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ORIGINAL PAPER

BIOPLASTIC PACKAGING MATERIALS IN CIRCULAR ECONOMY

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ABSTRACT. **Background:** The European strategy for plastics focuses on adjusting the EU regulations to the fulfilment of circular economy tasks. Circular economy is an approach that will soon lead to considerable changes in numerous branches of modern economy. To a large extent, they will also affect the packaging industry.
Methods: A particular interest has been attracted by aliphatic polyesters such as polylactide (PLA) and polyhydroxyalkanoates (PHA). This work presents the bioplastic market and the selected examples of the latest solutions in bioplastic packaging materials. In the near future, the presented bioplastics have a chance to become some of the most desirable packaging materials
Results and conclusion: Bioplastics seem to be an alternative to conventional plastics used for packaging production. As the focus shifts to creation of sustainable environment and prevention of plastic waste disposal in the environment, the production of bioplastics has gained much attention due to their biodegradability.

Key words: bioplastics, packaging materials, circular economy.

CIRCULAR ECONOMY IN PACKAGING

The global production of plastics has been growing for years. Packaging applications are the largest application sector for the plastics industry. They represent 39.6% of the total demand for plastics, which generates the increase in their production [PlasticsEurope, 2016]. As a result, over the last 70 years, global plastics production grew from nearly 0.5 million tons in 1950 to over 365 million tons in 2016. In 2017, it reached about 348 million tons per year. Europe is the second largest producer of plastics in the world after China with around a 40% market share for packaging purposes [Plastics Europe, 2017].

Although the use of plastics has many advantages in comparison to other materials [Andrady, Neal 2009], their drawbacks are becoming more visible. Most materials used in the packaging industries are produced from fossil fuels and they are practically nondegradable [Nampoothiri et al. 2010]. Despite that, for many years, the need to recover raw materials was not acknowledged. However, recently it has become necessary to change the approach to the management of packaging and packaging waste. In December 2015, the European Commission adopted the EU action plan for circular economy. Therefore, recently the EU has enacted two legal documents related to the packaging industry and waste management: Directive (EU) 2018/851 of the European Parliament and of the Council of 30 May 2018 amending Directive 2008/98/EC on waste and Directive (EU) 2018/852 of the European Parliament and of the Council of 30 May 2018 amending Directive 94/62/EC on packaging and packaging waste.

The EU regulations related to circular economy (CE) focus on packaging production and recovery of materials from waste.

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According to the European Commission, the potential for recycling plastic waste in the European Union remains unfulfilled. Europe generates 25.8 million tons of plastic waste per year but only 30% of it is recycled. Of 34 million tons of plastic waste, as many as 93% are stored on landfills and in the oceans [Pathak et al. 2014].

It is estimated that 100 million tons of polymers cause an ecosystem service damage amounting to approximately US$ 13 billion per year [United Nations Environment Programme 2014]. In circular economy (CE), products, materials and raw materials circulate as long as possible, which leads to minimization of waste. The main purpose of those actions is to increase recycling and limit the storage of packaging waste to 55% by 2025. Circular economy takes into consideration all stages of product life cycle, i.e. design, production, consumption and reuse of waste.

According to the European Union, CE has been introduced as a high-level strategy to move our societies beyond these limits [European Commisssion 2015]. Circular economy presents a solution to limit the excessive use of raw materials and makes it possible to reuse those materials that have already been used. However, only some types of materials can undergo repeated recycling or reuse. According to Ghisellini et al. [2016], CE will promote high value material cycles instead of recycling only low value raw materials as in traditional recycling). Furthermore, decomposition of plastics is known to be very difficult, and it involves high emission of CO_2 and many other toxic compounds [Emadian and Onay, 2017]. Many doubts are also raised by the fact that the majority of recycling methods cause an about 10% loss of material and material quality [Merrild et al. 2012]. Given the gigantic amount of plastics used for products, bioplastics may be a way to get a handle on our overwhelming waste problem. Recently the attention in the packaging industry regarding the use of bioplastics has been shifting from compostable/biodegradable materials towards bio-based materials [Nampoothiri et al.2010].

BIOPLASTICS MARKET

According to Mohanty et al. [2002], bioplastics are a new generation of plastics that limits environmental impact in terms of global warming and energy consumption. The term “bioplastics” is an umbrella term which describes several groups of materials: bioplastics from renewable resources, bioplastics from fuel resources, and bioplastics derived partially from renewable resources and partially from fuel resources. In terms of their susceptibility to disintegration, they can be divided into biodegradable materials, which include compostable materials, as well as non-biodegradable materials [Cooper 2013]. The non-biodegradable materials produced from renewable raw materials include, among others, polyethylene produced from bio-based ethanol. Materials that are biodegradable in the course of industrial composting but do not originate from renewable resources include, among others, PBAT or PCL, which may be obtained from natural products or petroleum. Biodegradable materials produced from renewable raw materials include PLA, PBS and PHA.

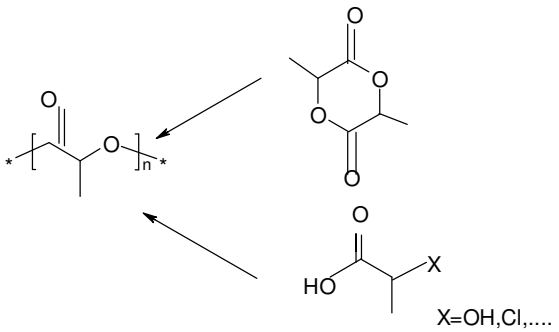
Biodegradable materials are undoubtedly eco-friendly but they have certain limitations such as high costs of production and low mechanical tendency. The decrease in the availability of gasoline and diesel due to higher costs exacerbates the shortage of resources, which promotes the need to create bioplastic materials [Thakur et al. 2018]. Therefore, natural polymers and polymers from renewable resources seem to be an alternative to conventional plastics. Their use is advantageous also from the economic point of view: their production requires less energy and does not result in toxic by-products.

The demand for bioplastics is constantly growing because they are applied in various contexts in order to manufacture more and more complex products. In 2017, the amount of biodegradable plastics produced at the global level was about 880 Gg [European bioplastics 2018], corresponding to less than 0.3% of the total amount of plastics produced that year (320,000 gigagrams). The demand for bioplastics is expected to grow to about 6 million tons per year.

The main recipient of bioplastics is the packaging industry. In the recent years, there have been developed many packaging materials based on starch, polylactide, polyhydroxyalkanoates, poly(glycolic acid) (PGA), aliphatic-aromatic polyesters, cellulose or lignin that are currently present on the market. Bio-based resources have a major role in the production of novel and bio-based materials [Brodin et al. 2017].

BIOPLASTIC PACKAGING MATERIALS

Bioplastic packaging materials, such as PLA and materials reinforced with natural fibers, have attracted particular attention over the past few years [Soroudi and Jakubowicz, 2013]. PLA is produced by conversion of corn, or other carbohydrate sources, into dextrose, followed by fermentation [Panseri et al. 2018] of polysaccharides or sugar, e.g. extracted from corn, potato, cane molasses or sugar-beet, into lactic acid [Murariu and Dubois, 2016]. PLA is produced as a result of ring opening polymerization and poly-condensation of lactic acid [Dubey et al., 2017] (Figure 1).



Source: Dubey et al. 2017

Fig. 1. Lactide and lactic acid monomer to form PLA

Studies show that PLA possesses good mechanical properties (high Young's modulus and tensile strength) and a high level of transparency. Similarly to conventional petrochemical plastics, it is easy to process [Auras et al. 2004]. The world literature presents many examples of PLA modification. One such solution was suggested by Przybytek et al. [2018], who combined polylactide (PLA), potato thermoplastic starch (TPS) and plant glycerin, and processed them by melt extrusion with epoxidized soybean oil as the reactive modifier. The presented studies have shown that it is possible to replace even up to 25% of PLA with mTPS. It will help to reduce product costs while retaining similar characteristics and compostability.

The addition of another biodegradable polymer-poly(ϵ-caprolactone) (PCL) changes the properties of PLA [Ostafinska et al. 2017] . The PLA/PCL combination was characterized by high rigidity (due to the presence of PLA) and hardness (due to the presence of PCL). The obtained material is promising as regards application in the packaging industry as well as other branches of industry. Spiridon and Tanase [2018] produced new poly (lactic acid)-lignin biocomposites. The addition of lignin to PLA in the amount of 7 to 15 wt% resulted in greater tensile strength. Biocompatibility studies evidenced that the addition of lignin to a poly (lactic aci d) matrix can allow for tailoring the final properties of the composites without inducing any significant changes in cell metabolic activity (compared to poly (lactic acid) itself).

Apart from PLA, the most commonly used biomaterials include starch. Starch has very favourable physical characteristics, such as high barrier properties and good rheological properties. Polymers combined with starch can be used without any limitations. The application of starch in the packaging industry dates back to the 90s. The first bags made from Mater-Bi became available in Germany in 1992 [Byun et al. 2014]. The studies carried out by Luchese et al. [2017] showed that corn and cassava starch may be considered promising alternatives for food packaging. The mechanical property values of starch based films were comparable to those of LDPE based film. The application of thermoplastic starch (TPS) offers numerous possibilities. Due to its characteristics, it may be processed in many ways, including casting, thermo-molding or extrusion and injection molding. As part of their studies, Sagnelli et al. [2016] produced starch-based bioplastic prototypes fabricated

from an almost amylopectin-free starch synthesized directly in the barley grain. Also modified starch, including starch modified with citric acid, turned out to be a very promising biopolymer. That modification improves the thermodynamic stability of starch [Qian et al.2015; Ban et al.2006]. Domene-López et al. [2018] obtained interesting biodegradable materials as a result of combining potato starch/PVA with different concentrations of rosin. The addition of 8% rosin to starch/PVA blends led to tensile strength values higher than 10 MPa and elongation at break values close to 2000% in comparison to LDPE. The addition of 8% rosin to starch/PVA blends led to increases in maximum tensile strength and elongation at break by 72% and almost 400%.

Polyhydroxyalkanoates (PHA) are biopolymers of hydroxyl fatty acids that act as storage compounds during unbalanced microbial growth [Wijeyekoon et al. 2018]. We know such polyhydroxyalkanoates as PHB (poly(3-hydro-xybutyrate) and its copolymers: poly(3hydroxybutyrate-co-3-hydroxyvalerate) (PHBV), poly[(R)-3-hydroxybutyrate-co-(R)-3-hydroxyhexanoate (PHBHx) and poly[(R)-3-hydroxybutyrate-co-4-hydroxybutyrate (P3HB4HB) [Gahlawat and Soni, 2017]. Polyhydroxybutyrate (PHB) bioplastics got the attention of the scientific community due to their low CO2 emission [Mostafa et al. 2015]. PHA polymers are fully biodegradable and compostable. Prokaryotes cause decomposition of PHA into carbon dioxide and water, which are consumed during plant growth. In addition, thermoplastic properties of PHA are similar to those of petrochemically derived polypropylene and polyethylene [Morgan-Sagastume et al. 2010]. To draw a comparison, polylactic acid (PLA) is compostable, but may remain in marine environments for up to a thousand years [DiGregorio, 2009]. However, the market size is still limited, mostly due to the elevated costs of PHA production, so many scientists are looking for manufacturing methods that require small outlays. For this purpose, PHA was produced with the use of such substrates as molasses, plant oils and fatty acids or microbes[Ntaikou et al. 2009]. Moreover, the use of waste substrates and/or mixed microbial consortia is of increasing interest as a strategy to reduce production cost [Burniol-Figols et al. 2018]. Challenges for PHA production include dependence on pure carbon sources, such as glucose; the requirement of organic substitutes for production of different types of PHA; the possibility of contamination; and the use of large amounts of solvents in downstream processing [Rodriguez-Perez et al., 2018]. One of the raw materials used for PHA production may be macroalgal biomass. This is exemplified by the studies conducted by Ghosh et. al. [2019], in which the authors evaluated the carbohydrate composition of 7 seaweeds to provide a carbon source for PHA produced by Haloferax mediterranei. Among the tested combinations, green macroalgae Ulva sp. had the best composition for the maximum yields of PHA. Sawant et al. [2018] proved that red algae Gelidium amansii can be used for PHA production without extensive hydrolysis.

Agro-industry waste has also been investigated as a low cost substrate for PHA production. Organic waste may be subjected to anaerobic fermentation or hydrothermal treatment to produce organic acid rich solutions. The volatile fatty acid rich liquors are an ideal feedstock for PHA production. Such a solution is illustrated in the work of S. Wijeyekoon et al. [2018]; the authors studied PHA production potential of substrates generated through subcritical wet oxidation (WO) of organic biomass. They presented two aspects of mixed culture PHA fermentation: first, the impact of a feed characterized by low carbon-nitrogen ratio, this being a natural characteristic of wet oxidized biomass conversion; and second, the influence of dissolved oxygen limitation as an alternative substrate limitation to enhance metabolism of carbon into PHA storage. The results of the studies showed that substrate feeding regime and oxygen concentration can be used to control the PHA yield and accumulation rate, thereby enhancing PHA production viability from nutrient rich biomass streams. The enzyme which produces PHA was extracted by Reddy et al. [2017] from two bacterial strains: P. pseudoflava and P. palleronii. As evidenced by the studies, P. palleronii showed higher PHA synthase enzyme activity than P. pseudoflava. It indicates the possibility of feeding the P. pseudoflava with cheap VFA rich fermented waste to produce PHA.

The scientific group of Bilo et al. [2018] obtained bioplastic from rice straw. Rice straw is considered agricultural waste, and its management is easy because it does not require separation from other waste [Dominguez-Escriba and Porcar, 2011]. The obtained bioplastic material exhibits good mechanical properties, with tensile strength and elongation at break equal to 45 MPa and 6.1% and 10 MPa and 63% for dried and wet dumbbell respectively. It proves that mechanical properties of bioplastic are comparable to those of polystyrene, while cast bioplastic in wet state is similar to plasticized poly(vinyl chloride). Moreover, the studies showed high mechanical performance of the newly obtained bioplastic both in dry and wet state.

Ramakrishnan et al. [2018] produced bioplastics with the use of chicken feathers followed by the mixing of different concentrations of glycerol. It was observed that the increased volume of glycerol reduced the time required for foil degradation. With 10 wt% glycerol content, the obtained foil disintegrated after 6 hours. This is the result of the poor strength and bonding between keratin and glycerol within the film [Ullah et al. 2011].

SUMMARY

Circular economy is one of the major principles of the economic policy of the European Commission. It is an approach adopted in order to tackle environmental challenges and support sustainable development. Circular economy promotes closing loops in industrial systems, minimizing waste, and reducing raw material and energy inputs [Stahel, 2016]. It pertains to industry, the entire economy and, to a large extent, the packaging industry. Circular economy introduces significant changes that have a huge impact on the future of the packaging industry. It is connected with the approach to environmental issues as regards composted polymer packaging materials that can minimize the increase of packaging waste currently generated from conventional plastics. In order to address those challenges, scientists all over the world are conducting studies to develop bioplastic materials whose properties will be similar to those of plastics. This article presents the results of the latest studies related to the development of bioplastic packaging materials with the use of such polymers as PLA, starch or PHA. In the near future, the bioplastic materials discussed in the text may replace plastics and become some of the most commonly used packaging materials.

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MATERIAŁY OPAKOWANIOWE Z BIOTWORZYW W GOSPODARCE O OBIEGU ZAMKNIĘTYM

STRESZCZENIE. **Wstęp:** Europejska strategia dotycząca tworzyw sztucznych skupia się na dostosowywaniu unijnych regulacji do realizacji zasad in circular economy. Circular economy to podejście, w ramach którego w niedługim czasie nastąpią znaczące zmiany w wielu gałęziach współczesnej gospodarki. Będą one dotyczyły w dużym stopniu branży opakowań.
Metody: Duzym zaniteresowaniem cieszą się poliestry alifatyczne jakie jak polilaktyd (PLA) oraz polihydroksyalkaniany (PHA).W niniejszej pracy przedstawiono rynek biotworzyw oraz wybrane przykłady najnowszych rowiązań w zakresie materiałów opakowaniowych z biotworzyw. Przedstawione biotworzywa w niedalekiej przyszłości mają szansę stać się jednym z najbardziej poządanych materiałów opakowaniowych.

Wyniki i podsumowanie: Biotworzywa wydają się być alternatywą dla konwencjonalnych tworzyw sztucznych stosowanych do produkcji opakowań. Aby stworzyć zrównoważone środowisko i zapobiec utylizacji odpadów tworzyw sztucznych w środowisku, produkcja biotworzyw zyskała wiele uwagi ze względu na ich podatność na biodegradację.

Słowa kluczowe: biotworzywa, materiały opakowaniowe, gospodarka o obiegu zamkniętym

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