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Recent Trends in The Use of Nanomaterials for Wastewater Treatment: A Minireview

Halima Saliu
Department of Materials Science and Engineering
African University of Science and Technology
Abuja, Nigeria
halima@aust.edu.ng

Ebenezer Annan
Department of Materials Science and Engineering
University of Ghana
Accra, Ghana
ebannan@ug.edu.gh

Abdulhakeem Bello
Department of Physics
African University of Science and Technology
Abuja, Nigeria
abello@aust.edu.ng

Michael Aperebo
Department of Materials Science and Engineering
African University of Science and Technology
Abuja, Nigeria
maperebo@aust.edu.ng

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***Abstract*— Man's life depends on water in many ways. Worldwide industrialization and water resource exploitation have accelerated during the past few decades. Heavy metals and other pollutants, including effluent wastewater, are released into water streams as a result of industrial activity all over the world. Due to their toxicity, these contaminants are thought to be hazardous to both man and the environment and lower the quality of water. Even in low concentrations, heavy metals can be extremely harmful to living things. Numerous methods have been investigated for treating wastewater for many years. However, the discipline of nanotechnology has recently shown the world how to solve the issue of wastewater treatment using creative and practical methods.**

Keywords*— Heavy metals, Nanomaterials, Nanoparticles, Nanotechnology, Wastewater,

I. INTRODUCTION

One of the major challenges that will be faced by the world's growing population in the next century is access to clean drinking water [1]. It is estimated that about four billion people across the globe face water scarcity for about a month every year [2]. The decreasing quality of fresh water could be attributed to rapid industrialization and the unsound exploitation of water resources for various human activities [3]. These processes lead to the generation of large effluents of wastewater. The United States Environmental Protection Agency (EPA) defines a water contaminant as anything that is not a water molecule [4]. These contaminants could be organic (pesticides, phenols, herbicides, petroleum, dyes, oils, biphenyls, fats, proteins), inorganic (heavy metals, chemical fertilizers, and excessive nutrients), biological, radiological or other physical substances [4], [5]. Their release into the water bodies degrades their quality and creates serious problems for the environment. Heavy metals as contaminants are of key interest due to the significant harm they can cause to man and his environment [6]. Many traditional methods/techniques have been developed to treat and reuse wastewater. Some of the physical methods include coagulation, flocculation, membrane separation and adsorption; chemical methods that have been explored includes advanced oxidation and ozonation; and biological methods includes the use of enzymes and microorganisms and the use of activated sludge [7], [8]. These methods have been associated with major challenges that decrease their overall efficiency in removing contaminants from wastewater. Some of the challenges include their low effectiveness, high operating cost, high energy requirements, the use of too many reagents, clogging of filter or membrane, reusability, generation of a large volume of sludge and so on [9], [10]. The use of some of the traditional means is not entirely sufficient f or treating polluted water comprising a wide range of pollutants. This has led to the search for water purification techniques that are technically feasible, cost-effective, and highly efficient in the removal of contaminants from wastewater. Researchers have discovered the use of nanosized materials for removing pollutants from the water as one of the most efficient ways of treating wastewater [11]. This review analyzes the recent trends and developments in the use of nanotechnology for wastewater treatment with a particular emphasis on the removal of heavy metals. The review highlights the various types of Nanomaterials that are commonly adopted for wastewater treatment, recent works on the synthesis of Nanomaterials for removal of heavy metal, challenges encountered in the utilization of Nanomaterials for treating wastewater, and the future prospects or gaps to be filled in this area over time.

1) NANOTECHNOLOGY IN WASTEWATER TREATMENT

Nanotechnology is the manipulation of matter at the molecular and atomic levels to generate a new structure or system that possesses better electronic, optical, mechanical and conductive properties [12]. Over the past two decades, Nanotechnology has been explored extensively in different fields including water treatment, and has shown remarkable and outstanding results so far [13]. Nanomaterials are materials with any internal or external structures on the

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nanoscale (1 to 100nm) [11]. They play a significant role in technological advancement due to their adjustable properties and enhanced performance when compared to other techniques [14]. Their large surface area enables them to possess the unique physiochemical properties [15] that accounts for their high efficiency in treating waste water. Due to their effectiveness in cleaning water contaminated by industrial waste processes and their sustainability, Nanomaterials have been used extensively for the adsorption process [16]. Adsorption is a purification method for removing different compounds from industrial wastewater. It is a process in which liquid or gaseous substances are concentrated on the surface of a solid to the extent of separating them from the other substances present [17]. Adsorption is the most widely explored method of treatment by researchers due to its effectiveness in removing pollutants, its wide range of applications, and the low cost of adsorbents [18]. It is expected that engineered nanomaterials will play a vital role in countering the challenges associated with the water treatment process across the globe. Different types of nanomaterials have been explored by researchers for treating contaminated water. These include carbon-based nanomaterials (CNTs), polymeric nanoparticles, metal nanoparticles, biopolymers, zeolites, mesoporous layers, dendrimers and many more. Some of these nanomaterials are shown in figure 1 below. They could be employed via adsorption and biosorption, nanofiltration, photocatalysis, disinfection, sensing and monitoring and so on [13].

A. *Carbon made nanomaterials*

The Nano structural forms of carbon include single and multi-walled carbon nanotubes (SCNTs & MWCNTs), graphene, fullerene, and carbon dots [14]. CNTs are cylindrically shaped nanomaterials that possess a relatively large surface area and numerous adsorption points. Depending on the method of synthesis, they can exist as single-walled, double-walled or multi-walled nanotubes [10]. CNTs can be functionalized to increase their surface area thereby improving their removal and adsorption capacities. Their outstanding mechanical properties coupled with their high conductivity and thermal stability enable them to be considered for a wide range of applications [19].

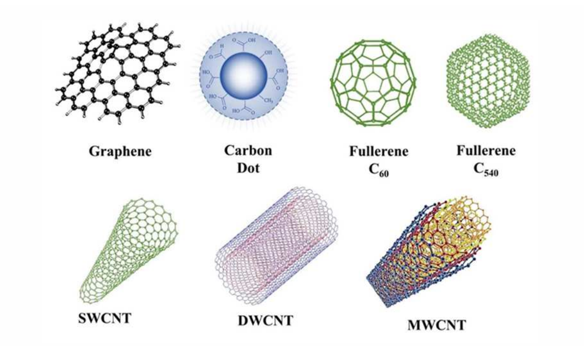

Figure 1:Types of Nanomaterials. Adapted from Iravani, (2021) [14]

Asides from CNTs, the interest in the use of carbon dots (CDs) nanomaterials has increased over the past few years. CDs are low-cost nanomaterials that are chemically stable and with superb surface properties. They are in synergy with living tissues and are therefore less risky or toxic to work with [20]

B. *Metal Nanoparticles/Metal Oxide Nanoparticles*

Carbon-based nanomaterials are costly to adopt for the water treatment process and thus, researchers are searching for more feasible alternatives which are equally as efficient as them. Metal and metal oxide nanoparticles have been explored extensively by researchers in the past decade. They have shown more effectiveness in removing heavy metal and radioactive materials from contaminated water than activated carbon [13]. Metal-based Nano-adsorbents include metal nanoparticles (Ag, Si, Fe) and metal oxide nanoparticles (TiO, FeO, CuO, ZnO). Metal oxide NPs stand out in terms of their high removal efficiency and heavy metals selectivity. Their small size and large surface area make it possible for them to be compressed without changing their surface area. [21].

C. *Polymer-made nanomaterials*

Polymeric nanoparticles are particles in the nanometer range (1-1000nm) that can be framed or entrapped on active compounds or absorbed onto their surface [22]. These small-sized materials have gained wide recognition among researchers over the years due to their outstanding properties. Functionalization of NPs with polymers gives a wide range of benefits such as better compatibility with living tissues, more mechanical strength and better chemical stability. The surface modification of magnetic NPs with polymers results in higher adsorption capacities [23]. Polymer-made NPs have been utilized for identifying and removing pollutants such as heavy metals, pesticides, pharmaceutical waste and organic compounds [14], [24].

D. Nanocomposites

A nanocomposite is a multiphase solid material in which one of the two phases has one, two or three dimensions in the nanometer range (<100nm). They are made by adding a nanoparticle into a material (or membrane) made of an inorganic or polymeric substance. The resulting mixture is usually of higher efficiency and possesses different physical and chemical properties [25]. However, their magnetic properties make it less difficult to retrieve them from the solution [26]. Also, their mechanical and thermal stability can be enhanced by adding metal oxides like Aluminum oxide (Al_2O_3) and Titanium oxide (TiO_2). They have also been very effective as adsorbents for removing nitrate from water [27].

E. Zeolites

Zeolites are microporous, 3D crystalline solids of aluminum silicate. They can be made naturally or from synthesis. Their porous nature makes them effective for iron exchange [28]. Natural zeolites are environmentally friendly, low-cost materials with excellent removal efficiency for heavy metals and ion exchange properties [14]. Their ability to remove anionic ions and organic compounds can be improved by modifying the surface of the zeolite to alter its surface charge [29].

F. Dendrimers

Dendrimers are a group of macromolecules with multiple branched chains and a well-ordered 3-D structure [30]. Their strong mechanical property, large surface area, high reactivity and hydrophilicity and ease of disposal have made them very effective in heavy metal removal [31]. They can also be used as adsorbents for removing organic pollutants [28]. Their adsorption capacity can be improved by functionalizing with other materials and they can be used to modify the properties of a material to enhance its productivity [32]. Dendrimers have been used to modify and fabricate membranes, thereby increasing their removal efficiency for removing contaminants from solution. They have also been explored as a possible solution to membrane fouling [33]

2) RECENT TRENDS/WORKS IN THE USE OF NANOMATERIALS FOR REMOVING HEAVY METALS

Adam et al., (2021) synthesized $ZnFe_2O_3$-carbon nanotube adsorbent material for the adsorption of heavy metals (Pb^{2+}, Cd^{2+}, Hg^{2+} and Sn^{2+}) from aqueous solution. The adsorbent was characterized using SEM/EDX and FTIR analysis. The effect of factors such as pH, contact time and adsorbent dosage on the adsorption process was studied. The adsorbent recovered the metal ions from the solution and the most favorable conditions for recovering the metal ions from the solution were observed to be a contact time of 15 minutes, an adsorbent dosage of 50 mg, and a pH of 5 (for Hg^2 and Pb^2) with a pH of 6 (for Cd^2 and Sn^{2+}).

Shen et al., (2020) prepared a composite with Fe_2O_3 and algae for immobilizing the microalgae and adsorbing heavy metal ions Cr^{2+}, Cu^{2+}, Pb^{2+} and Cd^{2+}. The composite was characterized by XRD, FTIR, XPS, SEM and TEM to assess its surface properties and morphology. The temperature of the Fe_2O_3 was controlled until the microalgae were greatly immobilized, and the removal efficiency of metal ions was examined. The composite was effective in immobilizing the microalgae and adsorbing the metal ions considered. Maximum adsorption capacities of 69.77, 62.63, 42.12 and 38.68 mg/g were recorded for Cr^{6+} Pb^{2+}, Cd^{2+} and Cu^{2+}. The adsorption process of Fe_2O_3-microalgae composite for all metal ions best fits the pseudo-second-order model.

El Mouden et al., (2023) carried out the synthesis of natural clay with Cobalt oxide, Co_3O_4 NPs to eradicate heavy metals Pb^{2+} and Cd^{2+} ions. The resulting matrix was characterized using SEM/EDS, TEM, FTIR, TGA and AFM. High removal efficiencies of 82.06 and 86.69 % was recorded for both ions. The kinetic data were best fit with the pseudo-second-order kinetic model. The synthesized material was observed to be biocompatible and easy to synthesize.

Zhang et al., (2023) synthesized Schiff-based side chain polymer with magnetic Fe_3O_4 NPs and utilized it for the removal of Cu^{2+} ions$^+$. The modified polymer-based nanocomposite proved effective as a reusable adsorbent with good selectivity for Copper ions. A maximum adsorption capacity of 59.98 mg/g for Cu^{2+} was recorded for this study.

Ha et al., (2020) synthesized a novel material by modifying activated carbon with pure nitrogen at high temperatures (500^0C and 1000^oC). The resulting mixture was further functionalized with amino-propyl triethoxysilane (APS) and used for the removal of heavy metals $Zn^{2+,}$ Cd^{2+} and Ni^{2+} ions. The modified material proved effective as an adsorbent for removing the heavy metals of interest. Maximum adsorption capacity (in decreasing order) of 242.5, 226.9, and 204.3 mg/g was recorded for Ni^{2+}, Cd^{2+} and Zn^{2+} ions. The decrease in adsorption capacity corresponds with the ions decreasing atomic radius and increasing ionic potential of the ions.

Ferrite NPs are Nanocomposites made by coupling metals with ferrite molecules. Their magnetic property makes it easy to separate them from the solution. They have been adopted by researchers for the removal of heavy metals such as Arsenic, cadmium, lead and so on [14]. [39] functionalized Fe_3O_4 magnetic particles (MPs) with dopamine (DA) and a detoxifying agent 2-3-dimercaptosuccine acid) for the removal of Pb^2, Cu^{2+} and Cd^{2+}. Maximum sorption capacities of 63.01, 49.46 and 187.62 mg/g were obtained for all three ions tested. The regeneration of the Fe_3O_4 MPs from this study was also really commendable.

Yan et al., (2023) designed a novel nanocomposite; a hydrogel-modified membrane prepared by dip-coating tannic acid, sodium alginate and amino-propyltriethoxysilane together. The modified membrane was used concurrently for removing Pb^{2+} ions and purifying oil sewage. Remarkable results were obtained from the study with an adsorption capacity of 39.9 mg/g for Lead ions and a flux recovery rate of 96.7% for the oil sewage.

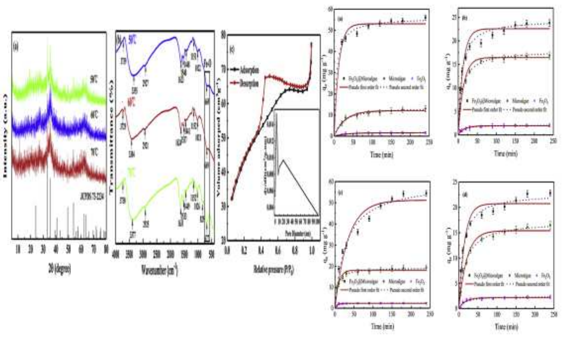

Figure 2: Characterization results for Fe2O3-microalgae composite: a) XRD pattern, b) FTIR spectra c) adsorption/desorption isotherm and pore distribution curves, d), e), f) and g) adsorption kinetic plots of Cr6+, Cu2+, Pb2+ and Cd2+. Adapted from Shen et al, (2021 [35].

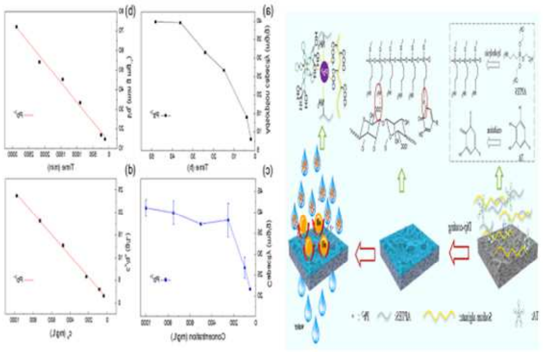

Figure 3: Illustration of the construction of nanocomposite, a) adsorption kinetic data of Pb (II), b) pseudo-second-order kinetics model c) adsorption isotherm of Pb (II) d) Langmuir isotherm model. Adapted from Yan et al, (2023) [40]

Zhao et al., (2023) modified a composite filter using MgO and biochar fibril (BCF) for treating wastewater. The composite proved effective for removing heavy metals Pb^{2+} and Cd^{2+} ions. The recovered metal ions were adsorbed on the BCF thereby making it easy to get rid of the extra waste generated in the process. Maximum adsorption capacities of 2035.4 and 3410.1 mg/g were recorded for Cd^{2+} and Pb^{2+} ions respectively.

Guo et al., (2023) utilized a zeolitic imidazole framework (ZIF) modified by a facile dip-coating method for the adsorption of heavy metals. The morphology and structure of the ZIF were modified from a leaf-shaped phase into a mixed phase in the hollow form to improve its effectiveness as an adsorbent. The resulting adsorbent was used for the adsorption of Pb^{2+} and Cu^{2+} ions. Batch and column experiments were investigated. The removal efficiency of 91.8% and above was recorded after ten cycles for both ions, thereby emphasizing the reusability of ZIFs as adsorbents for heavy metal removal.

As a possible solution to the challenges of reusability faced when working with zeolites in powder form, [43] modified a zeolite geopolymer foam adsorbent from natural clinoptilolite and calcined clinoptilolite. The material (clinoptilolite-based zeolite-geopolymer foams or CF) made was characterized by optical microscope, XRD, XRF, FTIR, BET, SEM and MIP techniques. It was used for the adsorption of heavy metals Cr^{6+}, Ni^{2+}, Cu^{2+}, Pb^{2+} and Cd^{2+} ions. A high removal efficiency (>60%) was observed for all metal ions considered after three consecutive cycles.

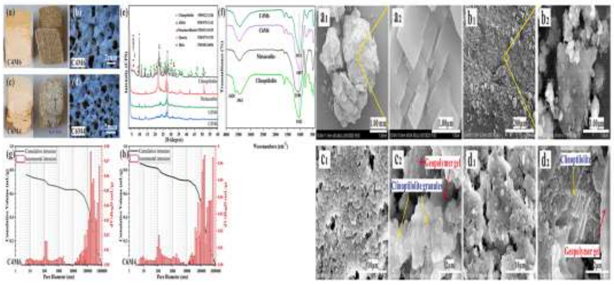

Figure 4: a)-d) XRD patterns, e)-f) FTIR spectra of raw materials and CF, g)-h) pore size distribution, a1)-d2) SEM images. Adapted from Liu et al., (2023) [43].

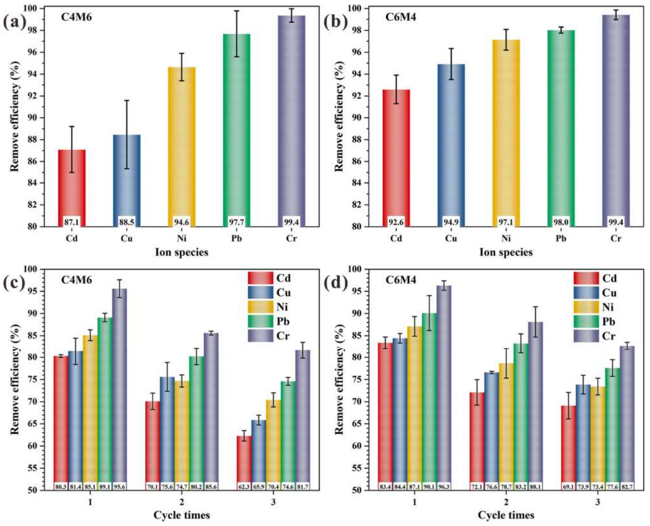

Figure 5: Removal efficiency (%) of metals ions and effect of cycle time on removal rates. Adapted from Liu et al., (2023) [43].

Patel et al., (2023) synthesized a higher-generation hydroxy-terminated-based dendrimer to remove Ni^{2+}, Co^{2+} and Zn^{2+} ions. Adsorption time, pH and varying generations were used to evaluate the sorption capacities of the ions. The dendrimers showed high metal sorption for all the metal ions tested. Removal efficiency increased with pH and time for all three generations of dendrimers used.

Chitosan-based adsorbents grafted by third-generation dendrimers were prepared by [44] for the recovery of Cu^{2+}, Pb^{2+} and Ag^{2+} metal ions from aqueous media. The prepared adsorbent was characterized by FTIR, XRD, TGA, and elemental analysis. Batch adsorption, desorption and recycling experiments were conducted. The adsorbent proved to be highly efficient as an adsorbent with a maximum adsorption capacity of 88.82, 97.87 and 88.82 mg/g for the metal ions considered.

II. LIMITATIONS AND FUTURE PROSPECTS

One of the key challenges faced with the use of nanomaterials is their toxicity to man and other living organisms when present in the body beyond certain limits. More strategies need to be put in place to monitor the extent to which these engineered materials are present in our water supplies. The high operational costs and possible toxicity to man and his immediate environment have limited the research on the use of these materials for wastewater remediation to laboratory and pilot scale. Also, existing methods for the recycling and reuse of Nano-adsorbents are minimal as they are quite difficult to fully develop. The commonly explored option currently is the use of magnets to remove metals with magnetic properties from the solution. Their small size also makes it challenging to separate and recover them from the solution. To enable the field of nanotechnology to be explored to its full potential and on a large scale, more research needs to be conducted to come up with possible solutions to the challenges currently being faced.

III. CONCLUSION

The use of nanomaterials for treating wastewater has proved to be a feasible alternative to another existing method over the years. It is still faced with many setbacks, and therefore a need for more research in this field to understand and derive the most acceptable means of adopting this technology to ensure the availability of water for generations to come.

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