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Recent Advances in Nanomaterials-Based Adsorbents for Organophosphorus Contaminant Removal in Water: An Overview

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Abstract: The presence of organophosphorus contaminants (OPCs) in water is a major concern due to their toxicity and adverse effects on human health and the environment. Traditional water treatment methods such as coagulation, sedimentation, and filtration are not always effective in removing OPCs from water, making it necessary to explore alternative methods. Nanomaterials, due to their unique physical, chemical and biological properties, have emerged as promising adsorbents for the removal of OPCs from water. This review article focusses on the recent developments in the use of nanomaterials as adsorbents for the removal of OPCs from water. The article covers various types of nanomaterials, including carbon-based nanomaterials, metal-based nanomaterials and other hybrid nanomaterials. The mechanisms of OPC adsorption by nanomaterials, such as electrostatic interactions, hydrogen bonding, and Van der Waals forces, are also discussed. The article further highlights the factors affecting the adsorption capacity of nanomaterials, such as pH, temperature, and concentration of OPCs. Additionally, the article examines the challenges associated with the application of nanomaterials in water treatment, such as the potential release of nanomaterials into the environment and the need for cost-effective and scalable synthesis methods.

Key words: Organophosphorus contaminants, nanomaterials, water treatment, adsorption.

Introduction

The escalation of the industrial revolution together with human activities has favourably influenced the expansion of the economy and social advancement. However, this great achievement comes with a price as the number of chemical contaminants in an environment especially in water rises drastically (Shukor et al., 2022). OPCs are a class of toxic compounds that can be found in water sources due to their widespread use in pesticides, herbicides, and other industrial chemicals (Norrrahim et al., 2020). OPCs are highly soluble in water and are resistant to degradation, making them persistent in the environment. The toxicity of OPCs

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is significant to non-target organisms, including humans, and can result in acute cholinergic toxicity (causing thousands of poisonings annually) and delayed polyneuropathy (Costa, 2018). Moreover, OPCs can have significant ecological impacts by harming aquatic organisms and disrupting ecosystems.

Conventional methods for the removal of OPCs in a water environment conducted by researchers have been reported such as electrochemistry (Hosseini et al., 2015), bioremediation, chemical extraction, coagulation (Saleh et al., 2020) chemical oxidation (Saleh et al., 2020), chemical precipitation (Ajiboye et al., 2021), photocatalysis and adsorption. Table 1 summarises several techniques for the removal of OPCs from water. However, these techniques are not always effective in removing OPCs from water due to their complex chemical structure and high solubility. Among this method, adsorption is regarded as the simplest and most convenient approach for contaminant removal due to its minimal chemical requirements and its high removal efficiency even at low concentrations. While some advanced treatment methods, such as ozonation and advanced oxidation processes, be effective, they are often expensive and can generate harmful by-products.

As a result, there is a growing need for alternative methods that can provide more efficient OPCs removal. In recent years, nanomaterials have emerged as a promising class of adsorbents for OPCs removal in water. Nanomaterials are materials with at least one dimension in the nanoscale range (1-100 nanometers) and exhibit unique physical, chemical and biological properties due to their small size and high surface area-to-volume ratio. Nanocellulose is one of the examples of nanomaterials that have been revolutionising conventional materials in many applications (Misenan et al., 2018, 2023; Misenan et al., 2022; Nor et al., 2021; Norrahim et al., 2022).

Nanomaterials offer several advantages over traditional adsorbents for OPCs and several other contaminants removal in water (Misenan et al., 2021). First, they have a higher surface area-to-volume ratio, which allows for more surface area available for OPCs adsorption. Second, their surface chemistry can be modified to enhance their adsorption capacity and selectivity for specific OPCs. Finally, nanomaterials can be synthesised from a wide range of materials, including carbon, metals, and polymers, allowing for a variety of properties to be tailored to the specific application. The current trends in the publications related to this area have increased extensively the past ten years. This can be seen by the survey done on Google Scholar using keyword "nanomaterials-based adsorbent for organophosphorus removal" as shown in Figure 1.

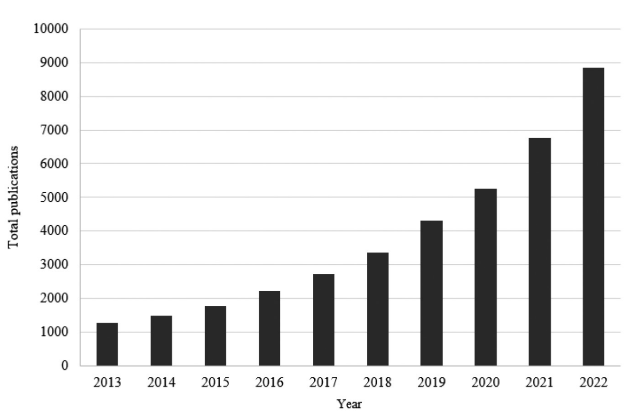

Figure 1:Total number of publications related to the nanomaterials-based adsorbent for organophosphorus removal.

Carbon-based nanomaterials, metal-based nanomaterials and hybrid nanomaterials are three of the most reported types of nanomaterials that have been investigated for the removal of OPCs in water. Carbon-based nanomaterials, such as activated carbon, carbon nanotubes, and graphene oxide, offer high surface area, tunable surface chemistry and excellent adsorption capacity. Besides, metal-based nanomaterials, including iron oxide nanoparticles and silver nanoparticles, have shown high adsorption capacity and selectivity for specific OPCs, as well as the ability to catalyse oxidation reactions for enhanced removal efficiency. Moreover, hybrid nanomaterials, such as carbon-based nanocomposites, metal-organic frameworks, and polymer-based nanocomposites, offer the advantages of both materials while addressing some of their limitations, making them promising candidates for the removal of OPCs from water.

Therefore, the article will cover the various types of nanomaterials that have been investigated for OPCs removal, including carbon-based nanomaterials, metal-based nanomaterials, and hybrid nanomaterials. Additionally, the article will discuss the mechanisms of OPC adsorption by nanomaterials, the factors affecting the adsorption capacity of nanomaterials, as well as the future perspectives of this field of study.

Organophosphorus Contaminants

OPCs are a diverse group of chemicals that are widely used in various industrial, agricultural, and household applications. These compounds are highly toxic and persistent in the environment, posing a serious threat to human health and the ecosystem. OPCs can be classified into several categories based on their chemical structure, Each of these categories contains numerous compounds with varying properties, including toxicity, solubility and persistence in the environment. Table 2 summarises some of the key properties of several common OPCs (Freed et al., 1979).

Each of these categories contains numerous compounds with varying properties, including toxicity, solubility, and persistence in the environment. For example, chlorpyrifos is a commonly used pesticide that has been linked to various health problems, including neurological damage, developmental disorders and cancer. Diazinon is another widely used pesticide that has been shown to have acute and chronic toxicity to humans and wildlife. Besides that, malathion is used primarily for mosquito control and is toxic to aquatic organisms. Parathion and methyl parathion are highly toxic pesticides that are no longer approved for use in many countries due to their health and environmental risks.

A key challenge in addressing the problem of OPCs contamination in water is the diversity of these compounds, which have different chemical structures, physical and chemical properties and toxicological profiles. The removal of OPCs from water requires a

Table 1: Several techniques for the removal of OPCs from water

Technique	*Description*	*Advantages*	*Disadvantages*
Adsorption	The use of solid adsorbents, such as activated carbon, to adsorb and remove OPCs from water	High removal efficiency, cost-effective, can be used for both point-of-use and point-of-entry treatment	Requires frequent replacement of adsorbent material, may generate secondary waste
Bioremediation	The use of microorganisms to degrade and remove OPCs from water	Natural process, cost-effective, can be used for both point-of-use and point-of-entry treatment	Requires a long retention time, may be limited by the availability of suitable microorganisms
Chemical oxidation	The use of oxidising agents, such as chlorine, ozone, and hydrogen peroxide, to degrade and remove OPCs from water	High removal efficiency, can be used for both point-of-use and point-of-entry treatment	May generate harmful byproducts, high operating costs
Membrane filtration	The use of membranes, such as reverse osmosis, ultrafiltration, and nanofiltration, to remove OPCs from water	High removal efficiency, can be used for both point-of-use and point-of-entry treatment	High operating costs, may require pre-treatment to prevent membrane fouling
Photocatalysis	The use of photocatalysts, such as titanium dioxide, to degrade and remove OPCs from water	High removal efficiency, environmentally friendly, can be used for both point-of-use and point-of-entry treatment	May require the use of UV light, high operating costs
Coagulation	The use of coagulants, such as alum and ferric chloride, to form aggregates with OPCs that can be removed by sedimentation or filtration	Effective for removing colloidal and suspended particles, can be used for both point-of-use and point-of-entry treatment	May require pH adjustment, high operating costs

Table 2: Properties of several common OPCs

OPCs	*Chemical formula*	*Molecular weight (g/mol)*	*Solubility in water (ppm)*
Chlorpyrifos	$C_9H_{11}Cl_3NO_3PS$	350.5	0.4
Fenitrothion	$C_9H_{12}NO_5PS$	277.2	30
Malathion	$C_{10}H_{19}O_6PS_2$	330.4	145
Parathion	$C_{10}H_{14}NO_5PS$	291.3	11.9
Methylparathion	$C_8H_{10}NO_5PS$	263.2	55

tailored approach that takes into account the specific properties of the compounds present. This has led to the development of a variety of treatment technologies, including adsorption, advanced oxidation processes and membrane filtration.

Recent Achievements on Organophosphorus Contaminant Removal in Water by Several Nanomaterials-Based Adsorbents

In this section, we will discuss the most reported nanomaterials which are carbon-based nanomaterials, metal-based nanomaterials, and hybrid nanomaterials for the removal of OPCs from water. The adsorption mechanisms of OPCs onto nanomaterials are primarily characterised by electrostatic interactions, hydrogen bonding, and Π-Π stacking (for carbon-based nanomaterials), as well as electron exchange or sharing between the vacant active sites of nanocomposites and OPCs molecules (in the case of hybrid materials). Figure 2 shows the possible mechanism of interaction between the nanomaterial (nanocellulose) with OPCs. Each section in this article provides an overview of the respective nanomaterials, their effectiveness in OPCs removal, and the underlying mechanisms of their adsorption capacity.

Carbon-Based Nanomaterials

Carbon-based nanomaterials are one of the most reported types of nanomaterials used for the removal of OPCs from water. These materials have several unique properties, including high surface area, tunable surface chemistry and a range of pore sizes and structures, which make them attractive candidates for water treatment applications. Activated carbon, carbon nanotubes (CNTs) and graphene oxide are the most widely used carbon-based nanomaterials for OPCs removal. The adsorption of OPCs onto these materials is primarily governed by physical adsorption mechanisms, including van der Waals forces, π-π interactions and hydrogen bonding.

Kodali et al. (2021) reported on their work investigating the use of Activated Coconut Charcoal (AcCoC) as an adsorbent for the removal of the OPCs. The results showed that AcCoC exhibited a high adsorption capacity for OPCs at pH 7.0, with a Langmuir adsorption capacity of 103.9 mg g^{-1}. The efficacy of the adsorption process was confirmed through the quantification of residual OPCs concentrations in water using triple quadrupole LC-MS spectrometry. The authors demonstrated the ability of AcCoC to effectively detoxify OPCs in samples of farm water collected from agricultural soils, highlighting its potential as a super adsorbent for the removal of OPCs from water.

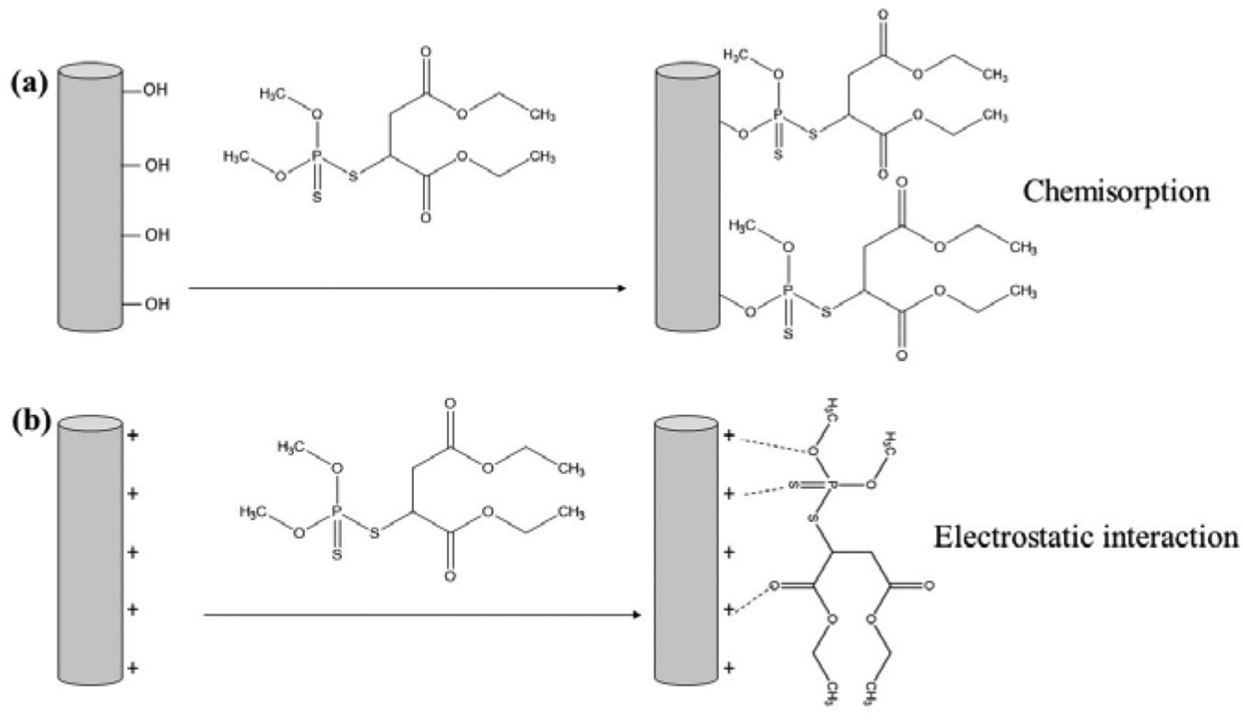

Figure 2: Mechanism of interaction between the nanocellulose and OPCs – (a) un-functionalised nanocellulose, (b) functionalised nanocellulose.

Liu et al. (2018) successfully prepared a functional adsorbent, namely metal–organic framework ZIF-8/magnetic multi-walled CNTs (M-M-ZIF-8) (Figure 3), for the removal of eight OPCs from environmental water samples. The high specific surface area and porous structure of M-M-ZIF-8 enabled it to exhibit high static adsorption capacities for OPCs. The Freundlich adsorption model was found to be a better fit for the static adsorption data as compared to the Langmuir model. The authors further applied M-M-ZIF-8 under optimised conditions and successfully removed the OPCs from environmental water and soil samples. The valence-electron-driven adsorption mechanism was proposed as the possible mechanism for the sharing or exchanging of electrons between the OPCs molecules and the vacant active sites of M-M-ZIF-8. The study concluded that M-M-ZIF-8 holds promise as a hybrid adsorbent for the adsorption and removal of organic pollutants from the environment.

Suo et al. (2018) developed an efficient adsorbent, activated carbon derived from sieve-like cellulose/graphene oxide composites (ACCE/G), for the removal of various OPCs. The maximum adsorption capacity of ACCE/G for chlorpyrifos was found to be 152.5 mg g^{-1}. The adsorption mechanism, thermodynamic properties and kinetics of the adsorption process were also investigated. The results showed that the adsorption mechanism of ACCE/G is dependent on the electron-donating abilities of the S and P atoms. Additionally, the Langmuir model provided the best fit for the isotherm data. Furthermore, the adsorption efficiency of ACCE/G was still over 80% after eight cycles of recycling, indicating its potential as a valuable candidate for the removal of OPCs. The findings of this study highlight the promising future of ACCE/G as an efficient and recyclable adsorbent for the removal of OPCs from contaminated water and soil.

Metal-Based Nanomaterials

Metal-based nanomaterials are another class of nanomaterials that have been studied for their potential to remove OPCs from water. These materials have unique properties, such as high surface area, high reactivity, and the ability to form various chemical bonds, which make them attractive candidates for OPCs removal. Silver nanoparticles (AgNPs) and iron-based nanomaterials are the most reported metal-based nanomaterials for OPCs removal. AgNPs have a high surface area-to-volume ratio, which allows for enhanced adsorption of OPCs. The adsorption of OPCs onto AgNPs is primarily governed by several chemical interactions. Other metal-based nanomaterials that have been studied for OPC removal include titanium dioxide nanoparticles (TiO_2NPs), gold nanoparticles (AuNPs) and copper nanoparticles (CuNPs).

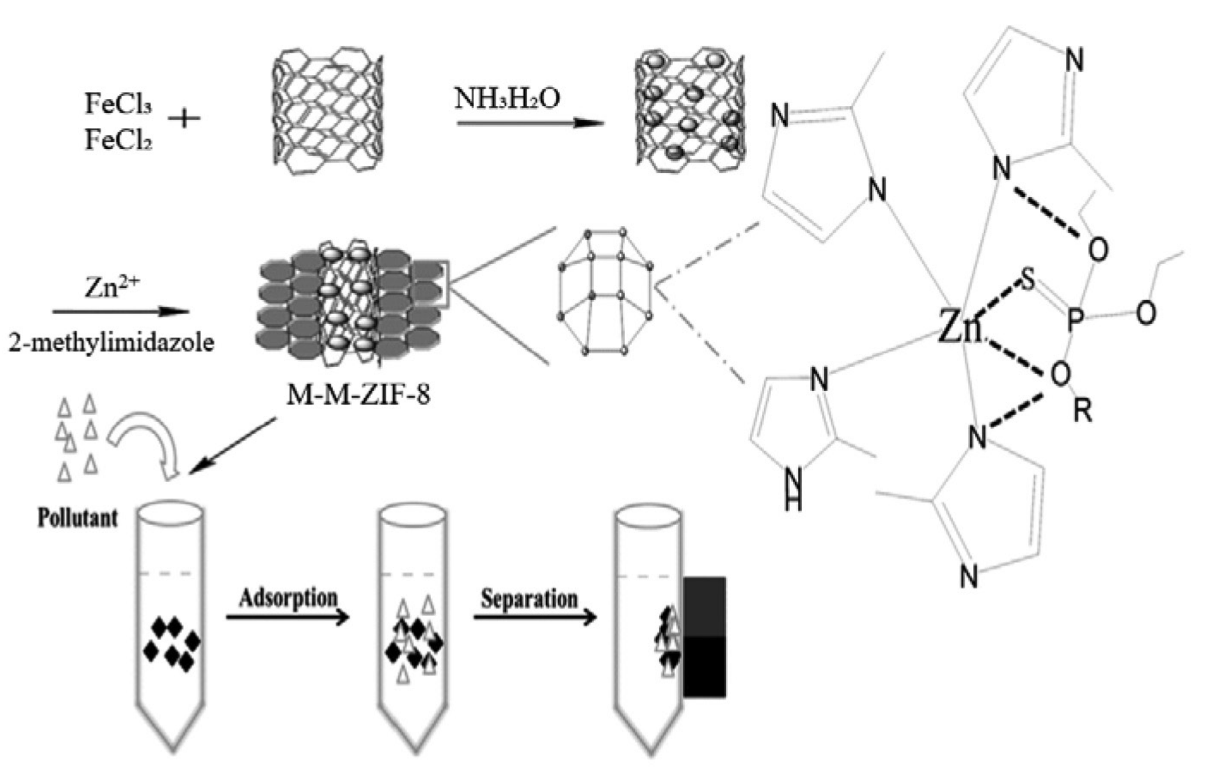

Figure 3: Procedure for M-M-ZIF-8 preparation and adsorption mechanism for pesticides.

Veerakumar et al. (2021) investigated the effectiveness of zinc oxide nanostars (ZnONSt) in degrading the OPCs methyl parathion, using noble metal nanoparticles (Ag and Pd) for conjugation. The noble metal nanoparticles were well-dispersed on the surface of ZnONSt using the microwave-hydrothermal method. Various analytical techniques were employed to evaluate the phase constitution, functional groups, optical properties, elemental composition, surface area and surface morphology of the resulting nanocomposites. The XRD patterns and FE-SEM/TEM micrographs indicated that the ZnONSt had a hexagonal wurtzite phase with a star-like morphology. Ag@ZnONSt and Pd@ZnONSt showed excellent photocatalytic activity in degrading the pesticides under visible light irradiation. The Pd and Ag nanoparticles acted as electron sinks and enhanced the interfacial charge transfer process by reducing the charge recombination rate. Pd@ZnONSt nanocomposite showed superior performance compared to other ZnONSt morphologies and commercial photocatalysts.

In another study, Li et al. (2019) prepared a magnetic nanoporous carbon material using Zn/Co-MOF as the precursor via a simple one-step carbonisation method, which effectively extracted OPCs through π–π and hydrophobic interactions. A magnetic solid-phase extraction technique coupled with gas chromatography and flame photometry was established for the quantitative analysis of OPCs in fruits using this material, which exhibited short extraction and total detection times, simplified operation procedures, and reduced reagent consumption and preparation costs.

Hybrid Nanomaterials

Hybrid nanomaterials, which consist of two or more different types of nanomaterials, have recently gained attention as potential adsorbents for the removal of OPCs from water. By combining different types of nanomaterials, hybrid nanomaterials can exhibit unique properties that are not present in individual nanomaterials.

Zhang et al. (2020) utilised an in-situ fabrication method to synthesise magnetic flower-like molybdenum disulphide hybrid materials (Fe_3O_4/MoS_2) (Figure 4), which were applied for magnetic solid-phase extraction of OPCs. They found that MoS_2 played a critical role in rapid and high adsorption capacity towards the

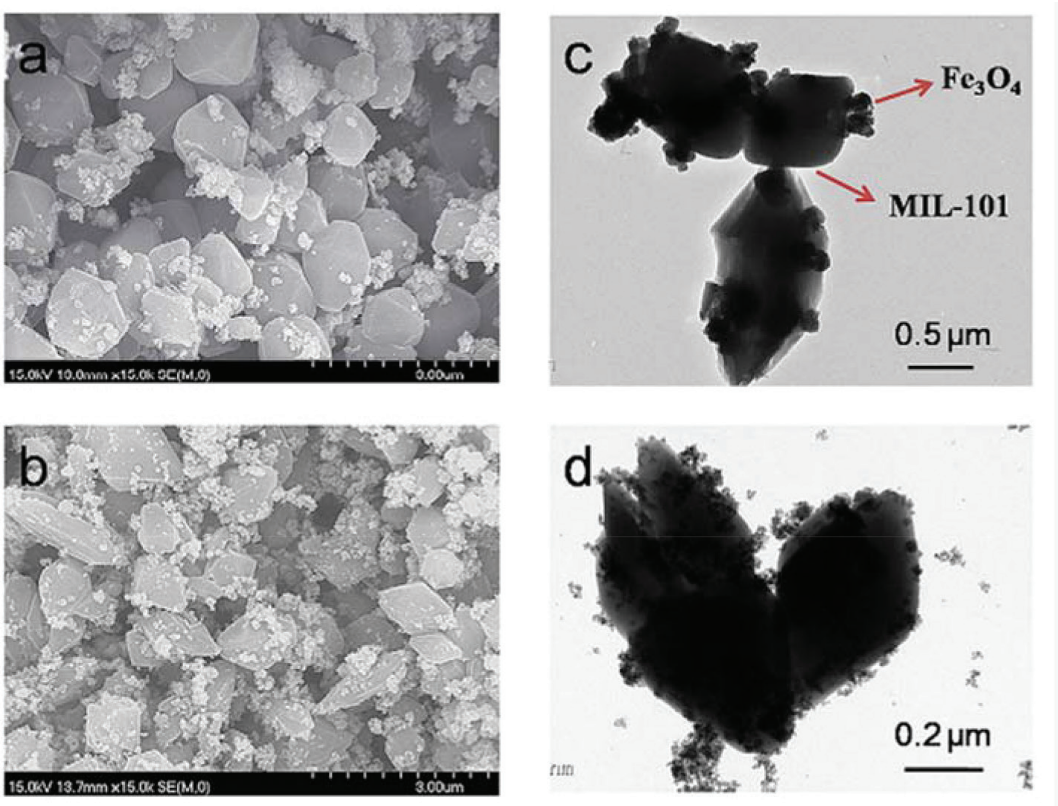

Figure 4: SEM images of Fe_3O_4/MIL-101 composites with Fe_3O_4 loading amounts of 50 mg (a) and 250 mg (b); TEM images of Fe_3O_4/MIL-101 composites with Fe_3O_4 loading amounts of 50 mg (c) and 250 mg (d). (Reproduced from ref. Zhang et al., 2020.)

OPCs. The best extraction efficiency and magnetic performance were achieved with a loading amount of 200 mg MoS_2. Fe_3O_4 was attached to the surface of MoS_2, forming a flower-like shape with a diameter of 500 nm. The researchers optimised various parameters such as amounts of adsorbents, adsorption time, solution pH, elution solvents, and sample volumes that affected magnetic solid-phase extraction efficiency. Under the optimum conditions, they established a magnetic solid-phase extraction coupled with a high liquid chromatography-tandem mass spectrometry method to analyse OPCs in environmental water samples. All the analytes showed good recoveries spiked at three different concentration levels (10, 20 and 50 ng/mL) in water samples, with relative standard deviations ranging from 1.4% to 6.8%.

Future Perspectives

The use of nanomaterials as adsorbents for the removal of OPCs from water is a promising area of research with significant potential for environmental remediation. While carbon-based, metal-based, and hybrid nanomaterials have shown promising results in OPCs removal, there is still much to be explored in terms of their synthesis, optimisation and application.

One emerging nanomaterial that has shown promise in OPCs removal is nanocellulose. Nanocellulose, a biodegradable and renewable material derived from plant sources, has a high surface area and tunable surface chemistry, making it a promising adsorbent for OPCs. Recent studies have reported the successful use of nanocellulose-based adsorbents for the removal of OPCs, such as malathion and chlorpyrifos, from water. In addition to nanocellulose, other emerging nanomaterials, such as MOF and covalent organic frameworks (COF), hold great potential as adsorbents for OPCs removal. These materials have highly porous structures and tunable surface chemistries, allowing for enhanced selectivity and adsorption capacity for specific OPCs.

Another area of future research is the optimisation of nanomaterials for large-scale and real-world applications. Challenges such as cost-effective synthesis, stability, and scalability must be addressed to ensure the practical application of nanomaterials for OPCs removal.

Furthermore, the development of new technologies for the regeneration and reuse of nanomaterial-based adsorbents can greatly reduce the environmental impact of OPCs removal. Regeneration methods such as photocatalysis, electrochemical regeneration, and microwave regeneration have shown promise in effectively regenerating nanomaterial-based adsorbents for multiple uses.

Conclusion

The removal of OPCs from water using nanomaterials as adsorbents is a promising area of research with significant potential for environmental remediation. In this review, we have discussed the three main types of nanomaterials used for OPCs removal, including carbon-based, metal-based, and hybrid nanomaterials, as well as the emerging material, nanocellulose. Carbon-based nanomaterials, such as graphene and carbon nanotubes, have shown high adsorption capacities and selectivity for specific OPCs. Metal-based nanomaterials, such as iron oxide nanoparticles and silver nanoparticles, have also demonstrated effective OPCs removal through adsorption and degradation processes. Hybrid nanomaterials, which combine the advantages of carbon-based and metal-based nanomaterials, have shown enhanced adsorption capacities and selectivity for specific OPCs. Despite the promising results reported, there is still much to be explored in terms of the synthesis, optimisation and application of nanomaterial-based adsorbents. With continued research and development, nanomaterial-based adsorbents can be optimised for practical and effective use in large-scale and real-world applications.

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Conflict of interest

The authors declare no conflict of interest in the preparation of this review.

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