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Remediation of nano- and microplastics in water environment using nanomaterials

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ABSTRACT

Micro- and nanoplastic pollution of the aquatic environment is a worldwide problem with long-term and dangerous natural consequences. The problems of micro- and nanoplastic pollution, including their bioaccumulation and ecological risks, have become a major focus of public and scientific attention. Over the years, several effective technological innovations have been developed to remediate harmful micro- and nanoplastics, most of which are able to remove this pollution to some extent, although they are not intentionally installed for this reason. Nonetheless, given the negative effects of micro- and nanoplastics contamination of the aquatic environment, further efforts to improve current technologies are still desirable. Thanks to their multifunctionality and structural flexibility, nanomaterials are increasingly being used for water and wastewater treatment to improve treatment efficiency. In particular, the unique characteristics of nanomaterials have been exploited in the manufacture of high-performance adsorbents, semipermeable membranes and photocatalysts for the removal of micro- and nanoplastics from the aqueous environment. This review examines the potential of nanomaterials to offer constructive solutions to solve problems and enhance the performance of existing materials used for micro- and nanoplastic removal. Current efforts and research directions that can be devoted to improving the removal of micro- and nanoplastics from the aquatic environment using nanomaterial-based strategies are discussed. Progress to date demonstrates the benefits of leveraging the structural and dimensional advantages of nanomaterials to enhance the performance of existing micro- and nanoplastic purification processes to achieve more sustainable purification. The control and elimination of micro- and nanoplastics depends on all of us; therefore, proper awareness of micro- and nanoplastic pollution must be provided to every person, as we are all part of the environment.

Keywords: Micro- and nano-plastics removal methods; Nanotechnology; Removal of nano- and microplastics using nanoparticles

1. Introduction

According to Plastic Europe, as of 2021, the annual global production of plastics was 390.7 million tons (Plastics Europe 2023) [1]. Today, plastics have become a part of our lives, mainly due to their properties: plasticity, lightness, durability and low cost [2]. According to available statistics, only about 14% of plastics are recycled, while the rest is incinerated, landfilled or gets into various environmental matrices. Plastic waste generates a mass of microplastics (MPs) into the environment. MPs are often defined as small plastic fragments <5 mm in size [3], with particles <100 nm in size referred to as nanoplastics (NPs) [4,5]. Despite their presence in personal care, cosmetic and pharmaceutical products, most MPs and NPs are derived from large plastic through fragmentation, weathering and degradation [6]. The most common plastic MPs/NPs found in the aquatic environment are polypropylene (PP), polyethylene (PE),

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polystyrene (PS), polyvinyl chloride (PVC), polycarbonate (PC), polyamides (PA), polyester (PES) and polyethylene terephthalate (PET) [7]. MPs/NPs have been globally detected in many environmental matrices, including oceans, freshwater, soil, atmosphere and even groundwater [8–10]. They can be released into the environment through several different routes, mainly through human activities in homes, industry and sewage systems. Approximately 700,000 MPs fibers are released from 6 kg of synthetic clothing during a single wash [11]. Wastewater is another important source of MPs in water bodies [12]. Although some of the current wastewater treatment processes used in industry show up to 90% efficiency in removing MPs, the remaining amounts are still a major problem [13].

MPs pose a direct risk to human health and the ecosystem, either directly or after interaction with persistent pollutants, such as pharmaceuticals, pesticides, heavy metals, dyes, as well as other emerging and conventional industrial pollutants [14–16]. The porous structure of MPs can also cause the transmission of harmful pathogenic microorganisms to humans [17]. In addition, synthesized plastics are usually enriched with additives such as plasticizers, pigments, flame retardants and antioxidants, which are toxic and can be easily released into the environment [18]. Many studies have shown the risk of human exposure to MPs and additives leached from them [19], such as the presence of MPs in drinking water [20,21], human feces and even placentas of children [22].

Environmental pollution of MPs is a global problem, and it is crucial to maintain environmental sustainability, protect public health and the food chain; so there is an urgent need to find more efficient methods of removing of MPs [4,5,23]. Tremendous efforts have been made, including reducing material consumption, increasing recycling capacity, and implementing a number of conventions and regulations to address MPs pollution. In addition to controlling the source of MPs discharge, water treatment is a simple approach to reducing the abundance of MPs and NPs in a wide range of water bodies, including industrial wastewater and natural waters. MPs removal technologies include coagulation, adsorption, magnetic separation, classical and membrane filtration, oxidation and photodegradation, etc. [4,5].

Over the past 5 y, there has been undertaken a lot of technological innovation in wastewater treatment and water treatment, where new materials and techniques are constantly being explored to accelerate current advances in the field. Nanotechnology is a key enabling technology that has the potential to solve a number of societal challenges [25,26]. Nanotechnology-based approaches to contaminant removal have emerged as viable options for solving problems facing water and wastewater treatment technologies by improving process efficiency and performance [27–30]. The size and dimension of the material are critical attributes affecting the performance and effectiveness of the material in their intended applications. Carefully designed and developed advanced nanometer-sized materials impart desirable properties for effective MPs removal. Although the application of nanomaterials in MPs removal is manifold, one simple strategy in which they can play a key role is the direct incorporation of functional nanomaterials into existing materials used for MPs removal. Nanomaterials can be easily incorporated into an existing cleanup process without significant changes to the overall operation. Studies on the engineering aspects of MPs removal technologies based on physical and chemical degradation process strategies emphasize the importance of structurally tuning nanomaterials to maximize their interaction with MPs for more effective removal [24,30–32]. Nanomaterials hold great potential for direct application to existing MPs removal processes, such as adsorption, membrane processes and photocatalysis, to address the limitations of their bulk counterparts. So far, the technologies used to remove MPs from the aquatic environment have been extensively discussed, but efforts to extend existing removal technologies to nanomaterials have not been thoroughly discussed, and a systematic evaluation of the role of nanomaterials in enhancing these methods has not yet begun.

Accordingly, this review aims to demonstrate the potential of nanomaterials in developing and enhancing the effectiveness of currently existing techniques for remediating the aquatic environment. First, the current knowledge of techniques used to remove MPs and NPs is briefly presented, followed by a discussion of the unique properties resulting from the structure and dimensions of nanomaterials, and their role in solving problems associated with existing techniques and processes is analysed. The development and performance of nanomaterials and their integration with MPs removal technologies are discussed. The review concludes with a discussion of key barriers to implementing MPs removal techniques using nanotechnology. Current limitations standing in the way of development are highlighted, and future research perspectives are recommended to expand the current state of knowledge.

2. Overview of microplastic removal technologies

Two systematic approaches should be considered in the fight against water pollution by MPs and NPs. The first is to stop or minimize further deposition of MPs in water bodies, while the second concerns their removal from the aquatic environment [4,5]. None of the water and wastewater treatment technologies currently in practice are designed to remove MPs and NPs, as they were developed to remove and neutralize dissolved and suspended contaminants [33,34]. In general, water and wastewater treatment technologies for the removal of MPs and NPs are commonly based on physical, chemical and biological processes, depending on the treatment mode (Fig. 1) [4,5,23].

Based on available research, it has been observed that physical methods are studied more, followed by chemical and biological methods. Most studies using physical principles such as adsorption, filtration, sedimentation, flotation, etc. are classified as physical methods. Most of these methods have been verified in the laboratory, on a pilot or even full-scale basis. Various physical methods with high MPs removal efficiency include carbon-based adsorbents such as biochar, magnetic carbon nanotubes, magnetic polyoxometalate-based adsorbents in the ionic liquid phase, disk filters, rapid sand filter and dissolved air flotation, and others [4,23,35,36]. In addition, among physical purification technologies, quantitative analysis has shown that filter-based methods show better MPs removal efficiency than others.

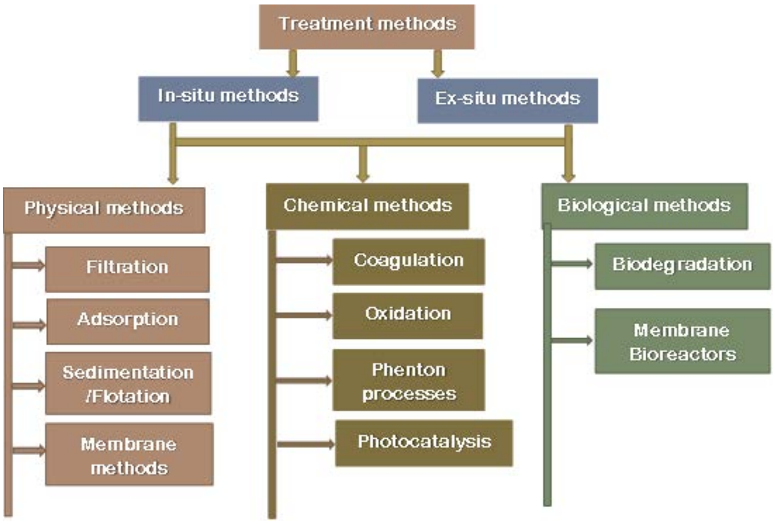

Fig. 1. Basic classification of MPs removal methods, adapted from [4,5,23].

MPs removal by physical methods follows the following sequence: filtration process > flotation process > adsorption process > membrane process > magnetic and density separation process [4].

In addition to physical and biological methods, chemical methods are also used in the treatment of water and wastewater containing MPs, either alone or in combination to enhance the effectiveness of physical processes (e.g., sedimentation, membrane processes). Several methods such as ozonation, advanced oxidation processes, coagulation and electrocoagulation, Fenton processes and photocatalysis are most commonly used for the removal/degradation of plastics [37]. The average efficiency of MPs removal achieved by chemical methods is as follows: photo-Fenton process > electrocoagulation > ozonation > electro-Fenton process > sol–gel agglomeration > coagulation > modified Fenton process [4]. Unfortunately, none of these treatment strategies can remove MPs from contaminated sludge and wastewater when implemented alone, without any other physical or biological treatment strategies. In addition, byproducts, as well as some secondary sludge generated by some methods, such as coagulation and electrocoagulation, require further treatment.

Biological methods use organisms to counteract the contamination of MPs present in the environment by degrading them. A number of organisms have been studied for their potential to degrade MPs present in water and wastewater, among them mainly microorganisms show potential to degrade MPs [3]. Biological methods of removing MPs have been used primarily to treat wastewater, both municipal and industrial. Wastewater treatment can be grouped into three main treatment methods: pretreatment, second-stage treatment, and third-stage treatment, also known as final treatment, used for reuse of treated wastewater [39]. In this last stage of purification, membranes have great potential [40].

3. Removal of MPs and NPs using nanotechnology

The development of nanotechnology is a promising alternative for increasing the efficiency of water and wastewater treatment. Nanomaterials (size: 1–100 nm) have unique physical and chemical properties, such as large surface area and special affinity for various types of chemical compounds, which translates into their use for removing inorganic and organic contaminants from water and wastewater [29,30]. Nanomaterials can be characterized by many parameters, including dimension, surface chemistry and crystal structures. Based on their geometry and dimension, nanomaterials are classified into four groups, that is, zero-dimensional (0D), one-dimensional (1D), two-dimensional (2D) and three-dimensional (3D). The functions and roles of nanomaterials are closely related to their chemical composition, dimension and geometry [27,30]. The chemical composition and corresponding surface chemistry, such as hydrophilicity and charge, are important aspects that affect their physico-chemical properties, such as antibacterial and antifouling properties, and easy reuse. On the other hand, the dimension and geometry of nanostructures have a major impact on their distribution, permeability and transport behaviour, as well as the interaction of nanomaterials with the surrounding matrix or types of impurities [41].

3.1. Types of nanomaterials used in water and wastewater technology

Considering the chemical composition, nanomaterials/nanoparticles used in water/wastewater technology are divided into three groups: inorganic nanomaterials (mainly metals/metal oxides), carbon nanomaterials (carbon nanotubes (CNTs), graphene and others), and nanocomposites, that is, nanoparticles embedded most often in polymer matrices [29,30].

Inorganic nanomaterials used in adsorption processes, semipermeable membrane modification and photocatalysis include zeolites [42,43], silicon dioxide [44] and metals and metal oxides, mainly zero-valent iron, silver and gold, as well as magnetic iron oxides, titanium dioxide, zinc oxide and others [30,42,45,46]. Some of them (e.g., nano-maghemite and nano-magnetite) exhibit paramagnetic properties, which facilitates their separation and recovery in a low-gradient magnetic field.

Carbon-based nano-adsorbents are mainly: carbon nanotubes (CNTs) (1D), graphene and graphene oxide (2D), graphitic carbon nitride g-C_3N_4 and others [47,48]. They are characterized by a large specific surface area, high thermal stability and the ability to bind organic and inorganic impurities, often after appropriate surface modification [47,48].

Graphene is an allotropic variety of carbon, made up of atoms arranged flat and connected by strong sigma bonds with sp^2 hybridization of very small lengths (0.142 nm) [27]. The graphene layer, forming a hexagonal matrix, resembles a honeycomb in its shape (Fig. 2a), and since it has a one-atom thickness of about 0.3 nm, it is simplistically referred to as a 2D structure [27]. Graphene has a high specific surface area, remarkable flexibility, relatively high electrical conductivity and high mechanical strength (100–300 times that of steel), and is an excellent mobile electric charge carrier [27]. The surface of graphene can be easily modified, as graphene's sp^2 carbon hybridization can exhibit various interactions with polymer matrices, such as electrostatic interactions, valence interactions, non-covalent interactions (e.g., π–π interactions) and ease of mixing with polymers [29,30,49]. Materials created by combining graphene with a variety of composite materials greatly expand its application potential [36]. Graphene is usually used as a derivative, such as graphene oxide (GO), reduced graphene oxide (rGO) or functionalized graphene oxide (fGO). Graphene oxide (GO) is a monolayer of graphene in oxidized form, containing oxygen-containing functional groups, mainly hydroxyl, carboxyl, carbonyl and epoxy (Fig. 2b) [27,29,30]. GO exhibits properties similar to graphene, except for the asymmetry of its sheets, which is due to the presence of functional groups located at the edges and in the basal plane [27], thus facilitating electrostatic interactions with organic and inorganic impurities. The presence of oxygen- and hydrogen-containing functional groups promotes dispersion in aqueous media and allows the performance of various surface modifications that can be used in the preparation of functionalized adsorbents or membranes [27].

Carbon nanotubes (CNTs) are an allotropic variety of carbon, whose walls are made of graphene coiled into a tubular structure [50]. Native CNTs can be divided into two main types, namely single-walled carbon nanotubes (SWCNTs) and multi-walled carbon nanotubes (MWCNTs) [29,30,50]. SWCNTs consist of a coiled single layer of graphene (Fig. 3b), while MWCNTs have multiple coaxially aligned sheets (Fig. 3a). The diameter of SWCNTs ranges from a fraction to a few nanometers (0.3–2 nm), while MWCNTs - from a few to tens of nanometers (2–100 nm) [50]. The lengths of both types of nanotubes can reach micro- or even millimeter values, so their length-to-diameter ratio is remarkably high, ranging from 10^3–10^5, which allows them to be classified as one-dimensional objects. SWCNTs consist of two separate regions with different physical and chemical properties. The first is the sidewall of the nanotube, and the second is the end cap (tip) of the tube [50]. The distance between adjacent graphene cylinders in MWCNTs is about 0.34 nm, and van der Waals forces interact between graphene layers to ensure that the cylinders are mutually ordered. In addition, CNTs have excellent water permeability and adsorption capacity, which is particularly important for their potential use in environmental engineering. Of great importance are functionalized CNTs containing oxygen functional groups that attach to the sidewalls, cores and ends of CNTs (Fig. 3c) [29,30,50]. The purpose of functionalization is also to eliminate the dispersion of CNTs and to

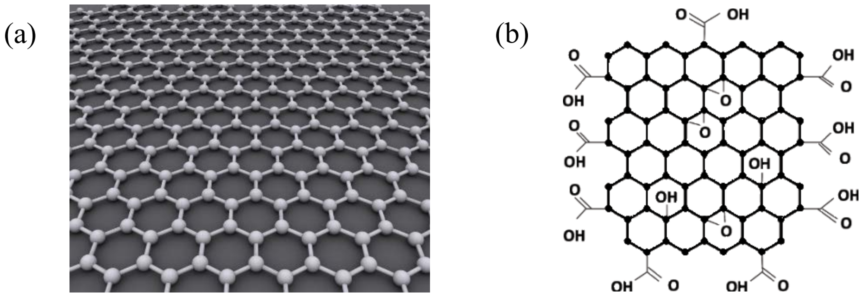

Fig. 2. (a) Two-dimensional graphene structure model and (b) graphene oxide model, adapted from [27,29,30].

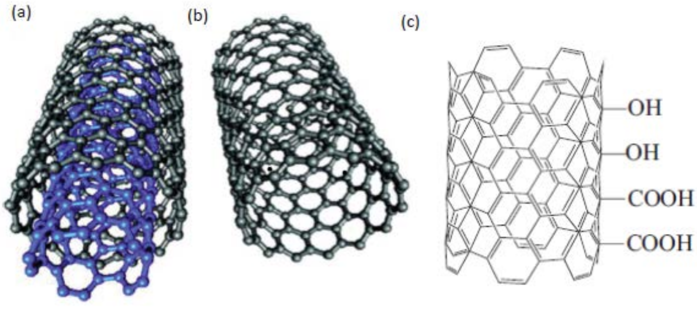

Fig. 3. Structures of multi-walled (a), single-walled (b) and functionalized (c) carbon nanotubes, adapted from [29,30,50].

facilitate the efficiency of adsorption or membrane separation in eliminating contaminants from water or wastewater.

Graphitic carbon nitride (g-C_3N_4) is a layered, stable and polymeric non-metallic material with a 2D structure that has been discovered as a photocatalyst responsive to visible light. The basic structure of g-C_3N_4 is graphite-like sp^2 planes of hybridized C and N atoms [52]. The inclusion of nitrogen atoms in g-C_3N_4 makes it more attractive for applications in photocatalysis [52], energy storage and solar cells. g-C_3N_4 shows enhanced activity in water and wastewater treatment due to its large adsorption surface area for a wide range of synthetic toxic contaminants, including microorganisms [52,53], and due to the appropriate value of the "bandgap" energy, Also, the synergistic effect of increased visible light absorption capacity, more exposed active sites and improved separation of photogenerated charge is responsible for excellent photocatalytic disinfection and removal activity of other contaminants [52,53]. Functional groups on the surface (–NH_2, >N–N<, =N–, –NH–, =C–N<, etc.) provide active sites for adsorption of toxic substances from wastewater [52,53]. Physical interactions (including electrostatic interactions, π–π coupled interactions, and hydrophobic interactions) and chemical interactions (including complex formation or acid–base interactions) on the surface of g-C_3N_4 enhance the adsorption of organic and inorganic pollutants from water.

Nanoparticles with a large specific surface area, used in water and wastewater treatment, have some limitations, which include ease of aggregation, the possibility of release into the environment, and potentially negative effects on the ecosystem and human health [26,54]. The problem can be solved or minimized by creating nanocomposite materials. Nanocomposites are multiphase materials in which at least one of the phases has dimensions in the range of 10–100 nm. Nanocomposite materials integrate the functional advantages of nanoparticles and matrices and show great potential in industrial-scale applications. Their classification can be made either by matrix material (inorganic or polymeric) and dispersed phase (CNTs, graphene, metals, metal oxides) [26,54]. Among various nanocomposites, polymer nanocomposites have become the subject of a number of research and development due to their many favourable properties, such as film-forming ability, dimensional variability and activation by functionalization [54]. Nanocomposite materials are promising materials in water and wastewater treatment because of their properties related to both nanoparticles and polymer matrix [26,54,55].

3.2. Efficiency of MPs/NPs removal using nanomaterials

Current research related to nanotechnology in water and wastewater treatment focuses on three areas: elimination of contaminants by adsorption, filtration through membranes, and catalytic degradation with simultaneous disinfection of microorganisms [3–5]. Functionalization or hybridization of classical materials with nanomaterials can improve the properties and performance of the former by removing their limitations [41], while surface functionalization of nanomaterials not only improves their stability and dispersion in aqueous media, but also introduces additional surface properties and facilitates integration with classical processes, such as oxidation or physical sorption [56]. By introducing specific ligands onto the surface of a nanomaterial, opportunities can be created to selectively bind target contaminants [57] and at the same time improve the stability and recovery of nanomaterials.

3.2.1. Adsorption

Adsorption is a process commonly used to remove organic and inorganic contaminants from water and wastewater. The performance of conventional adsorbents is usually limited due to low specific surface area and/or low number of active sites, lack of selectivity and slow speed, as well as short adsorption and regeneration cycles, which significantly affects the economics of the process. Biochar (BC) and activated carbon (AC) are widely used adsorbents for the treatment of water containing MPs and NPs [4,58]. However, the high cost of AC and BC and the other properties mentioned above limit the use of these adsorbents for the removal of MPs and NPs. Research is therefore being conducted on other alternative adsorbents [59], among which nanomaterial-based adsorbents, primarily nanometals and metal oxides, carbon nanotubes (CNTs), graphene, carbon nitride and nanocomposites are a new approach. Nanoadsorbents are characterized by high specific surface area and/or number of active sites, high selectivity and process speed, and long adsorption and regeneration cycles, and their adsorption efficiency against many contaminants is several times higher than that of traditional adsorbents [49,60]. In addition, nanomaterials exhibit high reactivity and sorption capacity and ease of functionalization [61]. Functionalization can improve adsorption capacity for a variety of contaminants, and specific functional groups. such as carboxyl and hydroxyl groups, provide the ability to target specific contaminants [59].

Carbon nanomaterials show wide application in adsorption of organic and inorganic pollutants from the aquatic environment, including MPs, due to their easy synthesis, high surface area, numerous active sites, hydrophobicity and tailored functionalization depending on the nature of the pollutants. [62]. The incorporation of hydrophilic groups into carbon nanomaterials also increases the overall hydrophilicity and porosity of the adsorbent and improves the mechanical strength of the nanocomposite compared to the original material [62,63]. Carbon materials that have so far been investigated as adsorbents for the removal of MPs and NPs include graphene, carbon nanotubes, and fly ash, often as composites with other materials.

Graphene-based materials take advantage of their intrinsic hydrophobicity, abundance of functional groups (GOs) and large surface area to remove organic contaminants. In addition, their special hexagonal honeycomb structure shows excellent stability in acidic and basic environments. Therefore, they have the ability to remove MPs under different pH conditions [64,65]. Recently, Yuan et al. [66] studied and modeled the adsorption of MPs from PS by 3D sheets of reduced rGO. The proposed 3D structure hindered the agglomeration of the sheets and thus facilitated the diffusion of microspheres in the adsorption process. The maximum adsorption capacity of 3D rGO was found to be a function of solution pH, temperature, initial concentration of MPs and ions in solution, and adsorption time. For a contact time of 120 min at 26°C, a pH of 6 and an initial concentration of 600 mg/L, the maximum adsorption capacity was found to be about 617 mg/g. The adsorption mechanism was dominated by strong π–π interactions between the benzene ring of PS and the carbon ring of rGO. These conclusions were confirmed by fitting the experimental data with a pseudo-second-order kinetic model and Langmuir adsorption isotherms, which confirm intramolecular diffusion in the monolayer adsorption process [66]. Combining CNTs with magnetic nanoparticles, greatly facilitates their separation after adsorption. For example, magnetic carbon nanotubes (M-CNTs) have been used to remove PE, PET and PA [67], with all MPs (5 g/L) being completely removed at an M-CNTs dose of 5 g/L over 5 hr. The maximum adsorption capacities of PE, PET and PA were 1,650; 1,400 and 1,100 mg/g, respectively. The adsorbent was thermally regenerated and retained its magnetic and adsorption properties like the original M-CNTs. It showed 80% removal efficiency even after 4 reuse cycles. The adsorption of PE and PET was attributed to hydrophobic interactions, while π–π interactions played a role in the adsorption of PA and PET [67]. Fly ash modified with Fe ions was used as an adsorbent to remove PS nanoplatelets from water. Modified fly ash typically has a large surface area, porous structure and good affinity for contaminants. Due to their low cost, these materials are largely being explored in water treatment. Fly ash modified with iron ions showed an adsorption capacity of 82.8–89.9 mg/g at pH 5–7, and after adsorption it can be easily recovered and reused several times [68]. In another study, a bimetallic carbon composite (Cu–Ni carbon material) was synthesized by a hydrothermal method and used to remove PS nanoplastics from water [69]. The removal efficiency of NPs from PS (10 mg/L) increased from 32.72% to 99.18% when the dose of CuNi@C increased from 0.1 to 0.3 g/L. Maximum removal efficiency was achieved at pH 4 and this was attributed to the opposing zeta potential of NPs from PS (positive) and Cu–Ni@C (negative), which leads to electrostatic attraction [69].

Metal–organic frameworks (MOFs) are excellent materials in the field of water and wastewater treatment due to their unique physicochemical properties, such as ultra-high surface area, porous structure, ease of functionalization and variable architecture through the selection of suitable metal ions and organic ligands. Many MOFs have been used for adsorptive removal of MPs from aqueous environments due to the formation of hydrogen bonds or van der Waals interactions with MPs [61]. Positively charged defects present in MOFs show high affinity for a wide range of negatively charged MPs, including polyvinylidene fluoride (PVDF), PS and poly(methyl methacrylate) (PMMA) [70]. MOFs show a high level of MPs removal from PVDF in the range of 61.8%–95.5%. Under optimal dose and operating conditions, consistent MPs removal efficiency was achieved for 10 high-capacity filtration cycles without change the structural integrity of the adsorbent. Zeolitic imidazolate framework (ZIF) MOFs have been widely used in the adsorption of inorganic and organic contaminants from water, due to the formation of hydrogen bonds, π–π type and electrostatic interactions [71,72]. ZIF-67 was used to remove MPs from PS, and the efficiency depended on the ZIF-67 dose, MPs concentration, sample pH, temperature and contact time. Under optimal operating conditions, removal of about 92.1% of MPs from PS was achieved [73]. ZIF-8-based wood aerogel was used to remove poly(1,1-difluoroethylene) (60–110 nm) and PS (90–140 nm), showing removal efficiencies of 91.4% and 85.8%, respectively [74].

Layered double hydroxides (LDHs) are excellent inorganic adsorption materials that consist of positively charged layers of metal hydroxides and negatively charged anions and water molecules to balance the total charge. LDHs have the general formula $[M^{2+}_{1-x}M^{3+}_{x}(OH)_2]^{x+}(A^{n-})x/n{\cdot}mH_2O$, where M^{2+} and M^{3+} are divalent and trivalent metal cations, and A^{n-} is an interlayer anion of value $n-$ [75]. Various divalent and trivalent cations can be used to produce LDHs if their radii are not significantly different from those of Mg^{2+} and Al^{3+} ions. LDHs are characterized by their excellent thermal stability, their ability to exchange anions and their ability to change their internal structure. During thermal treatment, LDHs lose interlayer anions and water molecules and transform into a new functional material known as double layered oxides (LDOs) [76]. Both LDHs and materials based on LDOs can be used in adsorption to remove organic contaminants, including MPs [77]. Zn-Al LDHs have been used to remove PS NPs from deionized water, synthetic fresh water and hard water. High adsorption capacities were observed and the removal efficiency depended on water type, anion concentration and pH. Electrostatic interactions played a major role in the adsorption of NPs onto Zn-Al-LDHs [78]. In most cases, LDHs can be regenerated thermally or by changing the pH.

Magnetic adsorption/separation based on nanomaterials has been described as a simple and inexpensive way to remove MPs/NPs [67]. Magnetized MPs and NPs can be removed from the aqueous environment in a magnetic field.

Magnetization of NPs and MPs from PE, PP, PS and PET of various sizes using Fe_3O_4 is an effective method for their separation from water samples [61,67,79–81]. Studies have shown that the removal efficiency of PET was much lower than that of PE, PP and PS due to its relatively poor hydrophobicity, and was 62.8% ± 8.34%, 86.9% ± 6.92%, 85.05% ± 4.70% and 86.1% ± 6.21%, respectively [79]. The removal efficiency also depended on the size and shapes of MPs/NPs as well as the type of water matrix. The removal efficiency of large (~900 μm) and small (~200 μm) MPs was higher than that of medium (~500 μm) MPs [79]. In seawater, electrostatic attraction between negatively charged MPs and positively charged Fe_3O_4 nanoparticles enhanced the adsorption and magnetism of MPs, thus improving the overall removal efficiency compared to freshwater. Heo et al. [80] evaluated the ability of magnetic Fe_3O_4 nanoparticles to adsorb PS MPs. Transmission electron microscopy images showed the formation of Fe_3O_4-PS complexes as a result of adsorption of PS particles onto the nanoparticles. The experiments concluded that the main interactions involved in the aggregation of Fe_3O_4 with PS particles were hydrophobic interactions. Ions present in the environmental freshwater sample inhibited the ability of Fe_3O_4 particles to form Fe_3O_4-PS complexes, but the adsorption capacity increased with increasing amounts of Fe_3O_4. Iron oxide particles could be recovered from Fe_3O_4-PS complexes by desorption. Martin et al. [81] have developed the synthesis of iron(III) oxide nanoparticles with various hydrophobic coatings to separate, concentrate and detach

NPs and MPs from both fresh and salt water by magnetic separation. They achieved 100% removal of particles in the size range of 2–5 mm and almost 90% of NPs particles in the size range of 100 nm to 1,000 nm in the magnetic field. Grbic et al. [2] modified Fe nanoparticles with hexadecyltrimethoxysilane (HDTMS) to improve their hydrophobicity to bind MPs. In a synthetic seawater solution, the resulting removal efficiency was 92% for PE and PS in the 10–20 μm size range and 93% for MPs including PE, PTFE, PS, PVC and PP of >1 mm. Also recovered were 84% and 78% of MPs (PE, PS, PU, PVC, and PP) ranging in size from 200 μm to 1 mm from freshwater and sediment, respectively [82].

Singh et al. [83] modified the surface of the biochar by depositing iron nanoparticles on its surface to increase surface properties and obtain magnetic properties. They achieved NPs removal efficiencies of about 100% compared to 75% for the raw biochar. It was found that the adsorption processes proceeded through complexation with the surface and electrostatic interactions between NPs and Fe nanoparticles. It was possible to regenerate the adsorbent by separating the NPs from the iron-modified biochar particles. After regeneration and reuse of the adsorbent, the adsorption capacity was maintained, making it recyclable.

Sepiolite is a naturally occurring green clay material consisting of hydrated magnesium silicate. It is useful in adsorption applications mainly because of its large surface area, porous structure and low cost. Magnetic sepiolite has been used to remove PE MPs with an efficiency of 98.4% [84]. After recycling the mixture of magnetic sepiolite and PE MPs, the removal efficiency was above 90% after 5 cycles of reuse and above 80% after 10 cycles of reuse.

Nanoadsorbents of biological origin that have been used to remove MPs and NPs are mainly chitin sponges, coral-inspired aerogels and biochar derived from biomass [61]. Naturally produced materials from renewable resources (e.g., cellulose, chitin) are used for MPs remediation because of their biocompatibility and biodegradability. Chitin, present in the internal and external skeleton of invertebrates, can be used to remove MPs [85]. The addition of GO to chitin sponges increases the strength of the composite due to hydrogen bonding between GO and chitin chains. In addition, the presence of functional groups on GO contributes to the removal of contaminants [86]. Similarly, oxygen-doped carbon nitride can also strengthen chitin and remove impurities due to its structural similarity to GO. Thus, chitin-based sponges composed with GO or oxygen-doped carbon nitride have been used for adsorptive removal of neutrally, positively and negatively charged MPs (PS, COOH-PS, NH_2-PS) [87]. The chitin-based sponges showed removal efficiencies in the range of 71.6%–92.1%, with the adsorption mechanism related to hydrogen, and π–π bond formation and electrostatic interactions. Chitin sponge combined with GO or carbon nitride showed similar performance. These modified chitin sponges can be used repeatedly without a significant decrease in performance.

Similar properties are exhibited by coral, chitosan aerogels and their composites with magnetic Fe_3O_4 nanoparticles, providing easy separation of aerogels after adsorption [88,89]. PDA-MCS aerogels removed PET MPs from water at pH values of 6–9, with removal efficiencies of up to 91.6%. Even after three cycles, they maintained a removal efficiency of 83.4%. MPs adsorption was mainly due to electrostatic interaction and physical adhesion. Under simulated aqueous conditions, PDA-MCS aerogels showed excellent PT, PE and PS MPs removal [210].

In conclusion, adsorption may be one of the promising technologies for removing MPs/NPs from the aquatic environment, mainly due to its simplicity of operation, high removal efficiency, flexibility in the choice of adsorbents and low cost. Adsorbents based on nanomaterials show greater efficiency in removing MPs/NPs from water than classical adsorbents. Most of the studies have been conducted at laboratory scale using simulated wastewater or seawater. In some cases, the presence of NOM and anions significantly reduced MPs removal efficiency. Although nanomaterial-based materials are emerging for adsorptive remediation of MPs, such research is still in its infancy and requires improvements in various aspects, such as synthesizing selective nanoadsorbents and conducting pilot-scale studies using a continuous flow system. Adsorbents with mechanical and chemical stability over a wide pH range are preferred. In addition, reusable adsorbents can provide environmental and economic impact benefits.

3.2.2. Membranes and membrane processes

A major challenge in the operation of semipermeable membranes is the relationship between membrane selectivity and permeability. The effectiveness of membrane systems is largely determined by the membrane material and thickness, and the ability to modify the physicochemical properties. Efforts to produce new semipermeable membranes with improved separation efficiency and anti-fouling properties are a key area of research in the field of membranes for water and wastewater treatment [90]. The new generation of membranes should meet the treatment of specific waters by matching their structural and physicochemical characteristics, including hydrophilicity, porosity, membrane charge and thermal and mechanical stability, as well as introducing additional functions such as antibacterial, photocatalytic or adsorptive properties. In recent years, there has been growing interest in the use of nanomaterials for the development of a new generation of membranes with advanced anti-fouling and anti-scaling properties and transport properties [91]. The most common applications in the fabrication of such membranes are zeolites, metals and metal oxides, and some carbon materials, that is, carbon nanotubes (CNTs) and graphene-based materials [30,50,92]. Significant improvements to the semi-permeable thin-film composite (TFC) membranes used today, involve the introduction of nanoparticles into the polymeric or ceramic active layer [93]. In the membrane structure, which is referred to as a thin-film nanocomposite (TFN), the nanoparticles provide preferential pathways for water transport, lower hydraulic resistance and retention of dissolved salt. Fig. 4 shows a schematic diagram of the structure of a TFC and a TFN membrane [30,50].

Recent studies have shown that inorganic two-dimensional (2D) nanomaterials such as graphene oxide (GO) and MXene have significant potential in separation and purification using TFN membranes due to their excellent chemical and thermal stability [94,95]. Mass transport in

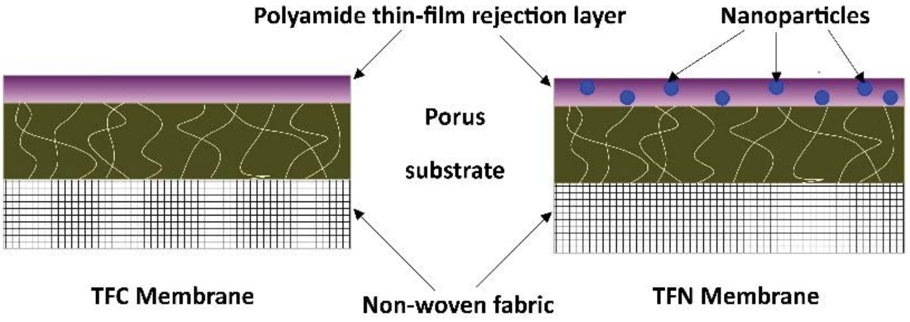

Fig. 4. Schematic diagram of the membrane structure of a thin-film composite membrane and a thin-film nanocomposite membrane, adapted from [30,50].

these membranes takes place mainly in the interlayer gaps of the nanoparticles, through which water flows during the separation process, but this results in low permeate fluxes.

MXenes are modern 2D materials due to their unique surface structure, which makes them combine properties such as metallic conductivity, hydrophilic surface and good mechanical stability [94,95], which creates their suitability for the preparation of separation membranes [96]. The most widely studied MXene is $Ti_3C_2T_x$, which has been used as a membrane material in various separations, especially in water purification, due to its excellent antibacterial properties and hydrophilic surface [96,97]. In addition, MXene's excellent mechanical flexibility and film-forming ability make it a great potential in creating membranes for water desalination with high efficiency and selectivity. Their strong tendency to overlap and lack of limited porous structure hinders their use in the formation of semipermeable membranes. Yang et al. [9] have described a new type of membranes based on porous $Ti_3C_2T_x$ (h-$Ti_3C_2T_x$) sheets, which were obtained by etching with Co_3O_4 nanoparticles deposited on $Ti_3C_2T_x$ sheets, followed by vacuum filtration using a polymer membrane as a carrier. $Ti_3C_2T_x$ nanosheets exhibit a planar porous structure with nanotubes with an average diameter of 25 nm, which facilitates the construction of membranes with higher water flux capable of separating MPs/NPs. Using PS microspheres (FPs) with different diameters as model MPs, the obtained $Ti_3C_2T_x$ membranes exhibited extremely high MPs efficiency (up to 99.3%) and high-water flux (196.7 L/h/m^2·kPa). Such properties make $Ti_3C_2T_x$ porous membranes, may have significant potential for practical applications in removing MPs from water.

Fryczkowska and Przywara [99] presented the results of a study of composite membranes made of polyacrylonitrile (PAN) with the addition of reduced graphene oxide (rGO/PAN) used to remove MPs from industrial wastewater. The study showed that higher amounts of rGO (0.11 to 0.83 wt.%) added to the PAN matrix result in the formation of a larger number of pores of similar size (~150 nm), which enables the separation of colloids in the environment (retention >82%) and, more importantly, MPs. An important feature of the rGO/PAN composite membranes studied are their anti-fouling properties and the ease of cleaning the filter cake layer, which allows their reuse. Other studies have shown that rGO/PAN composite membranes are not susceptible to fouling and can be used to remove PET fragments from wastewater from a recycling plant for this polymer [100]. The membranes under these conditions showed easy and effective cleaning, confirming the anti-fouling properties of rGO/PAN composite membranes formed with a small addition of rGO to the PAN matrix.

Membrane surface and pore cleaning performed with aggressive chemicals is used as a standard procedure in all membrane-based separations in industry [101]. As an alternative to energy-intensive membrane cleaning, membrane surfaces can be modified by both physical and chemical methods. A promising pathway for such membrane modifications is the introduction of inorganic nanoparticles (NPs) into the structure of the UF membrane to mitigate irreversible membrane fouling and maintain, or even improve, the membrane's ability to retain contaminants. This solution is considered as a way to mitigate/reduce membrane fouling caused by MPs [102]. Various nanomaterials are being considered in this regard, especially hydrophilic nanomaterials, which are of great importance in the context of nanocomposite membrane development. For example, incorporation of SiO_2 can prevent membrane compression under high pressure, reducing membrane pore size and water flux. Moreover, the incorporation of silver [103] and copper [104] nanoparticles can confer long-term resistance to bacteria on the membrane. Potential health risks associated with potential leaching of NPs have proven to be negligible, as the amounts of silver and copper leached are well below the thresholds set by the World Health Organization for drinking water safety. One issue encountered in all studies involving the use of inorganic NPs is that beyond a certain threshold, the addition of more NPs leads to inconsistencies in membrane fabrication, such as increased surface roughness, decreased hydrophilicity and NPs buildup. Therefore, it is important to determine the optimal amount of NPs to achieve the desired membrane properties.

Bionanomaterials are defined as nanomaterials of biological origin or inorganic nanomaterials combined with organic components. Bioremediation of MPs using bionanomaterials is still a growing field, and has a promising future, as many researchers around the world are working on scaling it up to the industrial level [105]. Jalvo et al. [106] have developed a nonwoven cellulose-based water filtration membrane. They impregnated various polysaccharide nanocrystals, namely cellulose nanocrystals (CNCs), cellulose nanofibers (T-CNFs) and chitin nanocrystals (ChNCs). The coating of the nanocrystals or nanofibers significantly increased the hydrophilicity, tensile strength and elastic modulus of the fabric. In addition, it changed the surface charge to be more negative (positive) for cellulose (chitin) based membrane modifications. CNC and ChNC modifications improved membrane permeability, and T-CNF impregnation improved stiffness under dry conditions. All of these biomaterials showed the potential to remove NPs less than 500 nm in size, while T-CNF was able to separate particles less than 2 μm in size [106]. Lignin-zeolite composite nanofiber membranes produced by electrospinning have higher flux and permeation rates compared to other commercially produced membranes. In addition, uniformly dispersed zeolite nanoparticles of 1 wt.%, combined with heat treatment, improve the mechanical properties of the nanocomposite. This bionanomaterial has been successfully used for pre-filtration of a wide range of MPs [105,107].

3.2.3. Catalysis and photocatalysis

Catalytic oxidation or photocatalytic oxidation belongs to advanced oxidation processes and is used to remove trace contaminants and pathogenic bacteria from the aquatic environment. In addition, photocatalysis is a useful pretreatment method for enhancing the degradation of hazardous and hard-to-biodegrade pollutants, and it can also be used as a pretreatment step in chemical and biological processes for the removal of organic compounds [108–110].

The process of photocatalysis is defined as "a change in the rate of a chemical reaction or its initiation under the influence of ultraviolet or visible light in the presence of a photo-catalyst that absorbs light and is involved in a 'chemical transformation'." [108,110]. During the photocatalysis process, the solid-state catalyst generates reactive oxygen species (radicals) on its surface, which in turn reduce and oxidize chemical compounds adsorbed on the catalyst surface [108–110]. When the energy of photons (*h*ν), delivered to its surface, is greater than or equal to the energy of the "band gap", free electrons are transferred from the valence band (VB) to the empty conduction band (CB). Generation of electron (e^-) - "electron–hole" (h^+) pairs occurs throughout the volume of the photo-catalyst, which has been reached by radiation quanta of sufficient energy. In order for the reaction with the substrate to occur, the electron and the "electron–hole" must be on the surface of the photo-catalyst. In the conduction band, the e– reacts with oxygen (O_2) and this leads to the formation of superoxide radicals ($^{\bullet}O_2^-$), or hydroperoxide radicals ($^{\bullet}HO_2$), which mineralize the impurities to water (H_2O) and carbon dioxide (CO_2) [108,110,111]. For a substance to be oxidized with a photo-catalyst, its redox potential must be above the valence band of the semiconductor, while for reduction processes, this potential must lie below the conduction band. This is due to the fact that after excitation in the valence band there are "electron–holes" involved in the oxidation process, while the electrons responsible for reduction, are transferred to the conduction band [108]. An extremely important factor affecting electron-"electron–hole" recombination is the size of the photo-catalyst particles, since the "forbidden band" energy, crystal structure and redox potential of semiconductors depend on their size [109]. Nano-catalysts are therefore of great interest in photocatalysis due to their high surface-to-volume ratio, which allows a higher degree of degradation of pollutants compared to their classical counterparts, providing better catalytic activity and efficiency [110]. The difference in the magnitude of the energy "refracted gap" (E_g) of semiconductors composed of nanoparticles with sizes on the order of single nanometers compared to solid materials of the same composition is quite significant. For example, for TiO_2 consisting of crystalline grains with a size of 1–10 nm, the "refracted gap" is about 0.2 eV larger than for monocrystalline [109]. In addition, the use of nano-structured semiconductor material for photocatalysis is more advantageous compared to their bulk counterpart, as most of the photo-generated electrons and "electron–holes" are available on the surface of the nano-photo-catalyst due to the high surface-to-volume ratio [109]. The doping of a wide range of functional nanomaterials into the base photocatalyst also makes it possible to activate the photocatalysts with visible light, instead of high-energy UV light, which opens the way to use solar energy to carry out photocatalytic processes [111–113]. In addition, the stability of the nano-catalyst is improved, and the nanocomposites formed are compatible with existing photo-reactors [109].

Photocatalysis has proven effective for degrading MPs and NPs in aqueous environments. It is widely believed that photocatalysis degrades NPs to a greater extent, due to its relatively large specific surface area compared to MPs, and the easier adsorption of photocatalysts on their surfaces to achieve their degradation [114]. TiO_2 has been widely used as a model photocatalyst due to its high oxidation capacity of organic pollutants [109], including for the elimination of MPs from wastewater with good degradation efficiency [4,115]. A number of studies have used TiO_2/plastic composites for photodegradation of plastics such as PP [116,117], PS [118] and high [119] and low-density PE films [120,121].

Thomas et al. [120] formed low-density polyethylene (LDPE) nanocomposite films by adding TiO_2 nanoparticles 15 nm in size to the LDPE matrix (LDPE-TiO_2). The weight loss rate of the nanocomposite film reached 68% under sunlight irradiation for 200 h, while it was only 1.02% for the non-irradiated film. In addition, it was found that the brittleness of the film increased, and the formation of micron-sized pores indicated the degradation of the nanocomposite film. In contrast, UV irradiation of PE film with TiO_2 nanoparticles with a particle size of 50 nm resulted in 18% degradation within 300 h. In [121], the photocatalytic degradation of PE was studied using semi-crystalline anatase-type TiO_2 nanoparticles synthesized by the sol–gel method with 0.1 wt.% TiO_2 and a particle size of 50 nm, and the degradation of PE was compared with commercially produced crystalline TiO_2 powder with a size of about 200 nm. The degradation was found to be size-dependent for TiO_2, and a higher rate of PE film weight loss of up to 18% within 300 h was observed for 50 nm diameter nanoparticles under UV irradiation. This is related to the higher band gap in TiO_2 nanoparticles. Zan et al. [122] studied the photocatalytic degradation of TiO_2/PE composite films under UV light or sunlight in air. The weight loss rate of the composite film reached 68.38%, the average molecular weight (Mw) of the composite film decreased by 94.56%, and the number of average molecular weight (Mn) decreased by 93.75% after exposure to UV light for 400 h. Asghar et al. [123], on the other hand, noted a significant increase in the oxidative degradation of polyethylene films when TiO_2 photocatalyst doped with Fe, Ag and Fe/Ag was incorporated. In previous PE degradation studies, TiO_2 has been used in the form of nanoparticles. However, there is an option to use a material with a larger surface area, in the form of nanotubes [124,125].

Considerable efforts have been made to exploit the visible portion of sunlight and alter the photocatalytic properties of TiO_2, since TiO_2 is only active in the UV wavelength range below 385 nm, which is less than 5% of sunlight. Dye sensitization is an effective method for increasing the photoreactivity of TiO_2 in the visible spectral range and has been successfully used for environmental applications [126–128]. Brilliant green (BG, bis (4-diethylaminophenyl) phenylmethyl chloride) has been used to sensitize TiO_2 to the visible light blue-green dye BG [127,128]. The degradation of pure and composite PE films was measured in terms of photoinduced weight loss and confirmed by FTIR, SEM, surface roughness and tensile strength tests. Thus, PE films with 10% dye-sensitized TiO_2 nanotubes showed degradation by about 50% under visible light in a short period of 45 d [127,128].

Polypropylene also undergoes photodegradation, as a number of molecular chains interact with TiO_2 in the wavelength range of 310–350 nm [129–131]. For this reason, some work has been directed toward studying the incorporation of TiO_2, in PP films [129,130]. PP film (thickness 44–47 µm) containing 0–25 wt.% TiO_2 powder was subjected to photon irradiation for up to 200 h at room temperature in air. It was found that only very fine particles of P-25 TiO_2 led to degradation of the film, as detected by a change in colour (bleaching) and a significant decrease in elongation at break. Measurement by transmission electron microscopy showed that voids around the P-25 particles in the film were formed by photocatalytic oxidation of TiO_2, causing bleaching and a decrease in mechanical properties. In [131], the photo-oxidative degradation of PP films with applied TiO_2 nanoparticles was studied in a chamber with xenon lamps as the radiation source. TiO_2/PP composites were prepared with 0.1, 0.5 and 1.0 wt.% TiO_2. It was found that the rate of photodegradation accelerated as the amount of TiO_2 increased from 0.1 to 1.0 wt.% due to the greater interaction of TiO_2 particles with PP chains.

Shang et al. [118,132] studied the photocatalytic degradation of polystyrene using nano-TiO_2 at ambient temperature under UV light. They found that as the TiO_2 content increased, the degradation rate of PS MPs improved. Compared to pure PS, the PS-TiO_2 composite showed a high mass loss rate and produced intermediates with low and medium molecular weights. In the paper [133], the solid-phase photodegradation of PS and PS-TiO_2 nanocomposite was studied under ultraviolet radiation at 253 nm. A pronounced decrease in the values of average molecular weights and greater weight loss of polymer samples were observed with increasing UV irradiation time, as well as with increasing percentage of TiO_2 content, both for PS and PS-TiO_2 composites. The band gap energies of PS and PS-TiO_2 decreased with increasing UV irradiance. Nabi et al. [115] studied the photocatalytic degradation of PS and PE microspheres using layers of TiO_2 nanoparticles under UV light. TiO_2 nanoparticles produced with Triton X-100 showed complete mineralization (98.40%) of 400 nm PS within 12 h. The photodegradation of PE proceeded at a good rate, and the main end product of the process was CO_2. Studies of the degradation mechanism and analysis of intermediates showed that hydroxyl, carbonyl and carbon-hydrogen radicals are generated during photodegradation of PS and PE. Recently, BiOCl was used to prepare PS-based nanocomposites, which showed significant degradation of PS after photodegradation [134]. The enhanced degradation of PS on BiOCl compared to non-irradiated PS was attributed to the former's better absorption of visible light (PS/BiOCl), which is undoubtedly due to the photocatalytic properties of BiOCl.

Cho i Choi [135] studied the photocatalytic degradation of poly(vinyl chloride) films deposited on TiO_2 in an air or nitrogen atmosphere. After 300 h of irradiation, the average molecular weight decreased by two-thirds and the weight loss of the composite film was 27%. The degradation performance showed almost no improvement in a nitrogen atmosphere, indicating the importance of oxygen in the photocatalytic process. A study was also conducted on the photodegradation of PVC under different UV radiation (UVA, UVB and UVC) under different environmental conditions [136]. Changes in the physical and chemical properties of PVC were more pronounced under UVC, and the amount of ROS generated was a function of exposure time. Thus, as irradiation time increased, PVC was found to contain more oxygen functional groups, a higher carbonyl index and higher crystallinity, especially under UVC. Based on Fourier transform infrared studies, the active sites and photodegradation sequence of functional groups under UVA, UVB and UVC radiation were confirmed. The amount of reactive oxygen species (ROS) produced increased with exposure time.

A study of the degradation of PMMA and PS nanoparticles by photocatalysis using TiO_2-P25/β-SiC foams under UV-A light was also conducted [137]. The effects of flow rate, initial pH and light intensity on degradation efficiency were studied. The results showed that about 50% of the nanoporous carbon contained in PMMA degrades within 7 h at an irradiance of 112 W/m^2, a flow rate of 10 mL/min and an initial pH of 6.3. Degradation is faster at low pH (4–6) and low flow rate. MPs with PS particle size of 140 nm degrade faster with particle size of 508 nm. In addition, the mineralization of PMMA was faster than that of similar size, probably due to the easier breaking of C=O and C–O bonds in PS than C–C, C=C and C–H bonds in polystyrene [137].

One nanomaterial that has attracted interest in recent years as a photocatalyst for the degradation of MPs and NPs is zinc oxide (ZnO), due to its excellent optical properties, high redox potential and good electron mobility [109,138], as well as its high contaminant degradation activity even in small amounts. Tofa et al. [139] studied the synthesis and use of ZnO nanowires as a photocatalyst for the degradation of low-density PE (LDPE) residual MPs in water in the presence of sunlight/Vis. The photocatalysis process increased the brittleness of the MPs surface and caused wrinkles, cracks and cavities in the MPs, leading to the formation of lower molecular weight compounds, taking as long as several months to complete the degradation process. Uheida et al. [116] investigated the photocatalytic degradation of MPs from PP under visible light using ZnO nanowires deposited on glass fibers in a flow photocatalytic reactor. The results of FTIR analysis confirmed the effective photodegradation of PP MPs due to the appearance of a carbonyl group with a higher carbonyl index (CI ~ 40) after 456 h of exposure to visible light, compared to control samples. The degradation of PP MPs proceeded through long-chain cleavage, which was confirmed by changes in crystallinity and generation of volatile organic products causing defects in PP. The results showed that photocatalytic degradation of PP MPs carried out continuously for two weeks under light reduced the average particle volume by 65% compared to the initial volume of PP MPs. Modification of ZnO with plasmonic metals is a promising method for accelerating the oxidation of MPs especially LDPE films in water under solar radiation. ZnO nanowires (3.26 eV band gap) were modified by depositing platinum nanoparticles [140], which slowed down electron-hole pair recombination, increased the absorption of visible light and thus the activity of $^{\bullet}OH$ radicals, which play an important role in the degradation of MPs. The results of the experiment showed that the ZnO-Pt photocatalyst effectively degraded MPs fragments from LDPE, and their composition changed significantly before and after photocatalysis under visible light. Ariza-Tarazona et al. [141] developed an eco-friendly photocatalysis strategy for degrading MPs from high-density PE using protein-based N-TiO_2 semiconductors. Environmental conditions, catalyst surface area, surface properties, as well as interactions between MPs and catalysts are extremely sensitive factors and can significantly affect the degradation rate. The degradation process using ZnO is economical and environmentally friendly, as zinc is a cheap and non-toxic element.

Other photocatalysts, including CdS, tungstophosphoric acid (HPW), H_3BO_3, goethite, and OMS-2 have also been studied for (micro)plastic degradation [138]. CdS and HPW show high photocatalytic degradation efficiency of MPs, while goethite shows relatively low efficiency. The degradation of PE films over cryptomelane-manganese oxide (OMS-2) photocatalyst under UV light in visible light has also been studied [142]. The weight loss of PE-OMS-2 composite films decreased systematically by 16.5% after 288 h of UV irradiation, but the composite films changed little under visible light irradiation. Under UV irradiation, the efficiency of PE degradation can be controlled by adjusting the OMS-2 content [142]. Another study proposed the development of a new ultrathin photocatalyst in the form of BiOCl, which showed strong potential to degrade MPs. BiOCl material is characterized by increased production of hydroxyl radicals ($^{\bullet}OH$), promoting the degradation of MPs [143]. Ding et al. [144] studied the photodegradation of PET using the clay minerals kaolinite and montmorillonite under UV light. Both clay samples, especially kaolinite, mediated the generation of $^{\bullet}OH^-$ and $^{\bullet}O_2^-$ radicals in reaction systems that vigorously participated in the photodegradation of PET. Other MXene/$Zn_xCd_{1-x}S$-type photocatalysts have been synthesized and used to degrade PET MPs [145]. In addition to the significant photocatalytic degradation of PET, the amount of hydrogen produced was also measured, which was 14.17 mmol/g·h. Other small useful organic products resulting from the transformation of PET MPs include acetate and ethanol. The high efficiency of MXene/$Zn_xCd_{1-x}S$ photocatalysts can be attributed to their enhanced photocarrier separation ability and optimal band structure with enhanced valence band oxidation capacity.

An alternative to metal catalysts in AOP is metal-free nanocarbon materials such as graphene and carbon nanotubes (CNTs), which have emerged due to their unusual dimensional structure, tunable physicochemical properties and environmentally friendly nature [4,27,109]. However, one of the challenges associated with nanocarbon materials is the trade-off between excellent catalytic performance and mediocre stability in oxidizing environments. Therefore, it is desirable to fabricate robust composite nanocarbon catalysts by environmentally friendly and easy methods without compromising the reactivity and stability of the.

Recently, the development of binary or ternary composites has begun, due to the possibility of synergistic effects and overcoming the limitations of pure nanomaterials. A stable hybrid structure is critical to the performance of a composite catalyst. Binary or ternary hybrid systems such as rGO/Fe_2O_3 or rGO/Fe_2O_3/ZnO have been synthesized to improve photocatalytic performance [146]. These composites not only improve photocatalytic performance, but also their sorption capacity. GO can act as an acceptor/transporter of photogenerated electrons for nanoparticles and reduce the recombination of photogenerated electron holes, which increases photocatalytic activity. Uogintė et al. [147] obtained mass loss of PE particles after 480 min by 48.06%, 39.54% and 50.46% for GO-Cu_2O, GO-MnO_2 and GO-TiO_2, respectively, compared to 35.66% for graphene oxide alone. FTIR results confirmed the effective degradation of PE MPs particles with the appearance of newly formed functional groups and an increase in the carbonyl index. The degradation process was found to follow a pseudo-first-order kinetic model. An experiment with different pH values showed that the degradation rate decreases at higher pH values. Optimal values of experimental variation were found at lower pH values and longer contact times of the nanomaterial with UV light. In conclusion, graphene oxide-based nanomaterials can be used to degrade MPs particles in aqueous environments.

Spiral CNTs were modified by nitrogen doping and encapsulation of manganese carbide nanoparticles to form magnetic hybrid nanocatalysts that were capable of degrading and mineralizing MPs derived from cosmetic products [148]. They achieved 50%wt. MPs removal at a catalyst concentration of 0.2 g/L and a microplastic concentration of 5 g/L. The synergistic effects of CNTs and Mn nanoparticles, together with the high graphite content of the nanocarbon, ensured the stability of the nanocomposite during the advanced oxidation process, in which the degradation yield could be maintained for 3 cycles. The encapsulated manganese carbide promoted electron migration to generate reactive oxygen species. The graphitic carbon coating provides protection for the Mn nanoparticles and prevents their leaching in solution. The spiral architecture enables high structural strength and particle aggregation during the hydrothermal process.

Photoactive micromotors have gained a great deal of attention in recent years due to their broad potential for remediation of environmental contaminants and water purification [149,150]. Micro-motors are very small particles (measured in microns) that can move on their own in a specific direction when placed in a chemical solution [150]. Recently, a number of studies have been conducted on the ability of TiO_2-based micromotors and the degradation

Table 1
Summary of performance of nanomaterials in technologies for removal of MPs and NPs

Process	Nanomaterials	Micro-plastics	Matrix	Removal efficiency	References
Adsorption	GO –OC_3N_4	PS	Synthetic water 1 ppm, pH: 7	Chitin: 63.3% Chitin/–OC_3N_4: 90.6% Chitin/GO: 89.6%	[87]
Adsorption	UiO-66-OH MOF	PVDF, PS, PMMA	Synthetic water 1 ppm	PVDF: Melamine: 33.0% Melamine/UiO-66-OH: 90.1% PMMA: Melamine: 46.1% Melamine/UiO-66-OH: 88.2% PS: Melamine: 44.7% Melamine/UiO-66-OH: 85.7%	[70]
Adsorption	3D rGO	PS	Synthetic water 0.25 ppm	PS: 72.63%	[66]
Magnetic adsorption	Fe_3O_4 composite C@Fe_3O_4	PE, PES	Real river water, domestic sewage, seawater	River water: 81.33% Domestic sewage: 82.28% Seawater: 80.56%	[79]
Magnetic adsorption	Fe-HDTMS	PE, PS	Synthetic seawater; 10–20 μm	92%	[141]
Magnetic adsorption	Magnetic CNTs	PA, PE, PET	Kitchen waste treatment plant 48 μm, 5,000 ppm	PA, PE, PET: 100%	[67]
Catalyse	Mn decorated N-doped CNT	MPs from cosmetics	Synthetic water 5,000	50%	[148]
Catalyse	TiO_2 NPs film	PS	5 μm	99.9%	[115]

mechanism of MPs in photocatalysis. MPs were degraded by photocatalysis through the use of micromotors constructed by coating TiO_2 particles (700 nm) with nickel (10 nm) and gold (30 nm) [151]. The micromotor drive is provided by photochemical reactions in water and hydrogen peroxide initiated by electron and electron–hole generation processes. The performance of micromotors in removing MPs has been tested on commercially supplied MPs from PS, primary MPs isolated from personal care products, and MPs collected from the Baltic Sea and Warnow River [150]. Since this method is very new in treating MPs compared to other conventional and advanced treatment methods, future research is definitely needed to obtain more effective and efficient advanced photocatalysts so that this method can be successfully applied to treat MPs in real wastewater.

4. Summary

MPs and NPs are abundant in the environment due to the excessive use of plastics in various types of products. Their release, from various sources, especially into water bodies, is becoming a serious threat to both living organisms and the environment, showing adverse effects on aquatic organisms and soil species. They have been found in various types of water, raising concerns even among the ordinary public about their impact on safety and health. They can reach the human body through the environment and food web. More recently, some studies indicate their presence in the human body, including blood, lung tissues and placenta. Given the multiple negative effects of MPs and NPs, it is very important to establish useful strategies to reduce their amount in the environment, especially in water bodies. While a preventive measure is the most effective strategy in remediating MPs and NPs, water and wastewater treatment are important engineering tools that can offer immediate solutions to control, mitigate and even eliminate MPs and NPs contamination.

The potential of nanomaterials in enhancing the performance of existing water and wastewater treatment technologies is undoubtedly enormous. This review presents the current state of technology for the removal of MPs and NPs from water systems based on a number of known and new functional nanomaterials used in both MPs and NPs capture and deactivation processes. The unique structural properties of nanomaterials make it possible to design hybrid structures that allow for synergistic effects, which can affect more efficient removal of MPs and NPs, compared to classical counterparts. It has also been noted that hybridization improves the recyclability of materials. While solving the limitations of conventionally used materials in typical water treatment processes, nanomaterials face several major problems, in particular the difficulty of large-scale processing and lack of reproducibility. In particular, large-scale fabrication of nanomaterials and the lack of practical experience in handling them remain formidable challenges in the field. Table 1 summarizes selected research results in this area.

Nanomaterial-based adsorption may be one of the promising technologies for the removal of MPs and NPs from aqueous matrices, mainly due to its simplicity of operation, high removal efficiency, flexibility in the choice of adsorbents and low cost. Most studies have been carried out at laboratory scale using simulated wastewater or seawater. Although nanomaterial-based materials are emerging for adsorptive remediation of MPs/NPs, such research is still in its infancy and requires improvements in various aspects, such as synthesis of selective adsorbents and pilot-scale experiments using a continuous flow system.

Polymeric/ceramic membranes containing nanomaterials are very often characterized by high efficiency, and have unique anti-fouling and anti-bacterial properties, high mechanical and thermal strength, compared to conventional membranes with comparable fouling retention. In recent years, there has been growing interest in the use of nanomaterials for the development of a new generation of membranes with advanced transport and anti-fouling and anti-bacterial properties. In the membrane structure, nanoparticles provide preferential pathways for water transport, lower hydraulic resistance and high retention of dissolved salt. The most common applications in membrane fabrication using nanomaterials are: zeolites, metals and metal oxides, and materials based on graphene and CNTs.

Photocatalytic degradation has emerged as an important technology for the removal of MPs and NPs. Although photocatalysts based on TiO_2 and other nanomaterials show satisfactory efficiency for the removal of MPs and NPs, the identification of intermediates, the reusability of photocatalysts and costs should still be the subject of further research. In addition, some TiO_2-based materials show good photocatalytic performance in the UV band, while more visible light photocatalysts with high efficiency need to be developed. The decomposition of MPs and NPs to small molecules is a complex process that involves many pathways, and their identification is a major challenge. At the same time, the toxicity of degradation products and the environmental friendliness of photocatalytic materials must be taken into account. Most photocatalytic materials are only applicable to the degradation of specific types of MPs and NPs, and there is a lack of systematic research on different types of mixed MPs and NPs.

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