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Review

# Sustainable Magnetic Nanorobots for Microplastics Remediation

Sarah Briceño <sup>1,\*</sup> , José Eduardo Arevalo-Fester <sup>2,3</sup>  and Ivan Andres Fierro-Sanchez <sup>1</sup> 

<sup>1</sup> School of Physical Sciences and Nanotechnology, Yachay Tech University, 100119 Urcuquí, Ecuador; ivan.fierro@yachaytech.edu.ec

<sup>2</sup> Division of Energy and Materials, Department of Chemistry and Chemical Engineering, Chalmers University of Technology, SE-412 96 Gothenburg, Sweden; jose.arevalo@chalmers.se

<sup>3</sup> Coordinación de Aprovechamiento de Residuos y Subproductos Industriales, Instituto Zuliano de Investigaciones Tecnológicas (INZIT), Km 15, 4033 La Cañada de Urdaneta, Estado Zulia, Venezuela

\* Correspondence: sarahbriara@gmail.com

## Abstract

Microplastics have become a major environmental concern due to their resistance to degradation, wide distribution, and potential uptake by organisms. Conventional mitigation strategies often exhibit limitations in efficiency, reuse, and scalability, and may generate secondary pollutants. In this review, we highlight the application of magnetically controlled, sustainable nanorobots based on magnetic hybrid nanoparticles with different functional groups to enhance the removal efficiency of microplastics from the environment. By leveraging hydrophobic interactions, surface modifications, and tailored additives, these magnetic nanorobots provide a sustainable, eco-friendly approach to mitigating microplastic pollution and offer improved magnetic separation performance. Bioinspired and biohybrid magnetic nanorobots, based on green synthesis principles, carbon-based nanomaterials, biochar, nature-inspired swarm motion, and collective behavior, present further advancements that mimic biological systems to capture microplastics with high efficiency and recovery. Achieving removal efficiencies often exceeding 90% in minutes, and maintaining the efficiency after several cycles. The synergistic integration of magnetic separability with tailor-made surface functionalities underpins the effectiveness of these magnetic nanorobots, setting the stage for their future commercialization and widespread adoption in water remediation technologies.

**Keywords:** nanoplastics; microplastics; magnetic nanoparticles; nanorobots; biochar



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## 1. Introduction

Nanoplastics and microplastics have emerged as pervasive contaminants in surface waters, wastewater effluents, sediments, and biological systems [1], and pose a significant threat to human health due to their slow degradation, high toxicity [2], and the potential to interact with organic pollutants, which can lead to the formation of even more hazardous substances [3]. Their widespread distribution has raised substantial concerns regarding trophic transfer and their role as vectors for polymer-associated additives and adsorbed environmental pollutants.

Traditional separation techniques, such as sedimentation [4] and granular filtration [5], exhibit limited effectiveness in dilute aqueous environments, particularly for particles smaller than 100 µm. The low effectiveness of these removal techniques is mainly observed when applied to wastewater, since this aqueous medium exhibits properties that can hinder removal, such as pH, organic materials, metals, and sedimentation, among others [6]. As

particle sizes enter the nanometer-to-micrometer range, a distinct separation gap becomes evident, making conventional removal methods ineffective [7]. In response, magnetic separation technologies, particularly magnetic-seeded filtration [8] and nanoengineered magnetic sorbents [9], have garnered attention as viable solutions to bridge this gap, offering high removal efficiencies under low-pressure conditions [10].

In response to these challenges, an emerging solution is the application of magnetic nanorobots, which are based on magnetic nanoparticles and serve as autonomous systems capable of actively seeking, capturing, degrading, and transporting microplastic particles [11]. These systems utilize a combination of the magnetic properties and advanced propulsion mechanisms to overcome the diffusion limitations inherent in passive remediation techniques [12]. Magnetic nanoparticles are often regarded as nanorobots because they exhibit the essential characteristics of nanoscale robotics: navigability, task execution, and remote control [13]. These magnetic nanoparticles can perform controlled, purposeful tasks at the nanoscale, acting like nanorobots when directed by an external magnetic field or functionalized with specific molecules [14]. Their movement can be modulated with a collective behavior by adjusting the strength, direction, or frequency of the magnetic field, enabling them to navigate through liquids, congregate in targeted areas, or move in specific patterns [15].

The efficiency of magnetic nanorobots for removing microplastics primarily derives from their ability to selectively adsorb plastic microparticles using bioinspired modalities, and subsequent recovery using external magnetic fields [16]. The adsorption mechanisms of the nanorobots are affected by several interface-related interactions, including hydrophobic effects [17], van der Waals forces [18], electrostatic attractions [19], hydrogen bonding [20], and, in carbon-based materials,  $\pi$ - $\pi$  interactions [21]. Magnetic separation is then achieved by applying an external magnetic field that induces the alignment and aggregation of the nanoparticle–plastic complexes into larger clusters, which can be easily extracted from the aqueous phase [22]. This method offers several advantages over traditional filtration and coagulation techniques, including rapid separation, inherent recyclability, reduced energy consumption, and ease of recovering the nanomaterials for subsequent reuse, which enables multiple consecutive cycles of microplastics removal without significant loss of performance, reducing secondary waste and lowering operational costs [23].

The utilization of waste-derived adsorbents has emerged as a sustainable and cost-effective strategy for removing pollutants, aligning with principles of the circular economy. Innovative approaches to mitigating the environmental impact of microplastics and nanoplastics are increasingly focusing on sustainable magnetic nanorobots as adsorbents for wastewater removal. Unlike conventional treatment methods, natural adsorbents are derived from renewable and biodegradable sources via eco-friendly processes, thereby minimizing secondary pollution [24]. These materials—such as biochar [25], carbon-based nanomaterials [26], and plant-based adsorbents [27]—possess unique surface properties that enhance microplastic adsorption efficiency. Given their abundance, low cost, and potential for performance optimization through modifications, they represent promising candidates for pilot-scale applications in water treatment facilities [28].

This review critically evaluates the use of magnetic nanorobots for removing microplastics, with an emphasis on developing sustainable nanomaterials based on green chemistry principles, hybrid magnetic nanomaterials, carbon-based nanomaterials, and complex biohybrid systems that utilize collective behavior and living organisms. Sustainability constitutes one of the primary advantages of modified magnetic nanorobots. In contrast to non-biodegradable nanorobots, these eco-friendly alternatives are derived from abundant natural resources and can be engineered for safe post-use degradation into biochar, algae, liquidbots, hydrogels, and other biomaterials, such as human hair waste.

Their low production costs, coupled with minimal environmental impact, pose a greater risk to public health than primary particles, due to their size and the toxicity they impart. Research gaps in the use of sustainable magnetic nanorobots are presented.

## 2. Microplastics and Nanoplastics

Microplastics are particles smaller than 5 mm in diameter but larger than 1 micrometer ( $\mu\text{m}$ ), and nanoplastics are plastic particles smaller than 1 micrometer ( $\mu\text{m}$ ), often considered in the 1–1000 nanometer (nm) range, that result from the breakdown of larger plastic, have emerged as pollutants from their persistence over time, environmental stability, ability to fragment, and potential to transport other pollutants [16]. Nanoplastics and microplastics are categorized based on their origin as primary and secondary. Primary source refers to plastics intentionally manufactured in small sizes for direct or indirect use as raw materials in products. Secondary sources are fragments of larger plastics that have degraded due to physical, chemical, or biological processes in the environment [29]. These secondary particles present a greater risk to public health compared to primary particles due to the size effect and the toxicity that affects both health and the environment.

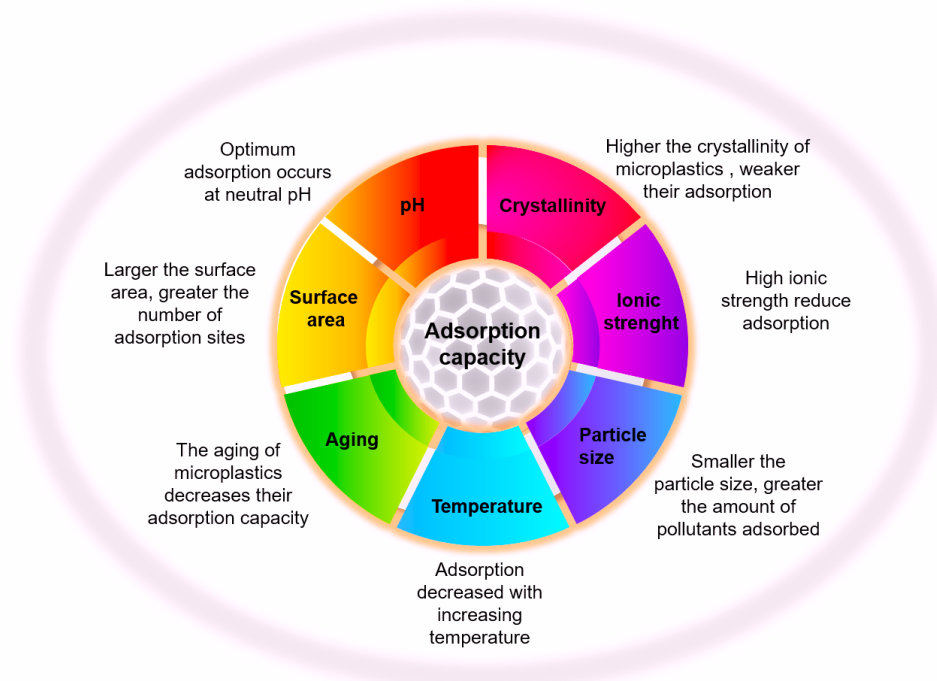
Nanoplastics and microplastics have a significant impact on human health [30], causing inflammation in the respiratory and gastrointestinal systems [31], compromising immune function [32], and increasing the risk of cardiovascular diseases [33] and neurotoxicity [34]. These effects are primarily attributed to the role of the nanoplastics and microplastics in disrupting hormonal regulation [35], which can lead to reproductive disorders [36] and an elevated risk of cancer [37].

The most commonly used polymers in plastic manufacturing are polypropylene (PP), polyethylene (PE), polyethylene terephthalate (PET), polyamide (PA), polystyrene (PS), and polymethyl methacrylate (PMMA), among others. As a result, these polymers are also the most frequently detected in the environment [38]. Apart from traditional plastic sources of microplastics, in recent years the proliferation of biodegradable plastics has emerged as an environmentally friendly solution. However, when left in natural environments, they can generate microplastics and even increase greenhouse gas emissions, especially if not processed in industrial settings [39]. Many of these bioplastics only degrade under controlled conditions. In an open environment, they can fragment more quickly, generating even more microplastics than regular plastics [40]. Moreover, additives such as Oxodegradable plastics exacerbate the issue by producing persistent nanoplastics [41].

In recent years, numerous technologies have been developed to remove nanoplastics from water environments, which can be categorized into physical, chemical, and biological technologies based on their removal principles [42]. Physical methods include filtration [43], coagulation [44], reverse osmosis [45], flocculation [46], and magnetic separation [47]. Chemical methods involve oxidation processes [48], such as electrochemical oxidation [49], electrocoagulation [50], photocatalytic degradation [51], and adsorption techniques [34]. Biological methods utilize microorganisms or enzymes to degrade microplastics [52].

The removal of microplastics and nanoplastics depends on several factors, such as pH, ionic strength, valency, concentration, charge density of interfering ions, and the presence of natural organic matter (Figure 1), all of which can influence the adsorption interactions between magnetic nanoparticles and microplastics [53]. For example, higher salt concentrations in marine environments promote the aggregation of microplastics, enhancing their adsorption efficiency. The presence of ions alters water's polarity and solvent environment, influencing the charge state and the hydration layer structure at the surface of microplastics. With increasing ionic strength, the polarity of water molecules diminishes, thereby weakening the interaction force between water molecules and microplastics [34]. At the

same time, excessive natural organic matter may compete for active sites on nanoparticle surfaces, slightly reducing efficiency [54].



**Figure 1.** Factors that influence microplastics and nanoplastics adsorption efficiency: pH, crystallinity, ionic strength, particle size, temperature, aging, and surface area.

The environmental pH significantly impacts the protonation state of functional groups on both adsorbents and adsorbates, leading to variations in their chemical properties, charge distributions, and ionic strengths [9]. Experimental findings suggest that both excessively acidic and alkaline conditions hinder the adsorption of microplastics due to their charged nature. At solution pH levels that are equal to, above, or below the isoelectric point of the microplastics, the surface charge of the microplastics can undergo protonation and deprotonation, resulting in neutral, negative, or positive surface charges, respectively. When both nanorobots and microplastics carry the same charge, electrostatic repulsion significantly impedes the adsorption process, leading to a substantial reduction in adsorption efficiency [34].

In general, the smaller the particle size and the larger the specific surface area of the nanorobots, the greater their number of adsorption sites and the amount of microplastics they can adsorb [55]. In the environment, microplastics undergo changes in their surface structure and oxygen-containing functional groups due to external environmental factors, including ultraviolet radiation, water, corrosion, and temperature. These factors also affect the mechanism by which the nanorobots absorb the microplastics [56]. During aging, some of the microplastics' chemical bonds are oxidized. The resultant oxygen-containing functional groups increase the hydrophilicity of the microplastics or strengthen the hydrogen bonds between the nanorobots and the microplastics, thereby enhancing the adsorption capacity of hydrophilic microplastics and decreasing the adsorption capacity for hydrophobic microplastics [57]. Temperature can significantly affect the adsorption capability of the nanorobots. At low-temperature conditions, the adsorption capacity of the nanorobots increases. In contrast, as the temperature increases, the adsorption capacity decreases due to a reduction in surface tension and van der Waals forces between the microplastics and the nanorobots [56].

### 3. Magnetic Nanoparticles

A magnetic nanoparticle is one with a size between 1 and 100 nm, in which the particles form single magnetic domains. When they are below a threshold of <20 nm, their properties become size-dependent [58]; magnetic nanoparticles are usually composed of iron, iron oxides (magnetite, maghemite), cobalt, and various ferrites [59]. The synthesis of magnetic nanoparticles can be achieved via multiple approaches, including co-precipitation [60], solvothermal [59], and thermal decomposition methods [61], often augmented by green chemistry principles that utilize non-toxic solvents and ambient conditions [62]. The morphology and size of the magnetic nanoparticles depend directly on the synthesis process, which includes temperature, pH, type of solvent, reaction time, and the presence of surfactants or stabilizers [60]. Therefore, they can take on various shapes, such as spherical, cubic, octahedral, disk-like, or rod-like, depending on the synthesis process. However, they are most commonly spherical, often consisting of a magnetic core and a functional outer layer [63]. The nanometric size of magnetic particles is important because it gives rise to unique phenomena such as superparamagnetism, quantum tunneling of magnetization, and enhanced magnetic coercivity [64,65].

#### 3.1. Magnetic Properties of Nanoparticles

At the nanoscale, magnetic nanoparticles behave differently from bulk ferromagnets because the energy required to maintain magnetic domains becomes comparable to thermal energy [66]. In single-domain nanoparticles, all magnetic spins are aligned coherently, leading to high magnetic anisotropy and potentially high coercivity, although a reduction in size below a critical threshold results in superparamagnetism [67,68]. Superparamagnetism is a key characteristic of magnetic nanoparticles, particularly for sizes below a critical diameter, typically a few tens of nanometers. Superparamagnetic nanomaterials exhibit zero coercivity and zero remanence [69]. Superparamagnetic behavior is particularly desirable for removing microplastics, as it prevents aggregation in the absence of a magnetic field and facilitates the recyclability of the material. For example, superparamagnetic iron oxide nanoparticles (SPIONs) typically exhibit saturation magnetization values ranging from 48.3 emu/g to 62 emu/g, making them highly responsive to magnetic manipulation [70,71]. These magnetic properties are a function of both the intrinsic material composition, such as magnetite ( $\text{Fe}_3\text{O}_4$ ) [72] or maghemite ( $\gamma\text{-Fe}_2\text{O}_3$ ) [73], the interparticle interactions, the shape, and the particle size, which is often in the range of 9–45 nm [66]. Detailed characterization using techniques such as SQUID magnetometry and vibrating sample magnetometry (VSM) confirms the superparamagnetic properties and the magnetic saturation of these nanoparticles, reinforcing their suitability for microplastics remediation applications.

#### 3.2. Functionalized Magnetic Nanoparticles

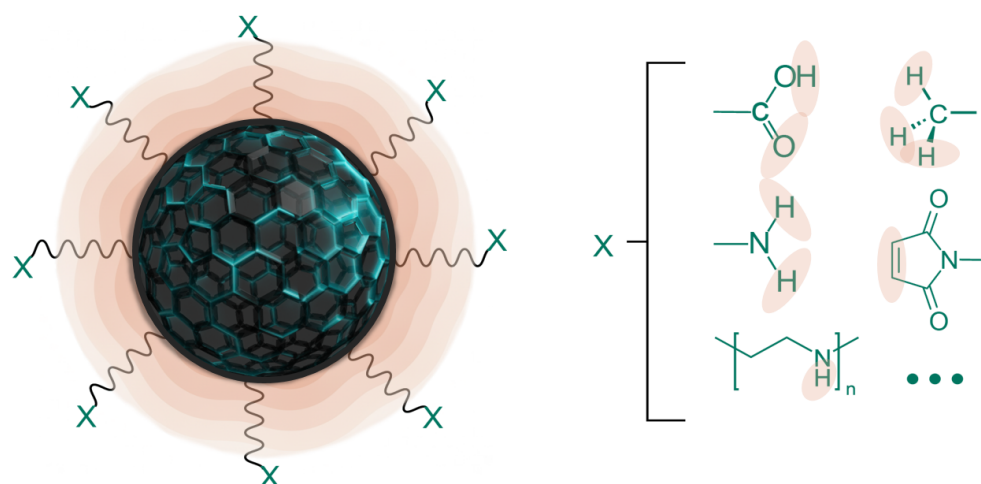
Magnetic nanoparticles have attracted growing interest due to various studies exploring their use for the rapid and low-cost removal of microplastics and nanoplastics, which are now found in nearly all ecosystems and living organisms [74]. This approach leverages the hydrophobic nature of the polymers most commonly used in plastic manufacturing [75]. The separation of nanoplastics using magnetic nanoparticles is based on two simple steps: the magnetic nanoparticles adhere to the surface of the nanoplastics, coating them; subsequently, the magnetized aggregate is attracted by an external magnetic field [76].

The interaction of magnetic nanoparticles with microplastics and nanoplastics is explained by magnetic dipole-dipole, van der Waals attractions, and hydrophobic attractions, which outweigh electrostatic repulsion [34]. In contrast, the surface organization of the magnetic nanoparticles results from balancing electrostatic repulsion forces and magnetic

attraction forces among particles [77]. However, when addressed in a general manner, it has been proposed that the EDLVO theory and the interfacial energy minimization theory can explain the adhesion mechanism. Reference [1], which describes the stability of colloidal dispersion through the superposition of van der Waals forces and double-layer forces. Reference [78], and hydrophobic interactions [1], respectively. These hydrophobic properties are enhanced by modifying the surface of the magnetic nanoparticles through the use of “functionalizers.” As a result, magnetic nanoparticles can better encapsulate and remove the microplastics and nanoplastics [76].

#### 4. Hybrid Magnetic Nanomaterials

When the surface of magnetic nanoparticles is modified with coatings (Figure 2 and Table 1), they can effectively bind hydrophobic microplastics via hydrophobic and van der Waals interactions [1]. This interaction facilitates the aggregation of microplastics onto magnetic nanoparticles, enabling their magnetic removal [79]. Once microplastics are bound to the functionalized magnetic nanoparticles, the resulting aggregates can be quickly and cleanly separated from water using magnetic fields, providing a rapid, energy-efficient method superior to filtration or sedimentation alone [80].



**Figure 2.** Surface modification of magnetic nanorobots with different surface groups (Carboxyl–COOH, amino –NH<sub>2</sub>, hydroxyl –OH, imidazole, polymers, etc.).

Magnetic Fe<sub>3</sub>O<sub>4</sub> nanoparticles functionalized with citric acid (Fe<sub>3</sub>O<sub>4</sub>@AC) were used to remove high-density polyethylene (HDPE), low-density polyethylene (LDPE), and polypropylene (PP) microplastics from an aqueous medium, achieving an 80% microplastic removal at a pH of 6, through hydrogen bonding, pore filling, and van der Waals forces that governed the adsorption mechanism. Reusability tests demonstrated that Fe<sub>3</sub>O<sub>4</sub>@AC could be reused up to five times, with a removal efficiency exceeding 50% [81].

Magnetic nanoparticles with hydrophobic fluorescent dyes and photoluminescence techniques are used for real-time monitoring of nanoplastic concentrations. This method integrates simple detection techniques with advanced robotic capture, providing a dual function that enhances both the quantification and remediation processes [82]. Velikov et al. (2024) [83] report the use of magnetite nanoparticles as magnetic nanorobots to capture and remove nanoplastics from aqueous environments, achieving 90% removal of nanoplastics from an aqueous solution within 120 min. This work demonstrates that easy-to-use, common fluorescent dyes, combined with photoluminescence spectroscopy methods, can serve as an alternative approach for detecting and quantifying nanoplastics

in water environments, and that swarming magnetic nanorobots can be used for efficient capture and removal.

Magnetic nanoparticles functionalized with Tannic acid (TA-Fe<sub>3</sub>O<sub>4</sub>) were prepared using the coprecipitation method for Polystyrene (PS) and polyethylene terephthalate (PET) microplastic removal. The maximum adsorption efficiency was 83% by 98% respectively, at a dosage of 2.5 g/L and an initial microplastic concentration of 1.25 g/L. The experiment was conducted in a medium with a pH of 6–7, and the contact time between the MNPs and the microplastics was 300 min. It is mentioned that the stable binding of TA-Fe<sub>3</sub>O<sub>4</sub> and the magnetic nanoparticles was due to electrostatic interaction, hydrophobic interaction,  $\pi$ - $\pi$  interaction, or hydrogen bonding. The functionalized nanoparticles were recycled five times without a significant decrease in removal efficiency [72].

Superhydrophobic magnetic cobalt ferrite nanoparticles coated with Lauric acid were used for highly efficient oil–water separation and fast microplastic Removal, reporting a microplastic separation efficiency higher than 99.6%. Due to the synergistic effect of the hydrophobic effect and van der Waals force, the functionalized magnetic cobalt ferrite had a high and stable microplastic removal efficiency. After ten microplastic removal cycles, the removal efficiency exceeded 98% [1]. Hydrophobically functionalized magnetic nanoparticles with Hexadecyltrimethoxysilane (HDTMS) (HDTMS-FeNPs) were used for polystyrene nanoplastics (PAN-Pd@NPs) removal, obtaining a removal efficiency of 84.9% [84].

Another interesting article was conducted by Vicente-Martínez et al. (2024). In this study, three magnetic materials were evaluated: iron oxide (Fe<sub>3</sub>O<sub>4</sub>), Fe<sub>3</sub>O<sub>4</sub>@Ag, and Fe<sub>3</sub>O<sub>4</sub>@Ag@L-Cysteine for the removal of polystyrene (PS). Comparative results show that Fe<sub>3</sub>O<sub>4</sub>@Ag@L-Cysteine MNPs are the best option, as 50  $\mu$ L of these achieved 100% removal of NPs at a concentration of 50 mg/L. This was achieved in 15 min at neutral pH. It is mentioned that the process is physisorption. It is worth noting that the other two magnetic materials also achieved 100% removal, but required higher MNP concentrations and longer times [80].

In the study by Zhang et al. (2023), magnetic nanoparticles (Fe<sub>3</sub>O<sub>4</sub>) functionalized with polyethylene glycol (PEG) were synthesized via coprecipitation for the removal of polyethylene (PE) microspheres. The functionalized MNPs achieved a maximum adsorption capacity of 2203 mg/g; moreover, they ranged in size from 20  $\mu$ m to 2163 mg/g at 5 °C. Neutral pH favored the removal of microplastics. The article proposes that the adsorption process was mainly due to intermolecular hydrogen bonding. It is worth noting that as the amount of MNPs increased, the adsorption rate also increased; however, for amounts greater than 2.0 g/L, the adsorption rate did not increase significantly [85].

Zandieh, M., & Liu, J. (2022) proposed the use of Fe<sub>3</sub>O<sub>4</sub> MNPs synthesized using the coprecipitation method using five types of plastics, which were prepared by mechanical grinding: high-density polyethylene (HDPE), polypropylene (PP), polyvinyl chloride (PVC), polystyrene (PS), and polyethylene terephthalate (PET). The microplastics had sizes ranging between 20  $\mu$ m and 800  $\mu$ m, while the MNPs had an average size of  $\approx$ 10 nm. In the experiments, a concentration of 60  $\mu$ g/mL of MNPs was used to remove 4 mg/mL of microplastics over 5 min. The removal efficiency for each type of plastic was  $\approx$ 100%; however, the removal was not simultaneous but followed this decreasing order of time: PET, PS, HDPE, PVC, and PP. It is mentioned that the magnetic nanoparticles benefited from the peroxidase-like activity of the exposed surface, allowing additional catalytic degradation [86].

Pasanen et al. (2023) synthesized magnetic nanoparticles by aqueous synthesis at room temperature of a nanocomposite “Zeolitic Imidazolate Framework-8 modified with iron” (Fe@ZIF-8). The MNPs were functionalized with n-butylamine to enhance their

hydrophobic properties and to remove Polystyrene (PS) microspheres with a diameter of 1.1  $\mu\text{m}$ . The size of the MNPs was approximately  $50 \pm 30$  nm. It was found that with a concentration of 20 mg and 30 mg of Fe@ZIF-8, a removal of  $98.0 \pm 0.2\%$  was achieved at a PS concentration of 25 mg/L, while for a concentration of 50 mg/L, a removal of  $88.7 \pm 1.5\%$  was obtained. The MNPs and microplastics were combined for 5 minutes under stirring at 700 rpm in Mili-Q water at neutral pH [87].

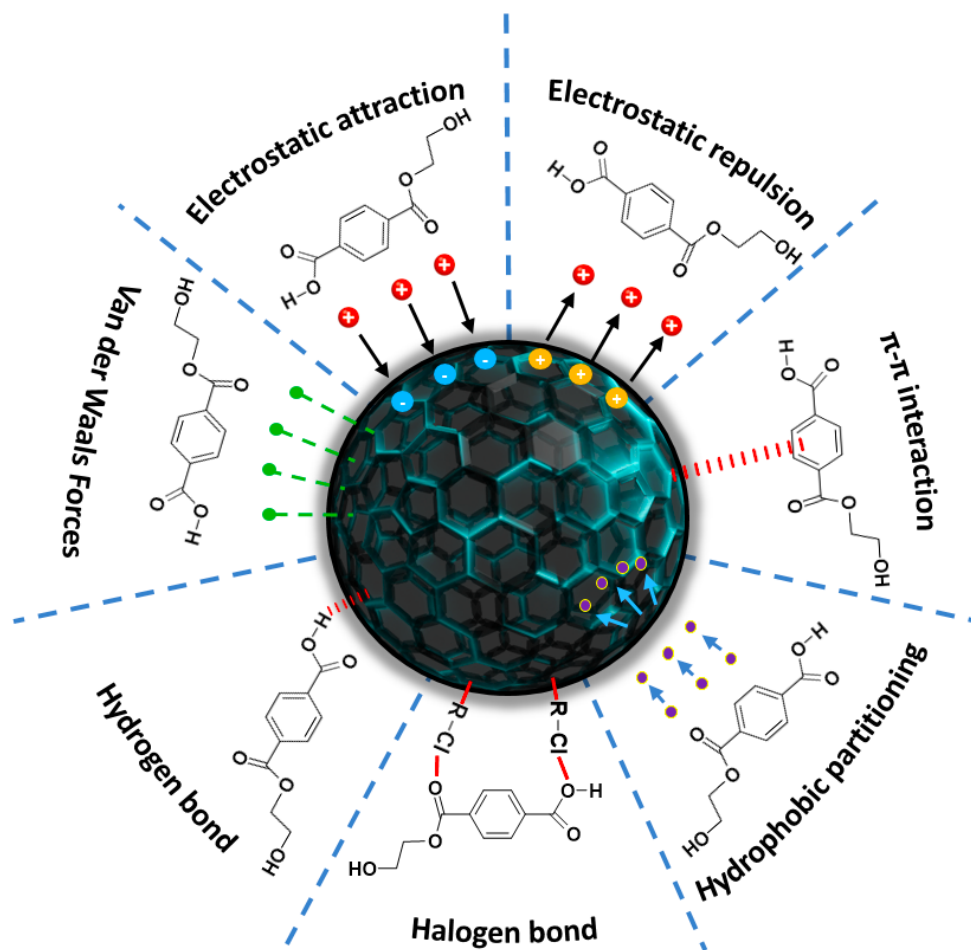
Bui, N. T., & Tran, T. D. (2022) reported the removal capacity of microplastics using a ferrofluid. Magnetic nanoparticles of cobalt ferrite ( $\text{CoFe}_2\text{O}_4$ ) were synthesized and functionalized using Sodium dodecyl sulfate (SDS). The nanoparticles were synthesized using the coprecipitation method, and subsequently the ferrofluid was prepared by mixing the MNPs with SDS in the following ratios: 1 : 4, 1 : 5, and 1 : 6; the  $\text{CoFe}_2\text{O}_4$  MNPs had a size between 15 and 20 nm. On the other hand, Polyvinyl chloride (PVC) microplastics were used, obtained by grinding PVC pipes and sieved using 50- and 100-mesh screens. The experiment was carried out simulating common domestic wastewater contaminants at neutral pH; the ferrofluid and microplastics were combined for 30 min at 90 °C with a ferrofluid concentration of 25 mg/L. It was found that the maximum removal percentage for MNPs-OH/SDS was 69.3%. For the ferrofluid without functionalization, it was 53.35%. Using vegetable oil as an additive (25 mg/L of ferrofluid with 100 ml/L of vegetable oil), the removal percentage was 51.2% [88].

Aragón et al. (2025) studied the use of magnetic nanoparticles ( $\text{Fe}_3\text{O}_4$ ) synthesized and functionalized with amino groups using TMPED (N-[3-(trimethoxysilyl)propyl]ethylenediamine) and with sodium alginate (SA) to remove polyethylene (PE) microspheres. The SA-functionalized MNPs were synthesized using the coprecipitation method, while the amino-functionalized nanoparticles were synthesized via the thermal decomposition method. The MNPs synthesized by coprecipitation had a size of  $8.9 \pm 1.2$  nm, and those synthesized by thermal decomposition had a size of  $11.7 \pm 1.3$  nm; moreover, the polyethylene microspheres had a size of  $232 \pm 8$   $\mu\text{m}$ . In the experiment, an MP concentration of 0.5 g/L was used, while the MNP concentration was 1.3 g/L, with a contact time of 20 min under stirring at 180 rpm, at room temperature, and at an optimal pH of  $5.2 \pm 0.3$ . It was found that the removal percentage for the SA-functionalized MNPs was  $82.4 \pm 0.3\%$ , while the amino-functionalized MNPs achieved a removal of  $75.5 \pm 2.3\%$ . The article mentions that the removal process is a consequence of the interactions between the functionalized nanoparticles with amino or carboxyl groups, as their surface charge interacts with that of the microplastics; additionally, van der Waals forces and hydrogen bonds between the functional groups –OH and –COOH allow the adhesion of the MNPs to the microplastics [89].

Microplastics exhibit hydrophobicity, allowing for strong interactions with hydrophobically modified magnetic nanoparticles [89]. These magnetic nanoparticles enhance adsorption through hydrophobic interactions, facilitating the aggregation of microplastics on their surface [79]. Electrostatic interactions also play a role, where oppositely charged microplastics and magnetic materials enhance adhesion. Once microplastics are magnetized through particle attachment or direct coating, an external magnetic field facilitates their separation from water [90]. This mechanism enhances the removal efficiency of microplastics while facilitating material recovery for potential reuse. For instance, hydrophobic iron nanoparticles have been developed to magnetize plastics using direct binding to the hydrophobic surfaces of microplastics, enabling their magnetic recovery [90].

The adsorption mechanism between microplastics and the surface of magnetic nanoparticles involves electrostatic and van der Waals forces [91], hydrogen bonding [10], hydrophobic interactions,  $\pi$ - $\pi$  conjugation [91], and other forces (Figure 3). Hydrogen bonding, recognized as a specific intermolecular force with a strength intermediate between

covalent bonds and van der Waals forces, stands as a primary mechanism governing the interactions of microplastics [34].



**Figure 3.** Summary of microplastics adsorption mechanisms OF Polyethylene terephthalate: Electrostatic and van der Waals forces [91], hydrogen bonding [10], hydrophobic interactions, and  $\pi$ - $\pi$  conjugation [91]

**Table 1.** Comparison of magnetic carbon nanomaterials for microplastics and nanoplastics removal.

Material Type	Removal Efficiency	Significant Impact
Hydrophobic Fe nanoparticles [92]	Recovered 92% of 10–20 $\mu\text{m}$ polyethylene and polystyrene beads and 93% of polyethylene, polyethylene terephthalate, polystyrene, polyurethane, polyvinyl chloride, and polypropylene from seawater.	Magnetic extraction of microplastics from environmental samples.
Magnetic Nano- $\text{Fe}_3\text{O}_4$ [54]	PE 86.9%; PP 85.1%; PS 86.1%; PET 62.8%.	Surface adsorption + magnet capture; validated in river/sewage/seawater.
$\text{Fe}_3\text{O}_4$ nanoparticles (FNPs) [93]	Removal 83.1–92.9%.	Charge neutralization; hetero-aggregation; pH < 6.7 best; salinity helps at alkaline.

Table 1. Cont.

Material Type	Removal Efficiency	Significant Impact
Fe <sub>3</sub> O <sub>4</sub> (Bare MNPs) [86]	HDPE, PP, PVC, PS, PET → ≈100% removal (order: PET > PS > HDPE > PVC > PP).	Fast removal in 5 min. The peroxidase-like activity of the exposed surface aids catalytic degradation.
Fe <sub>3</sub> O <sub>4</sub> nanoparticles with fluorescent dyes [83]	90% removal of nanoplastics within 120 min.	Fluorescent dyes combined with photoluminescence spectroscopy as an alternative approach for detecting and quantifying nanoplastics in water.
PEG-modified Fe <sub>3</sub> O <sub>4</sub> nanoparticles [85]	Maximum adsorption of 2203 mg/g for polyethylene microspheres.	These systems operate primarily through intermolecular hydrogen bonding mechanisms.
Ultra-thin Fe <sub>3</sub> O <sub>4</sub> Nanodiscs [94]	Adsorption capacity of 188.4 mg/g.	The removal mechanism involves both electrostatic and magnetic forces originating from the vortex domain of the nanodiscs.
Fe <sub>3</sub> O <sub>4</sub> @PDA [95]	Removal efficiencies up to 98.5%.	The PDA coating enhances adhesion through hydrogen bonding, $\pi - \pi$ stacking, and hydrophobic interactions.
Imine-functionalized mesoporous magnetic silica nanoparticles [96]	96% removal efficiency for microplastics loaded with organic pollutants.	Design of experiments and machine learning were used for multi-objective optimization in magnetic separation.
Magnetic sepiolite [54]	One cycle removal time is 600 s, and removal efficiency is 98.4%.	Magnetic carrier medium can be effectively recycled with the magnetic tube.
Fe <sub>3</sub> O <sub>4</sub> @Citric Acid [81]	HDPE, LDPE, PP → 80% removal at pH 6; reusable up to 5 cycles (>50% efficiency).	Removal is governed by hydrogen bonding, pore filling, and van der Waals forces. Reusability confirmed.
TA-Fe <sub>3</sub> O <sub>4</sub> [72]	PS (83%), PET (98%) at pH 6–7, 300 min.	High efficiency due to electrostatic, hydrophobic, $\pi-\pi$ , and H-bonding interactions. Maintained efficiency after 5 cycles.
Cobalt ferrite@Lauric Acid [1]	>99.6% microplastics removed; >98% after 10 cycles.	Synergistic effect of hydrophobicity and van der Waals forces. Excellent stability and recyclability.
HDTMS-FeNPs [84]	Polystyrene nanoplastics → 84.9%.	Hydrophobic functionalization enhanced PS adsorption.
Fe <sub>3</sub> O <sub>4</sub> @Ag@L-Cysteine [80]	PS → 100% removal (50 mg/L microplastics in 15 min, neutral pH)	Most efficient among the compared MNPs. Physisorption mechanism. Lower concentrations and shorter times are required.
Fe@ZIF-8@n-butylamine [87]	PS microspheres → 98% (25 mg/L microplastics), 88.7% (50 mg/L microplastics).	Hydrophobic functionalization enhanced adsorption. Effective at neutral pH in only 5 min under stirring.

Table 1. Cont.

Material Type	Removal Efficiency	Significant Impact
CoFe <sub>2</sub> O <sub>4</sub> @SDS ferrofluid [88]	PVC → 69.3% (functionalized), 53.35% (bare MNPs).	Performance improved with SDS functionalization. Efficiency is affected by wastewater contaminants. Oil addition reduced performance.
Fe <sub>3</sub> O <sub>4</sub> @Sodium alginate (SA)/Fe <sub>3</sub> O <sub>4</sub> @Amino (TMPED) [89]	PE microspheres: SA-MNPs → 82.4%, Amino-MNPs → 75.5%.	Removal favored at slightly acidic pH (5.2). Hydrogen bonding and van der Waals interactions between surface functional groups and microplastics enhanced adsorption.
Magnetic Coal gangue [97]	Adsorption capacity for polystyrene microplastics of 35.9 mg/g.	Coal gangue is a solid waste byproduct from coal mining and washing

## 5. Magnetic Carbon Nanomaterials

Magnetic carbon nanomaterials (MCNMs) are hybrid composites that incorporate ferri-magnetic iron oxide phases Fe<sub>3</sub>O<sub>4</sub> and/or  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> into diverse carbonaceous frameworks, such as biochar [98], 3D graphene-like carbons [99], carbon nanotubes [26], and carbon aerogels [100] (Figure 4 and Table 2). This synergistic integration combines field-responsive magnetization with high surface area and hydrophobic,  $\pi$ -electron-rich domains, promoting favorable interactions with polymeric microplastics. In magnetic-seeded filtration, magnetite particles heteroagglomerate with microplastics and are subsequently extracted via an external magnetic field, achieving removal efficiencies exceeding 95%. These efficiencies can be further enhanced by modulating ionic strength and particle surface charge [8].

Building upon these attributes, ongoing research has focused on optimizing the synthesis pathways of magnetic carbon nanomaterials to enhance structural uniformity, magnetic responsiveness, and pollutant selectivity for microplastics removal [101]. Strategies such as templated pyrolysis [25], solvothermal integration [47], and surface grafting [1] have enabled fine-tuning of pore architecture and functional group distribution, thereby improving adsorption kinetics and magnetic recovery rates. Moreover, the incorporation of redox-active moieties and catalytic centers within the carbon matrix has expanded the functional scope of magnetic carbon nanomaterials, allowing for the simultaneous capture and degradation of microplastic pollutants [102]. These advancements underscore the potential of magnetic carbon nanomaterials as both passive sorbents and dynamic platforms for integrated water purification technologies.

The versatility and efficacy of magnetic carbon nanomaterials across diverse water matrices and polymer types demonstrated compatibility with real-world conditions, ranging from beverage systems to sludge extracts, validating their robustness and operational relevance [103]. Importantly, the ability to tune surface chemistry, magnetic properties, and porosity enables magnetic carbon nanomaterials to be engineered for specific remediation targets, including co-contaminants such as microplastics and heavy metals [104]. From a systems perspective, magnetic carbon nanomaterials offer a modular platform for integration into existing water treatment infrastructures. Their magnetic responsiveness enables low-energy separation, while catalytic functionalities open up avenues for in situ degradation, thereby reducing downstream waste management burdens [105].

Ye Tang et al. (2021) [26] report the removal of polyethylene (PE), polyethylene terephthalate (PET), and polyamide (PA) microplastics from aqueous solutions using magnetic carbon nanotubes, achieving complete removal within 300 min. Bare Fe<sub>3</sub>O<sub>4</sub> nanoparticles magnetize and remove PE/PP/PS/PET in laboratory and environmental waters. The

removal mechanism is governed by aggregation-controlled kinetics and modulated by pH/salinity [54,93]. Extended performance was demonstrated by functionalized  $\text{Fe}_3\text{O}_4$  containing  $-\text{OH}/-\text{COOH}/-\text{NH}_2$  groups which enable co-removal of nutrients with favorable life-cycle implications [46], while PEG- $\text{Fe}_3\text{O}_4$  shows very high capacities for PE removal [85]. Wang et al. (2023) [106] used an eco-friendly and effective method to produce superhydrophobic fatty acid-grafted adsorbents  $\text{Fe}_3\text{O}_4$  ( $\text{Fe}_3\text{O}_4@\text{C}_{12} - \text{C}_{18}$ ) using a liquid phase deposition method capable of capturing 100 nm PS in high capacity and functions in real beverages, underscoring practical robustness.

In an innovative article, Xia et al. (2025) synthesized magnetic iron nano-oxides functionalized with biochar (MFe@BC) via thermal impregnation–pyrolysis to evaluate their capacity for removing microplastics of poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV), a biodegradable thermoplastic, from simulated wastewater. MFe@BC achieved an adsorption capacity of 13.14 mg/g and an efficient removal rate of 98.53% at a dosage of 1.5 g/L. The MP removal efficiency remained above 95% at a pH between 4 and 9 and a  $\text{COD}_0$  of 500 mg/L, indicating that the material was effective under conditions of high organic pollution. Furthermore, the MNPs were reused four times, maintaining a removal efficiency of >92% for PHBV. It is important to note that the approach is both innovative and significant, as under environmental conditions, the mineralization of biodegradable plastics is incomplete, making them a rapid source of MP generation and leading to rapid accumulation of microplastics in ecosystems [107].

Singh et al. (2021) explored the concept of functionalizing nanoplastics rather than MNPs. Iron oxide nanoparticles without functionalization were synthesized and incorporated into biochar using the in situ solvothermal method at two temperatures: 550 °C and 850 °C; the magnetic nanoparticles had a size of  $\approx 200$  nm. In the experiment, polystyrene (PS) nanospheres with three different functional groups were used: carboxylated and aminated, with sizes of 1000 nm, 1000 nm, and 30 nm, respectively. The MNPs and NPs were in contact for 30 min at room temperature (25 °C), at a pH of 5.5 and 200 rpm, with an NP concentration of 10 mg/L. For the carboxylated NPs, FB-850 was used and a removal of 100% or 225.11 mg/g was achieved; for the aminated NPs, FB-550 was used and a removal of 100% or 290.2 mg/g was achieved; for the other carboxylated NPs, FB-850 was used and a removal of 100% or 206.46 mg/g was achieved [108].

Wang et al. (2021) [98] highlight the development of magnetized biochars, specifically Mg/Zn-modified and chitosan-grafted variants, as highly effective adsorbents for microplastic remediation. These materials demonstrated exceptional removal efficiencies, achieving approximately 95–99% elimination of polystyrene (PS) microspheres from aqueous solutions. The performance of these adsorbents was evaluated using 1  $\mu\text{m}$  PS microspheres at a concentration of 100 mg/mL. Magnetic biochar (MBC) achieved a removal efficiency of 94.81%, while Mg and Zn-modified variants reached even higher efficiencies of 98.75% and 99.46%, respectively. The underlying adsorption mechanism is attributed to a combination of electrostatic interactions and chemical bonding between the microplastic particles and the biochar surfaces. However, the presence of competing ions and organic matter in real water samples, such as  $\text{H}_2\text{PO}_4$ , was found to reduce the performance of Zn-Mstrong affinity for various microplastics, forming stable M-CNT/MP composites that could be easily separated from solution using a magnetic field for degradation and adsorbent regeneration. This process not only eliminated the captured microplastics via catalytic degradation facilitated by the Mg- and Zn-active sites but also restored adsorption capacity by freeing up adsorption sites.

Recent studies have introduced advanced magnetic carbon-based nanomaterials as highly effective tools for the remediation of microplastics in aquatic environments. Two notable approaches involve magnetic carbon nanotubes (M-CNTs) [26] and cellulose-derived

materials [109], both of which offer promising performance and recyclability. For the first time, magnetic carbon nanotubes (M-CNTs) were synthesized by Tang et al. (2021) [26] and evaluated as adsorbents for removing microplastics, including polyethylene (PE), polyethylene terephthalate (PET), and polyamide (PA), from water. These M-CNTs displayed a strong affinity toward various microplastics, forming stable M-CNT/MP composites that could be easily separated from the solution using a magnetic force. At an optimal concentration of 5 g/L, complete removal of microplastics (5 g/L) was achieved within 300 min. The maximum adsorption capacities for PE, PET, and PA reached 1650, 1400, and 1100 mg of M-CNTs per gram of microplastics, respectively. Notably, this adsorption process demonstrated high robustness, maintaining effectiveness even in the presence of common water contaminants such as chemical oxygen demand (COD), ammonia nitrogen  $\text{NH}_3\text{-N}$ , and phosphate  $\text{PO}_4^{3-}$ . When tested at a real wastewater treatment plant that reduces waste, the M-CNTs successfully removed all detected microplastics. Moreover, the M-CNTs were thermally regenerated at 600 °C, retaining both their magnetic properties and MP removal capacity. After four regeneration cycles, they still achieved ~80% removal efficiency, highlighting their potential for repeated use and scalability in practical applications.

Building on biomass-derived materials, Liu and Wang (2023) [109] developed cellulose benzoate adsorbents through the esterification of cellulose in an ionic liquid (AmimCl), as confirmed by FTIR, XRD, and solid-state NMR analyses. When evaluated for polystyrene (PS) microplastic removal, the modified cellulose demonstrated a substantial improvement in efficiency, increasing from 9.0% for raw cellulose to 68.3% for cellulose benzoate. This enhancement was attributed to the aromatic ring structure of the benzoate groups, which facilitated  $\pi$ - $\pi$  interactions with PS, as well as to an increased zeta potential, leading to stronger electrostatic attraction.

Further modification with carbon nanotubes (CNTs) and magnetic carbon nanotubes (M-CNTs) elevated the PS removal efficiency to over 97%. This improvement was attributed to better particle dispersion and higher surface reactivity, resulting from increased zeta potential and a reduction in particle size. Kinetic and isotherm models confirmed that M-CNT-modified cellulose benzoate exhibited superior adsorption rates and capacities compared to other variants, making it a high-performance, sustainable material for MP capture.

Both magnetic carbon nanotubes and functionalized cellulose derivatives are powerful, recyclable platforms for removing microplastics. While M-CNTs demonstrate rapid, high-capacity adsorption with magnetic recovery even in real wastewater contexts, cellulose benzoate-based adsorbents offer a bio-based, scalable solution that can be tuned for enhanced efficiency through nanomaterial integration. Together, these innovations open new avenues for tackling microplastic pollution through advanced material science and sustainable engineering.

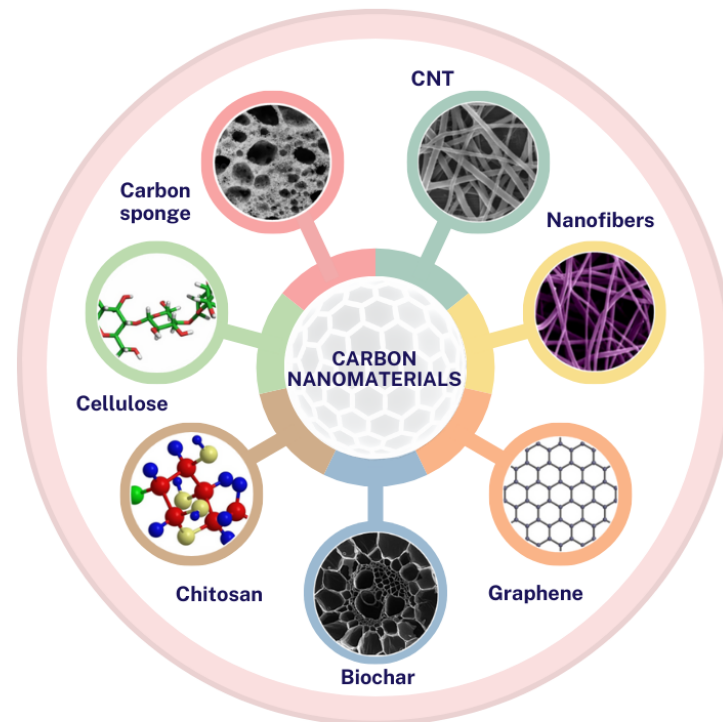
To address the pressing demand for technologies capable of both capturing and degrading microplastics (microplastics), recent studies have introduced iron-oxide nanoflowers (NFs) as a multifunctional material with dual capabilities: adsorption and mineralization. These multicore, flower-like magnetic nanoparticles, approximately 40 nm in diameter, offer a scalable, reproducible approach for removing and decomposing microplastics in aqueous environments, particularly those derived from cosmetic sources [110].

The NFs exhibit pronounced cooperative magnetic behavior and a high magnetic moment per particle, facilitating effective magnetic separation following MP adsorption. Under ultrasonic treatment, the nanoflowers readily adhere to polyethylene microplastic surfaces, achieving removal efficiencies of up to 1000 mg of microplastics per gram of NFs under optimal conditions (pH 7, 10 mg NFs, 30 min, magnetic field strength of

320 kA/m). These outcomes were obtained via gram-scale synthesis in a 1-L reactor, with 91% reproducibility in structural, colloidal, and magnetic characteristics—an essential factor for industrial scalability.

A key innovation of this method lies in its integrated degradation mechanism. After physical separation, microplastics suspended in water were hydrolyzed at 150 °C to cleave polymer chains, followed by Fenton-like oxidation catalyzed by the NFs. In this process, hydrogen peroxide reacts with the iron-based nanocatalyst to generate reactive oxygen species, which oxidize and mineralize the fragmented polymers. Mineralization efficiencies ranged from 20% at 25 °C to 75% at 90 °C. Notably, applying an alternating magnetic field (60 mT, 100 kHz) induced localized nanoheating around the NFs, enabling 78% mineralization without heating the entire water volume, thereby substantially improving energy efficiency. This approach not only removes microplastics but also converts them into benign mineral byproducts, offering a robust solution to the challenge of emerging contaminants. The scalability, reproducibility, and multifunctionality of iron-oxide nanoflowers underscore their potential as a promising platform for environmental remediation and public health advancements.

The co-occurrence of microplastics and heavy metals in aquatic systems presents a significant challenge for water purification technologies. To address this, Ullah et al. (2024) [111] developed a series of multifunctional nanocomposites capable of simultaneously removing both contaminants. These materials consist of polyoxometalate-based ionic liquids (POM-ILs) integrated with mesoporous silica-coated magnetite nanoparticles, forming composites denoted as Q8[Mo<sub>54</sub>]@SiO<sub>2</sub>@Fe<sub>3</sub>O<sub>4</sub>, Q10[Mo<sub>54</sub>]@SiO<sub>2</sub>@Fe<sub>3</sub>O<sub>4</sub>, Q8[Mo<sub>46</sub>]@SiO<sub>2</sub>@Fe<sub>3</sub>O<sub>4</sub>, and Q10[Mo<sub>46</sub>]@SiO<sub>2</sub>@Fe<sub>3</sub>O<sub>4</sub>. These materials were synthesized by combining large molybdenum oxide clusters ([Mo<sub>46</sub>]<sup>21-</sup> and [Mo<sub>54</sub>]<sup>26-</sup>) with tetraalkylammonium cations, followed by immobilization onto SiO<sub>2</sub>@Fe<sub>3</sub>O<sub>4</sub> cores. A comprehensive suite of analytical techniques, including FTIR, XRD, SEM, TEM, TGA, DSC, CV, ICP-AES, DLS, and GC-MS, was used to characterize the physicochemical properties of the composites. The results confirmed an amorphous surface structure, large specific surface areas, and strong adsorption capabilities. The performance evaluations demonstrated remarkable heavy metal removal efficiencies, ranging from 79.5% to 99.3% in synthetic laboratory water and from 92% to 99% in industrial wastewater samples. In parallel, dynamic light scattering measurements demonstrated 100% removal of polyvinyl chloride (PVC) microbeads in laboratory water and 95–98% removal in industrial effluents. These results emphasize the versatility and high effectiveness of these materials under complex water chemistry. Moreover, the composites exhibited excellent recyclability, maintaining their structural and functional integrity over at least five treatment cycles. The magnetic Fe<sub>3</sub>O<sub>4</sub> core facilitated easy separation and recovery, supporting the potential scalability of this approach in industrial applications. This study highlights the potential of magnetic composite nanomaterials as integrated solutions for multipollutant remediation, offering a promising platform for the simultaneous removal of emerging contaminants from water systems.



**Figure 4.** Carbon nanomaterials for microplastics removal: carbon nanotubes (CNTs) [26], nanofibers [46], graphene [112], biochar [98], chitosan [15], cellulose [109], and carbon sponges [113].

Magnetic carbon nanomaterials (MCNMs) offer a promising approach to overcoming three key limitations in the remediation of microplastics. First, they enable high-throughput processing, as magnetically assisted separation operates effectively even under low-pressure conditions, facilitating treatment of large fluid volumes [114]. Second, magnetic carbon nanomaterials exhibit strong selectivity, maintaining their performance in complex media such as sludge extracts and beverage samples, demonstrating their versatility across diverse environmental matrices [115]. Third, post-remediation management is greatly simplified through magnetic recovery and catalytic breakdown, minimizing secondary waste and enhancing downstream efficiency [102]. However, several hurdles remain before MCNMs can be widely adopted. Ensuring the environmental safety of residual nanomaterials is essential to avoid unintended ecological consequences [116]. Scalable and reproducible synthesis methods are also needed to support consistent performance in industrial applications [117]. Moreover, the long-term resilience of MCNMs under variable field conditions, including fluctuations in pH, salinity, and organic content, must be thoroughly assessed [118]. To support the practical deployment of these technologies, future research should prioritize techno-economic evaluations, life-cycle impact studies, and alignment with relevant regulatory frameworks. The convergence of nanotechnology, environmental science, and process engineering in MCNM development holds significant potential for advancing sustainable strategies to combat microplastic pollution [119].

Table 2 summarizes experimental findings across various families of magnetic carbon nanomaterials (MCNMs) examined in this section. It consolidates key performance data for functionalized  $\text{Fe}_3\text{O}_4$ , magnetic Biochars, Graphene/CNT hybrids and aerogels, and  $\text{g-C}_3\text{N}_4@\text{Fe}_3\text{O}_4$ , along with process comparators such as bare  $\text{Fe}_3\text{O}_4$  and magnetic-seeded filtration.

**Table 2.** Comparison of magnetic carbon nanomaterials for microplastics and nanoplastics removal.

Material Type	Removal Efficiency	Significant Impact
Magnetic/CNTs [26]	100% in 300 min for polyethylene (PE), polyethylene terephthalate (PET), and polyamide (PA) microplastics.	Strong affinity to the surfaces of all typical microplastics in testing solution and wastewater.
Graphene-like Magnetic Biochar [112]	The adsorption capacity of GLMB at 288 K (98.73 mg/g).	GLMB exhibited green, economical, high-efficiency, and reusable performance.
Black tea extract-based Magnetic Nanoparticles (BTMNP) [120]	90% microplastics removal efficiency.	Utilize polyphenol components as alternatives to toxic chemicals in the synthesis process.
Fe-doped porous carbon sponge [121]	$q_{\max} \approx 369 \text{ mg g}^{-1}$	Fe–C chemisorption (DFT) + $\pi$ – $\pi$ ; $\zeta$ effects; rapid adsorption; scalable synthesis.
–OH/–COOH/–NH <sub>2</sub> –Fe <sub>3</sub> O <sub>4</sub> [46]	$q_{\max}(\text{PE}) \sim 1611 \text{ mg g}^{-1}$ ; NH <sub>4</sub> <sup>+</sup> 13–18; PO <sub>4</sub> <sup>3-</sup> 24–30	Ion exchange; H-bonding; electrostatics.
Mg/Zn Modified Magnetic Biochar [98]	94.8–99.5% removal; $q_{\max} \approx 334\text{--}375 \text{ mg g}^{-1}$	Multipollutant removal; favorable LCA.
Chitosan-modified magnetic durian shell Biochar [15]	97.22%; $q_{\max} = 15.56 \text{ mg g}^{-1}$ ; 76.41% after 5×	Electrostatic + metal–O–PS; 5 cycles; real water tolerant.
PEG/PEI-coated Magnetic Biochar-Zeolite [122]	$q_{\max} 736\text{--}769 \text{ mg g}^{-1}$	Electrostatic, H-bonding, $\pi$ – $\pi$ stacking; green synthesis; low-cost reuse.
Magnetic Biochar (MBC) [123]	$q_{\max} \approx 234\text{--}248 \text{ mg g}^{-1}$	Electrostatic ( $\zeta$ +), heterogeneous sites; ultra-high adsorption; regenerable composite.
g-C <sub>3</sub> N <sub>4</sub> @Fe <sub>3</sub> O <sub>4</sub> & BNNS@Fe <sub>3</sub> O <sub>4</sub> [124]	>92–96% (Milli-Q); ~92% (effluent); ~80% after 3 cycles	Electrostatic + hydrophobic; Fe–O bands; rapid chemisorption; reusable.
Nanobiochar (OA-modified) [122]	93.4% (OA@NBA); 87.4% (OA@NBP)	Electrostatic, $\pi$ – $\pi$ , H-bonding, vdW; high efficiency; magnetic recovery.
Fe <sub>3</sub> O <sub>4</sub> -PWA/nOct [125]	99% (PS & PET); 83% (polysulphone)	$\pi$ – $\pi$ ; multilayer adsorption; acidic water removal; regenerable.
POM-IL@SiO <sub>2</sub> @Fe <sub>3</sub> O <sub>4</sub> [126]	PVC removed; metals 79.5–99.3%/92–99%	Hydrophobic adsorption; magnetic separation; scalable, robust.
Cellulose-Benzoate/MCNT [109]	>97%; $q_{\max} \approx 2.490 \text{ mg mg}^{-1}$	Co-removal platform; hydrophobic interaction; electrostatic.
Magnetic Seeded Filtration (MSF) [113]	Up to 95% (lab scale).	$\pi$ – $\pi$ /hydrophobic + magnetic; robust; 4× thermo-regen.
Scaled-up iron oxide Nanoflowers (NFs) [110]	Up to 1000 mg_MP g_NF <sup>-1</sup> ; 20–78% mineralization.	Hetero-agglomeration + magnetic separation; scalable; reusable; dual harvesting.
Fe <sub>3</sub> O <sub>4</sub> @C12 Superhydrophobic [15]	$q_{\max} \approx 809.29 \text{ mg g}^{-1}$	Hydrophobic + electrostatic + vdW; works in beverages; QA/QC.
Fe <sub>3</sub> O <sub>4</sub> @TiO <sub>2</sub> -CAN [127]	97.3% (PE); 96% (TC); 77.07% recyclability.	Photocatalytic degradation (S-scheme charge transfer; ROS generation); best-in-class TiO <sub>2</sub> performance.

Table 2. Cont.

Material Type	Removal Efficiency	Significant Impact
CTAB-Modified Magnetic Biochar [123]	The maximum microplastics removed were 98%.	The reusability results revealed that the developed RH-MBC-CTAB could maintain good stability for up to three reusability cycles.
Magnetic Biochar prepared by Red Mud [25]	The maximum adsorption capacities of original and modified biochar for PS were 227, 292, and 306 mg/g at pH 3–7, respectively.	The metal active sites provided by red mud noticeably contributed to the removal of microplastics.
Fe-modified lignin-based Magnetic Biochar [128]	The removal efficiency of polystyrene microplastics was 99%, with an adsorption capacity of 68.57 mg/g, which was 25 times higher than that of unmodified lignin biochar.	The effects of lignin/Fe salt mass ratios, adsorbent amount, adsorption time, adsorption temperature, solution pH, and presence of coexisting anions on adsorption efficiency were investigated.

## 6. Advanced Sustainable Nanorobots for Microplastics Removal

Nanorobots are small robots, typically measuring between 0.1 and 100 nm in size, and are built using nanotechnology [129]. Nanorobots are designed to perform specific tasks at the nanoscale by manipulating nanoscale objects or performing actions at the microscale level [130]. These machines are capable of detecting, capturing, and degrading pollutants, marking a groundbreaking shift from traditional treatment methods to more precise, efficient, and sustainable approaches [82]. They operate under diverse environmental conditions and can be tailored to address several types of nanoplastics. Their programmability, multifunctionality, and precision make them suitable for removing nanoplastics [131].

Nanorobots are classified based on their actuation mechanisms as chemically propelled robots or externally powered robots [132]. While chemically powered robots convert locally supplied fuels into force and motion, externally powered robots use magnetic, ultrasound, electrical, or optical fields to propel themselves and perform targeted tasks. This versatility in propulsion mechanisms has enabled them to engage in a variety of functions, ranging from detection to degradation or adsorption of nanoplastics. The unique ability of these robots to navigate and operate in complex environments enables efficient in situ cleaning without manual agitation, overcoming the limitations of traditional water treatment methods [82]. The use of the magnetic field for the propulsion of nanorobots is highly advantageous, offering wireless navigation without fuel, collective behavior can be easily configured into swarming abilities, stimuli-related memory, origami-like architecture, facile recollection, and recyclability, compatibility with other propulsion mechanisms that allow multiple modes of propulsion, and good permeability through the environment [133].

The need for renewable and efficient remediation approaches underscores the critical importance of developing new nanomaterials for this purpose. By exploiting suitable propulsion mechanisms, including light-powered and magnetically controlled modes, nanorobots can navigate turbulent and heterogeneous water bodies where nanoplastics accumulate [132]. Furthermore, several nanorobot designs incorporate surface functionalization with catalytic sites or enzymes to degrade the polymer chains of nanoplastics, thereby reducing their environmental persistence and toxicity [132]. Key considerations in the development of nanorobots include their sustainable design, the ability to degrade post-use to minimize environmental impact, and high specificity for binding or capturing microplastics. Advances in this field involve bio-inspired strategies, surface modification techniques to enhance plastic adhesion, and the integration of renewable materials.

Peng et al. (2023) [27] demonstrate the use of biohybrid nanorobots integrating algae platforms and magnetic nanoparticles for active capture and removal of microplastics from aquatic environments, achieving considerable efficiencies of 92% for nanoplastics and 70% for microplastics. The reusability of MARS was also tested over five cycles, maintaining 80% removal efficiency for nanoplastics and 54% for microplastics.

The design of magnetic nanorobots involves linking strands of a positively charged polymer to magnetic nanoparticles, enabling control via an external magnetic field. The polymer strands on the surface of the magnetic nanoparticles effectively attract microplastics. When subjected to a rotating magnetic field, the nanorobots can swarm together by self-organizing into flat clusters, allowing for precise control over the swarm's movement and speed [11].

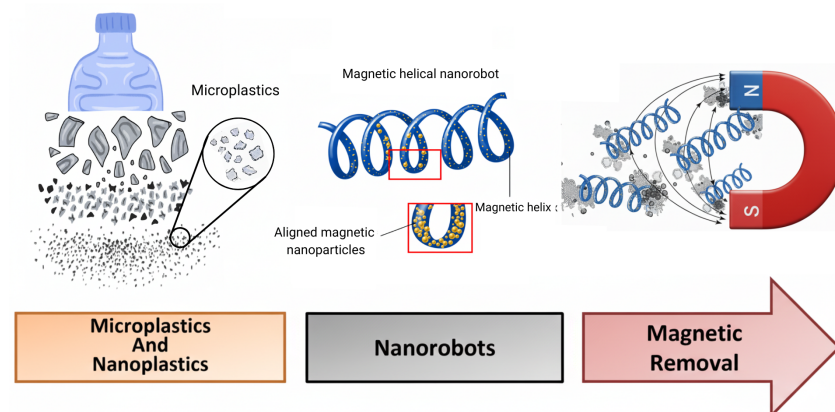
Swarm intelligence is a cutting-edge approach in nanorobotics that focuses on control and coordination, drawing inspiration from the collective behaviors found in biological systems [82]. The core principle of swarm intelligence in nanorobots is their ability to self-organize, communicate, and make decentralized decisions [134]. This ability enables them to engage in collective actions that can adapt dynamically to changing environmental conditions and tasks. In practical applications, particularly in combating water pollution, swarm intelligence enables nanorobots to identify, encapsulate, and degrade pollutants through coordinated behavior [12]. These swarms can cover larger areas, respond more effectively to the distribution of microplastics, and implement complex remediation strategies that individual robots cannot achieve independently [135]. Compared with chemical propulsion, magnetic nanorobots avoid residual fuel and enable deep-field penetration in turbid waters. Report actuation parameters (field amplitude and frequency) and water chemistry, as these govern capture kinetics, selectivity, and retrieval.

Su-Jin Song et al. (2025) [136] report the design of magnetically driven living bacterial nanorobots that exhibit a nature-inspired three-dimensional (3D) swarming motion, for the dynamic capture of aquatic micro and nanoplastics. By combining autonomous propulsion with magnetically guided navigation, they enabled the multimodal swarming manipulation of magnetotactic bacteria-based living nanorobots. The biobots achieved removal efficiencies of 83–89% for model PS microplastics and nanoplastics, and 60–96% for microplastics derived from PET water bottles and body scrubs. The actuation of 3D magnetic swarming motion resulted in a 6–43% improvement in removal performance, with more pronounced effects observed for larger microplastics and those with relatively weak electrostatic interactions with the bacterial surface, providing a straightforward and environmentally friendly approach for water purification.

Martina Ussia et al. (2024) [11] designed nanorobots based on spherical magnetic beads decorated with polymeric "hands" to capture both microplastics and free-swimming bacteria. The nanorobots respond to external rotating magnetic fields, forming organized, spinning planar swarms that enable controlled movement and collective behavior. In a simulated contaminated water tank containing 1  $\mu\text{m}$  polystyrene beads (as microplastics) and *Pseudomonas aeruginosa* bacteria, the microrobot swarm captured approximately 80% of the bacteria within 30 min at a concentration of 7.5 mg/mL. After use, the nanorobots were retrieved using a permanent magnet. The captured bacteria were then removed via ultrasound, and the water was UV-irradiated to ensure any remaining pathogens were eliminated. Microrobotic swarms engineered with magnetic cores and polymeric grips offer an innovative and efficient method for purifying water by capturing both microplastics and bacteria. Their ability to form coordinated swarms under magnetic influence enables effective contamination removal, and they can be retrieved and reused, offering an advanced, sustainable approach to water decontamination.

Bioinspired magnetic nanorobots have emerged as a promising strategy for removing microplastics, leveraging biomimetic magnetic actuation to enhance the efficiency of this process. By applying biological principles, these nanorobots enhance the separation of microplastics from water. They are designed to mimic the properties and functions of biological systems, using specially prepared magnetic nanocomposites to achieve optimal performance in microplastic removal [90]. These magnetic nanorobots replicate the movement and behavior of living organisms, enabling controlled motion in response to external magnetic fields (Figure 5 and Table 3). A novel approach to bioinspired magnetic separation utilizes reconfigurable liquid metal nanorobots, referred to as “LiquidBots” [137]. These nanorobots are coated in gallium oxide, which enhances their magnetic control and electrostatic interactions. This design, which mimics the mobility of living organisms, enables efficient removal of microplastic particles from water. Research has shown that these nanorobots can achieve removal rates of 74.3% for microplastics and 82.1% for nanoparticles (NPs) within just 2 h [137]. Further advancing the application of bioinspired methods, a magnetic microsubmarine based on a sunflower pollen grain was developed to remove large microparticles, achieving 70% removal efficiency via non-contact shoveling. This approach utilized the movement of microsubmarines to generate fluid flow. This flow facilitated the capture and transport of microparticles without direct contact as a promising strategy that does not require additional chemicals or invasive methods, thereby making it a sustainable solution for water remediation [138].

Geopolymers, a class of synthetic aluminosilicate materials, are distinguished by their exceptional mechanical properties, chemical stability, and inherently porous structure, making them promising candidates for adsorption applications [139]. This porous structure provides a large surface area, which is crucial for attracting and retaining micropollutants, including microplastics. Geopolymer adsorbents can effectively attract and retain microplastic particles due to their surface characteristics, such as porosity, charge, and chemical composition [140]. The use of geopolymers as sustainable materials for microplastic remediation has been documented [141]. With their high porosity, chemical stability, tunable surface chemistry, and regenerative properties, geopolymers show significant potential as both adsorbents and membrane materials. Extensive research has confirmed the effectiveness of geopolymers in eliminating microplastics, with findings indicating that silane-modified superhydrophobic geopolymer foam can achieve up to 99% removal efficiency for polyethylene microspheres in wastewater [141]. While this result underscores the viability of geopolymer-based solutions for microplastic remediation, focused research in this field remains limited.



**Figure 5.** Magnetic helical nanorobot for collecting microplastics.

The development of magnetically steerable Janus microparticles (MJMs) synthesized through a modified Pickering emulsion method employing aminated  $\text{Fe}_3\text{O}_4@\text{SiO}_2$  features a distinctive asymmetric, flower-shaped structure that optimizes microplastic adsorption, achieving removal efficiencies of up to 92% for polystyrene microplastics in just 20 min [121]. These MJMs illustrate the power of surface engineering in enhancing pollutant capture through hydrophobic and electrostatic interactions. These micromotors not only facilitate the mechanical separation of nanoplastics but also promote the catalytic degradation of organic pollutants, thereby enhancing purification. Their bubble-propelled mechanism further facilitates the efficient separation of suspended microplastics, resulting in a notable reduction in their concentration in polluted waters.

The combination of photocatalytic nanomaterials and magnetic components in nanorobots has led to the development of a novel class of environmental remediation agents. These nanorobots utilize light-driven self-propulsion and magnetic navigation to target, capture, and degrade nanoplastics in aquatic environments with impressive efficiency. Self-propelled, light-powered, and magnetic field-navigable Janus nanorobots made of hematite exhibit the ability to transport, capture, and degrade plastics actively [142]. These Janus nanorobots are fabricated by asymmetrically depositing different metals onto hematite microspheres, which are synthesized via low-cost, large-scale chemical methods. All nanorobots demonstrate fuel-free motility, with the capability of light-controlled activation and deactivation of movement, as well as magnetic field-controlled directionality. Notably, bimetallic coatings enable higher speeds than single-metal coatings. The nanorobots have shown an ability to completely degrade polyethylene glycol (PEG) chains into low-mass byproducts. This efficacy arises from their active motion, pH-programmable electrostatic PEG capture, enhanced charge separation at the hematite/metal interface, and the catalyzed photo-Fenton reaction. This research introduces a novel approach to the photodegradation of polymeric and plastic materials, as well as other pollutants in aquatic environments.

**Table 3.** Comparison of advanced sustainable nanorobots for microplastics removal.

Material Type	Removal Efficiency	Significant Impact
Living Bacterial Nanorobots [136]	83–89% for PS nanoplastics and 60–96% for microplastics derived from PET.	Nature-inspired three-dimensional (3D) swarming motion.
Nanorobots based on Magnetic beads [11]	Captured 80% of the bacteria within 30 min.	Magnetic beads with polymeric “hands” to capture microplastics and bacteria.
Biohybrid Nanorobots [27]	92% for nanoplastics and 70% for microplastics.	Algae platforms with magnetic nanoparticles.
PDA/PEI@ $\text{Fe}_3\text{O}_4$ MagRobots [143]	99% removal of polystyrene (PS) nanoparticles.	Sea-urchin-like structure, with a large surface area and an adsorption capacity up to 594.3 mg/g.
LiquidBots [137]	80% of micro/nanoplastics.	A reconfigurable and regenerable gallium-based liquid metal.
Keratin-based biohybrid nanorobots [46]	Removal efficiency for micro/nanoplastics with 95% and 82%.	$\text{Fe}_3\text{O}_4$ microspheres on waste human hairs as structural fiber supports.
Magnetic hydrogel Nanorobots (MHMs) [144]	95% removal efficiency within 3 min.	Removal efficiency through dynamic spinning that generates hydrodynamic flows.

Table 3. Cont.

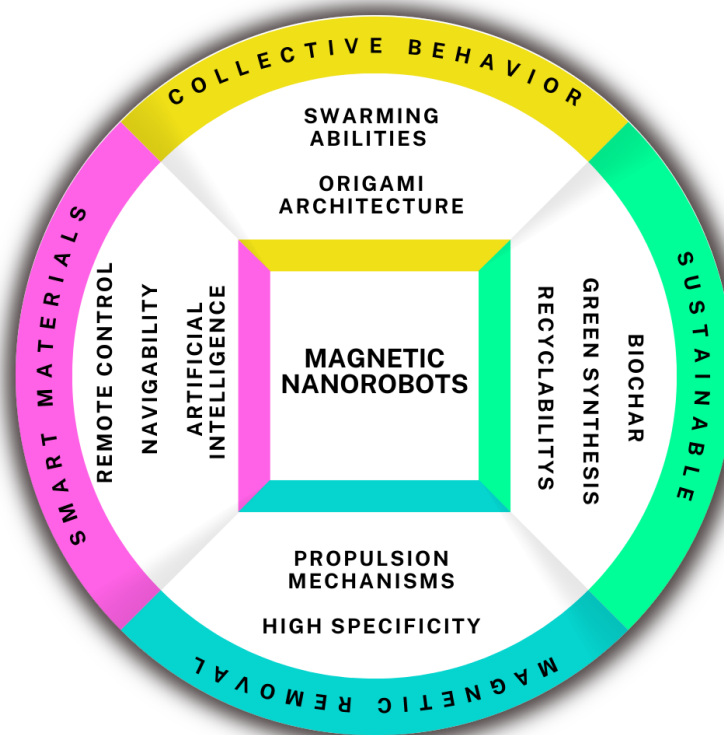
Material Type	Removal Efficiency	Significant Impact
Magnetic microsubmarine based on a sunflower pollen grain [138]	70% removal efficiency.	The fluid induced by the cooperative microsubmarines can remove the microplastics controllably in a non-contact method.
Ag@Bi <sub>2</sub> WO <sub>6</sub> /Fe <sub>3</sub> O <sub>4</sub> [145]	98% cleaning efficiency in 93 s.	Low-energy photoresponsive magnetic-assisted cleaning microrobot (LMCM) composed of photocatalytic material (Ag@Bi <sub>2</sub> WO <sub>6</sub> ) and magnetic nanoparticles (Fe <sub>3</sub> O <sub>4</sub> ).
Silane-modified superhydrophobic geopolymer foam [141]	99% removal efficiency for polyethylene microspheres in wastewater.	Geopolymers' transformative potential in addressing microplastic contamination.
Maifanite with a rotating magnetic field [146,147]	The removal efficiency ultimately achieves 100% when the concentration ratio with microplastics is set at 50%.	The influence of the flow field is remarkable in the magnetic removal process.
Iron–nitrogen co-doped layered biocarbon materials [148]	96.5% removal efficiency for PS microplastics.	Molecular dynamics simulations explain interactions between doped carbon and microplastics.
Polydopamine Enhanced Magnetic Chitosan (PDA-MCS) [148]	Removal efficiency of up to 91.6%.	Coral bio-inspired aerogels based on polydopamine and chitosan.
Magnetically steerable iron oxides-manganese dioxide core–shell micromotors [149]	Separated more than 10% of the suspended microplastics from the polluted water in 2 h.	Low-cost and scalable fabrication of bubble-propelled iron oxides-MnO <sub>2</sub> core–shell micromotors (Fe <sub>3</sub> O <sub>4</sub> -MnO <sub>2</sub> and Fe <sub>2</sub> O <sub>3</sub> -MnO <sub>2</sub> ) for pollutant removal.
Magnetic N-doped nanocarbon springs [150]	The Mn@NCNTs/PMS system can realize 50 wt % of microplastics removal by assisting with hydrolysis.	The magnetic nanohybrids were applied for peroxymonosulfate activation to generate highly oxidizing radicals to decompose microplastics under hydrothermal conditions.

Subsequent efforts over the past decade have led to impressive technological advances and powerful capabilities, including multifunctionality, collective behavior, intelligence, programmable navigation, hybrid control, biocompatibility, sensing, new fuels, transient behavior, and innovative manufacturing approaches, which enable advanced environmental applications of nanorobots [132]. Magnetic nanorobots operate at the nanoscale and interact directly with microplastics, reducing the release of excess treatment agents into the environment. However, the ecological risks associated with deploying nanorobots still need to be carefully evaluated [82]. One significant concern is the accumulation of nanorobots in natural water bodies, where they could become persistent pollutants if not effectively retrieved. Unlike conventional treatment residues, which can be contained and processed, nanorobots can move freely and disperse unpredictably, potentially leading to unintended environmental consequences [151]. For instance, if nanorobots are not biodegradable, they could accumulate in aquatic environments, enter food chains, and disrupt ecosystems [152]. This is especially concerning for nanoscale robots, which have greater mobility and may be ingested by microorganisms, leading to bioaccumulation and toxic effects at higher trophic levels [153]. Another critical issue is the potential formation

of toxic byproducts during the operation of nanorobots [82]. Many of these nanorobots rely on catalytic reactions to degrade microplastics [131]. While these reactions can be highly effective, they may also generate intermediate products whose toxicity is not yet fully understood [154]. Additionally, magnetic nanorobots that utilize metallic nanoparticles, such as iron or cobalt, may release metal ions that could accumulate in the environment, thereby raising further concerns about toxicity [155]. Efforts to address these challenges include developing sustainable, green-based, and low-cost nanorobots [131,156], improving production methods, and highlighting scalability. However, further innovation is needed to enhance material efficiency, reduce dependence on rare elements, and optimize recycling strategies [157].

After extensive revision of sustainable magnetic nanorobots for microplastics removal, gaps remain in a standardized, scalable, and real-world-validated process [74]. Most studies demonstrate impressive laboratory efficiencies and recoveries for microplastics; however, they are not yet optimized for real environmental conditions, continuous flow, safe recovery, reuse, or cost-effective energy at the plant scale. In general, there are no standardized methods for quantifying microplastics removal efficiency, making it challenging to compare the efficiency of different samples, methods, and processes for removing microplastics [158,159]. Swarms and magnetic nanorobots are typically evaluated in still or quasi-static reactors; robust guidance and integrity under flow, shear, and turbulence remain an open engineering problem for continuous treatment [160]. Beyond microplastic removal performance, it is imperative to evaluate environmental health, toxicity, and safety data on magnetic nanorobots, including release, persistence, and by-products [161].

Future research should focus on developing hybrid magnetic nanorobots that integrate detection, separation, and degradation within a unified system [162]. The advancement of intelligent nanorobots with responsive surface modifications could enhance their interaction with nanoplastics under real environmental conditions [163]. The investigation of bioinspired nanorobots for the targeted capture and degradation of nanoplastics represents an exciting new frontier in nanotechnology [164]. Furthermore, integrating AI-driven monitoring and control systems with nanoplastics remediation technologies could facilitate adaptive, real-time adjustments, thereby improving efficiency [15,165]. Artificial intelligence, particularly through machine learning, is emerging as a transformative solution [166–168]. AI enhances the identification of microplastics [169], enables predictive modeling for filtration systems [170], and optimizes operational parameters to boost efficiency [171]. Scaling these innovations requires a multidisciplinary approach that integrates nanotechnology, robotics, environmental engineering, and biotechnology [172]. Future developments emphasize circular-economy principles, focusing on full recyclability and minimal environmental impact. Combining multiple removal mechanisms. Conducting pilot-scale studies and life-cycle assessments to evaluate feasibility and environmental impact [76]. Collaborative efforts must focus on creating cost-effective, eco-friendly, and energy-efficient systems to ensure practical implementation [173]. By addressing these challenges, we can expedite the transition from laboratory-scale research to real-world solutions for the remediation of microplastics and nanoplastics, ultimately contributing to global environmental sustainability (Figure 6).



**Figure 6.** Summary of advanced sustainable magnetic nanorobots for microplastics.

## 7. Conclusions

Magnetic nanorobots represent a promising, sustainable approach for removing microplastics and nanoplastics from water. They offer high efficiency, reusability, and adaptability across diverse environments, thanks to programmable selectivity and multifunctional actions such as magnetic capture, degradation, and detection. They can be further improved in swarming, intelligent communication, and multi-pollutant targeting, opening the door to next-generation water-cleaning technologies. Sustainable magnetic nanorobots offer advantages such as low cost, environmental compatibility, renewability, and the ability to interact with diverse types of microplastics. Despite their promise, the application of sustainable magnetic nanorobots at scale faces several challenges. These include variability in adsorption capacity, lack of standardized testing protocols, and the need to assess performance under realistic operating conditions. Future research must prioritize the development of hybrid magnetic nanorobots that integrate detection, separation, and degradation within a unified system operating under real environmental conditions. This approach is essential for advancing global environmental sustainability.

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