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Nanotechnology-driven adsorption for the removal of pesticide contaminants from water - A review

Ulfat Jan^{1*}, Shivani Bhartiya², Nusrat Fatima¹

¹Division of Entomology, Sher-e-Kashmir University of Agricultural Sciences & Technology Kashmir, Srinagar-190025, Jammu and Kashmir, India, ²Department of Entomology, College of Horticulture & Forestry Neri, Hamirpur-177001, Himachal Pradesh, India

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***Corresponding Author:**

Ulfat Jan
E-mail: sheirulfatent@gmail.com

ABSTRACT

Pesticides are natural or synthetic compounds that are used to protect crops against pests. Pesticides are important as they help to grow more food and increase agricultural productivity by protecting crops from pests, diseases, and weeds. Pesticides have some harmful effects on the environment as they cause soil deterioration, water pollution, harm to non-target organisms, biomagnification, resistance development and disrupt ecosystem services. Pesticides also leave residues in the environment, food, soil and water sources. There are a number of methods for this pesticide elimination from the water. However, the development of a new scientific area, nanotechnology, has become widely used for environmental remediation in recent years. Nanotechnology offers promising solutions for removing pesticide contaminants from water through adsorption techniques. Nano-scale materials have a high specific surface area, changed quantum properties and their small size allows them to reach the target contaminant where micro materials cannot enter. The focus of this review is to study several nano-technology approaches like carbon-nanotubes, graphene-based nanomaterials, dendrimer nano-adsorbent and nano-crystalline metal oxides for pesticide removal from water by the adsorption process.

KEYWORDS: Pesticides, Residues, Water, Nanotechnology, Adsorption, Carbon nano-tubes, Nano-crystalline metal oxides

INTRODUCTION

Pesticides encompass a wide range of chemicals, including insecticides, fungicides, herbicides, rodenticides, molluscicides and nematicides (Figure 1). Pesticides are widely used around the world to protect crops from pests that inhibit plant growth. Since the mid-1940s, global pesticide use has risen dramatically and continuously, driven mostly by commercial farming. Pesticide use increases agricultural production by protecting crops from pests, diseases and weeds (Khan *et al.*, 2023). They can also help to improve food security by ensuring the amount and quality of harvested crops. Nevertheless, inappropriate and unauthorized pesticide usage has disturbed ecosystem services due to contaminated food and water, degraded soil, which ultimately affect beneficial insects and cause air pollution and biomagnification. The primary concerns, however, are the detrimental impacts of pesticide use on ecosystems, wildlife, and human well-being.

Pesticide residues are also present in different types of fruits, vegetables, soil, water, and processed foods. The environment, food, drinking water sources, as well as glaciers all include

residues due to the inappropriate use of pesticide combinations on crops (Pandiselvam *et al.*, 2020), which results in the contamination of drinking supplies, groundwater and soil (Nasrabadi *et al.*, 2011). There are several ways that pesticides can get into the environment, including dust, spray, runoff from farms, and industrial effluents. Pesticide residues have a substantial effect on aquatic ecosystems and mammals in addition to interfering with human life (Maddah *et al.*, 2017). It is important to take the required steps to get rid of the residual contamination in the environment. The initial step in reducing pesticide use is to minimize their application and implement regulations (Mahdavi *et al.*, 2021).

Multiple approaches have been developed to mitigate the effects of pesticide residues in the environment. On the other hand, nanotechnology has gathered significant attention and is a promising method to eliminate pesticides from the environment and water. Using techniques like photo-decomposition and absorption, nanotechnology may effectively remove pesticide residues from solids, water and the air. Because of their many diverse physical and chemical characteristics, nanomaterials offer a unique choice for pesticide elimination. They can also be

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modified with different chemical groups to enhance their efficacy in eliminating certain target chemical groups (Riu *et al.*, 2006).

In developing countries, one of the primary sources of human pathogenic microorganisms is water. Any contaminant found in a body of water that has an adverse effect on living things is considered water pollution (Figure 2). Therefore, in order to prevent public health issues, innovative lowest-cost treatment solutions are needed. This review's primary objective is to go over nano-adsorbent-based water filtration technologies.

UTILIZATION OF NANO-MATERIALS IN REMEDIATION OF ENVIRONMENTAL CONTAMINANTS

Particles with nano-metre sizes in at least one dimension are commonly known as nanomaterials (NMs) (Feynman, 1959). NMs are utilized in health sciences, electronics, aerospace,

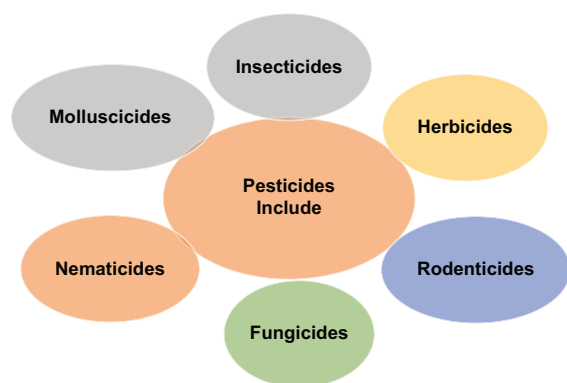


Figure 1: Types of pesticides used in agriculture

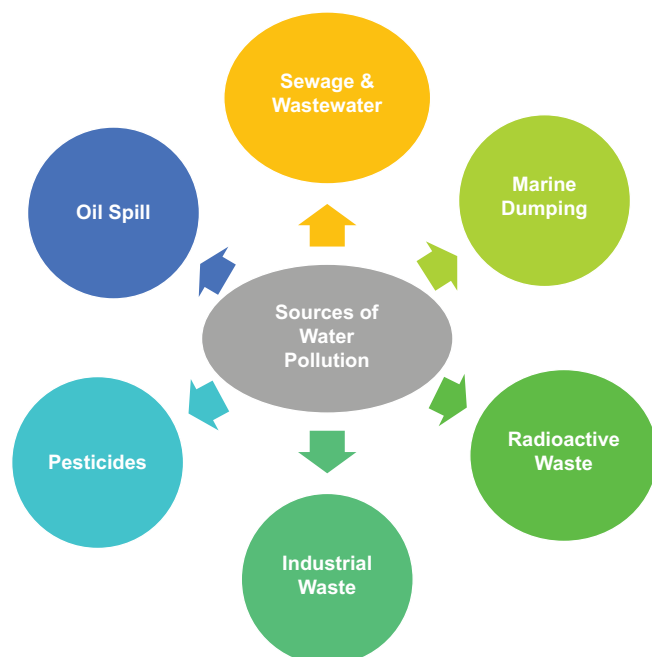


Figure 2: Sources of water pollution

chemical manufacturing, and agriculture, because of their small size (Pokropivny *et al.*, 2007). Numerous inorganic nanoparticles are used as nano-materials, such as carbon nano-tubes, which are typically utilized as carriers; iron nano-particles are used extensively because of their magnetism; and silica nano-particles have drawn interest from researchers because of their huge total surface area and rich pore structure. Other experiments have emphasized copper, gold, or silver nano-particles (Anandhi *et al.*, 2020).

Using the nano-materials in bioremediation of environmental pollutants is a very promising approach for a number of reasons. First of all, materials at the nano-scale are good adsorbents because of the huge area per unit mass. They can readily come in contact with the contaminant due to their large surface area. In addition, nano-particles have specific quantum properties and don't need high activation energy to start reactions. Another factor that makes nano-particles ideal for environmental remediation is that their size enables them to reach contaminated areas that are in-accessible to micro-particles. Particles at nano-scale show different characteristics like their mechanical and chemical features such as color and reactivity changes compared to their micro or macro scale, their color and reactivity.

Nano-materials, particularly membranes, adsorption, catalytic oxidation, disinfection, and sensing (Das *et al.*, 2017) offer a broad range of potential for water and wastewater remediation. Nano-materials have improved their competitiveness in water and wastewater cleaning by saving money. This review focuses on diverse nano-materials used for the elimination of pesticide pollutants from water.

NANOTECHNOLOGY APPROACHES IN PESTICIDE REMOVAL FROM WATER

Removal of Pesticide from Water by Adsorption

A lot of investigations have been carried out into the effective absorption of organic pollutants from aqueous solution (Khairy *et al.*, 2018). Sophia and Lima (2018) investigated that adsorption onto affordable materials is a useful and favorable technique for removing both organic and inorganic contaminants in water. Adsorption is an excellent method for water purification. Adsorption has been found to be more effective than other techniques for water cleaning in terms of original cost, flexibility, and simplicity of design, easy to use, and insensitivity to toxic pollutants. Adsorption also does not produce any harmful byproducts or residues. Among different nano-particle adsorbents, carbon-based nano-adsorbents (carbon nano-tubes, graphene) and nano-crystalline metal oxides (Figure 3) are mainly used in pesticide elimination from aqueous solution and are discussed below:

Carbon Based Nano-Adsorbents

Carbon Nanotubes (CNTs)

CNTs are unique mechanical, electrical, and chemical nano-materials. Carbon nano-tubes (CNTs) are nano-materials made

up of graphitic carbon with one or more concentric tubules. They can be described as a nano-scale-tube formed by rolling up a sheet of graphite. Carbon nano-tubes (CNTs) have a large specific surface area, excellent permeability, and strong thermal and mechanical stability. CNTs have a unique collection. CNTs as a nano-adsorbent are capable of eliminating heavy metal dyes and organic pollutants such as pesticide residues from wastewater. Carbon nano-tubes are capable of adsorption, enabling the removal of certain toxins that can be eradicated from wastewater. Due to their large surface area and layered, hollow structure, they possess a high adsorption capacity and can be utilized as absorbers for both organic and non-organic substances. Carbon nano-tubes (CNTs) are divided into single-walled carbon nano-tubes (SWNTs) and multi-walled nano-tubes (MWNTs) (Figure 4).

These are distinctive macro-molecules with one-dimensional structure, thermal stability and unique chemical characteristics (Firozjaee *et al.*, 2017 & Ren *et al.*, 2011). These nano-materials have an excellent capacity to eliminate different kinds of pesticides. The adsorption capacity of pollutants by CNTs is afflicted by the pore structure and the presence of a wide range of surface functional groups, which can be produced by chemical or thermal modifications to enhance the optimal

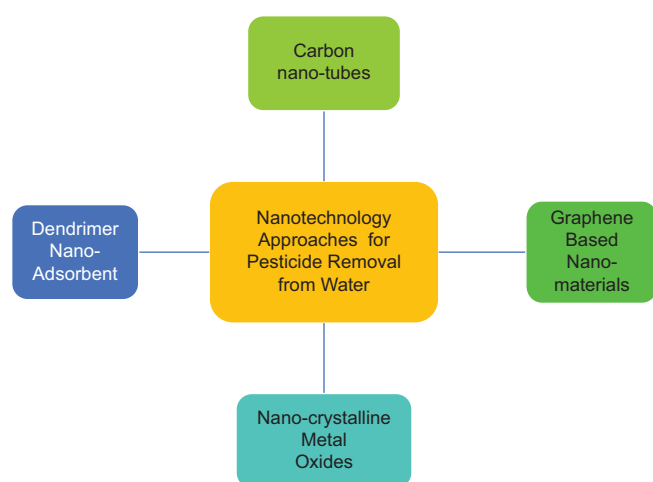


Figure 3: Nano-technology approaches for pesticide removal from water

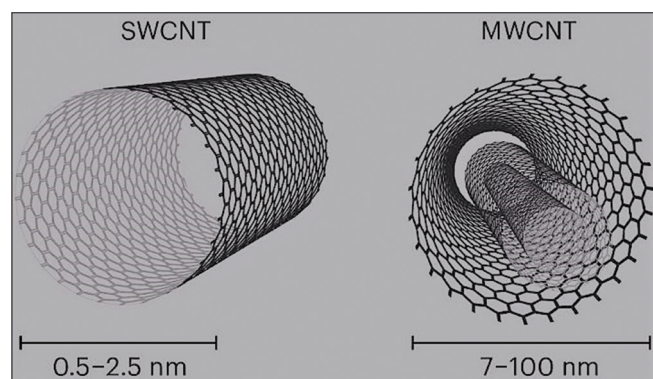


Figure 4: Carbon nano-tubes: Single-walled carbon nano-tubes (SWNTs) and Multi-walled nano-tubes (MWNTs)

performance for a particular objective. Hydrophobic effect, covalent bonding, (π - π) interactions, hydrogen bonding, and electrostatic interactions are the mechanisms involved in the adsorption of organic compounds on carbon nano-tubes (Yunus *et al.*, 2011). Some organic molecules with C=C bonds or benzene rings, such as polycyclic aromatic hydrocarbons (PAHs) and polar aromatic compounds adsorb on CNT through (π - π) interaction (Smith *et al.*, 2015). Adsorption capacity is also found in hydrogen bonds between organic molecules and functional groups like COOH, OH, and NH₂ (Yu *et al.*, 2014). Adsorption on Carbon nano-tubes (CNTs) has been evaluated for various compounds, for example, triazines, sulfonylureas, phenoxy-alkanoic acids, organo-phosphorous, organo-chloride pesticides and multi-class pesticides mentioned in Table 1.

Graphene-Based Nano-materials (GNMs)

Graphene, a type of carbon nanomaterial, has gained significant attention in water filtration and different applications because of its distinctive physical and chemical characteristics. As a single-layer nanoscale sheet of graphite, it offers substantial benefits for environmental protection. Graphene-based nanomaterials (GNMs) are being utilized by researchers to address water-related environmental challenges. Graphene can be synthesized through various methods, including the transformation of carbon nanotubes, chemical reduction, and mechanical exfoliation. Notably, graphene exhibits exceptional adsorption capacity for pesticides, varying between 600 and 2000 mg/g. Studies have explored its use in processes such as

Table 1: Latest examples for absorption of pesticides onto Carbon nano-tubes (CNTs)

Analytes	Sample	Reference
Sulfonylurea compounds	Water	Zhou <i>et al.</i> , 2006
Dicamba	Water	Biesaga & Pyrzynska, 2006
Atrazine, propoxur, methidation	Water	El-Sheikh <i>et al.</i> , 2007
Triasulfuron and bensulfuron-methyl	Water	Zhou <i>et al.</i> , 2007a
Sulfonylurea compounds	Water	Zhou <i>et al.</i> , 2007b
Phenoxyalkanoic acids	Water	Pyrzynska <i>et al.</i> , 2007
Multi-class pesticides	Water	Wang <i>et al.</i> , 2007
Thiamethoxam, imidacloprid, acetamiprid	Water	Zhou <i>et al.</i> , 2006
Organo-phosphorous pesticides	Fruit juices	Ravelo-Perez <i>et al.</i> , 2008
Pesticides	Mineral water	Asensio-Ramos <i>et al.</i> , 2008
Sulfonylurea compounds	Soil	Wu <i>et al.</i> , 2009
Chlorsulfuron and metsulfuron methyl	Water	Springer & Lista, 2010
Methyl parathion	Garlic	Du <i>et al.</i> , 2008
Organo-phosphorous pesticides	Seawater	Li <i>et al.</i> , 2009
Organo-phosphorous pesticides	Soil	Asensio-Ramos <i>et al.</i> , 2009
Atrazine and simazine	Water	Katsumata <i>et al.</i> , 2010
Chloroacetanilide	Tap and river water	Dong <i>et al.</i> , 2009
Organo-chlorine pesticides	Water, wastewater	Lü <i>et al.</i> , 2007

dehalogenation, halogenation, and the elimination of residual hydrocarbon pesticides from water. Graphene and related carbon nano-materials can adsorb contaminants, with aromatic rings through π - π interactions (Bjork *et al.*, 2010; Smith *et al.*, 2015). Graphene's adsorption capacity for pesticides can be significantly improved when combined with other substances. For example, graphene-coated silica (GCS) is an excellent sorbent for removing residual organo-phosphorus pesticides from water. The study highlights that the adsorption mechanism of OPPs on GCS is driven by the π -bonding network of benzene rings and the electron-donating properties of phosphorus (P), sulfur (S), and nitrogen (N) atoms (Pei *et al.*, 2013). Following the creation of graphene, various approaches were employed to develop graphene derivatives for pesticide removal from water. Table 2 provides details on the precursors, treatments, and synthesis methods used for these graphene-based materials. The different types of graphene family materials utilized for pesticide removal from water are outlined below.

Dendrimer Nano-Adsorbent

Another nano-adsorbent like carbon nano-tubes (CNTs) is a Dendrimer nano-particle used for eliminating organic pollutants and also heavy metals such as carbon nanotubes. Dendrimers are polymers that are sphere-like polymers and have three-dimensional, bifurcated structures. Dendrimers three-dimensional structure and numerous functional groups make it effective to attract ions and molecules, which can be adsorbed inside and outside cavities. Adsorption often involves electrostatic attraction, chemical interaction, and physical adsorption. Interactions are primarily determined by functional group chemistry, dendritic porosity, target species type, and solution pH. Dendrimers have limited practical application in water remediation due to their water solubility, preventing them from being easily separated from treated water. To solve this key problem, dendrimers are combined with various functional nanomaterials, increasing their applications due

to numerous benefits, including a high and reactive surface area, significant porosity, enhanced chemical, physical, and thermal resistance, efficient separation and reuse capabilities, and environmentally friendly processes. PAMAM is a popular dendrimer-based sorbent for removing heavy metals from wastewater due to its several cavities and ability to selectively chelate metal ions (Rether & Schuster, 2003; Ma *et al.*, 2009). Its high solubility in water and reactive amine or ester groups on the periphery enable the creation of poly-chelators with selective complexation of heavy metal ions (Rether & Schuster, 2003). Moreover, the high-water solubility and reactive amine or ester groups (found in intermediate generations) at the edges enable poly-chelators to be designed for selective heavy metal ion complexation (Rether & Schuster, 2003). After adsorption, the sorbent can be chemically separated from the remediated water, and the metal concentrations can be reused or disposed of at the same time, another adsorption-recovery cycle using the same sorbent.

Savage and Diallo (2005) incorporated dendrimers into ultra-filtration to effectively eliminate copper from water. The combined dendrimer ultra-filtration system successfully recovered nearly all copper ions. The adsorbent is easily regenerated by altering the pH. Sadeh *et al.* (2017) developed an effective bioadsorbent designed to eliminate anionic substances, including dyes, from textile wastewater through the creation of a chitosan dendrimer nano-structure. The bio-adsorbent is eco-friendly, bio-compatible, and non-toxic, achieving removal rates of certain dyes as high as 99%.

Nano-crystalline Metal Oxides

Nano-crystalline metal oxides are excellent adsorbents for various insecticides. Oxides such as iron, manganese, aluminum, titanium, magnesium, copper and cerium are not only effective but also cost-efficient (Figure 5). These materials are widely utilized for pesticide removal due to their exceptional adsorption capabilities, rapid kinetic properties resulting from their elevated

Table 2: Removal of pesticides by Graphene-Based Nano-materials (GNMs)

Graphene Material	Pesticide Removed	References
Reduced graphene oxide	Chlorpyrifos, endosulfan, and malathion	Maliyekkal <i>et al.</i> , 2013
Graphene-based magnetic nano-composite	5 different Carbamates	Wu <i>et al.</i> , 2012
Magnetite+SiO ₂ +TiO ₂ -reduced graphene	2,4-Dichlorophenoxyacetic acid	Tang <i>et al.</i> , 2013
Graphene coated solid phase micro-extraction fibre	Triazine herbicides	Wu <i>et al.</i> , 2012
Graphene	Six carbamate pesticides	Shi <i>et al.</i> , 2014
Graphene coated solid phase micro-extraction fibre	Six pyrethroid pesticides	Chen <i>et al.</i> , 2011
Cellulose/graphene composite	Six triazine pesticides	Zhang <i>et al.</i> , 2015
Magnetite+SiO ₂ +TiO ₂ -reduced graphene	2,4-Dichlorophenoxyacetic acid	Tang <i>et al.</i> , 2013
Graphene coated silica	Organo-phosphorous Pesticides	Liu <i>et al.</i> , 2013
Graphene sand composite	Chlorpyrifos	Gupta <i>et al.</i> , 2012

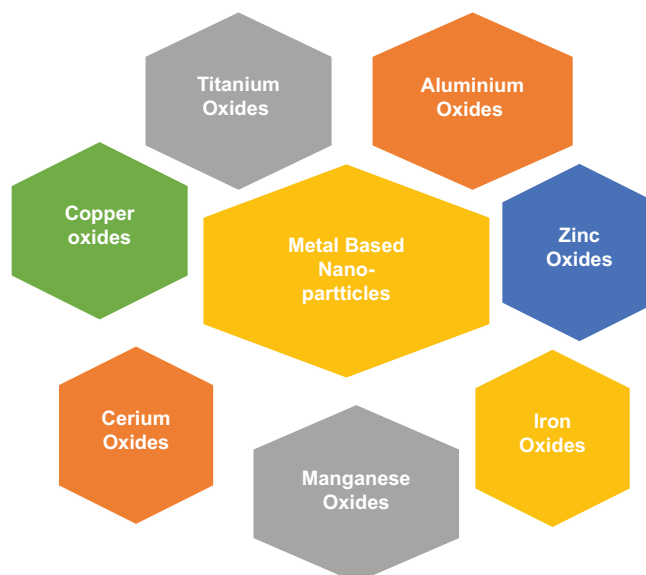


Figure 5: Metal based Nano-Particles

specific surface area, reduced intra-particle diffusion distances, and a greater number of active surface sites compared to their non-nano-commercial counterparts (Lan *et al.*, 2013; Moradi *et al.*, 2014). In addition to adsorption, nano-crystalline metal oxides can neutralize chemical hazards by transforming them into less harmful byproducts across a broad temperature range (Fryxell & Cao, 2013). Their extensive surface area and enhanced reactivity are attributed to the size quantization effect.

Research investigating the impact of particle size on the adsorption and desorption of as (III) and as (VI) revealed that as particle size decreased from 300 nm to 12 nm, the adsorption capacity increased nearly 200-fold (Mayo *et al.*, 2007). Some studies have explored the removal of organophosphorus pesticides using nano-metal oxides. While nano-scale metal oxides are highly effective destructive adsorbents for organophosphorus insecticides, the production of high-quality fine oxide powders remains a complex and costly process (Hua *et al.*, 2012). Researchers have explored the potential of magnetic nanoparticles for pesticide removal (Zeng *et al.*, 2011). Surface-modified magnetic core-shell nanoparticles demonstrate high adsorption efficiency and rapid contamination removal (Kaur *et al.*, 2014). Among these, Fe₃O₄ core-shell nanoparticles functionalized with C₁₈ are the most widely used for pesticide elimination. They are particularly effective for the separation or purification of non-polar and moderate polar pesticides due to their strong separation capacity, excellent stability, and ease of use. HaoYu and colleagues utilized magnetic Fe₃O₄-C₁₈ composite nanoparticles to remove organophosphorus insecticides (Shen *et al.*, 2007). When modified with C₁₈ silane, the Fe₃O₄-C₁₈ nanoparticles exhibited both hydroxyl and C₁₈ groups on their surface, enabling adsorption of hydrophilic and hydrophobic substances. These particles effectively collected organophosphates under the influence of a magnetic field. Additionally, alumina nanoparticles have shown significant efficacy in removing organophosphate pesticides (Wei *et al.*, 2012). The high surface area and abundance of hydroxyl groups on nano-crystalline alumina facilitate rapid adsorption of organophosphate insecticides.

DRAWBACKS IN THE USE OF NANO-ADSORBENTS FOR WATER PURIFICATION

The environmental and health implications of natural and engineered metal-based nanomaterials remain insufficiently studied. Nevertheless, their potential negative effects are largely associated with their nanoscale dimensions and chemical characteristics, especially in the case of mobile particles compared to those fixed within a matrix (Aragaw *et al.*, 2021). Nevertheless, their use in wastewater treatment will substantially raise their concentrations in aquatic environments, where they may infiltrate living organisms and lead to health issues (Cervantes-Avilés & Keller, 2021). For instance, various nanoparticles have been identified in influent, post-primary treatment, effluent from the activated sludge process, and reclaimed water at a full-scale wastewater treatment plant (WWTP). Specifically, the concentration of metal-based nanoparticles in influent wastewater has been reported to range

between 1,600 and 10,700 ng/L, while reclaimed water contained levels ranging from 0.6 to 721 ng/L.

One of the key drawbacks of using nano-adsorbents for contaminant removal is the difficulty in recovering and reusing them after treatment, as well as their tendency to agglomerate in aqueous media due to strong van der Waals forces (Kokkinos *et al.*, 2020). Consequently, the efficiency of water treatment processes can be significantly hindered, necessitating the adoption of strategies to stabilize nanoparticles. This is often achieved through surface coatings using materials such as polymers, secondary metabolites, carbon, or inorganic substances. Stability and recovery are further enhanced by incorporating the nanoparticles into polymeric matrices, including cellulose, chitosan, polyvinyl pyrrolidone, and similar materials. These methods help produce stable nanocomposites, improving their reusability and regenerative properties (Moreira *et al.*, 2022).

RECOVERY AND REUSE OF NANO-ADSORBENTS

Recovering nano-adsorbents from water after they have absorbed micro-pollutants is crucial to avoid downstream toxic effects on the natural environment. Likewise, regenerating the recovered nano-adsorbents allows for their reuse in new processes, thereby improving their sustainability. Effective nano-adsorbent recovery techniques must be efficient, eco-friendly, fast, and cost- and energy-efficient. The most common methods for nano-particle recovery include using a magnetic field to remove magnetic nano-particles. In addition to magnetic methods, filtration and centrifugation are also key techniques for recovering nano-adsorbents (Moreira *et al.*, 2022).

Various technologies, or combinations thereof, have been employed, as mentioned earlier, to develop a nano-adsorbent with high adsorption capacity and selectivity for specific emerging micro-pollutants. The choice of regeneration method depends on the type of adsorbent and adsorbate, as well as their stability and toxicity levels. Accordingly, thermal, chemical, and electrochemical methods have been utilized to regenerate nano-adsorbents from different media.

CONCLUSION AND FUTURE DIRECTIONS

The rapid advancement of science, technology and inventions has led to the evolution of nano-technology. Innovations in nanotechnology have increased the economic impact of research initiatives in this area. In conclusion, by improving the effectiveness and selective adsorption process for removing contaminants such as pesticides, fungicides, organic pollutants, and heavy metals, nanotechnology has transformed water purification and made a substantial contribution to clean water initiatives and sustainable development. Nano-materials, including carbon compounds (like carbon nanotubes, graphene-based materials, and dendrimer nano-adsorbents) and nano-crystalline metal oxides, provide excellent adsorption capabilities, quick kinetics, and reusability, rendering them very efficient in tackling water quality issues.

However, issues such as expenses, scalability, and possible environmental and health hazards need to be tackled to achieve its complete potential. Future studies ought to concentrate on creating environmentally friendly, affordable, and sustainable nano-materials while instituting strong regulations for their safe application. With ongoing progress, nanotechnology can transform water treatment, guaranteeing clean and safe water access for an increasing global population.

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