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Biodegradable polymer materials and modifying additives: state of the art. Part III

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ABSTRACT: One of the most demanded materials on the planet is plastic, the excellent performance of which contributes to the accumulationof a significant amount of waste on its basis. In this regard, a new approach to the development of these materials hasbeen formed in scientific circles: the production of polymer composites with constant performance characteristics for a certainperiod and then capable of destruction under the influence of environmental factors. Analysis of the current state of the industry of polymeric materials shows that the most urgent is the use of such classical polymers as polyolefins and polyvinyl chloride. First of all, the optimal solution to this problem due to the lack of a suitable replacement for traditional polymers is the development of composites based on them with the use of biodegradable additives. In this case, a set of problems associated with waste disposal issolved: the decomposition period of the recycled waste is significantly reduced, the territories required for plastic waste are reduced. The paper outlines the preconditions for the emergence and further development of the field of biodegradable polymers. The mainquantitative characteristics of the production capacities of manufactured bioplastics by types, regions and industries of applicationare given. Modern methods of reducing and regulating the degradation time of polymer materials are presented. The main global and domestic manufacturers of biodegradable polymers and their products are listed, as well as a list of the main manufacturers of biodegradable additives for polymeric materials. Modern types of bioplastics based on renewable raw materials, composites with their use, aswell as modified materials from natural and synthetic polymers are listed. The main methods for determining the biodegradability of existing bioplastics are described.

KEYWORDS: biodegradation, biodegradable additives, petrochemical raw materials, polymers, plasticizers, plant sources.

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NATURAL BIODEGRADABLE POLYMERS

Natural polymers are characterized by controlled biodegradation in nature, under the influence of microorganisms, water and soil, these materials decompose mainly into carbon dioxide and water. The main sources of raw materials are starch, cellulose derivatives, chitin, chitosan, lactic acid, hydroxyalkanoates.

High molecular weight carbohydrates. The main way of using these biopolymers for practical purposes is the development of biodegradable composites with properties similar to those of traditional plastics [55–57].

Starch. To create bioplastics, chemically modified starch is used, which leads to a change in its main properties: hydrophilic, rheological, physicochemical. The main technique is cross-linking due to hydroxyl groups [57]. Starch copolymers and starch-based composites are used as thermoplastic materials [58].When silica is added, thermoplastically processed starch is obtained in the form of nanocomposites with a water content of less than 10% [59].

Destructed starch is used in composites with synthetic polymers (polyethylene, polyvinyl alcohol, aliphatic polyesters, polyoxybutyrate) [59–62].

A wide range of complexing agents (deoxy succinates, epoxides, layered organosilicates) are used to regulate the biodegradation period [61–62].

Cellulose. Thermoplastic and soluble cellulose derivatives are obtained by chemical modification. In practice, cellulose ethers and esters are often used, obtained by interaction with OH-groups (3 alcohol hydroxyls) in each monosaccharide unit [63–66]. Other modifications of cellulose are also obtained: alkaline (under the action of sodium hydroxide solution), carboxymethyl cellulose (ether with glycolic acid), methyl cellulose of three modifications (methyl cellulose ether), mono-, di- and triacetyl cellulose (cellulose triacetate), nitrocellulose of three modifications (cellulose nitrates) [67].

Nitrocellulose is the basis of the first man-made plastic (celluloid). Its main disadvantage is easy flammability and release of toxic nitrogen oxides during combustion [67].

Etrol plastic is obtained on the basis of acetates, acetopropionates, acetobutyrates, cellulose nitrates and ethyl cellulose [67].

Chitin. There are three crystallographic modifications, which differ in the arrangement of molecular chains in the unit cell of a crystallite [68]. The most common modification, α-chitin, is characterized by close packing of macromolecules and is provided by antiparallel arrangement of chains in the unit cell. β-Chitin is an unstable crystal hydrate with a parallel arrangement of chains. The γ-chitin cell has two parallel chains and one antiparallel to them. The possibilities of chemical modification of chitin facilitate the production of materials of various structures and properties and solve environmental problems by using them in biodegradable polymers [69–70].

Chitosan. In contrast to chitin, chitosan has wider application possibilities [71]. After treatment with a deacetylation reaction, the crystalline regions will contain residual acetyl groups [72–73]. The distribution of these groups affects the deformation and strength properties of the fibers. During the enzymatic hydrolysis of chitosan, low molecular weight compounds are obtained that are biodegradable and biocompatible, and are non-toxic. The fibers obtained from a chitosan solution by the NANOSPIDER technology are distinguished by an overdeveloped surface and porosity, and have an effective wound healing and bactericidal activity [74].

Natural rubber. Rubber is isolated from the latex by coagulation with formic, oxalic or acetic acid and subjected to further processing. However, the production of stereoregular synthetic rubbers contributed to the reduction of the use of natural rubber in a number of industries [75–76].

Polyhydroxyalkanoates. The most promising is poly-3-hydroxybutyrate or polyhydroxybutyrate (PHB), discovered by microbiologists in 1925. Bacterial PHB is characterized by good elastic-strength properties, optical activity, thermoplasticity, and piezoelectric properties [77]. Articles made of such polymers are obtained from a melt or from its solutions in organic solvents in pure form and as a component of mixtures and copolymers based on it [78]. In terms of technological characteristics, it is practically not inferior to traditional thermoplastics, does not require special disposal, does not pollute the environment [79]. The production of PHB is waste-free and characterized by low energy consumption.

The compatibility of these biopolymers with industrial synthetic ones has contributed to the creation of new multicomponent composites, which reduces the cost of products [80-82].

Protein. The use of proteins for the production of biodegradable composite materials is not widely used. Block copolymers for medical purposes are known [76].

SYNTHETIC BIODEGRADABLE POLYMERS

Traditional polymers are characterized by enhanced physical, mechanical and performance characteristics, biological stability [10, 17]. The production of composites based on them is an effective and cost-effective method of modification, allows the maximum use of the properties of each, and also contributes to the disposal of industrial polymer waste.

One of the methods for obtaining biodegradable synthetic plastics is the synthesis of polyesters and polyesteramides, for example, copolyesters based on aliphatic diols and organic dicarboxylic acids [12, 25].

Another method for creating biodegradable PM is the development of composites based on natural polymers, for example, starch, cellulose, chitosan, or proteins [25, 31–33]. It is important to choose the right ratio of the components, at which the operational properties of the obtained PM approach the properties of the original conventional polymers.

Polyolefins (polyethylene and polypropylene). The use of starch in the serial production of biodegradable polyethylene-based PMs has been mastered by several companies: packaging material under the Mater-Bi brand from Novamont S.p.A. (Italy); Polyclean TM concentrate for the production of biodegradable films from Archer Daniels Midland (USA); Ecostar Plus concentrate from St. Sawrence Starch (USA). In addition to starch, the composition includes additives that act as a catalyst for the biodegradation of starch [83-85].

The most famous product with the addition of starch is the Mater-Bi material from Novamont S.p.A (Italy), which degrades in the soil in 60 days without the release of harmful products [83].

For packaging, composites are being developed based on polyethylene and polypropylene waste with the addition of wastes from flour and cereals, starch, sugar, and confectionery enterprises [86]. Chitin and chitosanare known to be used as fillers [69–70].

Photodegradable copolymers of ethylene with carbon monoxide have been developed. Photoinitiators of decomposition are vinyl ketone monomers in an amount of 2–5%, cellulose pulp, alkyl ketones or fragments containing carbonyl groups. The resulting films serve for

8–12 weeks, then they are photo- and biodegradable. Iron and nickel dithiocarbamate and their peroxides are also used as photosensitive additives.

Polyvinyl chloride. Basically, to accelerate the degradation process of composites with PVC, additives are used: natural high-molecular compounds that make up various food products – starch, rye, corn and wheat flour, barley, millet, buckwheat grain processing waste, wood processing products (wood flour), cellulose, etc. its derivatives, lignin, yeast, or blue-green algae [85–86].

The introduction of fillers weaken the polymer chain of PVC molecules. When injected into the soil, the formed monomeric fragments serve as a nutrient medium for microorganisms; the period of their biodegradation is from 6 to 36 months [87].

The nature of the plasticizer in the PVC composition also has a significant effect on biodegradation. PVC materials plasticized with phthalates of higher alcohols are the most resistant to the action of microorganisms, while the least biostable are compositions containing sebacates [87–88]. The authors of the article have obtained a class of non-toxic biodegradable plasticizers for adipates of oxyalkylated alcohols of the aliphatic and aromatic series. The use of such alcohols increases the degree of decomposition of the material. In the works, the biodegradation of PVC plasticates using the developed plasticizers is investigated, the ecotoxicity of their decay products is studied [89–92].

Polystyrene. Starch is mainly used as a biodegradable additive for the creation of biodegradable composites based on polystyrene [93–94]. To improve the compatibility of polymer components, copolymers of polystyrene and maleic anhydride have been proposed [95]. It is known to obtain photodegradable polymers based on PS by synthesizing copolymers of styrene with carbon monoxide [96]. The introduction of vinyl ketone monomers in an amount of 2–5% as a copolymer to styrene makes it possible to obtain photodegradable polymers. Of interest are the developed functionalized polymers and copolymers of styrene as polymer binders in biodegradable plastics. The range of such bioplastics is small, and their cost is 2–3 times higher than the cost of large-capacity polymers incapable of biodegradation [97].

Polyesters and polyesteramides. Polyethylene terephthalate (PET) and polybutylene terephthalate (PBT) are characterized by high biostability [98–101]. However, if the polymer envelops the filler well, the mushroom resistance of the resulting composition is high.

To create BPM based on polyesters, it is possible to use hydroxy acids as modifying components. Also, an expected method of increasing the biodegradability of PET is compounding it with a biodegradable component, for example, with starch. However, during the composting process, only starch is rapidly decomposed, and PET generally does not biodegrade [98–101].

Polylactides. The industrial synthesis of polylactide is carried out by polymerization of lactide [102–104] or by azeotropic polycondensation of lactic acid [103]. Today polylactide is one of the cheapest biodegradable plastics, but much more expensive than polyethylene and polystyrene. However, the development of a less energy-intensive industrial method for producing polylactide continues. For this purpose, to increase the stereospecificity of the process, compounds of zirconium, hafnium, gold, and platinum are used as catalysts [104–106].

Polylactide with a molecular weight of 102,000 and an extremely high melting point (210–218^{o}C) is known to be obtainedthrough formation of a special supramolecular structure [104–106].

Despite the advantages of polylactide, the rate of its biodegradation (half-life is 168 days) is quite high. For this reason, research is underway to obtain biodegradable copolymers of lactic acid with a controlled rate of biodegradation.

A promising comonomer is glycolide obtained from glycolic or monochloroacetic acids [107]. Polyglycolide due to hydrolytic instability (20 days) imposes certain restrictions on its use as a surgical material.

The synthesis of such copolymers makes it possible to successfully combine the properties of polylactide and polyglycolide and to control the rate of biodegradation [108–111].

The synthesis of copolymers based on dimethyl terephthalate, lactic acid and hexanediol using titanium butoxide as a catalyst has been developed [108–111].

CONCLUSION

Analysis of information sources showed that in the field of biodegradable plastics, there is a constant increase in the production capacity of already demanded polymeric materials, as well as the development and expansion of the range of new compositions that are characterized by environmental friendliness, the ability to modify the required specified service life and the ability to biodegrade without harming the environment. In connection with this, the range of developed biodegradable additives is also expanding.

In modern research, from our point of view, an important problem is to impart biodegradability properties to large-tonnage traditional polymers (polyethylene, polypropylene, polyvinyl chloride, etc.), the production of which is currently more environmentally friendly, energy- and resource-saving and provides them with undeniable advantages.

However, due to the depletion of petrochemical sources of raw materials for the production of traditional synthetic polymers, it is necessary to develop additional possibilities and promising directions for creating analogues of these polymers from plant raw materials.

2021; 13 (2):
73–78

At present, promising biodegradable bio-based plastics, for example, polylactides, polyesters and polyhydroxoalkanoates, are distinguished by wide possibilities for modeling the structure and properties of the materials obtained and are approaching the main indicators of traditional synthetic polymeric materials. But their serial production is not organized, which is primarily due to the availability of raw materials and its high cost, and also the reduction of the risk of the negative impact of the decay products of bioplastics on nature and the animal world is not fully justified.

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