

Recent Advances in Magnetic Polymer Nanocomposites for Water Purification Applications

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Abstract

Magnetic polymer nanocomposites (MPNCs) have attracted considerable attention as advanced multifunctional materials for water purification due to their high adsorption capacity, magnetic recoverability, and excellent reusability. This review presents a comprehensive overview of recent developments in the synthesis, characterization, and environmental applications of MPNCs for wastewater treatment. Particular emphasis is placed on the principal synthesis strategies, including *in situ* and *ex situ* approaches, and their influence on nanoparticle dispersion, interfacial interactions, and the physicochemical properties of the resulting nanocomposites. The review covers the most widely investigated magnetic nanomaterials, such as Fe_3O_4 , $\gamma\text{-Fe}_2\text{O}_3$, CoFe_2O_4 , ZnFe_2O_4 , and other ferrites, incorporated into natural and synthetic polymer matrices including chitosan, cellulose, alginate, polyaniline, polypyrrole, poly(vinyl alcohol), and polystyrene. Advanced characterization techniques, including X-ray diffraction (XRD), scanning and transmission electron microscopy (SEM/TEM), Fourier-transform infrared spectroscopy (FTIR), X-ray photoelectron spectroscopy (XPS), thermogravimetric analysis (TGA), vibrating sample magnetometry (VSM), and superconducting quantum interference device (SQUID) measurements, are discussed to evaluate the structural, chemical, thermal, and magnetic properties of these hybrid materials. The adsorption performance and underlying mechanisms of MPNCs for the removal of heavy metals, dyes, pharmaceutical compounds, organic pollutants, and oil contaminants are critically analyzed, highlighting the roles of polymer functionalization, nanocomposite architecture, and magnetic separation in enhancing treatment efficiency and reusability. In addition, the contribution of density

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functional theory (DFT) to understanding adsorption mechanisms and guiding the rational design of high-performance adsorbents is reviewed. Finally, current challenges and future perspectives, including green synthesis, multifunctional and stimuli-responsive materials, scalable manufacturing, and industrial implementation, are discussed. This review provides a comprehensive framework for the design and development of next-generation magnetic polymer nanocomposites for sustainable water remediation applications.

Keywords: magnetic polymer nanocomposites; magnetic nanoparticles; water treatment; heavy metal removal; wastewater remediation; functional materials

1. Introduction

Due to the rapid population growth around the world, industries have extended their projects to satisfy the requirements and demands of different sectors. This growth has caused many problems such as climate change due to gas emissions [1,2], and the extensive use of natural resources involved in heavy industries leading to an environmental issue related to is pollution by heavy metals, which adversely affect soil quality and pose a threat to human health requiring a rapid and comprehensive solution. Various human activities contribute to soil and water system contamination, including the discharge of effluents from mining operations [2], waste disposal site leakage, pesticide-laden agricultural runoff, and airborne pollutant deposition [3]. Heavy metals generate substantial environmental issues by entering the food chain, resulting in severe health problems in people at the top of the food chain [4]. Many different types of hazardous factors enter the environment during mining and smelting operations, posing significant risks to the environment. Dental procedures, textile, tanning, electroplating, and the paper and pulp sector emit heavy metals. Table 1 lists some of the hazardous pollutants that have been shown or have the potential to affect humans through drinking water.

Table 1. Major sources of contaminants.

Source	Type of Contaminants
Agricultural	Nutrients, pathogens, Sediment, Pesticides.
Industrial	Metals, Dyes, Organochlorines, BOD.
Construction	Export of sediment/soils and nutrients.
Land disposal	Pathogens, Hazardous chemicals, Nitrates.
Atmospheric deposition	Metals, Polar pollutants (POPs and PPOPs).
Resource extraction	Acidity, PAHs, Metals.

Heavy metals such as cadmium, mercury, lead, manganese, and chromium are very poisonous and are mostly released into the environment through various industrial operations [5]. Cadmium enters ecosystems through many paths, including industrial discharge, absorption mechanisms, and surface water runoff, which leads to sediment deposition. Mercury, on the other hand, is an exceedingly hazardous element present throughout the biosphere, and its atmospheric concentration is continuously growing as a result of human activity. When mercury comes into touch with aquatic sediments, it changes into highly poisonous methylmercury, which enters the human body via the food chain and causes neurological disorders [6]. Lead is non-biodegradable and hazardous to the human body when in large quantities. Manganese, the most prevalent hazardous heavy metal, exists in a variety of oxidation states and is necessary for physiological functions, but it may be deadly if taken in excess. Chromium, a cancerous and toxic element, exists in two stable oxidation states: chromium (III) and chromium (IV) (VI) [7]. Chromium is

widely used in a variety of industrial industries, some of which have a substantial influence on the local environment. The ferrochrome industry is one of the largest sources of chromium emissions [8].

Several strategies have been developed to mitigate heavy metal pollution, including biochar, zirconium-based metal–organic frameworks (MOFs), and natural biosorbents. Biochar is an effective adsorbent for heavy metals and pesticides due to its high porosity and ability to support microbial activity [7]. Functionalized Zr-based MOFs exhibit enhanced adsorption performance and selectivity [8], while biosorbents derived from natural resources provide a low-cost and environmentally friendly alternative for wastewater treatment [9]. For example, *Inula viscosa* leaves achieved approximately 85% Zn^{2+} removal [10], whereas an olive waste/sodium alginate composite removed 94.54% of Cu^{2+} from synthetic wastewater [11]. Recent advances in nanotechnology have enabled the development of high-performance materials with unique physicochemical properties for environmental remediation, particularly through the engineering of materials at the nanoscale [12,13].

Nanoscience and nanotechnology seek to create effective materials, systems, and technologies by manipulating matter at the nanometer scale, where unique physical, chemical, and biological features arise. To achieve this goal, we will need to work across disciplines, bringing together chemists, physicists, biologists, and engineers to gain a better understanding of the design, implementation, and impact of new nanotechnologies [9]. When the size of materials decreases to the nanometric scale, their physical and chemical properties can change significantly [10]. This transformation is attributed to several factors, including: a marked increase in the surface-to-volume ratio, resulting in a greater proportion of atoms located on the surface; modifications in the electronic structure of nanoparticles and their components; variations in interatomic distances as well as an increased presence of structural defects; and finally, quantum confinement effects, resulting from the limitation of charge carriers in dimensions close to the electronic wavelength [11].

Nanomaterials are used in a variety of sectors, but their application in environmental engineering is gaining popularity. Nanotechnology provides intriguing answers for tackling long-standing environmental issues, regulating pollutants, and remediating contaminated sites. A variety of remedial procedures have been developed to efficiently treat polluted soils, wastewater, and groundwater [12–14].

In recent years, numerous studies in environmental remediation have highlighted the potential of nanomaterials for the treatment of surface waters, groundwater, and industrial effluents. These materials have proven effective for the removal of various contaminants, such as toxic metal ions, radionuclides, organic and inorganic compounds, as well as microbial agents like bacteria and viruses [15]. Recent advances in the field of nanotechnology, particularly in nanofiltration and nanoparticle design, have led to the classification of nanometric-scale materials into four main categories: metallic nanoparticles or nanocomposites, carbon-based nanomaterials, zeolites, and dendrimers [16].

This study focuses on magnetic polymer nanocomposites (MPNCs), a potential subclass of nanomaterials that has received a lot of interest in recent years. These hybrid materials, created by incorporating magnetic nanoparticles into polymer matrices, combine the benefits of both components namely, the magnetic responsiveness and high surface reactivity of the nanoparticles, with the chemical stability, processability, and functional versatility of the polymers [17].

Because of their synergistic qualities, MPNCs have great potential for environmental applications, particularly the removal of heavy metals from contaminated water sources. Their ease of separation using external magnetic fields, combined with their capacity to be regenerated and reused, makes them ideal for long-term and cost-effective water treatment systems. This review aims to provide a comprehensive overview of the synthesis strategies, physicochemical characteristics, and adsorption performance of MPNCs.

Furthermore, it highlights recent advances, challenges, and perspectives associated with their practical implementation in environmental remediation technologies.

2. Nanocomposite Materials

Nanocomposites are considered emblematic materials of the 21st century due to their innovative architecture and the synergy of properties they offer, often absent in traditional composites [18]. Nanomaterials are used in a variety of sectors, but their application in environmental engineering is gaining popularity. Nanotechnology provides intriguing answers for tackling long-standing environmental issues, regulating pollutants, and remediating contaminated sites. A variety of remedial procedures have been developed to efficiently treat polluted soils, wastewater, and groundwater [19]. Compared to nanoparticles, nanocomposites generally exhibit a higher adsorption capacity, better selectivity, and increased stability. Their great diversity results from the variable combination between the nanometric phase and the host matrix, allowing the development of numerous types of nanocomposites tailored to specific applications [20]. Thus, depending on the nature of the matrix used, nanocomposites can be grouped into four main categories: (i) ceramic matrix nanocomposites, (ii) metallic matrix nanocomposites, (iii) carbon-based nanocomposites, and (iv) polymer matrix nanocomposites.

Ceramic matrix composites are materials in which one or more specific ceramic phases are incorporated to improve their wear resistance, as well as their thermal and chemical stability (Table 2). However, the main limitation of ceramics lies in their brittleness and low hardness, which restricts their use in many industrial applications. This weakness has been overcome through the development of ceramic matrix nanocomposites (CMNC). As for metal matrix nanocomposites (MMNC), they are multiphase materials consisting of a metallic matrix or a ductile alloy, in which nanometric-sized reinforcement particles are dispersed. These materials are distinguished by their excellent ductility, toughness, mechanical strength, and high elastic modulus. They find applications in various fields, particularly in aeronautics and the automotive industry [21]. Finally, in polymer matrix nanocomposites, the integrated nanometric elements generally play the role of reinforcement agents called nanofillers. These are classified according to their dimensionality: one-dimensional (1D) such as carbon nanotubes, two-dimensional (2D) like montmorillonite-type clays, and three-dimensional (3D) like silver nanoparticles in powder form [22].

Table 2. Matrix-based classification of nanocomposites and their practical uses.

Matrix Type	Strengths	Weaknesses	Key Applications
Ceramic (CMNC)	High thermal resistance	Brittleness	Cutting tools, coatings
Metal (MMNC)	High strength/ductility	Costly processing	Aerospace, automotive
Polymer (PMNC)	Lightweight, flexible	Lower thermal stability	Water treatment, packaging

Polymer Nanocomposites

Polymeric materials are widely used in industry due to their ease of manufacturing, low density, and generally ductile nature. Their insulating nature, along with the stability of their physical and chemical properties, makes them suitable for use in demanding environments [23,24]. The incorporation of inorganic fillers into polymers is a common method for modifying their mechanical, electrical, and functional properties [25]. The extent of the induced modifications strongly depends on several factors, including the nature, composition, morphology, and loading content, as well as the flow conditions at the time of integrating this load into the matrix. Nanometric-scale reinforcements have a strong potential to induce new behaviors, thereby imparting unique characteristics to the

obtained materials [26]. It is important to note that the reinforcement efficiency of these nanocomposites, even at low loading rates, can be comparable to that observed in fiber-reinforced microcomposites, achieving performances in the range of 40 to 50% [27].

Polymer matrix nanocomposites combine the exceptional properties of polymers and nanoparticles [28]. Their effectiveness relies on several advantages, including the high surface-to-volume ratio of the nanofillers, their high interfacial reactivity, as well as the good compatibility and excellent mechanical performance offered by the polymer matrix [29]. Moreover, these materials often exhibit a capacity for regeneration and reuse [30]. Thanks to these characteristics, they constitute a promising class of adsorbents for the removal of metals present in water and effluents [31].

Polymers adsorbed on surfaces, as well as polymer nanocomposites (PNC), are involved in numerous phenomena and application fields, such as adhesion, stabilization of colloidal dispersions, mineral flotation, oil recovery, smart materials, as well as in wetting and spreading phenomena [32]. In general, polymer matrix composites play a key role in modulating optical, magnetic, dielectric properties, as well as electrical and thermal conductivity [33].

The reinforcement elements used in the fabrication of polymer nanocomposites can be classified according to their dimensions [34]. Three-dimensional nanoparticles, whose three axes measure a few nanometers, form the first category. It notably includes spherical silica particles, metals, and certain semiconductor nanoclusters. The second category includes structures such as nanotubes and whiskers, which have two nanometric dimensions and a larger third dimension, resulting in elongated shapes. Carbon nanotubes and cellulose crystallites are commonly studied examples of reinforcing agents [35].

Finally, the third category includes sheet-like fillers, whose thickness is on the order of a few nanometers, while the length can reach several hundred, or even thousands of nanometers. These two-dimensional structures are the basis of layered polymer nanocomposites [36]. These materials are generally obtained by inserting the polymer, or a monomer intended to be polymerized later, into the intercalations of the multilayer structures of the host compound. A number of crystalline materials, whether natural or synthetic, have demonstrated their ability to accommodate polymers within their galleries under specific conditions [37]. Among the most frequently used hosts are graphite, metal chalcogenides (such as MoS_2), various types of clays, as well as layered silicates, such as montmorillonite, hectorite, saponite, fluorohectorite, fluoromica, kaolinite, and vermiculite, not to mention layered double hydroxides. Nanocomposites developed from clays and layered silicates are the subject of extensive research, particularly due to the availability of these starting materials and their well-established ability to form intercalated structures [37–39]. The synthesis of these advanced materials relies on tailored methods to ensure optimal dispersion of nanofillers within the polymer matrix. As illustrated in Figure 1, key approaches include melt blending (e.g., extrusion for thermoplastics), solution-based techniques (sol–gel, electrospinning) [40], and in situ polymerization, which enables precise control over nanoparticle formation directly within the polymeric network. For instance, melt blending is favored for industrial-scale production of clay/polypropylene composites, while sol–gel methods are ideal for creating homogeneous magnetic nanocomposites like Fe_3O_4 /chitosan for water treatment applications [41]. This methodological versatility allows engineers to fine-tune nanocomposite properties for specific functional requirements.

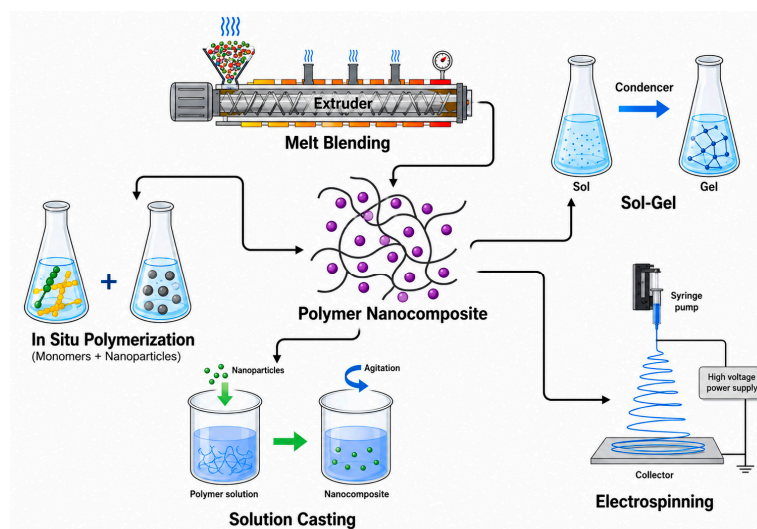


Figure 1. Schematic representation of polymer nanocomposite synthesis methods.

3. Magnetic Polymer Nanocomposites

The incorporation of ferromagnetic or superparamagnetic nanoparticles into polymer matrices proves to be particularly advantageous, as these particles have demonstrated strong potential in various emerging applications [41,42]. For many of these specialized uses, it is essential to be able to disperse the nanoparticles in a non-magnetic, easy-to-handle support. Polymers represent an ideal solution for this purpose. Numerous combinations of polymers and nanomagnetic materials have been explored for the design of magnetic nanocomposites, including magnetic nanoparticles associated with conductive polymers such as polypyrrole or iron, or with insulating polymers such as vinyl ester resins or polyurethane [42,43].

Polymer nanocomposites endowed with magnetic properties are generating significant interest, both for the in-depth study of their characteristics and for their numerous potential applications [44]. These materials represent a high-performance category of nanocomposites, incorporating magnetic nanoparticles as reinforcing agents. They have notably been highlighted for their effectiveness in the field of environmental decontamination as well as in certain therapeutic approaches [45]. Numerous studies have highlighted the use of polymer composites incorporating magnetic nanoparticles as charge agents, particularly iron-based compounds, as well as cobalt and nickel particles [46–48]. The adsorption process using these magnetic nanocomposites generally relies on four steps: (i) the capture of pollutants by adsorption, (ii) the magnetic separation of the adsorbent from the aqueous medium, (iii) the regeneration of the adsorbent, and (iv) the treatment of the solution resulting from this regeneration as well as the adsorbent itself. The effectiveness of these materials in the removal of contaminants is largely attributed to targeted functionalization and specific modification of the polymer matrix [48–50] (Figure 2).

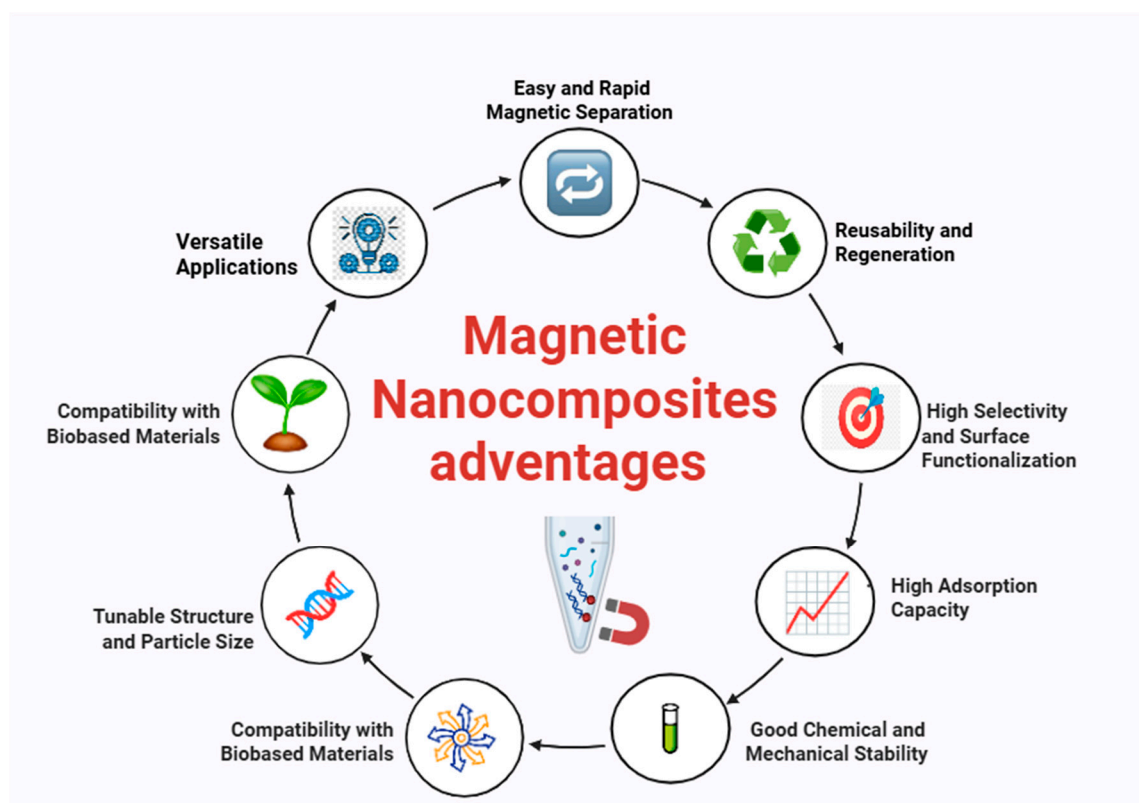


Figure 2. Functional Advantages of Magnetic Polymer Nanocomposites.

Extensive analyses have confirmed the efficacy of magnetic polymer nanocomposites in environmental and biomedical applications. A comprehensive meta-analysis comparing adsorption technologies demonstrated that these nanocomposites reduce operational costs by 30–50% compared to conventional adsorbents in large-scale wastewater treatment, primarily due to lower energy requirements for magnetic separation and enhanced reusability [51,52].

Systematic reviews have highlighted that iron oxide-based polymer composites achieve heavy metal (Pb^{2+} , Cd^{2+}) removal efficiencies exceeding 90%, with polymer functionalization ($-\text{COOH}$, $-\text{NH}_2$ groups) being the critical factor governing selectivity [53]. The surface modification of these nanocomposites significantly improves their adsorption capacity and target specificity, particularly for toxic metal ions in complex aqueous matrices [54].

One critical limitation of conventional and biobased adsorbents, despite their efficiency in pollutant removal, lies in their poor separability from aqueous media particularly in continuous flow systems [55]. To overcome this, magnetism has been increasingly explored as a recovery strategy. Magnetic separation, although historically used in water treatment since the late 19th century for coagulation and biological processes, has only recently been adapted for adsorption-based systems [56]. A key milestone in this field was the introduction of magnetic ion exchange resins (MIEX) in 1995 for the removal of natural organic matter [57]. Since then, polymer-modified magnetic particles have entered commercial use, further validating the practicality of magnetic separation techniques in large-scale operations.

4. Processing of Polymer Nanocomposites

Magnetic nanoparticles (MNP) can be synthesized using a wide variety of techniques, including the sol–gel method, flow injection, microemulsion, thermal decomposition, co-precipitation, microwave-assisted synthesis, vapor phase deposition, combustion, carbon

arc, or laser pyrolysis [58,59]. These MNP can be composed of pure metals (such as iron, cobalt, or nickel), alloys (like FePt, CoPt, MnAl, SmCo₅, Fe₁₄Nd₂B, etc.), metal oxides (for example: α -Fe₂O₃, γ -Fe₂O₃, Fe₃O₄, NiO, MnO₂, Mn₃O₄, Co₃O₄, etc.), or even MFe₂O₄-type ferrites, where M represents elements such as Li, Ni, Mg, Zn, Cu, Co, Mn, etc [60,61]. Finally, there are also complex magnetic molecular compounds, among which we can mention: {[Ni₂(mpba)₃] [Ni(dpt)(H₂O)]₆}(ClO₄)₄·12.5H₂O, 2-[4-(N-ethyl)-pyridinium]-4,4,5,5-tetramethylimidazoline-1-oxyl-3-oxide iodide, [Ni(1,3-propanediyl-bis(2-iminomethylene-6-methoxyphenol))Gd(NO₃)₃], or even [Ni(1,3-propanediyl-bis(2-iminomethylene-6-methoxyphenol))Dy(NO₃)₃], among others [62].

The methods for synthesizing polymers associated with magnetic nanoparticles reported in the literature are generally divided into two distinct categories: in situ and ex situ approaches, as illustrated in Table 3. In the context of the in situ method, magnetic nanoparticles (MNP) are formed directly within the polymer matrix, through decomposition or chemical reduction of a metallic precursor [63]. This strategy allows for precise control over the particle size, the morphology of the material, as well as other physico-chemical characteristics. The in situ approach also has the advantage of promoting well-defined interactions between the inorganic filler and the polymer, at the molecular or nanometric scale, which allows for the production of homogeneous nanocomposites and significantly limits the agglomeration of nanoparticles [64].

Table 3. Ex situ and in situ methods for magnetic polymer nanocomposites synthesis.

Approaches	Methods	Advantages
In situ	Thermal methods	- Controlled size and shape - Uniform distribution - Good contact between polymer and NPs - Well-ordered spatial arrangement
	Sol-gel processing	- High organized substances - Dense and packed layers
	Chemical vapor deposition	- High purity material (thin films, semiconductors industry)
	Co-precipitation	- Controlled size and shape - Additional thermal treatment required - Aqueous media
	Intermetallic synthesis	- High stability - Desired distribution - Core-shell NPs fabrication - Wide range of applications (better distribution for catalysis, nanoreactors)
Ex situ	Mechanical alloying	- Heterogeneity - Simplicity - Scaling up
	Reverse micellar	- Interfacial process - Controlled size
	Microencapsulation	- Stability against aggregation
	Sol-gel synthesis	- Narrow size distribution - Uniform dispersion - High porosity

Grafting	- Well-ordered packing
	- High density polymer brushes
	- Molecular scale applications

The ex situ method involves synthesizing nanoparticles beforehand, generally using soft chemistry techniques, before incorporating them into the polymer matrix [65,66]. In this context, various chemical and physical processes are used to develop polymer nanocomposites from the prepared materials [66]. This approach is widely adopted due to its flexibility, as it allows for the combination of a wide variety of nanoparticles with different types of host polymers, without major constraints [64]. Some examples of magnetic nanocomposites obtained by the ex situ technique are presented in Table 4.

One of the main challenges in the fabrication of organic-inorganic nanocomposites lies in creating a large interphase zone between the constituents. This interphase, often heterogeneous, strongly depends on the surface properties of the nanoparticles and the polymer matrix. Nevertheless, an appropriate surface modification of the nanoparticles can strengthen the interfacial interactions, improve the compatibility between the phases, and thus optimize the structure and overall performance of the nanocomposites [65–67].

Table 4. Example of magnetic polymer nanocomposites prepared by Ex situ methods.

Nanomaterials	Polymer Used	Synthesis	Ref.
Fe(III)/Ni(II) hydroxides	Alginate	To the homogeneous hydroxide suspension, sodium alginate powder was slowly added and the mixture was stirred until the total polymer dissolution was achieved. A peristaltic pump was used to dispense the suspension in a stirred reservoir containing CaCl ₂ solution used for gel formation	[68]
Fe ₃ O ₄	Polydimethylsiloxane	Oleic acid-functionalized Fe ₃ O ₄ NPs were dispersed in pre-polymer. Then, the polymer cross-linker was added and mixed. The final nanocomposite solution was casted into a mold and thermally cured	[69]
Fe/Fe-oxide	PMMA	The NPs were dispersed by sonication into a solution of PMMA powder in acetone. The solution was then spin cast until it hardened into a thin film	[70]
Fe ₃ O ₄	[Styrene- <i>b</i> -ethylene/butylene- <i>b</i> -styrene]	Fe ₃ O ₄ /polymer nanocomposites were prepared using a solution-casting method. Oleic acid modified Fe ₃ O ₄ and block copolymer were dissolved in THF, and then a film was cast over a period of 1 day	[71]
γ-Fe ₂ O ₃ /Fe ₃ O ₄ mixed phases	(Poly(ethylmethacrylate-co-methylacrylate))	NPs were dispersed in the copolymer solubilized in chloroform. After being sonicated, the resulting solution was drop casted on a glass substrate	[72]
Fe ₃ O ₄	Chitosan	NPs were dispersed in the chitosan solution by stirring at room temperature after which it was sonicated. Finally, a highly viscous solution of chitosan with uniformly dispersed Fe ₃ O ₄ NPs was obtained. The nanocomposite film was prepared by the dispersion of solution into ITO	[73]

5. Characterization Techniques for Magnetic Polymer Nanocomposites

The development of magnetic polymer nanocomposites (MPNCs) has garnered increasing attention due to their multifunctionality and potential applications in environmental remediation, biomedical devices, and data storage. A comprehensive understanding of their structural, morphological, chemical, thermal, and magnetic properties is essential for tailoring their performance. To this end, a wide range of advanced

characterization techniques is employed to prove the different physicochemical features of these hybrid systems [74].

At the core of any nanocomposite lies the polymer matrix, the magnetic nanofillers (e.g., particles, fibers, whiskers, nanotubes) [75], and the interfacial region between them. This interphase plays a crucial role in determining the overall properties of the material due to its distinct physicochemical nature compared to the bulk polymer, primarily stemming from the high surface area and strong interfacial interactions with the embedded nanoparticles. Even a low loading of magnetic nanoparticles (MNPs) can significantly modify the mechanical, thermal, and magnetic behavior of the polymer due to these interfacial effects.

5.1. Structural and Morphological Characterization

X-ray diffraction (XRD) is routinely used to assess the crystallinity, phase purity, and average crystallite size of the nanocomposite, both before and after the incorporation of MNPs. Crystallite dimensions can be estimated using the Debye–Scherrer equation. However, it should be noted that the crystallite size obtained via XRD may not directly correlate with the particle size determined by microscopic techniques [76]. Transmission electron microscopy (TEM) and scanning electron microscopy (SEM) provide high-resolution imaging for evaluating nanoparticle shape, size distribution, and dispersion state within the polymer matrix. Energy-dispersive X-ray spectroscopy (EDS), often coupled with SEM or TEM, enables semi-quantitative elemental analysis and mapping of the nanofillers.

For magnetic nanocomposites in which the matrix is soluble in volatile organic solvents (e.g., DMF, THF, CHCl_3), TEM sample preparation involves drop-casting a dilute dispersion onto a copper grid. In contrast, insoluble matrices such as Nafion require microtoming of ultrathin sections ($\leq 1 \mu\text{m}$), which are then mounted for microscopic observation. Additionally, atomic force microscopy (AFM) and scanning tunneling microscopy (STM) can be employed to investigate surface topography and local surface interactions at the nanoscale [77].

5.2. Chemical and Surface Analysis

Fourier-transform infrared spectroscopy (FTIR) is frequently used to identify functional groups present on the nanocomposite surface and to elucidate possible interactions or adsorption mechanisms with target pollutants [78]. X-ray photoelectron spectroscopy (XPS) provides complementary information regarding the surface composition and the oxidation states of the metallic elements within the nanocomposite. Nuclear magnetic resonance (NMR) spectroscopy may also be applied for studying molecular-level interactions, especially in functionalized systems [79].

5.3. Thermal and Magnetic Properties

Thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) are instrumental in assessing the thermal stability, degradation behavior, and thermal transitions of the composites. These methods also allow for the estimation of MNP loading by quantifying the inorganic residue after thermal decomposition. The incorporation of MNPs often induces notable changes in the thermal response of the polymeric host, underscoring the need for comparative studies before and after filler addition [79].

Magnetic properties are evaluated using vibrating sample magnetometry (VSM), which quantifies the magnetic moment of the material as a function of the applied magnetic field. Key parameters such as saturation magnetization, coercivity, and remanence can be extracted, offering insight into the magnetic response and potential application in magnetic separation or targeted delivery systems [80].

5.4. Simulation and Theoretical Modeling

To complement experimental studies, theoretical modeling and simulations (e.g., molecular dynamics or finite element methods) are increasingly used to predict stress-strain behavior, interfacial phenomena, and particle dispersion mechanisms [80]. These models help bridge the gap between nanoscale structure and macroscale performance, thereby supporting the rational design of advanced MPNCs [81].

Despite significant progress, achieving complete control over the structure-property relationships in MPNCs remains a major challenge. The final performance is strongly influenced by numerous synthesis parameters, including nanoparticle content, dispersion quality, interfacial interactions, and processing conditions. Therefore, an integrated approach combining multiple characterization techniques is vital to ensure a deep understanding and precise control over these multifunctional hybrid materials [82].

6. Magnetic Properties and Their Impact on Practical Applications

Magnetic metal nanoparticles exhibit a high sensitivity to oxidation due to their low stability in air. This vulnerability leads to a modification, or even a partial or total loss, of their magnetic properties. Consequently, the magnetic characteristics of these particles are closely dependent on the oxidation state of their constituents. Mastering these properties therefore requires a thorough understanding of the oxidation state of the nanoparticles, an essential condition for predicting the magnetic behavior of the obtained samples [81].

However, the precise evaluation of this oxidation state remains complex. Advanced characterization techniques, such as XANES spectroscopy (X-ray Absorption Near Edge Structure), allow access to this information. This method relies on the interaction between the incident radiation and the core electrons of the atoms, revealing details about the local electronic structure. By comparing the obtained spectra with those of well-characterized reference materials, it is possible to identify the oxidation states as well as the types of chemical bonds present in the sample [82].

Furthermore, for the study of magnetic properties, the use of a superconducting quantum interference device (SQUID) (Figure 3) is indispensable. This type of instrument is extremely sensitive to variations in the magnetic moment and allows for precise plotting of magnetization curves. During the analysis, the sample is subjected to a variable magnetic field at room temperature, which allows the magnetization loops to be recorded throughout the process [83].

As a general rule, ferromagnetic materials exhibit well-defined hysteresis curves, indicative of the remanence and coercivity typical of this type of behavior. In contrast, superparamagnetic materials often composed of ferromagnetic particles at the nanometric scale do not exhibit notable hysteresis, although they can display high magnetic saturation. This absence of hysteresis is due to the rapid fluctuation of individual magnetic moments under the influence of thermal agitation, which distinctly differentiates their behavior from that of conventional ferromagnetic materials [84].

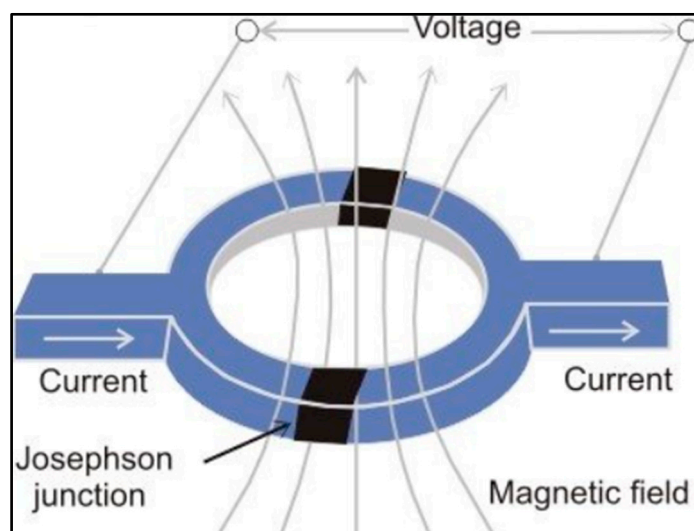


Figure 3. Schematic representation of a Superconducting Quantum Interference Device (SQUID).

The nanostructured material obtained from polyaniline and hematite nanoparticles, in the presence of naphthalene sulfonic acid as a dopant, exhibits electrical and magnetic properties at room temperature [85]. According to the synthesis conditions, this material consists of polyaniline nanotubes or nanorods, with a diameter between 80 and 100 nm, containing Fe_3O_4 nanoparticles of approximately 10 nm in diameter. For a Fe_3O_4 content of 20% by weight, the saturation magnetization (M_s) reaches $6 \text{ emu} \cdot \text{g}^{-1}$. Furthermore, the fabrication of Fe_3O_4 -PANI nanocomposites without the use of an external oxidant has been reported [84]. A mixture of ferrous and ferric chlorides has been used to simultaneously obtain PA and Fe_3O_4 nanoparticles. The conductivity of the resulting nanocomposite is $\sim 10^{-4} \text{ S cm}^{-1}$; $M_s = 48.4\text{--}80.4 \text{ emu g}^{-1}$ [86].

Burke and collaborators [87], reported saturation magnetization values ranging from 0.9 to $37.8 \text{ A} \cdot \text{m}^2/\text{kg}$ for different formulations of polymer-coated iron nanoparticles. The samples with the highest iron content exhibited the most significant magnetizations. The majority of these materials exhibited hysteretic behavior at room temperature, with coercivity dependent on the diameter of the magnetic cores, reaching a maximum when it approached 20 nm.

Furthermore, biocompatible ferrofluids containing between 10 and 40% by weight of hybrid Fe_3O_4 nanoparticles were obtained without exhibiting any hysteresis phenomenon [61,88], indicating superparamagnetic behavior. Similarly, core/shell nanoparticles composed of Fe_2O_3 coated with a polymeric layer, containing approximately 15.3% by weight of $\gamma\text{-Fe}_2\text{O}_3$, exhibited a saturation magnetization of around $30 \text{ A} \cdot \text{m}^2/\text{kg}$, also confirming superparamagnetic behavior [89].

In the case of polypyrrole (PPy)-based nanocomposites, the incorporation of 20 to 50% by weight of Fe_2O_3 nanoparticles resulted in saturation magnetizations ranging from 30 to $45 \text{ A} \cdot \text{m}^2/\text{kg}$. A notable improvement in electrical conductivity was also observed compared to pure PPy [61,90]. Finally, nanocomposites consisting of polyaniline containing 12.37% by weight of Fe_3O_4 have been identified as superparamagnetic, with a saturation magnetization of $3.88 \text{ A} \cdot \text{m}^2/\text{kg}$ [61,91].

6.1. Saturation Magnetization and Separation Kinetics

The saturation magnetization (M_s), typically measured in emu/g via Vibrating Sample Magnetometry (VSM), represents the maximum magnetic moment the nanocomposite can achieve in an applied external magnetic field. In practical wastewater treatment scenarios, M_s directly governs the kinetics of the solid–liquid separation process. MPNCs generally exhibit a lower M_s (often ranging from 15 to 50 emu/g) compared to bare magnetic nanoparticles (e.g., bulk Fe_3O_4 at ~92 emu/g) because the encapsulating polymer matrix acts as a diamagnetic, non-responsive “dead weight” that shields the magnetic domains. However, an M_s value exceeding approximately 15 emu/g is widely considered the threshold required to achieve rapid and complete magnetic separation from an aqueous medium within seconds to minutes using standard neodymium magnets. This rapid separation capability is what allows MPNCs to overcome the primary limitation of traditional nanoscale adsorbents namely, the need for costly, time-consuming ultracentrifugation or membrane filtration to prevent secondary nanoparticle pollution in the treated effluent.

6.2. Superparamagnetism and Adsorbent Regeneration

Beyond absolute magnetic strength, the specific magnetic behavior specifically superparamagnetism is a core critical factor for the lifecycle of the adsorbent. When magnetic nanoparticles (such as Fe_3O_4 or gamma- Fe_2O_3) are synthesized below a critical domain size (typically < 20 nm), they exhibit superparamagnetic characteristics, defined by near-zero remanence (M_r) and coercivity (H_c). In practical application scenarios, this means the MPNCs become highly magnetic when a field is applied (allowing for extraction) but lose all magnetic memory the instant the external magnet is removed. If the materials were ferromagnetic instead, they would retain residual magnetism and permanently clump together into large aggregates. Such agglomeration would drastically reduce the specific surface area and block the polymeric active sites, rendering the material useless for subsequent adsorption cycles. Thus, superparamagnetism is the fundamental mechanism that allows MPNCs to be easily redispersed in regeneration eluents and subsequently reused for multiple wastewater treatment cycles without a loss of adsorption efficiency.

6.3. The Protective Role of the Polymer Matrix in Extreme Environments

Finally, the design of MPNCs inherently involves a deliberate trade-off between maximizing magnetic responsiveness and ensuring environmental durability. Industrial effluents often present harsh, highly acidic conditions that can rapidly dissolve bare iron oxide nanoparticles, leading to a complete loss of magnetic separability and the unwanted leaching of iron ions into the water. By coating the magnetic cores with a dense polymer layer, researchers sacrifice a portion of the initial saturation magnetization to gain critical chemical shielding. The microstructural integrity provided by the polymer shell ensures that the core retains enough magnetic performance to remain recoverable even after prolonged exposure to corrosive pollutants.

7. Structure-Property Relationships in MPNC Adsorption

To understand the environmental application performance of the magnetic polymer nanocomposites (MPNCs) listed in Tables 5 and 6, it is critical to elucidate the intrinsic correlation mechanisms between their specific microstructural features and their adsorption capacities. The core critical factors governing adsorption capacity include the specific surface area, the density and type of functional groups introduced by the polymer matrix, and the structural arrangement of the magnetic nanoparticles.

7.1. The Role of Specific Surface Area and Porosity

The specific surface area is a primary determinant of a material's maximum adsorption capacity ($q_{\max}$) because it dictates the number of accessible active sites. For example, the ZnFe_2O_4 /chitosan magnetic particles exhibit a specific surface area of $57.4 \text{ m}^2/\text{g}$, facilitating physical adsorption mechanisms like pore-filling and hydrogen bonding, leading to a high diclofenac adsorption capacity of 188 mg/g . Conversely, the magnetic zinc ferrite-chitosan biocomposite possesses a much lower specific surface area ($5.187 \text{ m}^2/\text{g}$), which directly correlates to its substantially lower adsorption capacities for crystal violet and brilliant green. Furthermore, constructing porous 3D structures, such as the Chitosan/PVA magnetic hydrogel beads, provides a highly permeable microstructure that reduces intra-particle diffusion resistance, resulting in an exceptional Congo Red adsorption capacity of 470.1 mg/g .

7.2. Polymer Functionalization and Active Sites

The surface chemistry introduced by the encapsulating polymers drives the selectivity and primary interaction mechanisms between the MPNCs and the target pollutants.

- **Amine and Hydroxyl Groups:** Chitosan matrices are rich in $-\text{NH}_2$ and $-\text{OH}$ groups. When functionalized further, such as in the magnetic amine-functionalized chitosan or the amino-functionalized magnetic cellulose, the increased density of amine groups provides excellent electrostatic attraction and surface complexation sites. This microstructural enhancement explains the high diclofenac sodium capacity (469.48 mg/g) and the high Cr(VI) removal (171.5 mg/g).
- **Conjugated Polymers:** The use of conductive polymers like polypyrrole (PPy) and polyaniline introduces highly active nitrogen heteroatoms and π -electron systems. The polypyrrole-modified $\text{Fe}_3\text{O}_4/\text{SiO}_2$ composite achieves massive adsorption capacities for both Congo red (361.43 mg/g) and Cr(VI) (298.22 mg/g) due to robust π - π stacking interactions, hydrogen bonding, and ion exchange mechanisms driven by these functional groups.

7.3. Core–Shell and Multicomponent Architectures

The spatial arrangement of the magnetic core and polymer shell is critical for maintaining stability and reusability. Encapsulating Fe_3O_4 within robust matrices like silica, polyimide, or cross-linked copolymers creates a protective core–shell microstructure. This protects the magnetic iron oxide from acid dissolution or oxidative degradation during heavy metal removal (e.g., Pb^{2+} , Cr^{3+}). Additionally, incorporating 2D materials like graphene oxide (as seen in the Chitosan/Graphene Oxide/ Fe_3O_4 composite) creates a synergistic structure that facilitates multiple simultaneous mechanisms such as electrostatic interaction, surface complexation, and ion exchange preventing the agglomeration of nanoparticles and ensuring that the high specific surface area is preserved over multiple cycles.

8. Environmental Application

Environmental nanotechnology represents a major advancement in the field of engineering and research applied to environmental protection. The nanoscale allows for the design of materials with unprecedented properties, capable of effectively addressing complex environmental issues, such as the catalytic degradation of pollutants, selective adsorption, and even the ultra-sensitive detection of toxic substances [92,93].

Polymer nanocomposites (PNCs), which combine organic polymers with nanoparticles, have gained popularity due to their excellent mechanical stability, chemical compatibility, and high specific surface area. Nanofillers offer exceptional interfacial reactivities and enable the emergence of synergistic physicochemical properties. Moreover, the

integration of magnetic particles into these polymer matrices (MPNCs) addresses several constraints related to the direct use of nanoparticles, particularly their post-treatment separation and reuse [94,95].

Water treatment remains a critical issue in the current environmental context. Water sources are often contaminated by heavy metals, industrial dyes, persistent organic compounds, or pathogenic microorganisms. Conventional treatment technologies, although widespread, are often ineffective, costly, or not sustainable. In this context, materials derived from nanotechnology, particularly MPNCs, offer innovative, economical, and environmentally viable solutions.

Figure 4 illustrates the role of magnetic separation in the removal of heavy metals using magnetic nanocomposites. Various materials have been studied for this purpose, including maghemite (Fe_2O_3) and magnetite (Fe_3O_4) nanoparticles, modified and stabilized by agents such as humic acid, polyacrylic acid functionalized with amine groups, or multilayer magnetic carbon nanotubes incorporating Fe_2O_3 [96]. Other systems include magnetic composite particles based on Fe_3O_4 and activated carbon, as well as biopolymers such as gum arabic, chitosan, or various polysaccharides, which have been tested for their effectiveness in the adsorption of metal ions [97].

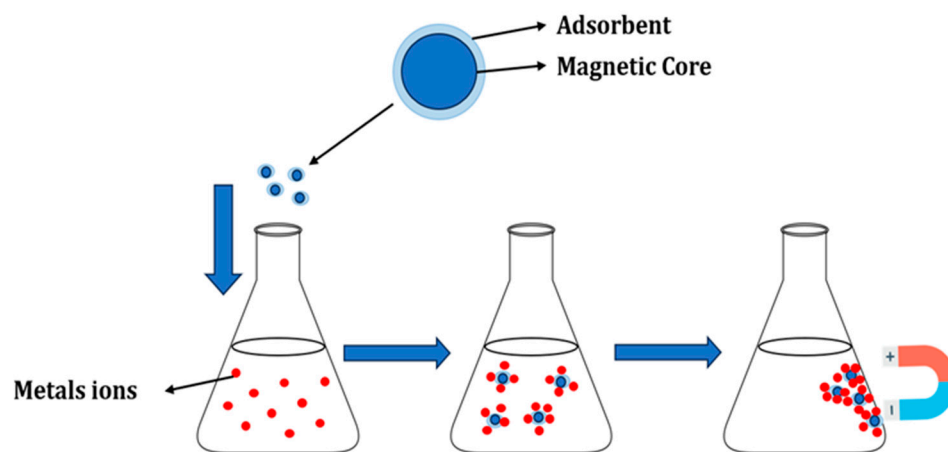


Figure 4. Schematic representation of magnetic separation of adsorbed metal ions from aqueous medium using magnetic Nanocomposite [98].

8.1. Removal of Toxic Dyes and Organic Effluents

The pollution of aquatic environments by synthetic dyes, pharmaceutical residues, and various organic effluents constitutes a major environmental issue [99]. These compounds are often resistant to natural biodegradation and can exhibit acute or chronic toxicity [100]. In this context, magnetic polymer nanocomposites (MPNCs) have emerged as efficient materials for the removal of these pollutants, thanks to their multifunctional structure and ease of separation by magnetic field [101].

Among these materials, chitosan-based composites, a biopolymer obtained from chitin, have proven to be particularly effective [102]. Thanks to the presence of amine groups, chitosan exhibits a strong affinity for anionic pollutants. Its association with magnetic metal oxides such as ZnFe_2O_4 or CoFe_2O_4 allows for the design of highly efficient hybrid adsorbents. For example, ZnFe_2O_4 /chitosan particles have shown an adsorption capacity of 188 mg/g for diclofenac, a commonly found anti-inflammatory in pharmaceutical effluents. Similarly, a biocomposite based on zinc ferrite and chitosan has demonstrated good efficiency in the removal of cationic dyes such as crystal violet and brilliant green.

Polypyrrole-doped composites (PPy), such as $\text{Fe}_3\text{O}_4/\text{SiO}_2@\text{PPy}$, offer a dual functionality of adsorption and reduction, with remarkable performances reaching 361.43 mg/g

for Congo red and 298.22 mg/g for Cr(VI). Their high porosity, along with the π - π interactions between aromatic pollutants and the matrix, explains their enhanced efficiency. Moreover, porous adsorbents based on magnetic β -cyclodextrin have demonstrated an adsorption capacity of 305.8 mg/g for methylene blue, confirming the interest of these functionalized macrocyclic structures. Finally, Zhu et al. developed magnetic hydrogel beads based on chitosan/poly (vinyl alcohol), achieving a remarkable adsorption capacity of 470.1 mg/g for Congo red, even surpassing the performance of chitosan/carbon nanotube composites.

Table 5 presents a summary of the most representative MPNCs used for the removal of toxic dyes and organic effluents, highlighting their specific surface area, targeted pollutants, and adsorption capacity.

Table 5. Magnetic Polymer Nanocomposites and Their Adsorptive Efficiency Toward Organic Contaminants.

Adsorbent	Specific Surface Area (m ² /g)	Target Pollutant	Adsorption Capacity (mg/g)	Primary Adsorption Mechanism	Reference
ZnFe ₂ O ₄ /chitosan magnetic particles	57.4	Diclofenac	188	Hydrogen bonding, pore filling	[103]
Magnetic zinc ferrite–chitosan biocomposite	5.187	Crystal violet/Brilliant green	14.3/20.0	Electrostatic interactions, hydrogen bonding	[104]
CoFe ₂ O ₄ –chitosan composite	2	Congo red/Methyl orange	15.60/66.18	Electrostatic interactions, hydrogen bonding	[105]
Polypyrrole-modified Fe ₃ O ₄ /SiO ₂ composite	-	Congo red/Cr(VI)	361.43/298.22	π - π interactions, hydrogen bonding, ion exchange	[106]
Magnetic amine-functionalized chitosan	-	Diclofenac sodium	469.48	Electrostatic attraction, surface complexation	[107]
Magnetic β -cyclodextrin porous polymer nanospheres	70.63	Methylene blue	305.8	Host-guest inclusion, electrostatic interaction	[108]
Chitosan/PVA magnetic hydrogel beads	-	Congo red	470.1	Hydrogen bonding, intra-particle diffusion	[109]
Polyaniline-coated Fe ₃ O ₄ nanoparticles	-	Polycyclic Aromatic Hydrocarbons (PAHs)	471	π - π stacking, hydrophobic interactions	[110]
Magnetic polyimide@Mg-Fe-LDH core-shell composite	-	Tetracycline/2,4-DCP/Glyphosate	345	Surface complexation, π - π interactions, electrostatic	[111]

It should be noted that the adsorption capacities ($q_{\max}$) summarized in Tables 5 and 6 represent the theoretical maximums achieved under the optimized experimental conditions specific to each independent study. Comparing these optimized Langmuir capacities allows for a reliable evaluation of the highest intrinsic potential of each nanocomposite. However, when assessing these materials for practical wastewater treatment, readers must consider the specific optimal conditions required to achieve these capacities; an adsorbent demonstrating a high $q_{\max}$ exclusively at extreme pH values may be less viable for large-scale environmental deployment than one operating efficiently at neutral pH.

8.2. Elimination of Heavy Metals by Magnetic Polymer Nanocomposites

The contamination of water by heavy metals constitutes a major environmental issue due to their toxicity, persistence in the environment, and ability to accumulate in living organisms [110]. Unlike organic pollutants, heavy metals such as Pb²⁺, Cd²⁺, Cr⁶⁺, Zn²⁺, Ni²⁺, or V⁵⁺ are not biodegradable and can cause serious health effects, including kidney failure, metabolic disorders, cancers, or neurological damage [111]. These metal ions mainly originate from industrial activities such as mining, electroplating, pigment and glass production, or battery manufacturing. Thus, their efficient removal from wastewater remains a critical environmental challenge [112].

Among the various treatment techniques chemical precipitation, ion exchange, membrane filtration, electrochemical methods adsorption stands out as the most promising due to its simplicity, high efficiency, and low cost. In this context, magnetic polymer

nanocomposites (MPNCs) are attracting growing interest, offering dual functionality: high adsorption capacity and easy separation from the aqueous medium using an external magnetic field [113].

These materials typically consist of magnetic nanoparticles (e.g., Fe_3O_4) embedded in or coated with a polymeric matrix such as chitosan, cellulose, polypyrrole, or synthetic copolymers which provides stability, water dispersibility, and abundant functional groups (hydroxyl, carboxyl, amine) that act as active adsorption sites for metal ions. The presence of a magnetic core allows rapid recovery of the adsorbent after saturation.

Numerous studies have investigated various MPNCs for the removal of toxic metals. For example, Ge et al. [105] showed that polymer-coated Fe_3O_4 nanoparticles exhibited significantly enhanced adsorption capacities for Cd^{2+} , Zn^{2+} , Pb^{2+} , and Cu^{2+} ions compared to uncoated Fe_3O_4 , with optimal adsorption at pH 5.5. Similarly, chitosan–graphene oxide–magnetite composites demonstrated a Pb^{2+} adsorption capacity of 76.94 mg/g [106], while chitosan-coated magnetite achieved 53.6% removal efficiency for the same ion [107].

Other functional polymers such as ethylenediamine or polypyrrole have also been used to enhance metal binding. Zhao et al. [108] developed ethylenediamine-functionalized magnetic polymers capable of effectively removing Cr(VI), although a decrease in magnetic properties was observed compared to unmodified Fe_3O_4 . In another approach, hybrid nanocomposites based on Fe_3O_4 @polypyrrole anchored on graphene nanosheets exhibited excellent Cr(VI) removal, attributed to the synergistic interaction between the components [109]. Table 6 summarizes representative MPNCs and their performances for heavy metal ion removal.

Table 6. Adsorption performance of selected magnetic polymer nanocomposites for heavy metal removal.

Material	Target Metal Ions	Adsorption Capacity/Observation	Primary Adsorption Mechanism	Reference
Chitosan/Graphene Oxide/ Fe_3O_4	Pb^{2+}	76.94 mg/g	Electrostatic interaction, surface complexation, ion exchange	[110]
Chitosan-coated Fe_3O_4	Pb^{2+}	53.6% removal efficiency	Chelation with amine/hydroxyl groups	[111]
Fe_3O_4 -modified copolymer (MMA/MAH)	Co^{2+} , Cr^{3+} , Zn^{2+} , Cd^{2+}	90.09–111.11 mg/g	Chelation, electrostatic attraction	[112]
Amino-functionalized magnetic cellulose	Cr(VI)	171.5 mg/g	Electrostatic attraction, reduction, surface complexation	[113]
Fe/resin composite	Cr(VI), Pb(II)	Remediation 30× faster than Fe powder; 21× better for Cr(VI)	Chemical reduction, coprecipitation, complexation	[114]
Fe_3O_4 @Polypyrrole/Graphene	Cr(VI)	Excellent efficiency due to synergistic structure	Ion exchange, electrostatic attraction, reduction	[115]
Polypyrrole/ Fe_3O_4 (PPy/ Fe_3O_4)	Cr(VI)	Efficient in batch; needs scaling for continuous systems	Ion exchange, reduction to Cr(III), electrostatic	[116]
Magnetic zeolite-polypyrrole (clinoptilolite/PPy)	V^{5+}	75% removal at pH 5; Langmuir isotherm confirmed	Ion exchange, electrostatic attraction	[117]
Polymer-modified Fe_3O_4	Cd^{2+} , Zn^{2+} , Pb^{2+} , Cu^{2+}	Higher removal than bare Fe_3O_4	Chelation, surface complexation	[108]

The reported adsorption capacities illustrate the efficiency of MPNCs, particularly those with carboxylic, amino, or pyrrolic groups. High values were observed for composites like amino-functionalized cellulose (171.5 mg/g) for Cr(VI) and magnetic hydrogel based on chitosan and cellulose (171.0 mg/g) for Pb^{2+} . These high performances stem from the strong affinity of metal ions with active functional groups.

A particularly notable case is the Fe/resin composite, which showed exceptional kinetic performance [118]. The initial removal rates of Cr(VI) and Pb(II) were 30 times faster than with conventional iron, and the Cr(VI) reduction was 21 times higher after two months of contact. While the adsorption capacity was not quantified, the rapid kinetics position this material as a promising option for long-term water treatment.

Additional innovations, such as magnetic zeolite-polypyrrole composites, have proven highly effective for vanadium (V^{5+}) removal, achieving 75% removal at pH 5, following Langmuir-type adsorption. Their low-cost base (clinoptilolite) and strong performance make them excellent candidates for large-scale treatment of industrial effluents.

Similarly, amino-functionalized porous magnetic glycidyl methacrylate (GMA) copolymers have emerged as another promising class of multifunctional magnetic adsorbents for wastewater remediation. Their highly porous structure, large specific surface area, and abundant amino functional groups provide numerous active binding sites for contaminants. These materials are typically synthesized by copolymerizing glycidyl methacrylate with suitable cross-linkers, followed by the incorporation of Fe_3O_4 nanoparticles and amino functionalization through epoxy ring-opening reactions. The resulting adsorbents exhibit excellent adsorption performance toward heavy metal ions, textile dyes, and a wide range of organic pollutants through chelation, electrostatic attraction, and hydrogen-bonding interactions. Furthermore, the incorporation of magnetic nanoparticles enables rapid and efficient separation of the adsorbent from aqueous media using an external magnetic field. Owing to their high adsorption capacity, fast adsorption kinetics, excellent selectivity, and remarkable regeneration and reusability, amino-functionalized magnetic GMA copolymers represent highly promising multifunctional materials for the sustainable removal of both inorganic and organic contaminants from wastewater. Representative examples of amino-functionalized porous magnetic GMA-based adsorbents and their adsorption performance are summarized in Table 7 [119].

A key reason why certain magnetic polymer nanocomposites outperform others is that adsorption capacity is not governed by a single factor, but by the synergy between surface chemistry, accessible surface area, pore structure, and interfacial architecture. In particular, materials based on chitosan, polypyrrole, polyaniline, or graphene oxide often exhibit superior performance because they provide a higher density of active sites such as amino, hydroxyl, and heteroaromatic groups, which promote electrostatic attraction, complexation, hydrogen bonding, and π - π interactions with target pollutants. By contrast, less functionalized systems generally offer fewer binding sites and weaker affinity for adsorbates. In addition, hierarchical porosity and good nanoparticle dispersion improve mass transfer and expose more active sites to solution, while core-shell or hybrid architectures help maintain structural stability and reusability during repeated adsorption-desorption cycles. Therefore, the superior performance of a given MPNC should be interpreted as the result of deliberate microstructural design rather than simply the identity of its components.

In conclusion, magnetic polymer nanocomposites offer a powerful and adaptable platform for the remediation of heavy metal-contaminated waters. Their high adsorption efficiency, magnetic separability, and tunable surface chemistry make them highly attractive for practical applications. Future research should focus on improving the recyclability and selectivity of these materials, as well as developing scalable synthesis methods for industrial deployment.

Table 7. Representative amino-functionalized magnetic glycidyl methacrylate (GMA) copolymers for the removal of textile dyes and other pollutants from aqueous solutions [119].

Adsorbent	Modification	Target Pollutant	Maximum Adsorption Capacity	Adsorption Model	Main Advantages
MCS-GMA-I	Magnetic chitosan-glycidyl methacrylate modified with diethylenetriamine	Remazol Brilliant Blue R (RBBR)	0.153 mmol g ⁻¹ (pH 2, 25 °C)	Langmuir isotherm; pseudo-second-order kinetics	Porous structure, high affinity for dye adsorption, spontaneous and endothermic adsorption, easy magnetic separation
MCS-GMA-II	Epichlorohydrin-crosslinked magnetic chitosan-glycidyl	Remazol Brilliant Blue R (RBBR)	0.153 mmol g ⁻¹ (pH 2, 25 °C)	Langmuir isotherm; pseudo-second-order kinetics	Higher amino-group density, porous morphology, excellent reusability, efficient magnetic recovery

methacrylate modified with diethylenetriamine

8.3. Oil Removal

MPNCs have emerged as highly effective materials for the removal of oils from contaminated waters, due to their unique combination of hydrophobic polymer matrices and magnetic cores. This synergy allows for selective adsorption of hydrocarbons, followed by easy recovery via external magnetic fields, thus representing a sustainable and effective alternative to conventional depollution methods [120].

For example, polystyrene-coated Fe_3O_4 nanoparticles ($\text{Fe}_3\text{O}_4@\text{PS}$) exhibit high hydrophobicity and can adsorb up to three times their weight in lubricating oil, with rapid separation by magnetization [121]. Other studies have highlighted the effectiveness of magnetite nanoparticles functionalized with polymethacrylate (PMA), which exhibit both a high oil absorption capacity and efficient recovery through superconducting magnetic separation [121].

Furthermore, magnetic nanocomposites based on polystyrene have been developed for oil spill cleanup operations, while alkyd resin-based composites incorporating Fe_2O_3 have been applied to the treatment of industrial wastewater contaminated with cutting oils. Owing to their high adsorption capacity, magnetic recoverability, reusability, energy efficiency, and selective affinity toward hydrocarbons, these materials offer significant advantages over conventional adsorbents, making them highly promising for oil spill remediation and the treatment of oily wastewater. To provide a systematic comparison of the representative magnetic polymer nanocomposites (MPNCs) reported for oil removal, their polymer matrices, magnetic cores, target oils, and oil absorption capacities are summarized in Table 8 [122].

Table 8. Representative magnetic polymer nanocomposites (MPNCs) used for oil removal from aqueous media [123].

Magnetic Polymer Nanocomposite	Polymer Matrix	Magnetic Core	Target Oil	Oil Absorption Capacity
WS@ Fe_3O_4 MNPs/SA	Wheat straw (WS) functionalized with stearic acid (SA)	Fe_3O_4 magnetic nanoparticles (MNPs)	Used motor oil (oil/water separation in seawater)	12.444 g g ⁻¹
WS@ Fe_3O_4 MNPs	Wheat straw (WS)	Fe_3O_4 magnetic nanoparticles (MNPs)	Used motor oil	3.90 g g ⁻¹ (at WS: Fe_3O_4 = 3:1)

9. Regeneration and Reusability of Magnetic Polymer Nanocomposites

Regeneration and reuse are among the most significant advantages of magnetic polymer nanocomposites (MPNCs), as they substantially reduce operational costs while minimizing secondary waste generation. Unlike conventional adsorbents, MPNCs can be rapidly and efficiently separated from aqueous solutions using an external magnetic field, which greatly simplifies the recovery process and enables multiple adsorption–desorption cycles without complex filtration or centrifugation steps. Consequently, the regeneration capability of MPNCs plays a crucial role in determining the economic viability and long-term sustainability of adsorption-based wastewater treatment technologies [123].

Regeneration generally involves the desorption of adsorbed pollutants from the active adsorption sites while maintaining the structural integrity and adsorption efficiency of the nanocomposite. The choice of regenerating agent depends primarily on the nature of the target pollutant and the adsorption mechanism involved. Dilute hydrochloric acid (HCl) or nitric acid (HNO_3) is commonly employed for the desorption of heavy metal ions, whereas sodium hydroxide (NaOH) solutions are preferred for acidic dyes. Ethylenediaminetetraacetic acid (EDTA) is particularly effective for removing strongly chelated metal

ions, while organic solvents such as ethanol and methanol are frequently used for desorbing organic contaminants. These regeneration processes are mainly governed by mechanisms including ion exchange, protonation–deprotonation, chelation, and electrostatic repulsion, which facilitate pollutant desorption and enable the adsorbent to be reused over successive treatment cycles with minimal loss of performance. For a systematic comparison of the regeneration conditions, mechanisms, desorption agents, and reusability of representative magnetic polymer nanocomposites (MPNCs), the available data are summarized in Table 9 [124].

Table 9. Regeneration of representative magnetic polymer nanocomposites used in wastewater treatment [125].

Adsorbent	Pollutant	Regeneration Agent	Adsorption–Desorption Cycles
Chitosan/Fe ₃ O ₄	Pb ²⁺	HCl	5
Amino-functionalized magnetic cellulose	Cr(VI)	NaOH	5
Magnetic zeolite–polypyrrole	V ⁵⁺	Acid washing	Several cycles
MCS–GMA-I	Remazol Brilliant Blue R	0.5 M NaOH	Multiple cycles

10. Contribution of Density Functional Theory (DFT) in the Study of Magnetic Polymer Nanocomposites

Following the discussion of regeneration and reusability, Density Functional Theory (DFT) has emerged as a powerful computational tool for understanding and optimizing the performance of magnetic polymer nanocomposites (MPNCs) [126–129]. By evaluating adsorption energies, charge transfer, frontier molecular orbitals (HOMO/LUMO), electronic structure, and active adsorption sites, DFT provides molecular-level insights into adsorption mechanisms and complements experimental investigations [130]. These computational studies support the rational design of MPNCs with enhanced adsorption capacity, selectivity, structural stability, and regeneration performance. Recent research has increasingly combined DFT with experimental studies to investigate porous amino-functionalized polymers for the removal of heavy and precious metals, textile dyes, pesticides, and other persistent organic pollutants [131]. For metal ions such as Pb(II), Hg(II), Cd(II), Cu(II), Au(III), Pd(II), and Pt(IV), DFT demonstrates that adsorption is mainly driven by coordination between amino nitrogen atoms and metal ions, resulting in stable metal–ligand complexes with high binding energies. For dyes and pesticides, adsorption is governed by hydrogen bonding, electrostatic attraction, π – π interactions, van der Waals forces, and pore-filling effects, while DFT identifies amino groups as the principal active adsorption sites and predicts the most favorable adsorption configurations [132]. Overall, the integration of experimental investigations with DFT and quantum chemical calculations has become an indispensable strategy for elucidating pollutant–adsorbent interactions and accelerating the development of next-generation porous amino-functionalized magnetic polymer adsorbents with improved adsorption efficiency, selectivity, regeneration capability, and long-term stability for sustainable wastewater treatment [133–136].

10.1. Elucidation of Mechanisms and Non-Covalent Interactions

The primary contribution of DFT lies in its ability to quantify the thermodynamic stability of the adsorption complex through the calculation of the adsorption energy (E_{ads}). By computing the energy difference between the isolated components and the formed complex, DFT allows researchers to definitively classify the dominant removal mechanism. Highly negative E_{ads} values typically denote strong chemisorption via electron sharing, whereas less negative values indicate physisorption driven by van der Waals

forces. Furthermore, Bader charge analysis and the mapping of charge density differences reveal the exact direction and magnitude of electron transfer between the pollutants and the active functional groups of the polymer matrix [137].

In addition to quantifying binding energies, advanced DFT-based visualization techniques are crucial for validating the complex mechanisms proposed in experimental studies. Tools such as Electrostatic Potential (ESP) mapping pinpoint the most nucleophilic and electrophilic regions on the MPNC surface, predicting exactly where a cationic heavy metal or an anionic dye will initially attack. Moreover, Non-Covalent Interaction (NCI) plots and Independent Gradient Models (IGM) are increasingly utilized to visually map the spatial regions of hydrogen bonding and pi-pi stacking interactions, which are critical for nanocomposites featuring conjugated polymers.

10.2. DFT-Guided Rational Material Design

Beyond explaining existing materials, DFT is now a critical driver for rational material design. Traditional trial-and-error synthesis of MPNCs is highly resource-intensive. DFT calculations circumvent this bottleneck by enabling the computational screening of various polymer functionalizations prior to laboratory synthesis. By modeling different spatial arrangements and evaluating how modifications alter the HOMO-LUMO gap (and thus the chemical reactivity of the composite), researchers can predict which structural configurations will yield the highest affinity and selectivity. This predictive capability ensures that future MPNCs are engineered with highly optimized microstructures tailored specifically for their intended target pollutants.

10.3. Validated Literature Case Studies

In an initial study, Zeinab Hosseini et al. [138], employed DFT calculations to explore the adsorption of asphaltenes on bare Fe_3O_4 surfaces as well as on surfaces coated with polyaniline (PANI). The results showed that the asphaltene/ Fe_3O_4 interactions were relatively weak ($E_{\text{ads}} = -54.39$ kJ/mol), occurring through aromatic-iron contacts and a low charge transfer ($\sim 0.10 e^-$). In contrast, the introduction of PANI on the surface of Fe_3O_4 significantly improves the adsorption energy ($E_{\text{ads}} = -96.23$ kJ/mol) due to a favorable HOMO-LUMO interaction between the π orbitals of the asphaltenes and the π^* orbitals of the PANI. This enhancement is due to a more significant electronic transfer ($0.32 e^-$) and polarization induced by the magnetic support, as confirmed by the analysis of projected density of states (PDOS) [137]. These results agree with the experimental data, confirming the synergy between Fe_3O_4 and PANI in the adsorption of heavy compounds from crude oils.

Another contribution, led by Zhengwen Wei et al. [138], combined phenanthrene adsorption experiments (a polycyclic aromatic hydrocarbon) with DFT simulations on a Fe_3O_4 nanocomposite modified by benzhydrylamine ($\text{Fe}_3\text{O}_4\text{-1.5BENZHY}$). Four adsorption configurations were modeled, revealing π - π interactions of the “offset parallel stacking” type between the aromatic nuclei of phenanthrene and the benzhydryl groups. The adsorption energies were all negative (up to -0.0169 u.a.), indicating a strong stability of the formed complexes [139]. The study of molecular orbitals showed an electronic transfer between the HOMO of $\text{Fe}_3\text{O}_4\text{-1.5BENZHY}$ and the LUMO of phenanthrene, accompanied by a reduction in the HOMO-LUMO gap (from 0.293 to 0.243 u.a.), indicating a more reactive electronic state after adsorption. These results were perfectly correlated with experimental data showing an increased adsorption capacity of the functionalized material and highlight the importance of aromatic groups to maximize π - π interactions with PAHs.

In a third study, Soleimani Lashkenari et al. [140], applied DFT to evaluate the interaction of lead (Pb^{2+}) with an $\text{Fe}_3\text{O}_4/\text{PTh}$ (polythiophene) nanocomposite. The calculations showed that the main active site was located on the surface of Fe_3O_4 , where Pb^{2+} forms

strong electrostatic interactions with the oxygen atoms (distance ~ 2.2 Å), with a high adsorption energy of -272.11 kJ/mol, compared to -53.44 kJ/mol on the surface of PTh alone. The electrostatic potential mapping confirmed the presence of localized negative zones around the Fe_3O_4 oxygens, identified as the preferential sites. Although the electronic contribution of PTh is limited, it contributes to the stabilization of the composite without significantly altering its electronic structure (HOMO–LUMO gap almost unchanged). This hybrid physisorption/chemisorption configuration provides the material with good thermodynamic stability ($E_f = -89.12$ kJ/mol) and an interesting regeneration capacity, experimentally validated over several cycles.

These studies demonstrate that DFT not only confirms experimental trends but also predicts and rationalizes the effects of polymeric functionalization, the crystalline structure of the magnetic substrate, and the nature of the target pollutant. The theoretical approach thus plays an essential role in the design of high-performance nanocomposites for the depollution of aqueous environments [141].

11. Current Challenges and Future Perspectives

The field of magnetic polymer nanocomposites (MPNCs) applied to water depollution is experiencing rapid growth, driven by the urgent need for sustainable and efficient solutions to combat water pollution. Future research should focus on the development of advanced surface functionalization techniques to improve the selectivity and adsorption capacity of MPNCs for a wide range of pollutants, including heavy metals, organic dyes, pharmaceuticals, and microplastics. In this regard, digital tools such as molecular modeling or artificial intelligence could play a key role in optimizing functional structures. It is also essential to promote green and scalable synthesis methods, with a low environmental impact, using little or no toxic solvents, and suitable for continuous and reproducible industrial production. The design of multifunctional MPNCs, capable of combining adsorption, catalytic degradation, and antibacterial activity, constitutes a promising approach for treating complex effluents. Moreover, the evaluation of their long-term stability, resistance to fouling or chemical degradation, as well as their ability to be effectively regenerated without significant loss of activity, is crucial for considering practical application. In-depth studies on the environmental and health impact of MPNCs must also be conducted, particularly on their life cycle, ecotoxicity, and the potential risk of nanoparticle release into ecosystems. The integration of MPNCs into conventional treatment processes such as membranes or advanced oxidation processes could lead to highly efficient hybrid systems. Finally, the development of smart materials, sensitive to environmental stimuli (such as pH, temperature, or pollutant concentration), could enable dynamic regulation of adsorption and regeneration cycles. Taking these research directions into account will facilitate the transition of MPNCs from the laboratory to concrete and sustainable applications, effectively addressing global challenges related to water pollution.

11.1. Deployment in Real Wastewater Systems: Bottlenecks and Challenges

While the laboratory scale adsorption capacities of magnetic polymer nanocomposites (MPNCs) are highly promising, the vast majority of current studies are conducted using simplistic, synthetic wastewater containing a single target pollutant. This represents a major disconnect from real engineering scenarios. Actual industrial and municipal wastewater matrices are highly complex environments, and this complexity introduces severe bottlenecks that remarkably impair the practical treatment efficiency of MPNCs.

11.2. Competitive Interference from Coexisting Ions and DOM

The primary bottleneck restricting the practical deployment of MPNCs is the presence of complex interfering factors. Real wastewater contains high concentrations of

background electrolytes (e.g., Na^+ , K^+ , Ca^{2+} , Mg^{2+} , Cl^- , SO_4^{2-}). These coexisting ions compete directly with target heavy metals or ionic dyes for the active functional groups (e.g., $-\text{NH}_2$, $-\text{COOH}$) on the polymer surface, significantly depressing the adsorption capacity through competitive electrostatic interactions. Furthermore, the presence of Dissolved Organic Matter (DOM), such as humic and fulvic acids, poses a critical challenge. DOM can bind to the MPNC surface, neutralizing its active charge, competitively chelating target heavy metals, and causing severe pore blockage, thereby severely hindering diffusion kinetics.

11.3. Fluctuating Physicochemical Conditions

Unlike controlled laboratory environments, real wastewater effluents experience drastic fluctuations in pH and temperature. Because the adsorption mechanisms of functionalized polymers (like chitosan or polyaniline) are highly pH-dependent, a sudden shift toward highly acidic or highly alkaline conditions can rapidly alter the protonation state of the adsorbent. This can lead to the sudden desorption of previously captured pollutants back into the effluent, or in severe cases, the chemical degradation of the polymer shell and the dissolution of the magnetic core.

11.4. Recent Advances in Real Wastewater Treatment

Despite these bottlenecks, recent advances are beginning to bridge the gap from lab to industry. For example, recent pilot studies have utilized surface-modified magnetic nanoparticles combined with polyacrylamide (PAM/ SiO_2 /MNPs) for the treatment of harsh “produced water” (PW) from the oil and gas sector. Even in this complex matrix containing heavy metals, phenolic compounds, and extreme turbidity, the nanocomposites maintained excellent structural integrity, successfully eliminating 98.7% of turbidity and 82.9% of chemical oxygen demand (COD). Such studies highlight that utilizing robust core-shell architectures (like silica encapsulation) combined with highly stable polymers is mandatory for resisting competitive interference [131].

11.5. Engineering and Scale-Up Challenges

The practical deployment of MPNCs is restricted by engineering limitations. Designing continuous-flow industrial reactors that can efficiently apply the necessary external magnetic fields to separate MPNCs from thousands of liters of rushing water per minute remains economically and mechanically challenging. Future research must pivot away from static batch experiments and focus on dynamic, pilot-scale column studies utilizing real wastewater effluents to accurately assess the long-term hydrodynamic stability, fouling resistance, and cyclic regeneration costs of MPNCs.

12. Conclusions

Magnetic polymer nanocomposites (MPNCs) represent a major advancement in the field of functional materials, due to their ability to offer innovative, effective, and sustainable solutions for the decontamination of polluted waters. This review has thoroughly explored the various synthesis methods, structural and magnetic properties, and environmental applications of these hybrid materials. It emerges that MPNCs are particularly effective for the removal of heavy metals, toxic dyes, persistent organic compounds, and hydrocarbons. The synergy between the properties of polymers notably their chemical stability, flexibility, and ability to functionalize and those of magnetic nanoparticles such as their high surface reactivity and easy separation under a magnetic field gives MPNCs strong potential in water treatment systems. Their regenerability and ability to be reused reduce operating costs and environmental impacts, making them promising candidates for large-scale applications. However, several technical challenges still need to be

addressed. Among these are the precise control of nanoparticle dispersion in the polymer matrix, the optimization of interfacial interactions, the improvement of selectivity towards specific pollutants, as well as the development of reliable, economical, and scalable synthesis methods [127]. The evaluation of their stability in complex environments and their long-term ecological impact remains essential as well. In summary, MPNCs pave the way for a new generation of water treatment technologies, combining performance, sustainability, and innovation. Their potential remains largely untapped, but their integration into modern industrial systems could radically transform approaches to managing aquatic pollution, while contributing to the preservation of natural resources and the protection of public health.

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