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Biodegradable Polymers: Present Opportunities and Challenges in Providing a Microplastic-Free Environment

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The stability of polymers against environmental factors, chemicals, microorganisms, and hydrolysis has challenged society with the accumulation of plastic waste and its management worldwide. Large amounts of plastic litter accumulate in the environment and disintegrate into microplastics (small pieces less than 5 mm in size), a topic of real concern especially for products and applications where the plastics are used for a short time before becoming waste, and where they are difficult to recover after use and remain in the environment. Whether biodegradable polymers can be one of the solutions to the problem of plastic waste is a question very often raised in this context. Although the use of biodegradable polymers appears to be highly promising based on recent and past studies, several aspects need to be considered further regarding environmental sustainability, acceptability, and degradability in the complex natural environment. Intensive efforts need to be invested in developing new environmentally biodegradable polymers and smart mechanisms of degradation after use in the environment. The present viewpoint article discusses the present scenario of the environmental acceptability of biodegradable polymers and the opportunities and challenges they offer regarding solving the problem of microplastics and their impact on the environment.

1. Introduction

Hermann Staudinger was awarded the Nobel Prize for Chemistry in 1953 for his pioneering research on macromolecules. We thank the "father of polymer science" for his great achievements that will never be forgotten. His work provided the basis for understanding and designing applications of polymers in everyday life as commodities and special functional polymers. Modern life is unimaginable without polymers. Despite all the benefits, plastics today are being severely discussed as materials responsible for harmful effects on the environment, plastic pollution, especially the plastic less than 5 mm in size (also called microplastics [MPs]) that remains in the environment.[1] In fact, the topic regarding the creation, collection, and undesirable effects of plastics on the natural environment and living systems has attracted a lot of attention from all sectors (scientists, industry, the public, policymakers) over recent years. However, the severity of MP pollution has different emphasis in different parts of the globe, but the concerns are similar.[2]

Most of the plastics detected as MPs in the environment are polyolefins (polyethylene (PE) and polypropylene) and polyethylene terephthalate (PET).[3] These high molar mass polymers with a very stable carbon–carbon or C-heteroatom backbone persist in the environment for a very long time spanning several tens and hundreds of years. Therefore, biodegradable polymers are very often discussed as one of the solutions to the present plastic pollution issues by substituting MPs creating polyolefins in general or for a few specific applications of polymers intended for use in the natural environment. Nevertheless, an obvious question arises: Would using biodegradable polymers create the opportunity of having an MP-free environment and would that represent a more sustainable overall situation? Although the use of biodegradable polymers appears to be highly promising based on recent and past studies, several aspects need to be considered about environmental sustainability, structure–property relationships, and biodegradation in the complex natural environment.[4] Intensive efforts need to be invested in developing new environmentally biodegradable and smart polymers with hidden triggers to initiate biodegradation in the environment. The present viewpoint article discusses the present scenario of environmental acceptability of biodegradable polymers and the opportunities and challenges they offer to solve the problem of MPs and their impact on the environment.

2. Biodegradation and Biodegradable Polymers

It is important initially to recapitulate some of the basics of biodegradation in order to understand the role of biodegradable polymers in the context of the issue of MPs, the present situation, and future directives. Degradation of macromolecular chains by the action of microorganisms is called biodegradation. On a molecular level, it is mainly a two-step process that can take place anywhere, for example, in soil, water, or human

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beings.[5] The first step is a fragmentation step, in which a high molar mass macromolecular chain is broken down to oligomers having polar functional chain ends and monomers with the loss of specific polymer properties, such as molar mass and strength. This step can take place due to hydrolysis (with or without enzymatic catalysis), oxidation, or any other means, depending on the chemical structure of the polymer backbone and the environment in which the polymer is being disposed and/or used. In the second step, oligomers with polar chain ends and monomers are mineralized by microorganisms forming ultimately carbon dioxide (CO_2), methane, water, and biomass (**Figure 1**). The product varies depending on the availability of oxygen. Several review articles are available describing in details the process of biodegradation.[6a–6c]

Aliphatic polyesters with easily hydrolysable ester units in the backbone are one of the classified biodegradable polymers.[6] By contrast, aromatic polyesters (e.g., PET) require very harsh conditions for hydrolysis (normally sulfuric acid at 150 °C) and are not classified as biodegradable. Aliphatic-aromatic polyesters with a limited number of aromatic units in the backbone are also classified as biodegradable polymers.[7] One of the examples of the latter is a copolymer of terephthalic acid, butanediol, and adipic acid (poly(butylene adipate-co-butylene terephthalate)PBAT). Some of the examples from the category of aliphatic biodegradable polyesters known in literature are semi-crystalline polycaprolactone (PCL), polylactide (PLA), polyglycolide, their copolymers, and special polyesters produced by bacteria: polyhydroxyalkanoates. These polyesters have been known for several decades and, up to now, have been mainly researched and used as biomaterials for different applications.[8] The most common uses, among others, are absorbable sutures, bone screws and plates, stents, carriers for drugs, and scaffolds for tissue engineering. The degradation behavior, toxicity of polymer and degradation products, mechanism of degradation, and kinetics of degradation under physiological conditions of the aliphatic polyesters listed above are well-documented for their acceptance as biomaterials. In fact, the term biodegradation has been mainly used to date in the context of biomaterials and biomedical applications.

Since the process of biodegradation is affected not only by the polymer properties but also depends upon the environmental factors, such as the availability of oxygen and light, pH, temperature, humidity, microorganism, and enzyme type and enzyme concentration, the same polymer shows different rates of degradation under different environments, such as water, soil, and physiological conditions. Therefore, the biodegradability of classified biodegradable polymers, which is shown under physiological conditions, cannot be taken as a direct measure of their biodegradability under environmental conditions. Furthermore, they might also be biodegradable under environmental conditions, but the rate of biodegradation and degradation profile might vary. Also, it is important in the case where the biodegradability of a polymer is proven under natural environmental conditions that the polymer should biodegrade completely in a short time so that it does not persist in the environment. This is of major significance when such polymers are considered instead of conventional, nonbiodegradable plastics as a solution to tackle the plastic pollution problem in an uncontrolled natural environment.

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3. Environmental Degradability of Classified Biodegradable Polymers

One of the commercially available classified biodegradable polymers that are greatly used as a biomaterial is poly(*L*-lactide) (PLA); this showed slow degradation in soil under Mediterranean field conditions.[9] The polymer fragmentation was observed for about 11 months. After this time, the fragmentation was still very little, dependent, of course, upon the thickness of the film. A similar trend regarding the biodegradability of PLA in soil and in artificial seawater and freshwater was seen in laboratory tests.[10] Further, it is not possible to distinguish clearly in these studies between fragmentation and biodegradation. The biodegradation should be evidenced by the conversion of organic carbon to CO_2/methane and biomass. Respirometry methods are normally used to record the quantitative formation of CO_2.[11] The CO_2 released is proportional to the percentage of biodegraded substrate. Furthermore, polymers labeled with the stable carbon isotope (^{13}C) should be used for the unambiguous proof of conversion of polymer carbon to CO_2. This

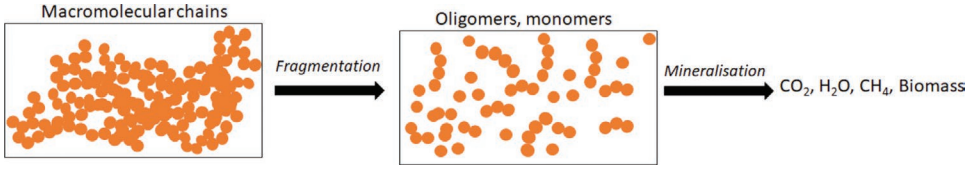

Figure 1. Two-step process of biodegradation—first step is fragmentation and the second step is the mineralization by microorganisms.

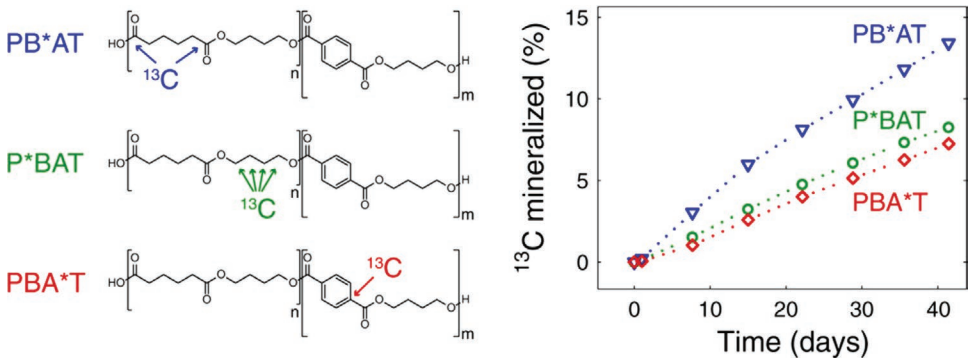

Figure 2. The $^{13}CO_2$ production results of soil incubation of ^{13}C-labeled PBAT (labeled at different carbons from each unit). The ^{13}C-labeled C-positions are shown in chemical structures (left). Evolved carbon dioxide was monitored by ^{13}C isotope-specific cavity ring-down spectroscopy as shown in the figure on the right. The results show the mineralization of each unit of PBAT. Reproduced with permission under the terms of the CC-BY-NC 4.0 License.[12] Copyright 2018, the Authors. Published by the American Association for the Advancement of Science.

will provide a clear distinction between the polymer-derived CO_2 and the one formed by mineralization of organic matter in the soil. This is the most ideal test for quantifying and proving the biodegradability of a polymer, but in reality, the use of ^{13}C-labeled polymers is not always possible due to the non-availability of the starting ^{13}C-labeled monomers for synthesis of the corresponding polymer and very high costs (5 g L-lactide ^{13}C-labeled monomer might cost up to 60 000 Euro). Laboratory experiments with ^{13}C-labeled aliphatic-aromatic polyester PBAT showed 10% carbon mineralization in about 6 weeks in agricultural soil with biodegradation of each repeat unit from the agricultural center at Limburgerhof (Rhineland-Palatinate, Germany) (**Figure 2**).[12] Although the experiments were not followed till complete degradation, the results regarding the biodegradability of PBAT are highly encouraging with a perspective to replacing polyolefins with such polymers for applications in soil. Promising biodegradation results in agricultural soil were also obtained for PBAT blends with PLA and starch. Mater-Bi CF04P (starch and PBAT blend: BioBag, Askim, Norway) and Bioflex F2110, made of PLA/PBAT in a 30:70 ratio (FKuR, Willich, Germany), films showed significant biodegradation in soil (taken from a vineyard located in Southern France near Carcassonne) over 2 years.[13]

In addition to laboratory tests, actual field experiments are also required before a new environmentally biodegradable polymer comes onto the market. The respirometry tests for quantitative estimation of biodegradation are carried out under controlled laboratory conditions using special devices and gas detectors but are not easy to design for field experiments. Auras and co-workers carried out soil-exposure and soil-burial tests in soil at a depth of 0.3 m for PBAT mulch films (PBAT with carbon black) for pineapple production at the EARTH University in Guácimo, Limón, Costa Rica, for 40 weeks from April 2008 to January 2009. The soil-exposed mulch films accumulated total solar radiation of 800 MJ m^{-2} and started losing physical integrity from the eighth week, suggesting a useful degradation time of 8 weeks of such mulch films, whereas samples buried in soil showed higher stability till about 24 weeks. Random chain scission was proved in samples both exposed and buried in soil in comparison to polyolefin, showing promise as biodegradable mulch film.[14] There are no indications of the biodegradation of PBAT in aquatic systems over a period of at least 1 year.[10] Currently, the biodegradation under actual environmental conditions is only tested by following the visual change in the form, mass, structure, and properties of the polymer under test. The additional environmental factors, such as sunlight, wind, wind speed, and humidity, can have a significant influence on the rate and mechanism of polymer biodegradation under natural environmental conditions as photodegradation, photooxidation, and cross-linking become significant in the natural environment. The environmental factors and the additional degradation mechanisms coming into play can have either positive or negative influence on the complete biodegradation process that will be dependent upon the type of the additional mechanism, polymer molecular structure, and the specific environment. Both theoretical and experimental correlation based on the laboratory experiments predicting biodegradation under environmental conditions beforehand are impossible.

4. Precise Classification of Environmentally Degradable Polymers

It is becoming clearer that biodegradation does not mean that a specific polymer will degrade at the same speed in each environment. Moreover, environmental differences are observed, for example, in different types of soils and water bodies with different pH, organic and inorganic rest, moisture rest in soil, and types of microorganisms. Aliphatic-aromatic PBAT and blends have shown promise as a biodegradable polymer in agricultural soil. This gives strong hopes for the expansion of an actual field of applications and degradation environments of biodegradable polymers with positive advantages over polyolefins and PET in terms of disposal and plastic waste management. Therefore, there is a need now to clearly subdivide biodegradable polymers into further categories with precise definitions for each category and to start emphasizing the time of complete biodegradation. The use of the general term biodegradable polymers might be misleading when used in the context of utilizing that polymer in different environments as a solution to MPs and the

plastic pollution issue. Therefore, it would be better to designate precisely, for example, soil biodegradability/biodegradable polymers and aquatic biodegradability/biodegradable polymers in which the degradation environment is a part of the name. This will make things a little easier and, at least, differentiate the environment of the biodegradation.

Current academic literature calls a polymer soil biodegradable if there is any proof of assimilation of its organic C to CO_2 in soil, even though the time of complete assimilation is not known.[12] This is a common notion. If the time of complete assimilation is very long (persisting in the environment for several years), then a question mark can be added to the utility of such biodegradable polymers in solving the issue of MPs. Therefore, the polymer should be ideally degradable in a defined time either in all types of soils or in water bodies so that a clear classification, that is, soil biodegradable or water biodegradable, can be assigned to it. Due to the complexity of our environment (different soil types and water bodies), a still existing challenge would be the unambiguous general classification of polymers into soil biodegradable and water biodegradable types. According to this argument, there is no polymer currently that can bear the tag of soil biodegradable or water biodegradable. This is an existing opportunity and synthetic challenge for polymer chemists. With the knowledge of chemistry and polymer science in hand and dedicated efforts, such polymers are sure to be developed in the future. Therefore, the only option at present is to narrow down the classification of biodegradable polymers according to the specific application(s) and the environment in which biodegradation is occurring, making sure that the time of degradation is defined precisely. At the present time, biodegradable polymers replacing PE mulch films for agricultural applications showing complete biodegradation in 6 months to 2 years under specified conditions according to European norm EN 17033 (biodegradable mulch films for use in agriculture and horticulture—requirements and test methods) and tested according to ISO 17556 (determination of the ultimate aerobic biodegradability of plastic materials in soil by measuring the oxygen demand in a respirometer or the amount of CO_2 evolved) and those passing ecotoxicity tests get an OK biodegradable soil certificate. The companies selling such polymers advertise them under the name "soil biodegradable polymer." There are questions raised about the biodegradability of such certified polymers in different types of soils. It is obvious that the same polymer might show different biodegradation behavior in different soil samples due to the presence of different types of, for example, microorganisms and pH. Therefore, why not narrow down the certification to the type of the soil and the specific application to avoid any confusion. The biodegradable polymer films tested in agricultural soil for biodegradation specifically for intended application as mulch film, for example, can be labeled as agricultural soil biodegradable—mulch film and not be given the general appellation as a soil biodegradable polymer. Application-oriented classification of biodegradable polymers in combination with the environment of biodegradation might solve several current misunderstanding issues.

Pure biodegradable polymer without additives is never used for any application. Additives fulfill different purposes depending upon the application, such as improving the mechanical properties, thermal stability, or gas barrier properties. Additives can also influence the mechanism and time of biodegradation by influencing, for example, the crystallinity and hydrophilicity of the base polymer. Therefore, a complete product with additives should pass the criteria of environmental degradability and gets the certification, which also strengthens the argument of classifying biodegradable polymers specifying the precise environment and application.

Defining the time of biodegradation is an important aspect in this classification. A polymer can be biodegradable as proved by the assimilation of organic C to CO_2 in an environment but might degrade very slowly and persist in the environment for several years. Such polymers although biodegradable will not be of any use as a solution to MPs and should not be given the classification of being biodegradable for any environmental application. The duration for complete biodegradation of mulch films specified in ISO17556 is currently a maximum of 2 years, but mulch films with complete biodegradation in a time matching the crop cycle would be the most beneficial. Otherwise, there will still be an accumulation of plastic fragments, and the migration of fragments from agricultural soil to land soil or water bodies cannot be eliminated. Tuning the biodegradability of biodegradable polymer films in soil for use as mulch films should be possible with chemistry tools and the existing know-how.

It should be very clear that a single polymer with a definite architecture and chemical composition might not be useful for all environmental applications, not even for all applications in one environment. The tuning of degradation kinetics using, for example, copolymerization, blending, polymer architecture, or additives, is required for making polymers suitable for specific applications. This is nothing new but is the same strategy as that applied for the use of biodegradable polymers as biomaterials for biomedical applications. In one of the studies, the blend of semi-crystalline and amorphous PCL showed enhanced fragmentation in a short duration in compost (**Figure 3**).[15]

5. Biodegradability as an End-of-Life Sustainable Option

It should be also obvious that the environment is not a dumping ground for plastic waste, and no environmentally biodegradable polymer will be developed with the aim of throwing and dumping into the environment after use. However, the property of biodegradability can be beneficially used for managing plastic waste originating especially from specific sectors such as food packaging as a managed end-of-life cycle option in controlled industrial compost (i-compost) plants. Packaging is one of the biggest sectors both using plastics and generating waste. Lot of plastic food packaging based on polyolefins and PET lands up in composting plants when food items are discarded together with packaging.

New i-compostable biodegradable polymers with fast biodegradation and suitable mechanical and gas-barrier properties as a substitute for polyolefins used presently would be a promising solution for MPs generated from packaging. This will also be advantageous for disposable items used for only

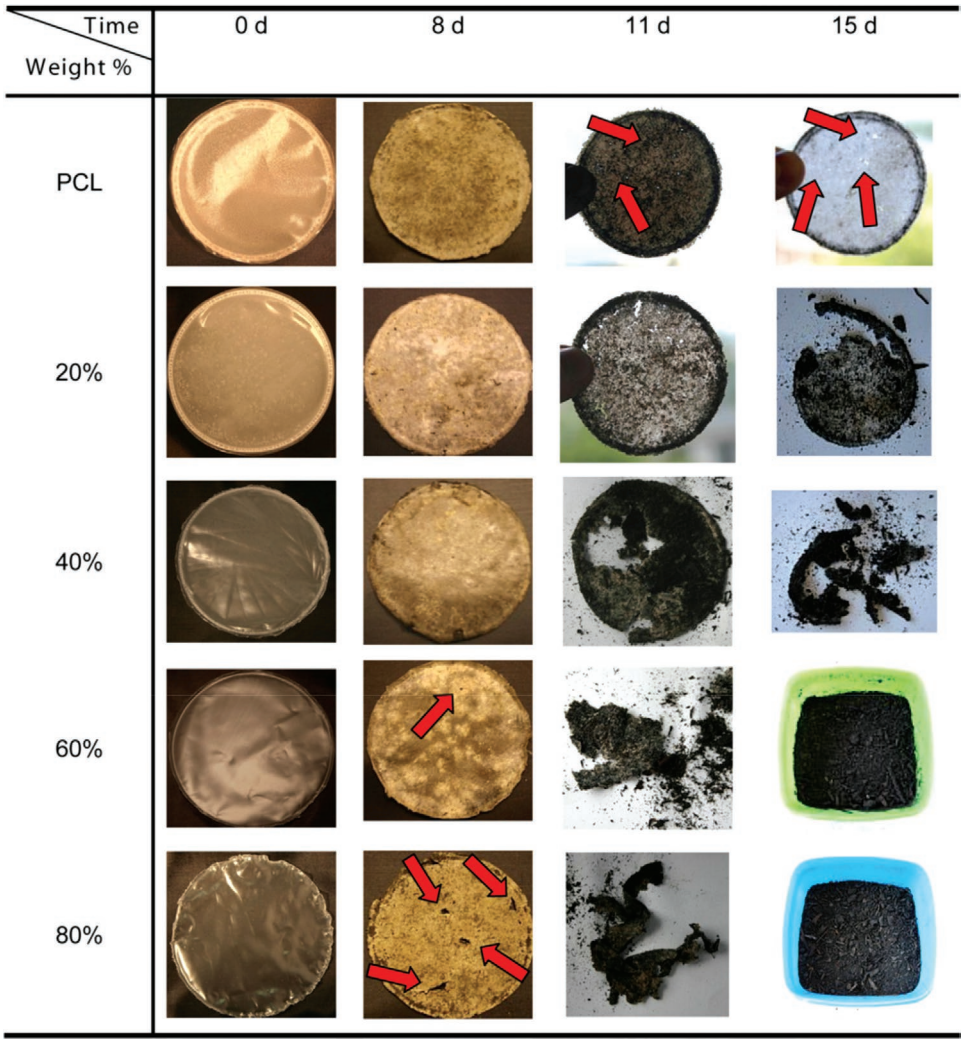

Figure 3. Fragmentation of PCL in industrial compost: effect of crystallinity. X%: % of amorphous PCL in a blend.[15] There is a fast fragmentation on blending amorphous and semi-crystalline PCL. Reproduced with permission.[15] Copyright 2010, Elsevier Ltd .

a short period of time. This is in accord with the E.U. strategy for circular economy utilizing organic recycling of such polymers in compost plants. At present, ASTM D-6400 (Standard Specification for Labeling of Plastics Designed to be Aerobically Composted in Municipal or Industrial Facilities) and the test procedure described in ASTM 5338 is used for testing the composting capability of plastics. According to the method, the sample is labeled positive (compostable in industrial facilities) in the case it fulfills the combined requirements: 1) after 12 weeks, not more than 10% of fragments more than 2 mm in size should be left; and 2) 90% of the organic carbon must be converted to CO_2 in 180 days (24 weeks). The problem is that real composting times in industrial compost plants are much shorter than the procedure allows. Most of these plants already distribute compost after 8–12 weeks. In some other compost plants, the time of composting is still shorter. This implies that the plastic with slow biodegradation in compost (biodegradation in compost not completed till the compost is ready in plants say in 8–12 weeks but which might be completed in 24 weeks) will also have the label of i-compostable biodegradation but will certainly be a source of MP as it will contain plastic fragments at the time of distribution. This is the present situation regarding several of the classified biodegradable polymers, one of which is PLA that is compostable but takes several weeks. The PBAT showed >90% conversion to CO_2 after 80 days in mature compost at about 58 °C in a laboratory test in a batch process.[16] The biodegradation profile of PBAT in controlled compost is highly attractive, with a caution that the compost in which PBAT was mineralized should not go onto the fields after only 8 weeks. If compost is distributed after 12 weeks, then PBAT is one of the promising compostable polymers available today. Similar to those mentioned above, real field tests are always required to confirm the biodegradability and degradability profile. In general, in case the time of biodegradation of plastics in compost plants matches the duration of a compost cycle in such facilities, replacing polyolefins and PET in packaging and disposable items with i-compostable biodegradable polymers would be one of the

significant steps forward. Therefore, there is a need to invest more research efforts in making i-compostable biodegradable polymers that work fast.

6. Final Remarks

Properly tested and precisely classified biodegradable polymers with complete biodegradation in a definite time matching to the application are surely an opportunity for stepping toward an MP-reduced environment. They can be especially of significant use in some specific scenarios. The first case is replacing nondegradable polyolefins with the appropriate biodegradable polymers for specific time-bound environmental applications, such as in agricultural fields as mulch film, slow release fertilizers, pesticides and water carriers, plantation pots and bags. The non-soil biodegradable polymers used for these applications stay in the environment and should be considered as intentional creation of MPs in the environment. Agricultural fields are gigantic, and the number of plastic products used for various agricultural applications are huge. It is not easy to recover plastics after use from such a huge area. Replacing polyolefins with agricultural soil-degradable, biodegradable polymers would eliminate the need to collect plastic fragments after use, and the plastic will not persist in the environment. There is a need to develop a series of agricultural soil-degradable biodegradable polymers with different physical properties and biodegradation profiles so that large number of applications can be covered.

Furthermore, biodegradability in combination with managed, closed-loop disposal systems, such as composting and anaerobic digestion, is highly promising regarding environmentally responsible solutions to MP problem when used for specific applications. In this way, biodegradable waste is recycled into useful products and not dumped in landfills. There is a need to use proper i-compostable polymers, as discussed above. Presently, industrial composting plants are very often seen coping with plastic bags, packaging films, and bottle caps, because they come with the biowaste, and the fragments from the plastic articles are even seen in the finished compost. Whosoever uses this compost either in agricultural fields, or public or private gardens bring MPs unintentionally into the environment. The use of i-compostable polymers with fast biodegradability matching the speed of the composting cycle would be a very big step to stop the leakage of MP through compost into the environment. Additionally, this has the advantage of organic recycling of the plastic, in which the plastic C is being converted to CO_2 and biomass and not landing in landfills.

Regarding short-term use items, the best scenario, like any other plastic, would also be to reuse and recycle even if they are made up of biodegradable polymer articles. Biodegradability can offer additional advantage in case there is an unintentional leak into the environment: such polymers will not persist unlike polyolefins and PET if suitable environmentally biodegradable polymers are used. For this intention, a polymer biodegrading in different natural environments (soils and water bodies) is required, which is still not available. Also, there is a serious concern about complications for existing plastic recycling systems in case biodegradable polymers enter the plastic waste stream. One should be encouraged with the beneficial degradation property of biodegradable polymers in case of their leak to the environment and should weigh the benefits against arising concerns and problems. In future, for short-term use items, environmentally degradable polymers with either mechanical recyclability matching with that of conventional plastics or advanced measures for collection and sorting of plastic waste are needed.

Further, the biodegradability property will generally only be beneficial in the case where the polymer and its degradation products do not exert any ecotoxicological effects at any stage. Up to now, there has been no study claiming any toxic effects from classified biodegradable polymers and this has been encouraging the use of such materials.

Few polymers have already shown very promising results regarding biodegradability in soil and industrial compost both in laboratory and field tests, motivating further research in this field. There is a need to put concentrated efforts into making new environmentally degradable biodegradable polymers with a fast degradation rate, if possible, degradability characteristics unaffected by the type of the environment—a dream that should come true in the future. More polymers also need to be developed showing fast and complete biodegradation in water bodies. In addition, polymers and new triggers are required making biodegradation in landfills possible.

It is also important to understand at the end that what is important is the environmental acceptability of the polymer for an MP-free environment—any mechanism of fragmentation is acceptable. There can be a combination of different mechanisms leading to the first step of the biodegradation in the natural environment accelerating the overall degradation process.

Finally, the more and more specific products balancing the physicochemical properties required during use and the biodegradability (after use) with complete mineralization in a defined period in a specific environment need to be researched, developed, and provided without any ecotoxicological effects during their entire life cycle. The use of appropriate biodegradable polymers alone can never solve the issue of plastic waste. The plastic pollution, waste management, and MP problem need to be tackled from different sides—use of biodegradable polymers in specific sectors is one of them. By replacing polyolefins and PET packaging, especially food packaging with the appropriate i-compostable polymer packaging that are both mechanical and organic recyclable; use of only soil-degradable plastic products for agricultural applications; and efficient sludge-degradable/waste water-removable polymers in cosmetics, laundry, and related applications in combination with general measures, such as reuse, recovery, and recycling of the plastic articles wherever possible, and anti-littering campaigns, we can strongly hope to have a MP-reduced environment.

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Conflict of Interest

The author declares no conflict of interest.

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biodegradable polymers, compostability, environmental acceptability, microplastics, mineralization

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