Removal Mechanisms of Polyethylene, Polypropylene, Polyvinyl Chloride, Polyamide (Nylon), Polystyrene and Polyethylene Terephthalate in Wastewater Treatment Plants by Chemical, Photocatalytic, Biodegradation and Hybrid Processes

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Abstract: - In this review study, polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), polyamide (nylon) (PA), polystyrene (PS) and polyethylene terephthalate (PET); Removal mechanisms through chemical, photocatalytic, biodegradation and hybrid were investigated in wastewater treatment plants (WWTPs). Plastic products are used in almost all aspects of our daily life. Due to their low cost, portability, durability and resistance to degradation, these plastic products affect the health of the environment and biota on a global scale. Therefore, the removal and mineralization of microplastics (MP) is an important necessity in the 21st century. The prevalence of MPs in aquatic ecosystems due to their high degradation resistance and bioaccumulation; It has become an important environmental problem in recent years. Plastic wastes; Gradual discharge, specific removal techniques, inadequate standard detection methods, and slow destruction rates of MPs lead to the ubiquity of these wastes. Evidence shows that MPs act as a potential vector by adsorbing different heavy metals, pathogens, and other chemical additives widely used in different raw plastic production. MPs are ingested by aquatic creatures such as fish and different crustaceans, and finally, people ingest them at the tertiary level of the food chain. With this phenomenon, MPs are responsible for the blockage of the digestive tract, the disruption of digestive behavior and, finally, the reduction of reproductive development of all living organisms. After all, MPs; has become an emerging potential threat and a source of increasing concern; This situation urgently requires the control of MPs in aquatic environments. Similar studies in the literature; It was evaluated in order to shed light on different studies that can be done on this subject in the future.

Key-Words: - Biodegradation; Chemical processes; Hybrid processes; Microroplastics; Photocatalytic; Polyamide (Nylon); Polyethylene; Polyethylene Terephthalate; Polypropylene; Polystyrene; Polyvinyl Chloride.

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1 Introduction

Synthetic polymers are made by linking together hundreds or thousands of organic subunits (“monomers”) via strong covalent chemical bonds. The first fully synthetic polymer, Bakelite (made by a condensation reaction of phenol with formaldehyde) dates to the early 20th century, but true mass production of polymers began only in the 1950s. Since then, global manufacturing has grown exponentially, reaching 380 million of tones per year (Mt/y) in 2015, [1]. Today, thousands of polymer types are produced on a commercial scale, [2]. The largest market shares belong to low-cost, commodity thermoplastic polymers, henceforth referred to as “plastics.” Thermoplastics and thermoset plastics are the two key plastic categories. While a thermoset plastic cannot be re-melted, a thermoplastic can be re-melted. The main categories of thermosets are epoxy resins, polyurethane (PU), silicones, and vinyl esters; thermoplastics include polypropylene (PP), polyethylene terephthalate (PET), polyethylene (PE) polyvinyl chloride (PVC), polystyrene (PS), polyamide (PA) (Nylon), and polycarbonate (PC), [3, 4]. The most common raw polymers include PET, high, low, and linear-low density PE [High Density Polyethylene (HDPE], Low Density Polyethylene (LDPE), and Linear Low-Density Polyethylene (LLDPE)], PVC, PP, PS and PA.

Thompson et al. [5] used the term “Microplastics” (MP) for small pieces of plastic that occur distinctly in nature. The origin of MPs was shown in Table 1. MPs include microfibers, micro-flakes, pellets, spheroids, bead granules or fragments; They are produced either directly by anthropological activity, or as secondary MPs as a result of the breakdown of larger plastic fragments by photodegradation, biodegradation, or mechanical means., [5-9].

* Table 1 can be found in Appendix section.

Plastics with different sources, shapes, and types decomposed into MPs with diameters smaller than 5 mm, [6, 10, 11], they are decomposed through photodegradation, thermal oxidation, thermal degradation and possible biodegradation, [6, 12]. MPs have the characteristics of small size, large specific surface area and remarkable chemical stability, [13]. Types of MPs in wastewater was illustrated in Fig. 1.

* Figure 1 can be found in Appendix section.

Degradation of plastic; The physicochemical properties of polymers depend on environmental conditions such as weather conditions, temperature, irradiation and pH values. MP particles in aqueous bodies are a potential threat to humans as well as other aquatic species; is a growing source of concern. MPs are considered an emerging environmental contaminant and their potential adverse effects on living things have received much attention. MPs are difficult to degrade, small in size and widely available in nature; It has become a big problem recently due to its impact on the environment, [14]. A new problem caused by MPs in 2018; This is due to the ingestion of pollutants and toxic substances that adhere to the surface of MPs by animals when they enter the food chain. These MPs are toxic therefore, they threaten animal and human health, [15-19]. For example, plastic particles less than 130 µm in diameter has been found to potentially trigger localized immune responses by translocating into human tissues, [20].

MPs have been detected in rivers, deep oceans, marine sediments and the atmosphere, [21]. However, MPs also have the ability to adsorb other harmful organic pollutants and trace metals, [22, 23], from the environment onto their surface, results in the transportation of these pollutants, [23]. The prevalence of MPs and its relationship with other pollutants poses a risk to the environment as marine life readily consumes MPs causing harm directly through physical interactions or by the adsorbed toxins and bacteria, [24]. MPs are consumed by humans via contaminated food and drinks, or via breathing contaminated air, [25, 26].

Conventional methods for the removal of MPs have been reviewed extensively and generally the relevant 3 classes: physical separation, chemical separation, and biological separation, [21-24, 27-34]. Physical separation includes techniques such as filtration, sedimentation and density flotation, [35]. High removal efficiencies are generally achieved with these methods. However, in these methods, while filtering is performed due to membrane contamination; There are disadvantages such as high energy, material cost, ineffectiveness for sedimentation and flotation against smaller particles (<100 µm), [36-38]. Chemical separation techniques, include coagulation and flocculation and photodegradation, [39]. These techniques can provide high removal efficiency; However, in the case of coagulation and agglomeration, they may be inefficient in removing smaller particles with a certain shape, and in the case of photodegradation, the risk of releasing metabolite contaminants into the environment increases, [40]. In biodegradation, lower efficiency is observed due to the resistance of these pollutants, [36, 37, 39, 40]. All MPs removal methods were summarized at Fig. 2.

* Figure 2 can be found in Appendix section.

MPs particles are used in a number of cosmetic and personal care products, including washing liquids, soaps, facial and body scrubs, toothpaste, and lotions. Most of the MPs used in personal care products are generally PE and PP which can end up in municipal wastewater treatment plants (WWTPs) and ultimately in the environment since present WWTPs are designed to remove organic matter but not MPs. As tertiary treatment processes commonly used to remove MPs from wastewater in WWTPs; Ultrafiltration (UF), coagulation, reverse osmosis (RO) and Membrane bioreactor (MBR) are used, [41-43]. 90–99% removal efficiency has been reported, [44-47], MPs of 20–300 µm in size still have problems to be removed, [44-47], and MPs in discharged water up to 0.25 particle/l has been detected, [43]. Furthermore, the sludge as residuals of WWTPs processes containing MPs may be used as agricultural fertilizers that subsequently finds its way into the groundwater, [48].

Magnetic nanoparticles have emerged as an alternative for the removal of MPs, [49]. In this method a magnetic sorbent is added to the polluted water and the analyte is adsorbed by the magnetic material which is then removed via an external magnet. This method ensures high MP removal rates, simple application, and highly efficient removal of smaller MPs, [50]. A range of nanomaterials have been employed for removal of MPs with the most common type being functionalised magnetic iron or iron oxide nanoparticles, [51]. Other magnetic materials based on biochar, zeolite can be also used. However, magnetic nanomaterials that enable efficient and simultaneous removal of MPs and their released small organic molecule-based components/additives are still lacking.

In the removal of MPs; Visible light photocatalysis is a promising environmentally friendly, low-cost and efficient process capable of mineralizing a wide range of organic pollutants into H_2O and CO_2, [52]. This process offers advantages such as the utilization of sunlight as a clean energy source, high degradation efficiency, and the generation of harmless by-products. It is based on the use of suitable wide bandgap metal oxide semiconductor materials such as titania (TiO_2) or zinc oxide (ZnO), that upon interaction with light give rise to the formation of different reactive species. When ZnO, TiO_2 or similar semiconductors are excited by light sources with energy greater than their natural band gap, charge separation is created in the form of free electrons excited from the valence band positions to the conduction band. This excitation simultaneously leads to a hole formation in the valence band. Both free electrons and holes react with H_2O, OH^- and O_2 adsorbed on the surface of the semiconductor, producing reactive oxygen species (ROS) such as hydroxyl ($OH^{\bullet}$) and superoxide ($O_2^{-\bullet}$) radicals. These species initiate the polymer degradation process, leading to chain scission and completion of mineralization to H_2O and CO_2, [53]. The photocatalysis process is described by the following in Eq. 1, Eq. 2, Eq. 3 and Eq. 4:

$$Photocatalyst \overset{hv}{\rightarrow} h^+ + e^- \quad (1)$$

$$h^+ + H_2O \rightarrow OH^{\bullet} \quad (2)$$

$$e^- + O_2 \rightarrow O_2^{-\bullet} \quad (3)$$

$$Organic\ pollutant + ROS \rightarrow CO_2 + H_2O \quad (4)$$

Recent methods investigated for the treatment of MPs waste are biodegradation and photocatalysis. Biodegradation of MPs can be achieved by microbes producing enzymes that break the macromolecules into smaller fragments which can potentially lead to complete mineralization, [54]. For instance, biodegradation of PP MPs using *Bacillus cereus* and *Bacillus gottheilii* bacteria has been investigated and it has been found that long exposure time is needed in order to achieve high removal efficiency, [55].

In this review study, polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), polyamide (nylon) (PA), polystyrene (PS) and polyethylene terephthalate (PET) in wastewater treatment plants (WWTP); Removal mechanisms were investigated using chemical, photocatalytic, biodegradation and hybrid processes. Similar studies in the literature; It was evaluated in order to shed light on different studies that can be done on this review subject in the future.

2 Degradation Mechanisms of MPs

The analysis of plastics degradation can be grouped into methods associated with assessing the elimination of small molecules, methods which assess chemical changes (hydrophobicity, functional groups) in the polymer structure, and methods which record physical changes in materials properties (tensile strength, surface morphology, crystallinity, etc.).

2.1 Assessing Bond Cleavage

2.1.1 Mass Loss

The simplest and most direct way to measure the degree of degradation of polymers is to measure changes in their mass. Quantification of mass loss has been used to assess degradation in soil, [56-58], in compost, [59, 60], and in microbially enriched lab settings, [61-64]. Because degradation takes place at the surface, the rate of mass loss is closely related to (and is typically proportional to) the surface area of the plastic piece, [65, 66]. Partial conversion to small molecules (including but not limited to CO_2 and H_2O) and their resulting volatilization or solubilization leads to a reduction in the mass of nonvolatile or insoluble polymeric material, [67]. Thus, the overall mass loss convolutes the liberation of small molecules with the flaking of larger, insoluble pieces, including microplastics (0.5- 5 mm) and mesoplastics (5−200 mm), [6, 68]. Currently, our knowledge of how plastic breaks down and MPs are formed is incomplete. Therefore, recent reports suggest that the shape of the plastic piece affects its fragmentation behavior in the ocean, and that small fragments with a low aspect ratio break down faster because their isotropic motion inhibits biofilm development, [69].

Little or no mass loss may be observed in the initial phases of degradation, [70]. Instead, the mass may actually increase at short exposure times due to O_2 incorporation and/or the attachment of microorganisms, [71, 72]. Clinging biomass and other debris can also accumulate in surface cracks and pits that develop during degradation. Thus, extremely long experimental times are usually necessary to obtain meaningful results. Since mass loss measurements alone are not easy to interpret or extrapolate, it is desirable to combine this method with some of the other analytical techniques, [73].

2.1.2 CO_2 Evolution

CO_2 is the ultimate fate of carbon under aerobic polymer degradation conditions, [74-77], (although polyesters can produce some CO_2 under anaerobic conditions), [78, 79]. Its formation is frequently used as an indicator of biological degradation. In anaerobic conditions, soluble carbon compounds are metabolized by methanogens or sulfate reducers, producing CH_4 and CO_2, respectively, [80, 81]. The polymer degradation rate is inferred by measuring the CO_2 liberated during abiotic or biotic mineralization in a controlled environment, [82-84]. CO_2 can be quantified by trapping and titration methods, [85], or by analytical techniques such as gas chromatography with thermal conductivity detection (GC-TCD), [77], and infrared (IR) spectroscopy, [86]. The CO_2 yield is defined as in Eq. 5:

$$CO_2(\%) = \frac{n_{CO_2,test} - n_{CO_2,control}}{n_{CO_2,theoretical}}\, x\, 100\% \qquad (5)$$

where, $n_{CO2,test}$: is the total accumulated amount of CO_2 product from polymer degradation, $n_{CO2,control}$: is the amount of CO_2 released in a blank experiment, and n_{CO2},theoretical is the total amount of CO_2 that would be liberated by complete mineralization of the polymer sample, [87]. However, on short time scales, polymer carbon is unlikely to be completely oxidized to CO_2. Therefore, to measure the kinetics of polymer degradation; The use of CO_2 evolution as a probe should be applied with caution.

2.1.3 Gel Permeation Chromatography (GPC)

This method reveals changes in the molecular weight, an important parameter in polymer degradation, by size exclusion, [82, 88, 89,]. Reduction in the molecular weight of partially degraded polymers has been observed during both biotic and abiotic degradation processes, which increase the concentration of chain ends and can lead to mineralization of the smaller polymer chains, [89]. GPC requires the polymer to be dissolved in a carrier solvent, which for polyolefins requires high temperatures. Care must be taken to ensure that dissolution of the polymer or the high-temperature measurement conditions do not cause further degradation, [90].

2.2 Assessing Changes in Chemical Functionality

2.2.1 Chemical Analysis

Nuclear magnetic resonance (NMR) and IR spectroscopies can easily detect the presence and concentration of certain functional groups in polymers, even at low concentrations, [91-94]. For example, 13C CP/MAS NMR reveals the formation of cross-linked polymer chains (via a peak at 39.7 ppm) upon γ-irradiation of HDPE, [95]. IR is particularly valuable for detecting polar functional groups, such as ketones and ester carbonyls (intense peaks at ca. 1715 and 1735 cm^{-1} , respectively), which are typical of oxidative degradation pathways, [96, 97, 98, 99]. The extent of oxidation during degradation can be measured from the change in absorbance for carbonyl stretching according to C–H stretching modes, [100, 101]. However, a recent study used this carbonyl index to measure the extent of oxidation relative to other modes; It suggests that it may be less accurate, such as methyl deformation in the case of PP, [102].

2.2.2 Contact Angle

Changes in the surface density of polar functional groups, for example, those formed during oxidative degradation, cause changes in the surface energy, which are reflected in the contact angle with liquids, [103, 104]. Hydrophilic surfaces, with their high wettability, have higher surface energies and give lower contact angles with water. Thus, formation of polar functional groups in polymers due to environmental weathering effects (e.g., UV exposure) results in a decrease in contact angle. Increased hydrophilicity promotes the attachment of microorganisms to the polymer surface, further accelerating the degradation rate, [105].

2.3 Assessing Changes in Materials Properties

2.3.1 Dynamic Mechanical Analysis (DMA)

This technique is typically used to characterize polymer strength. Changes in the tensile strength and elongation at break are also indicators of physical deterioration during polymer degradation, [106, 107]. Changes in these mechanical properties are associated with the formation of cracks and pores at the surface, as well as a reduction in molecular weight, [108].

2.3.2 Thermal Analysis

This method generally involves heating or cooling a sample at a controlled rate while monitoring its physical characteristics, [109-111]. Differential scanning calorimetry (DSC) measures heat capacity (C_p), melting temperature (T_m), and glass transition temperature (T_g), [112]. A decrease in T_g during polymer degradation results from a decrease in the average chain length, due to the higher motility of shorter chains, [113]. Thermal gravimetric analysis (TGA) records mass changes that occur upon heating. When coupled with product analysis, this method can provide information on the nature of the decomposition, such as oxidation or loss of volatiles, during thermal degradation, [114].

2.3.3 Surface Analysis

Surface modification of polymers during degradation can be detected with scanning electron microscopy (SEM) and atomic force microscopy (AFM), [115-117]. These methods can directly image topographic changes on the polymer surface, such as the formation of holes and cracks, increased roughness, and even the adhesion of microbes to the surface, [118]. Typical changes in polymer morphology are visible as cracks and cavities; surface degradation and deterioration of HDPE films can be seen after 6 months in the marine environment, [119].

3 Removal Mechanisms of MPs

The main MPs removal technologies in different aquatic environments was determined at Fig. 3.

* Figure 3 can be found in Appendix section.

In this article, chemical, photocatalytic, biodegradation and hybrid MPs removal processes are examined in detail.

3.1 Chemical Degradation Process

In this process, catalysts play significant roles to generate reactive oxygen species (ROSs) and thus trigger the degradation of MPs.

3.1.1 Advanced Oxidation Processes (AOPs)

AOPs are known as a powerful method to eliminate organic pollutants through generating ROSs with high standard reduction potentials, such as, sulfate radical ($SO_4^{-\bullet}$, $E_0 = 3.1$ V vs NHE) and hydroxyl radical ($OH^\bullet$, $E_0 = 2.7$ V vs NHE), [120-122]. Due to their strong oxidation capability, a large variety of pollutants including dyes, antibiotics, and POPs have been effectively degraded or mineralized by this technique, [123]. As a unique type of organic pollutant, MPs are obviously more challenging to be degraded due to their considerably higher molecular weights compared to other low molecular weight organic pollutants as mentioned above. Several recently reported pioneering studies assert the degradation of MPs via this protocol. Although previous studies have proven that ROS generated through AOPs can destroy the surface structure of MPs particles [124], investigating AOPs in MPs degradation of pollutants is extremely rare.

Wang et al., [125] were the first to use $SO_4^{-\bullet}$-based AOPs (SR-AOPs) to degrade MPs. In their report, a helical-shaped, N-doped carbon nanotube catalyst encapsulated with manganese carbide nanoparticles (Mn@NCNTs) was fabricated for SR-AOPs reaction, while PE beads obtained from a few commercial facial cleansers were used as MPs samples, [125]. Under a hydrothermal (HT) condition created in an autoclave, the PE-MPs beads first generated cracks, fused into a thin polymer film and then progressively developed into a film with a large number of cavities as evidenced by SEM analysis. A remarkable 54 wt% weight loss of MPs was achieved by this method after reaction for 8 h at 160°C. The decay of MPs removal efficiency in the first three cycles were slight, indicating the outstanding stability of Mn@NCNTs in the SR-AOPs system. From the perspective of the reaction mechanism, the authors verified that the presence of SR-AOPs is indispensable, which continuously supplies the free radicals ($SO_4^{-\bullet}$ and $OH^\bullet$) to oxidize MPs particles into small molecules and then mineralizing them into CO_2 and H_2O. A hydrothermal condition plays a crucial role in this process as well: The generated bubbles and vapor mechanically shear the PE beads causing chain scission of the macromolecules and triggering the degradation process; meanwhile, the generation of ROSs is also effectively accelerated given that the activation of peroxymonosulfate (PMS) can be heat-driven. The authors proposed that the mechanism of this process is as such: The C–C bond of PE backbone is first broken into two hydrocarbon radicals under a HT conditions. Subsequently, when further decomposition of the hydrocarbon radicals occurs, the lower-weight molecules are generated and are further converted into new shorter-chain hydrocarbon radicals via the β-scission and hydrogen abstraction routes, induced by other hydrocarbons. Finally, these intermediates radicals are attacked and ultimately mineralized by the $SO_4^{-\bullet}$ and $OH^\bullet$ generated from SR-AOPs system.

Alternatively, EAOPs based on Fenton or Fenton-like chemistry (electro-Fenton, EF) most frequently produce ROS (e.g., $OH^\bullet$) and persistent organic pollutants with properties such as versatility, superior efficiency, excellent environmental compatibility, and sustainability. It is used to decontaminate POPs, [126]. During this process, the cathode material plays the key role in determining the degradation efficiency of the system: H_2O_2 is generated in situ on the cathodes by reducing O_2 via a two-electron oxygen reduction reactions and is subsequently converted into $OH^\bullet$, [127-131]. Gao et al., [132], designed a heterogeneous TiO_2/C cathode in an EF-like system for degrading PVC-based MPs in a Na_2SO_4 electrolyte (0.05 m, pH=3.0), where a graphite electrode and Ag/AgCl electrode were used as the counter electrode and reference electrode, respectively. At the end of the reaction, the original smooth surface of PVC MPs was destroyed by showing a number of large holes according to SEM characterization. After potentiostatic electrolysis at −0.7 V under 100°C for 6 h, a dechlorination and degradation efficiency of 75% and 56 wt% were obtained, respectively, through quantitative analysis of the concentration of Cl^- in the electrolyte and weight loss of the PVC MPs. The evolution of organic intermediates (e.g., formic acid, acetic acid, propionic acid, oxalic acid) during the EF treatments further proved the destruction of PVC into small organics. The input of electrons in such EF-like systems continuously boosts the generation of catalytic species. By characterizing the chemical structures of degrading intermediates and residual material in the electrolyte, the authors proposed the reaction mechanism of dichlorination and degradation process. First, the direct electrons transfer from TiO_2/C cathode to the PVC MPs gives rise to the dechlorination under heating conditions. Meanwhile, the attack of oxidative $OH^\bullet$ toward PVC MPs results in the formation of oxygen-containing groups such as C=O and O–H, combined with the release of liquid shorter-chain intermediates.

Subsequently, the vulnerable hydrocarbon species are prone to be oxidized into small molecular weight organics and finally mineralized to CO_2 and H_2O. Given the advantages of the relative mild conditions under 100°C and −0.7 V (vs Ag/AgCl), such an EF-like technology is promising for potential applications in WWTPs to purify the MPs-contaminated water bodies.

3.2 Photocatalytic Degradation Process

When plastics are exposed to ultraviolet (UV) or sunlight irradiation alone, their polymer macromolecules can directly absorb photons and generate excited states, leading to chain scission, branching cross-linking and oxidation reactions, [133]. However, it takes at least hundreds of years for plastics to degrade, [134]. Consequently, to achieve the photodegradation of plastics, suitable catalysts play an essential role throughout the process. The mechanism of photocatalysis has been extensively studied, especially under the catalysis of semiconductors.

When plastics are destroyed in the presence of photocatalysts, their degradation can be briefly attributed to the following procedures: (i) capturing energy from light to produce electron–hole pairs, (ii) collecting charge from the internal homogeneous distribution of the photocatalysts on the surface, and (iii) stimulating redox reactions by excitation of charges at the interface between the reactants and photocatalysts is triggered, [135-137]. When the photocatalyst absorbs light with sufficiently high energy, electrons are excited from the valence band to the conduction band, $e_{(CB)}^-$, and subsequently holes are formed on the valence band, $h_{(VB)}^+$, [138]. One strategy for photocatalytic reaction is conducted in an oxygen-rich atmosphere, which is called the O_2 evolution reaction (OER), [139]. During the degradation process, O_2 reacts with free electrons and is reduced to $O_2^{-\bullet}$, in the valence band. Simultaneously, the holes in the valence band migrate to the surface and react with H_2O to form $OH^\bullet$, or directly with MPs (Eq. 6, Eq. 7 and Eq. 8), [140], (Table 2).

* Table 2 can be found in Appendix section.

Then, the $O_2^{-\bullet}$, react with the $e_{(CB)}^-$ and H^+ produced from H_2O to form H_2O_2 (Eq. 9), which further reacts with photons, $O_2^{-\bullet}$, and $e_{(CB)}^-$ to form $OH^\bullet$, OH^- and O_2 (Eq. 10, Eq. 11 and Eq. 12), respectively. $O_2^{-\bullet}$ and $OH^\bullet$ are among the most active photocatalytic oxidants, which can efficiently oxidize organic compounds (Eq. 13), [141, 142]. OER photodegradation treats MPs as wastes to be degraded and generates highly oxidizing radical species, subsequently leading to nonselective oxidation reactions, [143, 144].

Another type of photocatalytic reaction is carried out in inert atmosphere, usually in N_2 or Ar, which is usually called the H_2 evolution reaction (HER), [145]. In this case, MPs wastes function as misplaced hydrocarbon resources, enabling the production of value-added products and H_2, [146]. Different from photodegradation with O_2, the holes generated by photons on photocatalysts drive the transformation of MPs to produce relatively highly selective value-added chemicals (Eq. 16), while the photoinduced electrons react with protons simultaneously to form H_2 (Eq. 16), [147, 148]. The overall photocatalytic reaction process of MPs of these two different strategies is summarized in Table 2.

The photocatalytic effect is greatly related to the bandgap of photocatalysts, which can dominate the beginning and ending of the reaction process. A narrower bandgap allows MPs degradation to be conducted in a more facile way, and thus a broader range of materials can be chosen as photocatalysts (Fig. 4).

* Figure 4 can be found in Appendix section.

Such as, TiO_2 and ZnO have bandgaps of 3.2 eV, [149, 150], while that of C_3N_4 is 2.7 eV, CN-CNTs-NM is 2.4 eV, [151], polypyrrole (PPy) is 2.2 eV, V-substituted phosphomolybdic acid (VPOM) is 2.19 eV, [152], and PET-derived carbon nitride sheets (PCNS) is 1.82 eV, [153]. During degradation, the measured activation energies vary with different properties. In the case of high-density polyethylene (HDPE), the activation energy is in the range of 85.6 ± 27.6 kJ/mol for the formation of a carbonyl group and 71.5 ± 8.9 kJ/mol for the formation of vinyl, [154]. Meanwhile, in the case of low-density polyethylene (LDPE), the reported activation energy for carbonyl formation fluctuates in the range of 46 to 95 kJ/mol. For other degradation products, namely vinyl and hydroxyl groups, the activation energies are lower, which are between 20 and 46 kJ/mol, [155-157]. The Gibbs energy changes (ΔG^o) have also been reported in some studies. For example, the ΔG^o for the reforming of lactic acid (a monomer of PLA) is +27 kJ/mol, while that for ethylene glycol (a monomer of PET) is +9.2 kJ/mol, [158, 159]. In general, plastic disposal requires a lower potential, hence lowering the total potential required to accomplish the OER and HER reactions, making photocatalytic degradation more accessible, [146, 159]. Under these conditions, plastic wastes resemble sacrificial materials that are easily oxidized by holes induced by $O_2^{-\bullet}$, $OH^\bullet$ or low-energy light.

The photocatalytic degradation, allows breakdown of polymers when irradiated with high-intensity photons such that simpler monomers are derived. The current photocatalysts industries provide a wide range of options for carrying out this method of degradation of MPs. Therefore, only a handful of such photocatalysts have been able to exhibit high efficiency. For instance, a novel hydroxy-rich ultrathin photocatalyst, BiOCl, facilitated degradation of MPs due to enhanced production of surface $OH^\bullet$, [160].

An attempt at exploring metal oxide NPs (MONPs) for photocatalytic degradation of MPs have also been carried out. ZnO nanorods adhered onto glass fibers which demonstrated visible-light-driven degradation of PP spherical MPs, [161]. Their study reported that upon irradiation for two weeks, the generation of products, such as acetone, butanol, acetaldehyde, and formaldehyde, were additionally observed. These generated by-products exhibit significant potential for utilization in various industrial applications. More recently, due to the multi-fold advantages of photocatalysts, MXene/$Zn_xCd_{1-x}S$ photocatalysts which successfully exhibited photocatalytic H_2 production and PET degradation, [162]. It is evident that photocatalysts can prove to eliminate MPs from various systems, while simultaneously bringing forth additional benefits such as value-added products and energy generation.

3.2.1 Photocatalytic Reactors

Photochemical systems in addition to reactors have adopted conventional solar thermal collector designs, including parabolic troughs and non-concentrating collectors because the required hardware for solar photocatalysis is quite comparable to that required for thermal applications, [163]. However, it should be noted that contrary to solar thermal processes, solar photochemical processes utilize solely high energy with short wavelength photons. For instance, a TiO_2 photocatalyst operates under UV or near-UV sunlight (300 to 400 nm), and a photo-Fenton heterogeneous photocatalyst operates within sunlight wavelength radiation below 580 nm. The prime benefits and shortcomings of concentrating and non-concentrating collectors for solar photocatalytic applications are as listed in Table 3, [164]. Quantitative comparisons of photoreactor designs are quite challenging due to the broad diversity of photoreactor designs, operating conditions, photocatalyst design and preparation, changes in solar intensity, and kinds of pollutants. As a result, the comparisons are primarily qualitative and is focused on the practical aspects of each design, [165].

* Table 3 can be found in Appendix section.

3.3 Biodegradation Process

Various mechanisms for biodegradation of plastics and MPs have been proposed. Lucas et al., [166], suggested that the biodegradation of plastics and their fragments occurred first by cleaving enzymatically the polymers into their oligomers and monomers which were then assimilating by microbes. Similarly, Gu and Gu, [167], proposed that the biodegradation started with the cleavage of polymer backbone or its side chains by the act of extracellular enzymes, which resulted in the formation of smaller polymer units (i.e. monomers, oligomers). In most cases, it involves the hydrolysis of amides (in polyamides), esters (in polyesters), or urethane (in polyurethane, PU) bonds where the extracellular enzymes act as catalysts. In addition, abiotic hydrolysis can facilitate the polymers for cleavage, [168]. The cleaved and simpler molecules are then absorbed and metabolized by the microorganisms. These alterations further promote biodegradation (depolymerisation and mineralization). Plastics and MPs can undergo biodegradation both in the aerobic and anaerobic environments, [169], but three main conditions should be maintained: (1). Presence of appropriate microbes capable of depolymerizing polymer substrate and mineralizing monomeric fractions by enzymes through metabolic pathways; (2). Proper environmental conditions for the biodegradation (e.g. temperature, pH, nutrients); (3). Morphology of the polymer substrate should be favorable for the microbial attachments and formation of biofilms.

In addition, a number of biotic and abiotic factors can affect the uptake and biodegradation of MPs even though the appropriate microbes are present in the environment (Fig. 5). For the uptake of MPs (as a food of the microbes), physiological characteristics of MPs, their size and feeding type act as the major biotic factors; while temperature and pH are abiotic factors. The polymeric substrate's molecular weight, chemical composition, hydrophobicity, size of the invaded molecules, and other environmental conditions (e.g. temperature) were reported as essential factors. Obtaining an optimum environmental condition helps to improve the effectiveness of microbial treatments for MPs removal. A combination of field-based assessments and laboratory-based trials is required to facilitate the application of microbes for the degradation of different types of plastics and MPs under different environmental conditions.

* Figure 5 can be found in Appendix section.

An emerging green method of degrading MPs is through the usage of biological catalysts. This process of biodegradation can be carried out by using bacteria, enzymes, fungi, and even larvae. These biocatalysts can simply adsorb MPs as carbon source, as well as degrade MPs to produce simpler non-toxic monomers. For instance, novel bacterium *Ideonella sakaiensis* 201-F6 used PET as a source of carbon and energy to produce two benign monomers, [170]. Biodegradation proves to be a surprising method as polymers pose an inert and recalcitrant nature in our ecosystems. Being a relatively newer approach, with limited knowledge, researchers aim to explore this concept and develop sustainable and reliable methods for bioremediation of MPs using fungi, biofilms, bacteria and bacterial consortiums, [171].

Recently, Auta et al. [172] demonstrated that *Bacillus cereus* and *Bacillus gottheilii* were able to break down MPs. These bacterial strains consumed MPs by using them as a carbon source, [172]. It was observed that *Bacillus cereus* enzymatic mechanisms cause the weight loss of PE, PET, and PS by 1.6%, 6.6% and 7.4%, respectively, while *Bacillus gottheilii* causes the weight loss of PET, PP, PS, and PE by 3%, 3.6%, 5.8% and 6.2%, respectively. *Exiguobacterium* species for developing biofilms on the surface of PS, which successfully achieved weight loss of 8% and 8.8%, respectively, [173]. Fungal strains such as *Aspergillus tubingensis*, *Penicillium simplicissimu, Zalerion maritimum* and *Aspergillus flavus* are also capable of exhibiting efficient MP degradation, [174].

Besides microbes, microbial enzymes have also been explored as an alternative, such as lignin peroxidases, proteases and lipases for degradation of PE, PU and PET, respectively, [175-177]. The intrinsic chemical additives in MPs may reduce the effectiveness of microbial enzymes, [178]. The usage of enzymes proves to be time-consuming and a costly process. Additionally, in order for microbial enzyme colonies to function well, optimal conditions must be created, which is a difficult process in natural systems. Considering these limitations, microbes are a preferred choice for biodegradation of MPs as they obviate the need for laborious time-consuming procedures involved in the extraction and purification of microbial enzymes. Furthermore, microbes can be efficiently utilized in regenerative cycles, thereby enhancing the overall effectiveness and cost efficiency of biodegradation.

Research has been expanded into insects-mediated biodegradation techniques as well, [179]. A recent study shows that *Zophobas atratus* larvae were successfully able to degrade PS and LDPE without the generation of any MPs, [180]. Conjointly, enhanced efficiency of biodegradation of MPs may be achieved by pre-treatment through thermal and photoreactive methods. Thus, persistent research and development in the area of MP biodegradation have the potential to significantly accelerate the mitigation of MPs from all ecosystems.

3.4 Hybrid Process

Several technologies like UF, membrane fouling (MF), polymeric membranes, nanofiltration (NF), RO and hybrid techniques can be incorporated with the membrane technologies like electrochemical processes, adsorption processes, membrane fouling, and AOPs for superior MPs removal efficiency. For MBR, the design and performance of filters are essential for the effective removal of MPs, [181].

Membrane-based hybrid techniques have been considered to be a highly efficient and cost-effective manner for the removal of contaminants like MPs from various types of H_2O bodies. The most affected are the aquatic organisms who end up consuming materials already containing the MPs which can be very dangerous for them as well as humans. It can block the digestive tract and can negatively affect the reproductive functioning process, thereby affecting the reproductive growth of the offspring. Hybrid membrane technology has proven beneficial for wastewater treatment and counters the accumulation of MPs and Nano plastics in the water.

Magnetic extraction, adsorption, chemical, and electrocoagulation are promising, but they are limited in use due to longer contact time, chemical consumption, and lower removal rate. In this phenomenon, hybrid treatment technology should be incorporated for the complete removal of MPs from wastewater. The incorporation of microbial treatment with membrane technology is highly promising. Still, it is worth mentioning to investigate the impact of operational conditions, including membrane surface charge, membrane material, fouling phenomena, transmembrane pressure, pore size, and hydraulic retention time to MPs removal. Therefore, investigation on sample pretreatment (ozonolysis, solvolysis) is also notable before going for microbe's hybridization utilization to ensure maximum removal rate.

MPs-targeted wastewater treatment technology is not fully developed, and no specific treatment process aimed at MPs removal has been applied in full-scale WWTP yet. In wastewater treatment, significant improvements have been achieved during the last few years in the application of a variety of hybrid treatment technologies, which consist of the combination of different treatment technologies to obtain the maximum MPs removal efficacies, [182]. Recently, hybrid systems have been widely used for the removal of MPs from water and wastewater. Talvitie et al., [183] observed the removal efficiency of various types of MPs from WWTP effluents by advanced treatment technologies including disc filter (DF), rapid sand filtration (RSF), dissolved air flotation (DAF) and MBR. They concluded that MBR-UF hybrid process treating primary effluent could remove 99.9% of MPs of almost all sized (> 20 μm) and all shapes, and showed that DF-coagulation and flocculation-DAF hybrid processes removed 98.5% and 95% MPs respectively.

Porous membranes and biological process combination could enhance the MPs removal efficiency up to 99.9%, [183]. The performance of a municipal WWTP operating based on a combination of primary treatment and pilot MBR technology for the removal of MPs, [184]. This hybrid system was found to achieve high retention capacity of MPs over 98.3%. Similarly, the combination of MBR-RO is an effective advanced technology for the wastewater treatment to produce high-quality water, [185, 186]. RO influences the performances of MBR in wastewater treatment, and RO performance is commonly impacted by membrane fouling from inorganic, organic and biological fouling, [187]. MBR-based anaerobic/anoxic/aerobic (A/A/O-MBR) systems effectively removed MPs from influent by trapped in sludge and block into permeate (effluent) through micromembrane (pore size < 0.1 μm) filtration, [188]. This structure could eliminate necessarily all MPs from wastewater. A combination of sorption and filtration methodologies with biological and sedimentation processed showed an excellent efficiency for the treatment of MPs containing wastewater. MBR with other advanced physical and chemical treatment showed higher MPs removal efficiency in the WWTP. While MBR coupled with sorption and filtration process exhibited high removal percentage of MPs from the influent of water treatment plant, [189]. MBR based hybrid systems are more effective for the removal of high MPs concentrate influent.

Electrocoagulation (EC) and agglomeration, coupled with additional filtration stage, showed effective separation of MPs from water. Ma et al., [190] demonstrated that the coagulation process could remove more than 36.89% of MPs from water. Coagulation with sedimentation could enhance the removal efficiency up to 81.6% MPs of the secondary sediment effluent, [191]. Chemical coagulation treatment is the most widely used process to combine with the physical process including UF, DF, RSF, Granular activated carbon (GAC) and RO process to reduce the fouling problem or enhance the removal performance. Coagulation is often coupled with rapid sand filtration, membrane filtration and ozone oxidation in tertiary treatment of WWTPs, [191]. This hybrid treatment, including coagulation-RSF, coagulation-ozonation and coagulation-DF achieved the removal efficiency of MPs by 84.8%, 95.2% and 96.2%, respectively. Compared with coagulation (47.1-81.6% removal) and RSF (73% MPs removal), membrane filtration (MF) (79.4% MPs removal) and ozone oxidation (89.9% MPs removal) showed better performance to remove MPs. Small MPs are trapped with flocs which formed in coagulation and stopped during filtration. Primary and secondary treated three different WWTP in Daegu, South Korea was finally treated through different tertiary treatment process combined with coagulation, [191]. They showed that MPs' overall removal rate in different WWTPs is 99.2%, 99.1% and 98.9%, when using the ozonation, membrane disc filter, and RSF in the tertiary stage. UF process coupled with coagulation as a pretreatment which is one of the main water treatment technology to remove organic contaminants from wastewater and produce high-quality effluent in current WWTPs, [192]. UF-Coagulation hybrid system is not perfectly designed for MPs removal from wastewater, [193]. Recently Ma et al., [190] observed the performance of UF-coagulation to remove PE MPs for potential application in drinking water treatment. After coagulation slight membrane fouling was induced due to the formation of loose cake layer by flocs, although PE particles were completely eliminated during drinking water treatment. However, after coagulation with PE particles (especially small size), membrane fouling was gently eased to increase the action of UF membrane, [190].

Coagulation/flocculation combined with sedimentation (CFS) and granular filtration is applied to MPs (180 nm – 125 µm) removal in drinking water treatment, [194], but with unsatisfactory removal. MBRs combined the biological activated sludge process with membrane separation provided MPs free effluent. Many studies have shown MBR hybrid systems to be more effective in the removal MPs from water up to 99.9%, [195, 196]. GAC filtration could effectively remove contaminants such as MPs through a synergistic combination of physical adsorption and biodegradation from the effluent of ozonation, [197]. It is commonly coupled with ozonation process to remove some emerging contaminants, [198, 199], and larger molecular weight matter is converted into a small fraction to enhancing the biodegradability of the influent of GAC filter during drinking water treatment, [200]. When ozonation combined with GAC filtration, it increases the removal efficiency of MPs by approximately 17.2%-22.2%, [197]. So ozonation-GAC filtration couple process reduced 74-83.1% MPs from the final effluent of drinking water treatment plant. GAC filtration also coupled with sand filtration and sedimentation/flotation process for treated drinking water treatment plant, [201].

Coagulation/flocculation, sedimentation, sand and GAC filtration hybrid process removed 81% MPs and coagulation/flocculation, flotation, sand filtration and GAC filtration hybrid treatment reduce the MPs 83% in drinking water treatment. In an advanced drinking water treatment plant (coagulation + sedimentation + sand filtration + ozonation + GAC), the overall MPs removal efficiency was 82.1%-88.6%, of which 82.9%-87.5% 73.1%–88.9% fibers fragments and 89.1%–92.7% spheres were removed, [197]. With GAC filtration combined with coagulation and sedimentation only, the MPs removal was reduced to 56.8%-60.9% where 1-5 µm MPs removal was 73.7%-98.5%. Therefore, the combination of coagulation and sedimentation removed only fiber types MPs at 40.5%-54.5%. In the coagulation/sedimentation process, it was found that the larger size MPs had a higher removal efficiency. MPs > 10 µm were almost completely removed, followed by the removal efficiency of 44.9%–75.0% for 5–10 µm in this process. Despite the high removal efficiency of MPs about 99%, the conventional WWTP with primary and secondary treatment is not specially designed to improve the quality of final effluent. Different technologies could be combined before application to remove MPs in the WWTPs. Among them, a few physical and chemical treatment showed better performance when they use as combined with another process. The effective removal rates of MPs in tertiary stage of different WWTPs can be followed as hybrid MBR with RO or UF > coagulation-membrane disc-filter > coagulation- ozonation > Flocculation-DAF > constructed wetland > coagulation-RSF > ozonation-GAC filtration > coagulation-sedimentation. In summary, the combination of MBR with physical treatment such as RO/UF/NF has been found highly efficient in the removal of a wide range of MPs. Constructed wetlands (CWs) based hybrid treatment was found highly efficient, environmentally friendly and cost-effective. Moreover, coagulation with ozonation/GAC/DAF/RSF/ filtration processes are also more cost-effective than MBR based hybrid treatment.

4 MPs Removal Efficiencies of Each Step in WWTPs

A typical wastewater treatment process in WWTPs comprises 4 stages: Preliminary, primary, secondary and tertiary treatments (Fig. 6). The first step of

WWTPs is a so-called "pre-treatment" process, comprising a preliminary and a primary treatment step (Fig. 6a). At the first stage of the entire process, rags, sticks, and other large items are trapped during the preliminary treatment process to avoid their damage to pumps and interference of membranes in the subsequent purification processes. It typically comprises coarse screening (6–150 mm), fine screening (< 6 mm), and grit removal steps. Large flocs of fat, oil, and grease contained in wastewater can be helpful to trapping MPs during preliminary treatment, [202]. Thus, the suspended or floating MPs are removed along with other insoluble impurities affording ≈ 35%–59% MPs removal efficiency at this stage, [203, 204] Subsequently, a portion of the remaining MPs are removed by gravity separation and surface skimming operations in primary clarifiers (Fig. 6b), where the relatively heavier MPs can settle down or be trapped in sludge flocs, while the floating lighter MPs are trapped by the grease during surface skimming. As a result, ≈ 50%–98% of the MPs can normally be removed after primary treatment, [205]. During the pre-treatment process, the removal efficiency of MPs is influenced by several characteristics of the MPs, including their size, chemical compositions and morphology. The concentration of the relatively larger MPs with particle size of 1–5 mm was significantly reduced from 45% in the influent to 7% in the effluent during pre-treatment process, [206]. The lighter PE MPs floating on the water surface can be easily removed in the skimming process, while the heavier PVC and PET MPs can be separated by settling or being captured by sludge flocs, [207]. It is worth noting that the concentration of fibrous MPs in wastewater are markedly reduced in the pre-treatment effluent, which could be ascribed to the long-shaped fibrous MPs are more prone to be removed by coagulation trapping and following gravity separation, [208]. It has been reported that fibrous MPs remain to be the largest fraction after this stage, [202, 209]. In addition to the retention by pretreatment, the decrease of particle size of MPs in effluent could be ascribed to the fragmentation of large items by the physical forces through sand abrasion or water turbulence, [45]. This phenomenon leads to the newly generated smaller sized MPs debris passing through pre-treatment and proceeding to secondary treatment.

* Figure 6 can be found in Appendix section.

The next step of WWTPs is the secondary treatment, where the remaining suspended solids and dissolved organic pollutants in the H_2O can be further removed by the combined use of the activated sludge and the clarification tank (Fig. 6c). Aside from being captured by sludge, the microorganisms in the activated sludge can enhance the MPs removal efficiency. Due to growth of biofilms on the surface of MPs, the relative densities of MPs are normally dramatically altered, which facilitates the sinking and subsequent separation of suspended MPs, [210, 211]. It is known that there are a few bacteria, such as, *Bacillus, Rhodococcus*, and *Nocardia asteroids*, that are capable of degrading MPs, [171, 212, 213], however, the low biodegradation rates and short contact times between activated sludge and MPs result in negligible effect on the degradation of MPs in secondary treatment. The overall MPs removal efficiency is 86–99.8% relative to the pre-treatment effluent accompanied with decrease of the MPs' average size, [210, 211]. Sun et al. [214] reported the MPs with diameters > 500 μm were completely removed in this stage and the remaining ones were predominantly smaller than 190 μm. Talvitie et al. [196] revealed that only 8% MPs in the secondary effluent are > 300 μm in diameters. Similar to the pre-treatment stage, MPs undergo the continuous fragmentation under the comprehensive physical and biological factors. The formation of biofilms on the surface of MPs can facilitate the sedimentation of the MPs, the altered surface wetting properties and their relative densities might lead to MPs escape from skimming and settling processes, [214]. For many WWTPs, the secondary effluent is discharged into H_2O environment after disinfection, causing the leakage of large amounts of fibrous MPs and smaller-sized (i.e., < 300 um) MPs which accumulate in the environment. As indicated in a report, a high MPs removal efficiency of 98% was achieved for a secondary WWTP serving 650,000 people in Scotland. However, the daily seepage amounts of MPs (corresponding to the 2% remaining) into the environment was estimated to be 65 million pieces, [207]. In a separate report, the estimated median value of the amounts of MPs discharged from WWTPs with an average annual efflux of 5×10^7 m^3/y was 200,000 particles/d, [214].

Tertiary treatment, also called as advanced treatment, is the final stage of the whole process, which is frequently adopted to produce high quality drinking water (Fig. 6d). Typically, gravity filtration (GF), sand filtration (SF), DF, DAF, biologically active filters (BAF), MBRs, and other advanced treatments can be applied to decrease the concentration of suspended solid impurities, organic pollutants, heavy metals, and pathogens in H_2O, [209]. Based on these separation techniques, it has been reported that a high MP removal efficiency of 98–99.9% can be delivered. It is interesting to note that the fibrous MPs are difficult to remove in this process. By employing a sampling device with multiple mesh screens (500, 190 and 100 μm) to collect MPs from tertiary effluent was found that the MPs < 190 μm are the majority of the residual MPs in tertiary effluent, [215]. Meanwhile, the fibrous MPs have been proved to be the major species (54.50–88.90%) in tertiary effluent, which is thought to arise as these high-aspect-ratio materials are able to pass through the pores of filter or membranes longitudinally, [204, 209, 215, 216].

Despite the relative high MPs removal efficiency in WWTPs, the remaining MPs are extremely difficult to be removed by using currently available technology. The development of more advanced techniques to reduce the amount of MP in the wastewater of WWTPs is very necessary to prevent their further migration into the environment.

5 Conclusions

MPs are not degraded in the environment and can pose a serious threat to terrestrial and aquatic animals. The ready availability and diversity of plastic waste represent a largely under developed opportunity. Tailored upcycling processes for each type of plastic are the most likely approach to deliver maximal economic and environmental benefits.

In the MPs removal strategy, most conventional sewage treatment plants are being used to handle MPs pollution. To improve the efficiency, pretreatment with photocatalytic and biological degradation would reduce the MPs release in the environment.

MPs based on PS and PE are commonly found in the environment, mainly from the garment and household utensils, respectively. Source segregation on sites and awareness-raising campaigns should be carried out through the local authority to reduce MPs in both wastewater and surface water. Traditional methods for PET waste recycling still problematic because of the lethal effect on marine animals and humans. It is of great importance to find an effective and environmentally friendly strategy for green recycling of PET waste. The discovery of new PET biocatalysts and degrading microorganisms is an excellent movement toward a green recycling scheme for PET waste. Besides, studying their molecular mechanism extensively via solving their crystal structure will widen this research area to move forward the industrial applications.

The results of the chemical stability tests, chemical digestion utilizing mineral acids, such as sulfuric acid, can induce morphological deformation and transformation in the chemical structures. Thus, using sulfuric acid during the pretreatment of MPs should be avoided when researchers want to prepare MPs without chemical damaging based on findings on the results of chemical stability tests. Chemical structure changes of MPs particles showed almost the same results in their bulk states, such as film, which are reported in the related literature.

Flotation and agglomeration processes are commonly used in WWTPs to produce larger constituent particles that are easier to separate for MPs. Through the use of Fe- and Al-based salts along with other coagulants, these processes bind tiny particles by inducing uptake-complexation mechanisms that are initiated by exchanges of ligands, thus forming strong bonds between waste particles. In coagulation, colloids are broken apart and the surface charges of MPs are stabilized. Through van der Waals forces, the particles can interact sufficiently close to one another.

The optimization of the photocatalytic degradation of plastics requires the overall improvement of photocatalysts, reactors and reaction media. To realize the overall disposal of plastics and MPs through photocatalysis, more research should be conducted to achieve commercial success, together with the technique being suitable for mass manufacture. However, great physical and mechanical properties corresponding to typical plastics should also be achieved based on environmental protection. In addition to, photocatalysis can become widespread and gradually replace conventional plastics or conservative strategies of disposing plastics. Photocatalysis is quite promising in degrading plastics and it can be widely applied one day.

Photocatalytic degradation in abiotic degradation of MPs is a widely studied degradation technology, which has relatively high catalytic efficiency. Photocatalytic degradation and mineralization of MPs have the advantages of low energy consumption. The biodegradation of MPs was mainly focused on the degradation of MPs by plastic degrading bacteria or microbial community isolated from the environment. However, most of the biodegradation of MPs is still in the laboratory research stage. In the future, abiotic degradation technology as a pretreatment and biodegradation technology as subsequent mineralization can be combined to achieve complete degradation in the field.

Hybrid treatment for example, MBR-UF/RO system; coagulation followed by ozonation, GAC, DAF, RS, filtration and CWs based hybrid technologies have shown highly promising results for effective MPs removal. Among different physical and biological treatment technologies, the MPs removal performance decreases as membrane bioreactor (> 99%) > activated sludge process (~98%) > rapid sand filtration (~97.1%) > dissolved air floatation (~95%) > electrocoagulation (> 90%) > constructed wetlands (88%). Chemical treatment methods such as coagulation, magnetic separations, Fenton, photo-Fenton and photocatalytic degradation also show moderate to high efficiency of MP removal. Hybrid treatment technologies show the highest removal efficacies of MPs.

As a result, the design of the degradable polymer is also very critical. On the premise of not affecting performance, some inorganic or organic motifs can be introduced in the polymer framework as the switch of degradation to induce and accelerate the degradation of waste plastics in certain condition. For example, the introduction of inorganic matters with high expansion coefficient is capable to destruct the compact high crystalline structure, so that the degradation of waste plastics can be accelerated by the enhanced accessibility with the presence of UV or oxidation reagents. In addition to, more importantly, the production and disposal of biodegradable plastics need to be strictly regulated. Otherwise, it will worsen short-term MPs pollution, since the degradable polymers possess more rapid MPs generation.

Finally, in order to effectively degrade and remove MPs from our environment before they pose an unavoidable worldwide hazard to all life forms, reliable, efficient, cost-effective and green technologies must be developed. In addition, in education and awareness activities against plastic production and consumption; It is very important to adopt methods such as plastic waste management and recycling. Adopting novel and innovative approaches and policies would help develop clean and sustainable society, by diminishing accumulated plastic waste from the environment.

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Conflict of Interest

The authors have no conflicts of interest to declare that are relevant to the content of this article.

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APPENDIX

Table 1. The origin of MPs

Primary Sources	**Secondary Sources**
Clothing industry	Plastic fragmentation by mechanical friction
Cosmetic industry	Degradation by microorganism
Plastic production	Plastic breakdown by wave
Fishing industry	Fragmentation by UV and sunlight
Shipping line	
Airblasting	

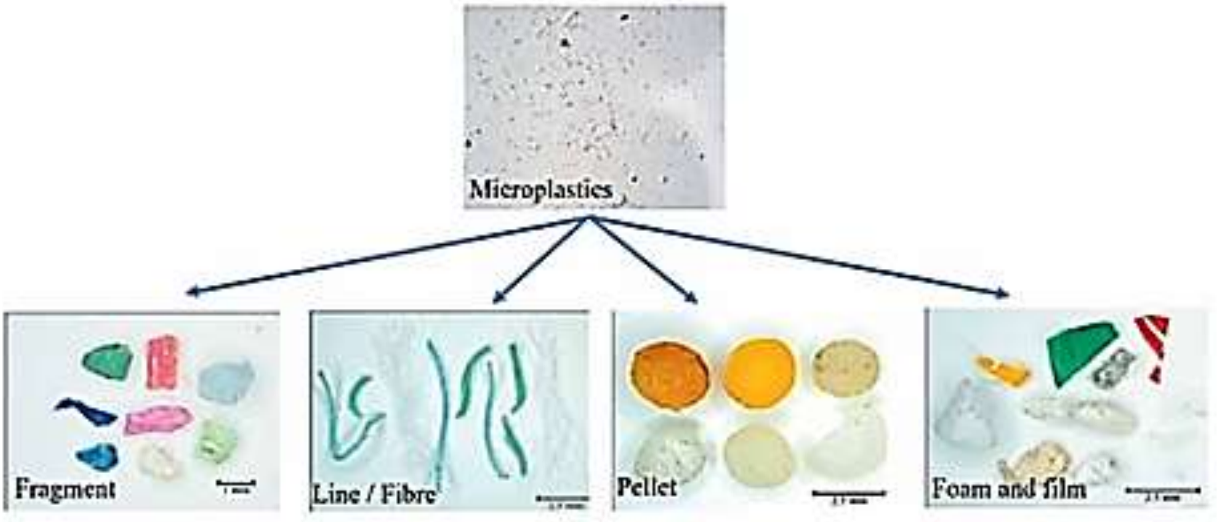

Fig 1. Types of MPs in wastewater

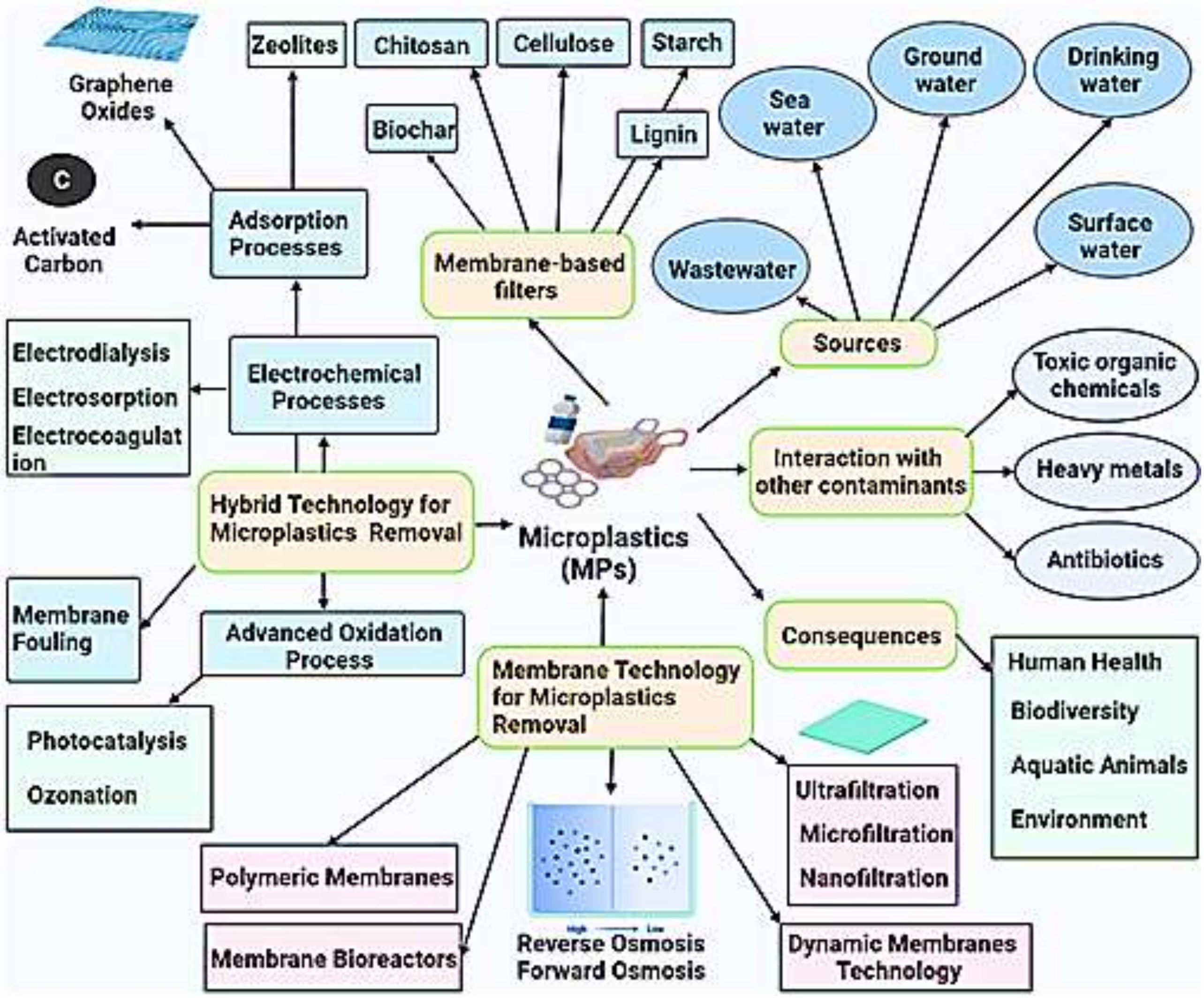

Fig. 2. All MPs removal methods (adopted from, [217]).

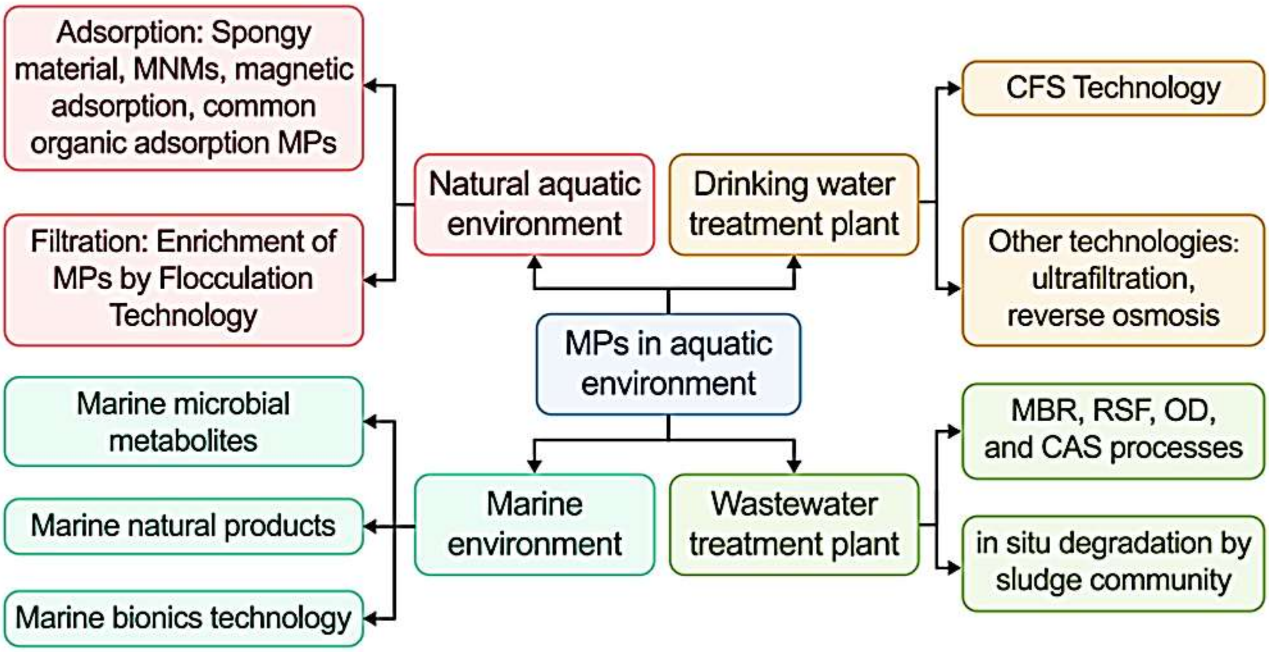

Fig. 3. The main removal technologies of MPs in different aquatic environments (adopted from, [218]).

Table 2. The general photocatalytic reactions of MPs

Photocatalytic reactions in O_2 rich atmosphere	**Equation Number**
$Photocatalyst + hv \rightarrow h^+_{VB} + c^-_{CB}$	6
$O_2 + e^-_{(CB)} \rightarrow O_2^{-\bullet}$	7
$H_2O + h^+_{(VB)} \rightarrow OH^{\bullet} + H^+$	8
$O_2^{-\bullet} + 2H^+ + e^-_{(CB)} \rightarrow H_2O_2$	9
$H_2O_2 + hv \rightarrow 2OH^{\bullet}$	10
$H_2O_2 + O_2^{-\bullet} \rightarrow OH^{\bullet} + OH^- + O_2$	11
$H_2O_2 + e^-_{(CB)} \rightarrow OH^{\bullet} + OH^-$	12
$OH^{\bullet}$ or $O_2^{-\bullet} + MPs \rightarrow Intermediates + Products$	13
Photocatalytic reactions in inert atmosphere	
$Photocatalyst + hv \rightarrow h^+_{(VB)} + e^-_{(CB)}$	6
$H_2O + h^+_{(VB)} \rightarrow OH^{\bullet} + H^+$	14
$H^+ + e^-_{(CB} \rightarrow H_2$	15
$h^+_{(VB)}$ or $OH^{\bullet} + MPs \rightarrow Selective\ valuable\ organic\ products$	16

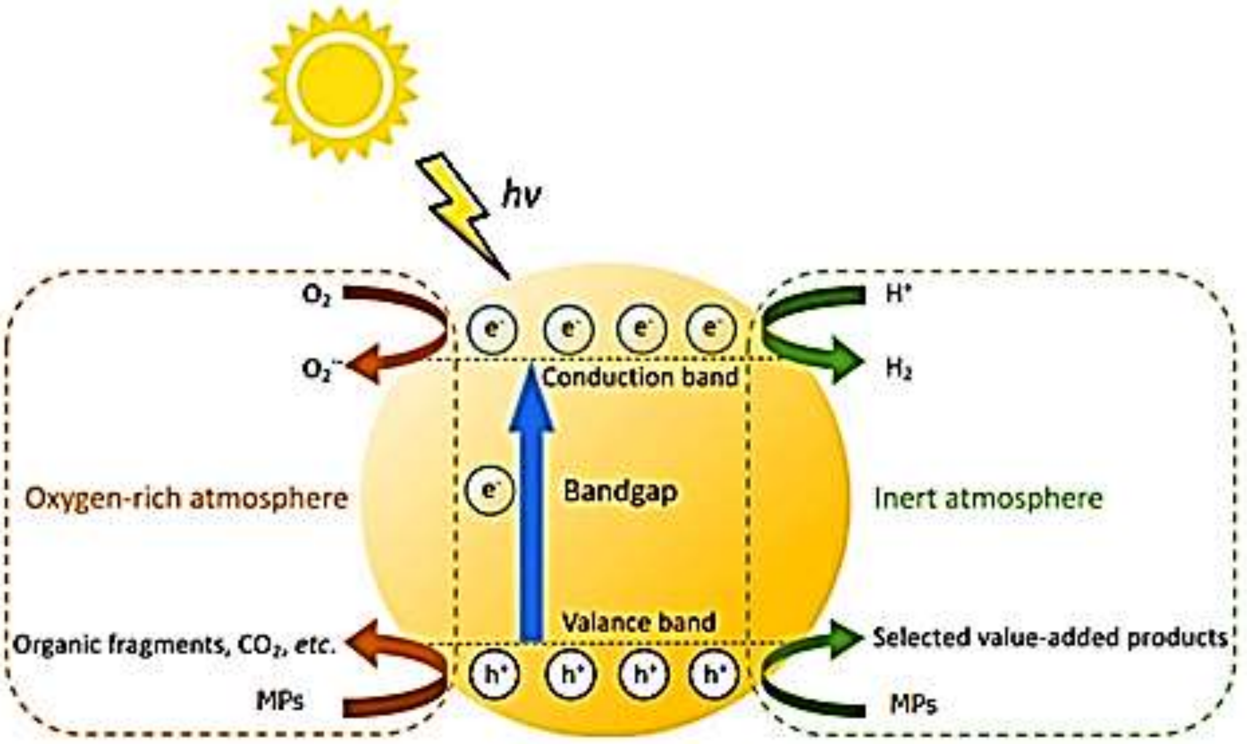

Fig. 4. Schematic representation of photocatalytic reactions for MPs degradation in different environments

Table 3. Comparisons between parabolic and non-concentrating solar photoreactors

Reactors	**Advantages**	**Disadvantages**
Concentrating collectors	Turbulent flow	Direct radiation solely in addition to water overheating
	Compounds free vaporization	High cost due to sun tracking system
	Smaller reactor tube area	Low quantum efficiency (r = k l < 1 with TiO_2)
	More practical usage of supported catalysts	Low optical efficiency
		Laminar flow with low mass transfer
Non-concentrating reactors	Diffused and direct radiation	Vaporization of reactants
	No heating	Contamination of reactants
	Cost effective	Weather resistance, chemical inertness, and UV transmission
	High quantum efficiency (r = k l with TiO_2)	
	High optical efficiency	

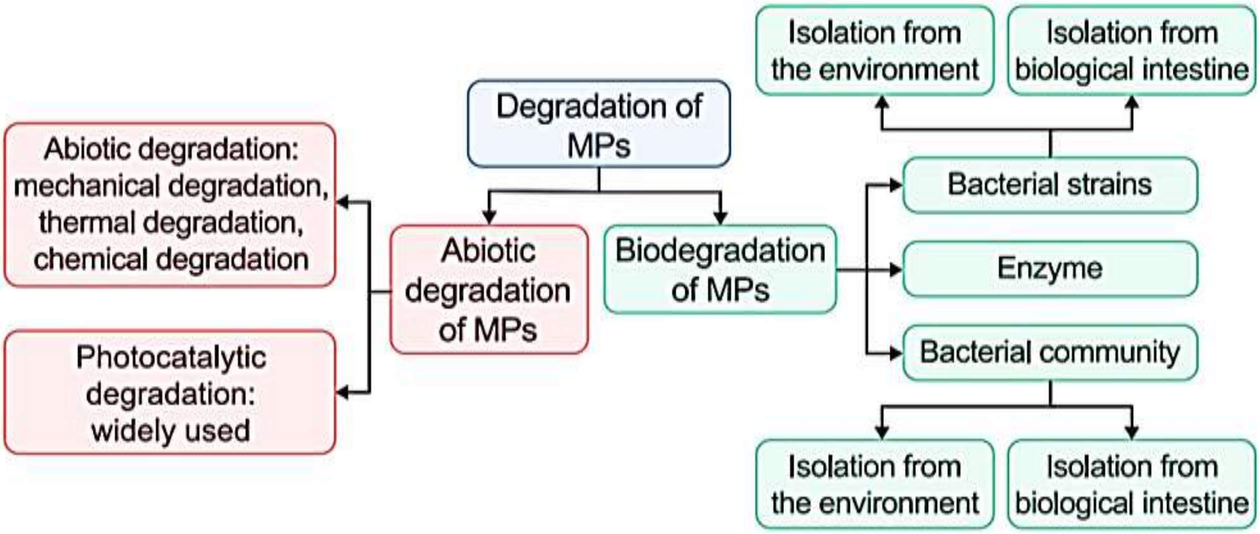

Fig. 5. Abiotic degradation and biodegradation methods of MPs (adopted from, [218]).

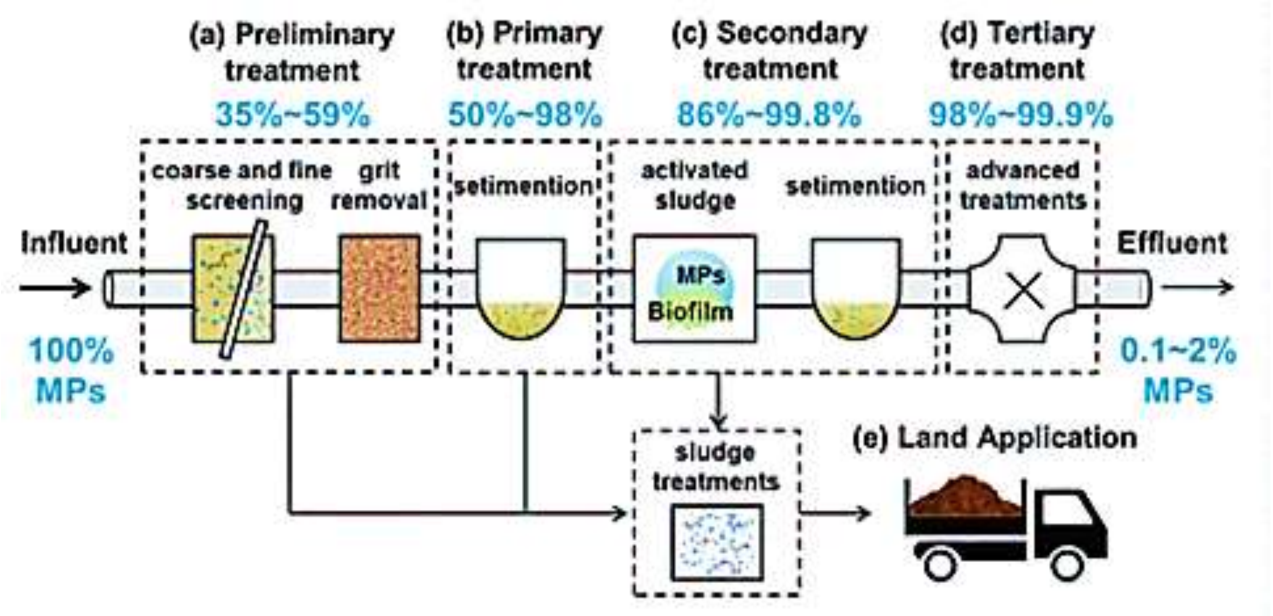

Fig. 6. MPs removals at different WWTP steps: a) Preliminary treatment, b) Primary treatment, c) Secondary treatment, d) Tertiary treatment and e) Land application (adopted from, [219]).