

REVIEW

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Biodegradation of polyethylene: a brief review

Sunil Ghatge^{1†}, Youri Yang^{1†}, Jae-Hyung Ahn^{2*} and Hor-Gil Hur^{1*}

Abstract

Plastic waste management and recycling became a serious global issue as it affects living beings from all the ecosystems. Researchers investigated biodegradation of polyethylene (PE) by measuring changes in various physico-chemical and structural characteristics using techniques like as fourier transform infrared spectroscopy (FTIR), scanning electron microscope (SEM), etc. However, these evidences are not enough to prove the exact biodegradation of PE. In this review, we summarized microbial biodegradation of polyethylene and discussed recent developments for the candidate microbial enzymes and their possible roles in PE degradation. In addition, we conversed the advanced technologies correctly used for measuring PE degradation using isotope-labeled PE to figure out its metabolism into the end products like as ¹³CO₂.

Keywords: Polyethylene, Biodegradation, Pre-treatment, Moth worms

Introduction

Researches on various synthetic polymers have been accelerated since DuPont had succeeded in mass production and an exclusive sale of nylon in 1940. Versatile properties of plastics, such as diverse application, convenience, non-degradability, and low price, have led to the replacement of natural materials and common use as household items since 1960. Worldwide the annual production of non-degradable plastic ranges from 350 million to 400 million tons out of that yearly, 5 to 13 million tons of waste plastic are released into the ocean, which is negatively affecting the ecological environment [47, 80]. In fact, a survey on the Great Pacific Garbage Patch has estimated 80,000 tons of waste plastics have found in the Pacific latitudes, including Hawaii, with 54% of the waste coming from North America and Asia [58]. Polyethylene and polypropylene represent about 92% of the synthetic

plastics produced, and they are used for production of plastic bags, disposable containers, bottles, packaging materials, etc. [16]. An estimated more than 500 billion to 1 trillion plastic bags used globally disturb the ecosystem and ultimately result into serious environmental issues of recycling these materials from the environment [4, 11, 20, 46, 91, 118].

The waste plastics that form particulate matter by UV irradiation and weathering increase surface area and mobility and thereby incorporate easily into the food chain, causing fatal injuries to all living organisms [14, 97]. In addition, the small size of suspended plastics causes the reduction of light transmission on the sea surface, photosynthetic efficiency of micro-algae, and the productivity of marine organisms. More seriously, the micro-plastic could act as a carrier to increase the adsorption of hardly decomposable hydrophobic chemicals such as polychlorinated biphenyl (PCB), when it is introduced into the food chain [23, 86]. The groundwater is also contaminated due to hazardous chemicals from the plastic waste in landfills [72].

Based on the annual production of various plastics such as polyamide (PA), polyethylene (PE), polyethylene terephthalate (PET), polypropylene (PP), polystyrene (PS), polyurethane (PU), and polyvinyl chloride (PVC), PE

*Correspondence: hyungz@korea.kr; hghur@gist.ac.kr

[†]Sunil Ghatge and Youri Yang are contributed equally to this work

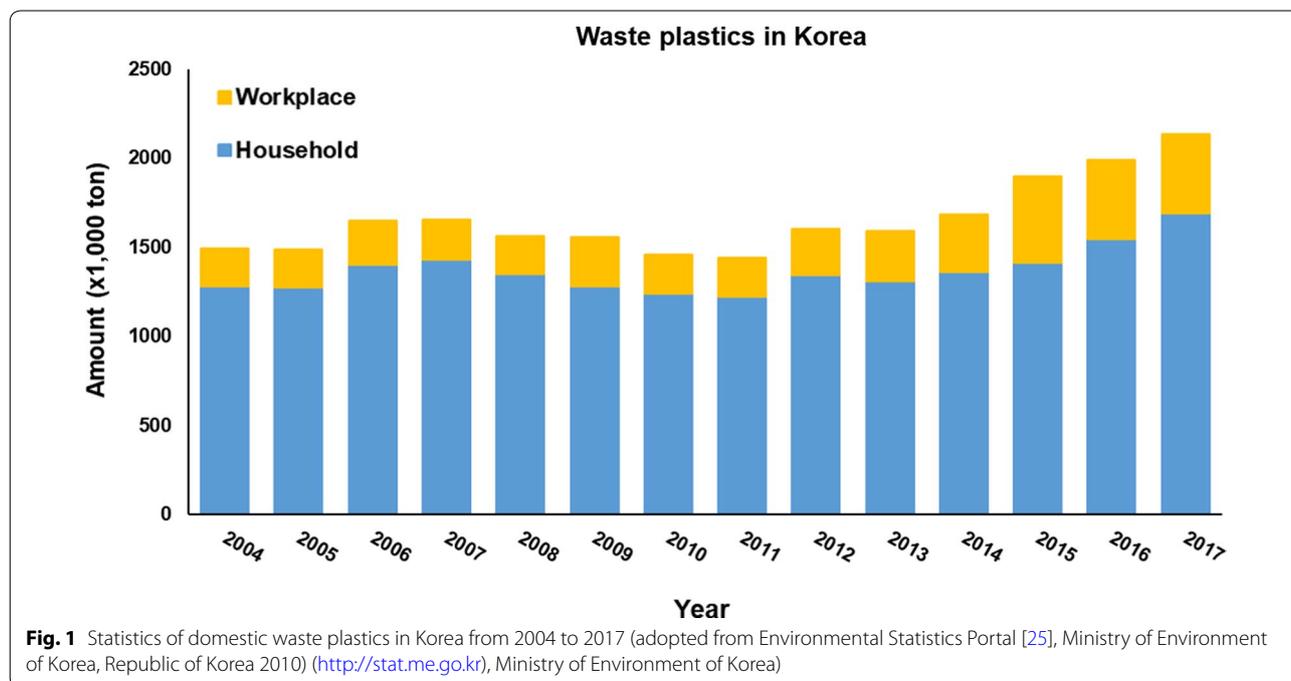
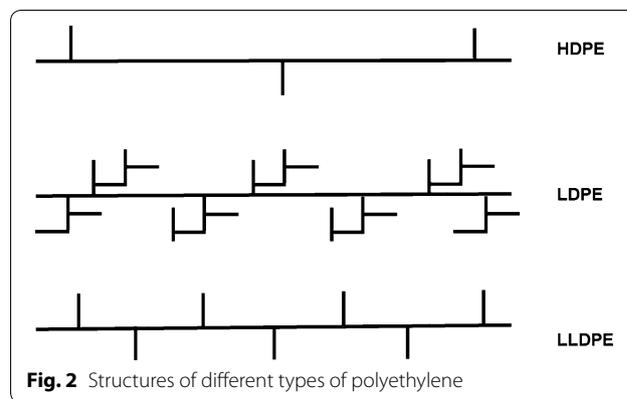
¹ School of Earth Sciences and Environmental Engineering, Gwangju Institute of Science and Technology (GIST), Gwangju 61005, Republic of Korea

² Agricultural Microbiology Division, National Institute of Agricultural Sciences, Rural Development Administration, Wanju 55365, Republic of Korea

and PP that have been widely used in Korea agriculture, comprise more than 60% of the total global plastic production and the annual production of PE is 116 million tons [21]. In 2015, 6.7 million tons of plastic resins were used in Korea, meaning that every single Korean consumed every single Korean consumed 132.7 kg of plastic per year. It represented that Korea is the second largest plastic consumer in the world. From the view on generation of waste plastics, about 10.1 million tons of waste plastic generated in 2016 in Korea, indicating 196 kg of the waste per person per year (EUROMAP [29]). The amount of waste plastics has been increasing every year in Korea (Fig. 1). According to a survey by ministry of environment in Korea, waste plastics from the agricultural environment were 310,000 tons and among them, 200,000 tons were collected and 170,000 tons were recycled (<http://stat.me.go.kr>). The non-recycled plastics left in the agricultural environment without any management will be getting worse due to a decrease in international oil prices and increase of plastic waste.

PE is highly recalcitrant and inert material hence it is very difficult to degrade in the environment even after buried for several years as landfill. A polyethylene sheet showed only partial degradation and negligible weight loss when kept in moist soil for 12–32 years [77]. The recalcitrance of PE is due to its water insolubility, the hydrophobicity because of presence of linear backbone of carbon atoms, degree of crystallinity, and its high molecular weight [86, 111, 114]. PE such as low density

polyethylene (LDPE) and high density polyethylene (HDPE), have been used for biodegradation studies [86, 97] (Fig. 2). LDPE was prepared by the high pressure polymerization of ethylene. The presence of branched chains is responsible for the low density of LDPE. Chemically, LDPE is inert at room temperature, however, it can be gradually attacked by strong oxidizing agents and some solvents, results in softening or swelling. It is intact for short period of time up to 95 °C and can be durable for longer hours at 80 °C. A degree of crystallinity of LDPE is within the range of 50–60% which provides several properties to the material such as opacity, tear strength, tensile strength, rigidity and chemical resistance, flexibility even at a low temperature [12, 33]. There



are two different types of LDPE, linear low density polyethylene (LLDPE) and branched low density polyethylene (BLDPE). They show different density, degree of branching and availability of functional groups on the surface. LDPE films are transparent, free from odor and toxicity. They have better ductility, low water vapor permeability, and heat seal ability [78, 109]. It is widely used for packaging food and non-food items, manufacturing trays and plastic bags. LDPE is utilized for mulching agricultural fields and constructing polyhouse. It is also utilized for coating on paper, textiles, and other plastics [97]. Notably, PE accounts for 97% of total agricultural waste vinyl and about 200,000 tons of LDPE were consumed, consisting of 64% of all types of agricultural waste vinyl (Fig. 3) (<http://stat.me.go.kr>). HDPE is a PE thermoplastic, produced by a catalytic process and having little branching. It has stronger intermolecular forces and greater tensile strength than LDPE. The higher density provides greater stability due to reduced bond length and compact packaging. Due to its hardness, opacity, and durability at higher temperatures (up to 120 °C), it is widely used in industrial and day-to-day applications such as production of carry bags, milk jugs, detergent bottles, margarine tubs, garbage containers, water pipes, etc. [8].

The extensive usage of both LDPE and HDPE poses severe environmental threats to the terrestrial and marine ecosystems, as experienced like blockages of PE in the intestines of fish, birds and marine mammals [10, 99]. In addition, several hundreds of different species from different ecosystems are on the edge of becoming endangered due to the ingestion of this waste [95, 104]. Inevitable use of PE to increase agricultural productivity in small territory has caused a significant social issue. According to a report by Chinese Academy of Agricultural Sciences in 2014, mulching waste vinyl reduced the movement of essential materials in soil such as air, moisture, and nutrients and the mobility of soil organisms including earthworms [60]. This led to a decrease in soil quality, physiological disorders in plant growth such as seed germination and root growth, which would which would decrease agricultural productivity in the end. Thus, the use and waste treatment for plastics have become a global problem. Therefore, it is of inevitable necessity to minimize PE and other plastics and to develop efficient methods for plastic degradation and recycling.

Extensive research has been carried out for degradation of PE either by physico-chemical or microbial methods or combination of both [14, 86, 97]. Physico-chemical technologies include thermal and UV treatment or combination of both, which reduces the polymer chain size and form oxidized groups such as carboxyl, carbonyl and hydroxyl, on the surface of polymer [19, 44, 57, 68]. These treatments modify the crystallinity and surface

morphology of the original polymer and facilitate the polymer biodegradation [59]. Oxidation of PE with nitric acid has been known to promote fragmentation of PE films followed by microbial degradation [68]. The biodegradation of PE involves use of microbes or microbial communities that modify and consume the polymer as a source of energy leading to changes in its physico-chemical properties such as weight loss, structural deterioration, and eventually carbon fixation as a biomass [86, 97]. However, the formation of a biofilm by polymer-degrading microorganisms on PE was restricted due to a high degree of hydrophobicity, a low specific surface area and smooth surface topography [62, 86]. In addition, productive adsorption and catalytic performance of polymer-degrading enzymes have been shown incompatible with a hydrophobic polymer surface [27, 87, 88, 92]. Besides, worms of the moth, *Galleria mellonella* and *Plodia interpunctella*, have been observed to degrade untreated LDPE [13, 63].

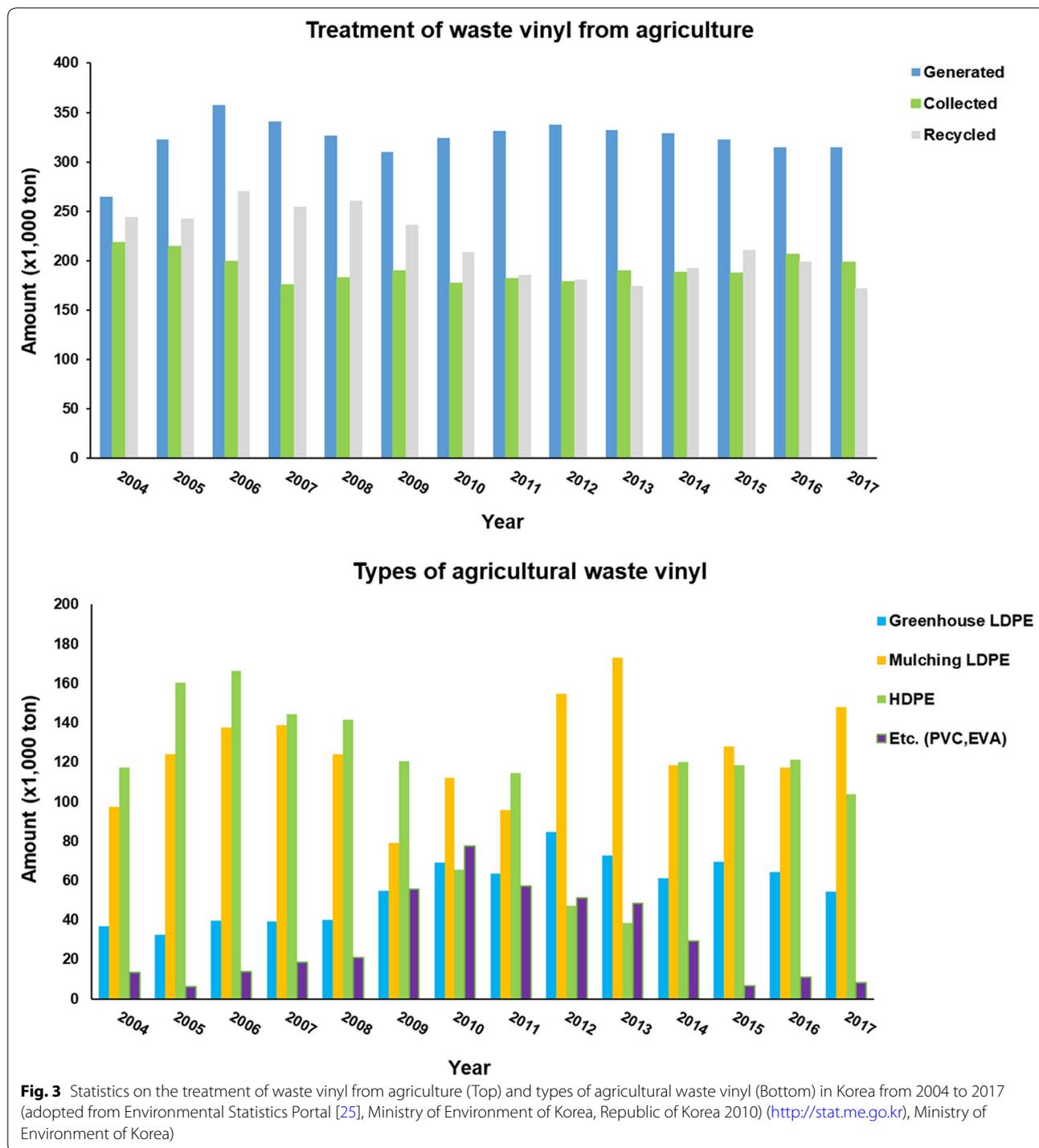
In most of these biodegradation studies, PE degradation was determined by measuring weight loss, observing polymer structural changes under SEM, and chemical modifications of functional groups using FTIR [86, 97]. It has been criticized that the weight loss and surface topography changes are probably derived from the degradation of various additives, which often contribute to significant fraction of the PE. Hence, the results from many of these studies need to be cross checked using more advanced physico-chemical, biochemical, and molecular biological technologies [21, 115]. Furthermore, the exact biochemical mechanisms and enzymes involved in PE breakdown are still unknown. From the view on the establishment of waste vinyl management, the researches on finding microorganisms effectively degrading plastics and analyzing their physiology and application are essential.

In this review, we described the effect of pretreatment and additives for enhancing biodegradability of PE and summarized recent studies on biodegradation of PE by various bacteria, fungi, and moth worms along with gut microorganisms from worms. In addition, we discussed candidate microbial enzymes involved in PE biodegradation and various advanced technologies used for evaluating the actual biodegradation of PE.

Pretreatments and effects of additives for enhancing biodegradability of PE

Thermo-UV pretreatment

Thermo-UV pretreatment was used to induce partial photolysis of the PE film and to simulate weathering of PE that occurs in nature (e.g., PE used for soil mulching or polyhouse cover). PE samples were treated in a QUV accelerated weathering tester developed by QLAB,



Homestead, FL. Herein, PE was alternatively exposed to cycles of UV and humidity: five cycles per d of UV exposure (four of 4 h each, one of 3 h at 70 °C) separated by 1-h intervals (50 °C). The pretreated PE film was tested for biodegradation and biofilm formation by *Rhodococcus ruber* (C208). FTIR spectra of UV-photooxidized PE

incubated with *Rhodococcus ruber* strain C208 showed that formation of the carbonyl residues on the surface of photooxidized PE plays an important role in initiation of biodegradation [38, 101]. In another study, *Bacillus amyloliquefaciens* strain isolated from composed plastic was studied for assessing deterioration effect

of thermo-irradiation pretreatment. Herein LDPE and LLDPE films were exposed to gamma rays followed by thermal treatment at 150 °C and 90 °C for 7 d, respectively. Furthermore, LDPE films were additionally exposed to UV irradiation in an oven at 60 °C for 7 d [73]. When the pretreated LLDPE was incubated with the isolated bacterium for 40–60 day, the dry weight of LLDPE was slightly decreased by 1.1 ± 0.3 to $3.2 \pm 1.3\%$, with flattening of carbonyl band ($1300\text{--}1100\text{ cm}^{-1}$) in the FTIR spectra, indicating a biodeterioration. Electrospray ionization-mass spectrometry (ESI-MS) analysis showed the release of 3-hydroxybutyrate oligomers only in the medium containing pretreated LLDPE and not with native LLDPE. These oligomers disappeared after incubation with *Bacillus amyloliquefaciens* indicating metabolism of low molecular LLDPE fractions.

Balasubramanian et al. [9] used combination of physical, chemical and biological treatment with *Aspergillus terreus* MF12 for enhancing HDPE degradation. HDPE film initially heated at 50 °C for 72 h followed by exposure to UV (312 nm) and humidity. Secondly, HDPE film was chemically treated by immersing into KMnO_4/HCl at a concentration of $0.25/0.5\text{ mol l}^{-1}$ at 45 °C for 8 h [32] and 10% citric acid for 8 h at 45 °C. The HDPE degradation gradually increased from 9.4 ± 0.1 to $20.8 \pm 0.1\%$ between the physico-chemical and biological treatments, indicating synergism between biotic and abiotic factors for HDPE degradation by *A. terreus* MF12 [9].

Treatment with pro-oxidants

All commercial prepared PE films contain a small amount of a stabilizer that prevents oxidation during processing and significantly prolongs its lifetime [15]. Supporting pro-oxidant additives to enhance the photo- and thermo-oxidation of PE films may lead to radical reactions that result in polymer chain cleavage [54, 55]. Conventional pro-oxidants include transient metal ions such as iron, manganese, titanium, cobalt in the form of stearate. Iron and titanium complexes as a source of radicals initiate photo-oxidation, while manganese and cobalt catalyze peroxidation without light. Study on biodegradation of PE (LDPE) treated with pro-oxidant, manganese stearate followed by UV irradiation and treatment with *Aspergillus oryzae* resulted in 62 and 51%, decrease in percentage elongation and tensile strength, respectively. Furthermore, FTIR analysis confirmed the formation of more carbonyl and carboxylic groups after treatment with pro-oxidant over UV treated film, which was completely degraded after incubation with *A. oryzae* thus confirming role of pro-oxidants in enhancing PE biodegradation [52]. Although it is clear that lower molecular weight products were formed by the catalytic action of pro-oxidants, which were consumed by the microorganisms,

however, it is not known how microbes participate in polymer chain cleavage and what kind of enzyme system is involved in this process.

Photo-catalysis using titanium dioxide (TiO_2)

TiO_2 is an ecofriendly photocatalyst that absorbs light in the UV region. Hence, TiO_2 -incorporated polymer films efficiently absorb UV light. TiO_2 mediated photocatalysis involves the absorption of photons of suitable energy leading to the generation of electrons and holes which promote the formation of free radicals, resulting in the oxidation of the polymer, followed by its degradation. Thankam thomas and Sandhyarani et al. (2013) investigated photocatalytic degradation of LDPE incorporated with titania nanoparticles after treatment with solar radiation. The composite PE film showed weight loss of 68% after exposure to the solar radiation for 200 h which is a significantly higher compared with a study where the similar rate of weight loss was observed after 400 h [108, 125]. FTIR and SEM analysis revealed the presence of carbonyl groups and the creation of holes at the interface, respectively indicating the degradation of the LDPE. Another study showed that solar irradiation of a PE film blended with copper phthalocyanine (CuPc) modified TiO_2 (TiO_2/CuPc) photocatalyst resulted in significant weight loss rate, rough surface texture, higher amount of generated CO_2 , compared to the original PE film [126]. Surface photovoltage spectroscopy (SPS) analysis suggested that CuPc promoted charge separation of TiO_2 . Reactive oxygen species generated on the surface of TiO_2 or TiO_2/CuPc particles are responsible for enhanced degradation of PE. Recently, Fa et al. [31] synthesized $\text{TiO}_2\text{-FeSt}_3$ ferric stearate-polyethylene (TFPE) composite film and studied photo-degradation by treating UV irradiation for 240 h and/or thermo-degradation at 70 °C for 30 d. FTIR spectroscopy confirms the formation of carbonyl and hydroxyl group which assist in biodegradation of PE films. The tensile strength and elongation at break of TFPE film reduced to 60% and 97.7%, respectively [31].

Biodegradation of PE

The biodegradation of recalcitrant PE has been investigated by many researchers [28, 40, 56, 62, 98, 100, 127]. Still, the complex biodegradation mechanism of PE is not yet fully understood. It is suggested that various abiotic and biotic factors play a vital role in the biodegradation of PE in the environment [62, 100]. Biodegradation studies have been accomplished either using pure cultures that are able to degrade PE [2, 5, 8, 34, 38, 41, 54, 81, 84, 101, 110, 113, 117, 119] or using complex microbial communities from various terrestrial (soil from landfill sites, composting) and marine habitats [1, 3, 6, 18, 49, 61, 71,

74, 76, 79]. In addition, it was reported that bioaugmentation with tailored microbial consortia could facilitate the biodegradation of PE [107]. Furthermore, significantly faster biodegradation of PE was observed by waxworms.

Bacterial biodegradation of PE

More than 20 bacterial genera have been shown to degrade different types of PE. Those include various Gram-negative and Gram-positives species belonging to the genera *Pseudomonas*, *Ralstonia*, *Stenotrophomonas*, *Klebsiella*, *Acinetobacter*, etc. and *Rhodococcus*, *Staphylococcus*, *Streptococcus*, *Streptomyces*, *Bacillus*, etc. [21, 39, 86, 97]. Most of these bacterial strains possess the ability to deteriorate surface and/or form a biofilm on PE. Table 1 summarizes bacterial strains associated with PE biodegradation. Studies on diverse activities of the genus *Pseudomonas* have been carried out to investigate their capabilities to degrade and metabolize a variety of synthetic plastics polymers and the by-products. *Pseudomonas* species have the unique ability to degrade and metabolize the polymers with extracellular oxidative and/or hydrolytic enzyme activities, which facilitate uptake and degradation of the polymer fragments, and control interaction between biofilms and polymer surfaces [117]. Complete degradation of PE in water was observed after treatment with *P. fluorescens* in presence of surfactant

and biosurfactant suggesting their importance in polymer oxidation and biodegradation [5]. Tribedi and Sil [110] showed that the addition of mineral oil to the LDPE degradation medium of *Pseudomonas* sp. strain AKS2 stimulated the hydrophobic interaction to form biofilms on polymer surfaces and degraded $5 \pm 1\%$ of the original PE material for 45 d, whereas Tween 80 had an adverse effect to the biofilm formation. A thermophilic bacterium *Brevibacillus borstelensis* isolated from soil has been reported to utilize BLDPE as the sole carbon and energy source, by which 30% of the molecular weight of PE film was reduced during an incubation period of 30 d [41]. [38] isolated a biofilm producing *Rhodococcus ruber* (C208) strain which degraded PE at a rate of 0.86% per week. A hydrophobic cell surface (e.g., mycolic acid layer) of this strain may play an important role in biofilm formation on PE surface [38, 101]. Awasthi et al. [7] reported that HDPE after thermal treatment was degraded by *Klebsiella pneumoniae*. This strain was able to strongly adhere to HDPE surfaces, leading to increasing biofilm thickness with decreasing weight and tensile strength of the HDPE film by 18.4% and 60%, respectively, in 60 d. Possible biodegradation of an HDPE film exhibits the SEM and atomic force microscopy (AFM) images of sub-surface corrosion, cracks, and surface roughness produced by bacteria.

Table 1 Bacterial strains associated with PE degradation

Bacterial strain	Substrate	Country of origin	References
<i>Streptomyces badius</i> , <i>S. setonii</i> ,	starch-PE	USA	[81]
<i>Arthrobacter paraffineus</i>	LDPE, HDPE	Sweden	[2]
<i>Brevibacillus borstelensis</i>	LDPE	Israel	[41]
<i>Nocardia asteroides</i>	LDPE, HDPE	France, Czech Republic	[54]
<i>Rhodococcus rhodochrous</i>	LDPE, HDPE	France, Czech Republic	[54]
<i>Bacillus halodenitrificans</i>	LDPE	India	[91]
<i>Bacillus sphaericus</i>	LDPE, HDPE	India	[105]
<i>Arthrobacter</i> sp.	HDPE	India	[8]
<i>Pseudomonas</i> sp.	HDPE	India	[8]
<i>Staphylococcus epidermidis</i>	LDPE	India	[17]
<i>Rhodococcus rhodochrous</i>	LDPE, HDPE, LLDPE	France, Belgium, Italy	[34]
<i>Arthrobacter viscosus</i> , <i>Acinetobacter baumannii</i>	LDPE	Poland	[74]
<i>Bacillus amyloliquefaciens</i> , <i>B. cereus</i> , <i>B. circulans</i> , <i>B. mycoides</i> , <i>B. pumilus</i> , <i>B. thuringiensis</i> , <i>M. luteous</i> , <i>M. lylae</i>			
<i>Pseudomonas fluorescens</i> , <i>Paenibacillus macerans</i> , <i>Rahnella aquatilis</i>	LDPE	Poland	[74]
<i>Staphylococcus cohnii</i>	LDPE	Poland	[74]
<i>Staphylococcus xylosus</i>	LDPE	Poland	[74]
<i>Microbacterium paraoxydans</i>	LDPE	India	[84]
<i>Pseudomonas aeruginosa</i>	LDPE	India	[84]
<i>Rhodococcus ruber</i> C208	LDPE	Israel	[93]
<i>Pseudomonas</i> sp. AKS2	LDPE	India	[110]
<i>Klebsiella pneumonia</i>	HDPE	India	[7]

Fungal biodegradation of PE

In addition to bacteria, several fungal genera including *Aspergillus*, *Cladosporium*, *Fusarium*, *Penicillium*, *Phanerochaete* have been reported for PE degradation [21, 39, 86, 97]. In general, fungi are thought to be more efficient than bacteria for the degradation of PE because they are capable of attaching to the hydrophobic surface of the polymers [51, 96, 97], producing extracellular enzymes targeting insoluble fibers, and surviving in stressful growth conditions [98]. Table 2 summarizes some fungal strains capable of degrading PE. Weight loss measurement is a commonly used method to analyze biodegradation of PE. For examples, biodegradation of LDPE by *A. niger* and *A. japonicas* in laboratory conditions have been found to decrease the dry weight by 5.8% and 11.1% per month, respectively [82]. Das and Kumar [66] studied microbial deterioration of LDPE by *Aspergillus* and *Fusarium* sp. Among them *Fusarium* sp. FSM-10 and *Aspergillus* sp. FSM-3 showed maximum weight reduction about 8–9%, while only 5% weight loss was observed by *Aspergillus* sp. FSM-5 after 60 d of incubation. Usha et al. [112] isolated strains belonging to *Aspergillus flavus* and *A. nidulans* through enrichment culture showing the clearing zone around their colonies on PE agar plates. Kathiresan [50] isolated PE degrading fungi from mangrove soils. Yamada-Onodera et al. [119] reported degradation of additives free PE by *P. simplicissimum* YK. Esmaeili et al. [26] isolated *A. niger* from soils of PE wastes landfills using mineral medium with PE powder as a sole carbon source. From SEM and AFM analysis of the PE surface, several strains of *Chrysonilia*, *Aspergillus*, and *Penicillium* species have been isolated using synthetic medium [67]. Among the isolated fungi, *P. chrysogenum* NS10 (KU559907), *P. oxalicum* NS4 (KU559906) have been evaluated for HDPE and LDPE degradation using the response surface methodology to optimize the growth media for increasing the mycelium weight. The AFM and SEM analysis is widely used to ratify PE degradation by

fungal strains leading to biofilm formation and morphological changes on LDPE and HDPE surfaces, including cracks, pits, and undulations [75].

Role of Waxworm and gut microbiome in biodegradation of PE

Larvae of *Galleria mellonella* and *Plodia interpunctella*, have been reported to degrade LDPE without pretreatment [13, 63]. The worms could soften thin-film PE shopping bags and metabolize them to ethylene glycol. Because there is a structural similarity between beeswax and PE, the biochemical machinery for beeswax metabolism of *G. mellonella* makes it useful for PE metabolism. About 100 worms of *G. mellonella* can cause a weight loss of 92 mg from a commercial PE shopping bag within 12 h. [115] criticized that the waxworm research lacked the necessary information to support the claims of the original *G. mellonella* report. Although the waxworm researches lacked information about the biodegradation mechanism of PE [115], cutting holes on the surface of PE by waxworms and FTIR analysis of degraded PE indicated that PE pieces break down with the carbon–carbon bond cleavage by mechanical force or enzymatic digestion or both. Recently, biodegradation of PE by *Enterobacter* sp. D1 from the guts of wax moth *G. mellonella* has been investigated [85]. The authors performed AFM and SEM analysis to show that the strain D1 was able to form colonies around a PE film after 14 d of incubation and disrupted the PE film surface. The treatment of a PE film with the strain D1 has highlighted that the appearance of carbonyl and ether functional groups on the FTIR spectra was concomitant with the release of oxidative cleavage products containing alcohol, ester and acid groups, analyzed by high-performance liquid chromatography-mass spectrometry. This study indicates the involvement of strain D1 in PE degradations. However, it is still unclear that PE is degraded by *G. mellonella* larvae or by the gut flora. Recently, complete genomic and

Table 2 Fungal strains associated with PE degradation

Fungal strain	Substrate	Country of origin	References
<i>Fusarium redolens</i>	LDPE C14 labelled; UV treated	Sweden	[1, 49]
<i>Verticillium lecanii</i>	LDPE	Sweden	[49]
<i>Phanerochaete chrysosporium</i>	LDPE/starch	Turkey	[76]
<i>Penicillium simplicissimum</i>	HDPE UV treated	Japan	[119]
<i>Aspergillus niger</i>	Thermal treated LDPE	USA	[113]
<i>Penicillium pinophilum</i>	Powdered LDPE	USA	[113]
<i>Cladosporium cladosporioides</i>	Degradable polyethylene green film	France, UK	[14]
<i>Glioclodium virens</i>	Thermal treated LDPE	Mexico	[64]
<i>Aspergillus flavus</i>	LDPE and HDPE film	France, Czech Republic	[54]

transcriptomic data of *G. mellonella* have been used to explore the beeswax and PE metabolism [53]. Notably, the long chain fatty acids from beeswax and PE were detected even in the absence of gut microbiota indicating that the PE metabolism can occur without help of gut microflora.

In another study, two PE-degrading strains, named *Bacillus* sp. YP1 and *Enterobacter asburiae* YT1, were isolated from the gut of *P. interpunctella* larvae [123]. They reduced the hydrophobicity and caused surface disruption of PE film after 28 d of incubation. After 60 d of incubation with these strains approximately $10.7 \pm 0.2\%$ and $6.1 \pm 0.3\%$ of the PE films (100 mg) were degraded, respectively. These findings demonstrated the importance of gut microbes of moth larvae for PE biodegradation. Yang et al. [120] explored biodegradation of PE and plastic mixtures by yellow mealworms i.e. larvae of *Tenebrio molitor*. Up to $49.0 \pm 1.4\%$ of the ingested PE was metabolized to CO_2 after incubation with larvae. PE-fed mealworms showed $40.1 \pm 8.5\%$ reduction in the molecular weights of egested polymer. The gut microbiome study with next generation sequence analysis has shown the abundance of *Citrobacter* sp. and *Kosakonia* sp. attached to PE. Both of them are members of the *Enterobacteriaceae* family which is known to contain PE degrading bacteria's [123]. In addition, *Citrobacter* sp. (aerobic), and *Kosakonia* sp. (facultative anaerobic) can utilize oxygen which suggests their involvement in plastic degradation [37, 56, 98, 111]. Further studies are required to reveal the mechanism of enzymatic degradation in the guts of mealworms and waxworms, which will facilitate the biodegradation of a variety of plastic materials.

Effects of microbial activity on PE

The effect of microbial colonization on the surface of PE was studied by monitoring seven different characteristics with respect to the degree of biodegradation of the

polymer: functional groups on the surface, hydrophobicity/hydrophilicity, crystallinity, molecular weight distribution, surface topography, mechanical properties, and mass balance (Table 3). The methods used to study these changes have been extensively reviewed in previous studies [86, 97]. Hence we will just briefly discuss each of these 7 characteristics for understanding PE degradation. FTIR spectroscopy is used to study the formation of various functional groups on the surface of PE after the abiotic and biotic oxidation by thermo-UV treatment and microbial degradation. For example, UV and nitric acid treatment to PE led to an increase in absorbance of infrared at $1710\text{--}1715\text{ cm}^{-1}$ (corresponding to carbonyl group) and 1640 cm^{-1} and $830\text{--}880\text{ cm}^{-1}$ (corresponding to --C=C--), which was then reduced after incubation with microbial consortia [43]. Similarly, Harshvardhan and Jha [42] reported PE biodegradation with an increase in the index of carbonyl bond, the keto carbonyl bond, and the vinyl bond, calculated using FT-IR spectra. These functional groups at the surface of PE are considered important because oxidized groups cause an increase in the hydrophilicity which in turns results in the efficient attachment of microbes to the PE surface thereby promoting the biodegradation [2, 110]. The hydrophilicity is usually estimated by measuring the contact angle of the surface with water. A small contact angle with water indicates a high hydrophilicity of the oxidized PE surface [91, 105].

Crystallinity is another important parameter to predict the extent of biodeterioration of the polymer, measured with the help of Differential Scanning Calorimetry (DSC) and FTIR analysis. Generally, the amorphous region is easily accessible and degraded by microorganisms, resulting in an initial increase in crystallinity [5, 44, 83]. After attack (or dwell) in the amorphous regions, microorganisms will start degrading the crystalline region and increase in the proportion of larger crystals [2, 105]. Size

Table 3 Techniques used for characterization of PE biodegradation

Techniques used	Changes in PE	Measured characteristics	References
FTIR	Functional groups on the surface	Keto-carbonyl index (I1715/I1565); Ester-carbonyl index (I1740/I1465); Vinyl-bound index (I1640/I1465); Double bound index (I908/I1465); C-O stretching (I1100)	[3, 6, 34, 38, 41, 74, 83, 93, 94, 105, 113]
Contact angle drop deposition	Hydrophobicity/hydrophilicity	Contact angle with water, Surface energy	[6, 91, 105]
FTIR/DSC/XRD	Crystallinity	Crystallinity; Melting temperature; Relative crystallinity; Lamellar thickness	[2, 76]
HT-SEC/GPC	Molecular weight distribution	Changes in the molecular weight	[38]; [41];
SEM/AFM	Surface topography	Molecular weight distribution topography	[6, 105]
Instron	Mechanical properties	Tensile strength; Strain energy; % Elongation and extension	[8, 14, 18]
Gravimetric CO_2 evolution	Consumption of the polymer	Weight loss	[34, 74, 93]

exclusion chromatography and time of flight mass spectrometry (TOF–MS) analysis gives an idea about molecular weight distribution of the PE after biodegradation. An increase in the average of molecular weight is witnessed after initial degradation of low molecular weight chains [5, 41, 81, 119].

SEM and AFM analyses are commonly employed to investigate surface topography of PE films during biodegradation. Biofilm formation on the surface of the polymers [14, 34, 38, 54, 101, 110] and penetration of hyphal structures [44, 83, 113] have been generally observed after incubation with microbes. The structural changes in the formation of pits, holes, and erosions have been observed under SEM indicating surface destruction of PE. Alterations in crystallinity and the average molecular weight as a result of oxidation modify the chemical and mechanical properties of PE. Universal mechanical testing system (UMTS) is preferentially used for studying changes in the mechanical properties of a polymer [74, 79, 105]. However, this method is prone to underestimate the local surface related damage caused by the microorganisms. Microorganisms utilize PE as a sole carbon source and metabolize it to CO₂ during respiration and hence measurement of released CO₂ can be linked to the amount of polymer consumed. Some studies reported a decrease in the weight of samples measured either by gravimetric measurements [6, 41, 74, 101, 105] or by CO₂ emissions from the samples [1, 3, 49, 96]. Progressive CO₂ emissions in the samples are measured to define the total degradation of the polymer along with its rate.

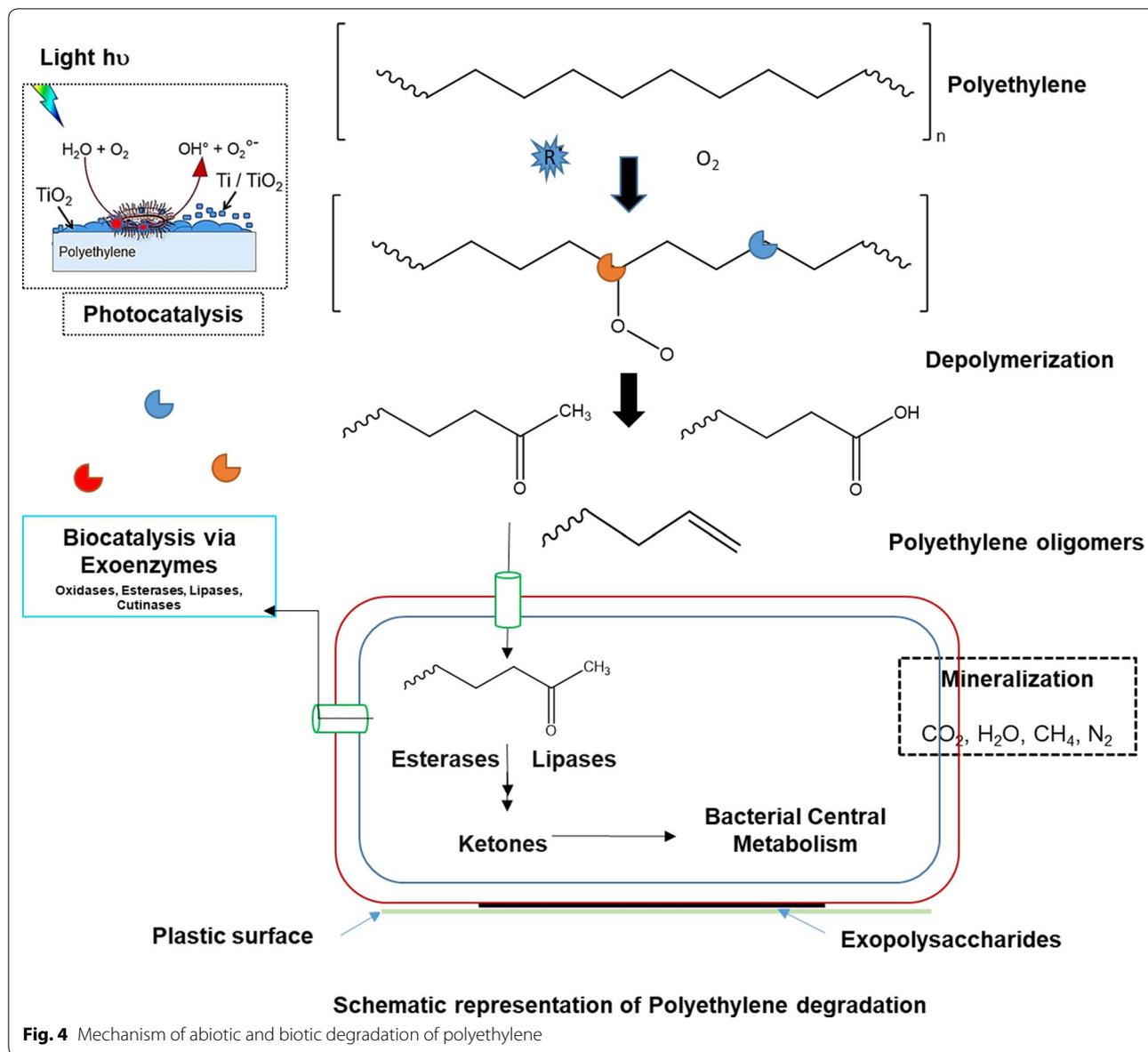
Microbial enzymes involved in PE biodegradation

The biodegradation of plastics is a complex process involving various abiotic and biotic factors [28, 40, 56, 62, 98, 100, 127]. The cooperative action of abiotic factors and microorganisms causes fragmentation of the bulk polymer to increase accessible surfaces for biodegradation. Some extracellular enzymes carry out further fragmentation of the polymers [62, 100] (Fig. 4). Several lignin degrading enzymes also participate in the breakdown of the PE thermoplastic [86, 100]. After the initial cleavage of the polymer into oligomers of 10–50 carbon atoms which can be transported into the cell for further metabolism [62, 86]. Biodegradation of PE is restricted by the absence of hydrolysable functional groups in the backbone [56, 86]. The carbonyl and hydroxyl groups formed from various pretreatment such as thermo-UV irradiation or addition of oxidizing agents could be adopted to stimulate biodegradation further [34, 54] a, b). Hence most of the biodegradation studies on PE have been carried out using substrate peroxidation [86].

Microbial enzymes capable of degrading lignin polymer containing oxidizable C–C bonds [35, 106]

have been involved in the biodegradation of PE [56, 86]. These include manganese peroxidase (MnP, EC 1.11.1.13), lignin peroxidases (LiP, EC 1.11.1.14), and laccases (EC 1.10.3.2.) [116]. A copper dependent laccase from *R. ruber* strain C208 was reported to degrade UV pretreated PE films [93]. Laccase mediated oxidative cleavage of amorphous region of PE films results in formation of easily accessible carbonyl groups and significant decrease in weight of a PE film. Fujisawa et al. [36] showed reduction of the molecular weight of a PE membrane after treatment with laccase from *Trametes versicolor* in presence of 1-hydroxybenzotriazole as a mediator. Degradation of a high molecular weight PE membrane by *P. chrysosporium* ME-446 and an isolate IZU-154 have been described and MnP from this white-rot fungus was found to be the key enzyme responsible for PE degradation [45]. A partially purified MnP stimulated degradation of PE in presence of various surfactants [24, 45]. The most active MnP from IZU-154 has been characterized in regard to the oxidation of 2,6-dimethoxyphenol [65] and the degradation of nylon-66 [22]. Enhanced extracellular secretion of laccases and MnP from *B. cereus* was observed when the strain was incubated with UV-irradiated PE [102]. However, the same PE film treated with a partially purified laccase and a MnP from *P. simplicissimum* showed negligible weight loss [103]. The LiP activity in the concentrated culture supernatants of lignocellulose degrading *Streptomyces* species has been reported to responsible for degradation of a heat treated PE [81]. Similarly, up to 70% degradation of a pre-oxidized high molecular weight PE sample has been reported after 15 d of treatment with *P. chrysosporium* strain MTCC-787. The extracellular peroxidases play a vital role in the biodegradation of PE by this strain [70].

AlkB family alkane hydroxylases (AH) (EC 1.14.15.3) can degrade hydrocarbon oligomers by terminal or sub-terminal oxidation [89]. Yoon et al. [124] reported action of a recombinant AH from *Pseudomonas* sp. E4 at 37 °C for 80 d converted 20% of the low molecular weight PE sample to CO₂. Expression of the complete AH system of *P. aeruginosa* strain E7 in *E. coli* showed 30% degradation of a PE sample [48]. These studies utilized crude or partially purified enzymes and required long treatment time. Notably, the use of tailored microbial consortia has shown promising degradation of PS and PE compared to the use of single microorganisms [26, 69, 91, 121]. Recently, transcriptome analysis of *G. mellonella* fed on beeswax similar to PE showed, upregulation of genes encoding carboxylesterase, lipase, and enzymes related to fatty acid metabolism. However, the detailed mechanisms of these enzymes have not been investigated.



Recent developments in the analytical methods to correctly estimate PE degradation

Many researchers investigated PE degrading microorganisms using commercially available polymers that possibly contain various chemical additives. The extent of degradation was estimated by calculating weight loss and functional group changes on the surface of polymer by FTIR. However, it is very obscure that the weight loss and surface structure changes result from degradation of additives, which often contribute a major portion of the polymer. Hence, more substantial changes are needed to distinguish the actual degradation of PE and minimizing chances of artifacts originated from the degradation

of additives [21, 86, 97]. In this regard, a robust, reliable method has been introduced to assess biodegradability of PE via the quantification of CO_2 using gas chromatography as a result of bacterial degradation and respiration [90]. Herein, the soil bacterium *R. rhodochrous* was grown in a defined aqueous medium with PE as the only carbon source and the production of CO_2 was directly related to the mineralization of the added carbon source via bacterial respiration. At stationary phase, no significant difference in the release of CO_2 between cells grown with no carbon source and with LDPE. This suggested that bioavailability of carbon was limited in bacterial growth on LDPE hence showing limited biodegradation.

Furthermore, the effect of UV pretreatment on biodegradability of LDPE was studied by incubating *R. rhodochrous* with UV pretreated LDPE and native LDPE for 35 d and released CO₂ was measured over the time. The biodegradation of UV treated LDPE was threefold greater than non-treated LDPE. Authors also tested growth of alkanes degrading marine bacteria *Alcanivorax borkumensis* on LDPE. Negligible difference between CO₂ generated by *A. borkumensis* on LDPE and the sample without LDPE indicating that the strain was not able to utilize LDPE.

In another study, 1320 oxidized oligomers from PE films before and after biodegradation with *R. rhodochrous* have been characterized by MS and nuclear magnetic resonance (NMR) spectroscopy [30]. The strain can assimilate 95% of the soluble oligomers after 240 d. Notably, longer molecules degraded quickly than the smaller ones, suggesting that both extracellular chain cleavage and intracellular β -oxidation mechanisms play an important role in PE biodegradation. In addition, there are several reports on assessing biodegradability of plastic based on carbon tracing from polymers into CO₂ and biomass. Zumstein et al. [128] used ¹³C-labeled polymer poly(butylene adipate-co-terephthalate) (PBAT), and investigated biodegradability in the soil. Various soil microorganisms and filamentous fungi were found to utilize carbon from each monomer unit of PBAT as a carbon and energy source. Biodegradation and mineralization of PS by *Tenebrio molitor* have been studied using ¹³C-labeled PS [121, 122]. The analysis of fecula egested from Styrofoam-feeding larvae was performed using ¹³C cross-polarization/magic angle spinning nuclear magnetic resonance (CP/MAS NMR) spectroscopy, which showed that 47.7% of the gulped Styrofoam carbon was transformed into CO₂ and 49.2% residue was excreted as fecula with only 0.5% incorporation into biomass. Thus, tests with labeled PS indicated the degradation of PS into ¹³CO₂ and incorporation into lipids [120]. Recently, a mass balance study showed that PE-fed mealworms of *Tenebrio molitor* converted 49.0 ± 1.4% of the ingested PE into CO₂ [120]. In addition, 1H-NMR data were used to determine the chemical modifications in the residual polymer from the excreta of PE-fed mealworms, in comparison with bran-fed mealworms.

Conclusion and future prospective

Until now many PE biodegradation studies had investigated changes in physico-chemical properties and structural deterioration using techniques such as FTIR, DSC, XRD, SEM, AFM, etc. Predominantly tested weight loss along with physico-chemical changes are insufficient to prove the real biodegradation of PE. There is a need for providing concrete and reliable evidence for

biodegradation of PE in order to minimize artifacts formed from degradation of additives rather than PE. Hence, upcoming research should be performed using additive-free PE. In addition, ¹³C-polyethylene degradation is proposed to show the formation of ¹³C-labeled metabolites including CO₂ emissions, progressively during an incubation period of time. Further investigations on the mechanism of enzymatic degradation will highlight the pathway for an efficient biodegradation of PE at molecular levels.

Abbreviations

PE: Polyethylene; FTIR: Fourier transform infrared spectroscopy; SEM: Scanning electron microscope; PA: Polyamide; PET: Polyethylene terephthalate; PP: Polypropylene; PS: Polystyrene; PU: Polyurethane; PVC: Polyvinyl chloride; LDPE: Low density polyethylene; HDPE: High density polyethylene; LLDPE: Linear low density polyethylene; BLDPE: Branched low density polyethylene; ESI-MS: Electrospray ionization-Mass spectrometry; TiO₂: Titanium dioxide; CuPc: Copper phthalocyanine; SPS: Surface photovoltage spectroscopy; TFPE: TiO₂-FeSt₃ ferric stearate-polyethylene; AFM: Atomic force microscopy; HPLC-MS: High-performance liquid chromatography-mass spectrometry; DSC: Differential Scanning Calorimetry; TOF-MS: Time of flight mass spectrometry; UMTS: Universal mechanical testing system; MnP: Manganese peroxidase; LiP: Lignin peroxidases; AH: Alkane hydroxylases; NMR: Nuclear magnetic resonance; PBAT: Poly(butylene adipate-co-terephthalate); CP/MAS NMR: Cross-polarization/magic angle spinning nuclear magnetic resonance.

Authors' contributions

SG and YY written the review and equally contributed for this manuscript; HGH and JHA revised the manuscript and share the corresponding authorship. All authors read and approved the final manuscript.

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Availability of data and materials

All data sets presented in this study are included in the published article.

Ethics approval and consent to participate

Not applicable.

Consent for publication

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Competing interests

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