240 %, respectively. This effect was due to the fact that the presence of hydrogen bonds in the structure of double helixes of crystalline sections of gelatin provided a high elastic modulus, more than 1000 MPa [36, 37] compared to PE. Therefore, it can be observed that as the content of grafted gelatin increased, the elastic modulus increased for formulations with gelatin content of 60 % by weight from 300 MPa to more than 700 MPa.

Based on the totality of all mechanical characteristics, compositions of polyethylene with gelatin with a content of 0.5 wt % MA in the polyethylene chain are of the greatest interest for applications.

Biodegradation of polymer composites

The biodegradability of LLDPE-g-MA/gelatin compositions is shown in Figure 3 as a function of residence time in soil. As is known, attacking agents of the biodegradation process are microorganisms such as actinomycetes, fungi and bacteria [18, 38]. Almost all compositions reached the level of maximum degradation on the 40th-50th day of testing. The level of degradation as the content of gelatin in the composition increased (30 %, 40 %, 50 %, and 60 % by weight) was on the order of 20 %, 35 %, 48 %, 58 %, respectively. The highest degradation rate was observed for a mixture containing 60 wt%. gelatin, less than 10 days degradation rate was 55 %.

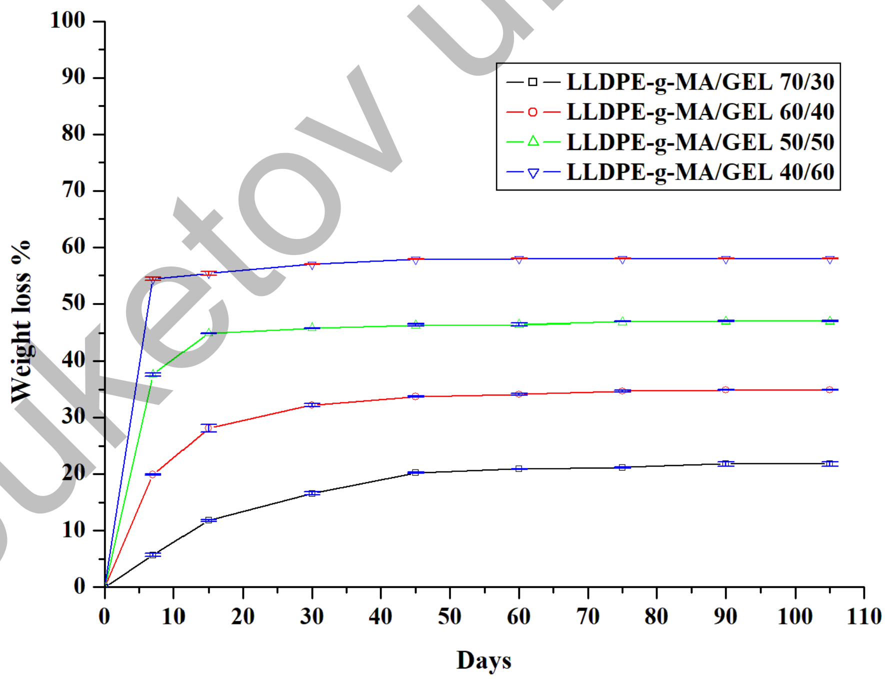


Figure 3. Weight loss of composites in the soil

Thus, the carried out studies showed the fundamental possibility of creating highly efficient biodegradable systems with a wide range of possible elastic and elastic properties. The achieved performance was due to the optimal morphology of the mixture and satisfactory interfacial adhesion of the components because of the formation of grafted LLDPE-g-MA/gelatin copolymers and segregated domains of grafted fragments and free gelatin.

Conclusions

The grafting of thermoplastic gelatin to maleated polyethylene promoted the formation of homogeneous compositions with a finer dispersion of the gelatin phase in the polyethylene matrix. Depending on the content of maleic groups in polyethylene and the content of gelatin in the blend, the grafting degree varied between 40–150 %. Three stages of thermal decomposition of the LLDPE-g-MA/gelatin composition associated with the presence of water molecules, degradation of the gelatin and polyethylene phases were identified. The presence of gelatin significantly shifted the onset temperature of decomposition towards low temperatures (from 242 °C to 201 °C), while the temperature range of degradation of the polyethylene matrix did not undergo significant changes. The maximum rate of degradation of the graft copolymer in the temperature range of 400–500 °C with an increase in the content of gelatin noticeably increased from –0.14 to –0.23 mg/K.

The presence of gelatin in the composition in the form of grafted chains, compared to conventional blends of polyethylene with gelatin, led to an increase in yield stress, stress at breaking and elastic modulus by 80 % and 240 %, respectively. This trend was the most pronounced for the composition of PE with the content of MA groups of 5 wt%.

In terms of applications, polyethylene compositions with 0.5 wt. MA-groups exhibited good strength properties and modulus of elasticity in combination with satisfactory deformability. The resulting compositions exhibited high biodegradability of up to 58 %, the level of maximum degradation was observed on the 40-50th day. The weight loss rate was observed for the composition with a gelatin content of 60 wt%.

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Полиэтилен және желатин негізіндегі биологиялық ыдырайтын композиттердің механикалық және термиялық қасиеттері

Синтетикалық және табиғи полимерлер негізіндегі биологиялық ыдырайтын композициялардың қалыптасу сатысындағы морфологияның бақылаусыз дамуы қанағаттанарлық физикалық, механикалық және эксплуатациялық сипаттамаларға қол жеткізу мүмкіндігін шектейді. Жұмыста қоспаның ұсақдисперсті морфологиясына қол жеткізу үшін функционалды полиэтиленді желатинмен реакциялық араластырып, егілген малеин ангидриді мен желатинмен (LLDPE-g-MA/GEL) егілген төмен тығыздықтағы сызықтық полиэтилен сополимерін қалыптастыру тәсілі ұсынылған. Қоспаның құрамдас бөліктерін селективті алу әдісімен егілген LLDPE-mma/GEL сополимерінің мөлшері, бос желатин, механикалық және термиялық қасиеттері, сондай-ақ биологиялық ыдырау деректері анықталды. Полиэтиленнің макромолекуласындағы малеиндік топтардың мөлшері артқан сайын егілген сополимер мөлшері көбейетіні, ал қоспадағы желатин мөлшерінің артуы серпімділік модулінің айтарлықтай жоғарылауына, кернеудің бұзылуына және бұзылған кезде салыстырмалы ұзаруға әкелетіні анықталды. Желатиннің ыдырауына байланысты композиттің термиялық тұрақтылығы (бастапқы температура) желатин мөлшерінің жоғарылауымен төмендейді. 400–500 °C температура диапазонында трансплантат сополимерінің максималды бұзылу жылдамдығы желатин мөлшерінің жоғарылауымен айтарлықтай артады. Қоспадағы желатиннің жоғарылауымен биоыдыраудың жылдамдығы өседі, деградацияның максималды деңгейі алғашқы 10 күнде байқалады және 50%- дан асады. ПОЭМА мен желатинді бір-біріне егудің максималды дәрежесі егілген сополимердегі малеин ангидридінің мөлшеріне байланысты екендігі айқындалды. Композиттердегі малеин ангидридінің мөлшерінің жоғарылауымен егудің максималды дәрежесі жоғары болғаны байқалды.

Кілт сөздер: биоыдырау, желатин, глицерин, полиэтилен, малеин ангидриді, полимерлі композит.

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Механические и термические свойства биоразлагаемых композитов на основе полиэтилена и желатина

Неконтролируемое развитие морфологии на стадии формирования биоразлагаемых композиций на основе синтетических и природных полимеров ограничивает возможность достижения удовлетворительных физико-механических и эксплуатационных характеристик. В настоящей работе для достижения мелкодисперсной морфологии смеси был предложен подход к реакционному смешению функционализированного полиэтилена с желатином с образованием привитого сополимера линейного полиэтилена низкой плотности с привитым малеиновым ангидридом и желатином (LLDPE-g-MA/GEL). Методом селективного извлечения компонентов смеси определяли количество привитого сополимера LLDPE-g-MA/GEL, свободного желатина, механические и термические свойства, а также данные по биоразлагаемости. Выявлено, что, по мере увеличения малеиновых групп в макромолекуле полиэтилена, количество привитого сополимера возрастет, увеличение содержания желатина в смеси приводит к заметному увеличению модуля упругости, разрушающего напряжения и падению относительного удлинения при разрушении. Ввиду деградации желатина термостабильность композита (начальная температура) при увеличении содержания желатина снижается. Максимальная скорость деструкции привитого сополимера в области температур 400–500 °C, по мере увеличения содержания желатина, заметно увеличивается. Обнаружено, что скорость биодеградируемости возрастет при увеличении содержания желатина в смеси, максимальный уровень деградации наблюдается в течение первых 10 дней и составляет более 50 %. Установлено, что максимальная степень прививки ПЭМА и желатина друг к другу зависит от количества малеинового ангидрида в привитом сополимере. Замечено, что максимальная степень прививки была выше с увеличением количества малеинового ангидрида в композитах.

Ключевые слова: биодеградация, желатин, глицерин, полиэтилен, малеиновый ангидрид, полимерный композит.

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