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Review

Status and Enhancement Techniques of Plastic Waste Degradation in the Environment: A Review

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Abstract: Plastic waste has been gradually accumulating in the environment due to rapid population growth and increasing consumer demand, posing threats to both the environment and human health. In this overview, we provide a comprehensive understanding of the degradation of plastics in real environments, such as soil, aquatic environment, landfill, and compost. Both conventional and biodegradable plastics exhibit limited degradation in real environments, except for biodegradable plastics during industrial composting with high thermophilic temperatures. Meanwhile, we also review techniques for enhanced degradation of plastics such as physical technologies (e.g., photocatalysis, mechanical degradation, and pyrolysis), chemical technologies (e.g., hydrolysis, alcoholysis, ammonia, strong oxidation, and supercritical fluids), and biotechnologies (e.g., microorganisms, microfauna, and microalgae). The future research directions for the enhancement of plastic degradation are also discussed, such as the establishment of equivalency standards, adoption of internal control techniques, the control of precise recycling of plastic products, and the ecotoxicology of their degradation products. Therefore, this review comprehensively summarizes the state of plastic degradation in real environments and proposes methods to improve plastic degradation, providing a theoretical basis for the future control and disposal of plastics.

Keywords: conventional plastics; biodegradable plastics; real environment; physical technology; chemical technology; biological technology

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1. Introduction

Plastics possess the characteristics of affordability, excellent performance, and diverse functionality. It is widely used in packaging, construction, electronics, and other fields and is an indispensable material and tool in the life of human society [1]. Global annual plastic production is currently around 400 million metric tons (Mt) and expected to nearly triple by 2060 due to economic and population growth [2]. Most of the global plastics are non-biodegradable polyethylene (PE), polypropylene (PP), polyethylene terephthalate (PET), polystyrene (PS), and polyvinyl chloride (PVC) [3,4]. Many countries and companies are actively researching the replacement of conventional plastics with bioplastics, which is a potential way to solve plastic pollution [5,6]. With increasing demand, global biodegradable plastics production will account for 62% of total bioplastics production by 2028, reaching nearly 7.43 million tons, mainly including poly (butyleneadipate-co-terephthalate) (PBAT), poly (butylene succinate) (PBS), polylactic acid (PLA), polyhydroxyalkanoates (PHA), and starch mixtures [7,8]. Although biodegradable plastics were once considered the best

alternative to plastic pollution, their actual environmental degradability is controversial yet [9–11].

Plastics are accumulating in the environment as a result of inappropriate waste management practices [12,13]. A total of 473,000–910,000 Mt of plastic waste are deposited in the terrestrial environment of the European Union annually [14]. China consumed 2347.3 Mt of plastic and generated 1420.8 Mt of plastic waste, of which 39.6% was landfilled from 1950 to 2020 [15]. The total amount of plastic waste transported from coastal areas to the oceans each year ranges from 4.8 to 12.7 million Mt. A total of 0.8–2.7 million Mt of plastic waste entered the oceans via rivers each year, of which 0.79–1.52 million Mt came from inland areas [16,17]. The accumulation of plastics in compost, landfills, soil, and aquatic environments affects virtually all aquatic and terrestrial ecosystems globally [6,18]. Plastic waste flows into aquatic and terrestrial ecosystems through the following pathways: (1) Litter and street runoff enter water bodies under rainfall, and some of them are weathered and decomposed to form plastic debris that enters the soil; (2) Plastic waste deposited in landfills generates plastic debris and toxic leachate due to a variety of factors, which can easily be transferred to surrounding soils and water bodies through rainfall and runoff [19]; (3) Residues produced through the composting process contain many plastic fragments are used as compost, resulting in many plastics to enter agricultural soils and migrate into water bodies; (4) Irrigation through the application of sewage sludge from sewage treatment plants leads to the introduction of plastics into soil and water bodies [20]. Waste plastics are also discharged into aquatic environments, such as rivers and oceans. The plastic waste disrupts natural flows, limits fish reproduction, destroys beneficial organisms, and even contributes to global warming by creating a shadowy canopy that hinders plankton growth [9]. Microplastics (MPs) (<5 mm) impede the digestive system of animals in the environment and even lead to death, disrupting the oxidation and genotoxicity, and affecting the growth of plants in the environment [18,21,22]. Therefore, it is crucial to improve the degradation of plastics in the environment in order to reduce the accumulation and hazards of plastics in the environment.

Various technologies have been proposed to enhance the degradation of plastics, such as physical, chemical, and biological technologies [23–27]. For example, physicochemical techniques, such as mechanical shredding, ultraviolet (UV) radiation, high temperature, and pH for enhanced degradation of plastics were summarized [24]. Chemical recovery technologies for the conversion of plastic wastes into chemical fuels and value-added polymers were comprehensively reviewed [27]. Advanced oxidation (direct photodegradation, photocatalytic oxidation, and electrochemical oxidation) and biodegradation techniques and mechanisms were discussed and summarized in detail [28]. The biodegradability and mechanisms of microbial species such as bacteria, algae, and fungi to plastics were reviewed [26]. Reinforced plastic non-biodegradation, such as photo-, thermo-, and mechanical degradation techniques and biodegradation techniques, were reviewed [23]. However, these reviews do not provide a systematic and comprehensive summary of the physical, chemical, and biological enhancement techniques for plastic degradation.

The main objectives of this review include the following: (1) to provide an overview of degradation behavior of conventional and biodegradable plastics in real environments, such as soil, water, landfills, and compost; (2) to summarize techniques for enhancing the degradation of plastics in terms of physical, chemical, and biological methods. Therefore, this study lays a solid theoretical foundation for understanding the degradation behavior and current status of plastics in the environment, as well as for developing efficient degradation methods for plastics, which helps to fill the gaps in this field.

2. Plastic Degradation in Real Environments

Plastics inevitably enter real environments after use, including natural environments such as soil and aquatic environments, and industrial environments such as landfills and compost. Therefore, it is important and necessary to understand the degradation behavior of plastics in the real environments mentioned above. The degradation behavior and

general status of conventional and biodegradable plastics in soil, aquatic environments, landfills, and compost are shown in Figure 1. Table 1 summarizes specific degradation of conventional plastics such as PE, PP, PET, PS, and PVC and biodegradable plastics such as PBAT, PBS, PLA, PHA, and starch mixtures in the above environments.

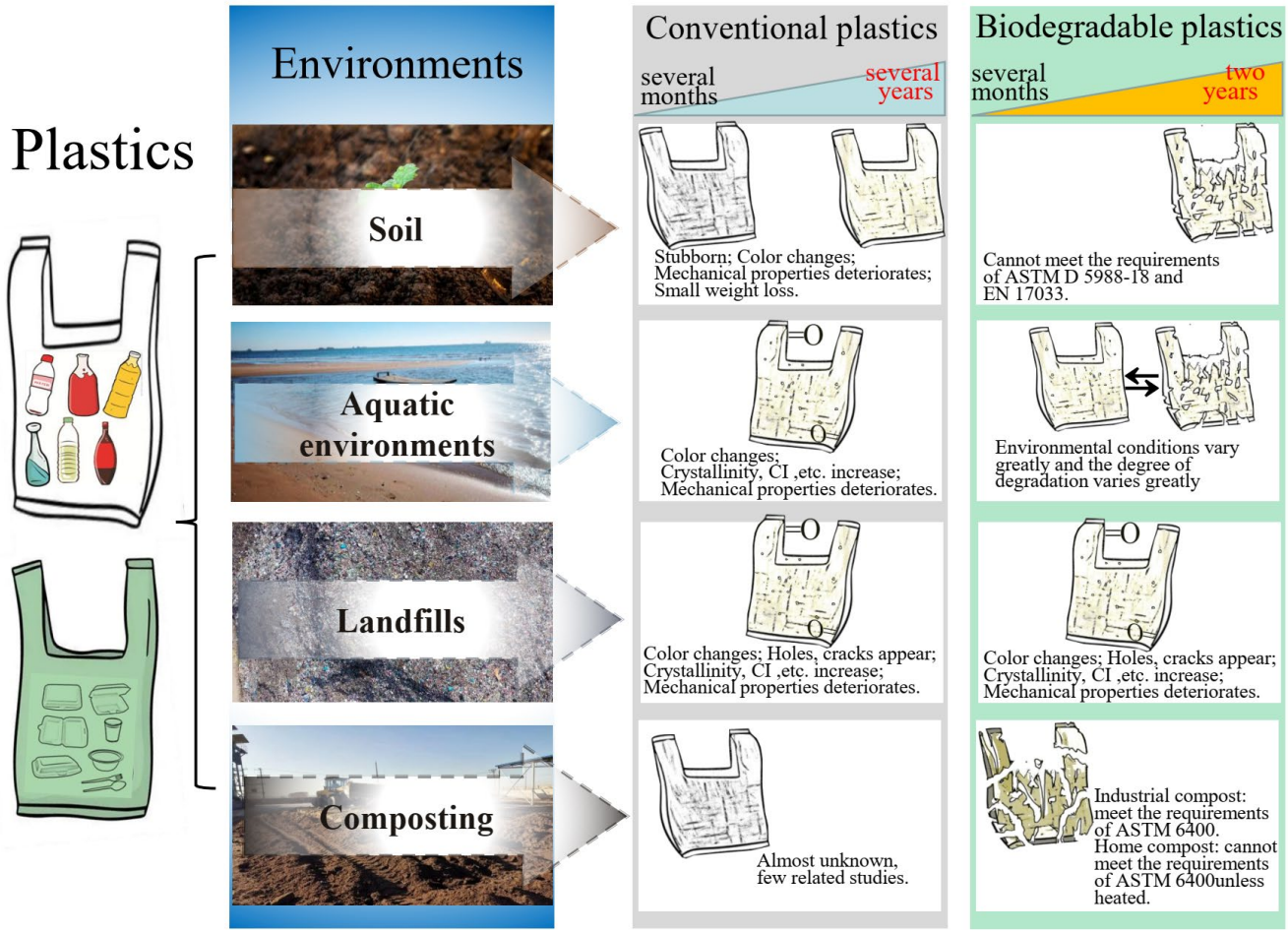

Figure 1. Schematic diagram of the degradation of plastics in the environment.

Table 1. Plastics degradation in real environments.

Plastic Types		Location	Condition	Environment and Period	Indicators	Degradation	References
Conventional plastics							
PE	PE	Washington State University	Temperature, 15 °C; Humidity, 82%	113 d in the soil	Surface topography	Almost 0%	[29]
	PE	Mediterranean Sea at the Bay of Fetovaia, Elba, Italy	Temperature, 14.9 ± 1.0 °C	33 d in 3 m—Water column and 6 m—sediment surface	Mechanical tests	Slightly change	[30]
	PE	The island of Elba in the Mediterranean Sea	Temperature, 19 °C	12.5 months in 25 m—water column	Tensile strength and disintegration	No change	[31]
	PE	Queens Anne Battery Marina	--	40 weeks in the sea	Tensile strength and surface area loss	Less than 2%	[32]
	PE	Czech Republic, Pilsen Region	Precipitation, 582 mm; Temperature, 8.0 °C	36 months in landfill	Surface topography	No change	[33]
	PE	Knoxville, TN, Mount Vernon, WA	Humid subtropical climate and cool mediterranean climate	36 months in the soil	Molecular weight	Almost 0%	[34]
	PE/PET	Outdoors in the University of Alicante	Temperature, 19.7 ± 1.5 °C	12 months in the soil	Loss of weight	Less than 2%	[35]
	LDPE	Marine station "STARESO"	Sea surface temperature, 20.8 °C	82 d in 4.5 m—water column and 8 m—sediment	Loss of weight	Less than 1%	[36]

Table 1. *Cont.*

Plastic Types		Location	Condition	Environment and Period	Indicators	Degradation	References
	LDPE	Mesodrome facilities of the University of Antwerp	Air temperature, 17.7 °C; Irradiance, 193 W/m^2	6 months in laboratory sea surface	--	Slightly change	[37]
	HDPE	Czech Republic, Pilsen Region	Precipitation, 582 mm; Temperature, 8.0 °C	36 months in landfill	Surface topography	No change	[33]
	HDPE	Compost provided by a full-scale aerobic composting plant	Temperature, 58.0 ± 2 °C	12 weeks in the composting	Loss of weight	Almost 0%	[38]
PP	PP	Outdoors in the University of Alicante	Temperature, 19.7 ± 1.5 °C; Natural rainfall, 545.1 mm	A year in the soil	Loss of weight	Less than 2%	[35]
	PP	Tambak Lorok coastal area	Temperature, 27 °C	60 d in 50 cm—sea surface area	An organic compound analyzed by EDX	3.15–16.67%,	[39]
	PP	Waste landfill	--	5 years in landfill	Surface topography	Almost 0%	[40]
PET	PET	Marine station "STARESO"	Sea surface temperature, 20.8 °C	82 d in 4.5 m—water column/ 8 m—sediment	Loss of weight	Less than 1%	[36]
	PET	Kernevel harbor	--	371 d in 1 m and 2 m depth	Surface topography	Almost 0%	[41]
PS	PS	Marine station "STARESO"	Temperature, 20.8 °C	82 d in 4.5 m—water column/8 m—sediment	Loss of weight	Less than 1%	[36]
	PS blend	Landfill	Humidity, 44%	3 months in landfill	Loss of weight	35.4%	[42]
PVC	PVC	Marine station "STARESO"	Sea surface temperature, 20.8 °C	82 d in 4.5 m—water column/ 8 m—sediment	Loss of weight	Less than 1%	[36]
		Kernevel harbor	--	371 d in 1 m and 2 m—sea	Surface topography	breaks slightly	[41]
Biodegradable plastics							
PLA	PLA	An open sea	Temperature, 30 °C	Several months	Loss of weight	Less than 10%	[43]
	PLA	Iowa State University compost site	Temperature, 55–60 °C	In 2.5 m × 1.2 m × 1.2 m —composting	Molecular weight	Reduction	[44]
	PLA	19 sites in Honshu Island, Japan	Temperature, 10 °C	12 months in the soil	Loss of weight	0–100%	[45]
	PLA > 30%	Stream, southwestern France	Temperature, 7.2 °C	77 d in the sea	Loss of weight	5%	[46]
	PLA/PHA	Knoxville	Humid subtropical climate and cool mediterranean climate	36 months in the soil	Molecular weight	20–70%	[34]
	PLA/PBAT	Washington State University	Temperature, 10.1 °C; Precipitation, 758 mm	60 months in the soil	Loss of weight	Less than 90%	[47]
	PLA/PHA and PBAT/starch	Mount Vernon, WA	Cool mediterranean climate	18 weeks in 2 m wide × 4 m long × 2 m high—composting	Molecular weight	85%–95%	[34]
PBAT	PBAT	The sea at Osaka Bay	--	6 weeks in the sea	Loss of weight	0–8%	[48]
	PBAT	Kernevel harbor	--	371 d in 1 m and 2 m depth—sea	loss of Thickness	20–40 μm	[41]
	PBAT	Bohai Bay, China	--	364 d in the sea	Loss of weight	Less than 2%	[49]
	PBAT	Bohai Bay, China	--	56 weeks in the sea	Loss of weight	Less than 2%	[50]
	PBAT/PLA	Knoxville	Humid subtropical climate and cool mediterranean climate	36 months in the soil	Molecular weight	20–80%	[34]
	PBAT/starch	Knoxville	Humid subtropical climate and cool mediterranean climate	36 months in the soil	Molecular weight	20–60%	[34]
PBS	PBS	The sea at Osaka Bay	--	6 weeks in the sea	Loss of weight	0–80%	[48]
	PBS	Bohai Bay, China	--	364 d in the sea	Loss of weight	Less than 2%	[49]
PHA	PHA	Washington State University's Research and Extension Center	Temperature, 10.1 °C; Precipitation, 758 mm	60 months in the soil	Mulch recovery	Less than 90%	[47]
PHB	PHB	SEA LIFE, Belgium	Temperature, 30 °C	43 d in the sea	Loss of weight	Almost 90%	[43]
	PHBHH	The sea at Osaka Bay	--	6 weeks in the sea	Loss of weight	20–100%	[48]

Table 1. *Cont.*

Plastic Types		Location	Condition	Environment and Period	Indicators	Degradation	References
Starch blends	Starch–PE blend	Washington State University Research and Extension Center	Temperature, 10.1 °C; Precipitation, 758 mm	60 months in the soil	Mulch recovery	Less than 90%	[47]
	Starch-based	Farm of the University of Bologna	--	3 months in the soil	Loss of weight	Almost 0%	[51]
	Starch-plastic	Agricultural experimental field, Shimane University	Temperature, 4–30 °C; Rainfall, 100–400 mm	48 weeks in the soil	Loss of weight	3.2–6.1%	[52]
	Corn starch	An aquaculture system in Rosignano Solvay	Seawater temperature, 16–25.8 °C	6 months in 0.5 m depth—shallow meadow bottom	Loss of weight	15%	[53]
	Starch	SEA LIFE, Belgium	Temperature, 30 °C	28 d in the sea	Loss of weight	More than 90%	[43]
	Starch and PCL	Czech Republic, Pilsen Region	Precipitation, 582 mm; Temperature, 8.0 °C	36 months in landfill	Surface topography	Pinholes and sized holes	[33]
	Starch	Compost provided by a full-scale aerobic composting plant	Temperature, 58.0 ± 2 °C	12—weeks in composting	Loss of weight	100%	[38]
	Starch and PP	Compost provided by a full-scale aerobic composting plant	Temperature, 58.0 ± 2 °C	12—weeks in composting	Loss of weight	99.60%	[38]
	starch-based mulch	Long Term Experimental field	--	108 d in the soil	Loss of weight	4–50%	[54]

Note: PE, polyethylene; LDPE, low-density polyethylene; HDPE, high-density polyethylene; PP, polypropylene; PET, polyethylene terephthalate; PS, polystyrene; PVC, polyvinyl chloride; PLA, polylactic acid; PBAT, Poly (butyleneadipate-co-terephthalate); PBS, poly (butylene succinate); PHA, polyhydroxyalkanoates; PHB, poly (hydroxybutyrate); PHBHH, poly (hydroxybutyrate—hydroxyhexanoate).

2.1. Conventional Plastics

Soil is a complex environmental system containing a wide range of mineral compositions, organic matter, microorganisms, and other substances [20,55]. However, conventional plastics tend to be stubborn in soil conditions at nearly all sites [56,57]. For example, PE showed little weight loss or change in the soil with an average daily temperature of 15 °C and a relative humidity of 82% for 113 d [29]. Similarly, weight loss of PE was less than 2% in the soil with an average temperature of 19.7 ± 1.5 °C for 1 year [35].

Aquatic environments have the characteristics of low temperature, low UV irradiation intensity, and low dissolved oxygen, which often hinder the degradation reactions of plastics, leading to the persistence of plastics [58,59]. In recent years, an increasing number of in-situ experiments have confirmed slow degradation of conventional plastics in real marine environments. For instance, the plastic samples such as low-density polyethylene (LDPE), PS, PET, and PVC exhibited minimal changes when exposed to either the water column or sediment for 82 d [36,60]. Vinyl polymer samples, such as PE, PP, and PS, showed only minimal changes in mechanical properties after 9 months of exposure to seawater surfaces and shorelines [61]. The PE degradation in various compartments of aquatic environments was investigated under different exposure times (33 d–20 years) [30,32,62]. PE consistently exhibited resistance to degradation in the aquatic environment after exposure to specific compartments, showing specifically either minimal surface area loss, little change in mechanical properties, or no apparent change in Raman spectra [31,62]. Interestingly, biofilms appeared on the surfaces of all PE samples from various compartments, but significant microbial biodegradation of PE was not observed [63].

Landfills are characterized by the complexity and heterogeneity of the waste mixture, variable temperature, lack of aeration and mixing, and long retention time [64]. These factors often result in extensive biodegradation processes for conventional plastics, usually lasting decades [65]. However, common conventional plastics such as PE and PP have low degradation rates in actual landfills [65,66]. For example, two types of carrier bags, i.e., high-density polyethylene (HDPE), PE, showed little degradation after 3 years in the landfill except for color changes due to the low temperature (≤20 °C) [33]. After 5 years in the municipal landfill, multiple cracks and plastic particles appeared on the surface of

the PP plastic samples, resulting in a 1.5-fold increase in crystallinity due to weathering and mechanical stress [40]. Additionally, the presence of microorganisms observed on the surface of PP implies the possibility of PP biodegradation. Both PP and PE specimens exhibited increased crystallinity and roughness with burial time, implying that the aging degree of plastics increases [66]. In summary, plastic waste in landfills exhibit only slight signs of degradation for almost a decade.

Composting is an autothermic process that proceeds through three main stages: (1) mesophilic (25–40 °C), (2) thermophilic (55–65 °C), and (3) maturation. The degradation of plastics under composting conditions has been extensively studied [67–69]. However, the composting process is usually not effective in degrading conventional plastics, and thus most research focused on degradable plastics [70,71]. For example, HDPE showed little weight loss in a full-size composting plant at 58.0 ± 2 °C for 12 weeks [38].

2.2. Biodegradable Plastics

Some biodegradable plastic polymers are marketed as potential alternatives that are degraded in soil after use without requiring additional removal and disposal [72,73]. For example, PBAT + PLA, PHA, and starch–PE blend showed less than 90% reduction in soil for 5 years [47]. The degradation rates of PLA and PHA were 70% and 20%, respectively, in 10 cm of soil for 3 years [34]. Starch plastics showed a weight loss of only 3.2% in an agricultural field from Japan for 48 weeks [52]. These results indicate that biodegradable plastics require up to 5 years or more to achieve nearly complete degradation in the soil. The biodegradability of biodegradable plastics in soil depends not only on their chemical properties but also on environmental factors such as oxygen content, pH, and microorganisms in the soil [74–76], as shown in Figure 2. Bioplastics degrade faster in soil than in water and air environments [77]. The possible reason is that a wide range of microorganisms in the soil have a significant effect on the degradation of plastics, among which *Aspergillus* spp. and *Penicillium* spp. are promising strains for biodegradation of plastics [78–80].

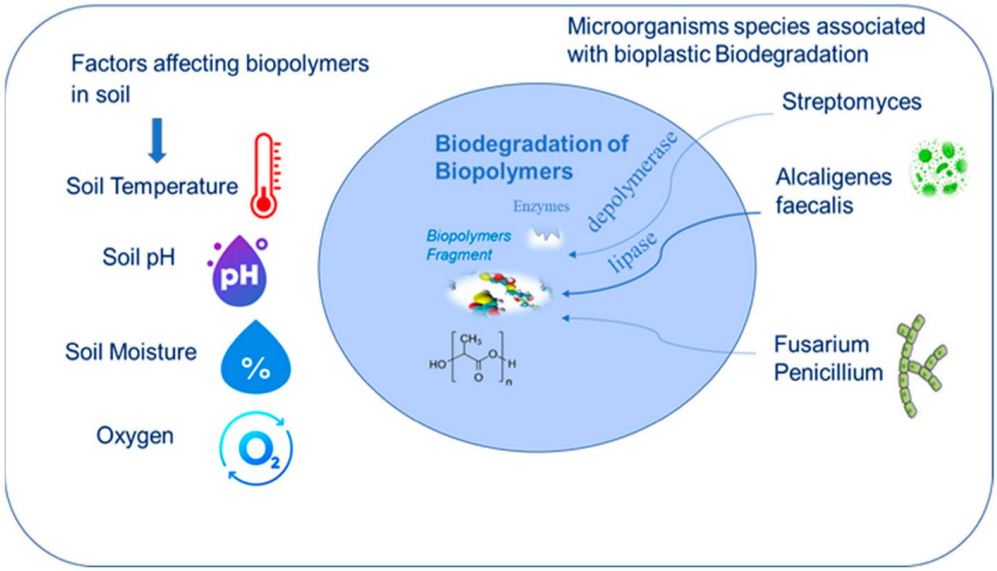

Figure 2. Interactions between microorganisms and environmental factors in MPs contaminated soils. Reproduced with permission from ref. [81].

Degradable plastics may take longer to fully degrade in aquatic environments compared to soil. For example, the degradation rate of starch-based bioplastics in the marine environment was only 1.5% for 90 days [82]. PLA degradation was slower than PHA and starch-based bioplastics in seawater environments at 25 °C, and the average estimated time for complete degradation of PLA exceeded ten years [82,83]. In addition to the proper-

ties of bioplastics, temperature, pH, nutrient content, microbial population density, and diversity are important factors influencing the degradation of bioplastics in aquatic environments [81,84,85]. Notably, photo-oxidative degradation occurs near the surface of the water (light zone), and biodegradation plays a dominant role in plastic degradation as the depth from the water increases [39,85,86]. Figure 3 depicts the degradation behavior of bioplastics in the bottom aquatic environments [81]. Thus, the regions of water had a significant impact on the breakdown of biodegradable plastics [87]. For example, PHA samples were almost intact after 22 months of incubation in the pelagic Mediterranean Sea, whereas the same samples were fully degraded in 3 months in the bottom waters of Southeast Asia.

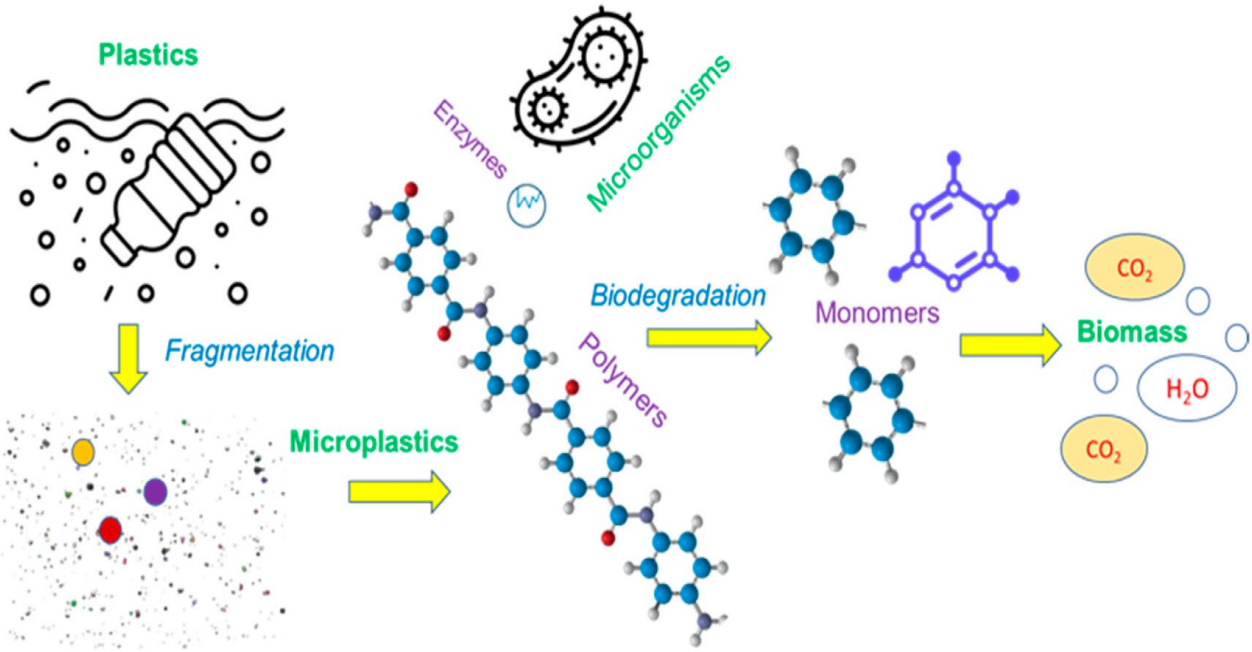

Figure 3. Bioplastic degradation behavior in aquatic environments. Reproduced with permission from ref. [81].

The degradation of biodegradable plastics was widely investigated in a landfill. For example, pinholes and sized holes appeared on the surface of the mixture (starch/PCL) in the landfill for 3 years [33]. PLA showed changes in color and mechanical strength and a 4.14% loss in mass after being buried on the surface of a natural landfill for 6 months [88]. Biodegradable plastics have a low degradation rate in actual landfills. Temperature, humidity, pH, and microorganisms have important influences on the degradation of biodegradable plastics in landfills, especially for microorganisms [77,89,90]. As a result, many studies have been conducted to isolate bacteria or fungi that have the ability to degrade plastics in landfills [89,91]. Actinobacteria, as a major bacterial taxon including *Streptomyces*, *Nocardia*, and *Rhodococcus*, were capable of degrading PHAs [90,92]. Fungi from landfills were available for degrading plastics, such as *Aspergillus* spp., including *A. nomius*, *A. carbonarius*, *A. fumiates*, and *A. terreus* [93,94]. In general, biodegradable plastics have low degradability in actual landfills, and microorganisms isolated from landfills have the potential for efficient biodegradation of plastics.

Composting is divided into industrial composting (large-scale, professionally managed) and household composting (small-scale) according to the control conditions [69,95]. Generally, industrial composting is a crucial method for managing biodegradable plastic waste, with a thermophilic temperature of 50–60 °C. However, home composting offers a sustainable alternative without the need to collect and transport the waste, with temperature spikes of less than 35–40 °C [96,97]. Biodegradable plastics were often designed to be compostable, which was verified through actual industrial composting studies [67–69].

For instance, PLA bioplastics were completely degraded by industrial composting in 4–6 weeks [98]. PHBV loses 59% of its mass for 186 days at 40–63 °C [99]. However, the biodegradability of plastics may not be suitable for home composting due to their relatively low temperatures inhibiting the activity of thermophilic bacteria [97]. For instance, the mechanical properties of the PLA film deteriorated, whereas the PLA film was not decomposed in a home/farm composting with a maximum temperature of about 42 °C for 7 weeks [100]. However, biodegradable plastics can meet the requirements of standard AS 5810 during home composting through enhancing thermophilic temperatures [29]. For example, commercial compostable bioplastic bags (starch/polyester) were decomposed after being incubated at elevated temperatures in three static home composters for six months, with degradation rates ranging from 90 to 96% [101,102]. In addition to temperature, the degradability of biodegradable plastics varies also with the plastic content in the compost, plastic properties such as chemical composition and crystallinity, and composting conditions such as carbon to nitrogen ratio and inoculum [8,97,103]. In summary, biodegradable plastics show excellent degradation in industrial composting. The biodegradability of biodegradable plastics is improved by raising the thermophilic temperature during home composting, which is a potential, feasible, and cost-effective alternative treatment technology [104].

3. Enhanced Methods for Plastics Degradation

Both conventional and biodegradable plastics exhibit limited degradation rates in real environments, besides the favorable degradation performance of biodegradable plastics under composting conditions. Therefore, various physical, chemical, and biological methods are used to enhance the degradation properties of plastics.

3.1. Physical Methods

3.1.1. Photocatalysis

Photocatalysis is an effective method to enhance the degradation efficiency of plastics through the addition of a photocatalyst to plastics. Catalytic additives tend to absorb energy from light radiation, such as ultraviolet light, sunlight, and fluorescent lamps, inducing generated holes and electrons to drive redox reactions. Depending on the reaction products of photocatalytic plastics, the above pathways are mainly divided into photooxidation (PD) and photoregeneration (PR).

During the PD process, the reaction of holes and electrons under O_2 conditions produces highly oxidizing radical oxygen such as hydroxyl radical (•OH), superoxide anion ($O_2\bullet^-$), monoclinic oxygen ($^1O_2$), and H_2O_2, which non-selectively oxidizes the plastic into plastic fragments or mineralizes it into carbon dioxide and water. For PD catalysts, semiconductors (e.g., TiO_2, ZnO, $NiAl_2O_4$, and CdSe) and metal catalysts (e.g., Au, Pb, and Fe) are predominantly used. For example, the addition of a TiO_2 catalyst to PVC increased the mass loss by 5.56% under UV irradiation for 30 h, compared to the addition of no catalyst [105]. The addition of Au and Pd nanoparticle catalysts to LDPE improved the degradation efficiency by 55.8% under UV irradiation for 240 h, compared to pure LDPE. Therefore, PD achieves efficient degradation of waste plastics under the condition of light and catalyst, and the direct use of solar energy is also more energy-saving and economical. However, the sustainable photocatalyst in PD remains in the environment and poses a risk to the environment [106].

PR is a method of oxidizing plastic substrates to valuable chemicals and reducing water to fuel-grade H_2 by controlling ambient pressure and temperature using sunlight and photocatalysts [107–109]. The development of PR technology will help to reduce the emission of CO_2 into the environment [110]. PR of plastics was initially proposed in 1981 using Pt-coated TiO_2 as a photocatalyst in aqueous NaOH solution (5 M) with moderate hydrogen precipitation rate. In recent years, several PR catalysts have been prepared, such as $ZnIn_2S_4$, MOS_2/CdS, RP@CoxPy/$Cd_{0.5}Zn_{0.5}S$, and SiC-g-C_3N_4 composites [107,111–113]. For example, plastic wastes such as PLA, PET, and PE were converted into a number of useful

compounds by PR using MOS_2-modified CdS-based catalysts with sequential precipitation of H_2 [112]. The PR reaction of PET and bisphenol A produced 18 and 12 $\mu molH_2/g_{cat} \cdot h$, respectively, using SiC-g-C_3N_4 composite photocatalyst [113]. A small amount of carbon nitride was added to SiC to make SiC-g-C_3N_4 have a good interaction and promote the H_2 precipitation of the PR reactions. In short, PR transforms waste plastics into high-value-added products and fuels. However, PR is subject to certain limitations, such as harsh reaction conditions, low hydrogen yield, and complex product collection [8,114].

3.1.2. Mechanical Degradation

Mechanical degradation in the environment refers to the breakdown of plastics into small particles due to physical forces such as wave and light action in the ocean, friction from wind, and sand in the air [115,116]. Plastics are more susceptible to rupture when subjected to low-amplitude forces such as friction and impact in the environment [117]. The sand grains combining UV make plastics prone to degradation and fragmentation [118]. However, UV irradiation causes cracks in the surface of the plastic but not outright fragmentation in the absence of mechanical abrasion by sand grains [24]. Plastics crack and tend to fragment after subjected to mechanical forces such as cutting, impact, and abrasion, and large plastic polymers degrade into smaller molecules [119]. Severe mechanical grinding conditions may initiate thermal degradation [120]. The degradation is carried out by mechanical forces generated by tools such as ball mills, vibratory mills, cryogenic extruders, hot and cold masticators, jet mills, disc mills, and wet mills [119,120]. Additionally, ultrasound has shown promise for degrading plastics through generating mechanical stress to shear polymer molecules and turn large molecules into small ones [121]. In general, the advantage of mechanical degradation techniques was that they were simple to operate. However, only the fragmentation of plastics is achieved [119]. Therefore, it is better to use it as a pretreatment for other plastic degradation technologies.

3.1.3. Pyrolysis

Pyrolysis is a process in which polymer molecules undergo chain-breaking degradation and crosslinking reactions due to the acquisition of activation energy required for chain-breaking at temperatures between 300 and 900 °C [121,122]. This process eventually leads to the deterioration of polymer properties and the formation of low-molecular substances. Waste plastics are converted into fuels, hydrogen-rich syngas, and liquid fuels through the pyrolysis process. Therefore, this method is considered an efficient treatment for enhancing plastic degradation [123]. However, the pyrolysis process requires high heat and advanced equipment, limiting its application for plastic degradation. Consequently, it deserves to reduce the reaction temperature of the pyrolysis [122]. LDPE was thermally degraded at 406–550 °C, mainly producing saturated alkanes and mono(di)-alkanes. The pyrolysis temperature was reduced to 370–430 °C by a heating combination of dry distillation combined with irradiation, thereby reducing energy consumption and equipment requirements [124,125].

Catalysts effectively reduce the pyrolysis temperature and increase the rate of plastic degradation, but they also make the recovery of monomers and closed-loop recycling of plastics possible [126]. The array of pyrolysis catalysts encompasses acidic solid catalysts, ionic liquid catalysts, organocatalysts, metal-loaded catalysts (Fe, Co, and Ni), and biomass (eucalyptus, corn cob) [127–129]. Meanwhile, the pyrolysis catalysts have the potential for reuse [130,131]. The ionic liquid catalyst [HO_3S–$(CH_2)_4$-mim] Cl–$FeCl_3$ was utilized to degrade PET at 190 °C, resulting in 98.7% degradation of PET after 6 h, and the catalytic activity can last for more than 6 cycles [132]. An organocatalyst (tetramethylammonium methyl carbonate) was reported to effectively degrade PLA, PCL, and PET, as well as to repolymerize dimethyl terephthalate into PET, demonstrating a complete chemical recycling process [133]. In summary, pyrolysis can not only achieve efficient degradation of plastics but also effectively convert plastics into fuel. However, harsh high-temperature conditions

and equipment requirements result in higher economic costs, limiting its application in plastic degradation [124,125].

3.2. Chemical Methods

Chemical degradation of plastics occurs when they come into contact or mix with chemical media, such as aqueous solutions, alcohols, amines, strong oxidizing agents, and supercritical fluids (SCFs). Eventually, plastics may degrade into oligomers, dimers, monomers, or value-added products with high recycling and reuse values [5]. Chemical methods of plastics mainly include hydrolysis, alcoholysis, aminolysis, strong oxidation, and SCFs.

3.2.1. Hydrolysis

Hydrolysis is the complete hydrolysis of covalent bonds, especially ester bonds, by contacting the polymer with water at high temperatures and pressures [134,135]. Hydrolysis of plastics such as PET and PLA in aqueous solution is a well-established and simple depolymerization process that allows the recovery of TA and ethylene glycol in high yields (>80%) [134,136]. As shown in Figure 4, hydrolysis of plastics can be carried out under neutral, acidic, and alkaline conditions [134,137–139]. Inorganic acids, such as sulfuric, nitric, and phosphoric acids, can accelerate the hydrolysis of PET [140]. Sulfuric acid is one of the most commonly used acids. For instance, PET degraded completely in the presence of 3 mol/L sulfuric acid at 190 °C for 1 h [141]. However, acid corrosion and disposal are also worthy of attention. Neutral hydrolysis of PET typically requires a temperature of 200–300 °C. The addition of an alkaline regulator such as NaOH and a co-solvent like ethanol reduced the hydrolysis temperature to 80 °C and enhanced the conversion rate to 95% for 20 min [142,143]. Similarly, PLA hydrolysis occurred slowly at 40 °C, with only 0.5% conversion to lactic acid for 120 d [144]. However, 95% of PLA was hydrolyzed at 160–180 °C for 120 min, indicating that high temperature is a favorable factor to promote the hydrolysis and degradation of PLA [145]. In addition, ultrasonic, microwave irradiation, and catalysts have been reported to accelerate PET hydrolysis [146,147]. Therefore, the hydrolysis and degradation of plastics are enhanced by regulating hydrolysis conditions like pH and temperature, as well as implementing auxiliary technologies such as ultrasonic, microwave irradiation, and catalysts. In summary, hydrolysis realizes efficient recycling of plastics in the process of plastic depolymerization. However, the technology needs to be further improved, such as by reducing harsh reaction conditions, inhibiting the generation of side reactions, and improving the purification and recovery of hydrolyzed monomers [135,148].

3.2.2. Alcoholysis

Alcoholysis is a method for degrading polymers through transesterification reactions, where alcohols react with ester bonds in polymer molecules [149]. These alcohols include methanol, ethanol, and ethylene glycol. Specifically, the use of ethylene glycol in alcoholysis is also known as glycolysis, but collectively referred to as alcoholysis in this review. Undoubtedly, alcoholysis is a valuable method to control the degradation of plastics, allowing recyclable monomers or high-value-added products to be obtained while degrading plastics. Typically, alcoholysis of polyesters is slow but can be greatly accelerated through the use of catalysts and/or reinforcement methods (e.g., microwave irradiation, supercritical state), achieving a conversion rate close to 100%. Alcoholysis catalysts are diverse, including metal salts (e.g., $FeCl_3$ and sodium metasilicate), transition-metal-substituted polyoxometalate, tetrabutyl orthotitanate, inorganic nanoparticles (CeO_2 NPs), and ionic liquids (e.g., Brönsted–Lewis acidic IL) [150–153]. With the presence of a catalyst, PET and PLA were nearly completely decomposed by alcoholysis at 190 °C and 130 °C for 80 min, respectively. This process yielded valuable products, including bis (hydroxyethyl) terephthalate (yield, 84.42%) and alkyl lactate (conversion, 97%) [154]. In addition, microwave irradiation can also improve the PLA alcoholysis to a certain extent

because of increased reaction efficiency and sensitivity [155]. Despite alcoholysis promise for polyester degradation and monomer recovery, it still faces various challenges, including harsh reaction conditions, difficult purification of monomers, and challenging catalyst regeneration [156,157].

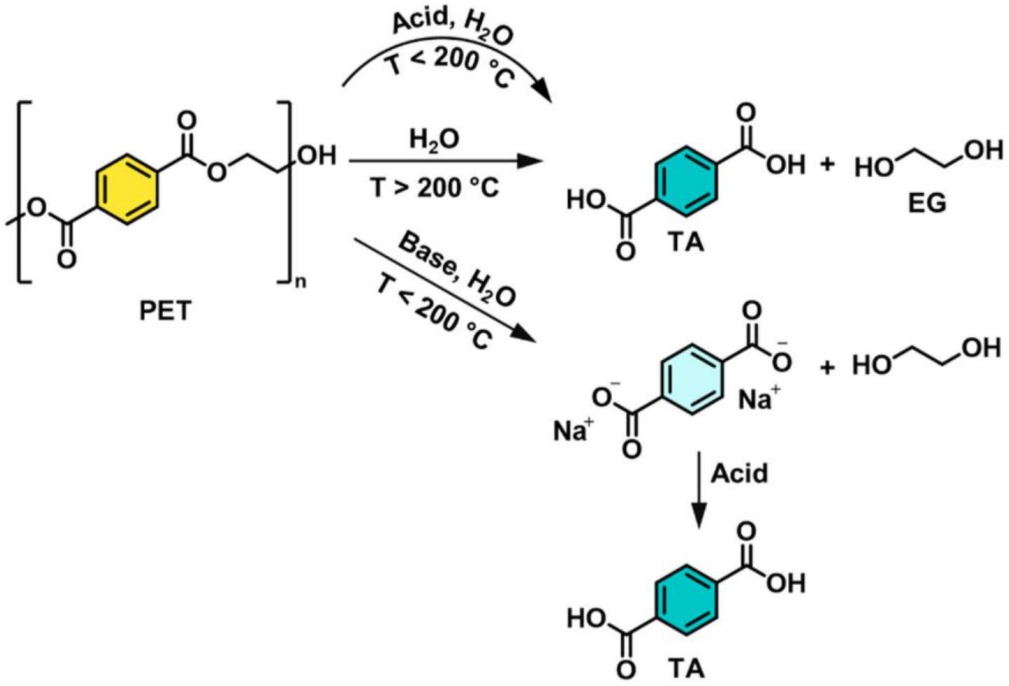

Figure 4. Hydrolysis of PET under acidic, neutral, or basic conditions. Reproduced with permission from ref. [134].

3.2.3. Aminolysis

Similar to alcoholysis, aminolysis is a method for degrading polyesters that utilizes the strong nucleophilic amino group of an amine molecule to cleave ester bonds. So far, there are only few studies on the aminolysis of plastics, mainly focusing on PET aminolysis [158]. PET can undergo aminolysis in the presence of various amines such as methylamine, ethylamine, diamine, and alcoholamine. The resulting products are corresponding amides or amines of terephthalic acid, which are used as plasticizers and modifiers [159,160]. PET was completely consumed through aminolysis with an ethanolamine and PET mass ratio of 1:3 at nearly 140 °C for 8 h [160]. The mixture of diamine and PET was heated at 130 °C for 24 h to obtain a water-soluble amide, which is condensed with salicylaldehyde to form a Schiff base [161]. In addition, catalyst also plays a significant role in the aminolysis reaction of PET, which is manifested in the reduction of reaction temperature and reaction time, or/and the increase of product yield. For instance, PET can react with hydrazine monohydrate for 3 h at 65 °C using sodium carbonate as the catalyst, resulting in an 84% yield of the aminolysis product (terephthalic dihydrazide) [162]. Similarly, using ionic liquids as catalysts, PET was aminolyzed with hydrazine hydrate at 110 °C for 60 min, and the yield of terephthalic hydrazide was 84%. In addition, glacial acetic acid and its sodium salts and potassium sulfate are also used as aminolysis catalysts. Overall, like hydrolysis and alcoholysis, aminolysis achieves polyester degradation and monomer recovery, but it is also limited by poor reaction conditions and difficult purification of monomers [160]. Notably, there are fewer studies on polyester aminolysis, and it has fewer applications on an industrial scale [163]. This may be attributed to the fact that amines are irritating and toxic and can be harmful to the human body, limiting its application.

3.2.4. Strong Oxidation

The strong oxidation process involves the addition of a strong oxidant that generates reactive oxygen species (ROS) for oxidation reactions when activated by light, heat, and transition metal catalysts. ROS has strong oxidizing ability and reactivity, causing chemical bond breakage of plastics and accelerating the oxidative degradation [164]. These ROS include $O_2\bullet^-$, $^1O_2$, •OH, and sulfate radical ($SO_4{}^-\bullet$) [165–167]. The common strong oxidants include O_3, Fenton's reagent, and persulfates. For example, UV activation of persulfate hastened the degradation of PVC, leading to a 20% decrease in average particle size and the generation of small molecules such as alcohols and carboxylic acids for 35 h [167]. Likewise, the PS experienced cracking and exhibited prominent wrinkles on the surface after 7 d of exposure to Fenton's reagent [165]. Although the strong oxidizer can effectively degrade waste plastics, the degradation efficiency of waste plastics is low, and the recycling of plastics cannot be realized [168].

3.2.5. Supercritical Fluids (SCFs)

SCFs are the state of a substance at conditions that exceed its critical pressure and temperature. SCFs have intermediate properties between those of a gas and a liquid, with molecular movement speed similar to that of a gas and density similar to that of a liquid, demonstrating significant potential for enhancing plastic degradation [169]. Solvent dissolution efficiency is improved when plastics are dissolved in a supercritical solvent [170]. This leads to a decrease in the reaction time and temperature and an increase in the conversion rate of waste. Common SCFs for degrading plastics encompass supercritical water, supercritical alcohol, and other supercritical organic solvents, such as ketones and aromatic compounds [171,172].

Polymer decomposition occurs rapidly and selectively in SCFs. Condensed plastics such as PET, nylon, and polyurethane are relatively easy to depolymerize into monomers in supercritical water or alcohols. The monomer components are recovered in high yields [173]. For example, PET was completely degraded in supercritical water at 275 °C and 150 MPa for 90 min [174]. The primary degradation products included diethyl terephthalate, ethylene glycol, and terephthalic acid. The yield of terephthalic acid reached 100% after the depolymerization of PET in supercritical water (400 °C/40 MPa) for 30 min [171]. Addition polymerized plastics such as phenol resins, epoxy resins, and PE are also decomposed into monomer fractions using SCFs with or without a catalyst [173]. Addition polymers are usually susceptible to form monomers in supercritical organic solvents and oligomers or small molecules in supercritical water. For instance, PS degraded at a rate of approximately 96% in supercritical ethyl cyclohexane at 400 °C for 1 h, with a styrene yield of 95% [175]. The carbon conversion rates of PS were 25% and 47.6% in supercritical water and supercritical water/CO_2 mixed media, respectively, at 700 °C for 20 min [176]. Undoubtedly, SCFs have the capability of enhancing plastic degradation and recycling. However, the SCFs for plastic degradation are still in an early stage due to harsh reaction conditions (high temperature and pressure) and equipment requirements [177].

3.3. Biological Methods

A wide range of organisms, such as microorganisms, small animals, and algae, have the ability to degrade plastics. This has sparked the interest of researchers in the ways in which these organisms can further enhance plastic degradability.

3.3.1. Microorganisms

Microorganisms promote the degradation of plastics mainly through the active participation of intra- and extracellular enzymes, leading to the mineralization of plastics [178,179]. These microorganisms are widely present in soil, aquatic environment, landfills, and compost. A total of 19 genera of bacteria are reported to have the capability for PE degradation, while 12 genera of fungi for conventional plastics [77]. More than 90 species of microorganisms are capable of degrading bioplastics. For example, plastics were effectively degraded

by actinomycetes and fungi isolated from landfills, with up to 39% of weight loss. Microorganisms such as *Exiguobacterium* sp., *Halomonas* sp., and *Ochrobactrum* sp. were isolated from more than 300 bay samples in China and showed significant degradation of PET and PE films. Selective in situ incubation of efficient plastic-degrading microorganisms is also an effective way to enhance plastic degradation [180]. Native microorganisms readily adapt to plastic pollutants and eventually degrade them since these plastics are present in the environment as pollutants for long periods of time [180]. The application of bacterial consortia is more effective in degrading plastics than any individual strain, since undesirable effects help to be eliminated through using the metabolites of one microorganism as a substrate for the growth of another in bacterial consortia. Bacterial consortia compensate for the deficiencies of single microorganisms to a large extent [181,182]. For example, *Souda* and *Agios* colonies led to 18% of PS loss after 2 months, which is higher than *Agios* colonies alone [183]. Therefore, artificially isolated and cultured microorganisms can enhance the degradation of plastics, and selective in situ incubation and bacterial consortia are ways to further enhance plastic microbial degradation. Microbial technology enables partial mineralization of waste plastics. However, microbial plastic degradation is a slow and time-consuming process compared to physical and chemical techniques [184]. The current study uses specific microbes to degrade and mineralize specific plastics. Microbial degradation of mixed types of plastics should be systematically studied in the future to improve their industrial feasibility [185].

3.3.2. Small Animals

Many small animals, such as insects (e.g., *Tenebrio molitor*, *Zophobas atratus*, *Plodia interpunctella*, *Tribolium castaneum*, *Galleria mellonella*), worms, fish, earthworms, and snails, are found to consume and degrade plastics [68,78]. For instance, Larvae of *Tenebrio molitor* from three locations, i.e., Beijing and Harbin, China, and Compton, California, USA, were reported to consume 23–31% of PS in a month [186]. As shown in Figure 5A,B, nearly 92 mg of plastics was consumed for 12 h after approximately 100 waxworms were placed in a PE shopping bag [187]. The mass loss of PE was higher by 13% than the control group without waxworm (Figure 5C) [187]. The researchers broke up the worms and smeared them in plastic to check whether the worms simply chewed the plastic or swallowed it, and found that about 13% of the plastic had disappeared for 14 h [188]. This suggests that certain compounds in the worms' digestive system can digest plastic [189,190]. *Enterobacter asburiae* and *Bacillus* sp. isolated from the intestines of wax worms (*Plodia interpunctella*) were able to degrade PE [191]. A bacterial strain, *Exiguobacterium* sp. strain YT2, isolated from the mealworm gut caused the PS weight loss of 7.4 $\pm$ 0.4% for 60 d [192]. The results also indicated that gut bacteria play a vital role in PS biodegradation and mineralization [188,190]. A large number of gut microbiomes such as *Citrobacter* sp., *Enterobacter* sp., and *Kluyvera* have been further isolated from insects and are effective in plastic degradation [193]. Therefore, the ability of small animals to degrade plastic is largely due to the role of their gut microbes. The advantage of using small animals to degrade plastics over microorganisms is that these invertebrates are readily available in nature [185]. However, the rate of degradation is slow compared to physical and chemical techniques. The current research on this technology has only been conducted for PS and PE, which is also limited. In the future, it should be expanded to include PVC, PET, PP, and other plastics [194].

3.3.3. Algae

Microalgae colonize on the surface of plastics, and induce plastic removal through precipitation, aggregation, adsorption, and enzyme pathways in the presence of sunlight, nutrients, and water [195–197]. Microalgal polysaccharides are essential for the vertical transport of PP-MPs from the water surface to the sediment, providing a possible pathway for MPs removal [198]. More than 12 groups of plastic-degrading algae have been found on the surface of plastic debris in marine and freshwater [199–201]. The microalga *Uronema*

Africanum Borge was reported to initiate the degradation of LDPE in 30 d [200]. Blue-green alga *Anabaena spiroide*, green algae *Scenedesmus dimorphus*, and diatom *Navicula pupula* caused the weight loss of 8.18%, 3.74%, and 4.44%, respectively, for LDPE sheets for 30 d [201]. In addition, many cyanobacterial strains were also capable of generating biofilms by photosynthesis on plastic surfaces and providing an important habitat for some plastic-degrading microorganisms [195,202]. In general, the growth environment of algae technology is simple, energy-saving and economic. However, the efficiency of algae to degrade plastic is low, and the growth of algae is easily affected by the concentration of plastic [203].

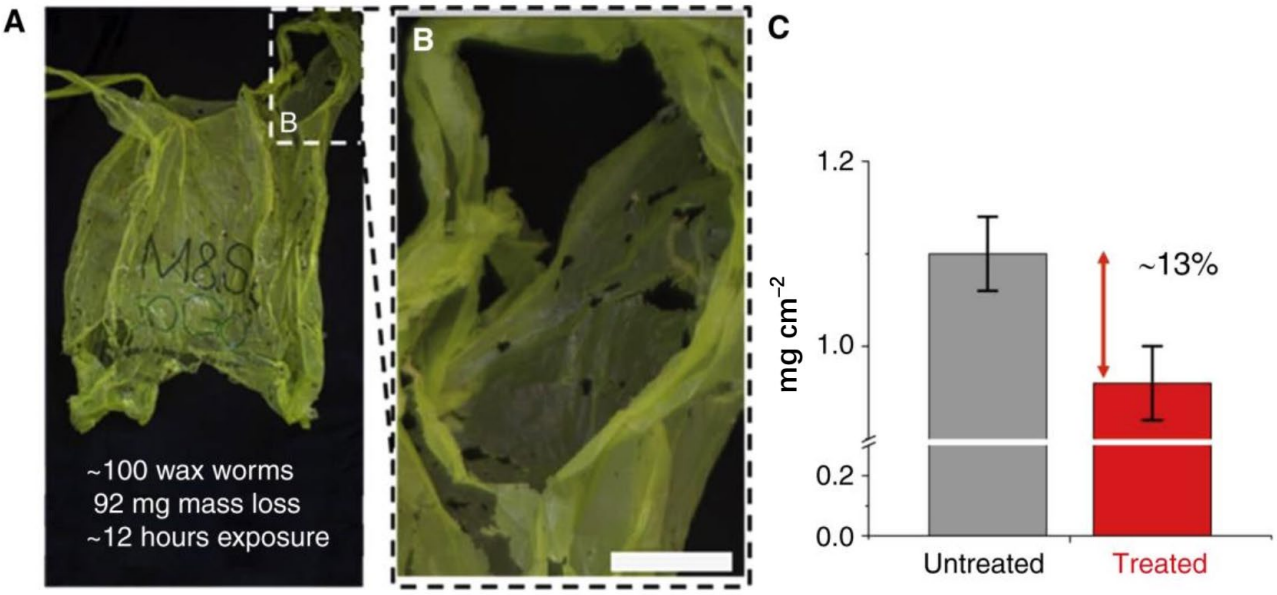

Figure 5. (**A**,**B**) Degradation of polyethylene (PE) by mellonella in a plastic bag after exposure to approximately 100 waxworms for 12 h. (**C**) Weight analysis of homogeneously treated and untreated PE. Reproduced with permission from ref. [187].

Recently, enzymes necessary for the breakdown of plastics are produced by genetic engineering (mutation) techniques using algae as chassis cells based on synthetic biology [204,205]. For example, the microalga *Chlamydomonas reinhardtii* has been genetically engineered to produce a PET hydrolytic enzyme, which can degrade plastic films composed of terephthalic acid [206,207]. Considering the simplicity of microalgal production and their ability to produce enzymes for dissolving plastics, microalgae may be used to achieve enhanced degradation of plastics.

4. Conclusions and Future Perspectives

Conventional plastics such as PE, PP, PET, PS, and PVC have low weight loss and little change in mechanical properties in soil, aquatic environments, landfills, and compost, but the crystallinity and roughness on the plastic surface may increase with burial time. Complete degradation of biodegradable plastics in soil environments usually takes at least five years, while in aquatic environments it may often take longer. Biodegradable plastics have a low degradation capacity in actual landfills but show considerable degradability under industrial composting conditions due to the high thermophilic temperature.

PD, mechanical techniques, and strong oxidation only facilitate the degradation of plastics into plastic fragments and thus are often used as pre-treatment techniques for plastic degradation. Meanwhile, PR and pyrolysis can convert plastics into fuels such as H_2 and hydrogen-rich syngas, while hydrolysis, alcoholysis, aminolysis, and SCFs can degrade plastics into monomers for recycling. However, the techniques often require stringent equipment, large quantities of chemical reagents, and harsh reaction conditions, such as

high temperature and pressure. In addition, plastics are also degraded by microorganisms (e.g., actinomycetes, *Exiguobacterium* sp., and fungi), small animals (e.g., waxworms, worms, and earthworms), and microalgae (e.g., cyanobacteria, green algae, and diatoms). The plastics biodegradation is further enhanced by microbial in situ cultures, bacterial communities, small animal gut microbes, and enzyme-producing microalgae.

Research should prioritize the following areas to enhance technologies for the plastic degradation:

(1) The degradation of conventional and biodegradable plastics in real environments is often complex and variable. For example, the degradation of plastics is affected by the characteristics of the plastic, the medium environment, and the region. Therefore, establishing an equivalence standard is required to accurately calculate and measure the degradability of specific plastics in various real environments. This will help to understand the accumulation of plastics in real environments and their potential ecological risk.

(2) This paper reviews external control techniques for enhancing degradation of plastics. Future research is suggested to further improve the plastic degradation through changing the polymer composition at polymer and molecular levels. For example, exogenous additives that promote plastic degradation, such as enzyme additives, are added inside the plastic. Alternatively, degradable plastics are prepared by introducing degradable groups into traditional plastic molecules through molecular techniques.

(3) Recycling of plastics is achieved by chemical technologies such as hydrolysis, alcoholysis, and aminolysis. However, many by-products are generated during plastics degradation through these technologies, resulting in a decrease in plastics recovery. In the future, more attention should be paid to the precise control of product recovery. For example, controlled recovery of a target product is achieved by controlling specific conditions (e.g., temperature, catalyst) to produce the target product.

(4) Degradation products of plastics often include MPs, plastic monomers, and additives in the environment, which are potentially harmful to the environment. Therefore, the impacts of plastic degradation products on the environment should be further controlled or eliminated by reducing the generation of risky products and recycling the products.

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