

Among the issues related to the disposal of polymers' waste is the design of cheap biodegradable polymeric materials, which are destroyed as a result of natural microbiological and chemical processes. Since the synthesis of biodegradable polymers is characterized by high material and energy costs, the filled biodegradable polymeric materials are more promising.

This paper substantiates the feasibility of using marble microparticles as a filler for the biodegradable polymeric material based on high-pressure polyethylene whose decomposition rate increases under the conditions of ultraviolet radiation.

Samples of the biodegradable polymeric material with the different content of a filler, the microparticles of marble (from 0 to 5.1 % by weight), were made; their physical-mechanical properties were investigated. It has been experimentally proven that UV radiation destroys polyethylene macromolecules into separate fragments with the formation of $>C=O$ bonds, making it possible to decompose appropriate waste under the influence of sunlight.

The feasibility of using the composition "high-pressure polyethylene – microparticles of marble" with a marble content of 1.78 % by weight has been confirmed for the manufacture of the sleeve and flat polymeric films for packaging and agricultural purposes, in particular, packing stretch film.

Recommendations have been given on using the proposed biodegradable polymeric material. In particular, it is proposed that the production of articles from the "high-pressure polyethylene – microparticles of marble" composition should utilize a pre-obtained granulate of the required formulation. When making articles from the composition obtained directly in the extruder that molds the products, it is possible to apply adhesive additives to improve the interaction between a polymeric matrix and the filler's particles

Keywords: extrusion, polymeric film, high-pressure polyethylene, marble microparticles, ultraviolet radiation, decomposition

UDC 539.216: [678.742.2+552.46]-046.64

DOI: 10.15587/1729-4061.2020.216835

DESIGN OF A COMPOSITION BASED ON POLYETHYLENE AND MARBLE MICROPARTICLES THAT DECOMPOSES UNDER THE EFFECT OF ULTRAVIOLET RADIATION

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Received date 19.08.2020

Accepted date 12.11.2020

Published date 09.12.2020

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1. Introduction

The history of mankind is conventionally distinguished by several long enough periods of development: the Stone Age, the Bronze Age, and the Iron Age [1]. At the same time, since the first half of the twentieth century, the Iron Age was followed by the age of polymers, the production volume of which at the beginning of the third millennium is com-

parable to the production volume of metals, and exceeded 300 million tons [2].

Significant volumes of production and consumption of polymers and plastics suggest devising effective measures for the disposal of polymeric products that lost their consumer properties [3].

One of the ways to dispose of polymeric waste, in particular containers and packaging, and, above all, polymeric

films, is the development of cheap biodegradable polymeric materials. These materials are designed for the manufacture of articles that have short service life (usually up to one or two years) and are self-destroyed as a result of natural microbiological and chemical processes [4].

The synthesis of biodegradable polymers is characterized by high material and energy intensity, so the filled biodegradable polymeric materials are more promising.

Therefore, the search for new biodegradable polymeric materials with high operational properties and adjustable service time is a relevant task.

2. Literature review and problem statement

Many types of biodegradable polymeric materials have been developed, that is, those materials that are destroyed intensively enough (compared to conventional polymers) as a result of natural processes [5].

Factors that accelerate the destruction of biodegradable polymeric materials (BPM) are, first of all, microorganisms, the oxygen of the air, moisture, the radiation of visible and ultraviolet spectra. These factors act mostly not so much directly on a polymer but the BPM components, in particular its filler. The BPM fillers that are widely used include additives that contain natural polymers, specifically cellulose, starch, chitosan, etc.

Study [6] terms the BMP with natural polymers “green composites” due to their natural origin. However, articles from such BPM have limited use because they absorb moisture actively enough and are intensively destroyed under the influence of sunlight. In addition, the corn or potato starch used in BPM reduces the volumes of food products whose shortage becomes one of the urgent problems of today. At the same time, cheaper inorganic substances, especially of natural origin, may become more promising fillers.

In addition, study [6] tackles the issue of handling the waste of polymeric materials based on polyethylene and polyamide filled with various dispersed fillers (sand, natural fiber, hemp fiber, metal powder, etc.). At the same time, attention is paid primarily to the task of recycling, rather than the disposal of these materials.

Work [7] analyzes the impact of various factors of the natural environment on the destruction of multilayered biodegradable films, primarily with the use of polymers such as polylactide, polybutylene adipate terephthalate, and polycaprolactone. These polymers are effective enough in the BPM composition but their processing causes some difficulties because they are prone to hydrolytic destruction. In addition, multilayered biodegradable films are much more expensive than single-layer ones.

Work [8] investigated the mechanical and degrading properties of BPM using the tubers of *Asphodelus*, starch, polyvinyl alcohol, titanium dioxide, and powdered dolomite. Dolomite, which contains calcium carbonate and magnesium, is used to enhance the mechanical properties of BPM, rather than controlling its biodegradation.

Study [9] reported a new class of polyester composites with a filler made of particulate substances in the form of dust-like marble waste with a filler content of up to 40 % by weight. It is shown that the strength of composites at stretching and bending slightly decreases compared to a pure polymer, while compressive strength, impact viscosity,

and hardness by Shore increase. The designed material is proposed to be used in friction units for the manufacture of wear-resistant elements. However, the impact of natural factors on the composite was not investigated.

Paper [10] examined the physical, thermomechanical, and tribological properties of epoxy composites reinforced with fiberglass containing the waste of marble dust with a filler content from 0 to 40 % by weight. It is shown that most physical and mechanical properties of composites improve at a temperature up to 60 °C. However, the effect of natural factors on the resistance of the composite was also not investigated.

Work [11] designed new polymeric composites with a filler in the form of waste of marble dust – solid waste. This waste forms when cutting and polishing marble pieces at construction sites, as well as at marble processing enterprises. The authors proved the feasibility of using the designed materials in the friction units of machine parts. No impact of natural factors on the resistance of the composite was examined.

Study [12] examined the coefficient of thermal conductivity of a nonwoven epoxy composite with particles of marble dust containing filler in the amount from 0 to 24 % by weight. It is shown that the coefficient of thermal conductivity of the composites increases with an increase in filler content. Similar to works [9–11], the impact of natural factors on the stability of the composite was not tackled.

Paper [13] investigated the tribological properties of epoxy composites filled with fiberglass and powdered marble with different filler content.

Work [14] examined the physical-mechanical properties of polypropylene filled with powdered marble containing filler in the amount from 0 to 80 % by weight. It is shown that the designed composite is characterized by high strength and heat resistance, as well as low water absorption.

Paper [15] shows that calcium carbonate nanoparticles improve strength, stiffness, size stability, abrasive resistance, and heat resistance of polymeric materials based on polyethylene, polypropylene, polyvinyl chloride, and polyamide. However, no information about the impact of this filler on the decomposition of relevant polymeric materials is given.

Typically, the following three main areas of research in the field of BPM are considered: the creation of polyesters of hydroxycarboxylic acids, the design of BPM based on renewable natural polymers, as well as rendering biodegradability to materials based on industrial synthetic polymers [16].

Given the widespread use of industrial synthetic polymers in various human activities, the third direction is the most appropriate. At the same time, it is important to determine the qualitative and quantitative composition of BMP which would demonstrate the required level of biodegradability and high physical and mechanical properties. In addition, important is the absence of toxicity, availability, and cheapness of BMP components, as well as the possibility of making articles from them using conventional technologies for processing polymeric materials [17, 18].

Powdered calcium carbonate may become such a component (including powdered marble), which is now used as a filler for polymeric composite materials to improve their physical and mechanical properties. At the same time, there are no studies on the possibility of using calcium carbonate in biodegradable polymeric materials as a substance that could accelerate the destruction of these materials under natural conditions.

3. The aim and objectives of the study

The aim of this study is to analyze the feasibility of using marble microparticles as a filler to form a biodegradable polymeric material based on one of the most common polymers – polyethylene.

To accomplish the aim, the following tasks have been set:

- to substantiate the possibility of using marble microparticles as a filler to form a biodegradable polymeric material based on polyethylene;
- to fabricate samples of the biodegradable polymeric material with different filler content and examine its physical and mechanical properties depending on the filler content.

4. Substantiation of the possibility of using marble microparticles as a filler to form a biodegradable polymeric material

One of the factors contributing to the destruction of macromolecules of synthetic polymers under natural conditions is solar radiation, in particular, the ultraviolet component of its spectrum [4].

The mechanical strength of many synthetic polymers, including polyolefins, depends on their structure, in particular the presence of macromolecules' bonds, the degree of crystallinity, the degree of macromolecule tension, as well as their mutual orientation. At the same time, a certain number of C–C and C–H bonds in macromolecules under mechanical stress are in a strained state, and, therefore, the energy of ultraviolet (UV) radiation is sufficient to break them.

UV rays cause the photochemical destruction of polymers, which is a radical chain process. Under the influence of UV rays, a polymer, in addition to breaking chemical bonds, experiences the crosslinking of macromolecules, the formation of double bonds and free radicals. However, the photochemical destruction of polyolefins mostly occurs in the surface layers of polymeric waste.

When polymeric waste is exposed to UV rays, some of them pass through the waste's material, a part is absorbed, and a part is reflected. The absorbed energy is spent to directly heat the waste's material (including the heating of the polymer), as well as to break the polymer's macromolecules.

At the same time, depending on the qualitative and quantitative composition of the polymeric material, the specified components of the UV ray energy may vary. To accelerate the destruction of polymeric waste, it is necessary to increase the proportion of absorbed energy. This can be achieved by reducing other components. It is for this purpose that we propose to use powder-like marble as a filler for a biodegradable polymeric material.

Due to the high reflective capacity of marble, as well as the significant specific surface of its microparticles, those UV rays are reflected that partially passed through the polymeric matrix, directly into the polymer. Thus, UV rays can be divided into "primary" and "secondary". The former fall on a sample that is subjected to destruction directly from the source of radiation; the latter are reflected from the marble particles inside the polymeric matrix, preventing them from passing unhindered passage through a sample of the polymeric material.

It is known that polyethylene is decomposed by some bacteria, as well as by insects, in particular, the caterpillar of *Galleria mellonella* [19]. Since this process of polyethylene destruction is slow enough, it can be accelerated by

pre-decomposing polyethylene waste into small fragments, in particular with the help of UV radiation.

Thus, combined with the strengthening effect, the microparticles of marble can become an effective catalyst for accelerating the destruction of polymeric waste under the influence of ultraviolet radiation in open dumps of solid household and industrial waste.

5. Procedure for fabricating samples of biodegradable polymeric material and for studying its physical and mechanical properties

The main consumers of articles from biodegradable polymeric materials are the packaging industry and agriculture. In this case, the basic type of such products is the sleeve and flat polymeric films made by extrusion [20–26]. At the same time, the use of various mixing and dispersing elements in extruders significantly improves the distribution of components of the processed composition in the polymeric matrix [27].

We made the polymeric composition at a worm-disk extruder (a worm diameter, 45 mm; a disk diameter, 135 mm); next, it was used to mold a sleeve film, 500 mm wide, in a folded form. Samples to be examined were cut from the resulting sleeve film. The melt temperature of the composition at the outlet from the extrusion head was maintained at the level of 180 °C. Geometric dimensions of the samples are as follows: length, 25 mm; width, 10 mm; thickness, 0.1 mm.

The main components of the resulting composition are as follows: high-pressure polyethylene (HPPE; polyethylene of low density), grade 15813-020, manufactured by Public Joint Stock Company "Kazanorgsynthesis" (Kazan, Russian Federation), and micro calcite, grade M40 (marble microparticles), manufactured by Niğtaş Mikronize Kalsit Ticaret Ve Sanayi Ltd. Şti. (Niğde, Turkey), with a CaCO₃ content of 99.4 % by weight, and an average size of marble particles of 40 µm.

To more accurately determine the content of marble microparticles in the samples studied, we burned the polymeric matrix out of them at the muffle furnace SNOL-1,6,2,5,1/9-14 (manufactured by Limited Liability Company "Bortek", Boryspil, Ukraine). The actual content of marble microparticles in the samples was less than expected, by 20–35 % by weight, which can be explained by that the fine-dispersed filler stuck to the wall of the loading hopper and other elements of the extruder.

The examined samples were exposed to UV radiation using the arc discharge high-pressure tubular mercury lamp DTR-1000 with a capacity of 1,000 W. During the experiments, the temperature of the samples was maintained within 50–60 °C. We determined the films' mechanical properties at the universal test machine TIRAtest-2151 (manufactured by TIRA GmbH, Germany).

The infrared spectrophotometer Specord 75 IR (manufactured by Analytik Jena AG, Germany) was used to analyze the impact of UV radiation on the examined samples.

Fig. 1 shows the dependence of the relative elongation of a sample with HPPE whose marble content was 0.42 % on the applied effort at the different duration of UV radiation.

The maximum sample elongation is demonstrated in the absence of exposure to UV radiation but the corresponding maximum load, in this case, is not maximal. A lower maximum load was demonstrated only by a sample that was irradiated over four hours; it began to deform only under a load of 7 N.

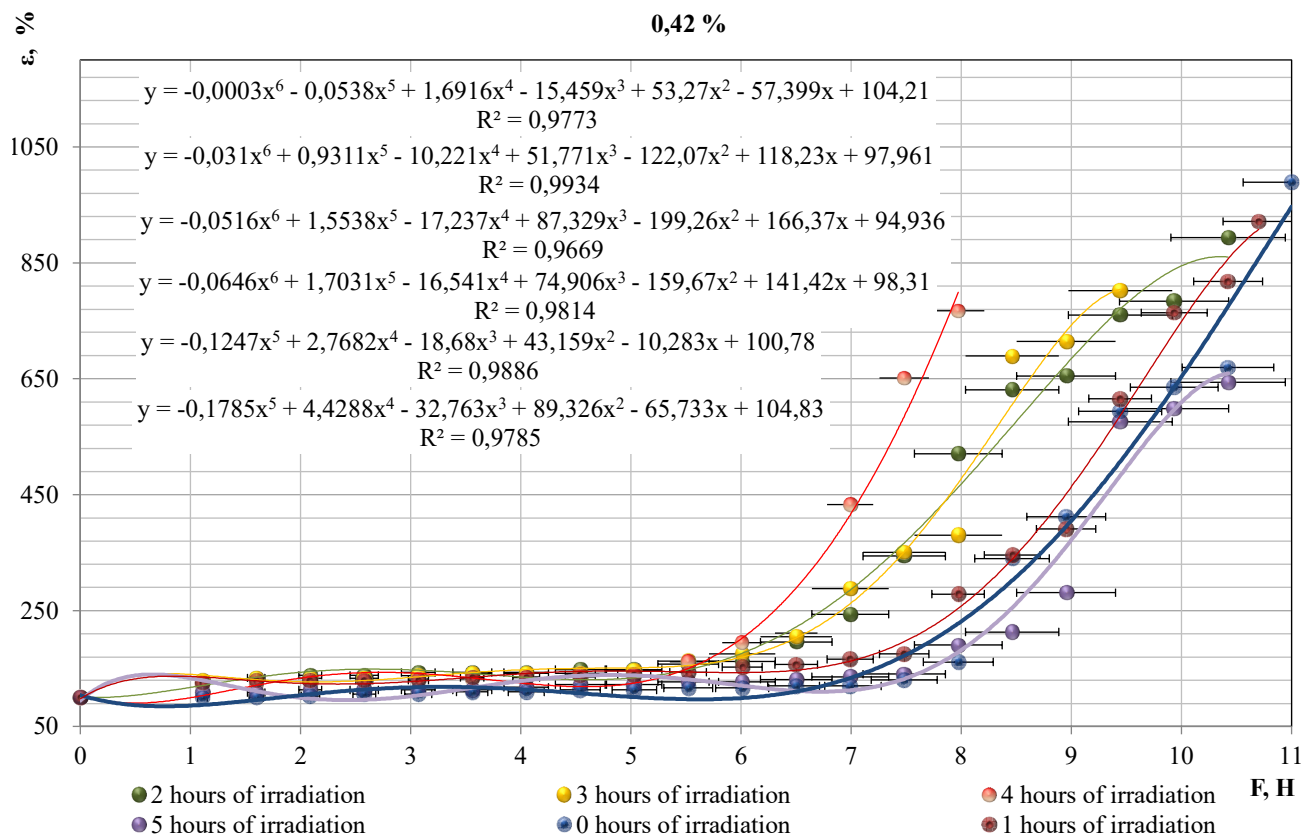


Fig. 1. Dependence of the relative elongation of samples with HPPE with a marble content of 0.42 % on the stretching effort

It is interesting to compare the dependences of the samples irradiated over one and two hours because they are characterized by the increased load that leads to sample deformation. While the non-irradiated sample demonstrated a noticeable deformation already under a load of 7.5 N, then for a sample irradiated within one hour, this value increased to 9.9 N. At the same time, for a sample irradiated over two hours – to 8.5 N. This indicates an increase in strength properties but the elastic properties of the samples decreased over time.

The best strength properties were observed for a sample irradiated over five hours. For this sample, the relative elongation changed significantly under a load of 10.4 N. In that case, the rupture occurred immediately after reaching a maximum elongation.

Fig. 2 shows the dependence of the relative elongation of a sample with HPPE, whose marble content is 1.14 %, on the applied effort at the different duration of UV radiation.

The nature of the deformation of the samples, not irradiated and irradiated over one hour, is similar, the difference is only the samples' elongation during destruction. Thus, the elongation of the sample, which was not irradiated, amounted to 700 %, and that of the sample, irradiated over one hour, to 670 %. At the same time, a load of destruction in both cases was 9.9 N, which indicates that the sample, irradiated over one hour, lost its elasticity.

The nature of the deformation of the samples, irradiated over two and three hours, is also similar. However, the deformation of the sample, irradiated over two hours, began under a load of 8.9 N, while the deformation of the sample, irradiated over three hours, – 9.9 N.

The sample, which was irradiated over four hours, demonstrates greater strength and less elasticity because it has undergone a significant deformation at a load of 9.9 N, which is the greatest value under these conditions.

The sample that was irradiated over five hours worsened both its strength and elastic properties. Its destruction occurred under a load that is less by 1.5 N compared to the non-irradiated sample.

Fig. 3 shows the dependence of the relative elongation of a sample with HPPE with a marble content of 1.78 % on the applied effort at the different duration of UV radiation.

The strength of all samples, except for the sample irradiated over five hours, is similar in nature, with a maximum load of 10.4 N. However, the onset of samples' deformation varies. Thus, for a sample that was not irradiated, and a sample that was irradiated over one hour, the corresponding load was 7.5 N (but in the second case, the dependence is more intense). For samples that were irradiated over two, three, and four hours, the respective load was 7; 6.5; and 9.9 N.

Fig. 4 shows the dependence of the relative elongation of a sample with HPPE with a marble content of 5.1 % on the applied effort at the different duration of UV radiation.

Fig. 4 demonstrates that the deformation of the samples varies significantly.

The irradiated sample began to deform under a load of 7 N; the samples irradiated over one, two, three, four, and five hours, – 7.5; 7.5; 9; 5.5; and 4.5 N, respectively.

Fig. 5 shows the dependence of the relative elongation of samples ϵ (%) on irradiation duration (h); Fig. 6 – the dependence of conditional tensile strength σ (N/cm²) on irradiation duration (h).

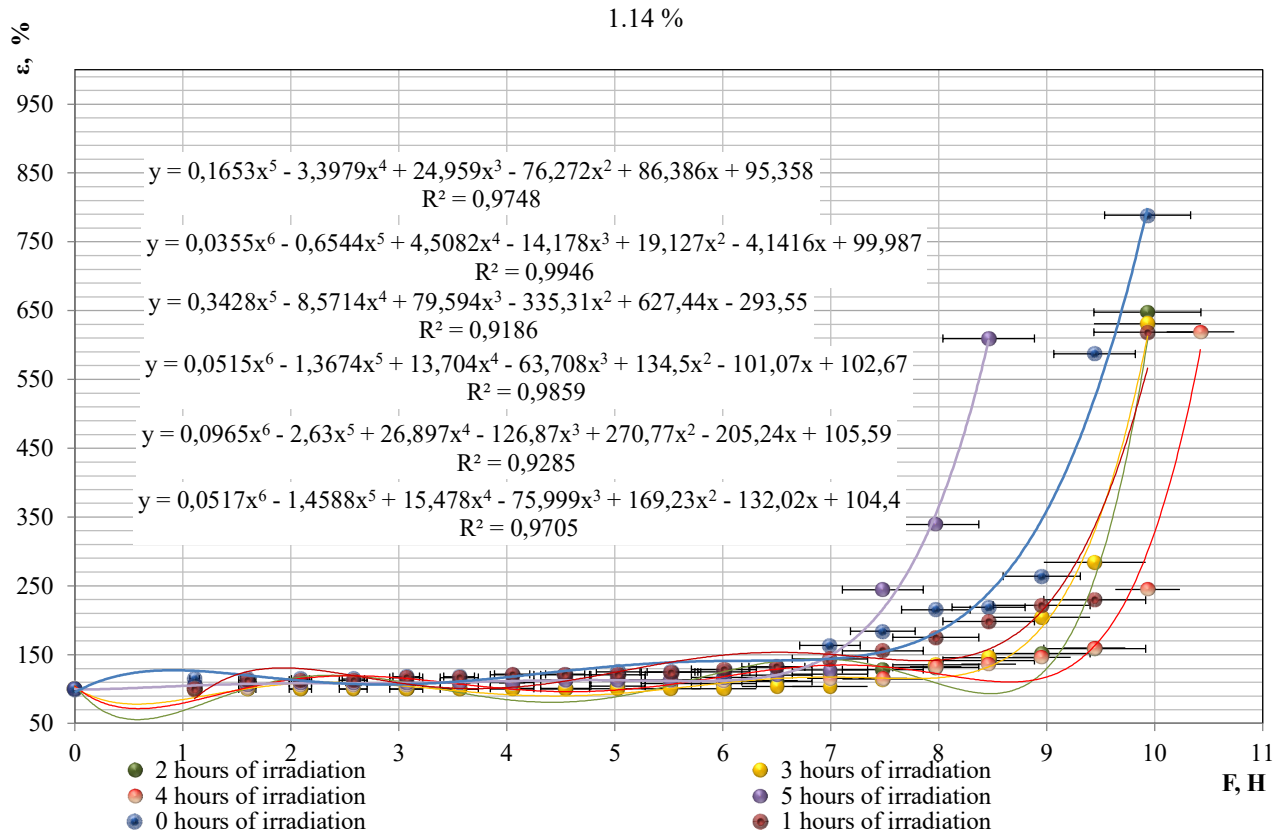


Fig. 2. Dependence of the relative elongation of samples with HPPE with a marble content of 1.14 % on the stretching effort

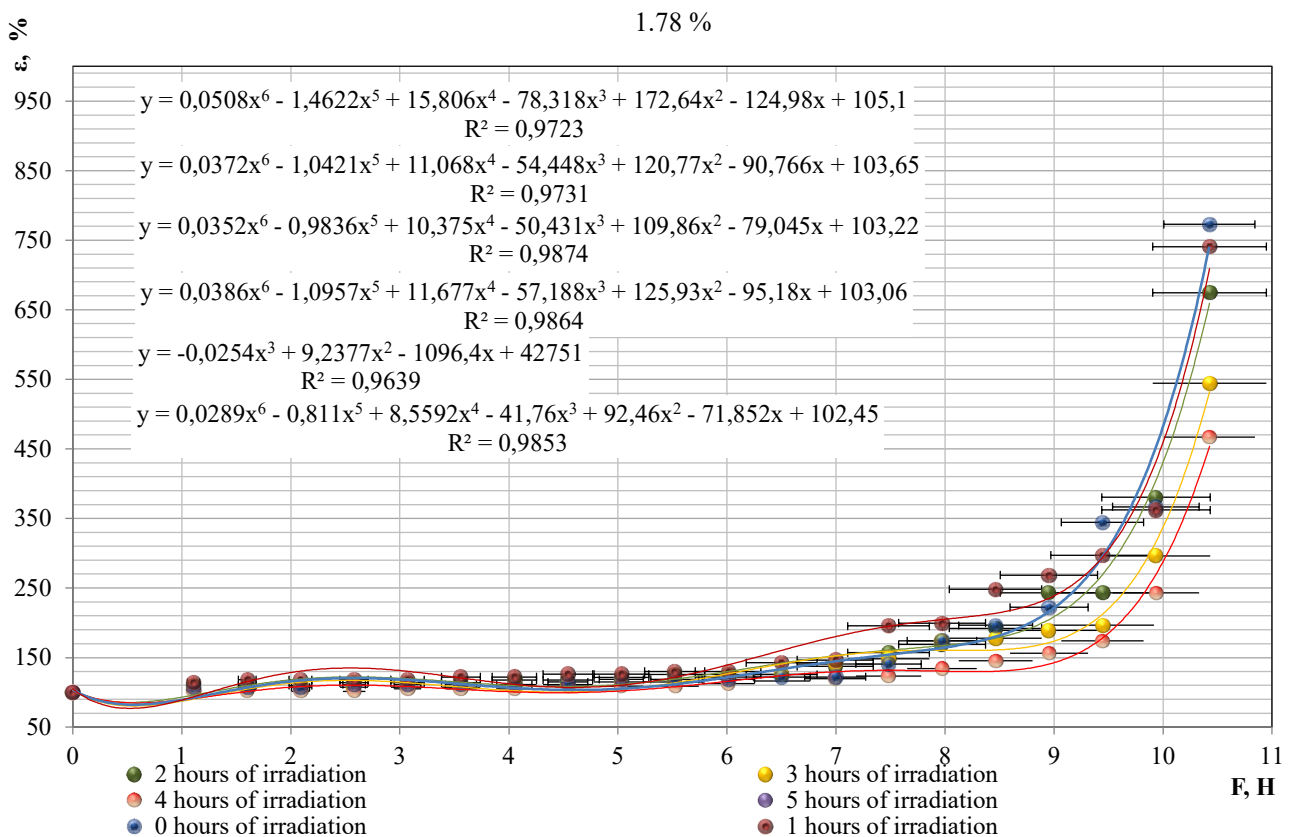


Fig. 3. Dependence of the relative elongation of samples with HPPE with a marble content of 1.78 % on the stretching effort

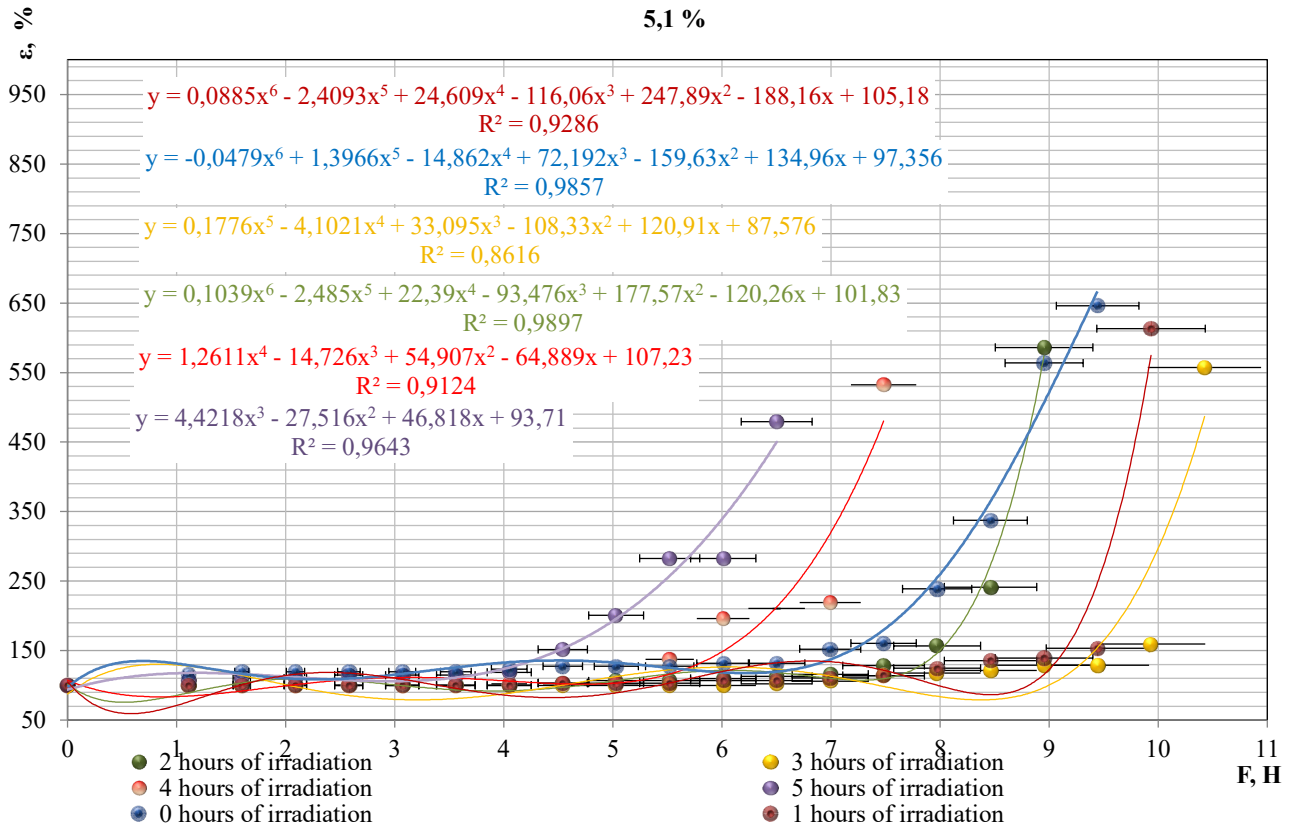


Fig. 4. Dependence of the relative elongation of samples with HPPE with a marble content of 5.1 % on the stretching effort

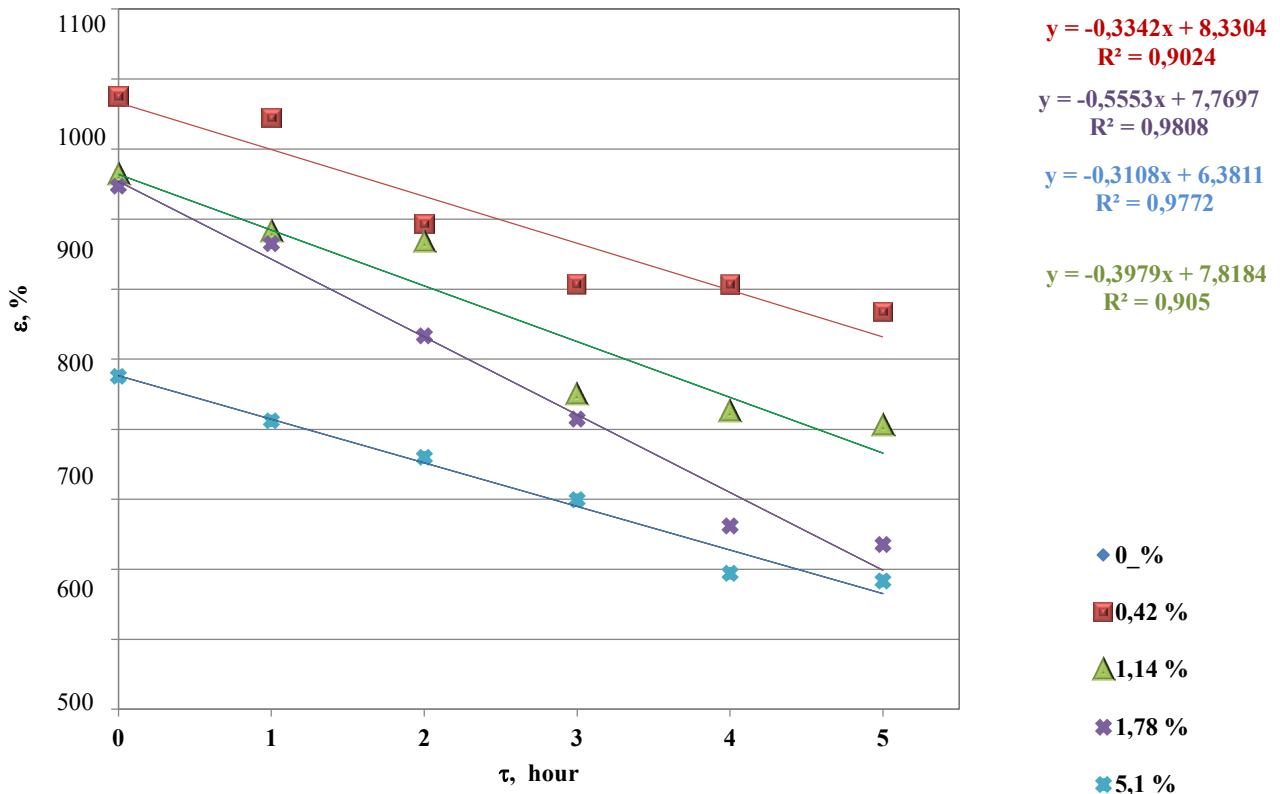


Fig. 6. Effect of UV radiation on the conditional tensile strength of samples with different content of marble microparticles

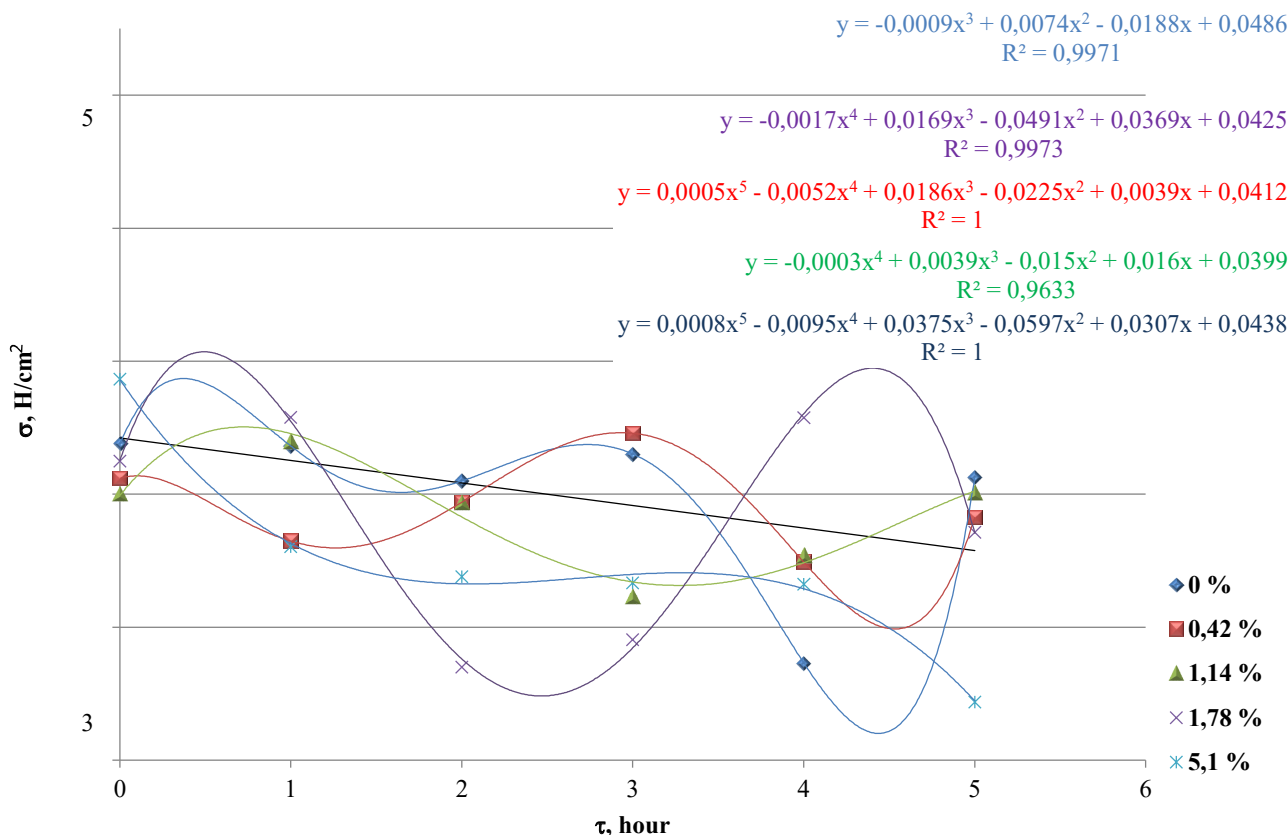


Fig. 6. Effect of UV radiation on the conditional tensile strength of samples with different content of marble microparticles

An analysis of our results reveals that the relative elongation of samples changes over the time UV radiation according to a linear law. At the same time, for samples containing marble within 0–1.14 % by weight, an increase in the concentration of marble decreases their relative elongation over the time of UV radiation.

For samples without marble, in the absence of UV radiation, the relative elongation is 1,002 %; hourly UV radiation leads to its reduction by an average of 7.2 %.

For samples with a marble content of 0.42 % by weight without UV radiation, these indicators are 851 % and 4.75 %, respectively. For samples with a marble content of 1.14 % by weight – 895 % and 2.6 %, respectively; and for samples with a marble content of 5.1 % by weight – 645 % and 4.8 %, respectively.

The nature of the dependence of the relative elongation of samples with a marble content of 1.78 % by weight on the duration of UV radiation differs significantly from previous samples. Thus, their relative elongation in the absence of UV radiation is 795 %, which is only 207 % less than the relative elongation of samples without marble. However, hourly UV radiation leads to a decrease in the relative elongation by an average of 9.43 %.

To determine the chemical effect of UV radiation on the samples of polymeric films, we examined the IR spectra of a sample, not exposed to UV radiation, and a sample that was exposed to UV radiation over 30 hours (Fig. 7) [28]. Fig. 8 shows that the IR-region of $1,720\text{ cm}^{-1}$ has a characteristic peak, which indicates the formation of a $>\text{C}=\text{O}$ bond in the film. In other words, polyethylene macromolecules

under the influence of UV radiation are destroyed with the formation of shorter macromolecules whose ends host a $>\text{C}=\text{O}$ group.

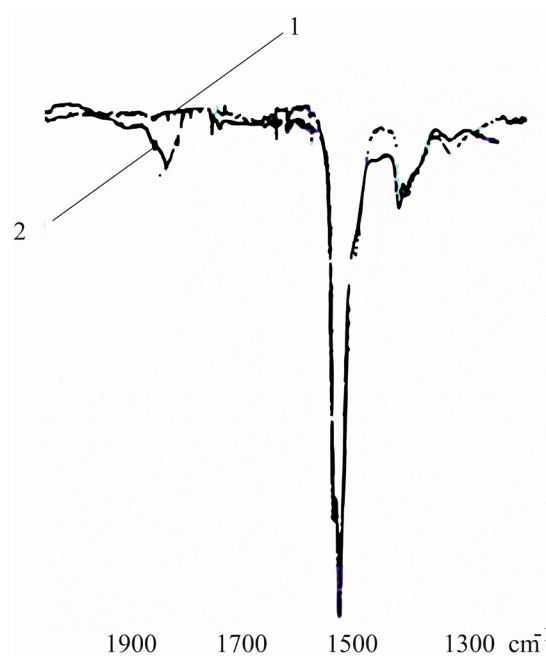


Fig. 7. The IR spectrum of a stretch film in the range of 1,900–1,300 nm: 1 – original sample; 2 – a sample exposed to UV irradiation over 30 minutes

Thus, the samples with a marble content of 1.78 % by weight are most suitable for the manufacture of a biodegradable polymeric material capable of collapsing under the influence of UV radiation.

7. Discussion of results of studying the samples of biodegradable material based on polyethylene and marble microparticles

Our analysis of the dependence of the relative elongation of samples on the duration of UV radiation shows that, for a sample containing 1.78 % by weight of marble microparticles, it differs greatly from other samples. In particular, for a sample containing 1.78 % by weight of marble microparticles, hourly UV radiation leads to a decrease in the relative elongation by an average of 9.43 %.

When UV rays reach the samples, some of them pass through the waste's material, a part is absorbed, and a part is reflected. At the same time, the destruction of polymeric macromolecules is facilitated by the absorbed energy. The above-described nature of the dependence of the relative elongation of samples on the duration of UV radiation allows us to assume that, when the content of marble microparticles is 1.78 % by weight, the share of the absorbed energy is maximal. At the same time, there is intense destruction of polyethylene macromolecules with the formation of shorter macromolecules with the end groups of $>C=O$.

Thus, if the content of marble microparticles in the composition increases, the share of energy that passes unhindered through the sample decreases while increasing the share of the absorbed energy. However, increasing the content of marble microparticles above 1.78 % by weight increases the share of energy, which is repeatedly reflected from the adjacent marble microparticles and, as a result, is not absorbed by the polymer.

Thus, the composition "high-pressure polyethylene – marble microparticles" with a marble content of 1.78 % by weight can be used for the manufacture of a packing stretch film.

In this case, the respective stretch film would have high elastic properties when used as intended and is prone to decomposition under the influence of solar radiation after it loses its consumer properties.

Unlike unfilled polyethylene, in which the photochemical destruction under the influence of UV rays occurs primarily in surface layers, in the designed composition the said destruction also occurs in deep layers. This is due to reflecting the UV rays in different directions inside the polymeric matrix and preventing them from passing through the composition without interacting with the polymeric matrix.

Marble microparticles are used as a filler for the proposed biodegradable polymeric material, which are an industrial waste, which would not lead to a significant increase in the cost of the said material. At the same time, destroying the waste of this material is greatly simplified.

The disadvantage of the studied composition is some difficulties in its manufacturing directly at conventional

technological extrusion equipment with the simultaneous formation of finished articles. This issue could be resolved through the use of adhesive additives to improve the interaction between a polymeric matrix and the filler particles; however, that could adversely affect the operational properties of the composition.

It is more expedient to preliminarily make the composition's granules with the required ratio of components and to subsequently process the resulting granules into finished products.

Further research is planned to determine the physical-mechanical properties of polymeric films with different ratios of film thickness to the average size of filler particles.

8. Conclusions

1. It has been shown that marble microparticles as part of a polymeric composition based on high-pressure polyethylene reduce the strength characteristics of this material under the influence of ultraviolet radiation. Reducing the strength of such a polymeric material facilitates the destruction of relevant waste and increases its specific surface, which would increase the speed of their decomposition under natural conditions. Therefore, the thermoplastic composition "high-pressure polyethylene – marble microparticles" can be considered to be a biodegradable polymeric material that is destroyed by ultraviolet radiation.

2. We have made samples of the biodegradable polymeric material with the different content of an additive – marble microparticles (from 0 to 5.1 % by weight) were obtained. The physical-mechanical properties of the fabricated samples with the different content of marble microparticles have been investigated.

We have experimentally proven that UV radiation destroys polyethylene macromolecules into separate fragments with the formation of $>C=O$ bonds, making it possible to decompose relevant waste under the influence of sunlight.

The expediency of using the composition "high-pressure polyethylene – marble microparticles", with a marble content of 1.78 % by weight, has been confirmed for the manufacture of a packing stretch film.

To produce articles from the studied composition, it is proposed to preliminarily make a granulate of the required formulation and/or apply adhesive additives to improve the interaction between a polymeric matrix and the filler particles.

Acknowledgments

We express sincere gratitude to Liubov Ivanivna Melnyk, Candidate of Technical Sciences, Associate Professor, and Iryna Viacheslavivna Nikolaieva, Magister, from the Department of Chemical Technology of Composite Materials at the National Technical University of Ukraine "Igor Sikorsky Kyiv Polytechnic Institute" for their assistance in conducting experimental studies.

References

1. Three-age system. Available at: https://ru.qwe.wiki/wiki/Three-age_system
2. Plastics – the Facts 2018. Available at: https://www.plasticseurope.org/application/files/6315/4510/9658/Plastics_the_facts_2018_AF_web.pdf

3. Mikulionok, I. O. (2011). Pretreatment of recycled polymer raw material. *Russian Journal of Applied Chemistry*, 84 (6), 1105–1113. doi: <https://doi.org/10.1134/s1070427211060371>
4. Kirsh, I. A., Chutkina, E. P. (2010). Biorazlagaemye polimernye kompozitsii na osnove othodov agropromyshlennogo kompleksa. *Plasticheskie massy*, 5, 45–48.
5. Chung, D. D. L. (2010). *Composite Materials: Science and Applications*. Springer Science & Business Media, 371.
6. Singh, N., Hui, D., Singh, R., Ahuja, I. P. S., Feo, L., Fraternali, F. (2017). Recycling of plastic solid waste: A state of art review and future applications. *Composites Part B: Engineering*, 115, 409–422. doi: <https://doi.org/10.1016/j.compositesb.2016.09.013>
7. Scaffaro, R., Maio, A., Suter, F., Gulino, E., Morreale, M. (2019). Degradation and Recycling of Films Based on Biodegradable Polymers: A Short Review. *Polymers*, 11 (4), 651. doi: <https://doi.org/10.3390/polym11040651>
8. Karaogul, E. (2019). Effects of asphodel tuber and dolomite on the properties of bio-hybrid films processed by a twin screw extruder. *BioResources*, 14 (2), 4473–4488. Available at: https://bioresources.cnr.ncsu.edu/wp-content/uploads/2019/04/BioRes_14_2_4473_Karaogul_Effects_Asphodel_Tuber_Dolomite_Props_Biohybrid_Films_15283-1.pdf
9. Nayak, S. K., Satapathy, A. (2020). Development and characterization of polymer-based composites filled with micro-sized waste marble dust. *Polymers and Polymer Composites*, 096739112092606. doi: <https://doi.org/10.1177/0967391120926066>
10. Choudhary, M., Singh, T., Dwivedi, M., Patnaik, A. (2019). Waste marble dust-filled glass fiber-reinforced polymer composite Part I: Physical, thermomechanical, and erosive wear properties. *Polymer Composites*, 40 (10), 4113–4124. doi: <https://doi.org/10.1002/pc.25272>
11. Nayak, S. K., Satapathy, A. (2019). Wear analysis of waste marble dust-filled polymer composites with an integrated approach based on design of experiments and neural computation. *Proceedings of the Institution of Mechanical Engineers, Part J: Journal of Engineering Tribology*, 234 (12), 1846–1856. doi: <https://doi.org/10.1177/1350650119896170>
12. Sharma, A., Choudhary, M., Agarwal, P., Kumar Patnaik, T., Kumar Biswas, S., Patnaik, A. (2020). Experimental and numerical investigation of thermal conductivity of marble dust filled needle punched nonwoven jute-epoxy hybrid composite. *Materials Today: Proceedings*. doi: <https://doi.org/10.1016/j.matpr.2020.07.097>
13. Ray, S., Rout, A. K., Sahoo, A. K. (2018). A study on erosion performance analysis of glass-epoxy composites filled with marble waste using artificial neural network. *UPB Scientific Bulletin, Series B: Chemistry and Materials Science*, 80 (4), 181–196.
14. Bakshi, P., Pappu, A., Patidar, R., Gupta, M. K., Thakur, V. K. (2020). Transforming Marble Waste into High-Performance, Water-Resistant, and Thermally Insulative Hybrid Polymer Composites for Environmental Sustainability. *Polymers*, 12 (8), 1781. doi: <https://doi.org/10.3390/polym12081781>
15. Sokolova, Yu. A., Shubanov, S. M., Kandyrin, L. B., Kalugina, E. V. (2009). Polimernye nanokompozity. *Struktura. Svoystva. Plasticheskie massy*, 3/4, 18–23.
16. Rybkina, S. P., Paharenko, V. A., Shostak, T. S., Paharenko, V. V. (2008). Osnovnye napravleniya v oblasti sozdaniya biorazlagaemykh termoplastov. *Plasticheskie massy*, 10, 47–54.
17. Mikulyonok, I. O. (2013). Equipment for preparing and continuous molding of thermoplastic composites. *Chemical and Petroleum Engineering*, 48 (11-12), 658–661. doi: <https://doi.org/10.1007/s10556-013-9676-x>
18. Mikulionok, I. O. (2015). Classification of Processes and Equipment for Manufacture of Continuous Products from Thermoplastic Materials. *Chemical and Petroleum Engineering*, 51 (1-2), 14–19. doi: <https://doi.org/10.1007/s10556-015-9990-6>
19. Bombelli, P., Howe, C. J., Bertocchini, F. (2017). Polyethylene bio-degradation by caterpillars of the wax moth *Galleria mellonella*. *Current Biology*, 27 (8), R292–R293. doi: <https://doi.org/10.1016/j.cub.2017.02.060>
20. Tadmor, Z., Gogos, C. G. (2006). *Principles of polymer processing*. John Wiley & Sons, 984.
21. Mikulionok, I. O., Radchenko, L. B. (2012). Screw extrusion of thermoplastics: I. General model of the screw extrusion. *Russian Journal of Applied Chemistry*, 85 (3), 489–504. doi: <https://doi.org/10.1134/s1070427211030305>
22. Mikulionok, I. O., Radchenko, L. B. (2012). Screw extrusion of thermoplastics: II. Simulation of feeding zone of the single screw extruder. *Russian Journal of Applied Chemistry*, 85 (3), 505–514. doi: <https://doi.org/10.1134/s1070427211030317>
23. Rauwendaal, C. (2014). *Polymer extrusion*. Carl Hanser Verlag GmbH & Co. KG, 950. doi: <https://doi.org/10.3139/9781569905395>
24. Mikulionok, I., Gavva, O., Karvatskii, A., Yakymchuk, M. (2017). Modeling and analysis of the process of polymeric film cooling on the drum with a liquid cooling agent. *Eastern-European Journal of Enterprise Technologies*, 5 (5 (89)), 67–74. doi: <https://doi.org/10.15587/1729-4061.2017.110687>
25. Agassant, J.-F., Avenas, P., Carreau, P. J., Vergnes, B., Vincent, M. (2017). *Polymer Processing*. Carl Hanser Verlag, 883.
26. Vlachopoulos, J., Vlachopoulos, N. D. (2019). *Understanding rheology and technology of polymer extrusion*. Dundas: Polydynamics Inc., 337.
27. Mikulionok, I. O. (2013). Screw extruder mixing and dispersing units. *Chemical and Petroleum Engineering*, 49 (1-2), 103–109. doi: <https://doi.org/10.1007/s10556-013-9711-y>
28. Nikolaieva, I. V., Petukhov, A. D., Shnyruk, O. M., Melnyk, L. I., Nudchenko, L. A., Musienko, O. S. (2018). UF promin – ruivnykh khimichnykh zviyaziv stretch-plivok polietylenu vysokoho tysku. XI Mizhnarodna naukovo-tekhnichna WEB-konferentsiya «Kompozitsiyni materialy» (kviten 2018 r.): zbirka materialiv. Kyiv: KPI im. Ihoria Sikorskoho, 51–54.