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DEGRADABLE PACKAGING MATERIALS - SOURCES, APPLICATION AND DECOMPOSITION ROUTES RAZGRADIVI AMBALAŽNI MATERIJALI – IZVORI, PRIMENA I NAČINI **RAZGRADNJE**

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ABSTRACT

There are many biodegradable and recyclable packaging materials available, alternatives for plastics: paper and cardboard; biodegradable polyethene (degradable due to additives incorporated during production, whose role is to lead to the polyethylene breakdown into CO2, H2O, biomass and minerals when in landfill) and biodegradable plastic (made from renewable biomassbiopolymers in a relatively energy-efficient process). The decomposition routes of degradable materials are reflected in the degradation for which realization a physico-chemical stimulus is required and biodegradation for which microorganisms are responsible. The global biodegradable plastic market was valued at \$1.6 billion in 2019 and it is expected to reach \$4.2 billion by 2027. The largest segment by application of biodegradable materials is in packaging with a market share of more than 60%. Some examples of degradable packaging existing on the market will be presented in the paper.

Keywords: packaging materials, degradable polymers, (bio)degradability, composting

REZIME

U današnje vreme mnogi reciklabilni i biorazgradivi materijali su dostupni, kao alternativa primeni polimernih materijala. Papir i karton se mogu ponovo koristiti, mogu se reciklirati i biorazgraditi. Biorazgradivi polieten je, takođe, biorazgradiv zbog dodatka aditiva tokom proizvodnje polietena. Dodatak aditiva dovodi do toga da se polietilen raspada na CO2, H2O, biomasu i minerale kada se nalazi na deponiji. Biorazgradiva plastika izrađena je od obnovljive biomase, odnosno može se dobiti iz nekoliko izvora: 1) polimeri ekstrahovani/izolovani direktno iz biomase; 2) proizvodnjom monomera na biološkoj osnovi i njihovom daljom polimerizacijom; 3) polimeri dobijeni direktno iz prirodnih ili genetički modifikovanih organizama. Svi dobijeni biopolimeri se sintetišu u relativno energetski efikasnom procesu, a osim toga nisu toksični za prirodno okruženje ili ljude.

Načini razgradnje razgradivih ambalažnih materijala ogledaju se u degradaciji za čiju realizaciju je potreban fizičko-hemijski stimulus i biodegradaciji za koju su odgovorni mikroorganizmi. Globalno tržište biorazgradive plastike procijenjeno je na 1,6 milijardi dolara 2019. godine, a očekuje se da će dostići 4,2 milijarde dolara do 2027. godine. Neki od ključnih faktora koji pokreću potražnju na tržištu biorazgradivih materijala uključuju nacionalne inicijative za uklanjanje plastike za jednokratnu upotrebu i propisi protiv upotrebe konvencionalnih proizvoda od plastike. Najveći segment primene biorazgradivih materijala je u oblasti ambalaže i pakovanja sa tržišnim udelom većim od 60%. U ovom radu će biti predstavljeni neki primeri razgradive ambalaže koja postoji na tržištu.

Ključne reči: ambalažni materijali, razgradivi polimeri, (bio) razgradivost, kompostiranje

INTRODUCTION

Packaging is an integral and final part of each production line and aims to enable the final product safer handling, storage and transport to the customer. Since the first plastic mass -Parkesine, a cellulose derivative was publicly presented at the Great International Exhibition in 1862 in London, the presence of plastic masses (polymers) has become irreplaceable on the world market (Šuput, 2016a). Plastics are polymers with additives commonly used in the packaging industry due to their easy molding and transformation, stability, resilience, and ease of production (Yadav et al., 2018). Synthetic polymers have long been the foundation of packaging materials. However, since they are non-biodegradable, reliance in the field of the packaging industry has led to serious ecological problems due to massive usage and long life-cycle (450 years for plastic beverage bottles, 50 years for plastic cups, etc.). Increased demand has appeared for biodegradable packaging materials, pose a very minimal threat to the environment and are manufactured from sustainable and renewable resources (Han et al., 2018). This replacement is justified by the fact that producing unsustainable (due to environmental issues) synthesized plastics consumes 65% more energy and emits 30%-80% more greenhouse gases than bioplastics (Tajeddin and Arabkhedri, 2020). The biodegradable materials market arose out of necessity but justifies its existence, and it has been continuously growing. The global biodegradable plastic market was valued at \$1.6 billion in 2019 is expected to reach \$4.2 billion by 2027, growing at a CAGR of 13.3% from 2020 to 2027 (Biodegradable Plastic Market). Some of the major manufacturers in the global biodegradable plastic market are BASF SE, Dow Inc., Novamont S.p.A., Mitsubishi Chemical Holdings, Natureworks, Corbion N.V., Eastman Chemical Company, Plantic, Danimer Scientific, Biome Technologies plc, etc. Industrial end-users are Danone (Germany), Ferrero (Italy), Lavazza (Italy), Reckitt Benckiser (Netherlands), Tetra pack group (Belgium), etc.

PARER AND CARDBOARD

Paper and cardboard are reusable, recyclable and biodegradable. Paper is a complex material made from renewable plant raw materials (plant fibers), that are obtained mechanically or chemically with the addition of various additives (adhesives, fillers, paints, etc.) that define the properties of the paper. It is formed on a sieve paper machine, by

draining the paper pulp. Advantages of paper and cardboard application as packaging materials rely on mechanical strength. biodegradability, light protection, "zero fold" properties, simple graphic processing and low cost. The main disadvantage of paper and cardboard usage is a poor barrier to moisture, gases, flavors, fats and oils. The continuous high consumption of paper-based products increases the demand for chemical additives in order to meet the needs of the (packaging) industry. Additives contribute to the advancement of many functions: surface engineering (surface strength, barrier properties, sizing, coating, enhancing the shelf-life, recyclability, etc.), mechanical strength enhancement (tear, tensile, etc.) and retention aid (for fines, filler, other additives, etc.) to paper rheology modifications. The production and specific utility of chemical massively utilized in paper-based product additives manufacturing are well known (Bajpai, 2015). The latest researches include bio-polymeric additives for a sustainable production strategy in pulp and paper manufacturing as retention aids, sizing agents, strength and coating additives (Basu et al., 2021). Apart from being eco-friendly, the most important feature of bio-polymeric additives is their versatility - one biopolymer can be used to impart diverse functions even at very low concentrations.

Over one-third of the new paper is produced with recycled fiber. Other fiber sources include whole trees and plants (onethird), as well as residue from sawmills (one-third). The paper recycling rate measured 33.5% in 1990, which was the base year against which the American Forest & Paper Association (AF&PA) began setting its recycling goals. In 2019 paper recycling rate was 66.2%. AF&PA member companies are continuing to work toward a goal to increase the U.S. paper recycling rate to more than 70% by the end of 2020. Data published by the U.S. Environmental Protection Agency (EPA) for 2017 indicates that paper and paperboard packaging accounts for three-quarters of all packaging materials recovered for recycling in the U.S. (https://www.paperrecycles.org/statistics/ paper-paperboard-recovery). Data for the year 2019 indicate that 37.8% of the paper and paperboard recycled in the U.S. went to produce containerboard (i.e., the material used for corrugated boxes); 35.7% net exports; 12.2% went to produce boxboard, which includes base stock for folding boxes like cereal or medicine boxes; 8.4% to produce tissues. Paper cannot be recycled indefinitely. With every recycling, fibers become shorter. After being processed five to seven times, the fibers become too short for the production of new paper, requiring the addition of new fibers. (https://www.thebalancesmb.com/paperrecycling-facts-figures-and-information-sources-2877868).

BIODEGRADABLE POLYETHYLENE

Among plastic packaging materials, polyethylene (PE) is the most commonly-used petroleum-based polymer in packaging applications (Emadian et al., 2017). The increasing demand for this polymer has created the need to convert it into biodegradable material in a significantly shorter time. A possible solution is the use of additives, called prodegradants, that accelerate this process. Degradable polyolefin is typically designed to oxo-degrade, followed by changes in chemical structure as a result of oxidation in air, which causes the molecules to break down into small fragments, which are finally bioassimilated (Ammala et al., 2011). While preserving the physical appearance of the polymer, this additive does not change the physical properties of polyethylene (Lazić et al., 2011). Commercially available additives are TDPA, Renatura, Reverte, AddiFlex, d2w, etc. The tendency of plastic products to undergo degradation induced by UV radiation, heat, or ozone is

increased by the addition of prodegradants to these polymers (Singh and Sharma, 2008). Transitional metal ions (iron, cobalt and manganese) are the most commonly used prodegradant additives. Iron is very effective in accelerating photodegradation, while manganese and cobalt are sensitive to thermal degradation. Metal ions are mainly introduced in traces in the form of organic complexes. In addition to metal ions, combinations of metal carboxylates and aliphatic poly hydroxy-carboxylic acids, fatty acid amides, ferrocene, metal oxides such as TiO2 and ZnO, additives having a chromophore group, benzophenone, βdiketones, some organic peroxides and hydroperoxides (Ammala et al., 2011). The other solution is to obtain bio-PE from various biological feedstocks like sugar cane, maize, wheat, lignocellulosic materials, etc. Siracusa and Blanco (2020) well explained mechanism: ethylene monomer is synthesized by dehydration of bio-ethanol. For example, sugar cane shredding and milling makes sugar cane juice products (containing 12-13% of sucrose) and the sugar cane fiber by-product (bagasse). The juice is anaerobically fermented in order to obtain ethanol, which is distilled in order to remove water, giving an azeotropic solution of hydrous ethanol, at 95.5 vol.-%, and a by-product named vinasse. The following ethylene monomer polymerization is the same that is followed when using ethylene derived from petroleum. The corresponding bio-polymer is identical in its chemical, physical, mechanical properties to fossil-based PE, also with regards to the mechanical recycling processes.

Initially, the production of bio-ethylene was not considered to be cost-competitive related to petroleum-derived ethylene. But, since 2008 the one-barrel price of sugar cane-derived ethanol has become competitive with the one-barrel price of crude oil (Siracusa and Blanco, 2020). With respect to the production of Bio-PE, the process followed to obtain bio-PP has been less explored, which explains why bio-PP has not yet been commercialized.

It is the same situation with PET which dominates as packaging material in the water and carbonated/non-carbonated beverages market. Bio-PET typically relates to a PET polymer in which only EG fraction is obtained from renewable sources. EG refers to 30% of the mass of the PET polymer and this is usually the maximum percentage of bio components encountered in bio-PET (Salvador et al., 2019). The aliphatic monomer Bio-EG could be obtained from renewable source by following steps: hydrolysis of ethylene oxide, obtained via oxidization of bio-ethylene obtained from the fermentation of glucose, followed by dehydration; or via sorbitol, based on hydrogenolysis or via the use of different types of microorganisms (Siracusa and Blanco, 2020; Salvador et al., 2019).

The increase in substitution of petrochemical plastics with bio-based plastics is estimated from 2030 onwards, thanks to the replacement of the monomers, which are chemically identical with the same equivalent functionality (Siracusa and Blanco, 2020).

BIOPOLYMERS

The new generation of the packaging materials fully consisted of biomaterials, can be divided into three main categories regarding their origin and production methods:

1. Polymers extracted/isolated directly from biomass

This category of biopolymers has been investigated extensively (Šuput et al., 2019; Popović et al., 2018) and it is most present on the market. Biopolymer film is usually produced from food-derived ingredients using a wet or dry manufacturing process and it is defined as a free-standing sheet that can be placed on or between food components (Šuput et al., 2015). They are usually classified according to the dominant building

material. The main groups of chemical compounds, which serve as sources for edible films and coatings, are polysaccharides (cellulose, chitin, pectin, starch, etc.), proteins (whey protein, casein, collagen, zein, soy protein, myofibrillar proteins of animal muscle) and lipids (free fatty acids, wax, paraffin, resin) obtained from plants, marine and domestic animals (Šuput, 2016a). These biopolymers can be used alone or as a mixture with synthetic polyesters such as polylactic acid (PLA) for material synthesis. Biopolymers have poor barrier, thermal and mechanical properties compared to their synthetic counterparts (Jabeen et al., 2015). Various chemical, enzymatic or physical treatments are available to improve properties of biopolymer materials (addition of reinforcing agents, crosslinking, heating, radiation, etc.) making them eligible for commercial applications.

The largest segment by application of biodegradable plastics is in packaging, both in terms of value as well as volume, with a market share of more than 60.3%. This is due to the fact that biodegradable plastics are being increasingly used to manufacture single-use packaging materials such as shopping bags, disposable cutlery, etc. Biopolymer significant feature is that they have a high potential to carry active ingredients: antibrowning agents, colorants, flavors, nutrients, spices, antimicrobial and antioxidant compounds that can extend product shelf-life, improve the organoleptic properties and food nutritional value (Šuput et al., 2016b), which leads to improving the quality and improving the safety of the packaged product (Šuput et al., 2019).

Popović et al. 2018 reviewed the application of numerous biopolymers obtained from biomass for various food products (fruits and vegetables, dairy products, meat industry products) packaging with special attention to quality changes (sensorial, chemical, oxidative, microbiological, etc.) during shelf-life (Popović et al., 2018). Biopolymers commonly used for industrial production of degradable and disposable products are usually starch: United Biopolymers SA (Portugal), Kompuestos (Spain), Indochine Bio Plastiques (Malaysia), Agrana Stärke (Austria), BIOTEC (Germany), cellulose: Futamura (UK), agar: Loliware, bio-resin: Pond (Denmark), biodegradable biocomposites: Sulapac (Finland), Promateris (Romania), biodegradable polyesters: Avantium (Netherland), Microtec SRL (Italy) and many others.

2. Polymers produced by classical chemical synthesis and bio-monomers

It is possible to get a large range biopolyesters by chemical synthesis. The most known is poly-lactic acid (PLA) and its copolymers, biodegradable thermoplastic linear polyester similar to polystyrene. The raw material for obtaining the lactic acid is obtained by fermentation of glucose or starch from another source (corn, wheat, or alternatively whey and molasses) (Wackett, 2008). PLA has been widely accepted as a biodegradable polymer for packaging materials due to its stiffness, transparency, processability and biocompatibility. When compared with other biopolymers, PLA exhibits better thermal processability, which allows for the various processing methods of PLA such as injection molding, blow filming, cast filming, fiber spinning, thermoforming, etc. (Rasal et al., 2010). PLA is mainly processed into thermoformed pads and containers for packing and serving food, films, transparencies and bottles and other packaging blown but also mixed with other materials (Ivanković et al., 2017). According to a recent market report on Biodegradable Plastics Market, the production of PLA is the largest segment by type with a market share of more than 45.1%. This is due to PLA's mechanical properties and ease of processability. In terms of value, starch blends are expected to account for the largest share in the market due to their comparatively high cost compared to PLA. There are many manufacturers and auxiliaries based on PLA: Zhejiang Hisun Biomaterials (China), Taghleef Industries S.p.A. (Italy), BIO-FED (Germany), Sidaplax (Belgium), BIOTEC (Germany), NatureWorks (Netherland), FKuR Kunststoff (Germany), Danimer Scientific (USA), CJ (Germany), CARBIOLICE (France), SELFECO (USA), etc.

The other biopolymers synthetized from bioderived monomers are:

Aliphatic polymers and copolymers: poly(glycolide) (PGA), poly(caprolactone) (PCL), poly(butylene succinate) (PBS), poly(butylene adipate) (PBA);

Aliphatic-aromatic copolymers:poly(trimethylene terephthalate) (PTT), poly(butylene adipateco-terephthalate) (PBAT), poly(propylene carbonate) (PPC), etc (Siracusa, 2019).

Among the biodegradable polymers, aliphatic polyesters represent good examples of biodegradable bioplastics, with proven biodegradability and characterized by good mechanical and gas barrier properties at competitive cost (Siracusa et al., 2017). Long-chain aliphatic polyesters well mimic the PE backbone, due to a large number of methylene units along the macromolecular chain. Long-chain aliphatic polyesters can be obtained by some of the listed pathways: a) the polycondensation of hydroxycarboxylic acids, b) the polycondensation of a dicarboxylic acid and a diol, or c) the ring-opening polymerization of lactones (Genovese et al., 2014). Aliphatic polyesters are odourless and biodegrade in soil and in water giving CO2 and H2O, in a period of 2 months (e.g. for a 0.04 mm thick film) (Siracusa et al., 2008).

Copolymerization represents one of the most adequate manners to tailor the properties of a material and improve its biodegradation rate, this last being generally attributed to the limited crystallinity degree of the resulting copolymers, for example by the introduction along their main chain new linkages (e.g ether and thioether linkages), whose presence has a significant effect on the final properties of the synthesized materials (Genovese et al., 2014).

3. Polymers obtained directly from natural or genetically modified organisms

Many bacteria accumulate these polymers as a source of energy and as a carbon reserve. This group includes polyhydroxyalkanoates (PHAs) - polyesters of various hydroxyalkanoates that are synthesized from microbial fermentation. PHAs are hydrophobic and insoluble in water, non-toxic and crystalline thermoplastic elastomers, whose properties depend on the PHA monomer compositions. They are biocompatible with good UV resistance, physical and chemical properties. PHAs application is constrained due to its poor mechanical properties, incompatibility with conventional thermal processing techniques, as well as their susceptibility to thermal degradation (Li et al., 2016). PHA, synthesized by microbial fermentation (Alcaligenes, Azotobacter, Bacillus, Halobacterium, Rhizobium, etc.) can be produced in large quantities biotechnologically using fermentation. Depending on the bacteria and the carbon source, the polyhydroxyalkanoate may be manufactured from rigid brittle to plastic to a rubber-like polymer. Have similar properties such as propylene and polyethylene; they are elastic and thermoplastic (Ivanković et al., 2017). Apart from renewable, biodegradable plastics can be produced from synthetic polymers by using bacteria. The bacterium Pseudomonas putida converts styrene monomer in the PHA. PHA is water-insoluble, biodegradable material and compostable whose improvement works intensively before its commercialization (Chiellini, 2008).

Another promising material applied in the packaging, medical and agriculture sector is polyhydroxybutyrate (PHB) - the most common derivative of PHA. It is obtained by bacterial fermentation of sugar or lipid. PHB is characterized by high crystallinity (up to 70%), which is the reason for its great mechanical properties (similar to polyethylene). It has been used for short-term food, cosmetics and pharmaceutical product packaging applications, as well as in agriculture (Lin et al., 2018). PHB is adequate for packaging applications due to its lamellar structure, which reflects its high barrier properties to water vapor and aroma. PHB completely degrades into water and carbon dioxide in aerobic conditions. Biodegradation in favorable conditions takes 5–6 weeks (Botana et al, 2010).

There are many manufacturers and auxiliaries for bioderived and biodegradable plastic suitable for various applications and industries (electronics, cosmetics, bio-med, aerospace, consumer, agriculture, packaging, etc) based on PHA and PHB: Kaneka Corporation (Japan), BIO-FED (Germany), CJ (Germany), Danimer Scientific (USA), EggPlant (Italy), etc.

THE MECHANISM OF POLYMER (BIO)DEGRADATION

Degradation is a major feature of biodegradable polymers and plastics. It is based on the fact that most of these materials are naturally organic or an additive for decomposition has been added to synthetic polymers, and they are subjected to physically, as well as mechanically and chemically induced degradation. The degradation process occurs due to abiotic or biotic activities, or most often as a combination of both (Lazić et al., 2011).

The degradation process consists of two phases: decomposition (depolymerization) and mineralization. The initial phase - depolymerization, is significantly associated with the deterioration of physical characteristics, such as brittleness and fragmentation. It is possible to classify environmentally degradable (bio)polymers and plastics according to the mechanism of the first phase of degradation (depolymerization) as "hydro-biodegradable" when this mechanism of the first phase takes place by hydrolytic processes, mediated or not by exoenzymes, and as "oxo-biodegradable" when it is thermally or photophysically induced oxidation, whether or not by exoenzymes (Krzan et al., 2006). The second phase is the final conversion of plastic fragments (mineralization). During degradation, the polymer is first converted to its monomers, and then these monomers are mineralized. Most polymers are too large to pass through cell membranes, so they must first be depolymerized before being absorbed and biodegraded within microbiological cells. Dominant groups of microorganisms and degradation pathways associated with polymer degradation are often determined by environmental conditions. When O2 is available, aerobic microorganisms are mainly responsible for the destruction of complex materials, up to biomass, CO2, and H2O as final products. In contrast, under anaerobic conditions, anaerobic microorganisms are responsible for the decomposition of polymers. The primary products will be biomass, CO2, CH4 and H2O under methanogenic (anaerobic) conditions (Shah et al., 2008).

Bio(polymer) characteristics, such as its mobility, crystallinity, molecular weight, type of functional groups and substituents in its structure, as well as plasticizers or additives added, play an important role in its degradation. In addition to the material characteristics, the factor that affects the course of biodegradation is the type of microorganisms in the mineralization process (Shah et al., 2008).

For all polymer and biopolymer materials, the degradation rate is increased by the addition of readily degradable components containing hydrolyzing functional groups, such as starch, polyesters, polyanhydrides, or polyamides. Another approach is the addition of active components in most commonly polyethylene, such as photo- or thermal-sensitive additives. After exposure to sunlight or heat, these additives accelerate oxygen uptake with the formation of peroxides and hydroperoxides that generate free radicals that randomly attack and break bonds leading to the formation of low molecular weight products, i. the first phase of degradation. (Krzan et al., 2006). This process is in full sense confirmed as biodegradation only when carbon compounds become food and microorganisms are transformed into water, biomass, or carbon dioxide (Barone and Arikan, 2007).

When a new biodegradable polymer is introduced into the market, two characteristics must be clarified: biodegradability and biodegradation (Siracusa, 2019). The first characteristic, examined in the laboratory by following standardized tests, is related to polymer chemical structure and the potentiality of such material to be degraded by a biological attack. The second characteristic refers to the degradation process, which could happen if certain conditions are present (temperature, pH, moisture, etc.). A polymer could be defined as biodegradable but can express very limited biodegradability if the environmental conditions are not proper. Consequently, not all biopolymers could be defined as biodegradable, which is related to their structure. Some polymers are resistant to biological degradation (their carbon linkages cannot be broken from enzymes and microorganisms) due to many reasons: the hydrophobic character limits enzyme activity together with other factors like low surface area, high molecular weight and crystallinity (Siracusa, 2019).

COMPOSTING

Biodegradable material is not necessarily compostable (can biodegrade during a time or under specific conditions), while compostable material is biodegradable. Composting is a biological process where, in controlled conditions (composting cycle), polymer degradation occurs, resulting in water, carbon dioxide and compost. Materials that are not suitable for composting can reduce the final quality of compost). The industrial composting cycle implies the application of composting temperature (can reach up to 70 °C) and humid conditions and the activity of certain microorganisms. The composting process takes place for months (Ivanković et al., 2017).

The resulting organic compost is completely environmentally neutral. The process of composting is a key segment of dealing with organic waste and return the remains of biodegradable materials to a new use (Xi et al, 2016). In Europe, the main leaders in composting are Germany and Netherlands, where for a very long time composting is carried out effectively and successfully.

Industrial composting is defined by national and international standards (eg. EN 13432, ASTM D-6900). The basic frame of rules relies on EN 13432 and EN 14995. Standard EN 13432 defines the characteristics of packaging materials that must fulfill to be labeled as compostable and acceptable for recycling organic solid waste. According to the EN 13432, a product to be defined as compostable must be biodegradable and disintegrable in a short period of time, or it must be turned from the microorganisms into water, carbonic and fertile anhydride compost. The product must result compatible with a process of composting, which means it must not release dangerous

substances and must not change the compost quality. In industrial composting bioplastics are converted into biomass, water and CO2 in about 6-12 weeks (Siracusa et al., 2008). EN 14995 extends the scope of plastic that is used for non-packaging applications. Besides industrial composting, another option is home composting, which differs since it takes place at lower temperatures in the compost heap. Materials must be specifically tested to demonstrate compostability in home conditions (Wiles and Scott, 2006).

CONCLUSION

It is an indisputable fact that polymer materials (plastics) rightly occupy a leading position in the field of packaging materials, but their main disadvantages are obtaining from renewable sources, as well as the very negative impact of the amount and accumulation of plastic waste on the environment. For this reason, there was a need for an increasing share of degradable packaging materials in the field of packaging. Degradable packaging materials are an environment-friendly substitute for synthetic polymers, due to biodegradability, agroindustrial waste (biomass) usage, and renewable raw materials. In addition, they are desirable in terms of availability and costeffectiveness. Degradation flows in two directions: as the disintegration of materials into physically smaller components and as biodegradation, where microorganisms play a decisive role. The goal is to close the cycle because the final components of biodegradation are CO2/CH4, H20 and biomass (humus in the case of composting). It is necessary to make an additional effort in terms of investing in the infrastructure needed for mass production of degradable packaging and to include government initiatives to eliminate single-use plastic and regulations against the use of conventional plastic products.

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