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Review

Plastic biodegradation: Frontline microbes and their enzymes

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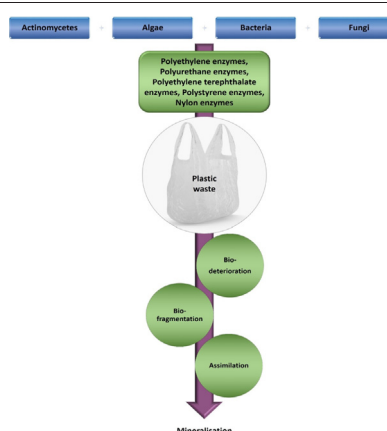
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HIGHLIGHTS

- Role of actinomycetes, algae, bacteria and fungi in plastic biodegradation assessed.
- Biodegradation involves biodeterioration, fragmentation, assimilation and mineralisation.
- PE, PU, PET, PS and nylon enzymes are major groups involved in plastic biodegradation.
- Microbial depolymerases, hydrolases, and peroxidases are the key enzymes in biodegradation.
- Polymer attributes, environmental conditions and chemicals are critical contributing factors.

GRAPHICAL ABSTRACT



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ABSTRACT

Plastic polymers with different properties have been developed in the last 150 years to replace materials such as wood, glass and metals across various applications. Nevertheless, the distinct properties which make plastic desirable for our daily use also threaten our planet's sustainability. Plastics are resilient, non-reactive and most importantly, non-biodegradable. Hence, there has been an exponential increase in plastic waste generation, which has since been recognised as a global environmental threat. Plastic wastes have adversely affected life on earth, primarily through their undesirable accumulation in landfills, leaching into the soil, increased greenhouse gas emission, etc. Even more damaging is their impact on the aquatic ecosystems as they cause entanglement, ingestion and intestinal blockage in aquatic animals. Furthermore, plastics, especially in the microplastic form, have also been found to interfere with chemical interaction between marine organisms, to cause intrinsic toxicity by leaching, and by absorbing persistent organic contaminants as well as pathogens. The current methods for eliminating these wastes (incineration, landfilling, and recycling) come at massive costs, are unsustainable, and put more burden on our environment. Thus, recent focus has been placed more on the potential of biological systems to degrade synthetic plastics. In this regard, some insects, bacteria and fungi have been shown to ingest these polymers and convert them into environmentally friendly carbon compounds. Hence, in the light of recent literature, this review emphasises the multifaceted roles played by microorganisms in this process. The current understanding of the roles played by actinomycetes, algae, bacteria, fungi and their enzymes in enhancing the degradation of synthetic plastics are reviewed, with special focus on their modes of action and probable enzymatic mechanisms. Besides, key areas for further exploration, such as the manipulation of microorganisms through molecular cloning, modification of enzymatic characteristics and metabolic pathway design, are also highlighted.

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1. Introduction

Microorganisms play important roles in the maintenance of many environmental processes, as they have evolved over millennia to transform and to mineralise different compounds including xenobiotics. They have been at the forefront of preventing the bioaccumulation of various materials as they consume these substances and recycle them into compounds that can be reutilised by nature. Hence, microbial communities tackle several environmental challenges by evolving their metabolic capacity through genome modification to allow the incorporation of new compounds into their metabolic pathways and by extension into the biogeochemical cycles. Thus, the ability of microbes to adapt to the metabolism of different anthropogenic compounds have been noted to be based on the natural selection of mutants possessing the necessary degradative enzymes with less specific substrate-specificities and probably novel metabolic pathways. However, the indiscriminate human exploitation of natural resources has generated unprecedented disturbances in nature through the introduction of xenobiotics at a faster rate than the adaptation and evolution of the microbes. As a result, the self-cleaning capacity of the ecosystems is overwhelmed and the accumulation of pollutants to problematic levels has become the norm. Top on the list of these ever-accumulating pollutants is the synthetic plastic which are human-made polymers derived from petroleum. In the last fifty years, plastic materials have grown to become indispensable in all aspects of human endeavours, replacing materials such as glass, metals and wood in different applications as a result of their low cost, durability and high strength. Hence, global plastic production has tripled in the last twenty-five years (Feil and Pretz, 2020). However, a majority of the estimated 8.3 billion virgin plastics produced so far are single-use convenience products that have ended up discarded in our natural environments (Nielsen et al., 2020). This has adversely affected life on land through their undesirable accumulation in landfills, leaching into the soil, and increased greenhouse emission. Recent investigations have shown the detrimental effects of plastics on the activity and diversity of soil microbiota, on reproduction

in soil organisms (Lahive et al., 2019) and leaching in soil invertebrates (Selonen et al., 2020). The damage done by plastic wastes to the aquatic environment is also as disturbing and has been shown to include the disruption of marine animals' endocrine system (Jung et al., 2020), intestinal blockage and false sensation of satiety in aquatic animals (Paço et al., 2019). Furthermore, they have also been found to interfere with chemical communication in aquatic ecosystems, to cause intrinsic toxicity by leaching, and by absorbing persistent organic contaminants as well as pathogens (Barceló and Picó, 2019). In recent times, microplastics, which are plastic particles less than 5 mm in size, have gained more attention, as they have been found causing irreparable damage in different ecosystems (Lwanga et al., 2017). Microplastics have also been noted to agglomerate toxic contaminants in water, including heavy metals and organic pollutants (Wang et al., 2019). The probability thus exists for plastics and the accumulated toxicants to enter various food chains (terrestrial and aquatic) and eventually find their way to the human body through the trophic transfer of microplastics, thus posing several potential health challenges (Lwanga et al., 2017). Industrial plastic manufacturing has also raised many environmental concerns, especially regarding the release of microplastics into the atmosphere and water systems. Many microplastics are generated from industrial precursors such as pellets, spherules, granules, discs and other plastic raw materials (Lechner and Ramler, 2015). Other detrimental wastes generated from industrial plants into the environment include mother liquors, organic halogenated solvents, washing liquids, waste hydraulic oils, to mention a few (Öncel et al., 2017).

In all of these, the bioaccumulation of plastic polymers in the ecosystems is a result of their inherent characteristics which hinder their disintegration, especially their high molecular weight and high crystallinity. The absence of favourable functional groups needed for oxidative reactions has also been identified as another hindrance since plastics are highly hydrophobic with stable functional groups such as alkane and phenyl (Devi et al., 2016). Current approaches aimed at mitigating their effects in the environment include incineration, recycling, landfilling, and

the emergent use of bioplastics. However, each of these methods comes with its demerits. For instance, incineration of different plastic polymers leads to the production of more toxic and volatile wastes such as furans, dioxins, heavy metals, sulphides which are all considered as potential carcinogens (Verma et al., 2016). The economy of plastic recycling has also been noted to be cost-ineffective as recycled plastics are more expensive than virgin plastic products (Gradus et al., 2017). Furthermore, “down-cycling” has been noted to be another undesirable consequence of recycling as recycled products are usually of lower value and functionality compared to the virgin products (da Silva and Gouveia, 2020). Thus, the application of biological systems as efficient alternatives to biodegrade these recalcitrant polymers has been a significant focus of scientific inquiry in the recent past. The primary mechanism behind the biodegradation of high molecular weight polymers such as plastic has been noted to be the depolymerisation of these polymer chains by enzymes into intermediates with modified properties, increasing their accessibility for cellular assimilation (Zhang et al., 2020). Different organisms, both higher and lower, capable of converting the plastic polymers into simple molecules like CO₂ and H₂O have been identified, with most of the attention being focused on insects with less emphasis on microorganisms. Insects in their larva form including the mealworm, superworm and waxworm, have been demonstrated with the ability to eat, degrade and mineralise various plastic polymers, albeit with the help of their gut microbiome (Zhang et al., 2020). In addition to the symbiotic microbes working in association with insects to degrade plastics, various microbes across different ecosystems have also been shown with the biodegradative potential. This review is therefore focused on the recent findings on the roles of different microorganisms as well as their enzymes in the biodegradation of synthetic plastics. Emphasis has also been placed on the mode of action of these biodegraders and the most significant factors affecting plastic biodegradation. Furthermore, the current and potential application of biotechnological tools in the modification of various organisms and their enzymes for enhanced plastic degradation is highlighted. This paper is expected to be an important reference for researchers and policymakers alike in charting a new course for the microbial war against plastic waste bioaccumulation.

2. Methodology

The review process was initiated by electronic searches using the ISI Web of Science (<http://apps.isiknowledge.com>), PubMed (<http://www.ncbi.nlm.nih.gov/pubmed>), Scopus (<http://www.scopus.com/>) and ScienceDirect (<http://www.sciencedirect.com/>) databases. The searches were conducted using the following keywords and strings: (plastic biodegradation OR degradation) AND (synthetic OR non-biodegradable) AND (microbe OR microbial OR microorganisms) AND (actinomycetes OR actinomycetal) AND (algae OR algal) AND (bacteria OR bacterial) AND (fungi OR fungus OR fungal) AND (enzyme OR enzymatic OR biocatalysts) AND (mechanism OR steps OR processes) AND (factors). This procedure allowed the filtering of published works on microbial and enzymatic degradation of synthetic/non-biodegradable plastic polymers. Publications on the degradation of biodegradable plastics were excluded and more focus was placed on publications within the last ten years except in cases where there is a lack of recent literature on the subject or for historical purposes. Two independent searches were made, and the conformity of the selected papers were validated, considering the inclusion criteria described. Furthermore, search results from predatory/unreputable journals, unpublished literature and publications in other languages besides English were not considered for this review. Finally, data from the search results were reviewed, analysed, categorised and presented under the appropriate sections to cover the scope of the paper.

3. Plastic types, properties and applications

The age of plastic began in 1869 when John Hyatt synthesized celluloid, the first plastic polymer, by the solvent action of camphor on

cellulose nitrate under mild temperature and pressure while searching for a replacement for ivory (Morris, 1988). Ever since this evolutionary discovery, different plastic polymers have been invented to enhance material science and to surpass the limits imposed by other materials such as wood, glass, wool and cotton. The global plastic market size was valued at USD 568.9 billion in 2019 and is expected to grow at a compound annual growth rate (CAGR) of 3.2% from 2020 to 2027 (Grand Review Research, 2020). Plastics comprise of a wide variety of long-chain polymeric substances, derived from different sources such as coal, oil and natural gas, with diverse applications in daily-life and industry. The synthesis of these polymers is basically along two reaction routes. The most common method involves addition polymerisation of carbon double bonds in the initial olefin to form new C—C bonds, the carbon-chain polymers (AlMa'adeed and Krupa, 2016). The production of polyolefins such as polyethylene, polybutene and polypropylene, which accounts for more than 60% of total plastic production is based on the aforementioned reaction. The other process is a condensation reaction between a carboxylic acid and an alcohol/amine group to form either a polyamide or a polyester. This second reaction forms the basis for the production of polyurethane which is produced from the condensation of isocyanate and polyol molecules (Akindoyo et al., 2016).

Synthetic plastics are commonly classified into two main groups viz., thermoplastics and thermosets, based on their thermal properties. Thermoplastics are plastic polymers which do not change their chemical composition upon reheating and thus can be remodified after melting. They include acrylonitrile butadiene styrene (ABS), polyamide, polyethylene (PE), polyimide (PI), poly-methyl methacrylate (PMMA), polypropylene (PP), polystyrene (PS), polytetrafluoroethylene (PFE), polyvinyl chloride (PVC), and polyvinylidene chloride (PVDC), (Li et al., 2019; Makhoul et al., 2016). The carbon backbone of thermoplastics makes them recalcitrant and resistant to the degradation or hydrolytic cleavage. Thermosets, unlike thermoplastics, cannot be remodified by melting as the heat generated chemical changes are irreversible. Furthermore, the backbones of thermosets are heteroatomic and highly cross-linked and thus are more susceptible to hydrolytic cleavage. Common thermosets include acrylic resins, epoxy resins, polyester, polyethylene terephthalate (PET), polyurethane (PU), silicone and vinyl resins (Jog, 1995; Ray and Cooney, 2018). The most commonly used plastics are PE, PET, polybutylene terephthalate (PBT), nylons, PP, PS, PVC and polyurethane (PUR), with PE and PP accounting for more than 50% of the total production. According to a recent report by Plastic Europe (2019), global plastics production was approximated at 360 million tonnes, with China producing around a third of the total. The report further showed that the packaging industry topped the demand for plastic with 40%, followed by building/construction, automobile, electrical/electronic, household/sports and agriculture. The properties and applications of the most commonly used plastics are summarised in Table 1 while an overview of the applications of different plastics is depicted in Fig. 1.

4. Microbial degradation of synthetic plastics

Microbes of all classes are in the forefront of preventing the bioaccumulation of various inorganic and organic compounds in the environment, hence, in the context of modern-day biotechnology, it is imperative to understand their roles in the biotransformation of xenobiotic compounds, such as plastic polymers. The biotic component of synthetic plastic degradation is mainly attributed to the action of various microbial communities which have been observed as potential degraders of xenobiotics based on their ability to adapt and use these chemicals as their growth and energy substrates. These group of organisms utilise their various enzymatic systems to degrade the polymers into intermediates which can be assimilated and metabolised subsequently for their energy needs. In this regard, different actinomycetes, algae, bacteria as well as fungi with the potential to biodegrade various plastic polymers have been investigated in recent times. However, the

Table 1
Properties and uses of some plastic polymers.

Plastic types	R-group	Tm (°C)	Tg (°C)	Xc (%) ^c	Reference
Acrylonitrile butadiene styrene (ABS)	C ₃ H ₃ N, C ₄ H ₆ & C ₈ H ₈	200	105–160	Low	(Wang et al., 2020)
Nylon	-NH ₂	220–280	160–220	20–35	(Huang et al., 2017)
Poly (methyl methacrylate) (PMMA)	-C ₅ O ₂ H ₈	98–171	95–113	Low	(Ute et al., 1995; Teng et al., 2009)
Polycarbonate (PC)	-CO ₃	200–300	140–200	Low	(Kyriacos, 2017; Lambert and Wagner, 2018)
Polyetherimide (PEI)	-C ₃₇ H ₂₄ O ₆ N ₂	340–360	218	0	(Gofman et al., 2013; Chen et al., 2019)
Polyethylene (PE)	-H	160–300	–120	45–90	(Li et al., 2019)
Polyethylene terephthalate (PET)	-COO & -OH	260	80	40–60	(Jog, 1995)
Polyimide	-ORCNCOR-	200–400	300–400	47.21	(Khalil et al., 2007; Lal et al., 2012; Zhang et al., 2017)
Polypropylene (PP)	-CH ₃	130	–10 - 18	40–60	(Makhlouf et al., 2016)
Polystyrene (PS)	-C ₆ H ₅	240	63–112	Low	(Lambert and Wagner, 2018; Wu et al., 2001)
Polytetrafluoroethylene (PTFE)	-C ₂ F ₄	342	–103	89–98	(Calleja et al., 2013; Sciuti et al., 2017)
Polyurethane (PU)	-	–55–100	–75–50	Low	(Foks et al., 1989)
Polyvinyl chloride (PVC)	-Cl	100–260	60–70	9–11	(Liu and Zhang, 2007)
Polyvinylidene chloride (PVDC)	C ₂ H ₂ Cl ₂	160–170	0–35	40–50	(Patterson and Dunkelberger, 1994)
Silicone	-R ₂ Si-O-SiR ₂	–47- 130	100–130	Low	(Rey et al., 2013)

*Tm: Crystalline melting temperature.

*Tg: Glass transition temperature.

*Xc: Percentage crystallinity.

rate of degradation of synthetic polymers by various microbes appear to be relatively slow which makes the biodegradative process not feasible for real-time industrial applications. This limitation has been ascribed to many factors, some of which have been highlighted in section 6.0 of this paper. Furthermore, this has resulted in the search for various additives to enhance the biodegradability of these materials (Selke et al., 2015), as well as the exponential growth of bioplastics with better biodegradability potentials (Thiruchelvi et al., 2020). Some of the microbes with plastic biodegradation potential and their characteristics are highlighted in Table 2, while Table 3 highlights some important patents that are associated with plastic degrading microbes and enzymes.

4.1. Actinomycetes

Actinomycetes form a diverse group of filamentous bacteria in soil, plant tissues and marine environments that have been well noted for their metabolic versatility and numerous biotechnological applications such as in bioremediation, medicine, and food industries. Actinomycetes including the *Streptomyces* groups, *Rhodococcus ruber*, *Actinomadura* spp., and the thermophilic *Thermoactinomyces* species have been isolated from different ecological zones and demonstrated to possess significant plastic biodegradative potentials (Auta et al., 2018; Jabloun et al., 2020). Their propensity to produce a wide variety of different hydrolytic enzymes as well as other bioactive metabolites have been highlighted previously (Gohain et al., 2020). These hydrolytic enzymes are one of the main factors responsible for their ability to grow on different plastic polymers and to degrade the high molecular weight compounds to simpler ones. Furthermore, they are known to produce extracellular polymers such as dextran, glycogen, levan, and *N*-acetylglucosamine-rich slime polysaccharides which probably facilitates their attachment to plastic surfaces for subsequent microbial action (Pujic et al., 2015). Similar to bacteria, biofilm formation has also been shown to be an important factor in the actinomycetal colonisation of plastics (Gilan and Sivan, 2013). *Streptomyces scabies*, isolated from potatoes was shown to degrade PET, together with other polymers including *p*-nitrophenyl esters, cutin and suberin using an esterase enzyme with a wide range of substrate specificity (Jabloun et al., 2020). An endophytic actinomycete, *Nocardopsis* sp. isolated from hibiscus was also demonstrated to degrade PE, as well as diesel (Singh and Sedhuraman, 2015). The effectiveness of actinomycetal plastic degradation has also been highlighted in a microbial consortium with a substantial fraction of actinomycetal species degrading polyurethane and different chemical additives (Gaytán et al., 2020).

4.2. Algae

Different algae, both photosynthetic and heterotrophic, have been well studied for their significant roles in bioremediation among their diverse industrial applications. They possess the capability to remove both inorganic and organic pollutants from various environments by accumulating, adsorbing, or metabolising them into relatively safer levels (Hwang et al., 2020). However, unlike other groups of microorganisms, only a few investigations have been carried out into the potentials of algae to degrade synthetic plastic polymers. Most of the studies have focused on their use for the production of green plastics. This is despite the different reports that have revealed the ability of marine algae to ingest plastic materials. Algae including the *Anabaena*, *Chlorella*, *Spirogyra*, *Nostoc*, *Oscillatoria* and *Spirulina* species have been shown to colonise different plastic surfaces even in terrestrial habitats, but there is no evidence to show that they metabolised the polymers (Sarmah and Rout, 2018). However, Kumar et al. (2017) showed the ability of *Scenedesmus dimorphus*, *Anabaena spiroides* and *Navicula pupula* to degrade both high density and low-density PE with the blue-green algae *A. spiroides* being the most promising, degrading 8.18% low-density polyethylene after 30 days. *Spirulina* sp. was also able to biodegrade PET and PP, however, the degradation rate observed after 112 days in the study, was significantly lower in comparison with bacterial and fungal cells (Khoironi and Anggoro, 2019). These results are not unexpected, as algae, unlike bacteria, utilise atmospheric CO₂ as their primary carbon source, and sunlight as the main energy source (Dineshababu et al., 2020). Hence, despite their ability to colonise plastic surface and assimilate microplastics, their metabolic pathways are not inclined to mineralise them, which is a major source of concern as this botched process has been identified to be an avenue of plastic to bioaccumulate and find their ways into the food chain (Hoffmann et al., 2020). However, a recent study has exploited the remarkable potential of *Phaeodactylum tricornutum* to serve as a genetic host and its inexpensive growth conditions for the biodegradation of PET with significant results. This was accomplished by expressing the gene for the widely popular PETase enzyme from *Ideonella sakaiensis* in the photosynthetic diatom (Moog et al., 2019).

4.3. Bacteria

Bacteria are noted to be the engine of the earth's nutrient as they are in the frontline for the transformation and cycling of nutrients through the environment. Like other microorganisms, their role in decomposition ensures that carbon and nutrients are liberated from different complex polymers, both natural and synthetic origin. They have been

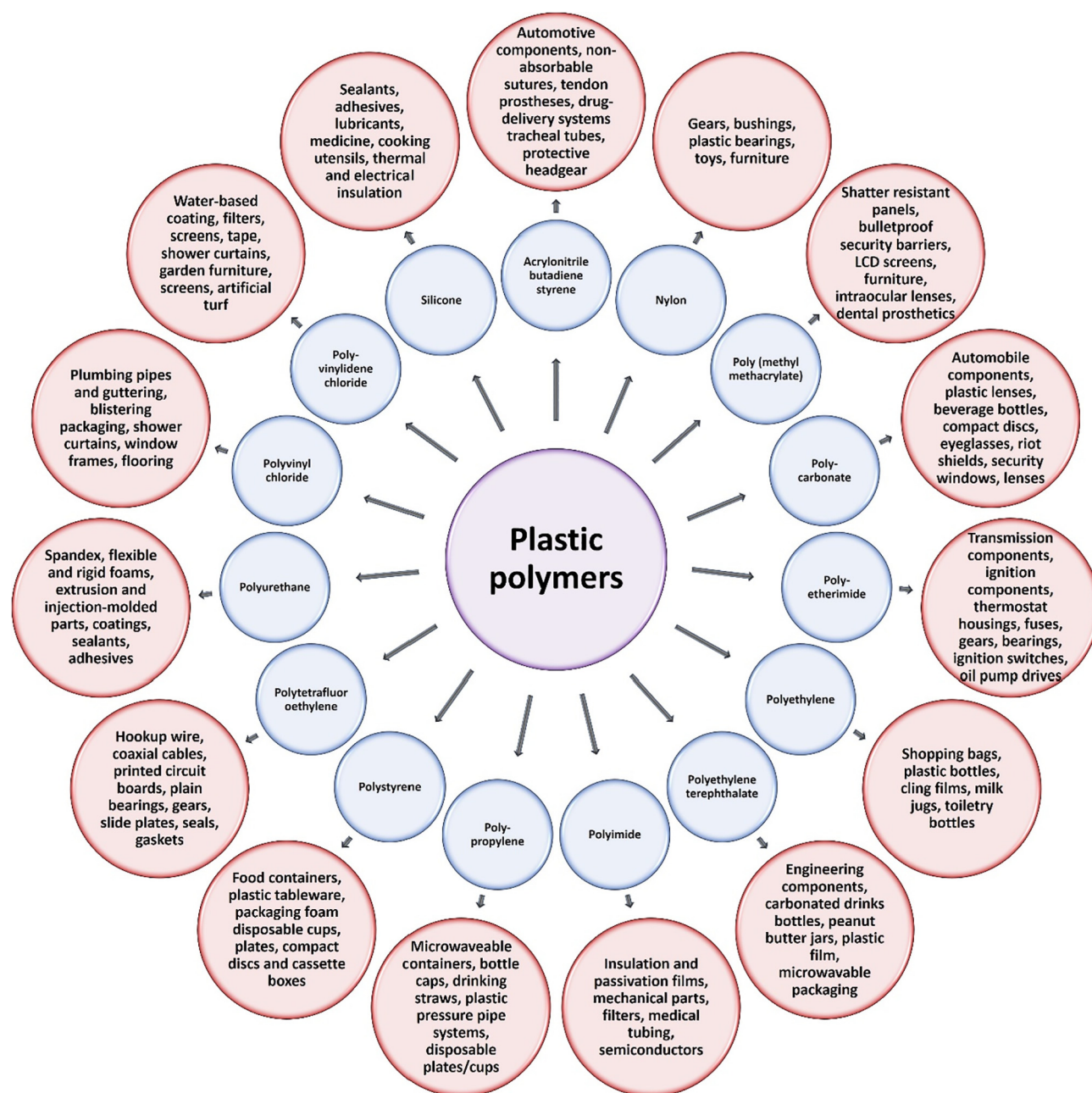


Fig. 1. Overview of synthetic plastic applications.

studied for their significant roles in bioremediation and have been shown to degrade various materials such as antibiotics, metal compounds, petroleum, plastic and other compounds that have gained prominence in this Anthropocene era. Using different approaches such as metagenomics, cloning, pure culturing, and even computational methods (Gan and Zhang, 2019), different bacterial species from the *Pseudomonas*, *Escherichia*, and *Bacillus* genera have been shown with significant potentials to degrade plastic polymers. Interestingly, these plastic degrading bacteria have also been isolated across different ecological niches such as dumpsites (Muhonja et al., 2018), recycling sites (Yoshida et al., 2016), landfills (Gaytán et al., 2020), cold marine environment (Urbanek et al., 2018) and insects' guts (Ren et al., 2019). Studies have shown that the ability of bacteria to degrade plastic is based on their natural capacity to degrade long-chained fatty acids, thus, it is not unexpected that *Pseudomonas* is the most prominent and studied bacterial genus with regards to plastic polymer degradation (Wilkes and

Aristilde, 2017). Biofilm formation has been noted to play a significant role in the bacterial decomposition of plastics, as it promotes the adherence of the colonies to the plastic surface as well as their persistence (Puglisi et al., 2019). As a result of the homopolymeric nature of thermoplastics, they have been known to be more resistant to microbial biodegradation, however, one of the most significant findings on plastic degradation is the ability of *I. sakaiensis*, a novel species isolated from a consortium of dumpsite bacteria, to degrade PET as it utilised the polymer as its primary source of energy and carbon (Yoshida et al., 2016). Polystyrene and polycarbonate are other common thermoplastics that have been shown to be degraded by *Pseudomonas aeruginosa*, *Bacillus megaterium*, *Rhodococcus ruber*, *Serratia marcescens*, *Staphylococcus aureus*, *Streptococcus pyogenes* and other bacterial strains (Arefian et al., 2020; Ho et al., 2018). Similarly, bacteria of different classes including *Bacillus*, *Pseudomonas*, *Micrococcus* have also been demonstrated to degrade various thermoset plastics, mostly polyurethane (Espinosa et al.,

Table 2
Plastic degrading microbes.

Microorganism	Plastic type	Degradative products	Reference
Actinomycete			
<i>Streptomyces scabies</i>	PET	Terephthalic acid	(Jabloune et al., 2020)
<i>Streptomyces</i> sp.	PE	Phthalic acid, Heneicosane, Benzoic acid, etc.	(Farzi et al., 2017)
<i>Streptomyces</i> sp.	PET	Ethyl benzene, o-Xylene	(Farzi et al., 2019)
<i>Streptomyces</i> sp.	PE	1,4-Epoxy naphthalene-1(2h)-methanol, 4,5,7-tris(1,1-dimethylethyl)23;2-t-butyl-5-chloromethyl-3- methyl-4-oxoimidazolidine-1-carboxylic, etc.	(Abraham et al., 2017)
Bacteria			
<i>Anoxybacillus rupiensis</i>	Nylon	6-Aminohexanoic acid	(Mahdi et al., 2016)
<i>Achromobacter denitrificans</i>	PE	Benzene, Tetrachloroethylene Heptadecyl ester, Benzene, 1–3, Dimethyl, Hexadecanoic acid, Eicosane, octane.	(Ambika et al., 2015)
<i>Acinetobacter baumannii</i>	PE	Methane, Dichloro-, Ethene, 2-Butene, Hexanal	(Pramila and Ramesh, 2015)
<i>Bacillus cereus</i>	PE	1-Trimethylsilylmethanol, 1,2,3,4 Tetra methyl benzene and Hexadecanoic acid, 1,2,3 Trimethyl benzene, 1 Ethyl 3,5-dimethyl benzene, 1,4 Di methyl 2 ethyl benzene, Dibutyl thalate	(Shahnawaz et al., 2016)
<i>Bacillus</i> spp.	PE	4,4-Dimethyl-2-pentene	(Muhonja et al., 2018)
<i>B. amyloliquefaciens</i>	PE	3-Hydroxybutyrate	(Novotný et al., 2018)
<i>Bacillus</i> sp.	PE and PVC	Carboxylic acids, alcohols	(Novotný et al., 2018)
<i>Enterobacter</i> sp.	PE	Monobenzyol phthalate	(Ren et al., 2019)
<i>Ideonella sakaiensis</i>	PET	Mono (2-hydroxyethyl) terephthalic acid (MHET), terephthalic acid (TPA) and bis(2-hydroxyethyl) TPA (BHET)	(Yoshida et al., 2016)
<i>Lysinibacillus fusiformis</i>	PE	1-Trimethylsilylmethanol, 1,2,3,4 Tetra methyl benzene and Hexadecanoic acid	(Shahnawaz et al., 2016)
<i>Pseudomonas</i> sp.	PS	2-(Nonyloxy carbonyl) benzoic acid, 1-chloro-2-methyl-cyclohexane, Dihexyl ester 1,2-Benzenedicarboxylic acid, etc.	(Subramani and Sepperumal, 2017)
<i>Pseudomonas aestusnigri</i>	PET	MHET	(Bollinger et al., 2020)
<i>P. protegens</i>	PU	Unspecified Impranil hydrolysis products	(Hung et al., 2016)
<i>Stenotrophomonas pavanii</i>	PE	4,6-Octadiyn-3-one, 2-methyl	(Muhonja et al., 2018)
Fungi			
<i>Aspergillus flavus</i>	PVC	Unspecified lower molecular weight oligomers	(Zhang et al., 2020)
<i>A. fumigatus</i>	PE	Unspecified lower molecular weight oligomers	(Muhonja et al., 2018)
<i>Aspergillus nomius</i>	PE	Phenol, 3,5-bis(1,1-dimethylethyl); 2-tbutyl-5-chloromethyl-3-methyl-4-oxoimidazolidine-1-carboxylic; dotriacontane; ethyl 14-methyl-hexadecanoate; diethyl phthalate; benzene, 1,3-bis(1,1-dimethylethyl); dodecane, 4,4-Dimethyl-2-pentene, 4,6-Octadiyn-3-one, 2-methyl	(Muhonja et al., 2018)
<i>A. oryzae</i>	PE	Dodecahydropyrido [1,2-b] isoquinolin-6-one	(Sangale et al., 2019)
<i>Aspergillus terreus</i>	PE	2 Naphthalene carboxylic acid; Dibutyl phthalate; 2-Cyclohexen; 1,2-Bis (Trimethylsilyl) benzene; Hexasiloxane and Hexadecanoic acid.	(Sangale et al., 2019)
<i>Aspergillus sydowii</i>	PE	7-Methylenebicyclo [3.2.0] hept-3-en-2-one; Dibutyl phthalate; 1,4-Benzenediol and	(Chaudhary and Vijayakumar, 2019)
<i>Cephalosporium</i> sp.	PS	Pyridine, Benzene, chloro, 2,4-Diphenyl-4-methyl-2(E)-pentene, Octadecanoic acid, ethyl ester, Methane, etc.	(Álvarez-Barragán et al., 2016)
<i>Cladosporium cladosporioides</i>	PU	2,2-Dimethyl-1,3-propanediol, hexane-1,6-diol, hexane 1,6-diisocyanate, and adipic acid di(oct-4-yl ester)-related compound	(Sumathi et al., 2016)
<i>Cochliobolus</i> sp.	PVC	hexane diisocyanate and other unspecified alcohols	(Jeyakumar et al., 2013)
<i>Engyodontium album</i>		Dimethylguanidine, 1,2-Benzenedicarboxylic acid, Diethyl [(phenyl sulfonyl) methyl] phosphonate, Diethyl [(phenyl sulfonyl) methyl] phosphonate, etc.	
		5-Octadecyne	
		1-Pentacosene	
		Dibutyl isophthalate	
		Acetoacetate	
		Propanoic acid	
<i>Mucor</i> spp.	PS	Benzene, Pyridine, 1,3,5 Cycloheptatriene, n-Hexane, etc.	(Chaudhary and Vijayakumar, 2019)
<i>Phanerochaete chrysosporium</i>	PP	Octane, Pentane	(Jeyakumar et al., 2013)
		9-Eicosyne	
		Acrylonitrile	
		Hexanoic acid, 2,7-dimethyloct-7-en-5-yn-4-yl ester, 3-Hexanol	

2020; Shah et al., 2008). Although most of the studies highlighted the biodegradability of pure bacterial strains, however, in nature, the bacteria usually act synergistically in consortia which have also been demonstrated in different studies (Lwanga et al., 2018; Shah et al., 2008). As earlier stated, the rate of microbial biodegradation can be affected negatively or positively by a range of external factors,

some of these factors have been subjects of investigations to accelerate bacterial degradation of different plastic polymers in vitro. The addition of specific additives such as food-grade dye-sensitised nanoparticles and starch revealed an improvement in the rate of PE degradation by some bacteria such as *Pseudomonas aeruginosa*, *Burkholderia seminalis* and *Stenotrophomonas pavanii* (Mehmood et al., 2016). Other

Table 3
Patents on plastic degrading microbes and enzymes.

Synthetic polymer	Patent number	Year	Patent description	Reference
Poly terephthalic acid second Diol ester, polybutylene terephthalate, polycaprolactam/nylon-6, polyhexamethylene adipamide/Nylon-66, polyhexamethylene sebacamide/nylon-610 and Poly L-lactic acid Polyester	CN107236147B	2019	A kind of method of crystalline plastics high-performance biodegradation	(Yang, 2019)
Polyester	US7960154B1	2011	Polyester-based-plastic-degrading bacteria, polyester-based-plastic-degrading enzymes and polynucleotides encoding the enzymes	(Toshiaki and Yukie, 2011)
Polyester	EP1849859B1	2014	Novel polyester plastic-degrading microorganism, polyester plastic-degrading enzyme and polynucleotide encoding the enzyme	(Toshiaki and Yukie, 2014)
Polyester	JP4625900B2	2011	Thermophilic polyester-degrading bacteria	(Tomisako and Kodaira, 2011)
Polyester	CN107532153A	2018	Novel polypeptide with polyester degrading activity and application thereof	(Li et al., 2018)
Polyester	US10287561B2	2019	Polypeptide having a polyester degrading activity and uses thereof	(Alvarez et al., 2019)
Polyester	WO2020021116A1	2020	Novel esterases and uses thereof	(Benoît et al., 2020)
Polyethylene	CN103980535B	2017	The method of bacillus extracellular laccase degrading polyethylene	(Yang et al., 2017)
Polyethylene	CN108633845A	2018	A kind of cultural method of the cured snout moth's larva of efficient degradation plastics	(Guo and Yi, 2018)
Polyethylene terephthalate	US10767026B2	2020	A process for degrading plastic products	(Desrousseaux et al., 2020)
Polyethylene terephthalate	ES2707304T3	2019	Method for recycling plastic products	(Boisart and Maille, 2019a)
Polyethylene terephthalate	JP6449165B2	2019	How to recycle plastic products	(Boisart and Maille, 2019b)
Polyethylene terephthalate	US10385183B2	2019	Process of recycling mixed PET plastic articles	(Maille, 2019)
Polyethylene terephthalate	WO2019053392A1	2019	Enzymatic process for depolymerization of post-consumer poly (ethylene terephthalate) by a glycolysis reaction	(De Castro et al., 2019)
Polyethylene terephthalate	WO2019168811A1	2019	Enzymes for polymer degradation	(Beckham et al., 2019)
Polyethylene terephthalate	FR3088070A1	2020	Process for the enzymatic degradation of polyethylene terephthalate	(Marty, 2020)
Polyethylene terephthalate, polyvinyl chloride, polystyrene, polypropylene, and polyethylene	WO2018143750A1	2018	Microorganism isolated from <i>Tenebrio molitor</i> larva and having plastic degrading activity, and method for degrading plastic using same	(Seo and Cheng, 2018)
Polystyrene, polyethylene, polypropylene, polyvinyl chloride, polyethylene terephthalate, and polycarbonate	US20150247018A1	2015	Biodegradation of petroleum-based plastic by microbial flora	(Yang et al., 2015)
Polyurethane	JP2004166542A	2004	New plastic decomposing bacterium	(Toshiaki and Yukie, 2004a)
Polyurethane	JP2004261102A	2004	Microorganism for degrading ester bond-containing plastic, plastic-degrading enzyme, and polynucleotide encoding the enzyme	(Toshiaki and Yukie, 2004b)
Polyurethane	JP2004166540A	2008	New plastic splitting enzyme and gene encoding the enzyme	(Toshiaki and Yukie, 2008)

pretreatment methods such as anionic surfactant addition, (Mukherjee et al., 2017) thermal treatment (Savoldelli et al., 2017) and UV pretreatment (Montazer et al., 2018) have all been shown recently to significantly increase the rate of bacterial degradation of various plastic polymers.

4.4. Fungi

Fungi, together with bacteria play the most dominant roles among all microbes in the maintenance of the biogeochemical cycles and essential nutrients on earth. The potentials of different fungal species to degrade various plastic polymers have been highlighted based on their ability to utilise these synthetic polymers as their primary/sole carbon or energy source. In this regard, a wide variety of fungal strains that cut across different classes, ecology and forms have been demonstrated to degrade plastics. Most recent studies have shown the *Aspergillus* genus to be the most prominent fungal group with regards to synthetic plastic biodegradation. *A. clavatus* (Gajendiran et al., 2016), *A. fumigatus* (Osman et al., 2018), and *A. niger* (Usman et al., 2020) are some *Aspergillus* species isolated from different terrestrial habitats and have been shown to degrade PE, PU and PP respectively. Interestingly endophytic fungi isolated from different plants have been shown in a study to degrade PU to varying levels under both solid-state and submerged fermentation conditions (Russell et al., 2011). Other fungal species with significant plastic degradability include *Fusarium solani*, *Alternaria solani*, *Spicaria* spp., *Geomyces pannorum*, *Phoma* sp., *Penicillium* spp.,

etc. (Muhonja et al., 2018; Zhang et al., 2020). Furthermore, in contrast to most of the studies that have been focused on the potentials of pure cultures, several fungal consortia have also been shown to synergistically degrade various forms of plastics such as PU (Cosgrove et al., 2007), and PE (Sowmya et al., 2015). Like all biological processes, the roles of fungal enzymes especially the depolymerases have been highlighted in all of these studies. Furthermore, the broad specificity of these enzymes which allows them to breakdown different polymers is significant (da Luz et al., 2019). The distribution and penetrative ability of fungal hyphae have also been noted to be an essential factor in their initial colonisation prior to subsequent depolymerisation as well as their ability to secrete hydrophobins for enhanced hyphal attachment to hydrophobic substrates (Sánchez, 2020). The enhancement of fungal biodegradation of plastics has been demonstrated by the pretreatment of the different substrates involving different factors such as photo-treatment and temperature (Corti et al., 2010), acid pretreatment (Mahalakshmi and Andrew, 2012), and various additives (Jeyakumar et al., 2013; Sánchez, 2020).

5. Enzymes involved in plastic biodegradation

Plastic-degrading enzymes, like other enzymes involved in the biological degradation of polymers, have since been classified into two broad categories, viz., extracellular and intracellular enzymes (Gu, 2003). However, the most studied group among the two are the extracellular enzymes which possess a wide range of reactivity, from

oxidative to hydrolytic functionality (Glaser, 2019). They are basically involved in the depolymerisation of the long carbon chains of the plastic polymers to a mixture of oligomers, dimers and sometimes, monomers. These diverse groups of enzymes have been found to act similarly to microbial laccases, peroxidases, lipases, esterases and cutinases as have since been classified as such (Gan and Zhang, 2019). Furthermore, these extracellular enzymes are posited to be involved in heterogeneous reactions occurring at the solid/liquid interface, as they act on the macromolecules available at the surface of the solid plastic while present in the liquid phase (Chinaglia et al., 2018). Other groups of enzymes are involved in the surface functionalisation of the hydrophobic plastic surfaces, degradation of the plastic metabolic intermediates into monomeric units, and the final mineralisation of the final monomeric intermediates. A large proportion of intracellular enzymes are responsible for the aerobic and anaerobic processes necessary to convert the intermediates to compounds which can be assimilated for the microbes (Pathak, 2017). However, not much information is available on the biochemical properties of plastic degrading enzymes as well as their structural characteristics. There have also been different classification schemes for these enzymes, however, in this review, the plastic degrading enzymes are grouped according to the polymer they act on, i.e. polyethylene, polyurethane, polyethylene terephthalate, polystyrene and nylon.

5.1. Polyethylene-degrading enzymes

Polyethylene degrading biocatalysts have been identified from actinomycetal, bacterial and fungal sources, which include hydroxylases, laccases, peroxidases and reductases. A manganese peroxidase was identified as the major enzyme involved in the degradation of PE by two lignin-degrading fungi, *Phanerochaete chrysosporium* and *Trametes versicolor* (Iiyoshi et al., 1998). Another enzyme, alkane hydroxylase was found to be important in the breaking down of PE by *Pseudomonas* sp. E4, which was further confirmed by cloning the gene in *E. coli* and evaluation of the recombinant protein (Yoon et al., 2012). Subsequently, an enzymatic system in *P. aeruginosa* was found to contain alkane hydroxylase together with rubredoxin and rubredoxin reductase which was revealed to be responsible for the degradation of low molecular weight PE (Jeon and Kim, 2015). Laccases from actinomycetes, *Rhodococcus ruber* (Santo et al., 2013) and fungi such as *Aspergillus flavus* (Zhang et al., 2020) and *Pleurotus ostreatus* (Gomez-Mendez et al., 2018) have also exhibited significant degradation of PE. It was proposed that they act via the oxidation of the PE hydrocarbon backbone.

5.2. Polyurethane-degrading enzymes

Different enzymes including cutinases, esterases, lipases, laccases, peroxidases, proteases and ureases from bacterial and fungal sources have been shown to possess PU-degrading abilities (Magnin et al., 2020). The activity of serine and cysteine hydrolase has been highlighted in the *Pestalotiopsis microspore* PU degradation (Russell et al., 2011). Recently, the synergistic effects of esterase and an amidase on the degradation of various PU-derivatives, via a proposed stepwise mechanism, was also demonstrated (Magnin et al., 2019). Using proximity ligation-based metagenomic analysis, a variety of enzymes involved in PU degradation in a consortium of microbes were identified. The enzymes were shown to be responsible for the metabolism of various PU intermediates and included different dioxygenases, decarboxylases, dehydrogenases, transferases, ligases, hydrolases, isomerases and peroxidases (Gaytán et al., 2020). However, it has been noted that most of the PU-active enzymes so far studied were active on ester-linked PUR and less likely on polyurethane ethers (Danso et al., 2019). It is posited that the carbonyl group present in the ester-linked PUR makes them more vulnerable to hydrolysis caused by enzymatic activities, unlike the ether-linked PUR.

5.3. Polyethylene-degrading terephthalate enzymes

Since the first description of the enzymatic hydrolysis of PET polymers using *Thermobifida fusca* hydrolases (Müller et al., 2005), many other enzymes including cutinases, esterases, (Jabloune et al., 2020) lipases and carboxylesterases have been shown with PET-degrading ability (Danso et al., 2019; Jabloune et al., 2020; Ru et al., 2020). The enzymatic hydrolysis of PET has been noted to be generally based on a surface erosion mechanism (Glaser, 2019). Perhaps one of the most studied plastic degrading enzymes is PETase, an aromatic polyesterase, from *I. sakaiensis* 201-F6 with a basic α/β hydrolase fold. This enzyme metabolises PET to bis(2-hydroxyethyl)-TPA (BHET), MHET, and TPA while its accessory enzyme MHETase acts on the MHET intermediate converting it to terephthalic acid and ethylene glycol (Austin et al., 2018). The enzyme was, however, thermolabile and subsequent engineering of the PETase enzyme aimed at increasing its thermostability resulted in a new variant which was found to be 14 times more active than the original biocatalyst (Son et al., 2019). A 25 kDa suberinase with remarkable stability and the capability to hydrolyse PET to terephthalic acid, as well as other polymers, was also sourced from *Streptomyces scabies* (Jabloune et al., 2020).

5.4. Polystyrene-degrading enzymes

Even though the microbial degradation of polystyrene by various bacteria and fungi has been demonstrated, the major enzymes involved in the initial depolymerisation of the polymers have not been clearly identified. However, an extracellular esterase from *Lentinus tigrinus* has been shown to breakdown PS (Tahir et al., 2013). In addition, some polymerases, which were sourced from *Bacillus* and *Pseudomonas* species, have also been noted to be responsible for PS degradation (Mohan et al., 2016). Other enzymes that are involved in the metabolism of styrene, the monomer of PS before its entry into the TCA cycle have been identified as styrene monooxygenase, styrene oxide isomerase, phenylacetaldehyde dehydrogenase, and phenylacetyl coenzyme A ligase (Ho et al., 2018). It is believed that these enzymes are involved in a series of reactions which include depolymerisation of the polymer to styrene, the oxidation of styrene to phenylacetate and the incorporation of phenylacetate into Krebs cycle.

5.5. Nylon-degrading enzymes

Some nylon-degrading enzymes have been identified from fungal and bacterial sources (Nomura et al., 2001; Yamano et al., 2019). Such enzymes include a manganese peroxidase from a white-rot fungus identified as strain IZU-154, which was shown to strip off the surface of Nylon 6 and cause deep horizontal grooves formation in the polymer (Deguchi et al., 1998). Peroxidases are a group of heme-containing enzymes, which catalyse the oxidation of a wide variety of organic and inorganic substrates using hydrogen peroxide as the electron acceptor. In addition, the metabolism of 6-aminohexanoate, an intermediate product of nylon has been shown to be catalysed by three different hydrolases in *Flavobacterium* and *Pseudomonas* strains (Negoro, 2000). These enzymes which were identified as 6-aminohexanoate-cyclic-dimer hydrolase, 6-aminohexanoate-dimer hydrolase and endo-type 6-aminohexanoate-oligomer hydrolase, acting in tandem converted the nylon intermediate to its monomer, 6-aminohexanoate, under mesophilic and neutral to alkaline pH conditions. Subsequent studies have also identified enzymes with similar properties with the aforementioned enzymes in *Arthrobacter* sp. KI72 (Yasuhira et al., 2007).

6. Mechanism of microbial plastic degradation

Microbes break down different compounds into simpler forms through biochemical transformation. The biodegradation of plastic polymers can be observed through an alteration in the physical

properties of the polymers especially by molecular weight reduction, loss of mechanical strength and change in plastic surface properties (Ho et al., 2018). As stated earlier, the objective of plastic biodegradation is the conversion of recalcitrant wastes to non-toxic lower molecular mass compounds that can return into the biogeochemical cycle. Thus, the different biochemical degradative pathways involved in plastic biodegradation can be classified into biodeterioration, biofragmentation, assimilation and mineralisation, all of these processes are executed via various enzymatic activities and bond cleavage (Gu, 2003; Pathak, 2017). A schematic representation of the processes involved in plastic biodegradation is shown in Fig. 2.

6.1. Biodeterioration

Biodeterioration is caused by the chemical and physical actions of microbes, or/and other biological agents, that result in the superficial degradation of the plastic polymer as well as modification in the chemical, mechanical and physical properties of the polymers (Anjana et al., 2020). The changes which are observed in the polymers during biodegradation are also enhanced by prolonged exposure to external conditions which include light, temperature and chemicals in the environment. The biodeterioration process is the first and it is initiated by the adherence and colonisation of microbes on the polymer surface with the sole aim of reducing the resistance and durability of the plastic materials. Thus, the introduction of hydrophilic functional groups to plastic surfaces is often required to promote the attachment of the microorganisms as plastics are naturally hydrophobic (Nauendorf et al., 2016). Furthermore, it has been observed that for plastics with higher surface hydrophobicity such as polyethylene, the formation of biofilms is necessary to increase the polymeric surface interaction with bacteria

(Schwibbert et al., 2019). Hence, biofilm-forming bacteria such as *Pseudomonas* have been observed to adhere more strongly and degrade low-density polyethylene in comparison to other bacteria in the planktonic mode (Tribedi et al., 2015). This is not far-fetched as bacterial biofilms are known to protect the microbial community from external fluctuations and enhance their persistence under different conditions. Tribedi et al. (2015) further established that biofilm promoting compounds such as mineral oil enhanced plastic biodegradation while surfactants which diminished biofilm attachment consequently reduced the degradative process. Fungal cells, on the other hand, are adapted to grow on almost all kinds of surfaces found in nature, hence they have been shown to attach to polymeric plastic surfaces through their hyphae (Sánchez, 2020). At this stage, the attachment and subsequent growth of the fungi on the polymer solids result in localised swelling, leaving a polymer with significantly decreased mechanical properties. As soon as the microbes attach to the plastic surface, they continue to proliferate using the polymers as sole carbon sources. Studies have shown that the attachment and initial growth of these microbes might be enhanced by the presence of other additives in the polymers, such as plasticisers which are more accessible and easily metabolisable by the microbes (Ru et al., 2020). Specifically, the capacity of a *Pseudomonas aeruginosa* strain to develop and maintain active biofilms on polyethylene surface, for two months was attributed to its consumption of low molecular substances in the polymer (Gupta and Devi, 2020). Furthermore, exopolysaccharides have also been noted to play significant roles during the attachment and biodeterioration of plastic polymers by promoting stronger biofilm adhesion (Anjana et al., 2020). These extracellular polymers also serve as surfactants which facilitate the exchanges between hydrophilic and hydrophobic phases to favour the penetration rate of microbial species (Lucas et al., 2008).

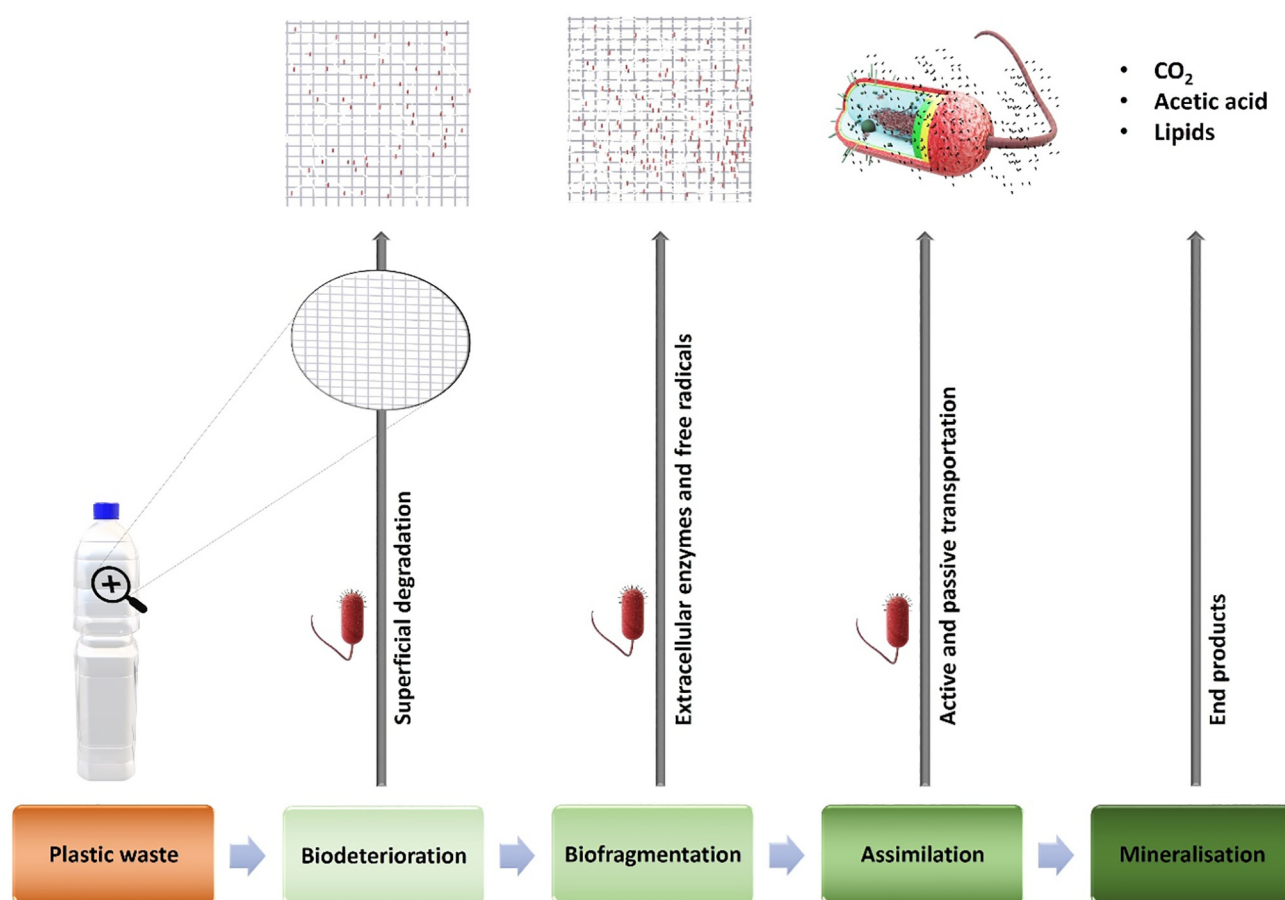


Fig. 2. Steps involved in plastic biodegradation.

6.2. Biofragmentation

Biofragmentation, the subsequent stage is a depolymerisation step involving the catalytic cleavage of bio-deteriorated plastic polymers into smaller units, by the action of extracellular enzymes, and free radicals generated by the microbes (Jenkins et al., 2019). The biofragmentation process is proposed to involve two principal reactions which are the reduction in polymer molecular weight and oxidation of the lower weight molecules. These reactions are necessary to facilitate the subsequent action of microbial enzymatic systems which are usually able to attack lower molecular weight compounds (Restrepo-Flórez et al., 2014). These enzymes mainly catalyse the hydrolytic cleavage of the polymers as the glycosidic, ester, and peptide bonds within the plastics are subjected to a hydrolytic nucleophilic attack on the carbonyl carbon. These hydrolytic reactions occur via two different modes of attack, the *exo*- and the *endo*- attacks which result in various products. While the former results into constituent oligomers or monomers, such as ethylene glycol and terephthalic acid, which the microbe can readily assimilate into the cell, the products of the *endo*- attacks must still undergo further degradation before they can be assimilated by the microbes (Pathak, 2017). The significant effects of the oxidation reactions have been shown in *Rhodococcus rhodocrous* which was able to degrade almost all of the previously oxidised oligomers from polyethylene (Gravouil et al., 2017). In addition, it is also posited that different inorganic and organic compounds released by the microbes could also be influential in facilitating the biofragmentation process. Various inorganic compounds (ammonia, hydrogen sulphide, nitrites thiosulphates), as well as organic acids (citric, fumaric, gluconic, glutaric, glyoxalic, oxalic acids, etc.), have been noted with the ability to scavenge cations from the polymer surface forming stable complexes that can induce surface erosion (Krause et al., 2020) and fragmentation. The positive relationship between the rate of plastic degradation and photooxidation has also been highlighted in previous studies, as deduced from the significant change in the carbonyl index for the UV-irradiated polymer (Sen and Raut, 2015).

6.3. Assimilation

The lower molecular weight compounds produced during biofragmentation are transported into the microbe's cytoplasm at the stage of assimilation. Although the assimilation process of plastic molecules across different microbial membranes have not been well elaborated, it is posited that just like hydrocarbons, the process involves both active and passive transportation. Octadecane, a degradative product of plastic polymers (Shahnawaz et al., 2019) has been shown to be taken up by *Pseudomonas* sp. DG17 via facilitated passive transport systems at higher concentrations, while at lower concentrations, it is assimilated via energy-dependent active transportation (Hua et al., 2013). Furthermore, the presence of many membrane-bound monooxygenases has been shown to be ubiquitous in alkene-assimilating bacteria for the initial oxidation of alkenes (Durairaj et al., 2016). Different membrane transport systems have also been shown to facilitate the movement of these substances into the cytoplasm for further processing. A specific transporter has been noted to be responsible for the inward transportation of terephthalic acid, a hydrolytic product of polyethylene terephthalate (PET), in a *Comamonas* species (Hosaka et al., 2013). Similarly, porins have been shown to possess the ability to move polyethylene glycol, a plastic degradative product, into the cytoplasm for further bioconversion (Duret and Delcour, 2010). In the study by Gravouil et al. (2017), different transporters mainly belonging to the major facilitator superfamily and the ATP binding cassette family of proteins, were identified to have been upregulated in *Rhodococcus rhodocrous* while assimilating polyethylene oligomeric intermediates. It was further proposed that some transporters could be involved in the dual role of intermediate trafficking and oxidation as exemplified by an identified transport protein with NADH dehydrogenase activity.

6.4. Mineralisation

Once these plastic derivatives are successfully transported into the cells, they undergo series of enzymatic reaction which lead to their complete degradation into oxidised metabolites which includes CO₂, N₂, CH₄, and H₂O (Ho et al., 2018). The complete mineralisation of plastic polymers has been shown by techniques such as isotopic tracing and the quantification of CO₂ release using Strum's method (Yang et al., 2020). Alternatively, the intermediates can be channelled into different chemical pathways. For example, polyethylene degradation has been proposed to proceed through the formation of acetic acid, which can enter the Krebs cycle through acetyl-CoA formation or channelled into lipid formation (Wilkes and Aristilde, 2017). Similarly, succinate, another Krebs cycle intermediate, is also generated from polyethersulfones degradation through the action of esterase in *Pseudomonas* sp. AKS2 (Tribedi and Sil, 2014). The well-elucidated biodegradation of styrene, the monomeric unit of the recalcitrant polystyrene showed that styrene is mainly oxidised to phenylacetate, which is then introduced into the Krebs cycle as phenylacetyl coenzyme A for complete metabolism (Ho et al., 2018). Studies on *I. sakaiensis*, also revealed that the internalised terephthalic acid is metabolised by TPA 1,2-dioxygenase (TPADO) and 1,2-dihydroxy-3,5-cyclohexadiene-1,4-dicarboxylate dehydrogenase (DCDDH) to produce protocatechuic acid (PCA) as the final molecule. PCA passes through a series of enzymatic reactions involving PCA 3,4 dioxygenase and dehydrogenase to form 2-pyrone-4,6-dicarboxylic acid which is channelled into the Krebs cycle as pyruvate and oxaloacetate, before final mineralisation to CO₂ and H₂O (Yoshida et al., 2016). The mineralisation step can either be aerobic or anaerobic but in both cases, it requires the activities of various enzymes which include esterases, lipases, cutinases, peroxidases and laccases (Alshehrei, 2017).

7. Factors affecting microbial biodegradation of plastics

Microbial biodegradation of plastic polymers, in nature as well as under controlled conditions is affected by a wide variety of factors that can all be grouped based on polymer characteristics, environmental factors and chemical factors. These factors function primarily to facilitate subsequent microbial action by increasing the surface area, hydrophilicity, as well as reducing the molecular weight.

7.1. Polymer characteristics

The rate of microbial degradation has been noted to decrease with increasing molecular weight as the polymers are needed to be transported across the cellular membrane to be metabolised. Hence, smaller units of polymers such as the monomers, dimers and oligomers are easier degraded and mineralised as has been demonstrated by *Rhizopus delemar* lipase (Tokiwa et al., 2009). The rate of plastic polymer degradation is also significantly affected by the morphology of the polymer, which includes the degree of branching, crystallinity, and its physical form. Plastic polymers with a higher proportion of side chains, and hence increased branching, are less assimilated for microbial degradation. Studies have shown that the non-crystalline portion of polymers is more susceptible to enzymatic degradation as they are more loosely packed and accessible, hence there is an inverse relationship between the crystallinity and rate of degradation (Devi et al., 2016). A study comparing the half-lives of different plastic polymers, assuming pseudo-zeroth-order kinetics, estimated that the specific surface degradation rate of polyethylene, a relatively more crystalline polymer, to be 9.5 $\mu\text{m year}^{-1}$ compared to 1105 $\mu\text{m year}^{-1}$ of PET under the same conditions (Chamas et al., 2020). The melting temperature of the respective polymer, T_m, strongly affects its rate of microbial degradation. It has been discovered that there is an inverse relationship between the T_m and biodegradation rates. However, T_m of plastic polymers is influenced by the change of enthalpy of melting (ΔH) as well as the change in

entropy of melting (ΔS) as shown in the equation; $T_m = \Delta H/\Delta S$ (Tokiwa et al., 2009). Although there are no findings to establish the relationship between the glass transition temperature (T_g) of synthetic plastics, it is believed that the structural changes that occur at the temperature may likely enhance microbial attack (Lucas et al., 2008). The extent of initial microbial colonisation of the plastic polymers is enhanced by the increase in the hydrophilicity of the material and the activity of the extracellular enzymes is also believed to be inhibited with higher hydrophobicity. Hydrophilic surfaces, with their increased wettability, possess higher surface energies and give lower contact angles with water, thus promoting the microbial attachment to the polymer surface, and accelerating the degradation rate (Chamas et al., 2020). Therefore, the presence and formation of polar functional groups in plastic polymers due to environmental weathering factors, such as UV exposure, has also been noted to result in a decrease in contact angle with water and hence an increase in hydrophilicity. The effect of polymer hydrophilicity/hydrophobicity has been demonstrated using molecular dynamic simulations, where highly hydrophobic plastic polypropylene had the least biodegradative potential compared to relatively highly hydrophilic nylon (Min et al., 2020).

7.2. Environmental factors

Biodegradation by microbes depends largely on the initial individual or synergistic action of different environmental elements on the polymers as the complexities of the surrounding environment play a key role in the kinetics of biodegradation. The characteristics of each environment differ significantly; hence the rate of microbial action will vary across a dry environment, humid air, a landfill, in compost, in the marine environment, etc. Factors including light, heat, moisture, pH, and biological activity have been found to enhance bond scission. They also affect structural homogeneities and new functional group formation (Siracusa, 2019). The presence of moisture in the environment will enhance the miniaturisation of plastic polymers through an increase in their solubility and also an increase in the hydrolysis rate. These, in turn, will lead to more chain scission and will eventually increase the sites of microbial action on the polymer chains for enhanced biodegradation as demonstrated by Chamas et al. (2020), which showed that degradation in the marine environment is significantly higher than on land, with all other factors kept constant. Different plastic polymers have been observed to be sensitive to electromagnetic radiation as they are able to absorb the stronger part of the tropospheric solar radiation. These synthetic polymers tend to absorb high-energy radiation in the ultraviolet region of the spectrum, which excites their electrons to higher reactivity and results in oxidation and scission (Brebou, 2020). Thermal degradation of plastic has also been observed to occur at high temperatures. Temperatures in landfills have been estimated at 100 °C, a condition which accelerates degradation rates provided sufficient moisture and oxygen and are available for the subsequent thermal-oxidative degradation and hydrolytic pathways (Hao et al., 2017). The increase in the kinetic energies of the atoms causes a disorderliness in the polymeric structure leading to molecular scission of the components of the long-chain backbones (Ray and Cooney, 2018). This induces chemical reactions between the different components which subsequently leads to changes in the physical and optical properties of the polymers. Specifically, thermal degradation affects the molecular weights of the polymers, reduces ductility and embrittlement, initial colour, cracking, etc. Subsequent to the effect of the other environmental factors, C—C and C—H bond scission initiates polymers degradation through a series of reactions involving free radicals (Devi et al., 2016).

7.3. Chemical reagents and additives

The presence of chemical reagents in the polymeric structures (additives) or the surrounding environment can either activate, inhibit or

catalyse the biodegradative process by affecting the functional groups as well as hydrophilicity/hydrophobicity (Fotopoulou and Karapanagioti, 2017). Recently, additives have been added to polymers to serve as pro-oxidants, flame retardants, pro-degradants, etc. Studies have shown some additives causing a significant reduction in plastic recalcitrance during subsequent reprocessing or degradation (Aldas et al., 2018). While others serve as inhibitors to microbial degradation, a notable example is dibutyl tin dilaurate, a highly toxic plastic additive in PU with antimicrobial effects, thus reducing microbial action on the polymer (Cregut et al., 2013). Furthermore, the presence of an alternative and simpler carbon source in the environment has been shown to affect microbial action on plastics (Mehmood et al., 2016). This effect is exerted through catabolite repression as observed when degradation of a polyethylene derivative by *Pseudomonas* strain was increased by 80% on the removal of glucose in the media (Tribedi et al., 2012). On the other hand, biodegradation was enhanced by the addition of biodegradable additives such as starch (Mehmood et al., 2016) and palmitic acid (Jayaprakash and Palempalli, 2018), which served as a nutrient source for microbes. Oxidative agents such as hydrochloric acid, hydrogen peroxide, sulphuric acid and nitric acid have also been shown to oxidise polymer surfaces by the addition of OH⁻ group radicals thereby enhancing biodegradation (Moharir and Kumar, 2019). Likewise, the addition of surfactants such as Tween 80 and sodium dodecyl sulphate, have also been observed to promote microbial degradation by causing an increase in the hydrophilicity of the polymer surfaces (Ghatge et al., 2020). The significant increase in mineralisation rate and carbon fixation observed during the biodegradation of polyethylene was ascribed to the introduction of additives which substantially induced the oxidation of the polymer (Jakubowicz, 2003).

8. Molecular aspect of plastic biodegradation

In order to increase production levels, ease product recovery as well as enhance the activities of various plastic degrading enzymes, different successful attempts have been made to manipulate the genes encoding for these enzymes using various molecular techniques. It has been observed however that most of the recombinant plastic degrading enzymes were cloned from *Pseudomonas* species. For example, *P. fluorescens* ST was used as the source of genes capable of polystyrene catabolism. The expression of genes in the host system showed that the gene products had monooxygenase, epoxystyrene isomerase and epoxystyrene activities (Marconi et al., 1996). A polyester polyurethane (PU)-degrading enzyme exhibiting remarkable activity against Impranil was also cloned from the same species (Vega et al., 1999), as well as from another species of the same genus, *Pseudomonas chlororaphis* (Howard et al., 2001). *Pseudomonas* sp. E4 was shown to be a potential degrader of polyethylene. Subsequently, its alkane hydroxylase gene was expressed in *E. coli* to produce a heterologous enzyme with the ability to mineralise close to 20% of the analysed low molecular weight polyethylene into CO₂ (Yoon et al., 2012). A polyester hydrolase, classified as a member of type IIa PET hydrolytic enzyme has also been cloned from *P. aestusnigri* and expressed in *E. coli* (Bollinger et al., 2020). The resultant recombinant enzyme was found to degrade PE, bis(2-Hydroxyethyl) terephthalate, amorphous PET but could not degrade commercial PET bottles. However, the activity of the enzyme was further improved to degrade films from PET bottle by site-directed mutagenesis (Bollinger et al., 2020). The remarkable ability of *Ideonella sakaiensis* to significantly breakdown PET has been a subject of many studies. In this regard, a relatively significant amount of genetic manipulation studies has been carried out on the PETase gene from the bacterium. The PETase gene from *I. sakaiensis* has been successfully cloned and expressed in different host systems including *E. coli* (Joo et al., 2018) and *Phaeodactylum tricornutum* (Moog et al., 2019). As a means of improving on the difficult enzyme recovery, instability and solubility challenges experienced with the wild type organism, the same PETase gene was also expressed in *E. coli* BL21 (DE3)-T1R (Seo et al., 2019). Recombinant enzymes with significant PETase activities have also been

cloned from different microbes besides *I. sakaiensis*. One of such genes was isolated from an actinomycetes strain of marine origin, *Streptomyces* sp. SM14 and expressed in *E. coli*, which resulted in an extracellular enzyme with similar native signal peptide sequence to the native enzyme (Almeida et al., 2019). An enzyme with the ability to metabolise PET together with other natural and synthetic substrates was also cloned from another actinomycete, *S. scabies* (Jabloun et al., 2020). Recently, the expression of a PETase of algal origin was also reported, the gene sourced from the green microalgae *Phaeodactylum tricornutum*, was expressed in a faster growing and environmentally friendly green algae *Chlamydomonas reinhardtii* (Kim et al., 2020). In order to fully explore the enzymatic degradation of PET, MHETase, a tannase-like enzyme which has been identified to work in tandem with PETase for the complete metabolism of PET, has also been cloned from *Ideonella sakaiensis* and expressed in *E. coli* (Janatunaim and Fibrani, 2020). In addition, several protein engineering strategies have been adopted to enhance the enzymatic activity of various plastic degrading enzymes. Site-directed mutagenesis has been employed to reduce Km value significantly and to increase the activities of *I. sakaiensis* by more than a hundred per cent (Ma et al., 2018). The substrate specificity of the enzyme has also been engineered to metabolise other plastic polymers, besides PET such as polyethylene-2,5-furandicarboxylate (PEF) (Austin et al., 2018). The PET degrading ability of a cutinase from *Thermobifida cellulolytica* has also been observed to be enhanced by 16-fold by the covalent fusion of its gene with hydrophobins (Ribitsch et al., 2015).

9. Conclusion and future prospects

This paper has highlighted significant research on the biodegradation of synthetic plastic wastes by microbes including actinomycetes, algae, bacteria and fungi. Some light was shed on the general mechanisms of this biodegradation and the roles of different enzymes involved. Based on the current literature, it can be deduced that the information on different microbes with plastic degrading potentials have been based on pure culture isolates. This clearly demonstrates that the high diversity of microorganisms across different natural habitats has not been significantly exploited, notably, no yeast organism has been identified as plastic biodegraders. The use of metagenomics which ensures the exploration of both culturable and unculturable microbes will enhance the identification of microbes and biocatalysts with potentials for plastic biodegradation. Furthermore, other -omic tools including genomics, transcriptomics, proteomics and metabolomics will aid in understanding biological interactions that occur between genes, transcripts, proteins, metabolites and external environmental factors during synthetic plastic degradation. It is also believed that the application of different microorganisms as a consortium will lead to greater efficiency in plastic degradation due to the synergism between the microbes and their enzymes. Although many plastic degrading enzymes have been identified from diverse sources, however, the biochemical and structural properties of these enzymes have not been well studied. These pieces of information are necessary to have a better understanding of the mechanisms involved in the biodegradation of recalcitrant plastics. This understanding will be useful in the modification of enzymes through protein engineering, designing of microbial cell factories with better degradation efficiency, as well as the development of novel plastic polymers with improved biodegradability. Also, of high importance is the investigation of the effects of different pretreatment methods and additives on the microbial degradation of synthetic plastics, as it is expected that the inclusion of the appropriate pretreatment/additives might yield better results. A lot of discrepancies were observed from the different studies with regards to the methods of assessing the degradation efficiency, thus, developing and adopting a standard or universal approach will go a long way in the harmonisation of data and the subsequent promotion of this area of research. Given the inexhaustible potentials of microbes and their continuous adaptation to the changing

environment, it is expected that more in-depth studies in this area of research will soon result in viable biodegradation processes that can be developed on a commercial scale.

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Ayodeji Amobonye: Conceptualisation; Writing - Original Draft. Prashant Bhagwat: Writing - Original Draft. Suren Singh: Resources, Writing - Review & Editing. Santhosh Pillai: Resources, Writing - Review & Editing, Supervision, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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