

Magnetic polymeric composites: potential for separating and degrading micro/nano plastics



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ABSTRACT

The pervasive presence of microplastics (MPs) and nanoplastics (NPs) across various ecological settings, organisms, and products has underscored the urgent need to collect data on their abundance and identify the polymer types in diverse substrates. NPs distinguish themselves from MPs by their smaller size. The application of magnetic composites in wastewater treatment has grown in popularity due to their remarkable flocculation capabilities and ferromagnetic properties. This comprehensive review centers on the synthesis methods, preparation technologies, and practical applications of magnetic polymeric composites for separating and degrading MPs/NPs. It offers an extensive overview of recent advancements in the separation and degradation of MPs/NPs within various wastewater sources. The review begins by consolidating efficient methods for the separation of MPs/NPs. This includes techniques such as adsorption, coagulation/flocculation, flotation, filtration, and magnetic separation. The subsequent section explores diverse approaches for the degradation of MPs/NPs, encompassing advanced oxidation processes (AOPs), electrochemical degradation, photocatalysis, and biological degradation. Additionally, the review offers in-depth insights into the synthesis and characterization of magnetic polymeric composites. Lastly, it discusses the current challenges and prospects associated with the separation and degradation of MPs/NPs in wastewater treatment.

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1. Introduction

The production of plastic has been steadily increasing over the past decade. In 2017, a significant 348 million tons were produced, and this figure is projected to double by 2025 [1]. The widespread use of polymers in various applications and the subsequent rise in their production have led to a corresponding increase in plastic waste. MPs which are small plastic particles of various shapes measuring from 0.1 μm to 5 mm in length, are now a well-known issue, with a staggering total of 51 trillion plastic fragments [2]. NPs are characterized by their size falling from 1 nm to 1 μm . These criteria distinguish them from engineered nanomaterials, as outlined by [3]. Nanoplastics stand out from microplastics due to their distinctive transport characteristics, interactions with light, and natural colloids. They exhibit a significant proportion of exposed surface molecules, increased bioavailability, and the release of plastic additives. Moreover, their characteristics differ from engineered nanomaterials, primarily in terms of pronounced particle diversity and the potential for rapid fragmentation in natural environments. These distinctive attributes have profound implications for their environmental destiny, potential impacts on ecosystems and human well-being, and the techniques employed for sampling and analysis [4].

MPs/NPs in the environment can lead to ecotoxicological concerns [5]. The presence of MPs/NPs has been linked to diminished growth, reduced reproductive capabilities, weakened immunity, and the development of deformities in both animal and human reproductive systems [6]. Furthermore, MPs/NPs can serve as carriers, accumulating substantial amounts of trace metals, persistent organic pollutants, and harmful additives far exceeding the concentrations typically in natural sediments [7]. The presence of MPs/NPs in wastewater treatment plants affects microbial communities, hinders sludge hydrolysis, and accumulates acidic substances [7].

Separation and degradation could be considered two different methods for removing MPs and NPs from aqueous media. The separation methods are primarily classified based on three separation principles: size, hydrophobicity, and density [8]. Common adsorbents face limitations, particularly their challenging separation from sample solutions or proposed media [9]. Numerous adsorbents and catalysts encounter challenges related to effective separation from the solution. This concern can be mitigated by integrating magnetic properties, facilitating the effortless extraction of the adsorbent from water through the application of an external magnet. [10,11]. Magnetic separation, known for its simplicity, minimal energy requirements, and cost efficiency, has been extensively utilized in various water treatment applications.

The selection of the magnetic material is a pivotal factor influencing the success of the magnetic separation process [12]. Magnetic particles typically contain ferromagnetic elements like cobalt, iron, and nickel, along with their composite oxides, or alloys, structures [13]. Composites are materials composed of two or more constituent components with different physicochemical characteristics that exhibit hybrid properties unseen in either constituent individually [14]. The selection of a polymer matrix for magnetic polymer composite particles is contingent upon their intended application, often involving a choice between commodity plastics like polystyrene (PS) and polymethyl methacrylate (PMMA) or conductive polymers such as polypyrrole (PPy) and polyaniline (PANI) [13].

Alternatively, degradation processes provide viable options for removing small microplastics and nanoparticles from water. These approaches offer potential eco-friendly solutions and address the challenge of managing microplastics generated during separation in the concentrated stream. [15]. Numerous techniques have been employed to date for both the elimination and degradation of MPs/NPs from wastewater [16].

The choice of separation techniques is contingent upon the physicochemical characteristics of the MPs/NPs, including their

hydrophilicity, size, shape, and magnetic attributes. For instance, plastics modified with nano-iron oxide can be easily isolated from water using magnetic separation, as demonstrated in work by Shi et al. [17]. In addition to standalone approaches, integrated methods that harness the strengths of various techniques are commonly adopted to augment separation efficiency. An illustrative example of this is the coagulation-flocculation-sedimentation-filtration procedure, widely employed in various water treatment facilities to separate MPs/NPs, as evidenced in a study conducted by Nikiema and Asiedu [18].

In the last five years, MPs [19–28] and NPs [29–31] has been well documented regarding their detection methods, fate, and remediation strategies, however, a common challenge many adsorbents and catalysts face is their difficulty separating from the solution. This separation issue can be effectively addressed by incorporating magnetic properties, allowing for easy separation of the adsorbents from water using an external magnet. Given the lack of a comprehensive review explicitly focused on the application of magnetic polymeric composites in the context of MPs/NPs, this review aims to provide a detailed account of the synthesis methods, characterization, and performance of magnetic polymeric composites, with a specific focus on their capabilities in terms of separation and degradation. Additionally, we shed light on the current challenges that impede the development of separation techniques in this field. Finally, we extend our discussion to explore the prospects and opportunities in this exciting and promising area of research. The review has been organized into sections; preparation processes and characterization techniques are then discussed (Section 2). Section 3 demonstrates the separation strategies of MPs and NPs in different wastewater, and then Section 4 highlights the degradation of MPs and NP-based adsorbents. Section 5 demonstrates the results of the review, deliberating the level of application of magnetic polymeric composites for micro/nano plastics separation and degradation. Section 6 discusses the elucidation of the mechanisms of removal and degradation of contaminants and Sections 7 and 8 presents the challenges and draws conclusions according to the findings.

2. Detection and physicochemical characterization of MPs and NPs

Plastics are categorized into thermosets and thermoplastics based on their behavior when exposed to heat [21,32]. The physicochemical structure of plastic particles is intricate and subject to variation, contingent upon their sizes, shapes, types, colors, sources, and production methods. Despite the many different types of plastic available, the market is primarily governed by six principal classes: polyethylene (PE), polyvinyl chloride (PVC), polypropylene (PP), polystyrene (PS), polyethylene terephthalate (PET), and polyurethane (PU). These plastics have different chemical structures and properties, which can affect their behavior and impact the environment [33]. These polymers can also be modified with various additives to enhance their properties, such as plasticizers, stabilizers, pigments for color, and flame retardants. In some cases, these stabilizers may also contain hazardous metals such as lead, cadmium, and mercury [34].

MPs are classified as primary and secondary according to manufacturing purpose and fragmentation [35]. MPs can originate from two primary sources. The first one, primary MPs, represents plastic particles intentionally produced in micrometric sizes for specific applications. They can appear as pellets directly released by various care products or fibers from the textile and laundry industry [36]. 'Primary microplastics' comprise particles intentionally manufactured for direct or indirect use as raw materials in the production of various consumer polymer products. These primary microplastics are commonly used in scrubs, facial cleansers, toothpaste, cosmetics, abrasives, and exfoliants [37]. Primary MPs are typically spherical or irregularly shaped, while secondary MPs are often jagged and irregularly shaped due to the mentioned fragmentation process. The composition of primary and secondary MPs can also differ, as primary MPs may contain additives

Table 1
Abundance, type, and size of micro-/nanoplastics in different aquatic sources worldwide.

Wastewater type	Geographical location	Period	MPs abundance (particles/L)		Shape	MP/NP type	Size	Reference
			Range	Mean				
Drinking water	North Western Germany	August 2014	0-0.007	0.0007	Fragments and fibers	Polyester (dominant type, 62%), PE, PA, PVC and epoxy resin	50-150 µm	[46]
Drinking water	Rüsselsheim, Germany	October 2019- March 2020	0	0	-	-	-	[47]
Drinking water	Skane, Sweden	May 2019	0-1.22	0.174	Fragments (81%) and fibers (19%)	Polyester and PA (dominant type), PVC, PS, acrylic acid, PE, PU, and PP	< 20 µm, dominant size	[48]
Groundwater	North China	2020	4-72	29	Fragments (dominant shape: 86.9%) and fibers	PA, PE, PP, PVC and PS	0-50 µm (dominant size, 80%)	[49]
Groundwater	Jeju Island, Korea	July-September 2021	0.006-0.192	0.007	Fragments and fibers	PP and PE (dominant types), PET, PS, PA, PVC, ABS and PU	20-100 µm (dominant size, 95%)	[35]
Groundwater	Haean Basin, Seoul	June-July 2021	0.02-2.56	0.64	Fragments (dominant shape) and fibers	PP and PE (dominant types), PET, PS, ABS, PA, PVC, PMMA and PU	20-50 µm (dominant size, 63.2-91.7%)	[50]
Groundwater	Haean Basin, Seoul	October-November 2021	0.20-3.48	1.15	Fragments (dominant shape) and fibers	PP and PE (dominant types), PET, PVC, PS, PA, ABS, PMMA and PU	50-100 µm (dominant size)	[50]
Groundwater	Tianjin, China	2022	17-44	27.9	Fibrous MPs (dominant shape, 44.7%), films (28.4%), fragments (20%), and pellets (6.9%)	PET (24.7%), PE (19.7%), PS (16.6%), PP (14.3%), cellulose (12.2%), teflon (8.2%) and Witcobond (4.0%)	10-200 µm (dominant size, 73.1%), 200-1000 µm (16.4%), 1-2 mm (7.4%) and 2-5 mm (3.1%)	[51]
Groundwater	Mexico City, Mexico	NR	5-91	18	Fibers (dominant shape)	PTT and epoxy resin	< 0.5 mm (dominant size, 50%), 0.5-1 mm (25%) and 1 - 5 mm (25%)	[52]
Mineral water (Single use/reusable PET-bottled)	Bavaria, Germany	NR	NR	2649 / 4889	NR	PET	< 5 µm (dominant size)	[53]
Mineral water (glass-bottled)	Bavaria, Germany	NR	NR	6292	NR	PE (46%), PP (23%) and styrene-butadiene-copolymer (14%) (dominant type)	< 5 µm (dominant size)	[53]
Mineral water (glass-bottled)	Germany	NR	4-156	50	NR	Polyester + PET (41%), PE (35%), PP (8%), PA (12%), and others (4%)	5-10 µm (dominant size, 45%), 10-20 µm (32%), 20-50 µm (14%), 50-100 µm (7%) and > 100 µm (3%)	[54]
Mineral water (single-use plastic bottled)	Germany	NR	2-44	14	NR	Polyester + PET (59%), PE (9%), PP (1%), PA (1%), and others (30%)	5-10 µm (dominant size, 45%), 10-20 µm (30%), 20-50 µm (22%), 50-100 µm (5%) and > 100 µm (2%)	[54]
Mineral water (returnable plastic bottled)	Germany	NR	28-241	118	NR	Polyester + PET (84%), PP (7%), PE (5%), PA (2%), and others (2%)	5-10 µm (dominant size, 56%), 10-20 µm (29%), 20-50 µm (12%), 50-100 µm (2%) and > 100 µm (1%)	[54]
Mineral water (beverage cartons)	Germany	NR	5-20	11	NR	PE (38%), polyester + PET (32%), PP (26%) and others (4%)	5-10 µm (dominant size, 39%), 10-20 µm (28%), 20-50 µm (16%), 50-100 µm (10%) and > 100 µm (7%)	[54]
Mineral water (PET-bottled)	Kermanshah, Iran	NR	0-36	8.5	Fragments (dominant shape, 93%) and fibers (7%)	PET, PS, and PP	1.28-4.2 µm (mean size = 2.44 µm)	[55]
Tap water	China	NR	0-1247	440	Fragments (dominant shape), fibers and spheres	PE (26.7%), PP (24.4%), PE+PP (22%), PPS (7.3%), PS (6.5%), PET (3.3) and others (9.8%)	520 µm (dominant size, range = 3-4453 µm; mean = 66 µm)	[56]
Tap water	North Dakota, United States	May 2019	66-472	182	Fibers (dominant shape, 48.6%), fragments (41.8%), pellets (5.7%) and filaments (3.7%)	NR	2.5-100 µm (dominant size)	[57]

(Continued on next page)

Table 1 (continued)

Wastewater type	Geographical location	Period	MP's abundance (particles/L)		Shape	MP/NP type	Size	Reference
			Range	Mean				
Water (PET- and glass-bottled)	Bangkok, Thailand	NR	NR	140 (PET-bottled) 52 (glass-bottled) 11.7	Fibers (62.8%) and fragments (37.2%)	PET, PE, PP and PA (dominant type)	6.5–50 µm (dominant size) and > 50 µm	[58]
Water (Single-use bottled)	Serdang and Kembangan, Malaysia	July 2023	8–22	16	Fragments (90.1%) and fibers (9.1%)	PET and PP	100–300 µm (dominant size)	[59]
Water (PET-bottled)	China	NR	2–23	16	Fibers and fragments	Cellulose (dominant type, 63.6%), PS (13.6%), PE (9.1%), PET (9.1%) and PU (4.6%)	100–300 µm (dominant size), 29.2–49.2%, 25–50 µm (0.4–14.6%), 50–100 µm (19.4–37.1%) and 1–5 mm (4.8–12.3%)	[60]
Water (Plastic-bottled)	North Dakota, United States	May 2019	49–166	101	Fragments (dominant shape, 51.7%), fibers (38.1%), filaments (9.1%) and pellets (1.1%)	NR	< 100 µm (dominant size)	[57]
Raw water	Úhlava River, Milence plant, Czech Republic	Winter 2019/2020	NR	23	Fragments (dominant shape) and fibers	NR	< 5 µm (dominant size, 50%)	[61]
Treated water	Úhlava River, Milence plant, Czech Republic	Winter 2019/2020	NR	14	Fragments (dominant shape) and fibers	NR	< 5 µm (dominant size, 65%)	[61]
Raw water	Úhlava River, Pízen plant, Czech Republic	Winter 2019/2020	NR	1296	Fragments (dominant shape) and fibers	NR	< 5 µm (dominant size)	[61]
Treated water	Úhlava River, Pízen plant, Czech Republic	Winter 2019/2020	NR	151	Fragments (dominant shape) and fibers	NR	< 5 µm (dominant size)	[61]
Raw water	Yangtze River Delta, China	December 2018–January 2019	NR	6614	Fibers (dominant shape), fragments and spheres	PET, PE, PP, and PA	1–5 µm (dominant size, 54.6–58.0%)	[62]
Effluent	Yangtze River Delta, China	December 2018–January 2019	NR	930	Fibers (dominant shape), fragments and spheres	PET, PE, PP, and PA	1–5 µm (dominant size, 84.4–86.7%)	[62]

ABS: acrylonitrile butadiene styrene, PE: polyethylene, PA: polyamides, PC: polycarbonate, PET: polyethylene terephthalate, PMMA: polymethyl methacrylate, PPS: polyphenylene sulfite, PS: polystyrene, PP: polypropylene, PTT: trimethylene terephthalate, PU: polyurethane, PVC: polyvinyl chloride, NR: Not reported

such as colorants, stabilizers, and flame retardants. In contrast, secondary MPs may have a mix of plastic types and additives [32].

Conversely, secondary microplastics arise from the fragmentation of larger plastic particles released into the environment. Once released, these plastic items undergo a series of processes, including exposure to solar radiation, photo- and thermal oxidation, and mechanical abrasion in the environment [38]. 'Secondary microplastics' encompasses particles formed due to the breakdown, fragmentation, and gradual deterioration of larger plastic pieces. Secondary MPs present a higher level of public health concern than primary microplastics. Most environmental microplastics belong to this category, stemming from the fragmentation of larger plastic debris in the environment [37].

The shape of MPs/NPs is a pivotal factor influencing their behavior and destiny in the environment. The structural manifestations of MPs exhibit diversity contingent upon the size of plastic particles, encompassing forms such as fibers, fragments, films, foams, pellets, spheres, and beads. As the particles increase, they tend to be thinner, longer, and have a rougher surface, while smaller particles tend to be more rounded. The shapes of MPs can change depending on the amount of time they spend in the environment and the degree to which they break down. Microplastic particles discovered in marinas, characterized by their sharp edges, are thought to have recently entered the sea. In contrast, smoother particles with rounded edges have been detected in sediment for an extended period, gradually becoming smoother through erosion processes [39].

The role of surface charge density in their colloidal stability and ease of separation is another decisive parameter. Due to the particle size, there is a disparity in diffusion and surface charge, leading to instability in polystyrene particles. Enhancing PS particle stability relies on increasing surface charge and density, influenced by particle size and surface treatment. Identifying the ideal aggregation for particle removal during water treatment is challenging. When particles are suspended in water, a surface charge emerges, forming an electrical double layer at the particle-water interface. Increasing ionic strength leads to double-layer compression, in which case more ions adsorb onto particle surfaces, reducing the double layer's thickness and enhancing its charge. This compression may elevate the zeta potential in certain instances. Determining the optimal coagulant dosage involves establishing the initial pH at which particle surface charges neutralize [40].

MPs/NPs persist in the environment for extended periods, causing ecosystem disruptions and affecting wildlife. The ecological impacts of these particles are diverse and can manifest in several ways. Plastic particles can harm wildlife through ingestion, causing physical damage or blocking the digestive tract [41]. MPs and NPs in aquatic systems can significantly influence these ecosystems' overall health and functioning. They can alter food web dynamics and lead to degradation in water quality. The accumulation of these particles in organisms' tissues can harm and disrupt biological processes. The potential effects of this contamination on human health remain largely uncharted.

Nevertheless, research has unveiled that MPs and NPs can inflict internal damage on marine animals, disrupt their reproductive systems, and even lead to fatalities [42,43]. These particles can enter the body through ingestion, inhalation, and dermal exposure and can cause inflammation, oxidative stress, and deoxyribonucleic acid (DNA) damage in cells. They can also disrupt hormone function and affect the immune system, potentially leading to chronic diseases like cancer, neurodegenerative diseases, and cardiovascular diseases [44,45]. Plastic debris

can also release harmful chemicals into the environment, which can have a long-lasting impact on local ecosystems. The characteristics that distinguish nano plastics from microplastics, according to [4], can be summarized into the following:

1. The predominant influence of Brownian motion, characterized by the unpredictable movement of particles in a liquid suspension, takes precedence over sedimentation and buoyancy effects.
2. Departure from the geometric/ray approximation regarding the interaction between light and matter. While microplastics can often be analyzed by assuming that light travels in a straight line, nano-plastics exhibit wave-like behavior when interacting with light.
3. A noticeable abundance of molecules on the particle's surface results in a relatively greater emphasis on surface interactions than physical ones.
4. The particle size may closely resemble the dimensions of environmental macromolecules, leading to potential adsorption and hetero-aggregation phenomena while remaining considerably smaller than microorganisms.
5. Facilitating particle uptake, translocation, and membrane transport in biological systems.
6. Compact length scales may expedite the diffusion-driven release of plastic additives and unintentional substances from the original bulk plastic.

Table 1 summarizes the abundance, type, and size of MPs/NPs in different water sources worldwide.

2.1. Synthesis and characterization techniques of magnetic polymeric composites

Soft magnetic particles are commonly present in materials characterized by a cubic crystalline structure, which includes elements like iron, nickel, cobalt, iron-nickel, iron-cobalt, and metal oxides such as M-iron oxide, where M is a divalent metal [13]. The production of magnetic polymer particles can be broadly classified into three categories, as illustrated in Fig. 1a particles featuring a magnetic core enveloped by a polymer shell, Fig. 1b particles with a polymer core encased in a magnetic shell, and Fig. 1c magnetic particles embedded within polymeric materials. [13].

2.1.1. Synthesis techniques of magnetic polymeric composites

The advancements in polymer research incorporating different NPs have led to several polymer composites showing "ad-hoc" mechanical, thermal, and electroactive properties [63]. However, several studies reported limitations in preparing nanocomposites with the desired properties. The uniform dispersion of nanoparticles in polymeric supports is the main obstacle to the mass production and commercialization of nanocomposites [64,65]. Understanding and developing efficient processing methods at the nanoscale and usable at the macroscopic scale is required to upgrade their wide applications. Some of the primary potential polymer composite synthesis and preparation methods are blending, *in-situ* polymerization, molding, coprecipitation, and grafting, as explained in the upcoming subsection (Fig. 2).

2.1.2. Blending method

Mixing filler material and polymer is the conventional and most straightforward way of synthesizing organic-inorganic nanocomposites.

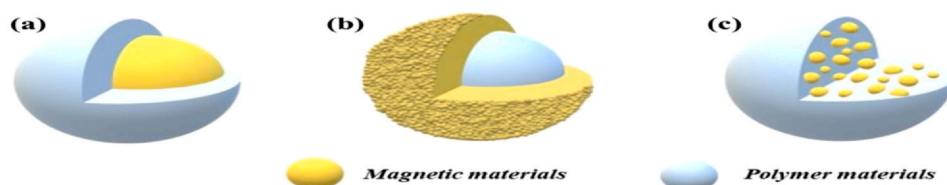


Fig. 1. Diverse morphologies of magnetic polymer nanoparticles include (a) Magnetic core-polymer shell, (b) Polymer core-magnetic shell, and (c) Magnetic particles embedded in a polymeric material. [13].

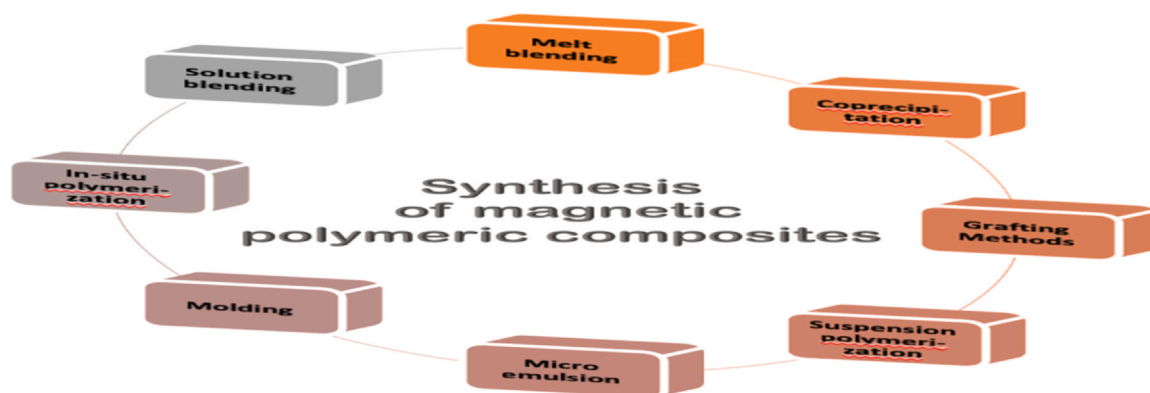


Fig. 2. Synthesis of magnetic Polymeric Composites.

The two most common and popular methods are melt and solution blending because there are no restrictions on the types of host polymers and NPs that can be used [66]. Since they do not restrict the chemical makeup of the material utilized, large-scale industrial applications have primarily used a blend of polymers and magnetic particles by encouraging their homogenous mixture for making nanocomposite materials.

Melt blending is more flexible, simple, cost-effective, and environmentally beneficial (since no solvent is required), an optimal method for bulk manufacturing of thermoplastic nanocomposite preparation under an inert environment created by neon (Ne), argon (Ar), or nitrogen (N₂) [67,68], [69,70]. The melt compounding method has broadly been used with many materials, including oxides and carbon nanotubes. So, the main advantage of this method is that a massive quantity of polymer composites are commercially manufactured through the extrusion process. Much research is dedicated to studying the effect of process parameters and preparing different composite polymers using this method. For instance, Chung et al. [71] used this method to synthesize cross-linked shape memory polyurethane/iron oxide magnetic compounds, observing good mechanical and shape memory qualities. Thus, melt blending enhanced magnetic quality with nanocomposite material's structural flexibility was obtained using components such as linear low-density polyethylene, polybenzoxine, and cobalt ferrite [72]. However, at high filler levels, the melt-blending process frequently results in inadequate filler dispersion, which leads to agglomeration and intercalation [73].

Solution-blending or solvent-casting is another approach considered one of the most straightforward ways to make polymeric nanocomposites [74,75]. This process involves physically entrapping nanostructures within polymeric matrices by casting and solvent evaporations, requiring less operation time and minimal equipment. A novel composite prepared using magnetite nanoparticles and polypropylene in a polymeric matrix has been synthesized via solvent casting and compression molding. According to research by Shirinova et al. [76], the magnetic properties and structural flexibility of polymeric composites depend on the percentage of magnetic fillers used. Additionally, Bayraktar et al. [77] used the solution mixing method to produce silver nanowire/poly lactide nanocomposites, which were 3D-printed to acquire the required structures. The final product showed outstanding antibacterial qualities and a nanofiller dispersion in the matrix. In this research, it could be observed that by combining the adaptability of 3D printed-polymers with some nanofiller's antibacterial properties, the solution mixing method is effective for use in 3D printing. However, several investigations continue to maintain the capability of generating NPs with improved polymeric matrix dispersibility and durability over time against aggregation despite the simplicity of these procedures [78].

2.1.3. *In situ* polymerization

In-situ polymerization is a method of polymer composite formulation that can particularly avoid the strict thermodynamic restrictions connected with the polymer intercalation process, making it more successful in creating various polymer composites with well-dispersed fillers [79]. It also provides flexible tuning of the matrix composition and structure, enabling the interface design between the nanoparticles and the polymers. It also delivers an efficient approach to synthesizing various polymer composites with an expanded property range. Many studies use the *in situ* polymerization method to create innovative polymer composites, showcasing its advantages over alternative synthetic techniques [80].

In situ polymerization entails directly scattering inorganic NPs to monomer or monomer solutions, followed by typical polymerization steps to polymerize further the monomer dispersion [64,81]. Applying a polymer coating to the surfaces of magnetic NPs enhances their interaction with inorganic molecules, reduces NPs release, and prevents oxidation in the surrounding medium. Similarly, in the manufacturing of polymer/magnetic particle composites, several synthesis techniques have been documented, including dispersion, suspension cross-linking, and inverse emulsion polymerization. Shabzendedar et al. [82] recently developed a superparamagnetic core-shell nanocomposite using poly (m-aminobenzenesulfonic acid) and iron oxide (Fe₂O₃, Fe₃O₄) nanoparticles through an *in situ* polymerization method. The magnetic polymer composite showed an increased power-conversion performance and a saturation magnetization value of about 40 emu/g. Due to these unique qualities, this material could effectively be used in polymer solar cell developments. By a similar method, Wang et al. [83] prepared innovative composite hydrogels using polypyrrole (PPy)- iron oxide NPs. Then, they added to a polyvinyl alcohol (PVA) matrix. The resulting hybrid hydrogel exhibited excellent mechanical, electrical, and magnetic properties. Hence, it could be a promising functional magnetic polymeric composite used as an electronic device in biomedical applications.

2.1.4. Molding

Molding represents a straightforward method for replicating patterns through soft lithography. In this process, the mold's walls are heated to a temperature exceeding the melting point of the mold material, facilitating a more rapid flow of material into the cavities [68]. A standard method to produce magnetic composites is molding, which thoroughly combines magnetic fillers and polymeric precursors before curing the mixture to create specified forms or structures [84]. Various molding techniques, including injection, resin transfer, and compression, fill a mold under pressure and heat [68]. Recently, this method was used to coat Fe₃O₄ nanoparticles using carbon nanotubes (CNTs) mixed with cross-linked poly(cyclotriphosphazene-co-4,4'-sulfonyldiphenol), being then used as a photothermal magnetic filler [85].

2.1.5. Coprecipitation

Coprecipitation is a simple and practical method for synthesizing magnetic polymer nanocomposites from aqueous salts, e.g., mainly Fe^{2+} and Fe^{3+} solutions [86,87]. Typically, the coprecipitation method of magnetic NPs synthesis yields polydispersed, spherical particles with dimensions ranging between 5–40 nm [88,89]. However, this procedure often entails the formation of concurrent nucleation, growth, coarsening, and aggregation processes [90]. For instance, Mehra et al. [91] developed a polymer matrix with uniform melamine cyanurate dispersion using an in situ coprecipitation method. This proper inclusion led to a 65% increase in thermal conductivity, providing a fresh approach to creating novel thermally-conductive materials.

A similar method was used to develop various magnetic polymer composites for different applications. Graphene oxide/chitosan-based magnetic polymers were prepared through a co-precipitation reaction between graphene oxide and Fe^{2+} and Fe^{3+} ions, which resulted in the development of a magnetic polymer composite with excellent adsorbent properties for dyes and heavy metals removal from aqueous solutions [92]. Fe_3O_4 -dispersed polylactic acid (PLLA)-based magnetic composite was developed using a solvent-precipitation process followed by a compression molding step, resulting in the enhancement of the properties of the material [93]. Also, an in-situ coprecipitation of Fe^{2+} and Fe^{3+} salts in the presence of polyurethane was accomplished to synthesize a polyurethane-based magnetic polymer to remove tetracycline-family antibiotics present in polluted wastewater [93].

2.1.6. Grafting methods

When a monomer is immediately adsorbed to the surface of a particle, it can be challenging to polymerize it using traditional methods. However, grafting techniques can be used to overcome this challenge. Using a grafting procedure, a polymer chain can be grown from the particle's surface, which allows the monomer to be polymerized. This process can result in a polymer covalently attached to the surface of the particle, which can provide several benefits. For example, the polymer can improve the stability of the particle, enhance its compatibility with other materials, or change its surface chemistry. Grafting methods can be carried out using various techniques, such as radiation, plasma, or chemical grafting. [94]. In "grafting to" methods, anchoring groups are added to polymer chains to bind to the particle's surface [95,96].

Similarly, in the presence of hydrogel precursors, the surfaces of magnetic NPs can be altered by incorporating suitable functional groups that act as cross-linkers, forming covalent bonds with polymer chains. This allows for the formation of magnetic NP-based hydrogels covalently bonded to the polymeric networks [97]. Due to these covalent interactions, magnetic polymer matrices synthesized by the grafting approach possess better dispersion stability properties.

Individually synthesized polymer side chains and backbones are combined during the grafting-onto processes [98]. Grafting does not entail a chain reaction [99]. According to Yang et al. [100] study, by using gamma radiation, filler was successfully grafted onto polymeric materials. Thus, Tanasa et al. [101] have reported the synthesis of a functionalized-magnetite NP hydrogel based on polyacrylamide. Additionally, Hu et al. [102] prepared a magnetic hydrogel from polyacrylamide (PAAm) and 3-(trimethoxysilyl) propyl methacrylate-coated Fe_3O_4 by using the grafting method.

2.1.7. Merits and demerits in preparation methods

It is essential to recognize that each available option possesses distinct advantages and limitations, both in terms of cost and effectiveness, suitability, and environmental impact, as highlighted by [103]. Table 2 provides a comparative analysis of the methods discussed, offering a concise overview of their merits and demerits and relevant references related to the subject.

2.2. Physicochemical characterization

The intricacy of various sample matrices has compelled researchers to employ multiple analytical techniques for identifying MPs. The characterization of MPs-related experiments is a decisive factor in ensuring the reliable interpretation of findings [105].

The analytical methods used for the identification of nanoparticles can be categorized as visual identification, microscopy, chromatography, and spectroscopy. Both MPs and NPs have been subjected to a diverse collection of characterization methods, including Fourier Transform Infrared Spectroscopy (FTIR), Thermogravimetric Analysis (TGA), X-ray Diffraction (XRD), Scanning Electron Microscopy (SEM), Small-angle X-ray scattering (SAXS), transmission electron microscopy (TEM) and others. These methods have yielded valuable insights into crystal properties, structural and chemical analyses, particle size, and morphology.

2.2.1. XRD

XRD stands as a potent technique for materials characterization, particularly in elucidating the crystallographic properties of various materials, including polymers, composites, and polymer matrix nanocomposites. This method proves instrumental in discerning both crystalline and amorphous phases within polymers and filler materials. Its durability, simplicity, availability of fascinating quantitative data, and nondestructive nature are a few of its main advantages. For instance, the semiconductor sector uses the XRD technique to determine crystal size and orientation, lattice strain, and structural makeup. So, the obtained diffractogram patterns can be understood as the "fingerprints" of the material's crystal arrangements [106]. Another benefit of the XRD technique is that it requires little sample quantity, and the obtained results are readily discussed. Thus, several investigations have been carried out to confirm reinforcements' crystallinity and crystallographic orientations. The level of crystallinity of the magnetic polymer composite, which is reflected in the homogeneous dispersion of the filler within the polymer, is another piece of information that can be obtained from XRD patterns. Sometimes, the availability of reinforcement with different crystal structures might partially or entirely impact the crystallinity of the matrix. Even distribution of the filler will guarantee uniform crystallinity across the matrix. Additionally, XRD patterns can provide insights into the purity of the dispersed filler.

2.2.2. SEM

This technique investigates how electrons and a material's surface interact. A succession of elastic and inelastic electron signals generate the micrographs, revealing morphological, compositional, and structural properties [107]. The topographic details of the sample surface can be observed in the resulting SEM micrographs. This analytical approach allows for evaluating the surface roughness of magnetic polymer composites and the morphology of the magnetic particles dispersed within the matrix (Shekhar et al., 2020). They consider that the conventional SEM characterization procedure requires vacuum conditions and totally-dried materials [108]. So, SEM micrographs reveal morphological properties like particles' shape and size and conditions like aggregation, which may be related to electrostatic contact between nanoparticle layers [109].

2.2.3. TEM

The element that gives a polymer nano-composites the vast majority of its unique features is the filler or reinforcement. Additionally, a potential fracture zone might appear if the fillers are agglomerated. If unnecessary, fillers should be dispersed uniformly throughout the polymer matrix to avoid the fractured area. In this regard, the TEM technique is a practical method for assessing the degree of filler dispersion along the polymer matrix. Besides, TEM provides further significant parameters, like fillers' orientation, size, and crystallinity. These characteristics affect the nanocomposites' performance by giving

Table 2
Overview of fabrication techniques for magnetic polymeric composites, including their merits and demerits [68,104].

Synthesis approach	Brief Description	Merits	Demerits
Molding	A solid material precursor comes into contact with a polymeric stamp.	<ul style="list-style-type: none"> • Replication of precise sizes and aspect ratios • This method facilitates pattern molding. 	<ul style="list-style-type: none"> • The fundamental atomic and molecular granularity of matter constrains its resolution.
Coprecipitation	Reducing a blend of metal ions in a low-temperature, inert atmosphere with a basic solution.	High product yield and purity No requirement for organic solvents Simple replicability Cost-effective	<ul style="list-style-type: none"> • The characteristics of the resulting particles (size, shape, and composition) are strongly influenced by reaction conditions. To stabilize the nanoparticles produced, it's necessary to introduce low-molecular-weight surfactants.
In situ precipitation	Nanoparticles are distributed within a monomer or monomer solution, followed by polymerization using conventional methods.	<ul style="list-style-type: none"> • Superior exfoliation is attained when compared to alternative methods 	<ul style="list-style-type: none"> • Requires appropriate solvents, which may include water and various mixtures
Blending	Polymers are melted with a specified quantity of filler in the presence of an inert gas and elevated temperature.	Straightforward Environmentally conscious Adaptable to industrial processes (suitable for large-scale production and cost-efficient)	Not environmentally friendly <ul style="list-style-type: none"> • There is a possibility of the degradation of the polymer matrix.
Grafting	The dispersion of nanoparticles on the surface of the polymer matrix is instigated through radical polymerization.	Filler covalently attached to the solid surface Precise control of polymer molecular weight Applicability with a broad spectrum of monomers, resulting in diverse functional nanocomposites Effective filler dispersion	<ul style="list-style-type: none"> • Harsh industrial operation conditions • Limited polymerization techniques available

them better or poorer mechanical properties [68].

2.2.4. FTIR

FTIR can detect chemical groups within the composite structure. This method produces a spectrum featuring vibrational bands associated with distinct bonds or functional groups, which can be employed to identify specific chemical compounds. Consequently, as previous studies have indicated, FTIR can be utilized to ascertain the functional groups and composition involved in capping and stabilizing magnetic nanoparticles within polymeric composites [110]. Hence, the intense attraction between Fe and the polymer can serve as evidence for the existence of particular groups such as amides, phenols, nitrogen-containing compounds, and aromatics, among others [111,112].

2.2.5. TGA

TGA studies the average mass loss of the thermostability of particular components. In this sense, the thermal degradation stages at certain temperatures can be identified using the obtained thermograms [113]. Two main thermal degradation types of polymeric composites can be considered: the main chain and the pendant groups (hydroxyl ethyl and amide groups) [114].

2.2.6. Magnetic field

Static or oscillating magnetic fields influence the behavior of magnetic nanoparticles. Fortunately, these magnetic fields possess a substantial penetration depth and serve as a contactless stimulation source. The interplay between the magnetic moments within the material and the magnetic gradient induced by the magnetic field leads to the material's magnetic behavior. [115]. The most often used magnetic NPs include metals, alloys, and metal oxides, with magnetite standing out due to its ease of synthesis, superparamagnetic characteristics, and high biocompatibility [116]. In this context, using polymers as a matrix to support and sustain evenly dispersed nanomagnetic particles to synthesize magnetic nanocomposites is generally advantageous. The nanocomposite's movement or change in shape has been observed to be the magnetic response of polymeric nanocomposites. It is a magnetically separable substance that can be usually used in applications such as drug delivery and purification.

3. Separation processes of MPs/NPs from different wastewaters

The core of the separation process primarily involves extracting minuscule particles of MPs/NPs from the liquid, with their inherent characteristics, such as size and chemical composition, typically undergoing little change during this separation procedure. Various advanced methods for separating solid particles, particularly MPs/NPs, from water sources have been applied. These methods include coagulation/flocculation, adsorption, filtration, flotation, and magnetic separation, as elaborated in the investigations conducted by Zhang et al. [16]. The selection of appropriate technologies and the design of an efficient separation process to eliminate and recycle plastics are crucial factors that depend on the properties of the source materials, effluent, facility capacity, and various other considerations. Specifically, additional separation steps are required to eliminate MPs because of their size, following conventional processes [8].

Magnetic adsorbents, leveraging their magnetic properties, have the potential to mitigate the limitations associated with traditional adsorbents. These materials remain suspended in the solution, facilitating their movement. The magnetic particles within these composite materials demonstrate magnetic separation capabilities, form stable complexes with pollutants, and exhibit chemical stability. [117,118]. Within the magnetic separation procedure, magnetic carrier media can attach to magnetic particles/nanoparticles (MPs/NPs) and imbue them with magnetism, a crucial aspect of magnetic extraction. Efficient magnetic carrier media for removing MPs/NPs include Fe-NPs [119], magnetic sepiolite [120], and micro-submarines [121]. The magnetic separation efficiency remains largely unaffected by the type of plastics used [119]. However, it's worth noting that larger plastics, such as those with a size of 900 μm , are more easily recoverable than smaller ones. Furthermore, an increased concentration of magnetic carrier media enhances separation ability [17].

Conventional magnetic separation usually includes an initial coagulation step followed by a separation process, but equipment costs hinder its widespread use. Broadening the horizons of magnetic separation applications, there is a demand for the creation of inventive and cost-effective magnetic carrier materials and separation infrastructure. [122].

Magnetic separation provides the potential to effectively remove a range of contaminants from water-containing MPs and NPs. As an example, the utilization of three-component magnetic polyoxometalate-supported ionic liquid phases facilitates the magnetic extraction of MPs, microbial organics, and inorganic substances from water. In the case of separating polystyrene (PS) MPs, the presence of small Fe-containing materials on a significant portion of PS beads' surfaces makes them responsive to magnetic separation. This method also efficiently eliminates heavy metals, dyes, and bacteria [123]. Sorbents based on magnetically responsive composites are highly appealing to removing pollutants, thanks to their substantial specific surface area, impressive adsorption capacity, and the added advantage of magnetic properties that facilitate effortless separation from sample solutions or media. Looking ahead, magnetic composites hold great promise when integrated with various materials, such as selective substances like molecularly imprinted polymers. The development of selective magnetic composites not only expands the utility of selective materials for the purification and separation of diverse analytes from complex samples but also streamlines separation processes and elevates method reproducibility. Furthermore, the future outlook for magnetic composites involves their scalable production and application to treat environmental toxic pollutants [124].

3.1. Adsorption onto activated carbons and biochars

Some adsorption studies regarding the efficient removal of MPs using different composites or biochars have been widely reported in the literature. The adsorption process is based on the efficient contact between MPs and adsorbents. For particles smaller than 10 nm, adsorption shows a relatively good efficiency in removing NPs and small MPs. Electrostatic, hydrogen bonds and π - π interactions are essential in adsorption [125].

3.1.1. Magnetic adsorbents

Wang et al. [126] have studied the adsorptive elimination of PS spheres using a magnetic biochar adsorbent synthesized by the impregnation method. High removal efficiencies (98.75%, and 99.46%) were obtained in batch mode with this material. In this case, the biochar played two roles simultaneously due to its Fe, Mg, and Zn content.

The presence of positively charged $\text{Mg}(\text{OH})_2$ and ZnO particles significantly enhances the efficiency of polystyrene (PS) adsorption, primarily attributed to electrostatic interactions and metal-O-PS bond interactions. Also, the active sites of the material showed high catalytic activity towards PS, resulting in the formation of smaller molecules.

Another example is the study accomplished by Tang et al. [127], which used magnetic carbon nanotubes with superparamagnetic characteristics to adsorb PE, polyamide (PA), and polyethylene terephthalate (PET). All the tested MPs were removed entirely within 300 min (with high adsorption capacities); the saturated adsorbents could be readily separated by using a magnet from the aqueous medium due to the magnetic properties of the carbon nanotubes. It was found that the adsorption process was strongly affected by $\text{NH}_3\text{-N}$, COD, and PO_4^{3-} substances.

The influence of dissolved organic matter (DOM) and competitive ions on the adsorption of MPs/NPs exhibits variation across different research studies. The existence of competitive ions or DOM can significantly diminish the sorption of MPs/NPs, primarily due to the specific affinity of these ions and DOM toward the adsorbents [128]. In Fig. 3, the presence of DOM and cations such as Ca^{2+} and Mg^{2+} in the water leads to DOM/NP aggregates forming through electrostatic attraction.

This process impedes the penetration of NPs into the inner or porous structure of the adsorbent (granular activated carbon, GAC). However, the presence of Mg^{2+} and Ca^{2+} is expected to create additional adsorption sites on GAC through bridging effects, thereby leading to an improvement in adsorption efficiency. [129].

Moreover, Singh et al. [125] used Fe-coated-magnetic biochars for the adsorptive removal, in batch mode, of NPs with different surface functionalities. High removal efficiencies ($\sim 100\%$) were obtained in all cases (compared to 75% removal reached for the un-modified material); furthermore, the spectroscopic analysis confirmed that surface complexation played the main role in the NP's adsorption. Additionally, column adsorption experiments were carried out by using Fe_3O_4 -modified biochars for the removal of PS MPs under various chemical conditions, i.e., humic acid, ionic strength, and cationic presence ($\text{Na}^+/\text{Ca}^{2+}$) Wang et al. [130].

Electrostatic adsorption forces and complexation are the main mechanisms involved in this process, finding an optimum retention value of 92.36%. Generally, it could be observed that other chemicals in the effluent negatively impacted the PS removal efficiency. In addition, Shi et al. [131] studied the removal of PS NPs by batch-mode adsorption using magnetic biochars obtained from pinewood sawdust by pyrolysis at three different temperatures (300, 500, and 700°C). The highest removal efficiency of pristine NPs (82.73%) was obtained for the biochar synthesized at 500°C and increased to 95.20% for removing an amine-functionalized NP (NP-NH₂). FTIR and XPS studies suggested that C=O bonds, -COOH, and C-C/C-H played an essential role in the adsorption process due to H-bonding formation, explaining the high adsorption of NPs onto the magnetic biochar. Hydrophobicity, electrostatic attraction, and π - π conjugation mechanism also contributed to adsorption.

In a separate study, a method for the removal of PS and carboxylate-modified PS (CPS) NPs from water involves the utilization of an efficient and reusable composite was reported. This composite is prepared from cetyltrimethylammonium bromide (CTAB)-modified magnetic biochar (CMB). [132]. In this study, the removal efficiency values reached up to 95.2% and 91.2% for PS and CPS, respectively, at pH 7. In this case, aggregation was the primary removal mechanism to remove PS and CPS from water when CMB was used. As water contact angle studies demonstrated, grafting CTAB increased surface hydrophobicity. So, PS/CPS nanoparticles were probably removed and aggregated with the help of CMB, which served as a bridge. Li et al. [133] assessed the removal efficiency of large polyamide microplastics (PA MPs) with varying aging times using magnetic corncob-based biochar (MCCBC) under diverse experimental conditions. The developed material demonstrated high removal performance, achieving an efficiency of approximately 97%, particularly for aged PA MPs across a broad particle size spectrum.

In contrast, the pristine polyamide (PA) exhibited a maximum removal efficiency of only 25% under identical treatment conditions. Observations indicated that tiny PA microplastics were adsorbed onto the surface of the larger magnetic corncob-based biochar (MCCBC) particles, whereas some smaller MCCBC particles adhered to the surface of the prominent PA microplastics. Consequently, the removal of PA by MCCBC involved two mechanisms: firstly, the prominent PA microplastics were magnetized by adsorbing tiny MCCBC particles, and secondly, the extensive MCCBC absorbed the smaller PA microplastics.

Moreover, a magnetic zeolite imidazolate nanocomposite (nano-Fe@ZIF-8) was used for the adsorptive removal of PS microspheres (1.1 μm particle size) from aqueous solution. The adsorption of MPs onto nano-Fe@ZIF-8 was proposed to be attributed to hydrophobic interactions between MPs and adsorbent surface due to the presence (and affinity) of 2-methylimidazole and n-butylamine. A peak removal efficiency of 98% was possible in just 5 min by applying an adsorbent dose of 0.8 g/L [134].

3.1.2. Other non-magnetic adsorbents

Other researchers, such as Bhagat et al. [135], proposed adsorption and aggregation as efficient technologies for removing MPs. Besides, other studies have reported efficient remediation of MPs by adsorption using different materials, i.e., cellulose, lignin-zeolite composites, carbon nanotubes, charcoal, biochars, clays, graphene, etc. [136,137]. Other composite materials, such as chitin-graphene oxide sponges, were

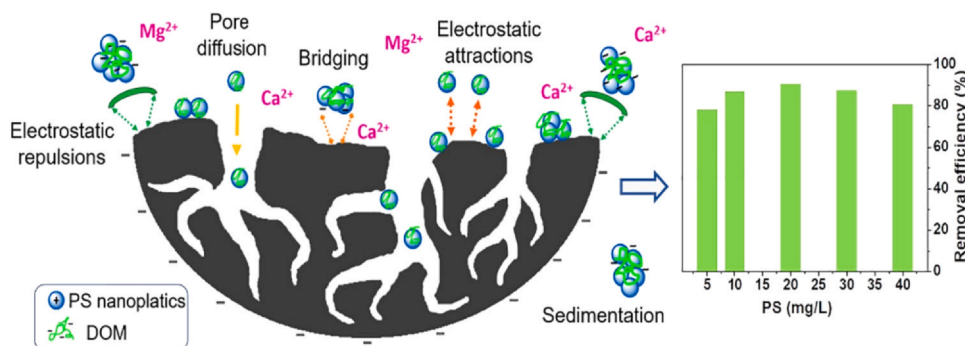


Fig. 3. Suggested adsorption mechanism of polystyrene nanoparticles using activated carbons [129].

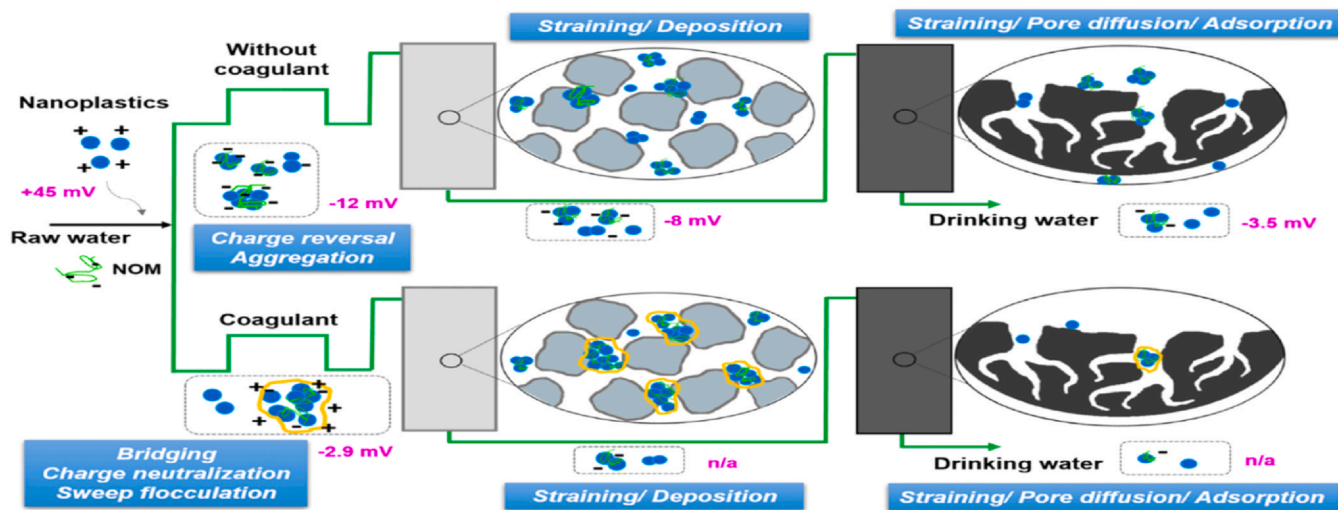


Fig. 4. Mechanisms for eliminating polystyrene nanoparticles (PS NPs) in a typical drinking water treatment plant (DWTP) with and without the coagulation process [146].

efficiently used [138]. Also, Siipola et al. [139] have reported the synthesis of activated carbons by gradual pyrolysis at 475°C from pine and spruce bark, followed by a steam activation at 800°C, which is a reasonably inexpensive synthesis method to create porosity in the material to improve their adsorptive properties. Different shapes of PE were studied, such as spherical PE, cylindrical PE pieces (2–3 mm), and microbeads (10 μm). The cylindrical PE fragments and almost 100% of the fleece shirt fibers were completely removed. Additionally, it has been found that livestock manure-based biochar could degrade poly (hydroxyalkanoate) MPs during composting [140]. Also, an interesting work focused on adsorption in fixed-bed columns using activated biochar from corn straw and hardwood feedstock for the removal of PS microbeads immobilized in the biochar was carried out [141].

The experimental data indicated that the obtained removal efficiencies were close to 100%, showing a much better performance than sand filters (60–80%) due to the morphological and textural properties of the synthesized activated biochars. Some authors synthesized several activated carbons from corncob via physical activation at different temperatures and then were tested in the adsorptive removal of PS NPs. In all cases, the oxidized biochar led to higher adsorption capacities; thus, porous structure, physicochemical properties, i.e., oxygenated surface groups, and electrostatic interactions highly influenced the PS adsorption [142]. Another biochar was synthesized from corncob via physical activation (CCBC) to remove PVC-MPs from water. It is evident that due to its robust adsorption capacity, this biochar effectively removes MPs from water. Furthermore, it can be affirmed that this material has the potential to mitigate the phytotoxic effects of PVC-MPs, as demonstrated in hydroponic experiments involving lettuce [143].

3.2. Coagulation/flocculation

Moreover, coagulation and flocculation processes can be accomplished to remove MPs from water efficiently. Zhou et al. [144] used commercial activated carbon coupled with FeCl₃ to deplete via PS and PE MPs coagulation. High removal efficiencies (~80%) for PS and around 35% for PE were obtained; this difference can be attributed to the higher density of PS. Therefore, the removal of MPs (PS spheres of 1 and 6.3 μm) from a secondary wastewater treatment plant (WWTP) effluent by coagulation/flocculation using inorganic and organic coagulants, i.e., ferric chloride, polyaluminium chloride, and polyamine was studied [145]. High removal efficiencies (~99%) were obtained in this case. It could be observed that parameters such as solution pH, turbidity, humic acid content, MP size, and type of coagulants, among others, highly influenced the process efficiency. Also, an ozonation process integrated with granular activated carbon (GAC) was studied, obtaining an increase of 17.2–22.2% in the MPs removal efficiency [62].

In the context of separating polystyrene nanoparticles within a pilot-scale drinking water treatment plant (DWTP), introducing a coagulation step elevates the removal efficiency from 88.1% achieved through filtration alone to an impressive 99.4%. In-depth investigations reveal that including powdered activated carbon (PAC) noticeably reduces the zeta potential of PS NPs, leading to the formation of larger aggregates through mechanisms like bridging, charge neutralization, and sweep-flocculation. Consequently, the coagulation process enhances NPs retention and streamlines their filtration in the subsequent stages (Fig. 4) [146].

3.3. Magnetic separation and filtration

Magnetic separation can be considered another novel approach for efficiently removing MPs from water. According to Shi et al. [120] study, magnetic separation shows a high capacity, is simple to use, and produces less waste sludge in eliminating MPs/NPs from wastewater. Additionally, combining magnetic separation with other procedures, such as magnetic seeded filtration and adsorption-magnetic separation, can make the MPs/NPs separation process [147]. The prerequisite for magnetic extraction is the ability of magnetic carrier media to connect to MPs/NPs during the magnetic separation and magnetize them. Magnetic sepiolite, hydrophobic Fe-NPs, and magnetic micro-submarines are effective magnetic carrier media for MPs/NPs removal [17,119]. Although this kind of plastic has minimal bearing on the effectiveness of the magnetic separation process, larger plastics (such as those with 900 m diameter) are more accessible to collect than smaller ones (200 m diameter) [17,119]. Additionally, it was determined that a more significant magnetic carrier medium density resulted in a higher separation capacity. In addition, in another study it has been used a high-intensity-magnetic filter (magnetic field strength of 4–8 T) to remove polyvinyl chloride (PVC) MPs, reaching up to 100% removal with the increase in the reaction time [148].

3.4. Other treatments

As has been reported in the literature, advanced final treatment stage technologies, after conventional biological processes, such as sand filters, disc filters, membrane bioreactors, and dissolved air flotation, are capable of removing > 95% of microplastics (> 20 µm); since these units can be scaled up to on-site WWTP [149]. Observations indicate that smaller-sized MPs managed to escape during the membrane bioreactor removal process. This inefficiency in removing MPs, particularly those with a high length-to-width ratio, can be attributed to the occurrence of membrane biofouling. Moreover, a disc filter with 18 µm pore size could remove the MPs > 10 µm with high efficiency, c.a. 89.7%. It was observed that the removal efficiency decreased for the prominent-size MPs due to the clogging of the filter pores [150]. Therefore, Mintening et al. [151] reported the use of a tertiary post-filtration unit to remove > 500 µm of MPs and 95% of 20–500 µm MPs; in the same way, Ziajahromi et al. [152] used an ultrafiltration process after reverse osmosis for the removal of MPs. Thus, electrospun nanofiber membranes that eliminate different MPs from aqueous media have been studied [153]. This work reported high removal efficiencies (> 99%) with low energy requirements. In addition, biofilters, a low-cost technology, have been established as an efficient technology to remove MPs and NPs [154,155]. Finally, Bai et al. [156] used a porous composite membrane (prepared with 4-benzenedithiol and 1,3,5-triethynylbenzene) as material to remove PS fluorescent microspheres (0.1 µm diameter). In this case, the filtration efficiency was 90.92%. Also, as reported in the literature [157], the removal efficiency achieved with filtration can be enhanced using solar energy.

This way, the conventional activated sludge treatment was seen as very effective, with an average removal efficiency of 98.3%. In contrast, this method generates a large amount of sludge, as has been reported [158]. The dissolved air flotation process is another technology in which air is liquefied at high pressure, generating bubbles that favor the flotation of smaller molecules, this is, low-density MPs; in this sense, the addition of polyaluminium chloride allowed an MPs removal value of 95% [149]. Moreover, the extraction process using ferrofluids mixed with iron-coated nanoparticles, which are super magnetic particles with high sorption ability and large surface-volume ratio, has been tested for the efficient removal of PET [159]. The PET removal efficiency by using this method decreased up to 64% when actual wastewater was treated.

Another alternative, non-considered as chemical, of separation of MPs from the aqueous medium is their deposition onto aquatic biota. Gopalakrishnan et al. [160] conducted a study involving the cultivation

of *Chlamydomonas* algae with varying concentrations of MPs (0–0.4 mg/mL) and temperatures ranging from 2.5 to 32.5°C. The aim was to comprehend the deposition dynamics and assess the impact of MPs on extracellular polymeric substances (EPS) production and algal density. The findings revealed an increase in algal cell density and MP deposition, particularly when the MP concentration reached up to 0.3 mg/mL. Additionally, this kind of enzymatic degradation of MPs by using different systems of particle-enzyme/polymer or microorganism complexes has also been studied by Tang et al. [127].

4. Degradation of MPs/NPs from different wastewaters

Notably, the degradation of NPs exhibits more significant promise than MPs, as the reactivity with hydroxyl radicals significantly increases with larger plastic surface areas. Three tested processes for the degradation of both MPs and NPs include catalytic degradation, photocatalytic degradation [161], and electrochemical degradation [15].

The predominant methods for breaking down MPs and nanoparticles primarily encompass AOPs, electrochemical degradation, photodegradation, and biological photocatalytic degradation. Electrochemical degradation and AOPs are distinguished by their capacity to achieve a relatively rapid degradation rate among these techniques. Meanwhile, biological degradation, known for its low energy consumption, has been the subject of extensive research for an extended period [162].

Numerous magnetic composite catalysts exhibit the potential to function as micro-scale motors or robots for the degradation of MPs/NPs. These catalyst systems have notable benefits, such as enabling efficient interactions between photocatalysts and MPs/NPs and requiring minimal energy consumption [163]. When subjected to sunlight, a composite catalyst containing magnetic (Fe₃O₄) and photocatalytic (BiVO₄) nanomaterials demonstrated autonomous mobility, marked by precise control using a magnetic field within macro-channels. Paired with the exceptional photocatalytic capabilities of BiVO₄, this micro-robotic catalyst effectively degrades MPs in intricate, confined environments [163].

4.1. Photodegradation

Prolonged exposure to light, particularly ultraviolet (UV) light, plays a significant role in initiating the degradation and transformation of microplastics/nanoplastics through the action of reactive oxygen species (ROS) and other active substances in nature [164]. The photodegradation of microplastics/nanoplastics can expedite chemical processes in aquatic environments. In particular, photo-degraded polystyrene microplastics can serve as photosensitizers in the solution, enhancing the photodegradation of organic pollutants by generating ROS and other active agents (e.g., carboxyl, oxygen, and polystyrene) [165]. Fig. 5 illustrates that the photodegradation of microplastics/nanoplastics typically results in surface oxidation, heightened hydrophilicity, reduced size, structural imperfections, the formation of organic compounds and gases like CO₂, as well as the persistence of free radicals, among other effects [165,166].

4.2. Photocatalytic degradation

Photocatalysis represents an environmentally friendly and energy-efficient technique for decomposing micro-/nanoplastics [167]. When exposed to appropriate light sources, photocatalysts or semiconductors produce excited electrons (e⁻) and holes (h⁺) [168, p.]. In the photocatalytic degradation of MPs/NPs, the highly oxidizing photogenerated h⁺ can directly trigger the oxidation of plastics. Moreover, the interaction of e⁻ and h⁺ with water, OH⁻, and oxygen leads to the formation of specific free radicals, which further contribute to plastic decomposition [169]. Various photocatalysts, including titanium, zinc, tin, bismuth, and copper-based oxides/sulfides, are commonly employed in

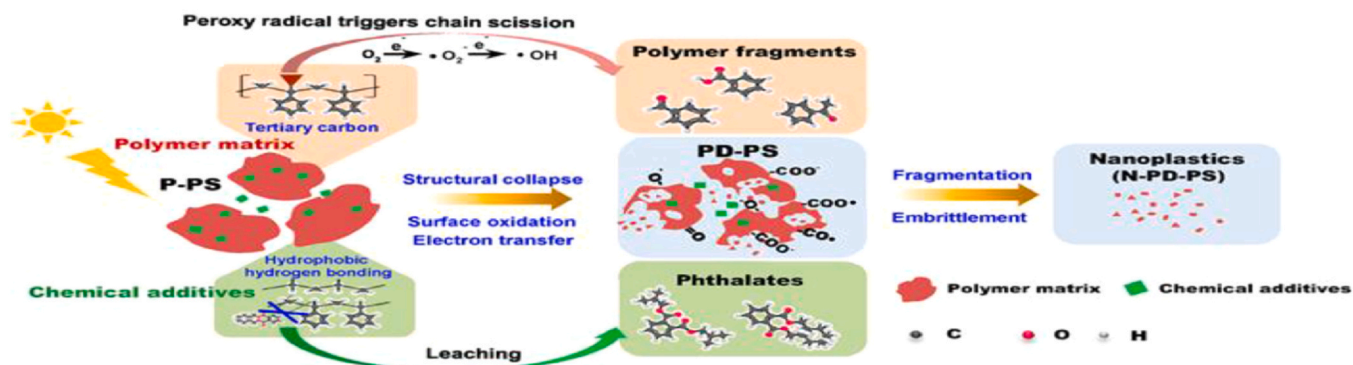
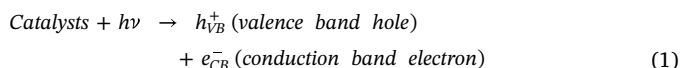


Fig. 5. Schematic representation of the photodegradation mechanism of microplastics (MPs) and nanoparticles (NPs) (P-PS: pristine polystyrene fragments; PD-PS: photodegraded polystyrene fragments; N-PD-PS: nanosized) [165].

the degradation of MPs/NPs from water [169], and the physicochemical properties of these catalysts significantly influence their catalytic performance.

Enhancing degradation efficiency can be achieved by modifying the chemical composition of titanium dioxide through doping, regardless of controlling the nanostructure. For instance, silver-doped titanium dioxide [170] and carbon, nitrogen-doped TiO_2 [171] have demonstrated effective performance in the degradation of MPs/NPs.

The photocatalytic degradation of MPs/NPs is influenced by various significant factors, encompassing the plastics' characteristics and the pH level. Plastics with smaller dimensions and darker pigmentation are prone to degradation because of their elevated specific surface area and enhanced light-absorbing capabilities, respectively [172]. Moreover, film-shaped MPs' degradation efficiency is lower than MP beads, primarily due to their limited oxygenation and reduced exposure to light [173]. A general photocatalytic degradation mechanism can be described as follows:



Thus, the research of Nakatani et al. [174] focused on the removal of polypropylene (PP) films through photocatalytic degradation. This process involved the use of a poly(styrene-block-acrylic acid, PS-*b*-PAA) matrix containing TiO_2 gel with a copper phthalocyanine catalyst under visible light irradiation. Notably, this method can utilize sunlight, thereby reducing the overall cost of photocatalysts and increasing their eco-friendly characteristics. The study results indicated that PP degradation occurred through chain scission, initiated by light irradiation or OH^* radical attack. Autooxidation produced hydroperoxide and carbonyl compounds, which served as degradation markers. The exfoliation of the PP surface appeared to result from a synergy between autooxidation and water, with water playing a significant role in surface exfoliation.

In the study by [175], the research delved into the photocatalytic degradation of polyethylene (PE) powder and polystyrene (PS) microspheres using TiO_2 -based films. These films were designed to decompose MPs directly under UV light irradiation, effectively avoiding the release of toxic intermediate compounds into aquatic environments. The results of this study indicated that the degradation predominantly occurred at the PS-air interface within 3 and 6 h of irradiation. In conclusion, the research revealed the pivotal roles of h^+ , OH^* , and

oxygen groups in the photodegradation of PS, with h^+ emerging as the dominant active species in the reaction.

Also, the photocatalytic degradation of polyethylene terephthalate (PET) using an MXene/ $\text{Zn}_x\text{Cd}_{1-x}\text{S}$ photocatalyst was studied by Cao et al. [176]. The catalyst synthesis involved Ti_3AlC_2 MAX powder and using the solvothermal preparation method. This work followed the photocatalytic H_2 generation, a helpful product coupled with MP degradation, under UV-light irradiation. As stated in the previous work, an excessive load of MXene might effectively block the absorption of visible light from the catalyst. It has been reported that only low molecular weight organic compounds, i.e., formate, methanol, acetic acid, and ethanol, were obtained after the reaction. The results confirmed that the hole in the valence band of $\text{Z}_x\text{C}_{1-x}\text{S}$ was the active species directly involved in PET degradation.

4.3. pAOPs

AOPs that utilize hydroxyl, chloride, and sulfate as their primary oxidizing agents have gained extensive use in wastewater treatment. They are esteemed for their exceptional ability to transform organic pollutants into harmless end-products [177]. Recently, there has been a significant surge in research focus on Fenton reactions and persulfate-based systems, driven by their outstanding efficiency, ease of application, and environmentally friendly characteristics, as explained further below.

4.3.1. Ozone-based systems

Ozone (O_3), known for its reactivity and oxidizing solid properties, has been widely employed in water treatment for an extended period [178]. Ozone serves as a discerning oxidizing agent, primarily aiming for electron-rich functional. Ozonation generally falls under the AOP category, as it produces carboxyl ($-\text{COOH}$) groups during its application [179]. Recent research has explored the efficacy of ozone treatment in degrading MPs/NPs. Compared to chlorination, O_3 proves to be more efficient in degrading higher molecular weight (MW) PS NPs, achieving a degradation efficiency of 96.3% in just 30 min, as opposed to 4.2% for chlorination [178]. The utilization of a combination of O_3 with ultraviolet (UV/ O_3) and hydrogen peroxide (H_2O_2) has the potential to enhance the degradation of MPs and NPs. In the UV/ O_3 system, the sequential application of ozone and UV has demonstrated superior efficacy in degrading polyethylene (PE) MPs when compared to the conventional simultaneous use of O_3 and UV. This sequential approach augments carbonyl ($\text{C}=\text{O}$) and vinyl ($-\text{CH}=\text{CH}_2$) groups while reducing hydrophobicity [180].

4.3.2. Fenton reaction-based systems

Fenton oxidation leads to significant changes in the physicochemical characteristics of MPs and NPs. As shown in the atomic force microscopy images (Fig. 6a), the MPs subjected to post-oxidation display a

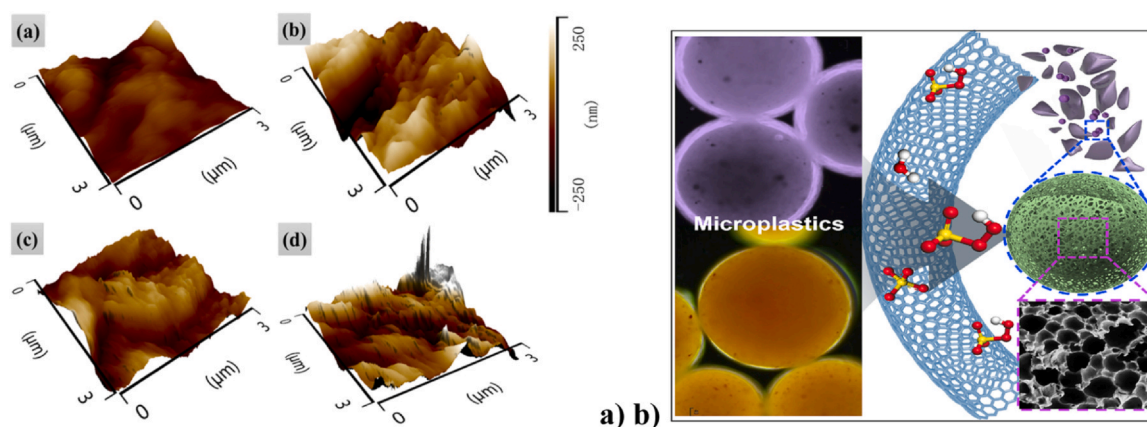


Fig. 6. (a) Atomic Force Microscopy (AFM) topographic representations of unaltered and oxidized microplastics (MPs) [181] utilizing Fenton reaction-based systems. (b) Illustration depicting the degradation process of cosmetic MPs via the integrated carbon/HT/persulfate system [182].

rougher surface with more pronounced irregular domains [181]. Additionally, the carbon catalyst utilized enhances the degradation of MPs, achieving an impressive degradation efficiency of 54% within 8 h by activating peroxymonosulfate to generate reactive radicals (Fig. 6.b) [182].

Fenton or Fenton-like reactions can generate carboxyl groups for the breakdown of various pollutants, including organic compounds and MPs/NPs. These reactions offer the advantages of being straightforward, highly effective, and environmentally friendly [183]. Fenton oxidation induces noteworthy changes in the physicochemical properties of MPs and NPs. Approaches combining Fenton reactions, such as electro-Fenton-like system [184], thermal Fenton system [185], and bio-photo-Fenton system [186], have demonstrated enhanced degradation efficiency against MPs/NPs. In the bio-photo-Fenton system, the impact of UV radiation results in a substantially higher production of butanoic acid and acetic acid from sulfonated polyethylene when compared to the photo-Fenton system, exhibiting a 17-fold and 21-fold increase, respectively [186].

4.3.3. Persulfate-based systems

AOPs reliant on sulfate radicals have received significant interest because of their elevated redox potential, swift pollutant degradation, and extended half-life [187]. This approach also yields valuable low-molecular-weight organic compounds, including alcohols, potentially motivating further exploration of repurposing waste plastics. In the catalyst/persulfate system, an integrated approach that combines hydrothermal hydrolysis and carbocatalytic peroxymonosulfate oxidation known as the integrated carbon/HT/persulfate method, was introduced for the degradation of cosmetic microplastics. The utilization of the carbon catalyst markedly enhances the efficiency of microplastic degradation, achieving a degradation rate of over 50% in a mere 8 h [182].

Several techniques have been documented, including heat, catalysts, and UV light activation of persulfate. Utilizing a UV/persulfate process, PVC MPs can be effectively broken down, achieving a dechlorination rate of approximately 58/L Cl^- within a 35 h treatment period. Additionally, the existence of sulfate radicals and reactive oxygen species indicates the fragmentation of the polymer structure and the formation of oxygen-containing functional groups [188].

5. Application of magnetic polymeric composites for micro/nanoplastics separation and degradation

This part discusses recent applications of magnetic polymeric composites for MP/NP separation and degradation processes, and some representatives are showcased in Table 3. According to Urso et al., it was reported that γ - Fe_2O_3 /Pt/ TiO_2 microrobots were able to catch 97%

of PS-COOH nanoparticles (50 nm) in their study. In contrast, MXene microparticles achieved a lower capture efficiency of only 50% within 5 min. The higher NP capture rate (97%) observed with γ - Fe_2O_3 /Pt/ TiO_2 microrobots, compared to MXene microparticles (50%), can be attributed to the stronger electrostatic interaction and the multi-layer stacks morphology exhibited by the former [189].

Sarcelletti and colleagues have created core-shell super-paramagnetic iron oxide (SPION) NPs with different functionalizations to eliminate MPs of various sizes from aqueous media efficiently. They demonstrated that the SPIONs adsorbed onto the surface of MPs, acting as a binding agent. Importantly, the functionalized SPIONs could eliminate MPs by utilizing bonds and interactions between the microplastics and the SPIONs. Furthermore, the SPION systems exhibited high elimination capabilities even in a mixture of melamine resin (MR) and PS. The functionalized SPION nanoparticles demonstrated a greater capacity for capturing MPs compared to nano- Fe_3O_4 particles. The SPION systems showed improved performance in removing MPs from surface water [190]. Grbic et al. proposed a magnetic extraction method for separating plastics, utilizing the hydrophobic nature of plastics to magnetize them. They synthesized hydrophobic iron NPs that selectively bind to plastic, enabling magnetic retrieval. By applying this principle to a simple method, they achieved a recovery rate of 92% for polyethylene and polystyrene beads with sizes ranging from 10–20 μm and 93% for larger MPs (> 1 mm), including polyethylene, polyethylene terephthalate, polypropylene, polyvinyl chloride, polyurethane, and polystyrene, from seawater. Additionally, they recovered 84% of microplastics with sizes between 200 μm and 1 mm from freshwater and 78% from sediments [119].

In another study, a magnetic magnesium hydroxide coagulant (MMHC) was developed to efficiently eliminate polyethylene from wastewater, a prominent component of microplastics that tends to float on the water surface. The production of MMHC involved incorporating Fe_3O_4 magnetic particles during the formation of $Mg(OH)_2$. The performance of MMHC in microplastics elimination was compared to that of the non-magnetic version. Three different types of MMHCs were synthesized by adjusting the ratio of Mg^{2+} to OH^- . The findings revealed that among the three types of MMHC, the highest microplastics elimination efficiency of 87.1% was achieved when the Mg^{2+} : OH^- ratio was 1:1, which was 14.7% higher than that of MHC alone. Notably, the elimination efficiency of microplastics remained relatively stable within the pH range of 5–9, indicating charge neutralization during the coagulation process [191]. Misra et al. introduced an innovative method for water purification, involving utilizing a magnetic nanoparticle composite to eliminate various types of pollutants such as organic compounds, inorganic substances, microbes, and microplastics. The composite comprises magnetic microporous core-shell Fe_2O_3 / SiO_2 particles coated with a polyoxometalate ionic liquid resulting in the

Table 3
Application of magnetic polymeric composites for MPs/NPs removal and degradation from different water sources.

A/A	Type of Wastewaters	Adsorbents	Polymers/M/NPs	Co-presence pollutants	Physicochemical Properties*	Removal efficiency (%) / Adsorption capacity (mg/g)	Removal Technology	Mechanism	Highlights	References
1	Kitchen wastewater	M – CNTs.	MPs PE, PET PA (48 µm)	COD (0–5 g/L), NH ₃ -N (0–0.2 mg/L), PO ₄ ³⁻ (0–1.2 g/L)	SSA: 197 m ² /g before and ~198 m ² /g after MPs elimination. TPV: 0.141 cm ³ /g before and 0.147 after MPs elimination. PS ~2.86 nm	100%/ 1650 mg/g (PE) 100%/ 1400 mg/g (PET) 100%/1100 (PA)	Physical Method/ Adsorption/ Separation Process	Combination of π-π electron interaction, complexation, electrostatic interaction, and hydrogen-bond interaction; PSO/ F interaction; PSO/ F Combination of electrostatic interaction and chemical bonding interaction; PSO/ Freundlich	High removal efficiency of 100% Magnetic carbon nanotubes can be reused repeatedly. Elimination rate decreases with number of applications	[127]
2	Tap water, river water, wastewater	MBC, Mg-MBC, Zn-MBC	MPs PS (1 µm)	Cl ⁻ NO ₃ ⁻ HCO ₃ ⁻ SO ₄ ²⁻	SSA: 363.8, 265.5, 329.9 TPV: 0.37, 0.41, 0.34 PS: 3.81, 3.83, 3.83	94.81%/ 100.6 mg/g (MBC) 98.75%/ 98.52 mg/g (Mg-MBC) 99.46%/ 99.21 mg/g (Zn-MBC)	Physical Method/ Adsorption-Thermal treatment/ Degradation process	Combination of electrostatic interaction and hydrophobic interaction;	The anchoring of metals and aging of biochar significantly improved the MPs and NPs adsorption. Zn-MBC demonstrated superior removal effectiveness (over 97.5%) at acidic and neutral pH levels.	[126]
3	Artificial sea water	Nano-Fe ₃ O ₄	MPs PE, PP, PET (200–900 µm)	NaCl (26.7 g/L), MgSO ₄ (3.2 g/L), MgCl ₂ (2.2 g/L), CaCl ₂ (1.1 g/L), KCl (0.7 g/L).	-	> 80 from real waters	Physical Method/ magnetization treatment/Separation Process	Combination of surface adsorption: electrostatic interaction and hydrophobic interaction;	The established method could efficiently eliminate MPs from river water, sewage and natural seawater with the elimination rate of higher than 80%. Extensive validation and extensive implementation of this approach are required, particularly across various types of aqueous media and for a broad spectrum of MP sizes and polymers.	[17]
4	Freshwater, groundwater, tap-water	Iron-modified biochar FB-550, FB-580	M/NPs PS-COOH, PS- NH ₂	NO ₃ ⁻ SO ₄ ²⁻ Cl ⁻ CO ₃ ²⁻	FB-550 = 25.8 m ² /g and FB-850 = 34.5 m ² /g.	99.5%/ 206 mg/g	Physical method/ magnetic extraction/ Separation Process	Combination of Electrostatic interaction, π-π conjugation and hydrophobicity; GO/ S	These FB composites have shown efficient elimination of NPs of different dimensions and functionalities. The amine-modified NPs particle was eliminated quicker with higher adsorption capacity than carboxylate-modified nanoparticles.	[125]
5	Simulated wastewater with PVC, PET, PS and ABS	Fe-kaolin	MPs PVC, PS, PET, ABS	Cl ⁻ : 0.2 g/L	SSA:7.5360, PV:0.031363, S:16.6471		Distinguishing between chemical adsorption and physical adsorption of microplastics by the modified kaolin materials posed a challenge in this study.	Electrostatic interaction; PFO/ F	Co/Mn-kaolin exhibited a higher adsorptive capacity (22 mg/g) compared to Fe-kaolin (~13.7 mg/g), primarily attributed to its larger surface area and presence of pores. It should be noted that the adsorption efficiency of microplastics using the modified materials can be further enhanced by optimizing certain external factors.	[193]
6	Wastewater	γ-Fe ₂ O ₃ /Pv/TiO ₂ microrobots Mxene microparticles	NPs PS-COOH	-	Specific Surface Area: 6.8 m ² g ⁻¹	0.5 ± 0.1 mg g ⁻¹ 50%(Mxene) – 97% (microbots) within 1 min	Physical Method/ thermal annealing /degradation + UV-light irradiation, magnetic collection.	Electrostatic interaction, negative photogravitaxis	This study introduces a novel approach for the removal and detection of nanoplastics in water, utilizing self-propelled light- powered magnetic microbots derived from MXene. It is noteworthy that the γ-Fe ₂ O ₃ /Pv/ TiO ₂ microbots exhibited a higher nanoplastic capture efficiency (97%) as opposed to MXene microparticles (50%), mainly attributed to their intense electrostatic interaction and morphology.	[189]

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Table 3 (continued)

A/A	Type of Wastewaters	Adsorbents	Polymers/M/NPs	Co-presence pollutants	Physicochemical Properties ^a	Removal efficiency (%) / Adsorption capacity (mg/g)	Removal Technology	Mechanism	Highlights	References
7	River water	SPIOs- PAC ₁₈	MPs PS, PMMA, and MR	-	-	1-2.25 MR 0.2-0.35 PS 0.2-0.3 PMMA 1.75-2.25 MR/PS blend	Physical Method/ Magnetic extraction	Combination of van- der-Waals interaction, electrostatic interactions, and hydrophobic attractions;	The functionalized superparamagnetic iron oxide nanoparticles (SPION) were able to effectively remove microplastics (MPs) owing to van der Waals forces, electrostatic interactions, and hydrophobic attractions between the microplastics and the SPION. It is anticipated that the surface properties of the spilled nanoparticles (NP) would alter their chemical fate, subsequently influencing the interaction between SPION and NP. The hydrophobic iron (Fe) nanoparticles, which can adhere to plastic surfaces, were utilized in magnetic recovery experiments to achieve high recovery rates for different sizes of microplastics (MPs) tested: large (greater than 1 mm; 93%), medium (200 µm to 1 mm; 81%), and small (less than 20 µm; 92%). It should be noted that the concept was evaluated under restricted conditions, and the method used for extracting the microplastics was relatively basic compared to using a more sophisticated engineered apparatus.	[190]
8	Seawater, freshwater, Sediments	Hydrophobic Fe nanoparticles	M/NPs PE, PET, PS, PUR, PVC, and PP	-	-	92-93 (artificial seawater) 84 (freshwater) 78 (sediment)	physical Method/ Magnetic extraction by hydrophobic Fe nanoparticles/ separation process	Hydrophobic interactions	The hydrophobic iron (Fe) nanoparticles, which can adhere to plastic surfaces, were utilized in magnetic recovery experiments to achieve high recovery rates for different sizes of microplastics (MPs) tested: large (greater than 1 mm; 93%), medium (200 µm to 1 mm; 81%), and small (less than 20 µm; 92%). It should be noted that the concept was evaluated under restricted conditions, and the method used for extracting the microplastics was relatively basic compared to using a more sophisticated engineered apparatus.	[119]
9	Simulated wastewater with humic acid and kaolin	Magnetic magnesium hydroxide coagulant (MMHC)	MPs PE	Humic acid and Kaolin	-	87.1% 92% (PAM)	Chemical Method/ Coagulation/ Separation Process	Electrostatic interactions	Among the three variants of modified metal hydroxide coagulants (MMHC), the highest removal efficiency of microplastics was achieved when the Mg ²⁺ : OH ⁻ ratio was 1:1, reaching 87.1%. This percentage was 14.7% higher compared to using conventional metal hydroxide coagulant (MHC) alone. The process of microplastics removal involves charge neutralization, which serves as a significant mechanism in coagulation. The magPOM-SiLP composite demonstrates the ability to effectively eliminate various types of pollutants, including organic compounds, inorganic substances, microbes, and microplastics, through a selection of targeted removal mechanisms. By optimizing the individual components, it is possible to enhance the capacity of these systems and explore their integration with electromagnetic recovery systems, thereby enabling their application under more realistic operational conditions.	[191]
10	Commercial colloidal solutions	Magnetic microporous core-shell Fe ₂ O ₃ /SiO ₂ particles	MPs PS (1, 10 µm)	-	Size:1 & 10 µm, SSA: ~270 m ² g ⁻¹ , PV:0.97 cm ³ /g;	100%	Physical Method/ Magnetic extraction by polyoxometalate- supported ionic liquid/separation process	Hydrophobic interactions	The magPOM-SiLP composite demonstrates the ability to effectively eliminate various types of pollutants, including organic compounds, inorganic substances, microbes, and microplastics, through a selection of targeted removal mechanisms. By optimizing the individual components, it is possible to enhance the capacity of these systems and explore their integration with electromagnetic recovery systems, thereby enabling their application under more realistic operational conditions.	[123]
11	Simulated wastewater	Functionalized carbon nano springs (Mn@ NCNTs/PMS) degrade MPs	MPs PE	-	SSA:217.1 m ² g ⁻¹	50 wt% of MPs removal	PhotoCatalytic degradation/ Degradation Process	catalytic activation of peroxymonosulfate	The Mn@NCNTs/PMS system exhibits the capability to achieve a 50 wt% elimination of microplastics (MPs) by aiding in the hydrolysis. Specifically, polyethylene-based MPs were effectively decomposed through the integrated carbon/HT/persulfate system. Additional investigation is required to assess the practical applicability of the AOPs technique for controlling MPs in real-life water matrices.	[182]

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Table 3 (continued)

A/A	Type of Wastewaters	Adsorbents	Polymers/M/NPs	Co-presence pollutants	Physicochemical Properties ^a	Removal efficiency (%) / Adsorption capacity (mg/g)	Removal Technology	Mechanism	Highlights	References
12	Wastewater	Fe ₂ O ₃ -MnO ₂ core-shell micromotors	MPS PE 0.8 μm	-	SSA: Fe ₃ O ₄ -MnO ₂ and Fe ₂ O ₃ -MnO ₂ micromotors are 63.2 and 71.3 m ² g ⁻¹ , respectively, volume: 0.29 cm ³ g ⁻¹ ,	up to 90% of organic pollutants, as well as more than 10% of the suspended microparticles in a few hours	hydrothermal ABS mechanism and catalytic degradation	Combination of synergy of catalytic degradation, surface adsorption, and adsorptive bubbles separations;	The micromotors based on magnetic MnO ₂ demonstrate the ability to effectively remove organic dyes and separate microplastics from contaminated water. Specifically, the ABS mechanism of the micromotors facilitated the removal of up to 90% of organic contaminants and more than 10% of the microparticles within a few hours.	[192]
13	Three distinct solutions, each consisting of 100 mL of E2, were prepared.	graphene-like magnetic biochar (GLMB)	MPS 17β-estradiol	Ionic strength and the presence of SO ₄ ²⁻ ions on the adsorption of estradiol was investigated. Additionally, the impact of humic acid and various background electrolytes (including Na ⁺ , Mg ²⁺ , Cl ⁻ , NO ₃ ⁻) was also examined.	SSA: ~ 828 m ² /g	98.73	physical adsorption	π-π interactions, micropore filling effects, electrostatic interaction, pseudo-second-order kinetics and Langmuir isotherm models	The adsorption capacity of E2 onto GLMB was found to be influenced by multiple factors, encompassing the solution's pH, ionic strength, and the presence of SO ₄ ²⁻ ions.	[194]
14	Synthetic water/ Surface water/ Wastewater	chitosan-based multilayer adsorbent (FCS-PD)	MPS PVC, PE on the FCS and FCS-PD	Various cations (Na ⁺ , K ⁺ etc) and anions (Cl ⁻ , NO ₃ ⁻ etc) and Diclofenac	SSA: 1.4 m ² /g, PS: 3.3 nm and PV of FCS-PD 0.01 cm ³ /g	434.78 mg/g	Physical method/ Adsorption process/ Separation process	chemisorption and intraparticle diffusion, which was emphasized by the pseudo-second-order kinetic and intra-particle diffusion analysis	Under neutral conditions, the maximum adsorption capacity of FCS-PD was determined to be approximately 434 mg/g. Fe ₃ O ₄ exhibited significantly lower adsorption capacities for all six pharmaceuticals. Notably, FCS-PD exhibited exceptional adsorption capacities specifically for DCF, surpassing the adsorption capacities of the other pharmaceuticals tested.	[195]

^a SSA: Specific Surface area, TPV: Total Pore volume, PS: Pore size

formation of a magnetic POM-supported ionic liquid phase (magPOM-SILP) [123].

Kang et al. discovered that persulfate oxidation can efficiently contribute to eliminating MPs/NPs present in water contaminated with facial cleanser. Experimental findings demonstrated that when utilizing a catalyst concentration of 0.2 g/L (manganese carbide nanoparticles), peroxymonosulfate solution concentration of 6.5 mM, and starting microplastic/nanoplastic concentration of 5 g/L, a degradation rate of 40% was achieved after an 8 h catalytic reaction at 120 °C. This study expands the application of AOPs and offers a viable strategy for extracting microplastics from water. Incorporating transition metal (Mn) encapsulation and utilizing a robust helical structure in the Mn@NCNTs catalyst considerably enhance its catalytic performance and stability in the degradation of microplastics, accompanied by a notable reduction in activation energy. The findings suggest that the Mn@NCNTs/PMS system can achieve a 50 wt% elimination of microplastics by facilitating hydrolysis [182].

Ye et al. conducted a study showcasing the efficiency of two bubble-propelled micromotors with a core-shell structure consisting of iron oxide and manganese dioxide ($\text{Fe}_3\text{O}_4\text{-MnO}_2$ and $\text{Fe}_2\text{O}_3\text{-MnO}_2$) in eliminating pollutants. The $\text{Fe}_2\text{O}_3\text{-MnO}_2$ micromotors, in particular, demonstrated efficient elimination of water-based organic contaminants and suspended microplastics through a combination of degradation, surface adsorption, and bubble separation methods. The latter method effectively removed over 10% of microplastics from sewage within a 2 h timeframe [192].

5.1. Co-presence of M/NPs and ions in different water sources

Tang et al. investigated magnetic carbon nanotubes (M-CNTs) to eliminate polyethylene terephthalate (PET), polyamide (PA) and polyethylene, (PE). They efficiently adsorbed the microplastics, which were subsequently separated through magnetic means. A dosage of approximately 5 g/L of M-CNTs eliminated all microplastics within a 5 h timeframe. The maximum capacities observed for PE, PA, and PET were approximately 1650, 1100, and 1400 mg/g, respectively. The elimination rate for PE, PET, and PA was 100%. Factors such as chemical oxygen demand (COD), ammonia (NH_3) and phosphate (PO_4^{3-}), had no significant effect on the adsorption of MPs. The adsorbent material demonstrated the ability to undergo thermal regeneration while retaining its properties, similar to the original M-CNTs (magnetic carbon nanotubes). Even after four reuse cycles, it maintained an 80% removal efficiency. The hydrophobic interactions explained the sorption of PE and PET, while for PA and PET, it involved π - π interactions. Carbon-based materials should be able to effectively remove various types of MPs in water resembling real-world MP-contaminated environments. The specific surface area of the adsorbent is crucial in determining its adsorption behavior, in this case being approximately 197 m²/g. Following the adsorption of MPs, the SSA and pore volume became slightly larger, indicating that these parameters remained largely unaffected by the adsorption process. The findings revealed that the MPs' surface became rougher due to the adsorption and agglomeration of M-CNT particles on a significant segment of the MPs' surfaces. [127].

The same year Wang et al. proposed magnetic biochars (MBC) to eliminate MPs due to the simple synthesis process, which involves incorporating magnetic components into biochar. MBC offer advantages such as easy removal after adsorption, higher adsorption capacity, and low toxicity. Mg/Zn-modified MBCs were used for the elimination of microplastics. MBC, Mg-MBC, and Zn-MBC demonstrated elimination rates of approximately 94.8%, 98.7%, and 99.4%, respectively for PS microspheres. They also exhibited significantly higher mesoporous surface areas than regular biochar, resulting in improved contact efficiency. The elimination rate of biochar was only 25.8%, nearly three times lower than that of the magnetic versions, highlighting the crucial role of the metals in enhancing their efficiency. Notably, the elimination rates of Mg and Zn-based biochars surpassed those of M-CNTs,

which are well-known for their excellent microplastic elimination capabilities. Characterization studies revealed that the metal-modified biochars were coated with Fe, Mg, and Zn particles. These particles were presumed to capture MPs on the BC surface and strengthen the attachment through interactions with the MPs. The introduction of metal modifications to BC resulted in a significant reduction in its specific surface area, primarily due to the decrease in the proportion of micropores. This reduction was attributed to the formation of metal oxide NPs, which caused the collapse of pore walls, converting micropores into mesopores. [126].

Shi et al. reported that Fe_3O_4 NPs have proven efficient in magnetizing diverse categories of microplastics via surface absorption. The focus was on magnetizing microplastics of various sizes and types. A magnet was employed to separate the magnetized MPs, and the elimination rate varied when pure water was used. The elimination efficiency for large and small sizes was higher than medium sizes, although this characteristic was not evident in the elimination of PET MPs. Furthermore, PET's elimination rate was lower than other types of microplastics. The elimination rates exhibited minimal variation among MPs with different shapes but the same polymer composition. The elimination rates of MPs were recorded as approximately 81.3% in river water, 82.2% in domestic sewage, and 80.5% in seawater [17].

Singh et al. found that Fe-modified biochar produced through pyrolysis at two different temperatures, 550 °C (FB-550) and 850 °C (FB-850), demonstrated exceptional efficiency in removing PS-based microplastics (1 μm) in less than 10 min. It is important to highlight that FB-850 exhibited a higher sorption capacity for PS-COOH compared to PS-NH₂. The interaction between adsorbent and adsorbate primarily occurred through an acid-base reaction. On the other hand, FB-550 demonstrated greater sorption of PS-NH₂. Regarding nanoparticles, FB-850 exhibited better removal rates of PS-COOH nanoparticles (30 nm) than FB-550. The pH of the solution did not considerably affect the elimination process in pH range 3.5–9.5. However, the same NPs were eliminated more under low pH conditions. Interfering ions had no impact on the removal of PS-NH₂ microplastics by both the modified biochars. The sorption of microplastics (1 μm) was worse in freshwater, and best in groundwater, because of the higher ionic strength of groundwater. However, groundwater had a higher natural organic matter content [125].

The role of iron-modified adsorbents is discussed by Huang et al. Moreover, the inclusion of various metals, minerals, and acids has been explored to impart hydrophilic or hydrophobic attributes and improve reactivity on the surface of the adsorbents. The research findings indicated that Co/Mn-kaolin and Fe-kaolin could efficiently eliminate various categories of nanoplastics. The adsorption capacity of Co/Mn-kaolin for NPs notably decreased as the pH increased from acidic to alkaline. Conversely, Fe-kaolin exhibited increased NP adsorption while increasing the pH level to 10. Nevertheless, the sorption ability of Fe-kaolin was notably hindered at pH levels above 10 [193].

5.2. Co-presence of M/NPs, pharmaceuticals and heavy metals in different water sources

Using magnetic biochar, Liu et al. investigated the influence of higher microplastic ratio on the sorption equilibrium of 17 β -estradiol (E2). The adsorption capacity of E2 was influenced by factors such as solution pH, ionic strength, and the presence of SO_4^{2-} ions. However, the impact of humic acid and electrolytes on the adsorption capacity was found to be negligible. The introduction of Al_2O_3 /MPs reduced the time required to reach adsorption equilibrium for E2 adsorption on graphene-like magnetic biochar (GLMB), but it did not notably enhance or hinder E2 elimination once equilibrium was achieved. The adsorption mechanism for E2 on GLMB involved multiple factors, including π - π and electrostatic interactions. It was revealed that the adsorption capacity of GLMB (approximately 65 mg/g) remained at higher levels than similar E2 adsorbents even after undergoing three cycles [194].

The following year Zhao et al. successfully synthesized a magnetic chitosan-based multilayer adsorbent (FCS-PD) with surface functionality and magnetic separation capability, showing great potential for water contaminant elimination. Diverse analytical methods were utilized to assess the attributes of the resulting material. Aqueous media thoroughly assessed its efficiency in removing diclofenac (DCF). The impact of MPs, inorganic ions, and humic acid on adsorption was examined. Remarkably, the adsorbent exhibited a highest adsorption capacity of approximately 434 mg/g under neutral conditions, surpassing the effectiveness of most reported adsorbents. Diclofenac was efficiently eliminated even at low concentrations (50 µg/L) in surface water. Furthermore, after undergoing five rounds of adsorption and regeneration, the produced adsorbent demonstrated less than 10% decrease in adsorption capacity, indicating satisfactory regenerative capabilities [195].

6. Elucidation of the mechanisms of removal and degradation of contaminants

6.1. Carbon nanomaterials assisted in the removal of micro/nanoplastics

Carbon nanomaterials are important in microplastics adsorption due to their elevated specific surface area and active sites. Among the different carbon nanomaterials, carbon nanotubes (CNTs), defined as hydrophobic materials with a 1 nm diameter and a length of hundreds of micrometers, show an outstanding affinity and in consequence adsorption capacity towards many compounds of hydrophobic nature; among these pollutants, MPs are included [196]. CNTs can remove non-polar MPs by hydrophobic interactions; however, polar/hydrophilic MPs, which do not perform hydrophobic interactions with CNTs surface, cannot be removed by CNTs.

So, magnetic carbon nanotubes (M-CNTs) have been developed to remove polar and non-polar MPs [127]. In this case, the MPs are removed due to the ability of the M-CNTs to magnetize them, converting the MP surface to more hydrophobic character. Thus, the defined method successfully treated seawater polluted with MPs from polyethylene, polyethylene terephthalate, polystyrene, polyurethane, polyvinyl chloride, and polypropylene. Specifically, the magnetic materials, typically iron oxide-CNTs, improve the adsorption efficiency of MPs/NPs, and then, the material can be conveniently isolated using a magnetic field [197].

In this sense, different hydrophobic Fe-nanoparticles that can effectively bind to plastics have been developed, being easy the subsequent recovery of the material from the aqueous medium by using a magnet. Therefore, iron oxide (Fe₂O₃) loaded on CNTs facilitates the removal of both polar and non-polar MPs from water [198]. In addition, it is noteworthy to highlight that the surface charge of the MPs could influence the adsorption mechanism. A study accomplished by Sun et al., 2020 [138] showed that the modification of polystyrene (PS) by carboxylation led to an adsorption mechanism based on electrostatic and pi-pi interactions, H-bonding, and intra-particle diffusion, while the amine modification of PS particles led mainly to hydrophobic interactions and an intra-particle diffusion mechanism. Fig. 7 illustrates the MPs/NPs removal mechanisms by carbon materials.

6.2. Adsorption mechanism of nanomaterials onto MPs-adsorption isotherms

Adsorption can be considered as the primary mechanism of MPs removal by magnetic nanomaterials. In this regard, adsorption isotherms provide helpful information about the adsorbate and adsorbent surface interaction and the adsorption mechanism [199,200]. Those interactions between adsorbate and adsorbent surface play a key role in the efficiency of the adsorption process. Several isotherm models, such as Langmuir, Freundlich, and Temkin equations, are typically used. The adsorption isotherm is the curve representing the equilibrium

adsorption capacity at constant temperature, a phenomenon which could consider mono/multi-layer, and chemical/physical adsorption. The Eq. (5) shows the linearized form of Langmuir isotherm equation:

$$\frac{C_e}{q_e} = \frac{1}{K_L Q_0} + \frac{C_e}{Q_0} \quad (5)$$

where, q_e is the equilibrium adsorption capacity (mg/g), C_e (mg/L) is the equilibrium concentration in the solution, K_L is the Langmuir constant (L/mg), and Q_0 is the maximum adsorption capacity (mg/g) [199].

In the case of Freundlich isotherm, a heterogeneous heat distribution in the adsorbent surface can be assumed. Eq. (6) shows the linear form of Freundlich equation:

$$\log(q_e) = \log K_F + \frac{1}{n} \log C_e \quad (6)$$

where, q_e is the equilibrium adsorption capacity (mg/g), C_e is the adsorbate concentration in the aqueous phase at equilibrium (mg/L), K_F is the Freundlich adsorption constant (mg/g), and n is the Freundlich exponent [199]. Also, the linear Temkin isotherm model can be expressed as follows:

$$q_e = B \ln K_T + B \ln C_e \quad (7)$$

where, q_e is the equilibrium adsorption capacity (mg/g), C_e is the adsorbate concentration in the aqueous phase at equilibrium (mg/L), K_T and B are Temkin isotherm constants.

According to the Langmuir model, adsorption occurs on an adsorbent uniform surface through monolayer formation and without interaction between the adsorbed molecules. On the contrary, the Freundlich model considers that the adsorption is occurring by forming a monolayer onto the adsorbent surface with a heterogeneous distribution and the subsequent formation of adsorbate's multi-layers within the available active sites [201]. Finally, Temkin adsorption model assumes the presence of chemisorption, considering electrostatic interactions, as the basis of the adsorption process. This model assumes a linear reduction of the adsorption energy due to adsorbent surface coverage during adsorption.

In the case of the MPs adsorption onto the magnetic carbon nanotubes (M-CNTs), the resulting experimental adsorption isotherms could be predicted by the Freundlich equation. A comparatively higher affinity of M-CNTs towards PE and PET than PA based on the affinity constant of the Freundlich equation indicated that the hydrophobic interactions between PE/PET and M-CNTs surface were stronger than the electrostatic and complexation forces between PA and M-CNTs [127].

6.3. Adsorption kinetic modeling

The kinetic modeling equations are feasible for the comprehension of adsorption kinetics, this is, the contact time and nature of adsorbate-adsorbent interactions related to the process efficiency. Several adsorption kinetic models, such as the pseudo-first-order, the pseudo-second-order, the Sweber, and the Elovich intraparticle diffusion model, are usually used to explain the adsorption kinetic process [202]. Thus, the pseudo-first order and pseudo-second-order kinetics models can be expressed following the Eqs. (8) and (9), respectively:

$$\ln(q_e - q_t) = \ln q_e - k_1 t \quad (8)$$

$$\frac{t}{q_t} = \frac{1}{q_e^2 k_2} + \frac{t}{q_e} \quad (9)$$

where, qt (mg/g) and q_e (mg/g) are the adsorption capacity values at any time t (min) and equilibrium time, respectively, k_1 (per min) and k_2 (mg/g.min) are the first-order and second-order rate constants. The pseudo-first-order kinetic model can describe PE adsorption onto M-CNT, while the adsorptive removal of PET and PE was well-fitted by the

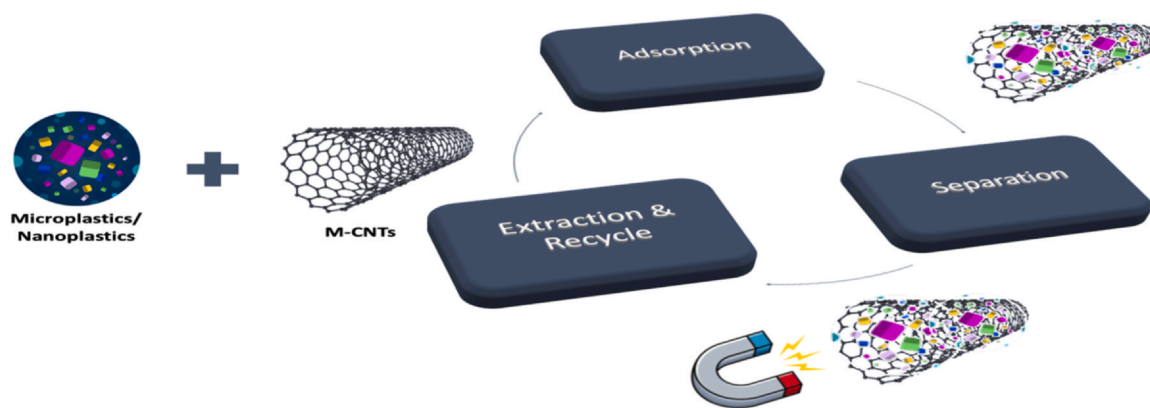


Fig. 7. Micro/nanoplastics removal mechanism by carbon materials.

pseudo-second-order kinetic model. M-CNTs are positively charged due to the presence of Fe_3O_4 on the surface. Fe_3O_4 is linked with the carboxylic group of the M-CNTs. The H-bond between M-CNTs and the PA amide group leads to M-CNTs adsorption on PA.

6.4. Magnetic separation procedure of MPs

A strong magnet can easily remove the complexes formed by the MPs adsorbed onto magnetic carbon nanotubes (M-CNTs) surface. A higher density of hydrophobic Fe NPs, magnetic sepiolite, and magnetic microsubmarines led to a higher separation efficiency. In this case, the plastic type (composition) has a minimal effect on the separation [119]. Magnetic nanomaterials-assisted separation is further supported by the covering of MPs by Fe-containing nanomaterials, which enhances magnetic separation [203]. In addition, M-CNTs have also been useful in removing heavy metals (Pb^{2+} , Ni^{2+} , Co^{2+} , MnO_4^-), industrial dyes, and microorganisms, such as *Escherichia coli* and *Bacillus subtilis*, from wastewater [123]. Indeed, magnetic microsubmarines constructed from sunflower pollen grains have the capability to extract oil [203] and also MPs. It is essential to incorporate a preliminary coagulation stage and a subsequent magnetic extraction process at a large-scale implementation, particularly when employing cost-effective magnetic nanomaterials.

6.5. Extraction of magnetic nanomaterials from micro/nanoplastics and recycling

The magnetic nanomaterials can be separated by a strong magnetic field and heat treatment, which could not cause any deterioration in the magnetic materials. Tang et al. applied the thermal technology (25 to 900 °C) and reported a 100% mass loss of MPs due to the water evaporation; thus, the total decomposition of MPs was observed at 600 °C. This is characterized by developing C-O groups of MPs that eventually disappeared at 600 °C, leaving magnetic nanomaterials clean for reuse [127,204]. One of the most striking features of M-CNTs is its reusability. In the wake of increasing prices of wastewater treatment, it is essential to develop an economical and sustainable technology.

Tang et al. used their exhausted M-CNTs and regenerated them five times by heating them at 400, 500, and 600 °C in a N_2 environment for 2 h. At least four times, the reuse of regenerated M-CNTs with greater than 80% microplastic removal demonstrates its potential in water treatment. However, a loss in magnetism of M-CNTs was observed with increased reuse. M-CNTs reuse makes them economical and eco-friendly materials to eliminate MPs in the reclamation of wastewater treatment. In addition to the extraction and degradation facilitated by M-CNTs, there are alternative approaches for the disintegration of microplastics and nanoparticles, including electrochemical degradation, photodegradation, photocatalytic degradation, and biological degradation [203].

6.6. Modification of M-CNTs for effective MPs removal

Significant improvements have been made in removing microplastics effectively, and most of the approaches are centered on modifying the hydrophobicity of nanoparticles for effective adsorption. Misra et al. [123] modified core-shell particles comprised of a superparamagnetic iron oxide (Fe_2O_3) core coated with a silica shell by magnetic polyoxometalate supported ionic liquid phase (POM-IL) surface-anchoring to form magPOM-SILP. This allowed adsorption on the spherical polystyrene bead (PS) and > 90% removal of 1 to 10 μm PS beads from water under the magnetic field. Grbic et al. [119] increased the hydrophobicity of the Fe nanoparticles coated with hexa decyltrimethoxysilane (HDTMS) for extracting microplastic from water.

The OH groups of iron (Fe) and methoxysilane groups ($-\text{Si}-\text{OCH}_3$) of HDTMS react to form siloxane bonds ($-\text{Si}-\text{O}-\text{Si}-$) [205], exposing the alkyl chain of HDTMS on the surface of the Fe nanoparticles to make them hydrophobic. 10–20 micres microplastic, including PE, PS beads > 1 mm MPs, including PE, PP, PS, PU, PV chloride, and 200 μm –1 mm MPs, including PE, PS, PU, PV chloride and PP were removed up to 92%, 93%, and 84%, respectively. The superhydrophobic surface has been thought to improve adsorption even in saline environments. MPs recovery from the saline environment has been reported by Rius-Arya and Llorca-Isern [206], where the hydrophobicity of the aluminum surface was enhanced by adding anodization and the liquid-phase deposition of lauric acid to remove 99% MPs. Singh et al. [125] developed an eco-friendly and renewable adsorbent via the pyrolysis of excessive biomass. It was shown that MPs greater than 10 μm were 90% removed through a biochar-sand filtration column system that trapped more prominent MPs. The adsorption process in biochar was mediated by the functional groups on the surface of biochar and through electrostatic attraction forces [125].

7. Challenges

Further research should focus on the following points:

- The consideration of MPs/NPs' impact on separation processes, notably membrane fouling, requires a deliberate approach to improve separation techniques.
- The surface chemistry, type, morphology, and size of MPs/NPs should be factored in when addressing membrane fouling. Priority should be given to developing precise and efficient methods for identifying and quantifying MPs/NPs, particularly the smaller nanoparticles, before evaluating separation efficiency.
- Additionally, establishing standardized analytical methodologies and protocols is highly advisable for objectively assessing various methods, each of which possesses its own strengths and weaknesses. Consequently, a viable approach may involve combining individual methods to achieve high separation efficiency at minimal costs.

Techniques such as flocculation-filtration coagulation, and adsorption-magnetic separation have proven effective.

- Functional materials are pivotal in numerous separation processes, with adsorption being a significant aspect. A strategic and cost-effective design of functional materials is strongly recommended to enhance separation efficiency.

7.1. Challenges on commercial magnetic polymeric composites for water processing industries

MPs/NPs are an ecotoxicological issue of great concern because they aggregate and disseminate toxic metals, stubborn pollutants, pharmaceuticals, and personal care products in wastewater media [207]. So, the efficient removal of MPs/NPs is essential. Although magnetic polymeric composites have demonstrated significant promise in removing microplastics and nanoplastics from water and wastewater, this area is still in its initial phases of development [208]. Therefore, more efficient, economical, and eco-friendly MPs/NPs separation methods are still required. There is no doubt that magnetic polymeric composites are promising materials because of their reactive surface and potential for efficient water remediation. However, certain limitations must be addressed for the large-scale application of these materials. These drawbacks include, but are not limited to, their high costs of production, associated with complex synthesis processes. Removal of MPs/NPs by using magnetic nanoparticles lead to fast, non-toxic, and eco-friendly processes, compared to physicochemical treatments; however, their mass production, optimized surface functionality, stability, and biocompatibility should be carefully considered before setting up an industrial-scale remediation process.

Apart from the material or technology used for MPs/NPs remediation, the size of micro/nanoplastic, presence of other pollutants in water, and fragmentation of the micro/nanoplastics while separation also influence the removal efficiency of the process. For instance, while larger (> 1 mm) and smaller (< 20 μ m) MPs can be effectively removed, medium MPs (200 μ m-1 mm) recovery showed low removal efficiency, and this could be attributed to the relatively lower surface area to volume ratio of this medium size fraction of MP. Moreover, the recovery of MPs from water can be negatively affected by the presence of soil and lipophilic particles of biological character (i.e., fat in fish tissues) in aqueous media, which led to non-specific NPs binding. Furthermore, MPs can be fragmented into smaller pieces during the magnetic separation, reducing their removal efficiency [119].

Therefore, we suggest specific prospects for further improving the current status of MP/NPs separation and degradation studies using magnetic polymeric composites. So, future research should focus on:

- i) Developing more cost-effective synthesis methods of magnetic polymeric composites. Incorporating biomaterials or bio-inspired materials into magnetic materials could enhance the biocompatibility of the tested composites.
- ii) Considering factors such as MPs/NPs size, chemical compounds present in the studied water matrix, and the possibility of MPs/NPs' fragmentation during the remediation process will be helpful to obtain higher removal efficiencies.
- iii) Use of real-water matrices for performing the separation or degradation experiments that would assure the real-world applicability of the remediation processes.
- iv) Developing efficient recovery steps for these composites to maintain the removal process's sustainability and enhance their commercial viability in water processing industries should be performed. For instance, adsorbent regeneration is crucial in adsorption since the reusability further provides critical insights regarding its real-practical application.
- v) Exploring the detailed mechanisms of magnetic nanomaterials, the nature of composites with high surface-to-volume ratio, and strategies to maintain the magnetic properties of the tested nanomaterials after consecutive adsorption-desorption cycles.

- vi) Exploring and proposing proper and regulated disposal methods of the magnetic composites after being used as an adsorbent or photocatalytic material in remediation processes.
- vii) Conversion of recovered MP/NPs into value-added products such as fuels (hydrogen and hydrocarbon fuels) [209] and chemical fuels (waxes and diesels) [210] should be explored.

In addition to this, considering the health issues related to the consumption of plastic particles and difficulties associated with their remediation, MPs/NPs should be proposed as a global concern on a similar magnitude as climate change and ozone layer weakening. All countries must make a global effort to reduce the use of MPs/NPs, setting a strict discharge limit. Dedicated funding should be allocated in the research area to develop effective MPs/NPs removal technologies, making them available worldwide before this contamination problem becomes an environmental catastrophe.

8. Conclusions and future perspectives

Functionalized magnetic polymer nanoparticles hold significant promise for various applications, particularly in addressing the issue of MPs and NPs in a wide range of wastewater sources. This comprehensive review explores advanced strategies for separating MPs and NPs, including adsorption, coagulation/flocculation, flotation, filtration, and magnetic separation. The efficiency of these separation processes is influenced by factors such as the reactivity of functional materials or reagents, the properties of MPs and NPs, and the chemical composition of the water.

Furthermore, the review delves into recent advancements in the degradation of MPs and NPs, encompassing AOPs, electrochemical degradation, biological and photocatalytic degradation. Notably, these degradation techniques have yet to achieve complete and efficient removal of MPs and NPs, with photo- and biology-driven methods requiring extended durations (ranging from several months to years) to achieve satisfactory remediation. The degradation of MPs and NPs remains incomplete, raising concerns about the potential generation of hazardous or toxic by-products. These by-products have the potential to introduce new pollutants and pose risks to both aquatic ecosystems and public health. Consequently, it is crucial to elucidate the degradation pathways, monitor potential intermediate products, and assess the toxicity of these degradation by-products.

The small plastic particles, infamously known as MPs/NPs, are contaminants of great environmental concern, persisting for long periods. In this way, the presence of MPs in wastewater is a global health problem because only a few of them show a biodegradable character, and their future environmental impact may be more devastating than the present predictions. They can accumulate in the tissues of several marine organisms, thereby entering the food chain. In addition, humankind can get exposed to MPs/NPs by ingesting polluted drinking water. In this sense, several methods and kinds of materials have been recently developed to separate and degrade MPs/NPs from an aqueous environment.

Among them, the use of magnetic polymeric composites has gained the attention of researchers owing to their outstanding physicochemical properties, such as a high specific surface area, showing exceptional properties in the purification of wastewater polluted with MPs/NPs, acting as adsorbents or photocatalysts. These materials are generally prepared by blending, co-precipitation, in-situ polymerization, molding, or grafting methods. The magnetic materials provide excellent removal efficiency values and offer the advantage of an easy separation using a magnet after the purification treatment. Nevertheless, the large-scale applicability of magnetic polymeric materials, including economic aspects and a further increase in the removal efficiencies, must be comprehensively explored. Meanwhile, steps should be taken by the concerned authorities to regulate the usage of plastics worldwide.

The advancement of magnetic nanocomposites for environmental applications has experienced substantial growth owing to their unique attributes, which include high chemical stability, ease of manufacturing, good recyclability, effective adsorption, and reusability. This review covers many studies related to the development of magnetic nanocomposites. The proven biocompatibility of magnetic nanoparticles ensures the safety of the resulting materials in aquatic environments, particularly in water and wastewater. Additionally, the ease of separation through a magnetic field enhances the value of magnetic materials. A diverse array of magnetic-based nanocomposites has been developed and is currently commercially available, catering to a multitude of real-world applications. However, a notable gap exists between theoretical research and practical applications.

Data availability

Data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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