



Insights into the removal of microplastics and microfibrils by Advanced Oxidation Processes



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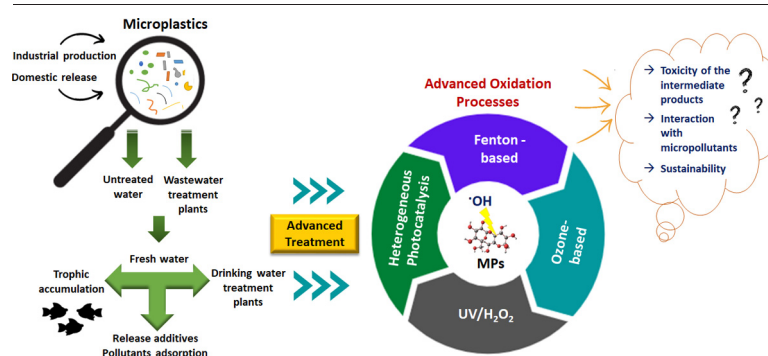
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HIGHLIGHTS

- MPs are less degraded by conventional Fenton compared to other AOPs;
- Smaller MPs including MFs increase abundance after ozonation.
- Lab studies use longer UV times than the requirements of treatment at full-scale.
- Oxidized MPs have lower interaction with hydrophobic micropollutants.
- There is a lack of studies on sustainability of AOPs applied for MP degradation.

GRAPHICAL ABSTRACT



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ABSTRACT

Water treatment plants' effluents are hotspots of microplastics (MPs) and microfibrils (MFs) released into the aquatic environment because they were not designed to capture these particles. Special attention should be given to MFs, since they mainly come from laundry and are related to one of the main MP shapes detected in water and wastewater treatment plants. In this sense, Advanced Oxidation Processes (AOPs) could be a feasible solution for tackling MP and MF pollution, however, it is still premature to extract conclusions due to the limited number of studies on the degradation of these particles (specifically MFs) using AOPs. This review addresses the impacts of AOPs on MPs/MFs, focusing on their degradation efficiency, toxicity, and sustainability of the processes, among other aspects. The review points out that polyamide MFs can achieve mass loss >90% by photocatalytic system using TiO₂. Also, the low oxidation of MPs (<30%) by conventional Fenton process affects mainly the surface of the MPs. However, other Fenton-based processes can provide better removal of some types of MPs, mainly using temperatures >100 °C, reaction time ≥ 5 h, and initial pH ≤ 3, achieving MP weight loss up to 96%. Despite these results, better operating conditions are still required for AOPs since the ones reported so far are not feasible for full-scale application. Additionally, ozonation in treatment plants has increased the fragmentation of MPs (including MFs), leading to a new generation of MPs. More attention is needed on toxicity effects of intermediates and methods of analysis employed for the analysis of MPs/MFs in wastewater effluent should be standardized so that studies can be compared effectively. Future research should focus on the sustainability of the AOP for MP removal in water treatment (power consumption, chemicals consumed and operational costs) for a better understanding of full-scale applicability of AOP adapted to MP treatment.

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1. Introduction

In 2020, the global production of plastics reached the estimated amount of 367 million metric tons (Tiseo, 2021). Plastic waste generated accumulates in landfills and in the natural environment where it degrades slowly. From this scenario, an increase of plastic particles is expected in the environment. Plastics <5 mm in size are denominated microplastics (MPs) whereas those <1 µm are referred to nanoplastics (Frias and Nash, 2019). However, the occurrence and detection of nanoplastics in aquatic environments is still a challenge due to the limitation of reliable analytical methods for such small particles (Mohana et al., 2021; Pulido-Reyes et al., 2022). One of the most prevalent MP sub-groups found in the aquatic environment are microfibrils (MFs), from which polyamide (PA), polyester (PEST), and polyethylene terephthalate (PET) have special relevance because of their abundance in the environment (Soltani et al., 2022). PEST contains the ester functional group and is a family of polymers that covers many plastics including PET. This last one has a compact structure, is highly recalcitrant and is developed for using as textile fibre and other applications, like food containers (Gewert et al., 2015; Hu et al., 2022a; Taniguchi et al., 2019). Natural fibres with micro-scale size are also denominated MFs but they are not MPs, which can lead to confusion. In this work, when referring to MP microfibrils (MFs), it will only be about plastic MFs. Also, the term MP will include MFs (when not, it will be highlighted).

Plastics and MPs can be transported with the wind (especially light ones like MFs) or run off to wastewater treatment plants, stormwater detention reservoirs, rivers, lakes, wetlands, estuaries and the sea (Braga Moruzzi et al., 2020; Capparelli et al., 2021; Russell and Webster, 2021). MFs are released from households (Hernandez et al., 2017; W. Liu et al., 2021; Llorente-García et al., 2020; Sørensen et al., 2021), including household laundries, fishing activities (Bui et al., 2020; W. Liu et al., 2021) and the textile industry, among other sources which imply lower release. One of the major concerns related to MPs is their toxicity: their toxic effects are not clearly understood yet (Aguar and Ascencios, 2021). MP degradation and potential impact is related to the type of polymer, density, size, exposure to weathering conditions, and their chemical additives (Munoz et al., 2021; Sørensen et al., 2021). Examples of chemical additives are diethylhexyl phthalate, Bisphenol A, brominated flame retardants (BFRs) (Kwon et al., 2017) (Jang et al., 2016) (Jeong and Choi, 2020) (Lambert

et al., 2017) and metals, like cadmium and chromium (Turner and Filella, 2021) or titanium (in PVC). Some of the chemicals used to produce textiles (e.g. benzophenone, phthalide and phthalate organophosphorus) can leach from MFs into seawater (Sørensen et al., 2021).

Once in the aquatic environment, MPs can accumulate, and trophic transfer could occur although biomagnification is difficult to assess due to the complexity of the food web and scarcity of studies on it (O'Connor et al., 2022). Benthic macroinvertebrates are especially exposed to MP pollution in contrast to fish species (O'Connor et al., 2022). MPs can be ingested by humans through various exposure pathways including drinking water or dust (Nematollahi et al., 2022). Recently, the presence of MPs was found for the first time in the human blood, with an average concentration of 1.6 µg/mL (A.Leslie et al., 2022).

Some countries have defined strategies aimed at reducing the effects of MPs in freshwater systems and pathways to the ocean. France and Australia, for example, intend to require all new washing machines to have MF filters to reduce the amount of MFs pollution from laundry (DAWE, 2021; Díaz Sánchez, 2020; France, 2020). Although policies on MF reduction have not yet been comprehensively implemented, some have been considered by countries seeking to reduce the MP emissions from end users (OECD, 2021). For instance, some policies have been proposed directly to consumers. For instance, in the New York State Bill (A1549), the Assembly Committee proposes additional labelling on clothes with >50% synthetic material, noting that plastic MFs are shredded from such clothes when washed (Environmental Conservation Law, 2020). A few countries will require filters in washing machines (DAWE, 2021; Díaz Sánchez, 2020; France, 2020). Kenya had a more overarching approach: it invested in a number of policies to decrease the flow of plastic into the ocean and other waterways. In 2017, the country banned the use, manufacture and importation of all plastic bags for commercial and household packaging (Republic of Kenya, 2017). Recently, plastic bottles, cups, and cutlery were also banned in its national parks and conservation areas (Republic of Kenya, 2019). Later, in 2019, the European Union approved a directive for the reduction of the impact of single-use plastic products (Directive (EU) 2019/904), giving priority use to “non-toxic” re-usable products (European Parliament, 2019). In 2017, the UK implemented regulation to reduce the quantity of plastics entering the marine environment and specifically microbeads, which were introduced via ‘rinse-off’ from cosmetics products. Plastic straws, cotton buds and drink stirrers were banned in

late 2020 (Department for Environment, 2020). Also, the UK proposed to decrease the production of solid MPs (Department for Environment, 2017), and from 2022, plastic packaging manufactured in, or imported into the UK, except those with at least 30 % of recycled plastic, will be taxed (UK Government, 2021).

MPs derived from urban activities can reach wastewater treatment plants (WWTPs). Several types of MPs have been found at contrasting concentrations in the influents and effluents of WWTPs (Blair et al., 2019; Carr et al., 2016; Mintenig et al., 2017; Murphy et al., 2016; Ruan et al., 2019; Talvitie et al., 2017). This variation may be partly due to the different methods used for MP quantification (e.g. detection capability of equipment and devices) or sample collection strategies (e.g. cut-off in filters or nets used for sampling) (Campanale et al., 2020; Halfar et al., 2021). But also due to the influences of the treatment plant location, local consumption habits and different economic levels. Considering the treatment plants were not projected to capture MPs, most of them can be release points of MPs (including MFs) to the aquatic environment (Blair et al., 2019; Carr et al., 2016; Gündoğdu et al., 2018; W. Liu et al., 2021; Lv et al., 2019). Examples of recent levels of MPs reported in European WWTPs are ~0.25 to 15 MPs/L (in all treatment stages) and <1–3 MPs/L (in the effluent) in UK WWTPs (Murphy et al., 2016) (Blair et al., 2019). In Canada, MFs were found in the range of 1.36–1.49 MFs/L (Prajapati et al., 2021), and there was higher presence of fibres than other shapes in the effluent. Indeed, several studies have indicated fibres and fragments as the most predominant shapes in raw water and WWTP effluents (Blair et al., 2019; Leslie et al., 2017; Murphy et al., 2016; Ruan et al., 2019; Xu et al., 2019; Yang et al., 2019). Some studies in drinking water treatment plants (DWTPs) have noticed that the quantity of fibres was higher than other MPs detected mainly in treated water (Dalmau-Soler et al., 2021; Sarkar et al., 2021; Wang et al., 2020). The lack of MF targeting technologies has been one of the consequences of MF abundance reported in WWTP effluents (Blair et al., 2019; Lee et al., 2020). For instance, polymers such as PEST, one of the main polymers making up MFs, are more resistant to removal/degradation by both conventional and advanced treatment processes (Dalmau-Soler et al., 2021; Murphy et al., 2016). Concerning the MP types detected in larger quantities in DWTPs, those correspond to polymers with relatively low density, such as PET (another polymer present in the MFs composition) (Dalmau-Soler et al., 2021; Pivokonský et al., 2020; Sarkar et al., 2021; Wang et al., 2020). Additionally, the effective removal of MPs < 10–20 µm and the micro – nanoparticles formation during some processes are a challenge for WWTPs and DWTPs (Pivokonský et al., 2020; Shen et al., 2021; Talvitie et al., 2017). Moreover, studies on fragmentation and removal of nanoplastics in DWTP are scarce, making it difficult to understand the scope of nanoplastics in those treatment plants (Mohana et al., 2021; Ramirez Arenas et al., 2022). The presence of MPs in sludge generated by WWTPs is also a concern, as the sludge can be recycled to agricultural land as a fertilizer resource, which can run off into waterways or seep into groundwater (Li et al., 2018; Lofty et al., 2022). Furthermore, the majority of MP content in sludge appears to be fibres. For example, 34 particles/g were found in the sludge of a WWTP, from which 90 % were fibres (Corradini et al., 2019). In this sense, it is essential to develop more efficient technologies than the usual conventional processes applied.

A well-established advanced treatment for the removal of recalcitrant contaminants in different aquatic matrices is Advanced Oxidation Processes (AOPs), which are capable of mineralizing pollutants. However, despite the increase in the number of research/publications focusing on the degradation of MPs, there is still a significant gap on the application of AOPs for the degradation of MPs and, mainly, MFs. To the best of our knowledge, the review articles published to date addressing the degradation of MPs using AOPs did not discuss (or discussed briefly) relevant aspects such as sustainability and environmental impacts of these processes used for MP treatment (Ahmed et al., 2021; Chen et al., 2022; Du et al., 2021; Lastovina and Budnyk, 2021; Ricardo et al., 2021; Shen et al., 2022; Xu et al., 2021). For example, there is a gap between treatment technologies to remove MPs and their impacts on carbon emissions. In addition, different

factors are involved in the sustainability of the processes, such as investment, operation conditions, energy consumption, generation of by-products and their toxicity (Chen et al., 2020; Krahnstöver et al., 2022). Moreover, reviews addressing the degradation of MPs using AOPs focusing on the formation of by-products and the sorption potential between oxidized plastic particles and other micro-pollutants are urgently needed. Also, there are no reviews discussing AOP for the degradation of MFs. Therefore, this review addresses the impacts of AOPs for MPs (including MFs) degradation for the first time, including the intermediate products formed, their toxicity and sustainability of the processes. AOP processes investigated are Fenton and its variations (photo-Fenton, electro-Fenton, thermal Fenton), UV/catalysis, UV/H₂O₂, and ozone-based processes.

2. Advanced Oxidation Processes for MP degradation

Several studies have proven the efficiency of AOPs (e.g. 50 % - 99 % removal) on recalcitrant pollutants (Bosio et al., 2019; dos Santos et al., 2021; Esplugas et al., 2007; Sánchez-Montes et al., 2022; Segura et al., 2013). In brief, AOPs are based on in-situ generation of oxidative radicals with different values of standard reduction potential (V), such as hydroxyl ($\cdot\text{OH}$) with + 2.8 V. Once these radicals are produced, a cascade of mechanistically complex reactions takes place. Such radicals are non-selective oxidants and are capable of converting organic compounds into simpler ones and/or mineralizing them (Deng and Zhao, 2015; Miklos et al., 2018). However, unlike other processes that are more mature for the removal of MPs (e.g. sand filtration), the application of AOPs for MPs degradation is recent. Recent works have reported advances in the identification of factors that play important roles in the degradation efficiency of MPs by AOPs (Hu et al., 2022b; Llorente-García et al., 2020; Miao et al., 2020; Nabi et al., 2020; Piazza et al., 2022). These factors are described in the following sections.

2.1. Fenton-based oxidation processes

2.1.1. Conventional Fenton process for MP degradation

In conventional Fenton process, highly reactive hydroxyl radicals are generated when H₂O₂ reacts with soluble iron (II) salts. These radicals can partially or totally oxidize the targeted organic contaminants to CO₂ and water (Dos Santos et al., 2021). As iron is a non-toxic and abundant element, this method has become one of the most investigated AOP for water/wastewater treatment (Ahmed et al., 2021; Deng and Zhao, 2015). However, it was reported that MPs are rarely affected by conventional Fenton processes (Mackulak et al., 2015). Precisely because of this, the use of the conventional Fenton process has been reported as an efficient method for isolating and purifying MPs from environmental and other biological materials by oxidising organic matter in the samples (Tagg et al., 2017), and to a less degree, MPs given that sample treatment seek to preserve the analytes. The Fenton process has also been used for “weathering” MPs when attempting to simulate natural oxidation of MPs in the environment (Liu et al., 2019).

The aging and degradation of nylon (PA6) and polystyrene (PS) MPs during 4 Fenton treatment cycles, each cycle lasting 5 h, were investigated. Under experimental conditions (pH 3, ~0.5 % H₂O₂ and 0.2 M Fe²⁺), the mass loss of PA6 was ~25 % and 22 % for PS. Gas chromatography-mass spectrometry (GC-MS) and surface Raman scattering (SERS) analysis indicated the formation of intermediate products, such as low molecular weight alkanes and the new presence of oxidized functional groups such as alcohols, aldehydes and carboxylic acids on the surface of the MPs (Liu et al., 2022).

The degradation of MPs achieved with conventional Fenton when carried out before anaerobic decomposition was studied (at pH 3 and ~0.2 % H₂O₂ for a reaction time of 20 min, followed by sedimentation (15 min). Under such conditions, PVC MPs fragmented, reaching a mass loss of 10 ± 2 % and some degradation products with low molecular weight were detected such as ethylbenzene and o-xylene. Additionally, anaerobic microorganisms digested part of the PVC fragments, and it was hypothesised that the Fenton degradation products were acting as a carbon

source for the anaerobic bacteria during the subsequent biological decomposition step (Mackuřak et al., 2015). The MP mass loss achieved was low if this treatment method is applied for MP degradation only (Mackuřak et al., 2015).

One of the first studies that reported PS degradation through Fenton, this time using ferric ions, did not obtain evidence of polymer degradation during 5 h of oxidative treatment, even at 75 °C (Feng et al., 2011). In that case, the experimental conditions were $[Fe^{3+}] = 0.42$ mM, $[H_2O_2] = 141$ mM at pH = 2. It is important to note that this type of Fenton process (known as Fenton-like), which uses ferric ions as a transition metal in the reaction, may be in some cases weaker than conventional Fenton (Jiang et al., 2010). On the other hand, the application of conventional Fenton for degrading organic pigments released by MPs to the environment confirmed to be an efficient process. The red pigment present in HDPE MP was degraded >90 % applying the conventional Fenton reaction (pH = 3, $[Fe^{2+}] = 1$ mM, $[H_2O_2] = 10$ mM, and total reaction time of 360 min) (Luo et al., 2021). The degradation of that pigment was greater than its leaching to the aqueous media. After the oxidation process, aged LDPE showed fragmentation and cracks on their surface (Luo et al., 2021).

In summary, even with the Fenton process being capable of accelerating surface aging of MPs, in general, results have shown a low degree of MP oxidation. Furthermore, there are still a limited number of published studies focusing on the mineralization of MPs through the conventional Fenton process (Fig. S1 in Appendix A). One of the disadvantages of Fenton oxidation is the high amount of sludge generated and the need for removing metallic iron in the form of particulate powder at the end of the treatment (dos Santos et al., 2021).

2.1.2. Heterogeneous Fenton and photo-Fenton processes for MPs degradation

A promising alternative for MP degradation is the application of solid catalysts to the Fenton system, which is generally named as heterogeneous Fenton. This process is efficient for the removal of organic compounds (Araujo et al., 2011; Dos Santos et al., 2021; Nidheesh, 2015; Teixeira et al., 2015). One of the main reasons for applying solid catalysts (such as iron minerals, nano-zero valent iron and other forms of metallic iron) as a source of iron, is to reduce treatment complexity and operational costs found in the conventional Fenton that produces sludge and leaves treated water with high acidic conditions. However, to the best of our knowledge, the application of the heterogeneous Fenton for the degradation of MPs and MFs in water has not yet been reported in the literature, it is therefore an interesting perspective for future studies. Alternatively, other variations of the Fenton process have been studied in recent years, such as the Fenton/heterogeneous Fenton processes under irradiation, known as the photo-Fenton. Unlike Fenton, the photochemical regeneration of ferrous ions occurs due to the photo-reduction of ferric ions in the photo-Fenton reaction. Thus, ferrous ions that form can react with H_2O_2 again, in a cyclic process, generating $\cdot OH$ and ferric ion favouring the extent of the cycle (Ameta et al., 2018).

The photo-Fenton process was evaluated for the degradation sulfonated PS beads (experimental conditions of UV irradiation from a 250 W Hg (Xe) light, volume of the reaction mixture of 25 mL, $Fe^{3+} = 0$ to 0.63 mM, $H_2O_2 = 0$ to 176 mM, and initial pH = 2) (Feng et al., 2011). After such work, photo-Fenton studies attempting MP degradation were carried out (Liu et al., 2020; Piazza et al., 2022). The photo-Fenton process led to almost complete removal of PS MPs after 300 min of UV irradiation (Feng et al., 2011). The degradation efficiency achieved was greater as the concentration of H_2O_2 and leached ferric ions increased. The degradation products identified on the polymer surface after 120 min of treatment corresponded to aromatic and phenolic groups. A recent study reported similar intermediate products such as benzoic acid, formic acid and acetophenone formed on the MP surface when treating PS by the photo-Fenton process (Liu et al., 2020). However, this study involved only photo-Fenton as the aging process (experimental conditions were UV-365 nm; 0.50 mL of 20 mM Fe^{3+} ; 0.51 mL of 30 % H_2O_2 ; pH 6.5; reaction time of 108 h) (Liu et al., 2020). The authors identified that the photo-Fenton treatment was producing effluents with some toxicity due to generation of intermediates even without

including toxicity test in their investigation. Despite previous studies reported toxicity of benzoic acid in some freshwater organisms (e.g. cladoceran crustacea and oligochaete worm) (Saha et al., 2006) and reported that acetophenone is nontoxic to freshwater invertebrates and fish (EPA, 2006), toxicity tests are needed to evaluate the effect of intermediates from the photo-Fenton process at the concentration level relevant to the degradation of MPs.

Recently, heterogeneous photo-Fenton using Fe^0 nanoparticles/zinc oxide nanorods coated with ZnO/SnOx was assessed (Piazza et al., 2022). The degradation of PP and PVC MPs was carried out in a continuous flow system (with $[H_2O_2]_0 = 35$ mM) for up to 30 days and achieved a reduction of the volume of PVC and PP MPs of 94 % and 96 %, respectively after one week of treatment. However, such a slow process makes it difficult to be implemented at full scale. In addition, the remaining particles were <53 μm , which would be problematic if these effluents were to be released into water bodies. The main surface oxidation indicators detected on the surface of PVC and PP were carbonyl and hydroxyl groups. Additionally, toxicity tests performed found that these carbonyl and hydroxyl groups were nontoxic to the evaluated aquatic organisms (bacteria, microalgae, and crustacean).

Even though the full mineralization of MPs by Fenton processes is still a challenge, the ecotoxicological data from the intermediates formed is useful for a better understanding of AOPs. However, despite the relevance of toxicity tests (evaluated by the inhibition of some organisms like *A. fischeri*, *D. magna* or *E. coli* bacteria e.g.) (Hu et al., 2022b; Piazza et al., 2022), there is scarcity of studies assessing the toxicity of MPs that have undergone Fenton processes.

2.1.3. Thermal Fenton and electro-Fenton processes

A recent strategy for MPs degradation is the thermal Fenton system (hydrothermal coupled Fenton process). This process was investigated for degradation of different MPs, such as LDPE, HDPE and UHMWPE (Ultra-High Molecular-Weight Polyethylene) (Hu et al., 2022b). It was efficient at 140 °C, which led to the generation of oxidative radicals. The polymers experienced relatively similar weight loss considering the different polymers, with MP range removal of ~40–80 % (experimental conditions were $[Fe^{2+}] = 4$ mM, $[H_2O_2] = 200$ mM, $[MPs] = 1$ g/L, 140 °C). In contrast, when the same experimental conditions were used with river and tap water samples, > 80 % of MP were removed. However, MPs from HDPE (debris from a milk plastic bottle) had a degradation <10 %. In this case, pH and temperature had direct influence on the treatment efficiency. In brief, the UHMWPE MP decomposition generated important oxidation markers (C=O and C—O) during the treatment. Additionally, the products formed during the reactions were nontoxic to *E. coli* bacteria.

The electro-catalytic process, known as electro-Fenton, is another Fenton-based process that has been tested for MP degradation. The decomposition of H_2O_2 from the reaction with the Fe^{2+} produces a high number of radical species with potential to degrade the target pollution. Miao et al. used electro-Fenton TiO_2 /graphite cathode process at an initial pH of 3 and 100 °C, for 6 h, for the degradation of PVC (size of 100–200 μm , initial concentration of 100 mg/L) (Miao et al., 2020). Under these experimental conditions, the oxidation of PVC occurred together with dechlorination, reaching 56 % of mass reduction and 75 % dechlorination (Miao et al., 2020). Such treatment involved cathodic reduction (with heating) of TiO_2/C acting as cathode. Among the intermediate products identified, there were alkenes and oxidized organics containing alcohols, monocarboxylic acids, dicarboxylic acids and esters. The toxicity of these intermediates and the identification of smaller MP fragments after treatment were not investigated. Fig. 1 explains the degradation mechanism of PVC MPs proposed by the authors.

Finally, Fig. 2 and Table 1 present a summary of Fenton-based processes investigated for the degradation of MPs in the present work.

Despite interesting outcomes (e.g. mass loss/volume reduction up to 96 %), from Fenton-based processes as potential oxidative treatments for MPs, there are still limited number of studies that have investigated variables that can affect the reaction, formation of intermediate products and their

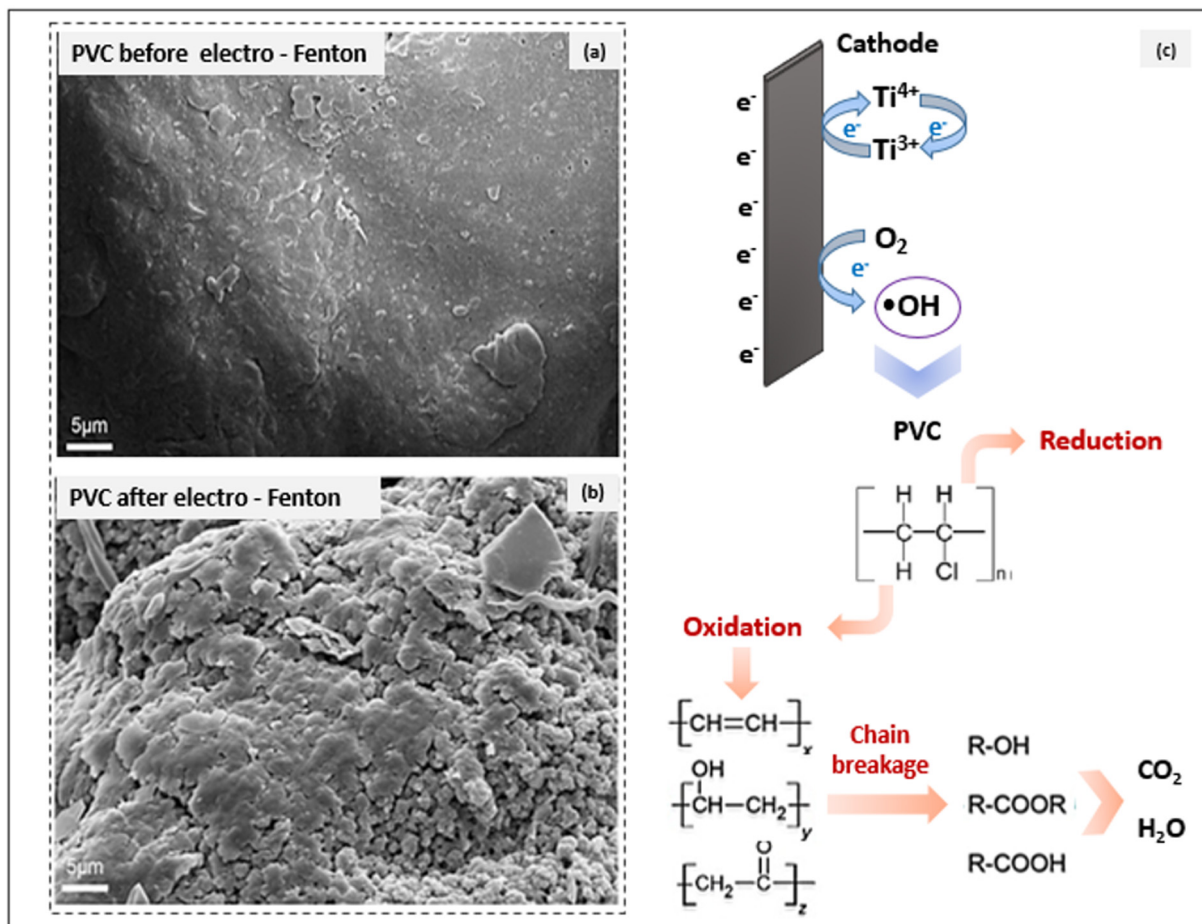


Fig. 1. Left side (a-b), Scanning Electron Microscopy (SEM) micrographs from the PVC MP surface (a) before treatment and (b) after 6 h of electrocatalytic treatment. Right side (c) proposed degradation process of PVC MPs using the electro-Fenton process. Adapted from (Miao et al., 2020).

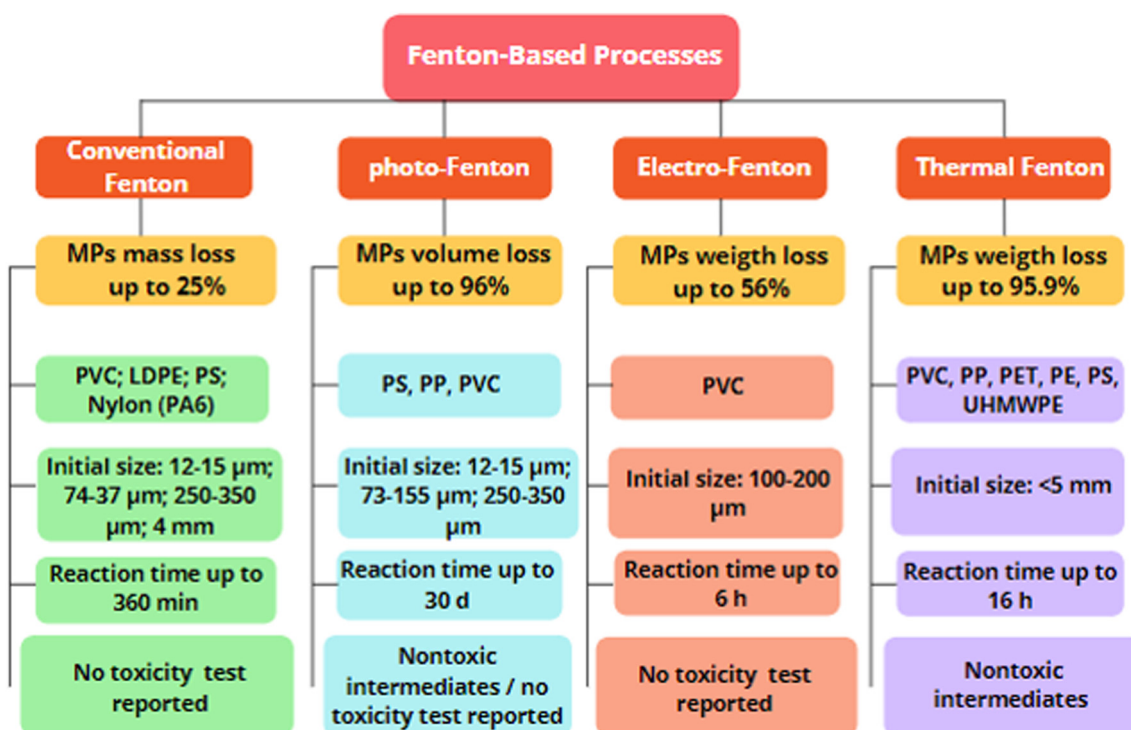


Fig. 2. Degradation of MPs by the Fenton-based processes. Initial size corresponds to the starting dimensions of the MPs subject to Fenton.

Table 1

Degradation of MPs (not including MFs) by the Fenton-based processes and their characteristics.

Experimental conditions of the AOP investigated	Matrix	Types of polymer	Size/concentration of MPs	Degradation and performance	Size of the MPs (after AOP)	Variables that influence the AOP	Intermediate products	Toxicity of the intermediates	Ref.
Fenton Fe ²⁺ = 0.264 g; H ₂ O ₂ = 0.80 and 1.60 mL (30 %, w/w) in 300 mL of deionized water. Initial pH 3 and reaction time of 20 min + sedimentation 15 min.	Deionized water	PVC	0.5 g/300 mL	PVC weight losses of 10 ± 2 %. Direct reaction between Cl ⁻ ions and •OH radicals decrease the reaction efficiency.	-----	Lower concentration of H ₂ O ₂ resulted lower degradation efficiency.	In the solution: <i>trans</i> -1,2-dichloroethene, <i>cis</i> -1,2-dichloroethene, trichloroethene and tetrachloroethene ethylbenzene and o-xylene	-----	(Mackuřak et al., 2015)
Fenton [Fe ²⁺] = 1 mM; [H ₂ O ₂] ₀ = 10 mM. Initial pH 3, 200 rpm, and reaction time 360 min	Deionized water	LDPE with composed of pigment red	0.5 g/100 mL	Concentration of dye leached: 0.28 mg/L by Fenton and 7.5 mg/L without Fenton treatment.	-----	Pigment leaching was best promoted <pH 3.	On the MP surface: C–N bonds	-----	(Luo et al., 2021)
Photo-Fenton UV irradiation from a 250 W Hg (Xe) light, [Fe ³⁺] ₀ = 0.42 mM, [H ₂ O ₂] ₀ = 141 mM/g of the sulfonated PS. Initial pH = 2 and reaction time of 300 min.	-----	PS microspheres and sulfonated PS beads	12–15 µm (PS) and 250–350 µm (PS beads)	DOC concentration detected in the degradation mixture: mineralization efficiency >89 %	-----	The process was not significantly sensitive to the particle size of the polymers.	On MP surface: aromatic C=C and alcoholic/phenolic stretching. In solution: hydroxylated and phenolated species.	-----	(Feng et al., 2011)
Photo-Fenton ZnO NRs/SnO ₂ / Fe ⁰ nanoparticles under visible light lamp (120 W); [H ₂ O ₂] ₀ = 35 mM. Flow rate of 300 mL/min; reaction time = 15 and 30 days ^a	Diluted H ₂ O ₂ solutions	PP and PVC	155 ± 1.4 µm (PP) and 73 ± 0.5 µm (PVC) ~ 19 particles/L	After 1 week of reaction: 94 % and 96 % of PVC and PP volume reduction, respectively.	After 1 week of reaction: PVC = 28 ± 3 µm and PP = 53 ± 0.3 µm.	Increase in exposure time of MPs in the presence of H ₂ O ₂ caused an increase of carbonyl and hydroxyl groups.	On the MP surface: PVC: prevalence of aldehyde.	Toxicity evaluated by inhibition tests (<i>A. fischeri</i> , <i>P. subcapitata</i> , and <i>D. magna</i>).	(Piazza et al., 2022)
Electro-Fenton TiO ₂ /C cathode at 100 °C; potential of –0.7 V vs. Ag/AgCl; initial pH 3; 800 rpm; reaction time of 6 h.	100 mL Na ₂ SO ₄ solution (0.05 M)	PVC	100–200 µm 100 mg/L	75 % dechlorination and 56 % weight loss of MP.	-----	Higher mineralization degree of PVC obtained with elevated temperature. The initial concentration of PVC influenced the efficiency of the process.	In the solution: alkenes, alcohols, and esters.	Nontoxic intermediates	(Miao et al., 2020)
Thermal Fenton 200 mL Teflon autoclave at 140 °C, 1 g/L of MPs dispersed in 150 mL of ultrapure water, [Fe ²⁺] = 4 mM, [H ₂ O ₂] = 200 mM, and initial pH = 1–3.	Ultrapure water	UHMWPE, LDPE, HDPE, PS, PVC, PP, PET	LDPE and HDPE = debris <5 mm 1 g/L for all MPs	UHMWPE = 95.9 % weight loss in 16 h and 75.6 % mineralization efficiency in 12 h.	Up to 12 h of reaction, the remaining fragments were aggregated in a larger size than the original particles.	At a pH value lower than 1, the MP decomposition rate decreased. <140 °C indicated almost no MP weight loss.	On the MP surface: methyl group.	Toxicity evaluated by filter paper disk method using <i>E. coli</i> . Nontoxic intermediates.	(Hu et al., 2022b)

HDPE = high density polyethylene; LDPE = low density polyethylene; PET = polyethylene terephthalate; PP = polyethylene; PS = polystyrene; PVC = polyvinylchloride; UHMWPE = ultra-high molecular-weight polyethylene.

----- Not informed or not evaluated.

^a Continuous flow system. The other systems presented in the Table were evaluated without continuous flow, on a laboratory scale.

toxicity and the formation of secondary MP. Furthermore, to the best of our knowledge, there are no studies on Fenton-based processes for MFs in the literature, even with recent studies confirming a high concentration of MFs in different aquatic matrices and the inability of total removal by water and wastewater treatment plants (Yang et al., 2019).

2.1.4. Sustainability of the Fenton-based processes for MPs degradation

The sustainability of the investigated Fenton processes for MP treatment should include an assessment of their sustainability in relation to degradation efficiency, power consumption, sludge formation, chemicals consumed and operational costs. The carbon footprint of some of the oxidative processes presented in Section 2 is relevant to WWTPs and DWTPs. For example, despite the high efficiency in removing different types of polymers in different waters, the Thermal Fenton system did not favour mass loss of MPs at temperatures <140 °C (Hu et al., 2022b). From the sustainability point of view, the energy demand for this AOP will be high and it will be affected by the duration of the process. In this sense, the operational costs and the carbon footprint produced need to be evaluated to verify the real benefits of this system in return for the removal efficiency achieved (Kang et al., 2020). Moreover, the number of investigations on photo-Fenton for the degradation of MPs is very limited and there is no data on its feasibility for real scale applications despite the fact that this process is widely used for the removal of different pollutants (Alalm et al., 2015; Murray and Parsons, 2004; Ribeiro et al., 2020; Rodrigues et al., 2013). For example, despite the promising results of MP degradation reported by Piazza and co-workers, and the use of visible light in their study, the adopted photo-Fenton method demands an overlong degradation period (over a week) (Piazza et al., 2022). Hence, photo-Fenton processes have potential as a low carbon technology if solar irradiation is used, but further studies with improved operating conditions are still needed.

2.2. Heterogeneous photocatalysis for MP degradation

Oxidation of MPs by photocatalysis has been studied (Lee et al., 2020; Llorente-García et al., 2020; Tofa et al., 2019b, 2019a); however very little is known of its application for MF oxidation (Lee et al., 2020). In general, heterogeneous photocatalysis degradation occurs using a semiconductor exposed to UV (by sunlight or artificial light) emitting photons (which need to have equal or higher energy than its band gap), causing the generation of positive holes in the valence band and excited electrons at the conduction band (e^-). The most commonly studied catalyst for photocatalysis is titanium dioxide (TiO_2) (Deng and Zhao, 2015). Catalysts with a larger surface area are generally beneficial since they enhance the availability of reactive species (such as hydroxyl radicals $\cdot\text{OH}$ and superoxide O_2^-) that will react with the polymer. Additionally, for better interaction between the catalyst and MPs, some studies have evaluated nanoscale semiconductors (e.g. TiO_2 and ZnO) to accelerate the degradation process in the presence of UV, and its immobilization, to avoid the separation and recovery step after the treatment (Nabi et al., 2020; Tofa et al., 2019a, 2019b). As shown in Fig. 3, the presence of the catalyst in the photolysis system potentiates the MP and MF degradation, indicated in this figure as size reduction and morphological changes. The respective catalysts in Fig. 3 used by Nabi et al. (TiO_2 nanoparticle films catalysts made with Triton X-100, an anionic surfactant) (Nabi et al., 2020) and by Lee et al. (TiO_2 powder) (Lee et al., 2020) were not immobilized on polymers.

2.2.1. Effect of the pH

The efficiency of the photocatalytic system is dependent on the experimental setup. For example, depending on the type of catalyst used, the pH of the photocatalytic system can affect its surface charge. The pH of the process had direct influence on the degradation results of polymethylmethacrylate (PMMA) and polystyrene (PS) (Allé et al., 2021).

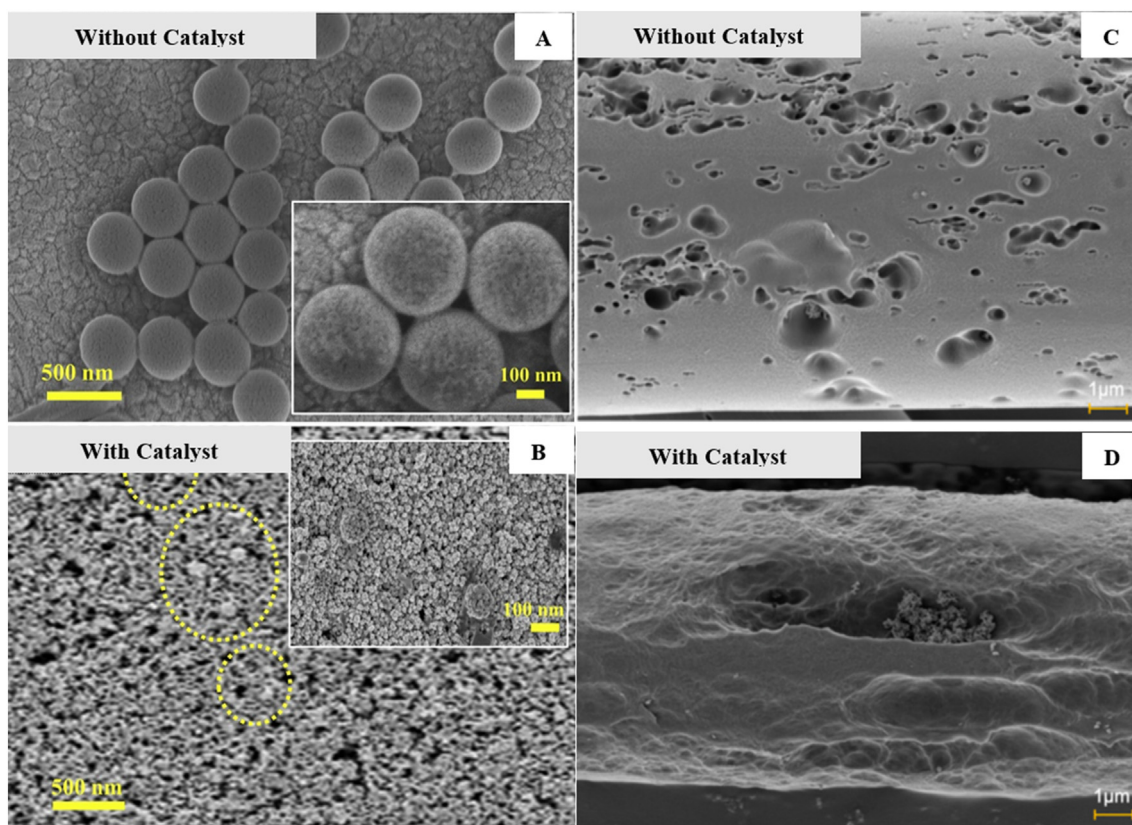


Fig. 3. SEM micrographs of MPs and MFs in heterogeneous photocatalysis. (A) PS spheres without catalyst after 12 h irradiation and (B) PS spheres with catalyst Triton X-100 based TiO_2 (TXT) after 12 h irradiation (adapted from (Nabi et al., 2020)); (C) PA66 Microfibre without catalyst after 105 h of irradiation and (D) PA66 Microfibre with catalyst TiO_2 after 105 h of irradiation (adapted from (Lee et al., 2020)). The catalysts were not immobilized on polymers.

At acidic pH, the surface of the catalyst ($\text{TiO}_2/\beta\text{-SiC}$: open-cell $\beta\text{-SiC}$ alveolar foams as TiO_2 support, with diameter 38 mm and length 30 mm) was positively charged, triggering competition between ions in the aqueous media (such as chloride- from the HCl solution used) and PMMA nanospheres towards the active sites of the TiO_2 surface. On the other hand, at pH 9, the TiO_2 surface was negatively charged and a fraction of the ester functions of the PMMA surface were hydrolysed to negative carboxylate groups, which caused electrostatic repulsion forces between the catalyst and the target contaminant, hindering photocatalytic reactions.

Improved efficiency in the degradation HDPE MP at pH 3 was found compared to pH 7 and pH 11 under different temperatures ($T = 0, 20$ and $40 \pm 2^\circ\text{C}$) (Ariza-Tarazona et al., 2020). A total of 71.7 % MP mass loss was achieved at pH 3 and at 0°C (Fig. 4). pH in the acidic range incorporated H^+ into the system, which lead to the formation of hydroperoxide radicals that facilitated the MP degradation. Regarding different temperatures applied (0°C and 40°C) with the same pH value (pH 3), higher degradation was reported at 0°C is probably due to the increase in the surface to the volume ratio of the MPs caused by fragmentation (Ariza-Tarazona et al., 2020).

2.2.2. Effect of UV irradiation

The lighting of the system is an important operating parameter in photocatalysis leading to the formation of oxidative species (Allé et al., 2021; Lee et al., 2020; Tofa et al., 2019a). In general, shorter UV wavelengths, compatible with TiO_2 band gap have been reported to be responsible for greater changes in the MP surface, such as cracks, wrinkles, and scission of polymer chains, compared to the application of longer wavelengths. A recent photocatalytic study reported greater MF degradation efficiency when 254 nm was applied compared to 365 nm (Lee et al., 2020). The application of UV-C lamps (254 nm) resulted in higher carbonyl index values and FTIR results showed more intense peak bands $\approx 1630\text{ cm}^{-1}$ compared to the application of UV-A, which correspond to the carbonyl group of the amide groups (NH-C=O) on the fibres after oxidation (Moezzi et al., 2020) (Fig. 5). The degradation of polyamide 66 (PA66) MFs occurred with a mass loss of 97 % in 48 h using UV at 254 nm (experimental conditions were UV-C lamps 8 W, 100 mg of catalyst TiO_2/L , MFs diameter of 10 μm , concentration of 1.3 mg/50 mL of deionized water, and reaction time up to 105 h). Similarly, there was greater PS and PE degradation at 254 nm compared to 365 nm and visible light, using TiO_2 nanoparticle film catalysts under reaction times of 12 h, 24 h, and 36 h (Nabi et al., 2020).

Regarding the efficiency of photocatalysis induced by visible light, some studies have shown lower efficiency when visible light was applied comparing to irradiation at 254 nm (Tofa et al., 2019b; Uheida et al., 2021). However, this type of irradiation has been increasingly investigated to simulate solar irradiation, which can make the photocatalytic process greener. The great challenge of applying visible light is that the UV region only accounts for <5 % of the entire solar spectrum. In this way, some of the strategies for extending the optical absorption of TiO_2 into the visible region can be metal-doping, non-metal doping, and direct reduction of TiO_2 (Shoneye et al., 2022). Recent studies have indicated promising results in the removal of MPs (PE and PP) using solar light/visible light; however, some of them had reaction times greater than those reported at 254 nm. For instance, the total reaction time required in the study from Tofa and co-workers (Tofa et al., 2019b) was 175 h (using Platinum nanoparticle-coated ZnO nanorods catalysts). The reaction time in the study performed by Uheida and co-workers using visible light was 168 h reaching 95 % of MP volume reduction (using zinc oxide nanorods (ZnO NRs) catalysts) (Uheida et al., 2021). Another study also applied visible light, but with a reaction time of 50 h, indicating mass loss up to 71 % for HDPE MP (Ariza-Tarazona et al., 2020). On the other hand, no significant mass losses for high density polyethylene (HDPE) and low density polyethylene (LDPE) MP were reported in an investigation using similar reaction time and visible light (Llorente-García et al., 2020). In this case, the results were due to a combination of factors during the reaction such as size and shape of the MPs and surface-bounded $\bullet\text{OH}$ with the MPs by radical diffusion and stirring (Llorente-García et al., 2020).

2.2.3. Effect of other variables (MP shape and reaction interface)

The shape of the MPs can affect photocatalysis efficiency. A recent research using a mesoporous N- TiO_2 coating (pH = 3, 50 W visible LED Lamp (400–800 nm), and 50 h of reaction time) indicated that the shape of MPs can influence their dispersion and contact on the semiconductor during the mechanical agitation (Llorente-García et al., 2020). These authors proposed that stirring (300 rpm) favoured collisions between the dispersed MP spherical-shaped (HDPE, density 0.95–0.97 g/cm^3) and surface-bound oxidative species of the N- TiO_2 . On the other hand, MP film-shaped (LDPE, density 0.91–0.94 g/cm^3) showed low dispersion in the reactor with the accumulation of buoyant films. The MP film-shaped might have accumulated in the liquid–gas interface, which could hinder the absorption of light by N- TiO_2 , and/or decrease the oxygen diffusion from the air to the liquid phase, both affecting the efficiency of the photocatalytic process (Llorente-García et al., 2020).

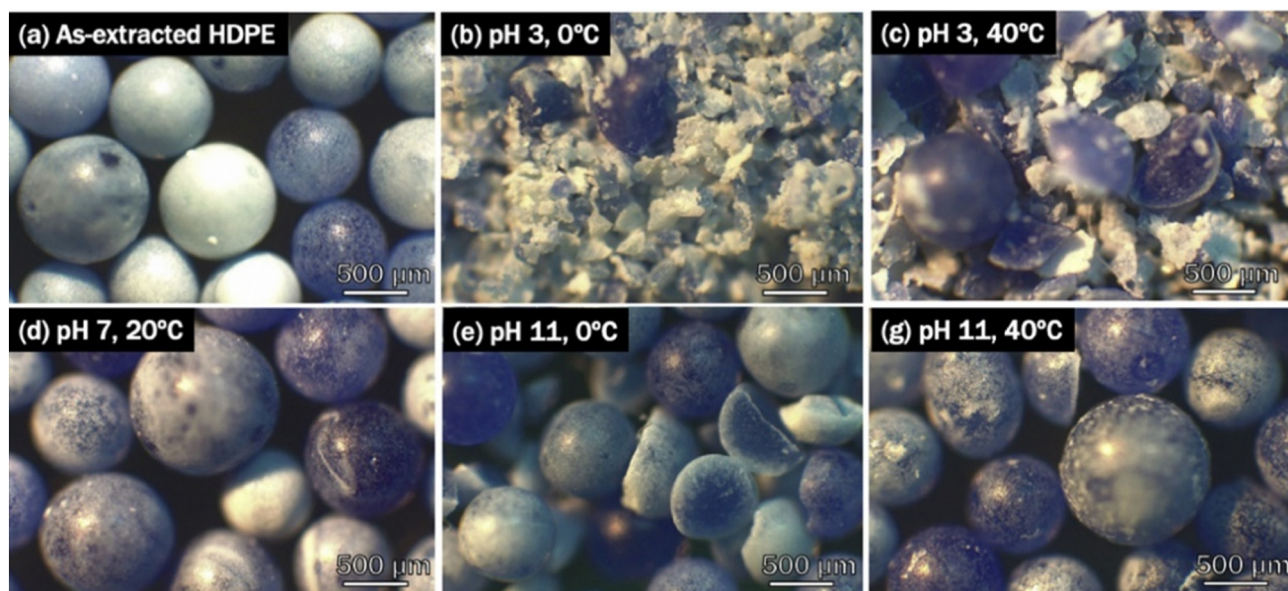


Fig. 4. Optical micrographs for high density polyethylene (HDPE) MPs as original HDPE and HDPE after different photocatalysis conditions (Ariza-Tarazona et al., 2020).

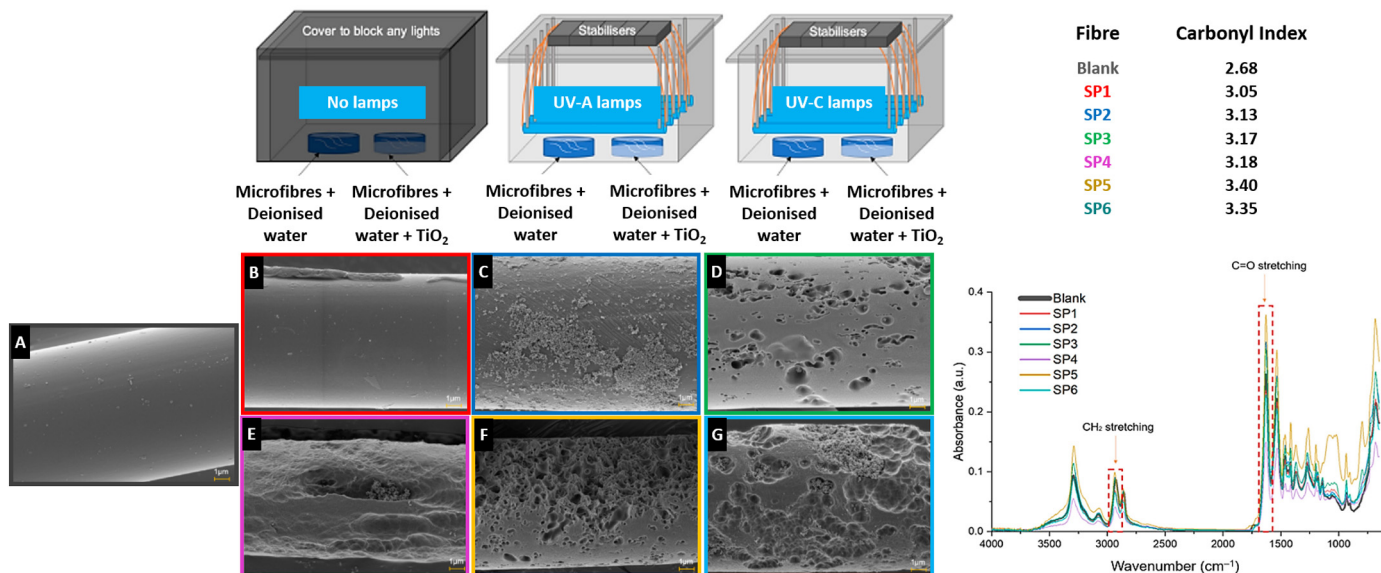


Fig. 5. Heterogeneous photocatalysis and control experiments using UV-A and UV-B lamps for microfibre PA66 degradation. SEM images (left side), FTIR spectra and carbonyl Index data (right side) show PA66 microfibre results after 105 h of treatments: (a) starting PA66 microfibre (blank), (b) PA66 microfibre suspended in deionized water (SP1); (c) PA66 microfibre in deionized water + TiO₂ (SP2), (d) PA66 microfibre in deionized water + UVA (SP3); (e) PA66 microfibre in deionized water + UVA + TiO₂ (SP4), (f) PA66 microfibre in deionized water + UVC (SP5), (g) in deionized water + UVC + TiO₂ (SP6) (Adapted from Lee et al., 2020).

The degradation efficiency of MPs can also change in different reaction interfaces (Nabi et al., 2020). PS MP mineralization through heterogeneous photocatalysis in aqueous solution showed slower kinetics because the contact/interaction between catalyst and MP particles becomes more difficult when in suspension. PS degraded by 44.66 % after 12 h of reaction in liquid phase. On the other hand, direct contact of MPs with the catalyst (TiO₂ nanoparticle films catalysts made with Triton X-100) in the solid phase favoured the direct attack of oxidative species (e.g. •OH) without any barrier/impeditive, reaching complete mineralization after 12 h and 36 h reaction for PS and PE MPs, respectively, under 254 nm. C—H stretching vibration of the aromatic rings and bending of the cyclic carbonyl groups (at 873 cm⁻¹ band), evidenced that the TiO₂ and UV irradiation accelerate bond cleavage of PS into complete mineralization (confirmed by thermogravimetric analysis). When the treatment was applied to ≤ 1 μm PS MPs, they were reduced to smaller particles (~250–400 nm). However, although total mineralization has been achieved (in MP particles > 1 μm), this study shows limitations in the feasibility of real-scale application, since for solid-phase degradation, the PS spheres solution (20 μL) was dropped onto the catalyst films. Therefore, for the treatment to reach such efficiency, MPs should be previously separated from the water body (e.g. by filters) before being subject to AOP.

In summary, the complete MP mineralization reported by Nabi and co-workers in solid system is still a challenge for liquid systems. A few studies have reported successful MP degradation in aqueous media: >70 % so far (Ariza-Tarazona et al., 2020; Kim et al., 2022; Lee et al., 2020). The MP removal results present in this section are summarized in Table 2.

2.2.4. Intermediate products and sustainability of the process

Photocatalytic systems achieve partial MP degradation in aqueous systems. However, most of these studies did not investigate intermediate products formed during the reactions. This is very important, because the partial oxidation may lead to the formation of intermediates, which are toxic to the environment and humans. Recently, results of PP degraded via photocatalysis with ZnO nanorods using visible light showed that intermediated products such as acetone, acetaldehyde, formaldehyde were formed (Uheida et al., 2021). In the same way, Nabi et al. and Tofa et al. identified some intermediates formed during the photocatalysis of PS and LDPE MPs, respectively (Nabi et al., 2020; Tofa et al., 2019a). In their studies, the organic species formed

on the PS surface were similar to those detected by Uheida et al., 2021, after photocatalysis.

There are no studies in the area of sustainability of photocatalytic systems applied for MP degradation. In general, greenhouse gas emissions in the TiO₂/UV-C treatment are mainly from UV lamp power consumption (Watt-hour) (Kang et al., 2020). Thus, the application of natural irradiation is an attractive route for the photocatalytic system. Additionally, it is important to include a sustainability assessment to the reaction time necessary for the improved efficiency of removal of the target compounds.

2.3. UV/H₂O₂ and ozone-based processes for MPs degradation

2.3.1. UV/H₂O₂ process

Ozonation and irradiation with UV are among the most advanced treatment processes applied in water treatment plants (Pivokonský et al., 2020; Wang et al., 2020; Yang et al., 2019). Concerning the UV system, the addition of chemicals to the water for the treatment can improve the degradation kinetics of the targeted pollutants. One of the chemical agents used in UV system is H₂O₂ (Deng and Zhao, 2015). The UV/H₂O₂ process has been intensively applied for environmental remediation, mainly for degradation of recalcitrant organic micropollutants (Dhawle et al., 2021; Lhotský et al., 2017). However, the application of this AOP for the removal of MPs has only been recently studied.

In order to understand how MPs become affected in drinking water treatment, the process involving UV/H₂O₂ for up to 6 h (with UV light-Xenon arc lamp, 300 W; temperature kept at 22 ± 1 °C; ~4 × 10⁷ microplastics/mL) was carried out after the stages of coagulation/sedimentation and sand filtration (Na et al., 2021). There, coagulation and sand filtration removed larger PS microbeads (>20 μm), and the UV/H₂O₂ treatment treated the smallest size fraction (10 μm) of MPs. The concentration of larger sized MPs (~10 μm) decreased considerably after 1 h exposure to UV/H₂O₂. Moreover, there was a gradual increase in particle concentration over time, especially within 1–2.5 μm size range, indicating fragmentation of PS.

The aromatic rings in PS (see Table S1) are sensitive to UV irradiation which facilitates their degradation (X. Liu et al., 2021). Also, subsequent formation of free radicals on the surface of MPs can favour the initiation of chain reactions up to the complete mineralization of the MPs, but only if the operational conditions are sufficient to do so (W. Liu et al., 2021).

Table 2
Heterogeneous photocatalysis for MPs degradation.

Experimental conditions (Photocatalysis)	Matrix	Types of polymer	Size/concentration of the MPs/MFs	Degradation and performance	Size of the MPs/MFs (after AOP)	Variables that influence the AOP/other observations	Impact of the treatment on MPs ^a	Ref.
Platinum nanoparticle-coated ZnO nanorods catalysts under visible light 50 W; reaction time of 175 h.	Deionized water	LDPE (film)	1 cm ² 50 µm thick	Carbonyl index and vinyl index increased by 15 % and 13 %, respectively.	-----	ZnO-Pt photocatalyst has 78 % enhancement in Vis absorption compared to ZnO photocatalyst	On the MP surface: oxygenated groups	(Tofa et al., 2019b)
TiO ₂ nanoparticle films catalysts - made with Triton X-100, under two UV lights lamps- 254 nm; reaction time of 12 h, 24 h, and 36 h.	Deionized water	PS (microsphere) and PE powder)	Liquid phase: 100 mL of PS solution (100 µL of PS -5 µm size) in 100 mL water	Solid system: complete mineralization (PS in 12 h and PE in 36 h). Liquid system: 44.66 % PS degradation after 12 h	PS diameter before and after solid system, respectively: 5 µm and 0 nm/1 µm and 401.28 nm	UV 254 nm was more effective than the 365 nm.	On the MP surface: PS: hydroxyl, carbonyl groups	(Nabi et al., 2020)
Five UVC lamps - 8 W; 100 mg of catalyst TiO ₂ /L; reaction ≤ 105 h	Deionized water	Microfibre (PA66)	1.0 m length (~1.3 mg, diameter of 10 µm)/50 mL of deionized water	97 % mass loss (48 h) and < 10 mg/L COD. Carbonyl index increased by 25 %.	-----	UV 254 nm was more effective than the 365 nm;	-----	(Lee et al., 2020)
Mesoporous coating of N-TiO ₂ catalysts under UV 50 W Visible LED Lamp; pH 3.0; 300 rpm; reaction time of 50 h	CH ₃ COONa/CH ₃ COOH buffer (pH 3)	LDPE (film) and HDPE (microbead)	Film sizes: 5 mm × 5 mm and 3 mm × 3 mm Microbeads sizes: 814 ± 91 µm and 382 ± 154 µm	No increase in carbonyl index observed for the larger films and for the larger and smaller microbeads	-----	Degradation was affected by MPs' size and shape	-----	(Llorente-García et al., 2020)
ZnO nanorods catalysts under visible light 50 W; reaction time of 175 h	Deionized water	LDPE (film)	1 cm × 1 cm	Mass loss 0.22–4.65 % 30 % increase in the carbonyl and vinyl indexes for longer ZnO rods	-----	Degree of oxidation directly proportional to the catalyst surface area	On the MP surface: carbonyl groups hydroperoxide, peroxides, and unsaturated groups	(Tofa et al., 2019a)
TiO ₂ - P25/β-SiC catalysts foams under UV-A radiation-112 W/m ² ; pH value of 6.3; reaction time of 7 h	Distilled water	PMMA and PS nanoparticle	600 mL/L of nanoplastic suspension	50 % of TOC conversion of the PMMA	-----	pH affected the surface charge of the catalyst	-----	(Allé et al., 2021)
ZnO NRs coated on glass fibre substrates under visible light 120W; flow rate of 300mL/min; reaction time of 456 h ^b	Deionized water	PP	154.8 ± 1.4µm 10 ⁴ particles (~70 mg/L)	Carbonyl index value >40 65 % reduction of particle volume	108.2 ± 2.5 µm after reaction	Increase in exposure time increased the intensities of the carbonyl band and the hydroxyl band and reduced the PP size.	On the MP surface: hydroxyl and carbonyl groups In solution: acetyl radicals, hydroxypropyl, acetone, pentyl group	(Uheida et al., 2021)
C,N-TiO ₂ catalyst under visible light 50 W LED lamp; pH 3, 7 and 11; and 0, 20, 40 ± 2 °C; 300 rpm; reaction time of 50 h	Dispersion of MP and catalyst (equivalent of 0.4 % w/v)	HDPE (spherical beads)	Average diameter:725 ± 108 µm 200 mg/50 mL of solution	71.77 ± 1.88 % of average mass loss (T = 0 ° C and pH = 3)	-----	MP degradation depends on pH and temperature	-----	(Ariza-Tarazona et al., 2020)

HDPE = high density polyethylene; LDPE = low density polyethylene; PA66 = polyamide 66; PMMA = polymethylmethacrylate; PP = polypropylene; PS = polystyrene.

----- Not informed or not evaluated.

^a There was no toxicity assessment of intermediates in the studies presented in this Table.

^b Continuous flow system. The other systems presented in the Table were evaluated without continuous flow, on a laboratory scale.

However, from the formation of smaller plastic particles, only a partial degradation of the MPs is expected (Na et al., 2021). In this way, the GC–MS analysis of the leachates revealed that a complex mixture of intermediate organic compounds such as benzaldehyde, methylbenzaldehyde, acetophenone and benzoic acid was formed (Na et al., 2021). Toxicity tests carried out from the intermediates in the water treated (evaluated from the inhibition of β -galactosidase synthesis in *E. coli*) indicated that 40–80 % of the enzymatic activity was inhibited after 6 h of UV and UV/H₂O₂ treatment, respectively. Similar oxidation intermediates have also been reported for other MPs (Capolupo et al., 2020; Castelvetro et al., 2021).

In water treatment plants, UV irradiation time for oxidation and disinfection purposes is much shorter (up to 1 h of UV exposure on average) (EPA, 1999) than the times investigated in some studies reported in this review article (generally, above 5 h of UV incidence). Thus, although there is some progress achieved on this subject so far, more studies are needed to better understand the toxicity potential of products released into waters through real operating conditions.

2.3.2. Ozone-based processes for MP degradation

Another promising strategy for MP degradation are the ozone-based processes. In short, ozone can react with pollutants via different ways, such as direct oxidation, where the oxidation of the carbon double bonds (e.g. aromatic hydrocarbons) can lead to the formation of smaller molecules. Furthermore, via indirect reactions, O₃ decomposes into free radicals (mainly •OH), which can react with the pollutants in water (Ikehata and Li, 2018). A recent study evaluated the ozonation of PE MPs for 60, 120 and 180 min (O₃ dose of 4 to 7 mg/min) (Zafar et al., 2020). The oxidant concentration and reaction time were relevant in the absorption of ozone by the PE MPs. The challenging removal of small MPs was addressed by ozonation, specifically for degrading nano-sized PS (≤ 200 nm) (Li et al., 2022). The treatment process was performed in a semi-continuous flow mode at initial pH 6.4, with 2.5 μ m/L of PS and ozone dosage of 4.1 mg/L. Fourier transform infrared (FTIR) and X-ray photoelectron spectroscopy (XPS) results showed that the ozonation introduced O-containing groups in the nano-sized PS, which altered the binding energy of carbon and increased the hydrophilicity of the PS. In the same investigation, GC–MS spectra confirmed the formation of intermediate products (such as formic acid, phenol, acetophenone, hydroquinone, methylbenzaldehyde, methyl acetophenone, and phenylpropionic acid) during the degradation steps; therefore, PS were not totally mineralized. The ozonation treatment reached 42.7 % mineralization in 240 min (Li et al., 2022).

The use of ozonation as an advanced treatment in full scale treatment plants has been a matter of concern due to the increase in MP formation. According to a recent study, the removal of MPs in a DWTP using ozonation integrated with GAC filtration as an advanced treatment stage showed an increase of small MP and MF particles in the size range of 1–5 μ m after ozonation (Wang et al., 2020). Another study carried out at a DWTP (located in China) evaluated the removal performance of 5 μ m – 5 mm MPs and 100 μ m – 5 mm MFs for all water treatment units, including ozonation (Wu et al., 2022). The authors identified that the steps of coagulation/flocculation and sedimentation combined with sand filtration could not effectively remove MPs ≤ 20 μ m (removal efficiency <50 %) (Wu et al., 2022). Besides, an increase of 13.8 % of granular MPs ≤ 20 μ m was observed after ozonation. It was noted that the ozonation and pre-ozonation stages might accelerate the MP embrittlement, resulting in the formation of smaller particles. In addition to the direct reaction of O₃ with the target pollutants, the production of •OH can be improved using alternative oxidants or irradiation (indirect approach). One way to achieve this is by combining the oxidants H₂O₂ and O₃, known as peroxone (O₃/H₂O₂) (Deng and Zhao, 2015; Ikehata and Li, 2018). A bench-scale study analysed the impact of the O₃/H₂O₂ process on PP, PE, and PS MPs (200–600 μ m) (under experimental conditions of 500 mg/L of MPs, O₃ dose of 88 mg/L for 10 min, and O₃:H₂O₂ molar ratio of 0.5) (Gomes de Aragão Belé et al., 2021). They found that PS MP presented greater susceptibility to chemical attack compared to PE and PP. This may have occurred due to the PS structure

including aromatic rings, from which it is inferred that the rings are a preferential site for reaction of oxidants such as O₃ by the introduction of OH groups on the PS surface (Murakami et al., 2003). Another study investigated the changes in the chemical structure of PE MPs by O₃/H₂O₂, using an ozone gas flow rate of 1, 3, and 5 L/min and a reaction time of 180 min, in such system, AOP favoured the increase of the intensity of the O–H and C=O bands on the PE surface. The highest carbonyl index (CI value = 1.33) was found when the treatment was carried out at initial pH 12 with a flow rate of 3 L/min, showing the degradation of PE MPs (Amelia et al., 2022). This method was suggested as a pre-treatment for a biodegradation process. PE degradation by ozone-based processes was recently evaluated applying UV irradiation (under ozone dosages of 4–7 mg/min during 180 min). The increase of O₃ dose was up to 6 mg/min and it showed a gradual increase of the levels of carbonyl (ketone and esters) and vinyl groups (Zafar et al., 2021). PE has single carbon bonds in its structure, which are quite resistant to photo-oxidative degradation due to the lack of UV-visible chromophores. However, it also contains an unsaturation (C=C), making it vulnerable to some oxidants (e.g. O₃) or unstable hydroperoxide. The latter can be transformed into a stable carbonyl group, which absorbs UV (Chamas et al., 2020; Lastovina and Budnyk, 2021).

Fig. 6 presents results of UV/H₂O₂ and ozonation processes investigated for the degradation of MPs.

It is worth highlighting that a very limited number of studies have reported the degradation of MPs by oxidative treatments in real water matrices (Wang et al., 2020; Wu et al., 2022). Most of the AOP studies use commercially sourced MPs, with pre-established concentrations and in synthetic solutions (Feng et al., 2011; Liu et al., 2022; Mackulak et al., 2015; Miao et al., 2020; Nabi et al., 2020). This makes it difficult to compare their efficiency with real high-complexity matrices. In addition, the quantification of MPs before and after degradation is not reported, and for real matrices, this quantification is more complex. To date, most studies have evaluated mass reduction of MPs using analytical balances (Hu et al., 2022b; Lee et al., 2020; Llorente-García et al., 2020; Miao et al., 2020) and the variation of MP size using optical microscopy with or without the use of software to support MP counting (Ariza-Tarazona et al., 2020; Llorente-García et al., 2020; Luo et al., 2021; Uheida et al., 2021). For samples of natural water, the identification of particle size is a challenge, requiring, for example, different steps to separate MP sizes (for example, through sieves columns) (Wu et al., 2022). Moreover, monitoring the degradation of MPs varies widely between studies, such as the analysis of total organic carbon (Allé et al., 2021; Na et al., 2021), molecular mass (Feng et al., 2011), and chemical oxygen demand (Lee et al., 2020), among others (Nabi et al., 2020). Hence, analytical methods and approaches to report the effect of oxidation should be standardized so that studies can be compared effectively.

2.3.3. Sustainability of UV/H₂O₂ and ozone-based processes for MP degradation

The sustainability of UV-C/H₂O₂ process and ozonation has been reported in a previous study where different AOPs were evaluated for the removal of Bisphenol-A from water (Kang et al., 2020). UV/H₂O₂ was the most effective in terms of the carbon footprint compared to ozonation (3 mM H₂O₂/UV-C, 4 mg/L ozonation), with a carbon footprint of 0.565 kg CO₂ eq/m³ and 3.897 kg CO₂ eq/m³ for UV/H₂O₂ and ozonation, respectively. The amount of greenhouse gas emission (GHG) from the UV-C/H₂O₂ process was less than that from the ozonation process. This may be due to power requirements in the O₃ treatment. Furthermore, with respect to the sustainability of the AOPs UV/H₂O₂ and ozone-based processes, some factors inherent to each treatment should be considered such as energy consumption, oxidant demand, total treatment time and the efficiency of MP removal by the process. It's important to highlight that the oxidant demand (i.e. O₃ and H₂O₂) might be higher when applying those AOPs for MPs compared to Bisphenol-A. According to the studies reported by this review (see Sections 2.1–2.3), due to the MP recalcitrant characteristics, most of them used significant doses of oxidant and UV irradiation for a long period. At last, UV/H₂O₂ and those O₃-based processes do not depend on extensive areas to perform the treatment process, which allows

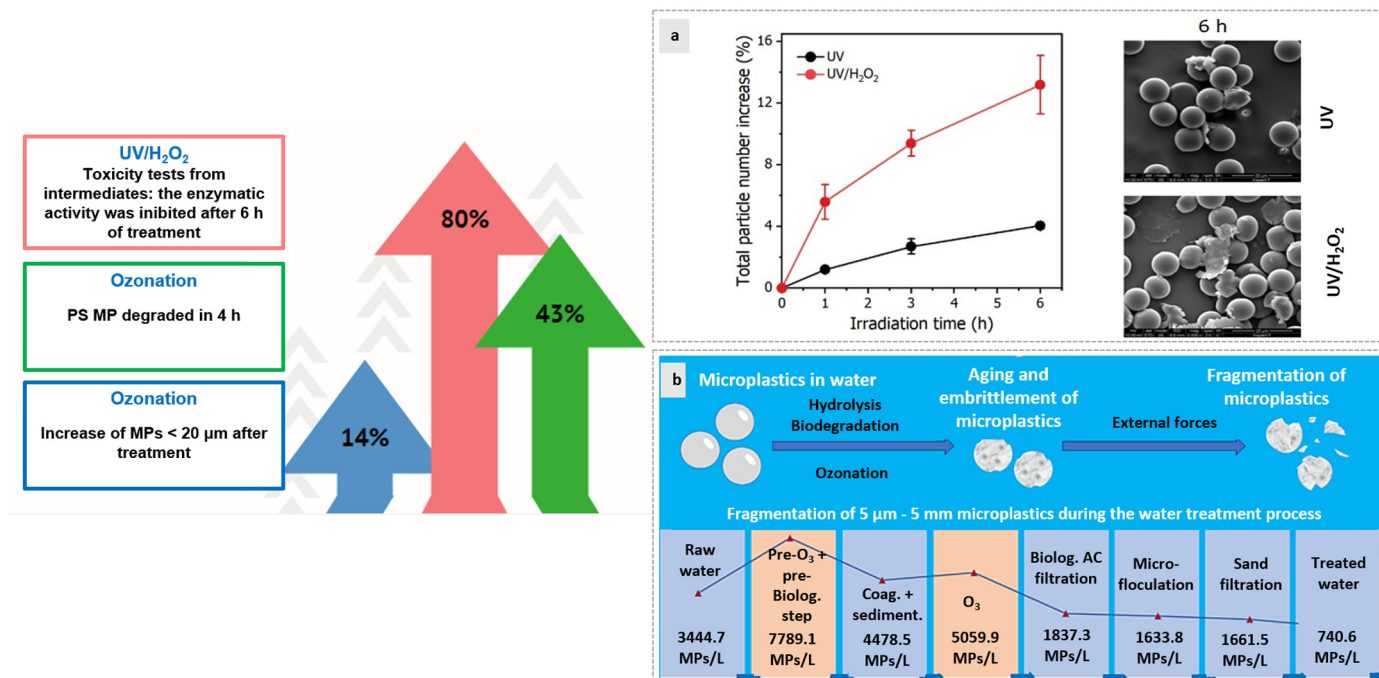


Fig. 6. Left side: results of UV/H₂O₂ and ozonation treatment for MPs. Right side: (a) particle number increase of 10 µm PS MPs during UV irradiation and UV/H₂O₂ treatment, and SEM micrographs of 10 µm PS MPs obtained during these treatments (adapted from Na et al., 2021), and (b) MPs fragmentation after ozonation treatment step in drinking water treatment (adapted from Wu et al., 2022).

both to be sequentially installed after conventional water treatment methods.

3. Interactions between MPs and micropollutants after AOPs

MPs weathered during environmental exposure or by AOPs acquire different sorption capacities (Li et al., 2020; X. Liu et al., 2021). Since the hydrophobicity of MPs is relevant to their sorption behavior with other chemicals, the potential for accumulation of organic pollutants on their surface can be high, especially when not being oxidized, and this favours the sorption of some pollutants such as persistent organic compounds, pharmaceuticals, and personal care products (Bui et al., 2020; Liu et al., 2020; Munoz et al., 2021; Yao et al., 2022). MFs can adsorb chemicals differently from natural-based fibres due to having different surface area and surface chemistry. Sorption properties will be influenced by the type and abundance of functional group and size and shape of the MPs (Grancaric et al., 2005). However, these properties are subject to change when the polymer is aged/oxidized. The oxidation of the MPs is expected to reduce the adsorption of chemicals based on van der Waals interactions. Moreover, interaction between MPs and organic pollutants will depend on both the properties of the sorbate and the sorbent, as well as the medium properties (e.g. dissolved organic matter, salinity and pH) (Menéndez-Pedriz and Jaumot, 2020).

When the AOP O₃/H₂O₂ (peroxone) was applied in the presence of PP, PE, PS MPs and two types of pesticides (chlorpyrifos-CPF and dichlorvos-DDVP) (Gomes de Araújo Belé et al., 2021), the presence of ketones on the surface of all oxidized MPs were detected. There was lower hydrophobic interaction between MPs and pesticides after the oxidation of MPs. Similarly, MPs submitted to the Thermal Fenton process also showed a reduction in their hydrophobicity after 8 h of treatment evidenced by the increase in O/C proportions (Hu et al., 2022b).

In relation to AOPs applied for the purpose of aging MPs, some of them have also indicated a change in the ability of MPs to interact with pollutants. Liu et al. reported that the aging of PS MPs using UV/H₂O₂ increased the abundance of groups like carbonyl or hydroxyl on the PS surface, and thus, increased its surface polarity (X. Liu et al., 2021). Also, the addition

of BPA in the aqueous medium together with the MPs after the AOP showed a significant reduction in the adsorption of BPA. One of the hypotheses suggested is that the carbonyl or hydroxyl groups formed after the photo-assisted treatment combined with water through hydrogen bonds began to compete with BPA for sorption sites. In contrast, increasing the degree of oxidation on the surface of PS and PE MPs after the Fenton process increased the adsorption capacity of MPs with the hydrophilic antibiotic ciprofloxacin (Liu et al., 2019). Furthermore, intermediate products that formed during the AOP can also affect the adsorption onto MPs. For example, intermediates released in the aqueous solution can compete for adsorption sites with some pollutants. A recent study showed that intermediates adsorbed on the MP surface with low aging degree decreased the adsorption of pharmaceutical Atorvastatin (ATV), hence there was competition for active sites, but increased the adsorption of pharmaceutical Amlodipine (AML) (Liu et al., 2020). On the other hand, when aged MPs were subjected to the same water, intermediates generated from the drug had lower sorption than AML. This lower adsorption can be explained by weaker interaction between the degradation products and weathered MPs caused by electrostatic repulsion (Liu et al., 2020).

The fate of micropollutants and the toxicity of MPs can be affected by the interaction of aged polymers with pollutants. More studies are needed to understand the mechanisms of interaction between MPs altered by AOPs and other substances, mainly micropollutants. Current information on sorption properties of MPs subjected to AOP treatment is summarized in Table S2.

4. Concluding remarks and research gaps

WWTPs and DWTPs are not able to efficiently remove MPs by their treatment units. Moreover, the higher presence of fibres than other MP shapes in the influent and effluent of some WWTPs and DWTPs has highlighted the need for investigating other technologies for MP degradation. Therefore, in this review, the current panorama of AOPs applied for MPs degradation (including MFs) has been highlighted.

Significant findings were presented based on a systematic review of recent studies:

- The efficiency of AOPs for removing MPs can differ in relation to the reaction conditions, such as temperature, pH, UV irradiation, reaction time, MP shape and reaction interfaces (solid or liquid).
- Heterogeneous photocatalytic processes have been the most studied AOP for MP degradation, while the Fenton process, and its variations, have been scarcely investigated.
- There is a negative removal of MPs (including MFs) from ozonation and heterogeneous photocatalysis mainly from MPs $\leq 10 \mu\text{m}$.
- Most toxicity tests of intermediates formed from the oxidation of MPs indicated non-toxicity to bacteria and microcrustaceans. However, more studies are needed since the AOPs reported by this review can only degrade partially MPs.
- Heterogeneous photo-Fenton and photocatalysis are capable of reducing the volume of some MPs (e.g. PVC and PP) by up to 96 %. However, reaction times are impractical at full scale (5 h – 400 h).
- Studies of AOPs in aqueous media have shown less efficiency in the degradation of MPs compared to studies in solid interfaces.
- Oxidized MPs showed lower hydrophobicity and sorption behavior with other micropollutants (e.g. BPA, pesticides, and some pharmaceuticals).

Despite AOPs being a promising alternative for the degradation of MPs, providing degradation efficiency on average of 10–70 %, there are still limited number of studies on this subject. Studies on AOPs addressing the degradation of MPs with realistic and detailed operational parameters are needed. It is essential that further studies focus on the fragmentation and formation of smaller sizes of MPs after oxidative treatments, in spite of current challenges in detecting, characterizing and quantifying nanoparticles and MFs. Also, investigations on the degradation of MFs by AOPs are urgent. Moreover, there is not a general rule on how AOPs affect the physico-chemical properties of MPs and their interactions with pollutants.

Investigations using real aquatic matrices and continuous systems are other knowledge gaps worth exploring. Additionally, most of the experiments were carried out in batch mode, which do not correspond to the treatment in use in wastewater and drinking water treatments. It is also worth noting that the degradation efficiency of MPs needs to be considered along with other parameters that can potentiate the carbon footprint of the process. Technologies that reduce electricity consumption, such as the application of solar irradiation in photolysis systems are an alternative.

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CRediT authorship contribution statement

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Naiara de Oliveira Dos Santos (Investigation and Writing) - conducting the literature review process, specifically data/evidence collection, and preparation of the manuscript, writing the initial draft and incorporating the suggestions/comments by the research supervisors.

Rosa Busquets (Supervision and Writing) - Oversight responsibility for the research activity planning and execution, and preparation of the manuscript through critical review, commentary or revision.

Luiza C. Campos (Supervision, Writing and Project Administration) - Leadership responsibility for the research activity planning and execution, preparation of the manuscript through critical review, commentary or revision, and management and coordination responsibility for the research.

Data availability

Data used in our manuscript have been published elsewhere and this is clearly referenced

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.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2022.160665>.

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