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Nano/Microplastics Capture and Degradation by Autonomous Nano/Microrobots: A Perspective

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The growing use of plastic materials has led to the continuous accumulation of wastes in marine environments, which fragment into hazardous micro- and nanoplastics. These plastic particles absorb toxic organic pollutants on their surface, support bacterial biofilms growth, and propagate through the food chain, posing serious risks for human health. Therefore, nano/microplastics pollution has become a global issue, making their definitive elimination compulsory. Self-propelled nano/microrobots have demonstrated efficient removal of nano/microplastics from water, combining enhanced physicochemical properties of nano/microscale materials and active motion. During the last year, the potential of this technology to degrade nano/microplastics has been investigated. Here, the most advanced strategies for nano/microplastics capture and subsequent degradation by autonomous nano/microrobots are critically reviewed. A short introduction to the main propulsion mechanisms and experimental techniques for studying nano/microplastics degradation is also provided. Forthcoming challenges in this research field are discussed proactively. This perspective inspires future nano/microrobotic designs and approaches for water purification from nano/microplastics and other emerging pollutants.

1. Introduction

Plastics are synthetic polymers constituted by hundreds to thousands of organic subunits, called monomers, linked by strong covalent bonds.^[1] Their excellent properties, among which high chemical and thermal stability, make them widely used and, at the same time, extremely difficult to eliminate. As a consequence, the amount of plastic waste accumulating in the world is continuously increasing.^[2,3] The images of marine environments full of plastic bags, bottles, and other products are particularly impressive.^[4,5] However, the real danger associated with plastic pollution results from the gradual fragmentation into smaller particles under the action of the sea and weathering (**Figure 1**).^[6] Plastic pieces with sizes below 5 mm are named microplastics.^[7] These can serve as a substrate for the adsorption of other pollutants or the growth of bacterial biofilms, becoming more toxic.^[8,9] Microplastics have already entered the food chain. They can be ingested by animals, such as fish, propagate through the food chain until reaching our table, or directly contaminate drinking water systems, posing severe threats to the health of all living beings.^[10,11] Unfortunately, microplastics can further break down into smaller, more hazardous pieces (<1 μm), termed nanoplastics.^[12–14] While microplastics tend to sediment on the seafloor, nanoplastics remain suspended in water due to

their lightness and diffuse rapidly because of the marine currents.^[15] They also adsorb larger amounts of pollutants and bacteria due to their higher surface-to-volume ratio than microplastics and penetrate tissues easily owing to their tiny size.^[15,16] On these bases, nano/microplastic pollution represents a rising concern.

Traditional approaches for separating suspended matter, such as filtration, are effective against microplastics but unsuitable for nanoplastics.^[17] Once removed, these have to be recycled or eliminated definitively. Incineration is efficient yet unsustainable due to the release of detrimental gases.^[18] For this reason, there is a pressing demand for the development of environmentally friendly solutions for nano/microplastics removal from contaminated waters and their subsequent total degradation.

Nano/microrobots are at the forefront of materials science and nanotechnology research. They are nano/micromaterials with the ability to harvest energy from their surroundings and convert it into self-propulsion while performing various tasks.^[19,20] Their active motion induces a local mixing effect, allowing them to overcome the diffusion limit that affects conventional water purification methods.^[21–24] This, in turn, promotes contact with pollutants and accelerates their degradation. Simmchen's group reported the first pioneering work on microplastics removal using UV-light-powered magnetic microrobots in the presence of H_2O_2 .^[25] Microplastics extracted from personal care products, rivers, and sea, were collected by phoretic interactions with microrobots or shoveling through self-assembled microchains formed under a magnetic field. Later, magnetic field-actuated microsubmarines based on sunflower pollen grains were used to catch, carry, and release microplastics.^[26] Another removal mechanism was the adsorptive bubble separation induced by iron oxide-manganese dioxide core-shell microrobots.^[27] The oxygen bubbles generated during their movement trapped microplastics and suspended them in a foam layer on the top of the vessel, facilitating their collection. Despite promising, these approaches do not consider the elimination of removed microplastics.

Recently, many efforts have been devoted to the “on-the-fly” degradation of microplastics by autonomous motile microrobots. This work briefly introduces their motion mechanisms and summarizes the experimental techniques to analyze nano/microplastics. Then it critically describes the most significant advances in nano/microrobotics for polymers and plastics capture and degradation. The future challenges towards the practical application of these strategies are also discussed. The considerations and conclusions of this work are not limited to nano/microplastics but, in principle, can be applied to other pollutants.

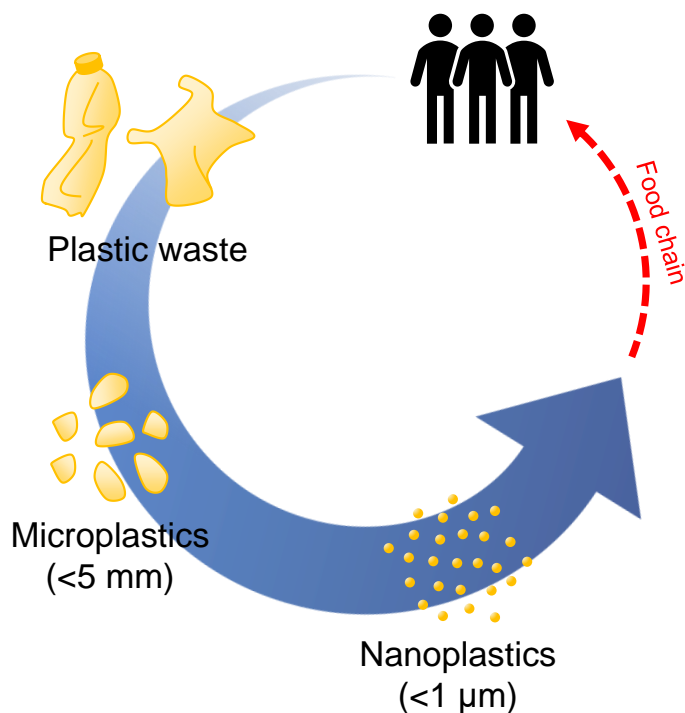


Figure 1. Plastic waste fragmentation into microplastics and nanoplastics.

2. Motion Mechanisms of Nano/Microrobots Against Nano/Microplastics

Nano/microrobots can be categorized into two main groups, whether their self-propulsion is obtained by external energy sources or by consuming chemical fuels, as bacteria do.^[28] Externally driven nano/microrobots convert energy from light, magnetic and acoustic fields into mechanical power. Catalytic ones move thanks to catalyzed chemical reactions involving fuels in their surroundings. For example, tubular microrobots propel through the continuous ejection of O_2 bubbles resulting from Pt catalyzed H_2O_2 decomposition.^[29] Janus nano/microrobots fueled by H_2O_2 can be prepared by asymmetrically coating nano/microparticles with a Pt layer.^[30] Alternatively, enzymes (glucose oxidase, urease) can be asymmetrically immobilized to power them by reacting with biological fuels (glucose, urea).^[31] Water represents fuel for disintegrating nano/microrobots, like Mg-based ones.^[32]

Nano/microrobots employed in plastic waste capture and degradation are principally propelled by photocatalytic reactions, eventually in the presence of H_2O_2 , or by magnetic fields. Therefore, the fundamentals of these externally driven motion mechanisms are presented in the following paragraphs.

Light, especially sunlight, is a powerful and abundant energy source to power nano/microrobots.^[33] These can be designed to respond to light stimuli by generating a

gradient of temperature or (charged) products, leading to thermophoretic, diffusiophoretic, and electrophoretic propulsion.^[34] Photocatalytic semiconductors are the key building blocks for light-driven nano/microrobots for water purification.^[23] They use light to move and, contemporarily, produce reactive oxygen species (ROS), such as $\cdot\text{O}_2^-$, $\cdot\text{OH}$, $\text{HO}_2\cdot$, inducing the photocatalytic degradation of pollutants in water. Typically, the photocatalytic semiconductor is not able to generate the motion alone. A noble metal layer can be deposited to form a “two-faced” Janus nano/microrobot, breaking its symmetry.^[35] The mobility mechanism of this type of nano/microrobot is illustrated in **Figure 2(a)**. Under light irradiation, the photocatalytic semiconductor absorbs photons with energy equal to or higher than its bandgap. Electrons are promoted to the conduction band, leaving holes in the valence band. The metal/semiconductor junction favors photogenerated carriers separation, suppressing detrimental recombination phenomena. Transferred electrons from the semiconductor conduction band to the metal and holes left in its valence band decompose water and H_2O_2 , if present. Protons (H^+) consumption at the metal side generates a protons gradient, establishing a local electric field that causes the nano/microrobot’s movement by self-electrophoresis.^[36] As a replacement for expensive noble metals, heterojunctions between two different semiconductors have been suggested.^[37,38] Instead, some photocatalytic semiconducting microparticles can propel without noble metals or heterojunctions due to their intrinsic structural asymmetry, which generates a products gradient driving their motion *via* self-phoresis.^[39]

Magnetic nano/microrobots offer the advantage of precise navigation, which is helpful in biomedicine and microplastics removal.^[40] They can be manipulated to reach targeted microplastics, capture and collect them. Moreover, magnetic fields can produce and control swarms of nano/microrobots to perform complex tasks that single entities can not accomplish.^[41] Magnetic actuation is achieved by integrating magnetic nanostructures into nano/microrobots, for example, magnetic Fe_3O_4 nanoparticles. However, Purcell’s scallop theorem states that, in a low Reynolds number fluid, the time-symmetric motion of a swimmer can not bring to a net displacement.^[42] Asymmetric geometries and flexible structures are accordingly required to break this theorem. The movement of most magnetic nano/microrobots is based on a magneto-phoretic mechanism in magnetic field gradients, i.e., spatially inhomogeneous fields, or the magnetic torque transfer induced by rotating magnetic fields.^[28] Three orthogonal coil pairs can generate rotating fields, as shown in Figure 2(b). These are characterized by a field vector rotating on a plane. Conical rotating fields are obtained by adding a magnetic field perpendicular to the rotating field plane. The latter are utilized to power the magnetic microrobots for microplastics removal in section 4.2.3.

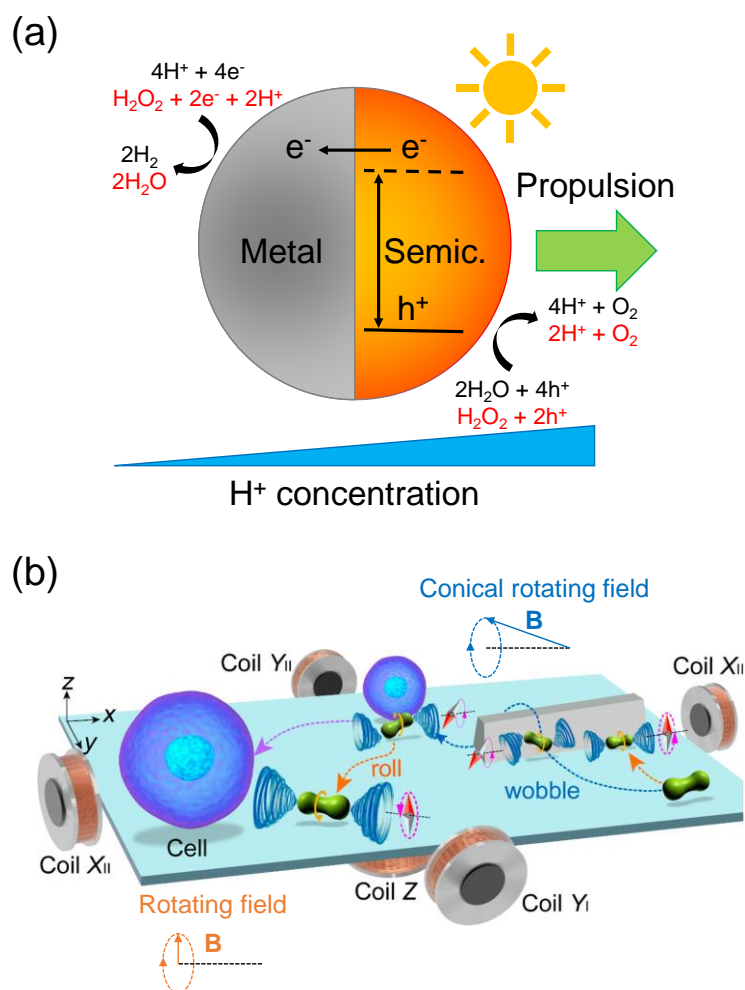


Figure 2. Schematic illustrations of (a) light-driven propulsion mechanism of metal/semiconductor Janus nano/microrobots and (b) magnetically actuated microrobots moving in rolling and wobbling modes under rotating magnetic fields for cell manipulation. Adapted with permission.^[43] Copyright 2018, American Chemical Society.

3. Measurement Techniques for Nano/Microplastics Degradation

This section lists the most valuable techniques for studying nano/microplastics and assessing their degradation, which helps interpret the results described in the following section. However, it is limited to illustrating their *pros* and *cons* rather than explaining their working principles.

The degradation of organic pollutants, such as dyes, is usually monitored by the decrease of their characteristic UV-Vis absorbance peaks.^[44,45] Most nano/microplastics do not possess these fingerprints or contain interfering additives, making UV-Vis spectroscopy unsuitable for evaluating their degradation. On the contrary, well-established techniques are Fourier-transform infrared spectroscopy (FTIR), Raman spectroscopy, and mass spectrometry

methods.^[46–48] The latter allow qualitative and quantitative determination of degradation products, which permits the evaluation of degradation pathways and potential secondary pollution. For instance, pyrolysis-gas chromatography-mass spectrometry (Pyr-GC-MS) provided information on byproducts formed during the aging of various microplastics,^[49] while size exclusion chromatography (SEC) disclosed the reduction of their molecular weight due to the induced oxidation. Smaller microplastics (<100 μm), and thus nanoplastics, are challenging to handle by Pyr-GC-MS.^[50] Contrarily, matrix-assisted laser desorption/ionization-time-of-flight mass spectrometry (MALDI-TOF MS) is an ionization technique that offers mass spectra of high molecular weight polymers, including plastics, with a resolution of the individual n-mers within the polymer mass distribution.^[51] Compared to typical ionization methods, it does not cause sample fragmentation. Hence, it is ideal for unequivocally investigating the degradation of nano/microplastics.

X-ray photoelectron spectroscopy (XPS) can be used to observe variations of the chemical composition of nano/microplastics related to their oxidation.^[52] Nevertheless, it is sensitive only to the sample's surface because of the small X-ray penetration. Thus, it can be used only to confirm their initial degradation.

Optical microscopy and scanning electron microscopy (SEM) visualize changes in the topography, morphology, and composition of microplastics before and after the degradation. These comprise the formation of microcracks and cavities, as reported for photodegraded microfibers.^[53]

Due to the minuscule size of nanoplastics, transmission electron microscopy (TEM) may be required for displaying them.^[54] Otherwise, nanoparticle tracking analysis (NTA) consents imaging nanoplastics by recording the diffracted light from an incident laser and furnishes information on their size distribution, concentration, and Brownian diffusion.^[55] It has been employed to examine the temporal fragmentation of disposable polystyrene (PS) coffee cup lid into nanoplastics.^[56] NTA shows potential in exploring the nanoplastics removal ability of nano/microrobots.

4. Strategies for Nano/Microplastics Capture and Degradation by Nano/Microrobots

This section presents the most innovative approaches for microplastics degradation using self-propelled nano/microrobots. They all show a common feature: the establishment of close contact between microrobots and microplastics before their degradation. In fact, for conventional photocatalysts, it is well known that this remarkably improves the efficacy of the

remediation process.^[57] Generally, it is obtained under constant (magnetic) stirring. Instead, microrobots self-propulsion creates a stirring effect locally in an energy-saving manner, enabling them to face more contaminants per unit time. To strengthen the contact, microrobots must be equipped with adhering properties. On these bases, first, the approaches for nano/microplastics capture are described, followed by the strategies for their successive degradation, as summarized in **Table 1**. Despite the reviewed examples referring to polymers and microplastics, they can be naturally extended to nanoplastics and other emerging pollutants.

Table 1. Nano/microplastics capture and degradation strategies by self-propelled nano/microrobots.

Year	Material	Fabrication method	Size (μm)	Powering source	Targeted material	Capture mechanism	Degradation mechanism	Ref.
2019	Au@Ni@TiO ₂	TTIP hydrolysis and condensation + thermal depositions	0.7	UV-light, magnetic field	PE, PP, PS, PET, PTFE	Phoretic interactions	/	[25]
2021	α -Fe ₂ O ₃ /Pt-Pd	Hydrothermal + sputtering	3-4	UV-light, magnetic field	PEG	Electrostatic interactions	Photo-Fenton reaction	[58]
2021	BiVO ₄ /Fe ₃ O ₄	Hydrothermal	4-8	Visible-light, magnetic field	PLA, PCL, PET, PP	Adsorption/precipitation	Photocatalysis	[59]
2021	PDA@Fe ₃ O ₄ /Lipase	Self-polymerization + lipase functionalization	/	Rotating magnetic field	Plastic centrifuge tube, PCL	Chemical adhesion	Enzymatic	[60]

4.1. Capture Strategies

Electrostatic interactions are promising to capture nano/microplastics. Nano/microrobots with tunable surface charge are required for this strategy. Their charge status must be programmed to be opposite to that of nano/microplastics. In this way, the attractive force between the nano/microrobots and nano/microplastics will lead to a firm contact. Certainly, the possibility to modify the charge of the robots at will is highly desirable for two reasons: 1) nano/microplastics are various; the same contaminated water sample could contain a mixture of positively and negatively charged plastics; 2) despite the ultimate goal should be their definitive elimination, the release of undegraded plastics may be wanted; this could be

obtained by switching the nano/microrobots' charge status, transforming the attraction into repulsion. Ideally, the nano/microrobots' charge should be externally and easily controlled. However, this surface charge reversibility is usually obtained by modifying the pH of the solution in which nano/microrobots and nano/microplastics are immersed. Some semiconductors (for example, TiO_2) are negatively charged at neutral-basic pH values and positively charged at acidic pH values (Section 4.2.1.).^[61] This behavior has also been observed for some polymers, among which polyethyleneimine (PEI), and used to fabricate self-propelled microrobots to capture, transport, and release nucleic acids.^[62] Nevertheless, compared to photocatalytic semiconductors, it does not offer the advantage of simultaneous degradation capability, necessitating the loading of other materials for this purpose. A main drawback of the pH-mediated capture and release approach is that controlling the pH of the solution can be challenging, even in confined spaces. Alternatively, the charge status can be determined by the material's design. On this subject, it is worth mentioning that static iron oxides nanoparticles have been functionalized with a monolayer of octadecylphosphonic acid (PAC_{18}) or (12-Dodecylphosphonic acid)-N,N-dimethyl-N-octadecyl ammonium bromide ($\text{PAC}_{12}\text{-NC}_{18}$), having, respectively, negative and positive surface charge values.^[63] A mixture of these particles could allow the simultaneous capture of all nano/microplastics in one step without any pH adjustment. Moreover, owing to their magnetic nature, they could be actuated exploiting rotating magnetic fields, permitting nano/microplastics capture in locations difficult to access, and finally magnetically collected.

Light-powered metal-capped semiconducting microrobots can collect microplastics due to attractive phoretic interactions, whose intensity depends on the chemical fuel. These are based on hydrodynamic effects such as the fluid flow around the microrobot due to the gradient of products generated by photochemical reactions. Particularly, it has been demonstrated that Au@Ni@TiO_2 microrobots can induce the formation of clusters of polystyrene (PS) particles and transport them.^[25]

As mentioned in the introduction, nano/microplastics act as platforms for accumulating other toxic substances in water, including heavy metal ions and organic pollutants, *via* an adsorption/precipitation mechanism. The same mechanism can promote the interaction between nano/microrobots and nano/microplastics and, accordingly, their degradation. In this regard, an example will be provided in Section 4.2.2.

Marine mussels stick on the surface of rocks, corals, and marine plants to stand against the strong sea waves. Their sticking ability is based on the protein 3,4-dihydroxyphenyl-L-alanine (DOPA) secreted from their feet. DOPA and the molecule dopamine have similar chemical

structures and, so, adhesive properties. This similarity has inspired the development of polydopamine (PDA)-based nano/microrobots applied from biomedical to environmental applications, including microplastics capture and degradation (Section 4.2.3.).^[64]

4.2. Degradation Strategies

4.2.1. Photo-Fenton Degradation of Polymer Chains

The electrostatic capture and subsequent degradation of polymer chains by self-propelled microrobots were demonstrated (**Figure 3(a)**).^[58] Although no microplastic is involved in this work, it provides direct proof of microrobots' capability to break solid chemical bonds as those present in polymeric and plastic materials. For this purpose, Janus microrobots were fabricated by asymmetrically depositing thin (~30 nm) metal layers (Au, Au-Pd, Pt, Pt-Pd) on α -Fe₂O₃ (hematite) microspheres (3-4 μ m in size) prepared by a scalable hydrothermal process. Microrobots showed fuel-free self-electrophoretic motion under UV-light irradiation (as schematically illustrated in Figure 2(a)) and magnetic field-controlled navigation (Figure 3(b)). Indeed, hematite is a visible light photocatalytic semiconductor and possesses a weak intrinsic magnetism.^[43,65] Consequently, it is a valuable component for designing microrobots because it harbors both photocatalytic and magnetic properties. Furthermore, in the presence of H₂O₂ and light irradiation, it catalyzes the photo-Fenton reaction, greatly enhancing the production of ROS involved in plastics degradation.^[66] The photo-Fenton mechanism is explained by the following reactions:^[67]



or



Briefly, electron-hole pairs are generated in hematite under light irradiation. Photogenerated electrons can be trapped by H₂O₂, creating OH• radicals, or surface Fe³⁺, leading to Fe²⁺ and then OH• upon reaction with H₂O₂. In this context, a recent work illustrates how vital it is in

water purification applications to balance properly H_2O_2 consumption for microrobots propulsion and photo-Fenton reaction.^[68] This is the only case in which the input energy is subtracted for the degradation in favor of the motion. Still, this drawback can be avoided by adjusting the metal layer thickness to ~30 nm, as in the case of the hematite/Pt-Pd microrobots.

Hematite/Pt-Pd microrobots exhibited the highest speed in fuel-free water and increasing concentrations of H_2O_2 , attributed to the largest electrochemical potential difference between the two components. Thus, they were tested to degrade polymers in 1% H_2O_2 under UV-light irradiation exploiting the photo-Fenton reaction. High molecular weight polyethylene glycol (PEG 4000) was selected as a model for polymer waste since its degradation can be easily studied by MALDI-MS. In addition, despite this polymer being widely used in cosmetics, its harmfulness at higher molecular weights is controversial.^[69] To favor PEG capture by microrobots, their electrostatic attraction has been intensified. The pH-dependence of microrobots Zeta potential was explored, indicating the acidic environment (pH 3) as the optimal one, being microrobots positively charged and PEG negatively charged (Figure 3(c)). Under these conditions, MALDI-MS spectra proved microrobots ability to break PEG 4000 chains into short oligomers within 24 h (Figure 3(d)). Although this method only qualitatively assessed the polymer degradation, it was noted that the broad distribution around m/z 4000 was disrupted into low-mass products ($m/z < 1000$), indicating the almost complete polymer degradation. This result was attributed to the synergistic combination of microrobots' self-propulsion, programmable and enhanced electrostatic interaction, and the photo-Fenton reaction, identified as the dominant degradation mechanism.

It is worth noting that H_2O_2 use in environmental applications is typically not recommended due to its toxicity. Nonetheless, in this case, it was shown that microrobots completely consumed H_2O_2 at the end of the process. So, it does not represent a limitation of the proposed strategy, differently from the obligatory use of a noble metal coating to enable microrobots motion. In fact, this heavily affects microrobots fabrication cost, and its corrosion during microrobots movement may be responsible for secondary pollution.

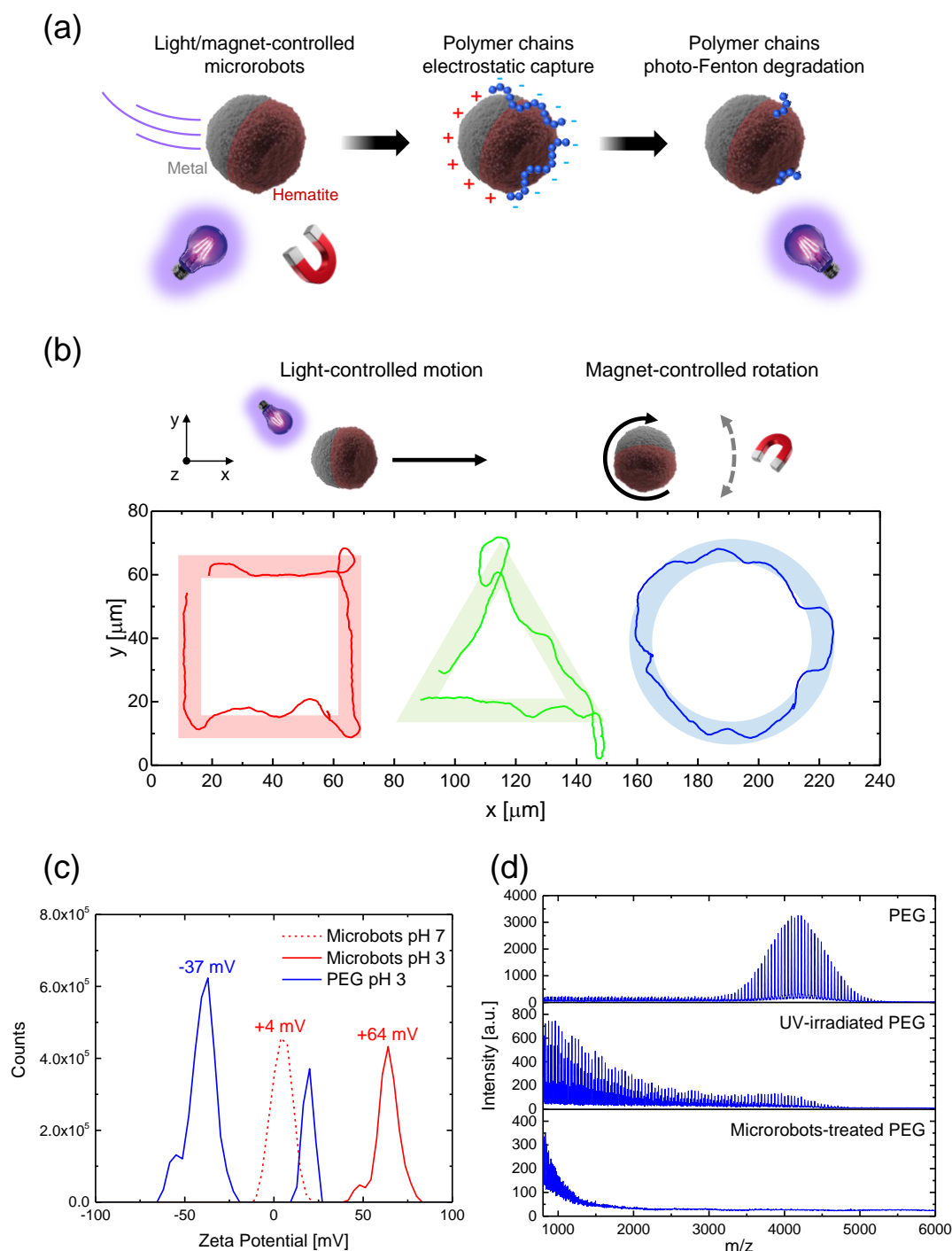


Figure 3. Electrostatic capture and photo-Fenton degradation of polymer chains. (a) Schematic illustration of the capture and degradation mechanism by self-propelled light-powered magnetic field-navigable hematite/Pt-Pd Janus microrobots. (b) Microrobots' motion resulting in square, triangular and circular trajectories in 1% H_2O_2 under UV-light irradiation using a magnetic field. (c) MALDI-MS spectra of untreated (PEG) and treated PEG 4000 under 24 h UV-light irradiation in 1% H_2O_2 in the absence (UV-irradiated PEG) and presence of microrobots (Microrobots-treated PEG). Adapted with permission.^[58] Copyright 2021, Wiley.

4.2.2. Photocatalytic Degradation of Microplastics in Confined Spaces

An important proof-of-concept study on the capture and photocatalytic degradation of microplastics in a confined space using smart microrobots was reported.^[59] These consisted of noble metal-free Fe_3O_4 nanoparticles-embedded star-shaped BiVO_4 microparticles (4-8 μm in size) prepared by a large-scale hydrothermal method (**Figure 4(a)**). BiVO_4 is a largely used visible light-activated photocatalyst.^[70] Due to their intrinsic asymmetrical geometry, microrobots were capable of autonomous motion in water under visible light irradiation in the presence of a small amount of H_2O_2 (0.1%). This result constitutes a considerable advance compared to the conventional light-driven metal/semiconductor Janus microrobots, which require expensive noble metals to unlock the self-propulsion ability. Additionally, $\text{BiVO}_4/\text{Fe}_3\text{O}_4$ microrobots could be easily navigated using external magnetic fields and collected through permanent magnets thanks to the incorporated magnetic nanoparticles. Such dual-movement was employed to attack various microplastics, including polylactic acid (PLA), polycaprolactone (PCL), polyethylene terephthalate (PET), and polypropylene (PP), in macroscale channels (**Figure 4(b)**). Specifically, it was shown that, when introduced to one end of the channel, these microrobots reached microplastics pieces due to their light-induced active movement. Microrobots attachment on the microplastics' surface was explained by the adsorption/precipitation mechanism introduced in Section 4.1. Their adhesion to the plastic pieces was strong enough that most microrobots were not detached even after 2 h intense shaking. By placing a magnet on the opposite end of the channel, they moved towards and cleaned it. The removal efficiency was higher for hydrophilic microplastics (~70% for PLA and PCL) than hydrophobic ones (~40% for PET and ~20% for PP) and decreased with the length of the channel. Capture and transport of PLA microplastics were verified in a system of 5 interconnected channels of variable sizes, proving their excellent performance in a complex maze of plastic waste.

Besides, the photocatalytic degradation of captured microplastics was investigated. Photocatalysis is a promising approach for the definitive elimination of microplastics since it necessitates only a photocatalyst, light, and water.^[71-73] It is based on ROS generation due to the reaction between photogenerated electron-hole pairs in the semiconductor and water, which oxidize and dissociate pollutants until they are mineralized into CO_2 and water.^[74] For light-powered microrobots, the ROS formation occurs simultaneously to microrobots motion. Upon exposure to microrobots under visible light irradiation in 0.01% H_2O_2 , microplastics

gradually lost weight. After 7 days, a maximum weight loss of 2-3% was observed for PCL and PLA (Figure 4(c)). The deterioration of microplastics' surface properties was detected. In particular, they exhibited enhanced hydrophilicity through contact angle measurements, surface oxidation as attested by XPS, and increased roughness as evidenced by SEM analysis (Figure 4(d)). SEC and liquid chromatography-high-resolution mass spectrometry (LC-HRMS) revealed the presence of oligomers and polymeric moieties in the treated microplastics solutions.

These findings unambiguously confirmed that the self-propelled microrobots partially degraded the big microplastic pieces. The degradation efficiency was relatively poor for all targeted microplastics due to the declining photocatalytic activity of BiVO_4 with time. The worse results for PET and PP compared to PCL and PLA are ascribed to their hydrophobicity. Therefore, more stable and efficient photocatalytic microrobots have to be developed to degrade real microplastics conclusively.

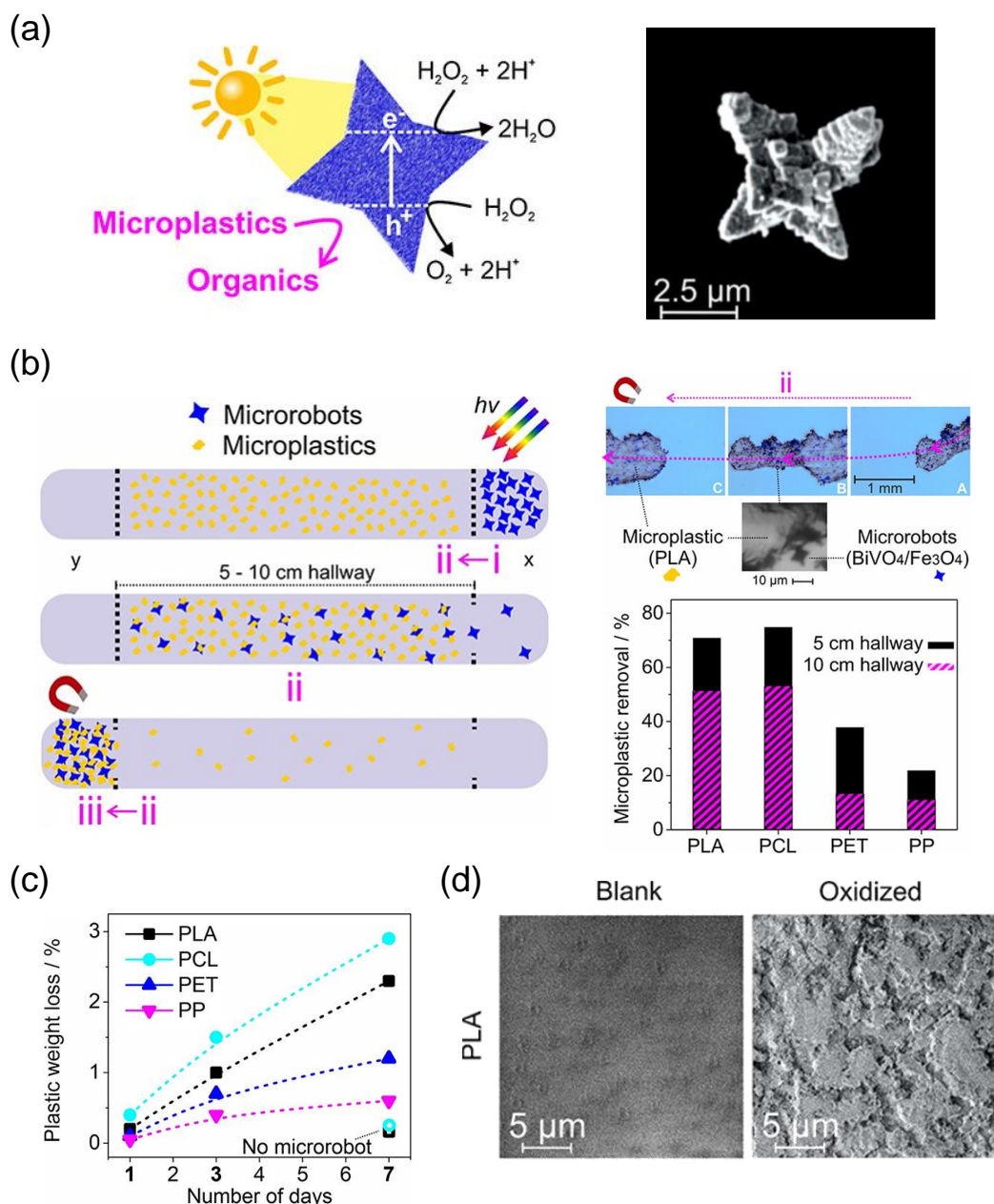


Figure 4. Photocatalytic degradation of microplastics in confined spaces. (a) Schematic illustration of the motion mechanism of $\text{BiVO}_4/\text{Fe}_3\text{O}_4$ microrobots for microplastics photocatalytic degradation and SEM image of a microrobot. (b) Microplastics collection in a homemade channel (5 or 10 cm in length, 0.5 cm in depth, 1 cm in width) filled with 0.1% H_2O_2 under visible light irradiation: (i-ii) movement and adsorption of microrobots on the microplastics' surface, as shown for a PLA piece by optical and SEM images, and (ii-iii) microrobots-microplastics collection using a magnet. Columns indicate microplastic removal efficiency for PLA, PCL, PET, and PP in 5 cm - (black) and 10 cm -long channels (magenta). (c) Microplastics' weight loss after the treatment with microrobots in 0.01% H_2O_2 under visible light irradiation. (d) SEM image of a PLA microplastic before and after the

photocatalytic degradation. Adapted with permission.^[59] Copyright 2021, American Chemical Society.

4.2.3. Enzymatic Degradation of Microplastics

A completely different approach consisted of magnetic field-powered mussel-inspired microrobots that chemically adhered to microplastics and enzymatically degraded them (**Figure 5(a)**).^[60] Mussel-like magnetic microrobots (MagRobots) were developed by coating magnetic Fe₃O₄ nanoparticles with a PDA layer through a simple, biocompatible, and low-cost self-polymerization process. PDA@Fe₃O₄ MagRobots were further functionalized with the enzyme lipase to induce microplastics enzymatic degradation by cleaving polymer chains. PDA@Fe₃O₄/Lipase MagRobots showed a “near-surface walking” motion under a conical rotating magnetic field generated by homemade triaxial coils, as illustrated in Figure 2(b). This allowed their precise navigation towards suspended microplastics (Figure 5(b)). Once MagRobots approached microplastic pieces, they firmly adhered to their surface and transported them independently from their size due to PDA strong adhesive force (Section 4.1.). Indeed, large microplastic fragments (>50 μm) were removed thanks to the coordinated action of several MagRobots. Afterward, MagRobots and captured microplastics were transferred and incubated for 24 h in a second vessel to perform their enzymatic digestion. Optical microscopy demonstrated the partial degradation of a PCL microplastic through evident structural variations (Figure 5(c)).

Enzymatic degradation is considered a safe, cost-effective, and reliable solution for the plastic pollution issue.^[75–77] The proposed strategy based on the combination of microplastics capture, collection, and separate digestion, permits better managing the problem of secondary pollution due to toxic byproducts. In fact, the latter would be eventually produced in a controlled environment, i.e., the vessel where the enzymatic degradation is conducted. Moreover, this mussel-inspired capture is more advantageous than the pH-triggered electrostatic interactions since it does not require pH adjustments. Instead, the main drawback is represented by the applicability of the magnetic setup used to power microrobots action in open water bodies like rivers, lakes, and seas.

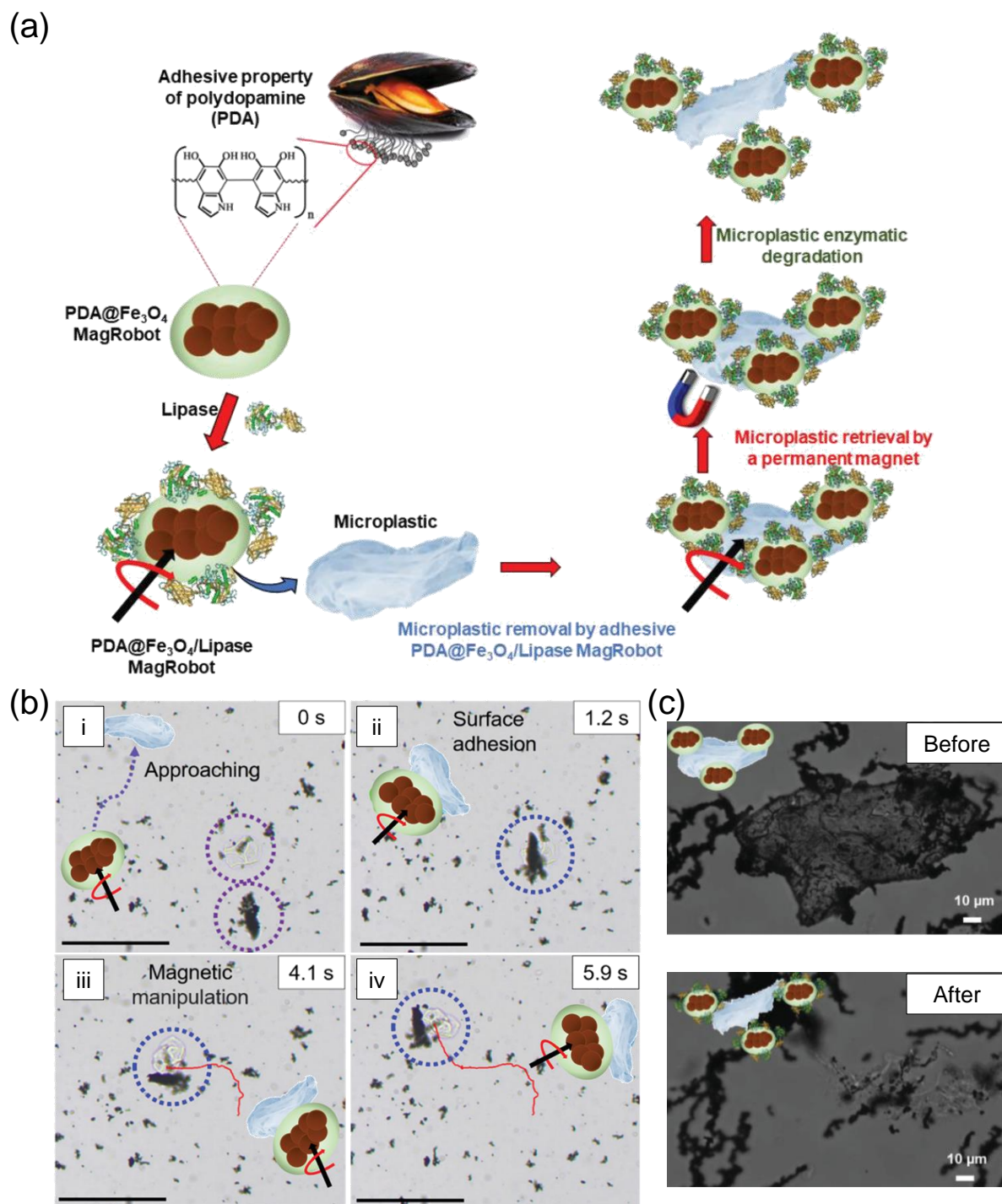


Figure 5. Chemical adhesion and enzymatic degradation of microplastics. (a) Schematic illustration of microplastics removal and enzymatic degradation by mussel-inspired adhesive PDA@Fe₃O₄/Lipase MagRobots. (b) Time-lapse images of a MagRobot approaching (i), adhering to (ii), and manipulating (iii-iv) a microplastic under a conical rotating magnetic field (scale bars are 50 μm). (c) Optical images of a PCL microplastic before and after overnight incubation with MagRobots. Adapted with permission.^[60] Copyright 2021, Wiley.

5. Future Challenges

Autonomous motile microrobots have been demonstrated to be effective tools for the degradation of plastic waste, from polymer chains to microplastics. However, there are several challenges to be faced towards the practical application of this technology:

- Light source and H₂O₂: to fully exploit the potential of light-driven nano/microrobots, visible light should be preferred to UV-light for their actuation since the former represents the biggest portion of the solar irradiance spectrum. H₂O₂ should be avoided unless mandatory, like in the photo-Fenton reaction. In this case, potential residues in the treated water have to be considered and managed appropriately. Furthermore, it is itself responsible for ROS production under light irradiation, which could make it hard to discriminate the effective contribution of microrobots' action in proof-of-concept studies;
- Reducing nano/microrobots components and cost: multifunctionality in nano/microrobots is usually achieved by combining several components with different properties (for example, photocatalytic semiconductors, metal layers, magnetic nanoparticles). As a result, the cost and complexity of nano/microrobots fabrication increase, limiting their commercial value. In this regard, the development of metal-free nano/microrobots through intrinsically asymmetric multifunctional components or semiconducting heterojunctions is promising;
- Large-scale fabrication: to treat large volumes of polluted water (m³), a massive quantity of nano/microrobots is necessary. Hence, large-scale preparation processes are required. Compared to physical deposition methods or templated-assisted electrochemical deposition, chemical syntheses, like hydrothermal, are very appealing. Nevertheless, high control over their parameters must be attained to ensure particles reproducibility in morphological properties (size, shape), crystallinity, and stability. 3D printing represents a valid approach towards low-cost, large-scale, and standardized manufacture of microrobots;^[78]
- Sedimentation: most reports on microrobots for water purification, including those on microplastics removal and degradation discussed in the previous section, deal with microparticles whose driving force is not strong enough to overcome the gravitation force exerted onto them. Consequently, they lie down at the bottom of the vessel. This problem may affect the speed and efficacy of the remediation process, in particular against suspended matter such as nanoplastics. Microrobots designed to move in the

3D space with six degrees-of-freedom and colloidal nanorobots able to swim in the bulk of the medium are practical solutions;^[79]

- Improving the degradation efficiency: photocatalysis, photo-Fenton reaction, and enzymatic degradation are promising ways to remove microplastics ultimately. Nonetheless, they rely on the accomplishment of microplastics mineralization before the photocatalytic semiconductor or immobilized enzyme loses its activity, which is far from reality. Thus, the degradation process must be accelerated by exploring better photocatalysts and enzymes or combining different mechanisms (for instance, photocatalytic and enzymatic degradation);
- Additional parameters influencing the degradation efficiency: so far, the influence of some critical parameters on the degradation efficiency, such as the irradiation time until total mineralization of microplastics, light source intensity, microrobots and microplastics concentrations, has not been investigated. In addition, only models for microplastics have been degraded. This is explained by the necessity of studying standards whose degradation can be easily monitored by experimental techniques. Still, real samples are much more difficult to degrade due to various stabilizers in the matrix (for example, UV stabilizers).^[80] The interference due to a mixture of pollutants, like persistent organic pollutants, heavy metals, and microorganisms, in the medium and on the surface of microplastics has not been considered yet. Light-powered photocatalytic microrobots with antibacterial properties may be helpful in this regard.^[81] Besides, the viscosity and salinity of water, affecting microrobots motility, have not been examined. Investigating these parameters is imperative for nano/microrobots devised to operate in oceans. Finally, microrobots reusability and performance deterioration (motility, photoactivity, etc.) have been rarely studied;
- Applicability in open water bodies: light-driven nano/microrobots are preferred over magnetic ones owing to their simpler actuation. The use of magnetic nano/microrobots on a large scale would need vast and expensive magnetic systems. In spite of this, magnetic properties in light-powered nano/microrobots are generally integrated to permit their collectability at the end of treatment. However, it is worth noting that the intensity and penetration of light in natural water bodies are reduced by obstacles (for example, aquatic plants) and depth;
- Secondary pollution: microrobots could release harmful substances in water because of their corrosion during utilization. For this reason, the development of nano/microrobots with low environmental impact is essential. Moreover, the

degradation of nano/microplastics could potentially liberate in the environment more dangerous compounds if not completely mineralized. These phenomena cause unwanted secondary pollution. Careful studies of plastic degradation byproducts have to be conducted by analyzing the treated waters through mass spectrometry techniques. Apropos, most of the strategies presented in this review foresee first a microplastics capture step, followed by their magnetic collection and transfer into a second vessel where the degradation is carried out. This allows to limit and control potential secondary pollution;

- Nanoplastics: nanoplastic pollution, the ultimate product of plastic waste fragmentation in the environment, is an emerging global issue due to their tiny size, which increases their hazard and the difficulty of removing them. Nano/microrobots have never been tested against nanoplastics. We expect that novel designs will soon be developed to trap and entirely degrade these elusive plastic nanoparticles, learning from the progress in the remediation of their bigger counterparts.

6. Conclusions and Future Outlook

Recent advances and future challenges on nano/microplastics capture and degradation by self-propelled nano/microrobots have been critically reviewed. Presently, only microrobots have been explored for this application. Their motility was triggered mainly by light irradiation. Magnetic properties were highly desired, especially for collecting microrobots together with captured plastics. All strategies were based on the synergy between microrobots active motion and programmed anchoring ability (electrostatic interactions, adsorption, and chemical adhesion) to “on-the-fly” capture polymer chains and microplastics rapidly. These were then transferred into a second vessel where the degradation was performed according to different mechanisms, such as photo-Fenton reaction, photocatalysis, and enzymatic degradation, avoiding secondary pollution resulting from toxic byproducts. The future challenges towards the practical application of autonomous nano/microrobots for the definitive elimination of nano/microplastics were exhaustively described. These relate to improving the degradation efficiency and the applicability of nano/microrobots on a large scale and reducing the environmental impact and costs associated with this technology. Possible solutions to overcome current limitations were also proposed. Indeed, the considerations in this review are of general validity and can be extended to other types of water contaminants. In the near

future, we expect the development of innovative nano/microrobot designs that induce nano/microplastics total mineralization due to the combined effect of multiple mechanisms.

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Nano/microrobots hold great promise for managing the increasing plastic pollution owing to their self-propulsion and programmable operation. In this perspective, future directions in this research field have been discussed after critically reviewing the very recent advances in the capture and degradation of nano/microplastics.

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Nano/Microplastics Capture and Degradation by Autonomous Nano/Microrobots: A Perspective

