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Review

Understanding plastic degradation and microplastic formation in the environment: A review☆

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ABSTRACT

Plastic waste are introduced into the environment inevitably and their exposure in the environment causes deterioration in mechanical and physicochemical properties and leads to the formation of plastic fragments, which are considered as microplastics when their size is < 5 mm. In recent years, microplastic pollution has been reported in all kinds of environments worldwide and is considered a potential threat to the health of ecosystems and humans. However, knowledge on the environmental degradation of plastics and the formation of microplastics is still limited. In this review, potential hotspots for the accumulation of plastic waste were identified, major mechanisms and characterization methods of plastic degradation were summarized, and studies on the environmental degradation of plastics were evaluated. Future research works should further identify the key environmental parameters and properties of plastics affecting the degradation in order to predict the fate of plastics in different environments and facilitate the development of technologies for reducing plastic pollution. Formation and degradation of microplastics, including nanoplastics, should receive more research attention to assess their fate and ecological risks in the environment more comprehensively.

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1. Introduction

Plastics are man-made long-chain polymeric materials that are extensively used in many aspects of life due to their excellent characteristics such as light weight, strong plasticity and flexibility, thermal and electrical insulation, corrosion resistance, and low cost. Global production of plastics (excluding fibers)increased from 1.3 million tons in 1950 to 359 million tons (including Polypropylene (PP) fibers) in 2018 (Plastics Europe, 2007; 2019). However, the production data of man-made fabrics including polyester, nylon, acrylic, cellulosic, and PP fibers is not readily available. The first commercial production of cellulosic fibers began in 1892, nylon was not discovered until the 1930s, and man-made fiber production has been increasing rapidly ever since (Woodings 2001; Reimschuessel 1977). It was reported that a total of 60.4 million tons of synthetic fibers were in demand in 2014, and the production of polyester surpassed cotton in 2002—with a significantly faster growth rate than those of all other fibers—and will reach an estimated 50 million tons in 2020 (Carmichael 2015).

Because of improper management and disposal practice, a large amount of plastic waste enter the environment through various pathways and cause serious environmental pollution problems (Jambeck et al., 2015; Geyer et al., 2017). Once in the environment, plastic wastes can slowly break down and generate numerous smaller plastic debris under the action of physical, chemical and biological processes (PlasticsEurope 2019). The particles were termed as “microplastics” and microplastic pollution becomes a global issue of emerging concern (Galloway and Lewis 2016; Eerkes-Medrano et al., 2015; Abel et al., 2018; Thompson et al.,

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2004).

Although a considerable amount of ecotoxicological studies have already been carried out, the real ecological and environmental risks of microplastics are still up for debate due to mismatching of microplastic concentrations and characteristics used in the laboratory experiments with those observed from the field (Andrady 2017; Lenz et al., 2016). Microplastics from real environments are more diverse and complex in types, shapes, sizes, and compositions, which are believed to be related to their toxicity (Lambert et al., 2017). In a recent review work, it was concluded that smaller and fiber shape microplastics are more toxic in general, which are related to the exposure duration and mode of actions of microplastics (Pirsaheb et al., 2020). Therefore, it is important to understand the processes relating to characteristics of microplastics in the environment for a better assessment of their true risks. Degradation of plastic wastes in the environment is considered to be a major process contributing to the formation of microplastics (Ivleva et al., 2017). Degradation and persistence of plastics in the environment is critical in determining their fate and effects, even though, knowledge on the environmental degradation of plastics and formation of microplastics is still limited and requires better understanding (Andrady 2017; Wu et al., 2012; Aragaw 2020).

Given the above background, the main aims of this review were to 1) identify hotspot areas for the accumulation of plastic wastes in the environment; 2) synthesize major mechanisms involved in the environmental degradation of plastics; 3) evaluate methods of advancement for the characterization of plastic degradation and microplastic formation; and 4) summarize studies investigating the degradation of plastics in the environment. At the end, research gaps were identified, and future research priorities were proposed. Only conventional plastics are considered, and biodegradable plastics are not the subjects of this review.

2. Hotspots for the accumulation of plastics

Plastic products turn into waste when their service life ends. The lifetime distributions for plastics of different industrial uses were estimated to vary from 1 year for packaging to decades for building and construction (Geyer et al., 2017). Though many efforts have been made to reduce the input of plastic waste into the environment, including by recycling, incineration, and landfills, plastic pollution is still inevitable. The overall recycling rate of post-consumer plastics was only 9%–30% in 2014 (Geyer et al., 2017). It was estimated that 60–99 million tons of plastics were inappropriately disposed of in the environment worldwide in 2015 (Lebreton and Andrady 2019).

When plastics enter the environment, they can be transported with wind and water flow due to their light weight (Law 2016). Precipitation, surface runoff, and riverine transport could be the main routes that transfer plastics from land to waters (van Emmerik et al., 2019; Rech et al., 2014). Although plastics in most scenarios have been transported from land to water, they can also be transported from water to land (Zhang et al., 2016a, 2019; Turrell 2018; Moreira et al., 2016). Within aquatic environments, there are many physical processes governing transport of floating plastic debris, such as open ocean processes, Stokes drift, internal tides, wind force, Langmuir circulation, ice formation, melting and drift, etc.(-van Sebille et al., 2020). Vertical transport is also possible due to biofouling or aggregation with other denser particles (Fazey and Ryan 2016). Besides, hyperpycnal flows could play an important role in vertical transport of plastic particles (van Sebille et al., 2020).

As a result of their various sources and multiple transport pathways, plastics have been detected worldwide, even in remote areas far away from human activities. The occurrence and abundance of plastics and microplastics in different environments worldwide have been reviewed in many previous works (Kumar et al., 2020; Wong et al., 2020; Pico et al., 2019). In aquatic environments, plastic accumulation could be largely affected by hydrodynamic conditions. Ocean gyres transport floating plastic debris into certain areas, often the center of the gyres—forming "garbage patches" (NOAA 2020). High abundances of plastic debris were discovered in the South Pacific subtropical gyre and the North Atlantic subtropical gyre (Eriksen et al., 2013; Law et al., 2010). On smaller scales, plastics could be carried by water current and accumulate in locations where current velocity has decreased, such as intersections of two or more water currents (Xiong et al., 2018a), a habitat behind a dam (Zhang et al., 2015), or estuaries (Zhao et al., 2019).

Sediment, including deep sea could be a sink for plastic debris. High density ($\rho > 1$ g/cm^3) plastics could settle at the bottom due to gravity, whereas low-density ($\rho < 1$ g/cm^3) plastics can also settle by a biofouling effect (Kooi et al., 2017; Chen et al., 2019). Flow velocity can significantly affect phytoplankton biomass, and a strong negative correlation could be found between phytoplankton biomass and the flow rate (Li et al., 2013). A lower flow rate could facilitate the formation of biofilm on the surface of plastics debris, resulting in a higher chance of settlement in a water column with a decreased flow rate (Zhang et al., 2017). Tidal zones and beaches are another important habitat for plastic accumulation. Plastics could be transported by high tide and be stayed during ebbs (Costa et al., 2011). In general, the habitat for accumulation of plastics is related to the input of plastic wastes, topographical features, and hydrological conditions.

3. Degradation mechanisms of plastics

Conventional plastic materials are very resistant to degradation in general. The longevity of plastics is estimated to be hundreds or even thousands of years depending on properties of the plastics as well as the surrounding environmental conditions (PlasticsEurope 2019). Although at a very slow rate, environmental weathering still causes the breakdown of plastics, which induces changes in polymer properties due to biological and/or abiotic processes. General processes of plastic degradation are illustrated in Fig. 1. More extensive degradation mechanisms and pathways for different plastics have been reviewed elsewhere (Debroas et al., 2017; Shah et al., 2008; Singh and Sharma 2008; Gu 2003).

3.1. Abiotic degradation of plastics

Abiotic degradation of plastics refers to change of physical or chemical properties that occurs for plastics due to abiotic factors such as light, temperature, air, water, and mechanical forces. Generally, abiotic degradation is expected to precede biodegradation due to a poor bioavailability of plastics (Andrady 2015).

3.1.1. Photodegradation of plastics

In the environment, photodegradation is recognized as the most important processes that initiate plastic degradation. Photodegradation of plastics usually involves free radical mediated reactions initiated by solar irradiation (Fig. 2). High energy ultraviolet (UV) irradiation UV-B (290–315 nm) and medium energy UV-A (315–400 nm) are mainly responsible (Liu et al., 2019a).

Polyethylene (PE) is resistant to photodegradation due to the lack of chromophores, but the presence of impurities or structural defects in polymers during manufacture or weathering can act as chromophores (Fairbrother et al., 2019). Carbonyl groups within the PE backbone can act as chromophores. Through the Norrish Type I and II reactions, radicals, end-vinyl, and ketone groups are formed

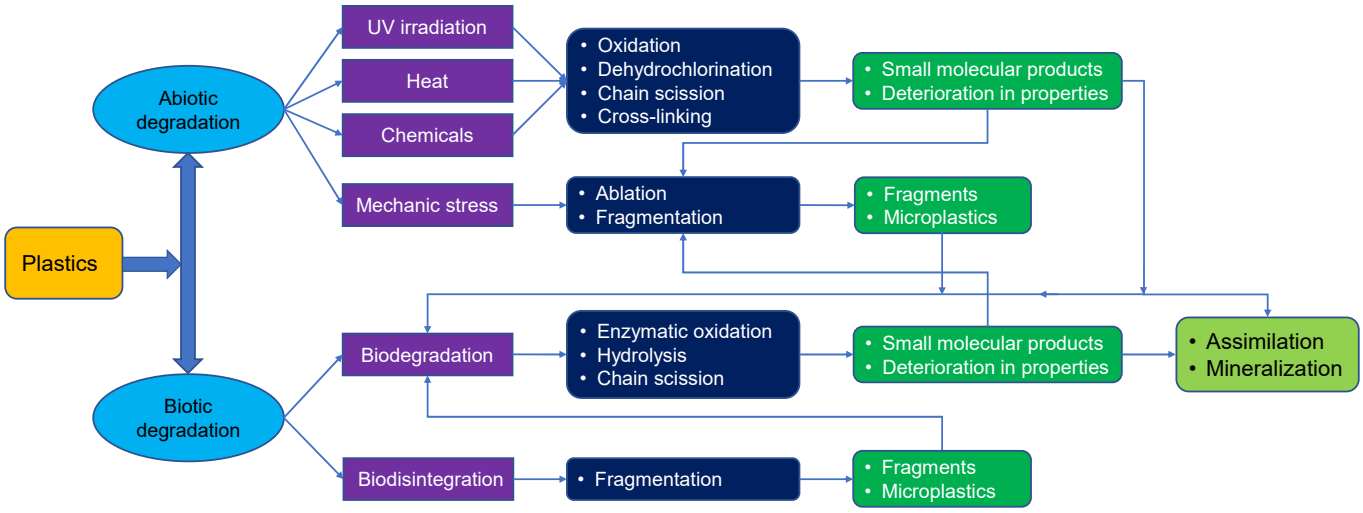

Fig. 1. A schematic diagram showing the general processes involved in the degradation of plastics.

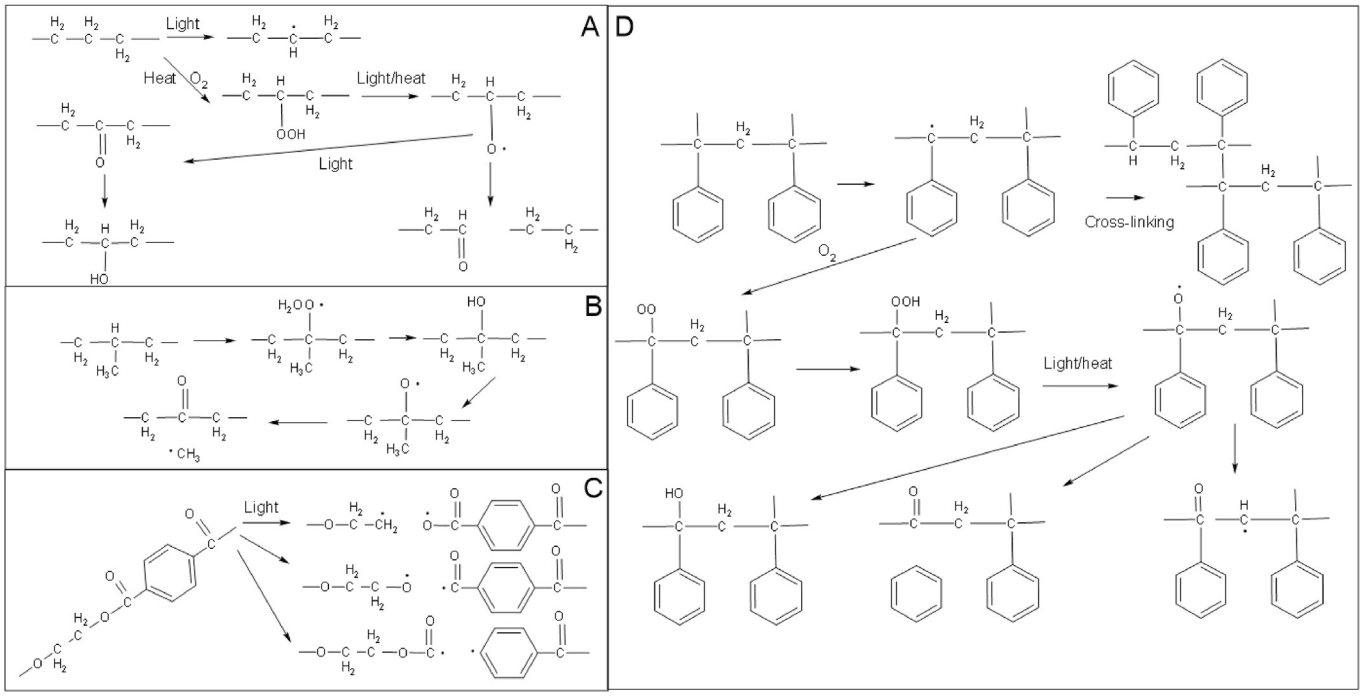

Fig. 2. Degradation of PE (A), PP (B), PET (C), and PS (D). Modified from (Fotopoulou and Karapanagioti 2017; Karlsson and Albertsson 2002; McKeen 2019a).

and cause a main-chain scission (Karlsson and Albertsson 2002). Free radicals can react with oxygen to produce peroxy radicals, which are converted to a peroxide moiety by hydrogen abstraction (Fig. 2A). The peroxide moiety dissociates into macro-alkoxy and hydroxyl radicals, which catalyze the subsequent reaction sequence. During the reaction sequence aldehydes, ketones, carboxylic acids, esters, and alcohols can be produced, and chain scission and cross-linking of polymers occur (Torikai et al., 1986).

PP is less stable than PE due to the presence of tertiary carbon, which is more susceptible to oxygen attack (Weber et al., 2011). The photodegradation mechanisms of PP are similar to those of PE (Fig. 2B). The presence of chromophores in PP due to impurities allows the formation of radicals under UV radiation. Subsequent radical mediated reactions lead to random chain scission and cross-linking, and degradation products of lower molecular weight are usually generated (He et al., 2019; Su et al., 2019).

Polyvinyl chloride (PVC) undergoes rapid dehydrochlorination under UV irradiation, and generates short sequences of conjugated unsaturation in the polymer. Unsaturated C=C double bonds are less stable and can be further photodegraded. Similarly, in PE and PP the presence of chromophores in PVC due to impurities can absorb UV radiation and generate free radicals. The free radicals form hydroperoxides that can break the double bonds of the backbone chain and generate smaller degradation products (Law 2016; Yang et al., 2018). A recent study revealed a new degradation route for PVC under environmentally relevant conditions, during which polyene structures were formed, and then oxidized to ketone and alcohol by O_2 and OH• radical (Wang et al., 2020).

Polystyrene (PS) is susceptible to photodegradation due to the presence of phenyl rings, which get excited and form triple state under UV radiation. Triplet energy of the excited benzene can lead to the dissociation of the phenyl group or be transferred to the nearest C–H or C–C bonds. In the absence of oxygen, a polystyryl radical is produced due to the cleavage of a C–H bond. In the presence of oxygen, the polystyryl radical is converted to peroxy radical, which reacts with the surrounding polystyrene molecule. Chain scission and cross-linking occur, and carbonyl compounds, styrene monomer and olefins can be produced (Fig. 2D) (Kumar et al., 2020; Dris et al., 2017).

Polyethylene terephthalate (PET) consists of alternating ethylene glycolate and terephthalate subunits, linked via ester bonds. Photodegradation of PET leads to cleavage of the ester bond directly, forming CO, CO_2, terephthalic acid, anhydrides, carboxylic acids, and esters (Fairbrother et al., 2019). PET can also be photodegraded via radical reactions. Hydroperoxide is formed through oxidation of the CH_2 groups adjacent to the ester linkage. Hydroxyl radicals formed by the cleavage of hydroperoxide can react with the aromatic rings in the polymer backbone to form hydroxyterephthalate groups (Fig. 2C). Radical intermediates and products can recombine to form cross-linked chains (Wong et al., 2020; Pico et al., 2019).

3.1.2. Thermal degradation of plastics

Thermal degradation refers to the breakdown of plastics due to energy input stemming from elevated temperature. Plastics can undergo thermo-oxidative reactions at high temperature. When sufficient heat is absorbed by the polymer to overcome the energy barrier, the long polymer chains can be broken generating radicals (Pirsaheb et al., 2020; Peterson et al., 2001). These radicals can react with oxygen and produce hydroperoxide in a process similar to what happens in the photodegradation of plastics, which cleave to form hydroxyl free radicals and alkoxy radicals. The reaction can self-propagate until the input of energy is discontinued or inert products are formed by collision of two radicals. During thermal degradation, both molecular reduction and enlargement may occur as a result of chain scission and cross-linking (Crawford and Quinn 2017).

The temperature requirement for the thermal degradation is related to the thermal properties of plastics and oxygen availability (Crawford and Quinn 2017). The glass transition temperature (T_g) and melting point (T_m) are important thermal properties of plastics. T_g is the temperature at which the polymer transforms from a glassy state to an elastic one. Plastics are flexible at temperatures above T_g but become rigid at temperatures below T_g due to changes of molecular mobility (Rudin and Choi 2013). Activation energy determines the temperature that initiates the thermal degradation of plastics. In the presence of oxygen, typical plastics containing hydrogen as a constituent give rise to exothermic oxidation at a temperature close to 200 °C, and higher temperatures are usually required for plastics with a higher T_m (Kotoyori 1972).

In the environment, exothermic oxidation is unlikely to happen due to a high temperature requirement, but slow thermal oxidation of plastics may occur in concert with photodegradation, especially on beaches or pavements that are exposed directly to sunlight. Meanwhile, temperature and UV radiation can have synergic effects on the degradation of plastics, and the rate of oxidative reactions also increases with the temperature (Kamweru et al., 2011; Andrady et al., 2003). In addition, increasing humidity was found to reduce the activation energy for the thermal degradation of plastics (Kotoyori 1972). 3.1.3 Chemical degradation of plastics.

In the atmosphere, the presence of pollutants such as ozone (O_3), sulfur dioxide (SO_2), nitrogen dioxide (NO_2), and volatile organic compounds (VOCs) can either attack plastics directly or catalyze the formation of radicals by photochemical reactions, which may also lead to the degradation of plastics (Crawford and Quinn 2017).

O_3 can be produced from O_2 by the action of UV and lightning, which is present at low concentrations in the atmosphere naturally, but the ground O_3 level increases due to air pollution by NO_x, SO_2, and VOCs (Placet et al., 2000). O_3 attacks the unsaturated C=C double bonds of the polymer, even at low concentrations. The reaction of O_3 with double bonds causes chain scission on the polymer chain. O_3 also reacts with saturated polymers, but at a much slower rate (Cheremisinoff 2001).

SO_2 can be excited by UV radiation, producing a reactive singlet or triplet state — which reacts with the unsaturated C=C double bonds directly — or producing O_3 via photochemical reaction with O_2. NO_2 is very reactive due to the existence of odd electrons in the molecule, which can easily react with the unsaturated C=C double bonds in the polymer. Similarly to SO_2, photochemical reaction of NO_2 with O_2 also produces ozone (McKeen 2019b).

In water environments, the most important chemical factors influencing plastic degradation are pH value and salinity of the water. High concentrations of H^+ (acidic) or OH^- (basic) in an aquatic environment may be able to catalyze the degradation of plastics that are susceptible to hydrolysis such as polyamides (PA) (Hocker et al., 2014; Wadsö and Karlsson 2013). These two factors can also alter the surface of other types of plastics and microplastics and influence their behavior in a water environment and toward other constituents and pollutants in the water (Liu et al., 2019b).

3.1.3. Mechanical degradation of plastics

Mechanical degradation refers to the breakdown of plastics due to the action of external forces. In the environment, external forces can come from the collision and abrasion of plastics with rocks and sands caused by wind and waves. Freezing and thawing of plastics in aquatic environments can also result in the mechanical degradation of polymers (Pal et al., 2018). The effect of the external forces depends on the mechanical properties of the plastics. Elongation at break, also known as fracture strain, reflects the capability of a plastic product to resist changes of shape without crack formation. Elongation at break varies from 1% to about 900% for different plastics (Crawford and Quinn 2017). Plastics with a lower elongation at break value are more likely to fragment under external tensile forces. Continued stress on plastics eventually results in chain scission of polymers (Sohma 1989).

Synthetic fibers are responsible for over 60% of world fiber consumption, with polyester, polyolefin, acrylic, and PA being the most common ones (Salvador Cesa et al., 2017). Mechanical degradation is particularly important for the degradation of synthetic fibers. Domestic washing has been found to be a major source of microplastic fibers, likely due to the shear, abrasion, and impact stresses on the synthetic fibers during laundering (Cesa et al., 2020). Moreover, wear and tear of garments were found to be capable of releasing microplastic fibers directly into the air, which is of equal importance to the release of microplastic fibers by laundering (De Falco et al., 2020).

Mechanical forces are mainly responsible for the generation of tire wear, brake wear, and road wear particles (Wagner et al., 2018). The interactions between a tire and a road surface as well as a brake pad and a brake disk yield frictional stresses, which tear fragments off the rubber surface directly. Tire treads are subjected to continuous stress during driving, and the rubber is pressed into a bulge, which creates prolonged stretching and causes material fatigue. Stress generated by abrasion with a road surface can reach the limiting strength of the material, resulting in micro-cutting or scratching of the tire and creating elongated rubber particles (Frank

Sommer 2018).

Chain scissions of polymers during photo, thermal, and chemical degradation will affect the mechanical properties of the plastics and particularly their tensile elongation at break and tensile modulus (Andrady 2017). Degradation in the environment was found to be able to decrease the elongation at break values of the plastics (O'Brine and Thompson 2010), which lowers the requirement of external forces for the fragmentation of plastics and facilitates the mechanical degradation of plastics.

3.2. Biotic degradation of plastics

Biotic degradation of plastics refers to the deterioration of plastics caused by organisms. Organisms can degrade plastics either physically by biting, chewing or digestive fragmentation (Cadée 2002; Dawson et al., 2018; Jang et al., 2018; Porter et al., 2019; Cau et al., 2020; Mateos-Cardenas et al., 2020) or biologically by biochemical processes (Danso et al., 2019). Microorganisms, including bacteria, fungi, and insects, can be mainly responsible for the biological degradation of plastics (Crawford and Quinn 2017). Strains with plastic biodegradation capabilities and their biodegradation pathways have been presented in several review papers (Shah et al., 2008; Sánchez 2019; Pathak and Navneet 2017; Wu et al., 2019).

Conventional plastics usually have an extremely low bioavailability due to their solid nature, as only a very small fraction of the polymer is exposed to the potential degraders (Battin et al., 2016). In addition, macromolecule polymers could not be directly used by microorganisms, and extracellular enzymes are required initially to break macromolecule polymers into small molecular products for cellular uptake and further metabolization (Chen et al., 2019). Abiotic degradation of plastics, which produces degradation products of low molecular weight and creates fractures and pores on the polymer surface, can accelerate the biodegradation processes (Wu et al., 2019).

Plastics can be categorized as hydrolysable or non-hydrolysable according to the presence or absence of ester or amide groups, which can be attacked by various extracellular hydrolases. Degradation of non-hydrolysable polymers such as PE, PP, and PVC by extracellular enzymes can be more complicated. Non-hydrolysable polymers resemble lignin due to their structural similarity. Therefore, enzymes involved in lignin biodegradation may also contribute to the biodegradation of non-hydrolysable polymers. Previously, laccase was found to play a major role in the biodegradation of PE by the actinomycete *Rhodococcus ruber* (Santo et al., 2013). Hydroquinone peroxidase was found to be responsible for the biodegradation of PS by *Azotobacter beijerinckii* (Nakamiya et al., 1997). It has also been suggested that several enzymes excreted by fungi are capable of decreasing the length of PE polymer chains (Sánchez 2019). Non-hydrolysable polymers can be oxidized by O_2 with the catalysis of those enzymes, resulting in the formation of degradation products of low molecular weight.

Hydrolysable polymers such as PET, PA, and polyurethane (PUR) are usually more susceptible to biodegradation due to the presence of existing biodegradation pathways such as extracellular hydrolases involved in the degradation of cellulose and proteins (Chen et al., 2019). PETase, an enzyme capable of hydrolyzing PET initially identified in *Ideonella sakaiensis* was found to be ubiquitous in the environment (Danso et al., 2018). Enzymes such as cutinase, lipase, serine esterase, and nitro-benzyl-esterase have also been found to be capable of hydrolyzing PET, whereas protease, cutinase, amidase, and hydrolase are involved in the hydrolysis of PA (Guebitz and Cavaco-Paulo 2008). Meanwhile, esterase and polyester hydrolase from bacteria and fungi might be responsible for the hydrolysis of PUR (Akutsu et al., 1998; Russell et al., 2011). In addition to hydrolysis, enzymatic oxidation may also contribute to the oxidative degradation of hydrolysable polymers (Magnin et al., 2020).

Hydrolytic and oxidative degradation of plastics by various extracellular enzymes results in chain scission of the polymer chain, producing short-chain polymers and small molecular fragments (e.g., oligomers, dimers, and monomers), and the degradation products can be taken up by microorganisms when their molecular weight is small enough (Chen et al., 2019). Small molecular degradation products can be assimilated and be subject to intracellular metabolism inside the microorganisms (Wilkes and Aristilde 2017). Eventually plastics can be mineralized into CO_2 and H_2O under aerobic conditions and into CH_4, CO_2, organic acids, H_2O, and NH_4 under anaerobic conditions due to both extracellular and intracellular processes, leading to the growth of microbial biomass. Nonetheless, biodegradation of plastics under anaerobic conditions is energetically unfavorable compared with that under aerobic conditions and may take a much longer time for complete mineralization (Gu 2003).

4. Degradation of plastics in different environments

Studies have been carried out to investigate the degradation of plastics under natural or simulated environmental conditions (Table 1). Most of those studies focus on the degradation of plastics in marine environments especially seawater. Plastics exposed on the surface or in the photic zone of seawater can be photodegraded by UV and result in weight loss, changes in appearance and texture, deterioration in mechanical properties, signs of oxidation, and alteration of physicochemical properties (O'Brine and Thompson 2010; Zhao et al., 2018; Iñiguez et al., 2018). Plastics exposed in seawater can be colonized by microorganisms that form biofilm, which affects the light transmittance of plastics and causes biodegradation (Sudhakar et al., 2007; Eich et al., 2015; Horton and Dixon 2018). Biodegradation by microorganisms in biofilm is the main cause of plastic degradation in seawater in the aphotic zone, and while photodegradation is mainly responsible for the initial degradation of plastics floating on the surface of seawater, biodegradation may take over once the plastic surface is covered with biofilm (Khoironi et al., 2020). In accelerated weathering conditions, formation of nanoplastics from degradation of plastics has been confirmed, and the concentration of nanoplastics increased over time during the degradation of the PS coffee cup lid (Lambert and Wagner 2016).

A few works have studied the degradation of plastics in marine sediment. PET bottles collected from the seafloor remained robust for approximately 15 years, and a significant decrease in the native functional groups was observed afterward (Ioakeimidis et al., 2016). PE carrier bags incubated in oxic and anoxic sediments from Eckernförde Bay showed no sign of biodegradation in 98 days (Nauendorf et al., 2016). Exposed in simulated water column and seafloor conditions for a year, plastics showed no significant weight loss trend but changes in carbonyl and crystallization indexes indicated early signs of degradation (Beltrán-Sanahuja et al., 2020). Plastics may be very persistent on the seafloor and in marine sediment due to shelter from UV, low temperature, low oxygen, and slow biodegradation.

In air, photodegradation of plastics also resulted in changes in appearance and texture, a decrease in mechanical properties, and alteration of physicochemical properties (Ojeda et al., 2011). Plastics in air sheltered from UV also showed color changes during exposure, and visible light and NO_2 were found to be the most important variables related to the degradation, followed by O_3

Table 1
Degradation of plastics in different environments.

Plastic	Exposure	Characterization	Consequences	Mechanisms	Reference
LDPE, PP, PS	24 h in a rotating laboratory mixer with sediment added	Visual inspection, fluorescence microscope, balance,	The coarser is the sediment, the more microplastics are generated.	Mechanical fragmentation	Chubarenko et al. (2020)
PE	33 days in seawater at 3 m depth and seafloor	SEM, Dynamometer	Biofilm attachment; Decrease in maximal elongation at break and force needed to break; Signs of degradation on the plastic surface	Unspecified	Eich et al. (2015)
PP	60 days exposure on surface water, at 50 and 170 cm depth in seawater	FTIR, Microscope, SEM-EDX	Decrease in organic carbon content; Changes in functional groups; Observation of erosion, discoloration, uneven blasting, and irregularity on surface	Photodegradation on surface and at 50 cm depth; Biodegradation at 170 cm depth	Khoironi et al. (2020)
PE	98 days in marine sediment	SEM, Raman, balance, Goniometer	Biofilm attachment; Decrease in contact angle	No sign of biodegradation	Nauendorf et al. (2016)
PE, PP, PS	3 months UV exposure in air, ultrapure water and seawater	FTIR, Raman, SEM	Changes in functional groups; Observation of granular oxidation, flakes and cracks on surfaces	Photodegradation	Cai et al. (2018)
LDPE	4 months buried in river bank, suspended from a tree and, tethered to the bank	Instron universal materials testing machine	Reduction in tensile strength and elongation at break	Photodegradation	Williams and Simmons (1996)
LDPE, HDPE, PP	6 months in seawater at 3 m depth	FTIR, DSC, SEM, AFM, Goniometer, Instron universal materials testing machine, Balance	Biofilm attachment; Weight loss; Changes in bond structures; Surface erosion; Increase in contact angle; Decrease in Crystallinity and T_m; Reduction in tensile strength and elongation	Photodegradation and biodegradation	Sudhakar et al. (2007)
Nylon, PE, PP, PET	6.5 months UV exposure in seawater	Instron universal materials testing machine, TGA, SEM, AFM	Loss of elasticity; Increase in rigidity; Weakening of the plastics; Observation of granular oxidation, flakes and cracks on surfaces; Loss of homogeneity on surface	Photodegradation	Iñiguez et al. (2018)
PVC	210 days thermal and UV exposure in seawater and air	SEM-EDX, surface area and porosimetry analyzer, FTIR	Changes in morphologies, surface area, pore volume, and pore size; Changes in functional groups;	Photodegradation and thermal degradation	Tang et al. (2018)
LDPE, HDPE	8 weeks in mangrove soil	Balance	Weight loss	Biodegradation	Kumar et al. (2007)
HDPE, PP, PS	32 weeks in an intertidal salt marsh	FTIR, SEM, Digital camera, Balance, Photometric radiometer	Weight loss; biofilm attachment; Decrease in light transmittance; Fragmentation and formation of microplastic fragments and fibers	Photodegradation and biodegradation	Weinstein et al. (2016)
HDPE, LLDPE, PP	280 days in air	FTIR, DSC, GPC, Instron universal materials testing machine	Reduction in average molar mass; Increase in carbonyl index; Decline in strain at rupture; Changes in Crystallinity	Abiotic degradation	Ojeda et al. (2011)
PP	40 weeks in seawater at 0.6 m depth	UV spectroradiometer, Instron universal materials testing machine	Decrease in maximum extension; Decrease in UV transmittance	Photodegradation	O'Brine and Thompson (2010)
LDPE, PP	12 months in air and seawater	Instron universal materials testing machine	Decreases in ultimate extension; Biofouling	Photodegradation	Pegram and Andrady (1989)
LDPE, PP, foamed PS	12 months on the flat roof and floating in an enclosed section of sea.	Instron Mechanical Testerc	PE: an increase in tensile strength after 2 months PP: extension loss PS: rapid yellowing, biofouling	Thermo degradation, biofouling	Andrady (1990)
PE, PP, EPS	UV exposure for up to 12 months followed by mechanical abrasion for 2 months	SEM, FTIR, Fluorescence microscope	Increase in carboxyl index; Observation of cracks on surface; Yellowing of surface; Fragmentation and formation of microplastics	Biodegradation and mechanical oxidation	Wu (2017)
PE, PP, PET, PA	One-year exposure in simulated euphotic, aphotic, and seafloor conditions	Balance, FTIR, DSC	Weight loss; Changes in functional groups; Changes in the melting enthalpy and crystallinity	Unspecified	Beltrán-Sanahuja et al. (2020)
PA, PVC, PS, PE, PP, PC, PUR	2-year in air sheltered from UV and precipitation	Colorimeter	Yellowing and changes in lightness	Chemical degradation and photodegradation	Pastorelli et al. (2014)
PE, PVC	24 months in soil at 5 cm depth	FTIR, AFM, spectrophotometer	Surface roughness increase; release of plasticizers	Biodegradation	Sullivan et al. (2019)
LDPE, HDPE, PP	36 months on roof or in tanks with seawater	FTIR	Changes in hydroxyl, carbonyl groups and carbon-oxygen bond structures; increase in opacity, yellowness, and brittleness	Unspecified	Zhao et al. (2018)

PET	Several years on seafloor at150–350 m depth	FTIR, SEM	Changes in functional groups; Development of cracked and uneven surface	Unspecified	Ioakeimidis et al. (2016)
PE	3 years in soil, open air, and marine environment	tensile testing machine, FTIR	Decreased tensile stress	Photodegradation and oxidation	Napper and Thompson (2019)
LDPE, PS, PVC	32 years in soil at 10 and 50 cm depth	FTIR, DSC, SEM, microscope, XRD	Whitened; Fragmentation; Observation of holes and ditches; Changes in functional groups; Decrease in initial oxidation temperature; Changes in crystallinity	Biodegradation and thermal oxidation	Otake et al. (1995)

*PE: polyethylene; LDPE: low density polyethylene; LLDPE: linear low density polyethylene; HDPE: high density polyethylene; PP: polypropylene; PS: polystyrene; EPS: expanded PS; PVC: polyvinyl chloride; PET: polyethylene terephthalate; PC: polycarbonate; PA: polyamide; PUR: polyurethane.

(Pastorelli et al., 2014). Photodegradation of plastics was inhibited in seawater compared to that in air, probably due to a lower heat buildup in samples exposed in seawater (Pegram and Andrady 1989; Tang et al., 2018). It was also found that the photodegradation level of plastics was higher in air and ultrapure water than in simulated seawater, and salinity and oxygen level were important factors affecting degradation (Cai et al., 2018).

Fewer studies have investigated the degradation of plastics in soils. In a biodegradation experiment for plastics buried in soil over 32 years, no evidence of biodegradation was observed for PS and PVC, but remarkable whitening was observed on LDPE film directly in contact with soil as a result of biodegradation (Otake et al., 1995). Nearly 5% weight loss was observed for commercial carry bags made of low-density PE (LDPE) and high-density PE (HDPE) buried in mangrove soil over a period of 8 weeks, which was related to the heterotrophic bacteria capable of producing hydrolytic enzymes isolated from the soil (Kumar et al., 2007). PE bags and PVC sheetings buried in soils of different environmental conditions for 2 years showed increased surface roughness over time, the degradation of PE was enhanced by increased moisture and an elevated soil pH, leaching of plasticizer from PVC was observed, and a moist environment had a more pronounced effect on the removal of plasticizer (Sullivan et al., 2019).

Degradation of plastics in an inland freshwater environment receives even less attention, where plastic pollution has also been found to be prevalent (Eerkes-Medrano et al., 2015; Horton et al., 2017; Wagner et al., 2014). LDPE samples positioned at natural riverine stranding levels underwent a rapid initial breakdown during the first week of exposure, followed by little further loss of tensile strength over a period of 4 months, and samples buried in the river bank retained the greatest ensile strength than those suspended from a tree and tethered to the river bank, suggesting that photodegradation is the principal cause of sample deterioration (Williams and Simmons 1996). The freshwater environment can be different from the marine one in several aspects, including sunlight spectrum and intensity, water physicochemical properties, and biological characteristics. Therefore, different plastics' degradation behaviors could be expected in freshwater environment.

In the majority of studies, only the early degradation stage was investigated, probably due to the very long time period required for the complete degradation of plastics (Garnai Hirsch et al., 2019), and the formation of microplastics has only been characterized in a few works. Weinstein et al. (2016) found that plastic strips exposed in a salt marsh were degraded abiotically in the first few weeks, but biotic degradation took over after 4 weeks due to biofilm attachment and microplastic fragments, and fibers were produced after 8 weeks as a result of surface delamination. In a simulated beach environment, a 12-month UV exposure and 2-month mechanical abrasion of PP and PE produced 6084 ± 1061 and 20 ± 8.3 particles/pellets, while a 12-month UV exposure and 2-month mechanical abrasion of expanded PS (EPS) produced 12,152 ± 3276 particles/pellets, and the number of fragmented microplastic particles produced by UV exposure and mechanical abrasion increased with decreasing size for all polymer types (Wu 2017). Advanced degradation and mineralization of plastics and microplastics in the environment have been insufficiently studied (Oberbeckmann and Labrenz 2020). The specific surface degradation rate (SSDR), a metric to harmonize disparate types of measurements, was proposed to extrapolate the half-lives of plastics, and linear extrapolation leads to estimated half-lives ranging from 58 years for bottles to 1200 years for pipes using a mean SSDR for HDPE in the marine environment (Aragaw 2020). A data-driven approach was used to elucidate the degradation trends of plastics by linking abiotic and biotic degradation behaviors in seawater with their physical

properties and molecular structures, a hierarchy of predictors such as T_g and hydrophobicity were revealed to quantify surface erosion, and plastics were ranked into fast, medium, and slow degradation categories (Min et al., 2020). However, more accurate prediction of plastics' fate in different environment is still difficult because plastics' degradation is a complex issue that depends on their own features and interactions with environmental conditions.

5. Characterization of degraded plastics and microplastic formation

Degradation causes the oxidation and chain scission of polymers, leading to changes in chemical composition, appearance and texture, physicochemical properties, and mechanical properties of the plastics. Therefore, changes in those properties during degradation can be used to characterize the degree of plastics' degradation (Tosin et al., 2012). In addition, degradation of plastics can result in weight loss and gas (e.g., CO_2 and CH_4) evolution, which can also be determined to reflect their biodegradability (Wu et al., 2012). Deterioration of plastics and subsequent mechanical surface ablation during degradation lead to their fragmentation and microplastics of various characteristics can be generated (Andrady 2017). Parameters and methodologies used for the characterization of plastic degradation and microplastic formation during the degradation processes are illustrated in Fig. 3.

5.1. Chemical composition

Plastics are composed of polymers, which are large molecules formed by the polymerization of monomers. The chemical composition of plastics depends on the chemical composition of monomers and the degree of polymerization. Meanwhile, chemical additives such as plasticizer, stabilizer, antioxidant, pigment, and flame retardants are commonly added to plastics to improve their performance and to facilitate their manufacture (Hermabessiere et al., 2017; Wei et al., 2015). Reactions may happen in the polymer chain as well as among the plastic additives during degradation that alter the plastics' chemical composition.

Degradation causes chain scission and the cross-linking of polymers, which directly lead to changes in their molecular weight. An average molecular weight is usually used because all polymer chains do not have the exact same mass (Ebnesajjad 2016). A decrease in the average molecular weight of the polymers has been observed as a result of photo- and biodegradation (Jabarin and Lofgren 1994; Tian et al., 2017). Size-exclusion gel permeation chromatography (GPC) is commonly used to determine polymers' molecular weight distribution, which separates polymer molecules based on their size as they pass through a column packed with porous resin beads. Viscometry and light-scattering measurements might also be used to measure the average molecular weight of polymers due to the quantitative relationships between viscosity,

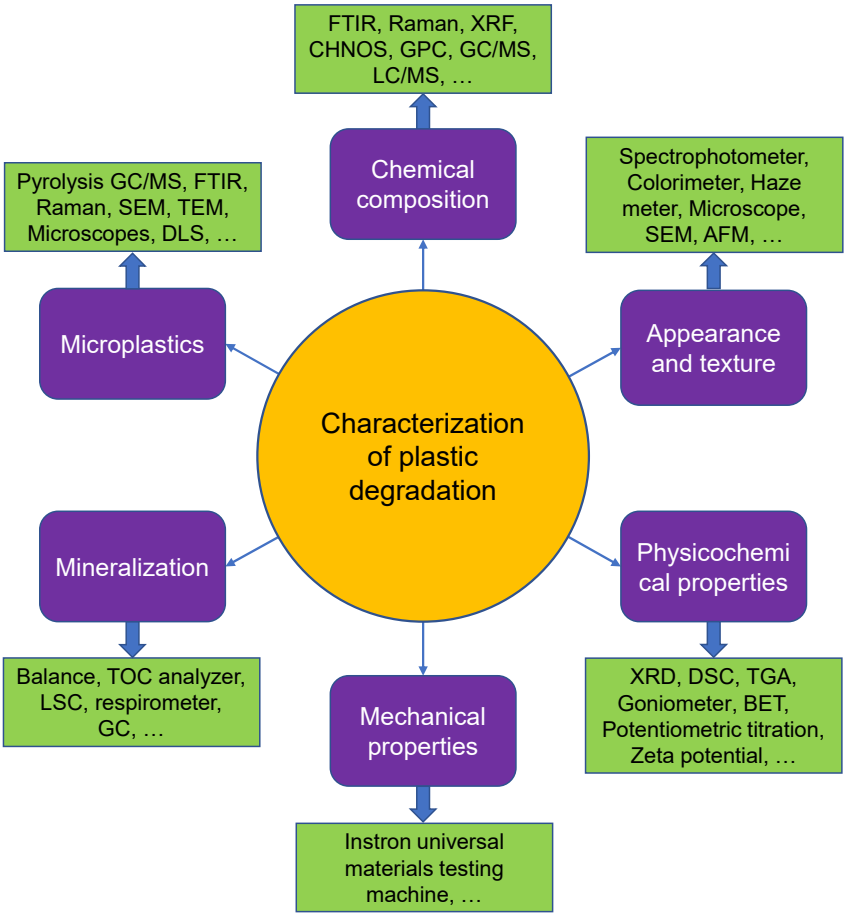

Fig. 3. A schematic diagram of parameters and methodologies used for the characterization of plastic degradation and microplastic formation during the degradation processes.

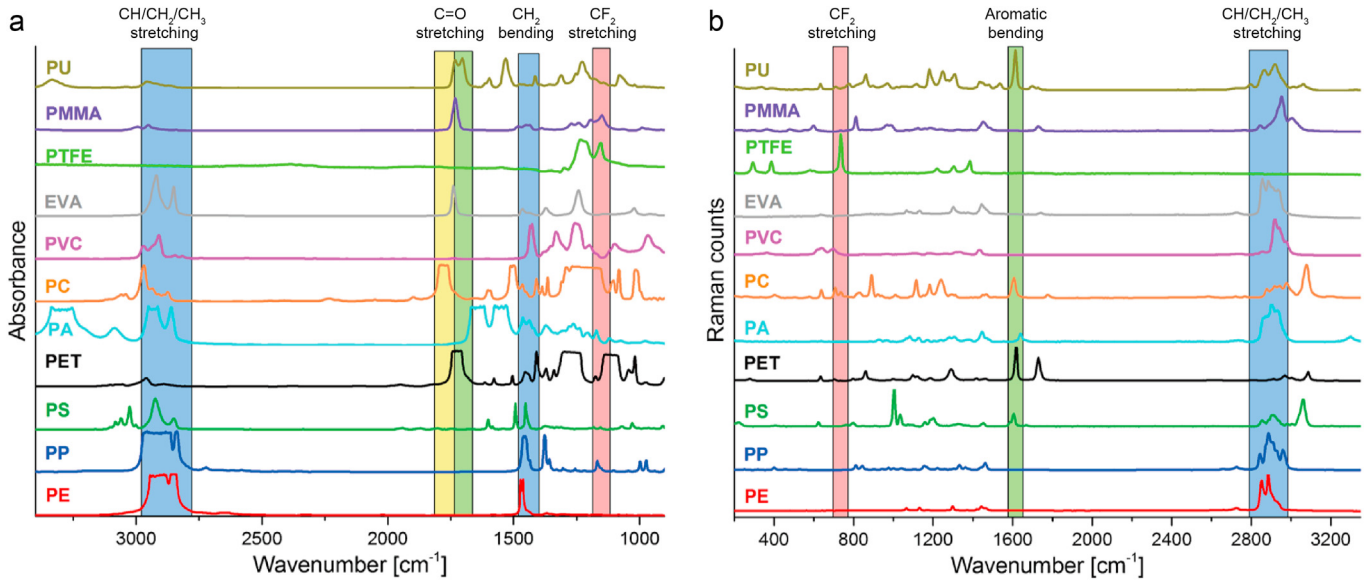

Fig. 4. FTIR and Raman spectra of the most common plastics (Modified from (Käppler et al., 2016)).

light scattering and molecular weight (Lewis and Gagg 2010).

Molecular spectroscopy technologies, such as Fourier transform infrared spectroscopy (FTIR), Raman spectroscopy, and nuclear magnetic resonance spectroscopy (NMR), are useful tools for detecting polymers' molecular bond information. FTIR measures absolute frequencies at which a sample absorbs infrared radiation, Raman spectroscopy measures relative frequencies at which a sample scatters radiation, and NMR measures the interaction of nuclear spins in organic compounds under a powerful magnetic field. FTIR and Raman spectra of the most common plastics are represented in Fig. 4, both of which are commonly used for characterizing the chemical changes of plastics during the degradation and identification of microplastics (Xu et al., 2019; Käppler et al., 2016). Carbonyl index (CI), defined as the ratio of the band area attributed to the carbonyl group −C═O and the band area attributed to the methylene group $-CH_2$, can be determined using FTIR and used to indicate the oxidation level of polymers (ter Halle et al., 2017).

The elementary composition of polymers can be determined using Elemental analyzer and X-ray fluorescence spectrometry (XRF). Elemental analyzer is the most common technique used for the elemental analysis of CHNOS in a sample based on the combustion of the sample. Upon combustion, C, H, N and S are converted to gaseous combustion products (e.g. CO_2, H_2O, NO_2, etc.), which are quantified using a gas chromatography (GC) subsequently, whereas O can be determined using a GC after pyrolysis of the sample in the absence of O_2 (Krotz and Giazz 2017). XRF is a simple, accurate, and nondestructive technique for the elemental analysis of many types of materials. XRF is based on the phenomenon that individual atoms emit X-ray photons of a characteristic wavelength when excited by an external energy source, and the elements can be quantified by counting the number of photons of each energy emitted from a sample (Aidene et al., 2020).

Small molecular degradation products and released additives can be analyzed using GC or liquid chromatography (LC) coupled with mass spectrometry (MS). Nonpolar and volatile compounds can be analyzed using GC/MS. Polar and non-volatile compounds are usually analyzed using LC/MS but may also be analyzed using GC/MS after being derivatized to volatile products. Degradation products and released additives can be analyzed quantitatively by comparing to standards of known concentrations, and qualitatively by matching them with the reference mass spectra for GC/MS (by electron ionization) and LC-MS/MS (by tandem MS) from the MS database such as the one provided by the National Institute of Standards and Technology (NIST). High resolution mass spectrometry (HRMS), such as time-of-flight (TOF), Fourier-transform ion cyclotron resonance (FT-ICR) and Orbitrap, allows more accurate mass measurement and can further facilitate the identification of degradation products (Prasse and Ternes 2016).

5.2. Appearance and texture

Changes in the appearance and texture of the plastics during degradation can be observed more easily. Appearance properties such as the color, gloss, haze, and yellowness of plastics can be measured instrumentally following standard methods developed primarily by standard organizations such as American Society for Testing and Materials (ASTM) and International Organization for Standardization (ISO) (McKeen 2014). Color is usually measured with colorimeters or spectrophotometers. Gloss can be determined using a glossmeter by projecting a beam of light at a fixed intensity and angle onto a surface and measuring the amount of reflected light at an equal but opposite angle. Haze describes the amount of light scattered when light passes through a transparent object, which reflects the plastics' internal bulk property. Haze meters and spectrophotometer can be used to measure the level of haze. The yellowness indicates the degree of color shifting from whiteness to yellowness or from transparency to dark yellow. Yellowness index can be calculated from spectrophotometric data obtained using a spectrophotometer.

Cracks, flakes, pits, and groove textures can be developed on the surface of the plastics during degradation and are usually observed in the plastic samples collected in the field (Xiong et al., 2018b; Zbyszewski et al., 2014). These surface textures can be examined with the assistance of optical microscopes, scanning electron microscopes (SEMs), and atomic force microscopes (AFMs) (Bläsing and Amelung 2018). Surface textures of typical plastic samples are presented in Fig. 5. Optical microscopes use visible light and a system of lenses to magnify an object, which have a resolution limit of 200−250 nm. SEM uses a focused beam of electrons as a probe to

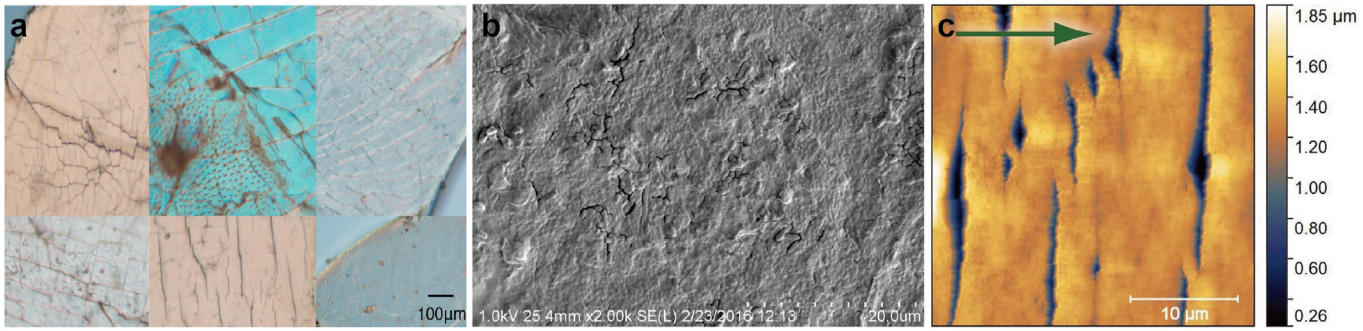

Fig. 5. Surface textures of typical plastic samples characterized using optical microscope (a), SEM (b), and AFM (c) (Modified from (Xiong et al., 2018b), (Zhang et al., 2016b), and (Julienne et al., 2019), respectively).

generate images of a sample by scanning the material surface, and its resolution can be down to below 10 nm. AFM, a type of scanning probe microscopy, maps the material surface using a micro-fabricated cantilever as a probe. AFM provides high resolution images at the nanometer scale and can measure the surface roughness (Nayfeh 2018).

5.3. Physicochemical properties

Physicochemical properties such as crystallinity, thermal properties, and surface properties of plastics can be altered during the degradation. The polymer chains present in either amorphous or crystalline forms in plastics. Amorphous polymer chains have no order, and their arrangements are random, whereas crystalline polymer chains have short-range order and align together to form ordered structures. Crystallinity reflects the fraction of the plastics' crystalline polymer chains and can vary from 0% to >90%, which determines their rigidity and T_m (Balani et al., 2014). Increased crystallinity of plastics was observed during the early stage of photodegradation because the amorphous region is usually more susceptible and may be degraded preferentially (Jabarin and Lofgren 1994). The crystallinity of plastics is usually determined using X-ray diffraction (XRD) and differential scanning calorimetry (DSC), but Raman spectroscopy and FTIR might also be used (Kann et al., 2014; Stuart 1996; Aggarwal et al., 2008). XRD is a relatively accurate method for polymer crystallinity analysis, which measures intermolecular order as a result of chain packing. DSC provides a rapid method for determining polymer crystallinity based on the heat required to melt the polymer. The heat can be evaluated in terms of % crystallinity by ratioing against a polymer of known crystallinity to obtain relative values (Andrady 2017).

T_g and T_m are important thermal parameters of plastics. In the amorphous region, the polymer molecules are in a frozen state at lower temperatures, referring to the glassy state. However, the polymer chains can wiggle around when heated, and the polymer becomes soft and flexible, entering the rubbery state. The glass transition occurs only in the amorphous region of polymers, and the crystalline region remains unaffected during the glass transition. When the temperature reaches T_m, all of the ordered polymer chains begin to change to a disordered phase and the plastic changes from solid to liquid (Hale 2002). Only semi-crystalline polymers have true T_m, whereas amorphous polymers possess only T_g. T_g and T_m depend on the mobility and flexibility of polymer chains, which are affected by intermolecular forces, pendant groups, stiffening groups, and cross-linking (Balani et al., 2014). The T_g and T_m of plastics can be measured using DSC by monitoring the heat flow associated with phase transitions as a function of temperature (Müller and Michell 2016).

Thermal stability is another important thermal property that indicates the ability of a plastic material to resist the action of heat and to maintain its chemical composition and mechanical properties. Thermal stability can be determined by thermogravimetric analysis (TGA), which monitors the weight that changed over time as a function of temperature changes. The thermal stability of polymers is related to their chemical structure, crystallinity, and molecular weight, and it can be modified by incorporating plastic additives (Sinha Ray 2013). Additionally, TGA can be used in combination with FTIR and MS to simultaneously measure thermal stability of plastics and characterize their degradation products generated at different stages of the thermal degradation (Peñalver et al., 2020).

Surface properties of plastics such as hydrophobicity, surface charge, surface area, and porosity are related to their interactions with other pollutants and microorganisms in the environment (Cai et al., 2019; Fotopoulou and Karapanagioti 2015). Hydrophobicity reflects the ability of a plastic surface to repel water, which can be determined by the contact angle measurement between the water droplets and the surface using a goniometer. Plastics such as PE, PP, PS, PET, and PVC are highly hydrophobic, and degradation can reduce their hydrophobicity by introducing hydrophilic functional groups to the polymer chain (Wilkes and Aristilde 2017). The charges on the plastic surface depend on the ionization of end-groups and ionizable moieties in the polymer. It was found that virgin PE was neutral at seawater pH but that weathered PE was negatively charged due to the formation of ketone groups (Fotopoulou and Karapanagioti 2012). Surface charge can usually be determined through Zeta potential analysis or potentiometric titration. The Brunauer-Emmett-Teller (BET) method has been used extensively to characterize the surface area and pore size of a material by measuring the physical adsorption of inert gases using a surface area and porosimetry analyzer. Environmental exposure leads to changes in plastics' surface area and pore volume probably due to the reshaping of the polymer surface by degradation reactions (Fotopoulou and Karapanagioti 2012).

5.4. Mechanical properties

Mechanical properties of plastics are critical in determining their applications. Mechanical properties are usually determined using an Instron universal materials testing machine following standard methods developed by ASTM and ISO (McKeen 2014). Tensile properties, such as elongation at break and tensile modulus, can be derived from a stress-strain experiment. Elongation at break, as mentioned above, reflects the percentage changes in the length of the material before fracture. Tensile modulus, also known as Young's modulus, is the ratio of stress to the strain in the linearly

elastic region. The primary measures of shear properties include shear strength and shear modulus, which can also be determined in a stress-strain experiment. Shear strength is the maximum shear stress a material can withstand without structural failure, whereas shear modulus is the response of a material to the shear deformation calculated as the ratio of shear stress to shear strain.

Plastics' mechanical properties are related to their chemical composition and physicochemical properties, such as molecular weight, crystallinity, cross-linking, and additives. Therefore, deterioration in mechanical properties can usually be observed during the degradation of plastic products in the environment due to the impact on their chemical composition and physicochemical properties (Arhant et al., 2019; Adelhafidi et al., 2015). Highly degraded plastics will eventually lose their integrity when the external forces from the environment exceed their mechanical strength, leading to fragmentation of the plastics and the formation of microplastics (Andrady 2017).

5.5. wt loss and gas evolution

Plastics can lose their total mass during degradation and be converted to CO_2 or CH_4 under different environmental conditions. Weight loss is commonly used to indicate the degradation of plastics, which can be directly measured using a balance of high accuracy. Additionally, weight loss can be reflected indirectly through total organic carbon (TOC) measurement using a TOC analyzer that determines the organic carbon concentration of a sample by analyzing CO_2 produced by combustion. Weight loss may result from not only the mineralization of plastics but also the release of additives. Therefore, weight loss may not be adequate to prove mineralization (Oberbeckmann and Labrenz 2020). Polymers labeled with radioisotopes such as ^{14}C may be used to characterize the mineralization of plastics unequivocally by measuring the changes in radioactivity during degradation by liquid scintillation counting (LSC) (Wan et al., 2016). However, radioisotope-labeled polymers are complex to synthesize, are not available for all plastics, and may have safety issues (Wu et al., 2012).

The production of CO_2 or CH_4 can also be used to indicate the mineralization of plastics (Rose et al., 2020). Standard methods based on the measurement of CO_2 or CH_4 evolution for biodegradability test have been developed by ASTM and ISO (Wu et al., 2012). CO_2 can be quantified using GC or a respirometry system with an infrared detector, whereas CH_4 is usually analyzed using GC. However, CO_2 or CH_4 evolution may require a considerably long time when studying the mineralization of conventional plastics due to their inherent low biodegradability.

(Silva et al., 2018; Shim et al., 2017; Möller et al., 2020; Prata et al., 2019).

6. Conclusions

In the terrestrial environment, soils under intensive human impact are hotspots for the accumulation of plastic waste. Inland waters, urban lakes and riverbanks have been found to be more vulnerable to plastic pollution. While in the marine environment, ocean current convergence zones, beaches, and seafloors are potential destinations of plastic waste. Plastics are subjected to abiotic and biotic degradation processes involving chemical, physical, and biological reactions in the environment. UV radiation is mainly responsible for the initiation of degradation. Many advanced technologies have been developed to characterize the degradation of plastics. Degradation causes oxidation and chain-scission of plastic polymers, forming low molecular weight degradation products and causing changes in physicochemical and mechanical properties. Deterioration in tensile strength and shear strength as a result of degradation leads to the fragmentation of plastics, and small sized plastic debris can be generated with the assistance of external forces. Digestive fragmentation is proven to be an important pathway of forming plastic debris. Advanced degradation of plastics can produce degradation products with a low enough molecular weight that can be assimilated and mineralized by microorganisms. In the natural environment, the degradation of conventional plastics is extremely slow and affected by their own features and the conditions of the exposed environment.

The degradation of plastics is critically important in determining their fate and effects in the environment. Microplastics produced during the degradation of plastics have become an issue of increasing concern in recent years because they are more prevalent and may cause potential health risks to the ecosystem and humankind. Most of the current research on the environmental degradation of plastics has been focused on the early stages of degradation, thus, the formation of microplastics under different environmental conditions is still poorly understood, and the persistence of microplastics in the environment is virtually unknown. Future research should further identify the key environmental parameters and properties of plastics affecting the degradation so as to predict the fate of plastics in different environments and to facilitate the development of technologies for reducing plastic pollution. The formation and degradation of microplastics, including nanoplastics, should receive more research attention in order to better assess their fate and ecological risk in the environment.

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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