

## Review Article

# End-of-Life Options for (Bio)degradable Polymers in the Circular Economy

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End-of-life options for plastics include recycling and energy recovery (incineration). Taking into account the polymeric waste, recycling is the intentional action that is aimed at reducing the amount of waste deposited in landfills by industrial use of this waste to obtain raw materials and energy. The incineration of waste leads to recovery of the energy only. Recycling methods divide on mechanical (reuse of waste as a full-valuable raw material for further processing), chemical (feedstock recycling), and organic (composting and anaerobic digestion). The type of recycling is selected in terms of the polymeric material, origin of the waste, possible toxicity of the waste, and its flammability. The (bio)degradable polymers show the suitability for every recycling methods. But recycling method should be used in such a form that it is economically justified in a given case. Organic recycling in a circular economy is considered to be the most appropriate technology for the disposal of compostable waste. It is addressed for plastics capable for industrial composting such as cellulose films, starch blends, and polyesters. The biological treatment of organic waste leads also to a decrease of landfills and thereby reducing methane emissions from them. If we add to their biodegradability the absence of toxicity, we have a biotechnological product of great industrial interest. The paper presents the overview on end-of-life options useful for the (bio)degradable polymers. The principles of the circular economy and its today development were also discussed.

## 1. Introduction

Reducing waste production, increasing the use of biodegradable and/or biobased feedstock, and development of recycling methods are a challenge for a modern and environmentally friendly economy. The circular economy is a model of economy in which all products, materials, and raw materials should be used as long as possible. The idea is that the used product should not become waste but a raw material for further production. This is a departure from the current “take-produce-use-discard” system in favour of reuse model of raw materials. Thus, it is alternative to the linear economy, in which waste is often treated as the last stage of the life cycle of products. The circular economy is an economic concept in which waste generation should be minimised as much as possible. It means that circular economy promotes reducing, reusing, and recycling as alternative options of waste treatment [1]. This idea takes into account all stages of the product’s life cycle, from

design, through production, consumption, waste collection, to its disposal. In circular economy, it is important that waste, if it is generated, be treated as secondary raw materials. All the activities preceding waste generation are to serve this purpose [2]. The European Environment Agency (EEA) recommended the model of a circular economy in relation with bioeconomy and presented the challenges and benefits of transforming the traditional linear economy system into a circular economy. The main benefits of circular economy implementation should be reduction of waste and emissions of harmful pollutants. The European Strategy for Plastics in a Circular Economy leads, by committing to separate waste collection and improving extended producer responsibility systems, to increased recycling of plastics, in particular packaging recycling at 55% in 2030 [3]. The reducing of the petrochemical feedstock use and replacing them by green plastics are an important aspect of the circular economy model, especially as we are currently overexploiting nonrenewable resources.

Moreover, the circular model often defined as closed loops is directed on a zero waste economy. It is a fact that landfills of waste contribute negatively to the environment and climate especially when depositing of biodegradable waste that produces harmful gas such as methane. On the other hand, not all materials can be recycled and, in this case, landfilling is the most appropriate solution. In this context, the new segregation technologies and the modern technological solutions for sustainable landfills should be developed to enable this. However, for recyclable materials, the main challenge is to extend the life of products by reusing them or introducing them into the natural life cycle of matter where it is possible and economically profitable. This strategy includes research programs and introducing innovations aimed at increasing the use of materials such as plastics in an environmentally sustainable way. In the European Union, the most commonly used goods are wood products, textiles, and plastics. All this should lead to the increasing of the efficiency of the recycling system and the improvement of the waste management system [4]. According to the EEA, 118 to 138 million tonnes of biowaste every year in the European Union is produced, of which 100 million tonnes is food waste. Only about 25% of them are recycled, and still, the majority of biowaste goes to landfills or is incinerated. This situation is a threat to the environment and a source of infectious diseases, and leachate from landfills can contaminate surface and groundwater [5]. The European Commission has recommended the Circular Economy Package presenting a new approach to waste management through closing the loop of product life cycles along with increasing of the reuse and recycling methods [6, 7]. Simultaneously, the use of the (bio)degradable polymers and the goods made from them is a way to reduce the pollution of the environment with conventional plastics. Biobased polymers can be produced from different renewable resources, such as first-generation feedstock (sugar, sugarcane, beet, soy, cassava, rice, wheat, potato, corn, hemp), more preferred second-generation feedstock (nonfood crops such as waste from food crops, agricultural, and wood residues), and third-generation feedstock (biomass derived from algae) or methane ( $\text{CH}_4$ ) made from waste, and they are divided into nonbiodegradable or biodegradable [8, 9]. For example, biobased poly(ethylene terephthalate) (biobased PET), biobased polypropylene (biobased PP), or biobased polyethylene (biobased PE) belongs to biobased nonbiodegradable polymers. Such polymers, although produced from renewable resources, are not degradable. They have the structure of their counterparts from petrochemical sources and can therefore be recycled with conventional polymers.

Due to their origin, (bio)degradable polymers are divided into polymers obtained from petrochemical (nonrenewable) resources and from renewable resources (of biological origin) as well as natural polymers (biopolymers). The most popular (bio)degradable polymers are polyesters that are grouped into aliphatic and aliphatic-aromatic ones (Figure 1).

The aliphatic-aromatic polyesters represent poly(butylene terephthalate) (PBT) and their copolymers: poly(butylene terephthalate-*co*-adipate) (PBTA, Ecoflex® type) or poly(butylene terephthalate-*co*-succinate) (PBTS). The aliphatic polyesters include polyhydroxyalkanoates (PHA) (for example,

poly(3-hydroxybutyrate) (PHB)) and polylactide (PLA) as well as poly(butylene succinate) (PBS), poly(butylene succinate-*co*-adipate) (PBSA), and poly( $\epsilon$ -caprolactone) (PCL). The PHA and PLA are often called green polymers because they are both (bio)degradable and obtained from renewable raw materials in a sustainable way [10, 11]. Decomposition of polyesters in environment occurs by enzymatic process as a result of the action of specific microorganisms (biodegradation) or by hydrolysis of ester bonds (hydrolytic degradation), but most often both of these mechanisms occur in the appropriate sequence [12–14].

The plant fibres are an example of the natural (bio)degradable polymers. Natural polymers are obtained from different parts of plants, for example, from seeds, leaves, fruit, stems, or other grass fibres. Cotton, milkweed, kapok, hemp, flax, jute, ramie, bamboo, kenaf, nettle, sisal, abaca, manila, and coir can be distinguished here [15, 16]. The most important feature of (bio)degradable polymers is their degradation time, which is from several months to several years and is much shorter than the degradation of conventional polymers, the decomposition of which can take even hundreds of years. For this reason, the production of (bio)degradable polymers is one of the more developing sectors of the global plastics market. The currently observed dynamic development of the production of these polymers means that the application of plastic products made from them also extend. EEA notes that the market of products manufactured from (bio)degradable polymers is growing [5, 8]. However, developing the biodegradable plastic market requires caution and learning from mistakes so that, as part of the assumed improvement in the state of the environment, it is not to cause even greater damage to it or to disturb the food market. The forensic engineering of advanced polymeric materials can help to understand the relationships between the structure of the (bio)degradable polymer material used, its properties, and behaviour for practical applications [17–19].

Plastics which are commercially available belong mainly to the nondegradable group of materials, and their recycling often is not justified economically [20]. Polymers produced from renewable raw materials (those that can be replenished at the same time or less than the time needed for their consumption) are now an increasingly significant market among all polymers produced in the European Union, whereas products made of (bio)degradable polymers are increasingly used in agriculture and industry as mulch films and various types of packaging.

The overview presents the recycling methods used for the (bio)degradable plastics such as mechanical, chemical, and organic. Mechanical recycling (typically leading to regranulated products) is the processing of waste materials, resulting in a reproduction of the original product or a production of the new one. Thus, the mechanical recycling is a process in which postused products and their waste are processed into a secondary raw material. Mechanical recycling cannot be limited to only one of the activities such as segregations and sorting waste, necessary to carry out the restoration of the used product to a condition that allows its reuse or obtaining secondary raw material not used in the production of the final product or use of waste as an energy source. Those

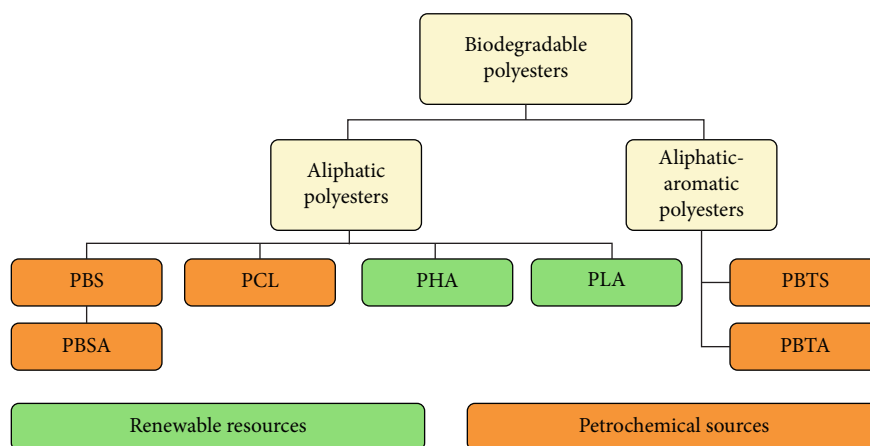


FIGURE 1: The division of (bio)degradable polyesters.

forms are not in themselves the mechanical recycling. The essence of the mechanical recycling is a reproduction of the original product or a production of the new one. In the case of biodegradable polymers, mechanical recycling is rarely used on a commercial scale, but taking into account the fact that it is (along with upcycling technologies) the most environmentally friendly way of processing plastic waste, in this overview, was described [21]. Chemical recycling (feedstock recycling) is a process that involves the depolymerisation of the material to low-molecular compounds or to the starting compounds or their derivatives. For this type of recycling, clean and homogeneous polymer waste is necessary. Organic recycling includes the aerobic degradation (composting) and wet/dry anaerobic digestion of biodegradable waste, carried out under controlled conditions and with the use of microorganisms, resulting in the production of stable biomass as well as water ( $H_2O$ ) and carbon dioxide ( $CO_2$ ) or  $CH_4$ . The disposal to landfills is not considered a form of organic recycling.

## 2. Mechanical Recycling

Mechanical recycling is one of the methods in which the basic structure of the material is not changed when processing waste into secondary raw materials [22]. This type of recycling contains many steps which can occur from zero to multiple times and in the various orders. These steps are collecting, separation, sorting, cleaning, and grinding of the waste (Figure 2).

Scheme of recycling process depends on the place from where the wastes are obtained and their composition [23]. The European Union directive concerning legal framework of mechanical recycling is included in the Packaging and Packaging Waste Directive 94/62/EC, the Waste Framework Directive 2008/98/EC, and the Landfill Directive 1999/31/EC [22, 24–26].

The information concerning the ability of “market for the recycle” and its “economic viability” belongs to the main factors influenced on the recycling sustainability. If there is problem with one of them, there is a high probability that recycling plan will fail [27]. When the recycling of selected material is economic viability and there is market for the

recycle, the recycling process itself should be considered. The type of the waste stream, contamination of feedstock, and the ability to recover affect the practicality of the processing. Starting from the waste stream process can use mainly two types of material: postconsumer and postindustrial waste. When the factories use the postconsumer waste stream, its usage history, homogeneity, and contamination should be balanced by the ecological and economic factors (e.g., reducing waste in landfills). Utilisation of the waste generated during processing (postindustrial waste), with minimised contamination and clear history, can be an attractive field of recycling scheme [22, 28, 29].

During mechanical recycling of plastics, some issues can appear. Mainly, they are caused by two types of degradation: thermal and mechanical. Mechanical shear and heating throughout processing can lead to the lowering of molar mass or cross-linking. Also, during lifetime of plastic products, some changes in the material, especially for the (bio)degradable polymers, can occur caused by the influence of the environment. The low-molar mass compounds which arise during lifetime or recycling process of plastics can compromise properties of the product as well as lead to the processing equipment corrosion. Proper preparation of the installation for recycling can minimise these issues [23].

*2.1. Influence of Mechanical Recycling on (Bio)degradable Material Properties.* Plastic industry should change the strategy whose consequence is environmental pollution, to the approach of more friendly to the environment, conducive to its protection. The (bio)degradable polymers can constitute the potential solution of the mentioned problem. Thus, such polymers are the subject of many studies, also in the area of their recycling. The researchers focused their investigation on multiple reprocessing of (bio)degradable polymers. PLA is one of the most important (bio)degradable, biocompatible, biobased materials obtained on the route of the ring-opening polymerisation of lactide. PLA technological waste can be used as a suitable material to be reused as an additive to make a new product [30, 31]. During reprocessing, the heat, oxygen, and mechanical damages can lead to the degradation of material [32]. The influence of the

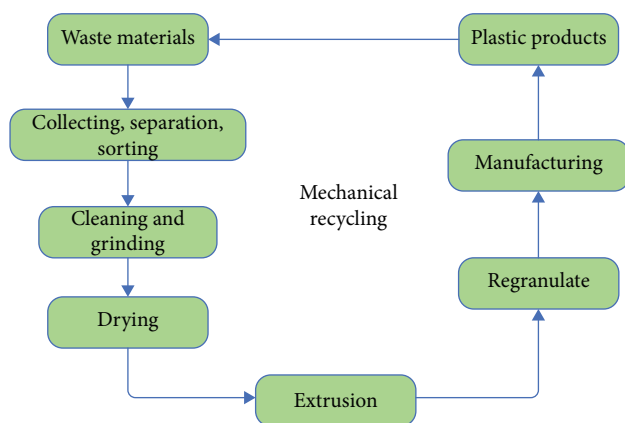


FIGURE 2: Scheme of mechanical recycling.

recycling on the mechanical properties of PLA was conducted by the multiple extrusion method. During the processing, the deterioration of mechanical properties was observed. The study indicated that mechanical properties such as the tensile strength, tensile strength at break, and melt flow index can depend on the number of the extrusion cycles (up to 10 times). However, the biggest difference was observed in the impact strength, up to 20% after 10 cycles of extrusion process. The tensile strain at tensile strength changed mainly at the beginning and not dependent on the number of extrusion cycles. The reprocessing had also the influence on the thermal properties but not in a significant way. A slight decrease in the thermal stability of PLA and a clear decrease in the temperature of the cold crystallisation were observed as the extrusion cycles increased [31]. Another approach was applied to the improving properties of reprocessing material by the addition of 50% neat PLA. Commercial PLA was processed to prepare the recycled material, taking into account aging, washing, and reprocessing of the recycled material. The thermal and hydrothermal aging steps as well as photochemical degradation had an influence of the material during this process. The decreasing of molar mass of PLA during recycling was observed; also, cold crystallisation for this material appeared at lower temperatures. The optical properties of material after the recycling steps did not change in comparison to neat PLA. Blending of neat PLA with material after mechanical recycling in a 50/50 ratio caused the improving of recycled material properties [33]. Thermal, mechanical, and rheological properties were investigated for PLA after seven recycling cycles. The recycling was simulated by the injection cycles with addition of oxidative stabilisers and residual catalyst stabilisers for achieving PLA's better stability during moulding or extrusion. Increasing of crystallisation during cooling and deterioration of mechanical properties were caused by this type of reprocessing. However, application of stabilisers limited the degradation of material [34]. The influence of poly(*L*-lactide) (PLLA) multireprocessing on the organic recycling was also investigated. The research was conducted under industrial composting conditions. The obtained results indicated that the multireprocessed PLLA degraded in these conditions with almost the same rate regardless of number of processing [20].

In recent years, reducing of manufacturing costs paves the way for research in the field of (bio)degradable packaging industry. The composites of (bio)degradable polymers with natural fillers meet the directions of action in accordance with the principle of sustainable development. Also, the mechanical recycling of composites belongs to more and more frequently discussed issues. One cycle of PLA extrusion procedure was used as simulation of recycling to prepare the matrices for composites that are rice hull-filled. Additionally, the composites with neat PLA have been subjected to reprocessing up to 2 times. The thermal behaviour which was examined after recycling showed that thermal stability of investigated materials was almost unchanged. A decrease of flexural strength and slight increase of flexural modulus were observed for recycled composites [35]. Two types of recycling which were used on PLA/clay nanocomposites caused an improvement in the clay nanoparticle dispersion in polymer matrices. This behaviour of the filler during reprocessing led to increase of optical, thermal, and gas barrier properties [36]. Composites which contain sisal as a filler belong to a very attractive alternative in the packaging industry. Not only due to the price reduction but also by the improvement of mechanical properties in comparison to neat PLA and poly(3-hydroxybutyrate-*co*-3-hydroxyvalerate) (PHBV) were matrices and sisal in the amount of 10, 20, and 30% used as fillers. Obtained composites were subjected to three recycle cycles. For both types of composites with different filler amounts, the deformation at break and tensile strength values notably lowered after first cycle. The recycling process did not significantly affect the thermal properties of all materials. Usage of different matrices had an impact on the tensile modulus, while an increase in this parameter was observed for composites with PLA, and there were only slight differences in tensile modulus when using PHBV [37].

Mechanical recycling was also investigated for poly(3-hydroxybutyrate-*co*-4-hydroxybutyrate) (P3HB4HB). During multiple injection moulding up to 10 times for the investigated material, the increasing of mass flow rate values was observed. No substantial effect was detected on mechanical properties up to 6 injection cycles. With the increasing of reprocessing cycles, the storage modulus value at 120°C increased. Changes in thermal properties indicate a decrease of thermal stability and degree of crystallinity of P3HB4HB [32]. The same procedure was used to PCL. Thermal and mechanical properties were investigated after injection moulding cycles up to 10 times. Increasing the extrusion cycles resulted in a decrease in the thermal stability of PCL. The differential scanning calorimetry (DSC) results indicated the influence of the number of processing on the temperature of crystallisation ( $T_c$ ), but other values such as enthalpy of crystallisation ( $\Delta H_c$ ), crystallinity ( $X_c$ ), and enthalpy of melting ( $\Delta H_m$ ) did not notably differ depending on the reprocessing number. Insignificant decrease in the value of impact strength and tensile strength at break was observed for the investigated material. With the number of injection moulding repetition, the melt flow rate of PCL steadily raised [38]. Mechanical recycling simulation of the most popular (bio)degradable plastics shows that reprocessed polymers (or its composites) in the form of technological waste are

adequate as an additive to a neat material. From the presented research, a conclusion can be drawn indicating additional possibilities of reducing the production costs of (bio)degradable materials by mechanical recycling at the stage of polymer production.

*2.2. Influence of (Bio)degradable Material Addition on Mechanical Recycling of Conventional Plastics.* The presence of (bio)degradable products in everyday life may cause them to unintentionally appear in the waste stream among the conventional plastics. The investigation of the influence of mixing biodegradable waste with conventional plastics gives a picture of potential changes in the recycling of pellets. Addition of two (bio)degradable materials, PLA and plasticiser-free thermoplastic corn starch (Bioplast GS), to the high-density polyethylene was used to simulate the contamination of recycled material. Samples for further analysis were prepared by the blending of PE with 10 and 20% *w/w* of (bio)degradable materials. Mechanical properties of the obtained materials decrease significantly in comparison to neat polymers. Surface analysis showed clear immiscibility and incompatibility of blend. Hence, changes and issues with replicability of mechanical property values were observed. Also, in the thermal properties, the effect of the addition of (bio)degradable polymers on the degree of crystallinity of PE was observed. The blends show a dramatic reduction in  $X_c$ . Conducted research has shown that disposal of (bio)degradable materials in the general waste stream can have a negative effect on the material properties after mechanical recycling [39]. Therefore, the segregation system plays an important role in the proper management of biodegradable polymer waste. Presence of (bio)degradable polymers in the waste stream can also affect recycled PP. Hence, the blends of PP with PLA, thermoplastic starch (TPS), and PHB were subjected to the simulated recycling. The amount of (bio)degradable polymers was applied in the blend up to 15%. Mixed materials were processed by melt extrusion followed by an injection moulding. The significant changes in the mechanical and thermal properties were observed when more than 5% *w/w* of (bio)degradable polymer was added. A way to detect the presence of (bio)degradable polymers in the PP recycling process seems to be Fourier transform infrared spectroscopy (FTIR). FTIR is a technique that allows to detect (bio)degradable polymer contamination in recycled PP. Their presence can be identified by clear signals indicating a characteristic band ( $-C=O$ ) of the PHB, PLA, and TPS. Elimination of impurities from recycled PP is however the best option to receive the material with a wide range of applications [40]. Nevertheless, there is no problem with contamination of petrochemical polymers by the biobased polymers during recycling. Mechanical recycling of biobased ones does not influence on recycled petrochemical polymer properties [41, 42]. From this point of view, production of conventional polymers from renewable resources is a good direction of development in this field.

*2.3. The (Bio)degradable Polymer Recycled Application.* The essence of the mechanical recycling is to recreate the original product or create a new one. The recycled PLA can be used as

a filament in 3D printers which lowers the high cost of the original PLA filament. A number of studies have been carried out to characterise the mechanical, rheological, and molecular properties of the recycled PLA [43]. The research has shown a decrease in the molar mass of PLA after subsequent recycling cycles with decrease in the mechanical properties of this material. In order to reduce degradation and obtain better mechanical properties of 3D printed objects fabricated from recycled PLA, polydopamine (PDA) was applied to promote adhesion. The study demonstrated that the addition of PDA improved the tensile strength of the material; therefore, PLA recycled pellets coated with PDA can be used to make filament for 3D printing. Polymer recycling should not influence the material properties' interests from the engineering point of perspective that allow the use of the recycled polymer. However, sometimes, the use of a fully recycled material might not be possible due to the loss of mechanical properties. In this case, it is possible to use a blend of neat and recycled materials [44]. PLA recycled with five consecutive extrusion cycles was used to prepare the nanocomposite with graphene nanoplatelets. The thermal, rheological, and mechanical tests, morphology analysis, and intrinsic viscosity measurements were performed. Comparative studies of the nanocomposite and the melt reprocessed neat polymer matrix suggest that recycled PLA can be used to make polymeric matrix for nanocomposite containing graphene nanoplatelets. The study demonstrated that the reprocessed nanocomposite samples revealed good particle dispersion and lower presence of aggregates. The loss of viscosity and molar mass was also observed, depending on the number of reprocessing cycles for pure PLA and its nanocomposites. However, the addition of graphene nanoplatelets slowed down the rate of PLA degradation as dependent on the amounts of recycling cycles [45]. Another ecofriendly composite was prepared from recycled PLA and kenaf with the use of a lab scale corotating twin screw extruder. It has been demonstrated that depending on the process parameters and screw configuration, it was possible to increase the mechanical strength properties of the composite, for example, an increase in tensile strength of about 65%. Moreover, it was observed that the natural filler functioned as a plasticiser. PLA is also one of the promising renewable polymers to be used in the automotive sector [46]. The next studies demonstrated that recycled polycarbonate (PC), PC/PLA blends, and recycled PC/recycled PLA blends or cellulose/polymer composites prepared with PC/PLA blends as matrix can be used in the automotive sector. The addition of cellulose fibres to the polymer blends resulted in an increase of the elastic modulus, which indicates the potential for obtaining the necessary strength of the final product, but with a smaller thickness, thus allowing for the reduction in the vehicles mass [47].

PCL is another biodegradable and biocompatible aliphatic polyester that can be used after the recycling process. This polymer is bioresorbable and nontoxic for living organisms. The mechanical properties, such as the tensile strength and the elongation at break of PCL after 8 recycling cycles, were found to be unchanged. In addition, recycled PCL can be blended with neat polymer to obtain a new material or can be used for the synthesis of biobased thermoplastic

polyurethanes. Biobased thermoplastic polyurethanes with PCL-based soft segments are used in medical applications such as orthopaedic splinting or casting and plastic. But the practical application of PCL-based thermoplastic polyurethane secondary blends for orthopaedic splints generates great amount of waste that is put on to landfill. Furthermore, due to the complex nature of polymer decomposition, some of the degradation products can be toxic. Therefore, research has been undertaken into the recycling of these polymers. The thermoplastic polyurethane secondary blends were prepared, and their mechanical properties and shape memory behaviour have been examined. The study showed that after the recycling process, the blends show significant shape memory and can be more than once used for orthopaedic splint production [48]. In another study, it was found that recycled PCL-based thermoplastic polyurethane/PCL blends can be used to make composite filled with montmorillonite. This work showed that the additive in nanocomposites slightly reduces the tensile strength and elongation at break of recycled thermoplastic polyurethane/PCL blends. Moreover, the influence of additive on hydrolytic degradation, elastic modulus, and melt flow index properties was observed [49].

Cellulose is the next material that can be used after the recycling process. For example, research has been conducted to prepare the ecofriendly aerogels using recycled cellulose fibres derived from waste paper. The aerogels were prepared by a freeze-drying method. As a matrix for aerogels, the carboxymethyl cellulose was used whereas to increase the thermal stability and fire retardancy, sodium montmorillonite and ammonium polyphosphate were added. This work demonstrated that biobased aerogels are potential course of action of replacement for petroleum-derived foams and can be potentially used as lightweight constructions, sensors or supercapacitors, and separation agents [50]. In another study, it was found that biobased cellulose aerogels from paper waste can be potentially used as isolation agent for heat insulation applications in a loaded water bottle. The study showed that water bottles obtained from the cellulose aerogel isolated are more economic and lightweight as well as can offer the better heat isolation than commercial bottles [51]. Another work demonstrated that it is possible to obtain of sustainable ecofriendly composite materials containing the recycled cellulose fibres from recovered papers and boards or other lignocellulosic materials. Composite containing recycled cellulose fibres, red peat, and other additives was used to prepare the biodegradable nutritive pots used in the production of vegetable seedlings. The study showed the same growth of plants as commercially available plastic or biodegradable pots. Moreover, the biodegradable pots containing natural raw materials can be impregnated with various components; they can be different kinds of fertilisers, plant growth regulators, fungicides, and insecticides. These substances can be released during plant growth and could increase the productivity of the manufacturing system. These studies have shown that the biodegradable pots derived from recycled polymers can be good alternative to conventional pots available on the market [52]. Furthermore, recycled cellulose can be used to make new textile fibres with superior properties. The textiles were obtained by Ioncell technology.

It is a technology that makes possible the sustainable conversion of cellulose and old textiles into new, high-quality textile fibres [53]. Moreover, Mattel Inc. sources 93% of paper and wood fibre used in its packaging and products from recycled materials. The company intends to use 100% recycled, recyclable, or biobased plastic materials in its products and packaging by 2030 [54]. Another study presents an original, environmentally safe approach to advanced recycling of natural cellulose fibres obtained from waste paper. Novel ecofriendly nanocomposites were obtained from upgraded (by increasing their alpha cellulose content and restoring their natural nanoporous structure) recycled cellulose fibres filled with kaolin in the presence of molasses. The nanocomposites exhibited high strength, and extraordinarily tremendous retention of inorganic fillers used in the manufacture of paper [55]. Due to the depletion of fossil fuels and ecological concerns with synthetic polymers, a lot of research addresses the green composites that comprise (bio)degradable polymers as matrix and biodegradable fillers, for example, cellulose fibres. The properties of the green composites made from TPS reinforced by recycled cellulose fibres were investigated. The cellulose fibres were extracted from used newspaper, and TPS was obtained from corn starch. The incorporation of recycled newspaper cellulose fibres into composites showed an effect on mechanical properties and thermal resistance as well as effect on water absorption. The studies have demonstrated that composites containing 8% *w/w* of the cellulose fibres to matrix had increased of mechanical properties and thermal resistance and showed the highest decrease of percentage water absorption than other composites. These composites can be used for the production of organic waste bags and seeding grow bags, because they are cheap and recyclable [56]. The effect of using cellulose fibres as cement replacement on the lightweight cement composite properties was also examined. The fibres were obtained after recycled process of waste paper and packaging, and their content in the composite was up to 16% by mass of cement. The obtained samples were tested after 28 days of curing. The experimental investigation showed decrease in the compressive strength with the increase of fibre content, but the thermal insulation properties of concrete were improved. This study demonstrated that it is possible to use this material for the construction of nonload-bearing walls, partitions, roofs, and ceilings [57]. The cardboard boxes waste can be recycled to manufacture paper and cardboard. The high-quality cellulose fibres received from recycling can be used for the production of paper towels, writing papers, and tissue papers. Waste cartons containing aluminium were used to prepare boards with urea-formaldehyde resin or poly(vinyl acetate)-based glue. The boards could be located behind radiators or electrical radiators for preclude heat loss [58]. In another study, compressive strength and microstructure of the composites obtained from recycled papers, cartons boards, Tetra Pak, and gypsum were evaluated. The results demonstrated that this composite materials can be used to prepare building materials. The influence of nanocellulose fibres and nanoclay particles on the mechanical and physical properties of biodegradable composites obtained from recycled thermoplastic starch and sawdust

was also examined. Moreover, effect of working temperature was investigated, which is an essential parameter affecting the mechanical properties of these composites and limiting their use in various applications. The study showed that depending on the type and amount of nanoparticles, the mechanical and physical performance of biocomposites changed, which can be used to tailor the desired properties [59].

### 3. Chemical Recycling

Due to the rapid depletion of natural resources, waste valorisation as an end-of-life route is necessary to obtain cost-effective and sustainable waste management options, renewable energy production, and also production of high-value chemicals in the circular economy [60]. There are many methods for recycling of plastic solid waste. Chemical recycling is considered to be an important way of reducing waste and greenhouse gas emissions as well as promoting circular economy [61]. It is an interesting end-of-life option for materials that cannot be mechanically recycled [62]. The success of this solution as an alternative approach to the processing of solid plastic waste depends on the affordability of processes and the efficiency of catalysts [63]. The main approaches to chemical recycling include depolymerisation (glycolysis, hydrolysis, solvolysis, or acidolysis), partial oxidation, and cracking (thermal, catalytic, and hydrocracking). Chemical recycling technologies with the highest technology readiness level (TRL) such as pyrolysis, catalytic cracking, or conventional gasification as well as mild solution-based catalytic depolymerisation can bring economic and environmental benefits [61, 63]. However, chemolysis is only suitable for processing of homogenous plastic waste. A separation of polymeric materials is therefore needed. However, when separating polymers such as PLA and PET, visual discrimination based on appearance is not possible because both materials are transparent and very similar. Additional labelling is therefore needed. PLA is potentially recyclable, but there is no separate waste stream for this polymer yet. When recycling other materials, the proportion of PLA mixed with them should be limited so that it does not contaminate them due to the lower transition temperature, causing agglomeration and sticking of PLA. A greater share of PLA in postconsumer waste would encourage the creation of separate PLA recycling streams as recycling would become economically feasible [64]. Chemical recycling called tertiary recycling is a process by which polymers are broken down into single monomers and then transformed into new polymers to produce a high-quality product. Chemical recycling of (bio)degradable polymers involves the recovery of monomer and/or low-molar mass depolymerisation products and includes thermal and chemical processes (Figure 3) [65].

Chemical depolymerisation is possible with monostreams of waste. It applies not only to conventional polymers but also to (bio)degradable polymers such as PLA or PHAs as well as polycarbonates. Depolymerisation is mostly used for polymers formed in the process of polycondensation and occurs with the participation of heat and catalysts. It is usually better not to lead to the monomer stage (total depo-

lymerisation), but to oligomers (partial depolymerisation). In most cases, the reactant that allows the bonds to break is a solvent (hydrolysis, alcoholysis, or aminolysis). Supercritical fluids, enzymes, reduction reactions, or metathesis are also studied [66]. Dry-heat depolymerisation in the melt is used to break down aliphatic polyesters such as PLA and poly(glycolic acid) (PGA) at a temperature above melting temperature and leads to a cyclic dimer. Dry-heat depolymerisation usually causes racemisation. Hydrothermal depolymerisation involves the hydrolysis of aliphatic polyesters with steam or hot water under high pressure and leads to hydroxy acids. The reaction can be carried out under subcritical and supercritical conditions, with or without oxidants [65]. Chemical recycling processes that break down PLA into high-purity lactic acid or lactide, in which monomers may then be repolymerised in closed loop into PLA, can be called a cradle-to-cradle processes [64]. Chemical recycling of PHAs through thermal degradation using alkali earth metal catalysts leads to vinyl monomers. PHBV depolymerisation leads to crotonic and 2-pentenoic acids at relatively low degradation temperatures and in the presence of CaO and Mg(OH)<sub>2</sub> as catalysts. The resulting crotonic acid was copolymerised with acrylic acid to obtain water-soluble and high glass transition temperature copolymers, poly(crotonic acid-co-acrylic acid). Copolymerisation of crotonic acid derived from PHA pyrolysis is an example of cascade utilisation of PHAs [67]. The enzymatic transformation of PHA into oligomers was also used as the recycling method [68]. The chemical recycling of commercial PLA products, including mixed wastes, blended materials of various polymers, and plastic retrieved from anaerobic digesters, also in the presence of PET or PP includes high-temperature hydrolysis to lactic acid and solvolysis [62]. In the case of (bio)degradable polymers, mechanical and chemical recycling has not yet been implemented for large-scale postconsumer recycling. In contrast, thermochemical processes with energy recovery are already widely used not only for conventional plastic but also for biobased and (bio)degradable plastic [69]. Mixing of different (bio)degradable polymers such as PLA, PHBV, other PHAs, starch, or natural fibres can complicate recycling processes. In terms of energy balance and cost-effectiveness, these processes stand between pure remelting and combustion [64].

Higher homogeneity of industrial waste contributes to higher recycling rates for postindustrial plastic waste compared to household waste. Thermal processes have higher tolerance to mixed and contaminated plastic waste streams [61]. Most often, plastic waste is used in blast furnaces, where they replace coke, coal, or natural gas to act as a reducing agent for conversion of iron ore and other oxidised metals into pure metals [70]. Solid municipal waste generated by households, commerce, offices, and public institutions as well as agricultural waste and waste from the food industry is a potential source of renewable energy. The need to increase the share of renewable raw materials, while at the same time reducing greenhouse gas emission as well as raising environmental awareness to protect the environment from pollution and unsustainable practices such as landfilling, will contribute to the development of the waste-to-energy idea. Waste-

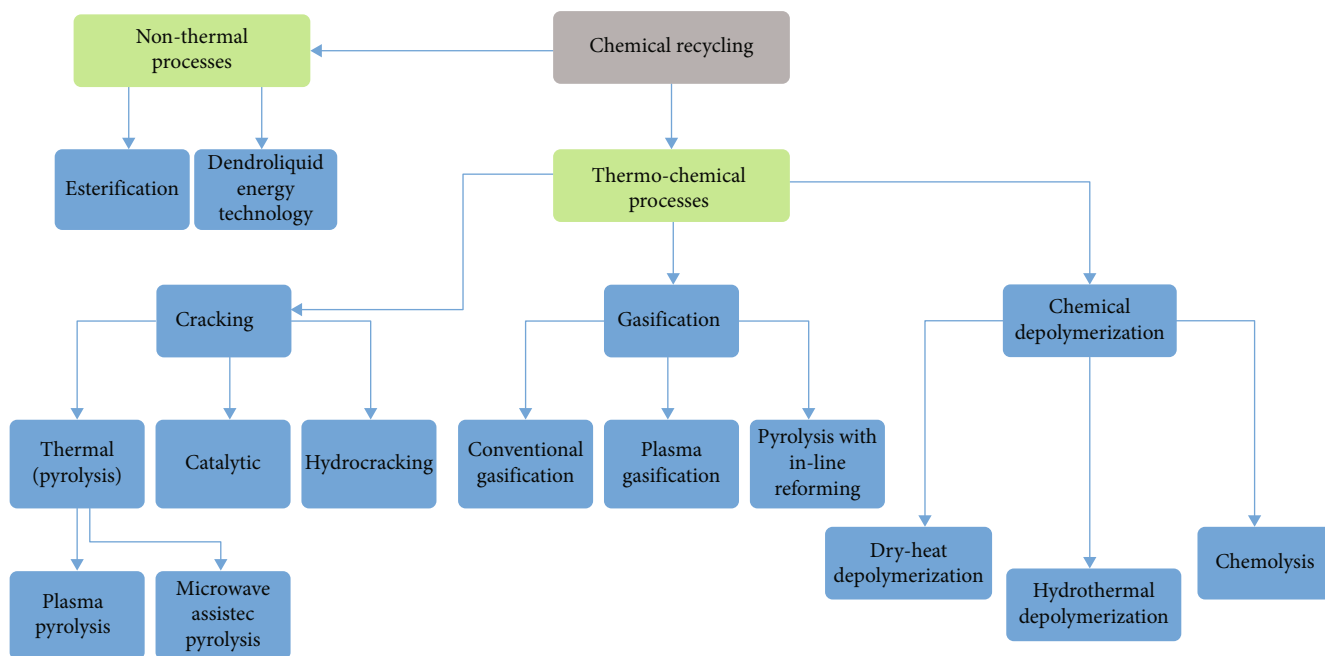


FIGURE 3: Chemical recycling technologies.

to-energy technologies are nonthermal, thermal, and thermochemical processes that generate energy from the conversion of waste into electricity, heat, biofuels, or synthetic fuels. Thermal waste-to-energy technologies produce electricity directly by combustion or by producing a combustible fuel commodity such as  $\text{CH}_4$ , methanol, ethanol, hydrogen gas ( $\text{H}_2$ ), or synthetic fuels. Modern combustion, pyrolysis, torrefaction, and gasification (plasma arc technology) are thermal processes to break down waste that use high temperatures and reduced oxygen compared to traditional direct incineration [71–73].

Plastic waste deposited in landfills is mainly composite packages or mixtures with noncombustible wastes such as glass, metals, and ceramics and can be degraded by liquefaction. After hydrothermal treatment, including a steam-explosion process, the separation of mixed waste into organic and inorganic substances becomes simpler. However, the effect of hydrothermal pretreatment on the subsequent liquefaction of organic substances and determination of optimal liquefaction conditions for organic substances from mixed waste subjected to hydrothermal treatment is not obvious [74, 75].

The difference between combustion, pyrolysis, and gasification of solid municipal waste, including polymers and biomass, depends on the process conditions and in particular the amount of oxygen (usually in the form of air) supplied to the thermal reactor and the temperature of the process. Municipal solid waste contains large amounts of cellulose, hemicellulose, and lignin as well as biobased plastic [76]. If the biobased waste is treated as a fraction of residual waste, it can be combusted with energy recovery as a high calorific value material or separated as a reducing agent for use in blast furnaces. Combustion temperature is around  $1000^\circ\text{C}$ .

Gasification, the process of converting hydrocarbon materials in the substoichiometric presence of air (limited

air), leads to carbon monoxide ( $\text{CO}$ ),  $\text{CO}_2$ ,  $\text{H}_2\text{O}$ ,  $\text{H}_2$ , and a mixture of impurities at high temperature, usually  $800^\circ\text{C}$  [69]. Synthetic gases can be used as a substrate for the production of PHA. The syngas conversion technology with gases obtained from the gasification of corn seeds leads during fermentation using *Rhodospirillum rubrum* bacteria to short-chain length PHA and to  $\text{H}_2$  as an additional product. The PHA obtained consists of approximately 90% 3-hydroxybutyrate units [77]. Gasification of lignocellulosic biomass leads to the production of various liquid fuels, primarily through the synthesis of Fischer-Tropsch, in which the syngas is transformed into usable liquid fuels as a result of the water-gas shift reaction combined with hydrogenation of  $\text{CO}$  or the synthesis of mixed alcohols produced from syngas. Gasification can be carried out by the biomass preprocessing, direct biomass gasification, syngas purification, and reforming as well as gas utilisation [78].

Pyrolysis is thermal cracking in an inert atmosphere at a temperature from  $300^\circ\text{C}$  to  $3000^\circ\text{C}$  depending on the technology used. The pyrolysis of waste from polymeric materials includes anaerobic controlled thermal decomposition and breaking the C-C bonds of macromolecules into molecules with lower molar mass, resulting in products, such as  $\text{H}_2$ , hydrocarbons, coke, and others. Nanocatalysed pyrolysis is a recommended solution for polymers with low thermal conductivity, because it promotes faster reactions at lower temperatures. It also indicates lower energy consumption which increases the selectivity of the process, so that products with higher added value are generated with increased efficiency [79]. The main advantage of nanomaterials in energy production is the increase of the process efficiency due to their large surface area per unit volume, which results in higher surface activity. Fly ash and nuclear waste management, hydrothermal carbonisation, pyrolysis, and high-



energy ball milling are examples of technologies using nano-technology to better waste management [80].

Nonthermal processes including mechanical and some chemical methods (esterification) as well as biochemical technologies can generate more electricity from the same amount of waste than would be possible with direct combustion and are able to efficiently convert waste into liquid or gaseous fuels. The relatively new dendroliquid energy technology, involving bioprocessing of mixed wastes, is close to the idea of "zero waste." In this case, all types of organic waste, including plastics and wood, are processed in the reactor by oxidizing bacteria or enzymes to CO and H<sub>2</sub>, which are clean fuels for electricity generation. Inert postprocess residues in the form of sand, gravel, etc. represent 4 to 8% and are used as aggregate or landfilling. The technology works on the fuel cell principle in small, decentralised, and low-cost units. In the microbiological chambers of fuel cells, proton exchange membranes separate the anode and cathode chambers. The first chamber is maintained anaerobically, and the second is immersed in aerobic solutions or exposed to air. The external circuit regulates the flow of electrons from the anode to the cathode [81]. It is four times more efficient than other waste-to-energy technologies in terms of near-zero emissions in the power generation process compared to anaerobic digestion, with almost no emissions due to no incineration needed and no on-site effluent problems. Both wet and dry waste can be processed at moderate temperatures of 150-250°C depending on the type of input materials, with high energy conversion efficiency of about 80%, and the resultant syngas is free of tar and solid particles. Therefore, this process of transforming waste into energy is a low-cost process. For paper, plastic, textile, and wood with a low moisture content, dendroliquid energy technology is an alternative to organic recycling [71, 82].

#### 4. Organic Recycling

The organic recycling is seen as the most suitable disposal technology of the organic waste from (bio)degradable polymers. The key factors responsible for the growing interest in the (bio)degradable polymer market include too much growth of municipal waste landfills, dependence on gaseous and fossil fuels, the need to stop greenhouse gas emissions, and introduction of the legal regulations regarding certification and commercialisation of new (bio)degradable polymers, as well as growing consumer interest in the sustainable development issues. In addition, a significant number of composting plants in Western European countries also contribute to the increase in the market importance of polymeric packaging that are subjected to organic recycling, whereas in Central Europe, despite the existence of a strong and specialised research base in the field of (bio)degradable polymers, research and development works on new solutions, and their application in practice, they do not progress at a pace commensurate with their scientific potential and production capabilities. The works are being made to develop the production of environmentally friendly polymers based on new "clean technologies." Thus, (bio)degradable polymers can be considered "polymeric materials of future." The use

of the products made of (bio)degradable polymers in many applications may provide a solution to the environmental problems. The (bio)degradable materials are adequate for biological waste treatment, especially through industrial and/or home composting, and obtained compost can be utilised as the soil fertilizer [64]. Organic recycling can be realised as composting, i.e., biological transformation of the biowastes under aerobic conditions into CO<sub>2</sub>, H<sub>2</sub>O, and biomass (organic matter) or anaerobic digestion of the organic fraction of wastes in the presence of microorganisms, with the biogas production [83].

**4.1. Composting.** Composting is a controlled degradation of organic wastes under aerobic conditions, which requires an appropriate chemical composition of the starting waste with an optimal carbon, nitrogen, and oxygen content. In the composted material, a proper pH (optimal pH = 6.5-7.5) and most favourable humidity in the range of 40-50% should also be kept. Selectively collected organic wastes suitable for composting include biowaste from households; plant waste from parks, lawns, and home gardens; plants waste from agricultural production as well as dehydrated sewage sludge. Microorganisms that break down organic waste use carbon as an energy source, and the nitrogen are built into the cell structure with simultaneous energy releasing. Almost all energy is released as heat, which can raise the temperature of the compost to 60-70°C [84]. The industrial composting leads to the utilisation of municipal waste suitable for this type of process with production of a sufficiently stabilised product for introduction into soil or storage, which is environmentally friendly and safe for human health and life. The benefits of organic recycling as well as the use of compost itself can be considered on many levels. Waste from compostable plastics is not directed to landfills, which protect environment and reduce the emission of pollutants, and in the same way, the idea of circular economy *via* recycling of valuable organic components is realised. Such proceedings may also significantly reduce the amount of solid municipal waste disposed in landfills [85]. The increasing amount of collected and separately recycled biowaste contributes to climate protection, because uncontrolled decomposition of organic material in landfill or under home composting conditions generates CH<sub>4</sub> which, according to United States Environmental Protection Agency data, is approximately 25 times more harmful to the climate than CO<sub>2</sub> [64]. If all organic waste will be collected more and more, the greenhouse gas emission from waste storage would be significantly decreased. In the European Union, requirements on industrial compostability of plastic packaging were introduced by an EN 13432:2000 harmonised standard [86, 87]. EN 14995:2006 is also approved by the European Committee for Standardization (CEN) and contains similar requirements on industrial compostability of nonpackaging plastics. These standards provide the basis for assessing the suitability of plastic items for recovery by organic recycling (industrial composting) [88]. The disintegration of plastic items should be assessed; the chemical composition, content of volatile compounds, and heavy metals as well as other environmentally hazardous components should be specified. The content

of hazardous ingredients in the plastic items or the possibility of their occurrence during biodegradation disqualifies the packaging as compostable. Plastics and plastic items are considered compostable if they meet the requirements of the norms: EN 13432:2000 (for plastic packaging only) and ISO 14855-1:2012 norm (for other plastics); it means that the tested material should reach at least 90% of decomposition within lasting a maximum 6-month test under industrial composting conditions. During 3 months of the composting process, it is required that no more than 10% of the dry mass of the tested material remain on the 2 mm sieve. The thickness of the material plays also an important role in meeting the requirements of EN 13432:2000. The 3-month test duration is concerned with a maximum thickness specified for the test. Under anaerobic conditions, during 2 months of testing, the degree of degradation (determined on the basis of separated biogas) should be 50% [86]. The ISO 14855-1:2012 norm describes a method of determination of plastic biodegradability under controlled composting conditions in which the composting of organic fraction of solid mixed municipal waste occurs, measured as the amount of released CO<sub>2</sub>. The samples are incubated for a period not exceeding 6 months at a constant temperature of 58°C. Controlled composting conditions are maintained throughout the test, including oxygen, moisture content, temperature, and pH [89]. Recently, the indication of the degree of aerobic biodegradability of water-soluble plastic materials, containing formulation additives, used the ISO 14852:2018 norm. The norm looks at the biodegradation under standardised laboratory conditions in activated sludge, mature compost, or soil under aerobic, mesophilic conditions. The product is placed in an aqueous medium and analysed for the amount of carbon evolved to CO<sub>2</sub>. The test duration cannot exceed 6 months [90]. It should also be kept in mind that the accordance with the EN 13432:2000 standard does not mean that these biodegradable products could be composted under home composting [91]. Products intended for collection as biowaste must be easily recognised by the special logo as biodegradable and compostable and collected together with organic waste. For this reason, many countries have introduced certification systems. Certified products should possess a special logo "Seedling" informing users that they are subjected to collection together with organic waste and use for composting (Figure 4) [92].

In Europe, TÜV Austria, AIBVINÇOTTE Belgium, COMPOSTABILE-CIC Italy, and DIN CERTCO Germany (Deutsches Institut für Normung) carry out certification on the largest scale. The certification of products useful for composting with a registered trademark of European Bioplastics e.V. "Seedling" is only given by DIN CERTCO. The other certifying bodies use different labels, e.g., OK compost through the AIBVINÇOTTE [93]. Certified biodegradable packaging bearing a special logo can be separated from the municipal waste stream along with organic waste generated in households and directed to organic recycling without removing the remains of their content [94–96]. The agreement with the European Union regulation system of the waste collection in many European countries is based on using the special containers that ensure efficient and contin-

uous waste segregation. The biowaste (green waste and organic kitchen waste) arising in households should be collected in brown containers. The paper is collected in the blue containers; metals and plastics are collected in the yellow containers; glass go to the green one; and mixed wastes are collected in the black containers. The mixed waste means the waste that cannot be placed in any of the mentioned above containers for segregated waste [97].

The basic composting systems are the classic composting windrows, from which most of the composting systems used today such as pile composting, row composting, tunnel composting, or drum reactors are derived. The various technological solutions of those systems were caused by the need to intensify the composting process and better homogenisation of composted waste, but in all systems, the process is based on the organic recycling mechanism and the final products are the same [96]. The static and dynamic composting systems can be distinguished. Static systems include open-air pile composting, layered composting, and composting in containers. Two methods of the waste aeration can be used in static composting systems [97]. The first one concerns composting in open-air piles and consists in natural aeration and oxygen spreading in the waste pile. In the second one, mechanically forced aeration is used, where the transport of oxygen to the interior of the piles is assisted by forcing or sucking air or as in the case of layered composting, the aeration is realised by loosening. In static open-air pile, the composting process is carried out on concrete ground. Each plate is equipped with holes through which the biomass on it is aerated, without mixing. The compost pile consists of wet organic matter (green waste nitrogen-rich) such as leaves, wood chips, grass, fruit and vegetable waste, and coffee grounds as well as brown matter (carbon-rich) that included dry leaves, wood chips, branches, newspapers, and cardboard [98, 99]. As the airflow decreases, the temperature in the pile increases, causing the chimney effect, and self-aeration of the pile occurs without mechanical mixing. In addition, the air flow in the pile and throughout the pile is constantly regulated by the activity of microorganisms. As the temperature rises, the microbial populations in the composting pile change from mesophiles to thermophiles. From pile formation, it is not sprinkled with water, so there is no seepage of excess water into the ground, and the moisture contained in the pile itself is "pulled" up and absorbed by microorganisms. Industrial composting in a static open-air pile differs from other composting methods in that the waste is only mixed together once. Perfect composting takes place at a temperature of 45–60°C, while the minimum oxygen level needed to initiate the composting process should be not less than 5%. When the active composting phase is finish, the oxygen level increases to around 21%, while the correct proportion of carbon to nitrogen, expressed as the C:N ratio, determines the compost maturity. The optimum C:N ratio value in the composted matter should be within the range of no more than 30:1 at the beginning of the process to 15:1–20:1 in mature compost. If the C:N ratio is too high, the reactions in organic matter slow down because of too little nitrogen, while the N excess may lead to the formation of ammonia toxic to microorganisms, which in turn leads to the



FIGURE 4: The international logo printed on the compostable products certified according to EN 13432/14995 standards, licensed by DIN CERTCO (®registered trademark of European Bioplastics e.V.) [85].

emission of upsetting odours and inhibition of organic matter decomposition processes. The composting process in static open-air pile is ongoing about 3-4 months [13]. The dynamic systems contain composting in towers and drum reactors as well quasidynamic systems as turned windrow, row composting, and tunnel composting. In tower composting system, the wastes are put on the top of the tower and moved from up to down in counter current fair. In the drum composting plants, the basic, multifunctional device of the system is a biostabiliser. Precomposting of such wastes as cattle manure, municipal biosolids, brewery sludge, chicken manure, and food residues causes their fragmentation and ensures high homogeneity of the composting products thanks to the rotational movements of the biostabiliser drum [100]. In quasidynamic systems, aeration can be realised by shifting from place to place of the wastes (turned windrow). The composting process in turned windrow compared to the static open-air composting pile allows the better aeration of the pile, reduces the risk of odour formation, also shortens composting time, and increases the porosity of the material, which makes the compost received more fragmented and homogeneous. The quasidynamic row and tunnel composting are a special form of composting in piles. In row composting, the waste is piled into windrows separated by walls, and in the tunnel composting, the rows are additionally covered with a roof. The composting process, in such cases, can be controlled by appropriate aeration, irrigation, and turning. PEABODY tower composting plants, MUTHERHOF and HORSTMANN-KNEER container composting plants, and SUTCO-BIOFIX or BIODEGMA tunnel composting plants operate in a closed system. The closed MUT-HERHOF system is based on stationary concrete-metal bioreactors, each of which constitutes a separate technological unit [101]. The closed HORSTMANN-KNEER system consists of working containers, a blower station container, a biofilter container, and a control unit. In the container, KNEER system can also compost organic kitchen waste, manure from horse race tracks, and food processing wastes. During 21 days, the precomposting process is completed and the biomass is stabilised. During precomposting in biomass, the pathogenic bacteria die off and the odours emitted are significantly lower. At this stage of the composting process, the working containers are emptied and the bio-

mass is transported to the technological yard where it matures. Composting in containers leads to a full-valuable compost, free from impurities. An example of a closed system with aeration can be the BIODEGMA tunnel system where composting process takes place in roofed modules made of reinforced concrete. The system processes mixed municipal waste, solid waste, sewage sludge, and organic waste and allows adding to the compost mass the biodegradable fraction separated in the process of segregation from municipal waste [102]. This is a relatively new solution in the field of industrial composting. The solid municipal waste generally contains organic C, carbonates, and other C forms, which may include “organically complexed” metals as well humic acids [103, 104]. The last phase of the composting process is maturation, which starts when the active composting ends both in static and dynamic systems. The compost is stabilised on flat technological yards during a 3-4-month storage period.

The ability to utilise waste generated from (bio)degradable polymeric materials in the organic recycling process is a unique opportunity to adapt the life cycle of this type of polymeric materials to the natural life cycle of matter. This life cycle cannot be achieved for conventional plastics. Thus, in the case of the compostable product, each of its components should be biodegradable, and materials combining conventional and (bio)degradable polymers should not arise [105]. The compostable products should also be made with biodegradable additives. In the literature, the numerous publications on the degradation behaviour of polymers in different environments such maturing compost (after the active composting stage), industrial compost, compost with the addition of activated sludge, and in home-type composters can be found [106–118]. For example, the studies of the whole and complete package of PLA Biota bottles conducted under real composting condition in a special preparing piles show that (bio)degradation degree of the tested PLA bottles is about 85% after 58 days of process [106], while for the Novamont plastic, Mater-Bi degraded only in 7.1% within a period of 72 days of composting [114]. The studies aiming at generating reliable information on the aerobic biodegradation of the biodegradable polyesters under industrial composting conditions were also performed. Comparative studies of (bio)degradation in the composting pile, the KNEER container system, and the BIODEGMA system of PLA and

PLA/85% (*R,S*)-PHB (where (*R,S*)-PHB is a synthetic analogue of natural PHB) rigid film samples have shown that in all tested environments, the biodegradation process of these materials occurs with successive decrease of the average molar masses of the tested films and changes in their surface morphology. The fastest biodegradation takes place in the environment with the highest humidity—in a container. In addition, the further research has shown that rigid films and final products made of them by thermoforming are characterised by a similar dynamic of biodegradation under industrial composting conditions which may indicate that thermoforming has no effect on the degradation rate [12, 108]. The negligible impact of the processing was also observed during the (bio)degradation test of multiprocessed PLA [20]. The study of composting process in the BIODEGMA system of the ecovio® cosmetic packages with longer time application (ecovio®—commercial blend of PBAT with PLA, BASF Company) points a certain influence of the cosmetics on occurring process. It has been found that the presence of the cosmetic (ionic surfactant) on the surface of the sample tested causes local changes in the form of pits and cracks [109]. The composites made of P3HB4HB with wood flour were also subject of industrial (BIODEGMA system) and laboratory composting process. It was concluded that both hydrolytic degradation and enzymatic degradation occur in the compost. Moreover, the production of such (bio)-degradable materials could be a way for utilisation of waste from the wood industry fit with the idea of a circular economy [112]. Recently, the degradation tests of PLA prototype packaging in industrial compost (in a composting pile and in a BIODEGMA system) and, for comparison, under laboratory, controlled conditions, using respirometric method, were conducted. Empty containers made by 3D printing from commercial PLA filament (PLA/12% PHA) and filled with cosmetic ingredients were used. The test results showed that the PLA/PHA container contaminated with paraffin in laboratory compost degrades faster than a clean container, and its disintegration was observed after 12 weeks of incubation [116]. The controlled composting tests were also performed for PHA samples. It was found that their biodegradation depended on the molecular structure of the samples studied in order of degradation from P3HB4HB to PHB *via* PHBV with decreasing HV units' content, and the process was catalysed by enzymes [117]. The influence of the plant origin compounds as functional, natural additives on polymer composite degradation profile was also studied. It was concluded that polyester materials containing phytochemicals with antimicrobial properties are still biodegrade; however, they are slower compared with the polymeric matrix [118].

The assessment of the degree of biodegradation under laboratory conditions is most often performed by determining the amount of produced CO<sub>2</sub>, which is evaluated experimentally, e.g., by means of titration analysis or gas chromatography. Biodegradation tests in conditions simulating the intensive process of oxygen composting are also performed automatically with the use of various types of respirometers. The differences between them concern both the construction and operation of the respirometer itself and the conditions of the experiment. The amount of released

CO<sub>2</sub> obtained in the test is used to determine the degree of biodegradation of the tested materials, which is calculated as the ratio of the total amount of CO<sub>2</sub> released in the test to the theoretical amount CO<sub>2</sub> obtained after complete biodegradation of the material [106, 110].

*4.2. The Wet/Dry Anaerobic Digestion.* The anaerobic fermentation process takes place in separate, closed fermentation chambers—bioreactors in the temperature range of the so-called mesophiles 33–35°C. The type of anaerobic digestion depends on the amount of water contained in the waste, and therefore, two processes can be distinguished: wet and dry. The term of the wet fermentation is used for the fermentation in which the substrate has a suspension form, with the dry matter content in up to 15% that allows it to pump. The waste with a dry matter content from 15 to 40% is processed by the dry fermentation. Above 40% of waste content, there are phenomena of inhibition of biological processes resulting from water shortage. The wet process requires minimal contamination, so presorting of the substrates is needed. By comparison, the dry process is much less demanding and can be used to treat organic waste products, even contaminated with other solids such as sand or fibres. Anaerobic digestion is used in biogas plants to produce methane, which accounts for approximately 70% of the resulting biogas [119, 120]. The solid fraction from the solid-liquid separation of digestate, containing about 20% of dry matter, including 40–86% of organic dry matter, containing highly available nitrogen and phosphorus, can be processed into compost or used as an organic fertilizer for plant nutrition [121, 122]. For food waste, as a preliminary stage, sometimes the thermal hydrolysis is used. The thermal hydrolysis significantly accelerates the anaerobic fermentation process and increases the amount and efficiency of the biogas production [123, 124]. One of the innovative, patented technologies of dry anaerobic fermentation at a solid content of 25 to 40% is the DRANCO (Dry Anaerobic Composting) process developed by Organic Waste Systems (Belgium company) and used by Dalkia Wastenergy (France company). The uniqueness of this technology is the use of a vertical installation, which allows a very high dry matter content of the waste charge and continuous gravitational movement of the charge, which in turn changes into the efficiency of the entire recovery process [125]. Dalkia Wastenergy installations reach a minimum of 180 N·m<sup>3</sup> of biogas per tonne of waste input and are not very emergent (all sensitive technology elements are outside the reactor) and do not generate odours or significant amounts of technological wastewater. An example of the effective use of the DRANCO system is the plant in Chagny (France), which is responsible both for waste sorting and for anaerobic digestion and composting. The annual processing capacity of this investment is as much as 73000 tonnes of mixed waste and 8000 tonnes of green waste, and the end result is around 5 million m<sup>3</sup> of biogas, then processed into methane. Another investment is the plant in Bourg-en-Bresse (France), capable of converting 66000 tonnes of mixed waste and 7500 tonnes of green waste annually into compost and electricity [126]. Despite many activities, the waste of compostable polymers

is not properly segregated and much of it is disposed of in landfills. For this reason, the PLA samples were tested in the digesters filled with pretreated municipal solid waste fraction and it was concluded that any (bio)degradation of PLA in a landfill starts from abiotic hydrolysis of material [127]. Moreover, in the composting process of nonsegregated waste stream containing also the nonbiodegradable materials, the contaminated compost with low stability could be obtained. And compost, not devoid of pathogens or heavy metals, can pose a threat to crops as fertilizer. It should also be noted that in the composting process, the greater part of the energy is the waste heat, whereas in anaerobic fermentation, the energy is transformed into biogas able for further using [119, 128].

## 5. Conclusions and Future Perspective

The circular economy is a concept aimed at rational use of resources and limiting the negative impact of manufactured products on the environment that should remain in the market as long as possible, and waste generation should be minimised as much as possible. The natural polymers and polymers from renewable resources such as plant fibres, starch, cellulose, PLA, and PHA, as well biodegradable aliphatic-aromatic polyesters, fit well with the concept of the circular economy and seem to be a good alternative to conventional plastics. The use of (bio)degradable polymers results in the reduction of environmental pollution and gives opportunity to close the life cycle of products obtained from them. The goods from (bio)degradable polymers can be mechanically and chemically recycled and organically recycled (composted or anaerobic fermented) and also can be used for recovery of energy. For the development of the (bio)degradable polymer market, it is important to conduct dynamic research and development activities aimed at cheaper and simpler production methods, mainly such (bio)degradable polymers as PLA, PHA, and PBTA, together with development of modern recycling methods of products made from them. The development of new technologies related to the production of (bio)degradable polymers can significantly affect the competitiveness of European enterprises or processing companies in various industries, including chemical. Also, the consumer demand and technological progress in the area of environmentally friendly polymers result in new strategies for replacement of petrochemical feedstock by biobased feedstock. The new polymers should not only be biodegradable but also pose specific properties properly for various range of applications. The key to the success of the development of the circular economy is to understand both the advantages and limitations of using (bio)degradable polymers. Their limitation depends from the type of polymer and mainly concerns mechanical properties, that is why many (bio)degradable plastics are used in the form of blends or composites. Also, the biodegradability which is their most important advantage may be a problem in mechanical recycling for mixed stream of wastes in which biodegradable and conventional items are together. Thus, the waste sorting systems must be taken into consideration when placing (bio)degradable products on the market. This approach is

consistent with the concept of the forensic engineering of advanced polymeric materials and also complies with the strategy of the Circular Economy Package recommended by the European Commission. The European market for (bio)degradable products is dominated by compostable plastic bags, which are primarily used for shopping or biowaste collection. The smaller market for the ecomaterial application is the construction and transport (including ground and aviation). Sectors such as electronics and the production of household goods also require materials with excellent properties, susceptible to organic recycling and therefore environmentally friendly. PLA finds application in the production of beverage bottles, rigid containers, bags, food containers, disposable cups, coatings, garbage bags, and packaging foams. Due to its easy access and relatively low price, starch is also good candidates for the production of biodegradable food packaging. Polyhydroxyalkanoates such as PHB and its copolymers: PHBV, poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) (PHBH), poly(3-hydroxybutyrate-co-3-hydroxyoctanoate) (PHBO), poly(3-hydroxybutyrate-co-3-hydroxyoctadecanoate) (PHBOD), and P3HB4HB, can be used in the food industry as the bottles, disposable cups, laminating films, and packaging for fast food products. Thus, aliphatic polyesters such as PLA and PHA are prospective biobased plastics for agricultural, medical, pharmaceutical, and packaging applications. Although PLA exhibits a high tensile strength modulus, UV and fat resistance, and the ability to process by conventional methods, there are cases where PLA-based products do not meet the packaging material requirements, especially for long-life application. In the production in these cases, the packaging from aliphatic-aromatic copolyesters is one of the ways to solve this problem. Aliphatic-aromatic copolyesters combine the good functionality of aromatic polyesters with the biodegradability of aliphatic polyesters and can be also used in the form of blends with other polyesters an example of which is *ecovio*<sup>®</sup>. *Ecovio*<sup>®</sup> (BASF product) is a certified compostable polymer, partly obtained from biobased raw materials. *Ecovio*<sup>®</sup> is used for production of the organic trash bags as well as shopping bags that can be used twice: first to pack purchases and then to throw away organic waste. Thus, *ecovio*<sup>®</sup> is a (bio)degradable material that enables the closing of the product life cycle. The increasing demand for ecofriendly products as well as shift in consumer preference is driving the global market of green materials. However, (bio)degradable polymers from renewable resources are still under development and commercialisation and their widespread use is still insufficient. Nevertheless, the goods from them fit in the circular economy concept, enabling the transition from the linear economy to the circular one.

## Conflicts of Interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

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