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Iron-Based Nanomaterials as Wastewater and Pollutant Adsorbents

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Abstract

Environmental and human health are seriously threatened by the release of heavy metals into the environment through mining, metal finishing, welding, and alloy manufacturing. Heavy metals, in contrast to some organic contaminants, cannot be metabolised or destroyed and are not biodegradable. Through the water and the food chain, organic contaminants that are persistent in the environment, such as antibiotics, endocrine disrupting substances, and organic chemicals, represent a serious threat to the ecosystem's health. Pharmaceuticals like paracetamol, carbamazepine, and sulphanilamide are produced in vast quantities each year to treat diseases in humans and improve livestock breeding. Dyes, are basic chemical compounds and widely applied in numerous areas, but pollute the environment. The majority of dye compounds are mutagenic, teratogenic, and carcinogenic. The most practical method for treating wastewater at a cheap cost and with a high level of efficiency was found to be adsorption. Nanomaterials are used in a variety of technical domains because they exhibit a number of special enhanced features that bulk materials can not. Due to their distinct physicochemical features, iron-based nanoparticles have recieved significant attention recently, particularly in practises for environmental remediation. There is an increase in demand for new, and environmentally friendly methods for generating these nanoparticles.

Keywords: Nanomaterial, iron oxide, adsorbent, heavy metal, wastewater

1. Introduction

Due to the diverse range of pollutants produced by industries including the chemical, biomedical, pharmaceutical, textile, and other sectors, wastewater treatment is getting more complex. Biological methods, such as evaporation, have fallen short of meeting strict treatment standards and methods, and incineration has fallen short of its promise because of the secondary pollution it produces. The advanced oxidation process, either by itself or in conjunction with a biological process, can significantly contribute to assisting businesses with demanding treatment standards. It is essential for the chemical industry's long-term sustainability to have minimal environmental impact, nonselective pollution destruction, and high adaptability (Bhuta, 2014).

Pharmaceutical residue contamination is now a major concern in many nations. This has motivated scientists all around the world to develop novel methods for managing both the potential environmental effects of harmful contaminants as well as their remediation. Medicines are ingested by humans and animals and end up in surface water, groundwater, drinking water, soil, and other environmental media. For their removal from aquatic systems, a number of treatment methods have been used, including oxidation, photolysis, UV degradation, membrane processes, reverse osmosis, adsorption, coagulation, and flocculation. Despite this, there are several opportunities for research and development of cutting-edge treatment to protect the environment from pharmaceutical discharges. Fenton is a modern treatment approach that has shown great effectiveness. In the Fenton oxidation, hydrogen peroxide, ferrous or ferric ions, and free radicals interact to form hydroxyl radicals. This is an oxidation process that is metal-catalyzed and uses iron as a catalyst. The main disadvantages of the Fenton process are the high cost of H_2O_2 and the significant amount of ferric sludge generated during the neutralisation step of the treated solution before disposal. The classic Fenton method may be combined with ultrasound, electrochemical processes, UV radiation, and other wastewater treatment technologies to produce the enhanced Fenton process in order to overcome the aforementioned constraints (Masood et al., 2023).

The use of disinfection agents like chlorine, chlorine dioxide, or ozone results in the development of disinfection by-products (including trihalomethanes, halo-phenols, ketones, and aldehydes) that have a significant potential for mutagenicity and/or cancer. In addition, even when wastewater treatment procedures are applied on a wide scale, many harmful substances, such as heavy metal ions and azo dyes, cannot be entirely eliminated from wastewaters. Heavy metal ions and azo dyes are the most prevalent hazardous substances in wastewaters that cause specific issues (Sadegh et al., 2017). High levels of arsenic (As) in copper smelting wastewater provide a significant environmental concern that must be overcome (Luo et al., 2010). Toxic substances must be removed from wastewater in order to safeguard human health and the environment. Because of its great efficiency and economic consideration, the adsorption process is the best approach. Adsorbents have been utilised extensively for wastewater treatment, including activated carbon (AC), zeolites, biomaterials, and polymers. However, these materials' adsorption effectiveness is not very high. Consequently, it has become crucial to discover more effective adsorbents (Sadegh et al., 2017).

2. Nanomaterials as adsorbents

Nanomaterials are being utilised and have been exclusively developed and used extensively in a wide range of products, such as those used in medicine, industry, personal care, cosmetics, sunscreen, toothpaste, paints, optics, and electronics, as well as photocatalysts, antiultraviolet light agents, food packaging, medical devices, bandages, clothing, dental restoration material, antibacterial agents, drug delivery systems, artificial organs, and tissue adhesives. There are currently more than 1000 items or product categories on the market that contain nanoparticles, and it is predicted that in 2015, the yearly profit from engineered nanomaterials exceeded 2.5 trillion US dollars. One of the most widely produced and utilised nanoparticle in the world is titanium dioxide nanoparticles (TiO_2 NPs) (Shah et al., 2017).

For the removal of heavy metal ions and dyes from wastewater, nanomaterials have been widely researched. For improving the effectiveness and adsorption capabilities of removing pollutants from wastewater, various innovative nanomaterial adsorbents have been developed in recent years. Development and application of innovative technology is essential for treating water with high efficacy and minimal energy use. The following requirements should be met by an ideal adsorbent used to treat wastewater: A product should: 1) be environmentally friendly; 2) show a high sorption capacity and high selectivity, especially to the contaminants present in water at low concentration; 3) allow for the easy removal of the adsorbed pollutants from its surface; and 4) be recyclable. Numerous researches conducted in recent years have demonstrated that the majority of these requirements can be met by nanomaterials. The nanostructured adsorbents exhibit substantially greater efficiencies and quicker adsorption rates in water treatment, primarily because of the extraordinarily high surface area. The waste water treatment procedures may benefit from the use of nanomaterials including carbon nanotubes, graphene, ferric oxide (Fe_3O_4), manganese oxide (MnO_2), titanium oxide (TiO_2), magnesium oxide (MgO), and zinc oxide (ZnO) (Sadegh et al., 2017). Environmental cleanup can be accomplished by using nanomaterial adsorbents, such as metal oxides, carbon nanotubes, and graphene sheets but organic dyes, aromatic substances, phenolic derivatives, medicines, and antibiotics all have different adsorption capabilities (Awad et al., 2020).

3. Metal oxide-based nanomaterials

Metal oxides are the most diverse class of materials due to their unique features, which span practically all areas of solid-state physics and material science. Because of their ability to be oxidised or dissolved in water and release metal ions, several types of metal oxide nanoparticles, including iron oxide nanoparticles, ZnO nanoparticles, copper oxide nanoparticles, silver oxide nanoparticles, and titanium oxide nanoparticles, have been utilised for wastewater treatment. These chemically stable metal oxide nanoparticles are employed in numerous applications, including adsorption, photocatalytic activities, antibacterial and antifungal activities. (Naseem and Durrani, 2021).

Transitional metal oxide nanoparticles must have very good absorption capabilities in order to be used in modern energy and environmental products. High activity can also be attained using amorphous phases, despite the fact that most metal oxides are crystalline in nature (Li et al., 2015). Inorganic nanomaterials with a metal or metal oxide basis are frequently employed to remove dyes and heavy metal ions. Fe_3O_4, MnO_2, TiO_2, MgO, CdO, and ZnO are nanoscale metals or metal oxides that offer high surface area and specific affinity. Metal oxides have been used as sorbents to remove heavy metals and dyes because of their poor solubility, low environmental impact, and none of these characteristics contribute to secondary pollution generation (Sadegh et al., 2017).

Copper oxide, silver oxide, zinc oxide, iron oxide, and titanium oxide are five important metal oxide nanoparticles. ZnO is the most often used of these five metal oxide nanoparticles for various wastewater treatment methods. CuO and TiO_2 are the most often utilised metal oxides for wastewater treatment after ZnO. ZnO, CuO, and TiO_2 are used in a wide range of applications in the literature, including adsorption, photocatalytic activity, antibacterial, and antifungal activities. The main applications of silver oxide are in photocatalytic and antibacterial processes. Iron oxide is mostly utilised in adsorption (Naseem and Durrani, 2021).

4. Iron oxides nanoparticles

One of the earth's most common elements is iron. Iron oxides are found in nature in a variety of forms, including hematite (alpha-Fe_2O_3), magnetite (Fe_3O_4), and maghemite (gamma-Fe_2O_3). Due to their greater specific surface area, high porosity, and powerful magnetic response, iron-based nanomaterials have recently demonstrated outstanding capabilities for sorption activities, leading to an extraordinary sorption capacity. According to reports, 10–20 nm nanoparticle sizes offer the best performance. Such particles have been claimed to exhibit superparamagnetic behaviour, a type of magnetism that has been observed in ferromagnetic particles (Nizamuddin et al., 2019). Nanosized ferric oxides are inexpensive adsorbents for toxic metal sorption due to the ease of synthesis and the availability of readily available resources. Due to the low risk of secondary contamination from elemental iron, nanosized ferric oxides can be injected directly to contaminated locations. Numerous studies examined the impact of various parameters on Fe_3O_4 magnetic nanoparticles' ability to remove metal ions. For instance, pH, temperature, the amount of the adsorbent, and the incubation duration all had a significant impact on the efficiency of the adsorption of Ni(II), Cu(II), Cd(II), and Cr(VI) ions by Fe_3O_4 nanoparticles. Surface functionalized Fe_3O_4 nanoparticles have been employed extensively for the removal of hazardous metal ions in compared to bare Fe_3O_4 nanoparticles. For the simultaneous adsorption of Cr(III), Co(II), Ni(II), Cu(II), Cd(II), Pb(II), and As3+ from wastewater, surface-engineered Fe_3O_4 nanoparticles exhibit a significant affinity. These nanoparticles were shown to preferentially adsorb metal ions, and it was discovered that the adsorption process was highly dependent on the quantity, surface functionality, and pH of the medium (Sadegh et al., 2017).

Because micro- and nanometer-sized particles are more likely to form agglomerates, particle size may lead to instability. Additionally, iron oxide-based nanoparticles lose their magnetic and dispersibility as a result of their active reaction to chemical reactions. These materials have increased stability when coated in carbon or silica. For the synthesis of magnetic nanomaterial, the right ratio of ferromagnetic and antiferromagnetic elements can produce a highly stabilised morphology. Material size, morphology, phase purity, and imperfections in the surface of the materials are a few other variables that can affect magnetic characteristics. Contrarily, it is still difficult to produce magnetic materials with a clear structure, greater physiochemical stability, a regulated composition, and a surface that can be tailored. A magnetic material's surface energy increases as its size reduces, from the micrometre to the nanoscale range, but the material's overall stability declines. To solve this problem, researchers have produced high-quality inverse-spinel $NiFe_2O_4$ using the hydrothermal co-precipitation method with graphene oxide as the feedstock. In this approach, utilising it as a sorbent has produced results that are both economically viable and make it easier to separate from adsorbed heavy metals. The magnetic moment of the material directly relates to the removal of heavy metals. However, in an aqueous media that also contains greasy oil and suspended particulates, a higher magnetic moment makes it simple to separate the adsorbent using a magnet. This strategy works well for avoiding high pressure drops and long separation processes like filtration. The extremely efficient adsorbent can also be used again, which lowers the overall cost. To remove colours from wastewater, iron-based nanoparticles are actually utilised. The interaction between the chemicals and the functional groups on the absorbent's surface makes this feasible. The functional groups define an absorbent's efficacy, selectivity, capacity, and reusability (Nizamuddin et al., 2019).

The textile and pulp industries are the principal users of dyes, which are primarily organic compounds. Nanomaterials made of iron-based oxide shows a variety of sorption capabilities that made them excellent in concurrently removing colours, organic pollutants, and inorganic pollutants. Methylene blue is a common heterocyclic aromatic chemical that is employed in many chemistry and biological fields. Additionally, methylene blue is used to make drugs (Nizamuddin et al., 2019). Singh et al. (2017) developed a wet-chemical process for the synthesis of superparamagnetic Fe_3O_4 nanoparticles coated with Green Tea Polyophenols (GTP), or Fe3O4@GTPs NPs. Due to their hydrophilic coating of GTPs, the synthesised NPs were extremely stable and evenly disseminated in aqueous medium. In the wastewater treatment process, these particles demonstrated a high adsorption capacity (7.25 mg/g) for the removal of the methylene blue dye. The pseudo-second-order kinetic and Langmuir isotherm models, respectively, and the sorption dynamic and equilibrium data, showed good agreement in both cases. The particles might also be readily magnetically removed from the liquid medium and possibly recycled for several cycles.

In 2013, Singh et al. revealed a simple soft-chemical method for manufacturing Fe_3O_4 embedded ZnO magnetic semiconductor nanocomposites (Fe_3O_4-ZnO MSN), and they compared their efficacy for water purification to that of their separate counterparts (Fe_3O_4 and ZnO). The formation of Fe_3O_4–ZnO MSN was evident from the detailed structural analyses by XRD, TEM and magnetic measurements. Due to their porous network structure, surface polarity, and large surface area, it has been found that these nanocomposites have a great potential to simultaneously remove Ni_2+, Cd_2+, Co_2+, Cu_2+, Pb_2+, Hg_2+, and As_3+ from waste water. These nanocomposites also show a good photocatalytic activity for the degradation of organic dyes under UV irradiation, and are found to be efficient in the easy and rapid capturing of bacterial pathogen. It has been observed that the efficiency of capturing bacteria is strongly dependent on the concentration of nanoadsorbents and their inoculation time.

By employing Fe-based MOF as a precursor, Angamuthu et al. (2017) developed a simple method to manufacture composite materials with mesopores that represent an iron oxide core and are encapsulated in carbon nanodisk shells. Fe_3O_4 nanoparticles encapsulated in mesoporous carbon matrix (FeMCNA-300) were produced by catalytic carbonization of Fe2+ coordinated 1,4,5,8-naphthaleneteracarboxylic dianhydride MOF at 725 °C under N_2 environment followed by air treatment at 300 °C. FeMCNA-300 displayed exceptional catalytic activity towards the degradation of 150 ppm of methylene blue (MB) dye in water with 84% TOC conversion in 15 min at ambient temperature. Through a regulated mesoporous channel in the FeMCNA-300 carbon nanodisk, methylene blue and the oxidant H_2O_2 interact with iron oxide nanoparticles.

In the study of Ebrahiminezhad et al. (2018), a unique iron nanostructure was effectively produced utilising a rapid and environmentally friendly method employing an aqueous extract of Mediterranean cypress (Cupressus sempervirens). In actuality, extremely reactive ultra-small iron nanoparticles with diameters of less than 1.5 nm were assembled into nanoclusters. Iron nanoclusters were found to range in size from 9 to 31 nm, with a mean diameter of 19 nm. Additionally, the produced nanoclusters shown tremendous potential for dye removal from aqueous solution in a time-dependent way. For the elimination of methyl orange, decolourization efficiency was estimated to be 95% after a 6-hour operation.

Eshaghzade et al. (2017) synthesised ferro magnetic iron oxide (Fe_3O_4) nanoparticles (NPs) using a simple and inexpensive electrochemical approach. The synthesised substance was utilised as a heterogeneous electro-Fenton catalyst to decolorize solutions containing C.I. Acid Red 14 and C.I. Acid Blue 92. Graphite electrode Modified with carbon nanotubes (CNTs) were used as cathode in both synthesis and electro-Fenton processes. The higher catalytic potential of magnetite nanoparticles was demonstrated by the remarkable discolouration across a wide pH range. Additional benefits include the ability of Fe_3O_4 NPs to be recycled following magnetic separation, the prevention of their release into the environment as a secondary pollutant, and the enhanced electro production of hydrogen peroxide at the cathode surface as a result of the presence of CNTs.

The ammonium ion, one of the inorganic contaminants in aquatic ecosystems, can impede the self-purification of lakes and rivers and lead to eutrophication. Therefore, it is crucial to minimise or completely stop ammonium leakage into aquatic systems. The traditional ion-exchange method is limited to application in smaller quantities because ammonium will be exchanged by other high-valence ions first in water and running cost tends to be high. The removal of ammonium using a biological filter is a highly successful technology that greatly outperforms conventional procedures. But building the filter comes at a considerable expense, and the subsequent processing comes with a bigger danger to safety. Ammonium ions have been widely adsorbed using activated carbon, zeolites, biomaterials, nanoparticles, and polymers, however their adsorption effectiveness has been observed to be relatively low. Additionally, nanoparticles utilised as adsorbents for extracting ammonium ions from wastewater should meet the following criteria: (I) The actual nanosorbents themselves ought to be safe. (II) The sorbents have relatively high sorption capabilities and are selective for contaminants with low concentrations. (III) The pollutant that was adsorbed onto the surface of the nano adsorbent was simple to remove. (IV) The sorbents could be regenerated indefinitely. Fe_3O_4 nanoparticles were employed in the work by Zare et al. (2016) as an effective adsorbent for the speedy removal of ammonium ion from the solvent phase. The developed adsorbent was synthesized using a chemical co-precipitation method from its precursor mixtures i.e. $FeCl_2 \cdot 4H_2O$ and $FeCl_3 \cdot 6H_2O$.

Superparamagnetic ascorbic acid-coated Fe3O4 nanoparticles with a high specific surface area were successfully synthesized by Feng et al., (2012) via an environmentally friendly hydrothermal route in the absence of any templates. The as-synthesized ascorbic acid-coated Fe_3O_4 nanoparticles have a diameter of less than 10 nm, thus leading to a high specific surface area of about 179 m^2/g, which is even larger than those of well-defined mesoporous structures. The ascorbic acid-coated Fe_3O_4 nanoparticles exhibit superparamagnetic properties at room temperature and saturation magnetization approaches 40 emu g_{-1}. Fe_3O_4 nanoparticles exhibit good adsorption behavior for removing arsenic ions.

Although a lot of methods, such as chemical precipitation, ion exchange, liquid– liquid extraction, resins, cementation, and electrodialysis have been developed for the extraction of the heavy-metal ions from industrial wastewater. However, from a practical point of view, the application of such adsorbents for treating wastewater has a major drawback, which is that it requires an additional separation step to remove the adsorbent from the solution. One of the new developments for removing heavy-metal ions from wastewater in recent years is to use magnetite as adsorbents, due to its high adsorption capacity for heavy-metals and organic pollutants. The most important advantage of using Fe_3O_4 as adsorbents or magnetic supports of catalysts is that it can be easily separated from the reaction system with an external magnetic field. However, for the practical application of magnetic powders as adsorbents, there is still a great challenge to solve the dispersibility of magnetic powders into the sewage water without any extraneous forces. Instead, Fe_3O_4 nanopartciles (NPs) with large solubility and high stability in water can be synthesized in large amounts. These water-soluble Fe_3O_4 NPs used as a fast recyclable tool for Pb_2+ and Cr_6+ removal from sewage water. It is noteworthy that the adsorption ability of the water-soluble Fe_3O_4 NPs to Pb_2+ and Cr_6+ is stronger than water-insoluble Fe_3O_4 NPs. Water-soluble iron oxide NPs can be directly dissolved and well stabilized in sewage water, and then an effective magnetic separation was used to call back the absorbent after complete adsorption (Wang et al., 2012).

EDTA Fe_3O_4 nanomagnetic chelators (NMCs), show a strong tendency towards the adsorption of Cr(III), Co(II), Ni(II), Cu(II), Cd(II) and Pb(II) from wastewater (Sadegh et al., 2017).

The adsorption of Eu_3+, La_3+, Co_2+ and Ni_2+ ions from aqueous solution by these citrate-coated magnetic nanoparticles was studied by Ngomsik et al., (2012). The kinetic study showed a rapid adsorption due to the presence of active sites on the external surface of the magnetic nanoparticles. The adsorbed amount of cations increases drastically with pH from pH 2 to 3.5 and remains constant over pH 3.5. These results show that these nanoparticles could be used in water treatment process for removal of pollutants. Indeed, this magnetic adsorbent is efficient and easily removed from the aqueous solution by applying a magnetic field.

Warner et al. (2010) demonstrated the synthesis and characterization of high-performance, superparamagnetic, iron oxide nanoparticle-based, heavy metal sorbents, which exhibit outstanding affinity for the separation of heavy metals in contaminated water systems (i.e., spiked Columbia River water). The magnetic nanoparticle sorbents were manufactured from an easily synthesised iron oxide precursor, and then a straightforward, one-step ligand exchange reaction was used to add an affinity ligand that is unique to a particular heavy metal or group of heavy metal pollutants to the nanoparticle surface. The designed magnetic nanoparticle sorbents have higher binding capabilities due to their naturally high active surface areas.

Asfaram et al. (2017) examined the simultaneous, ultrasound-assisted adsorption of brilliant green (BG) and malachite green (MG) onto Mn-doped Fe_3O_4 nanoparticle-loaded activated carbon (Mn-Fe_3O_4-NP-AC) as a new adsorbent. It was noted that cationic dyes were rapidly and simultaneously removed from their binary solutions. Small amount of adsorbent used with high adsorption capacity. Mn-doped Fe_3O_4-NP-AC was a newly developed and effective adsorbent.

5. Conclusions

Due to their large surface areas and small particle sizes, which result in a large number of adsorption active centres, nanomaterials have been extensively explored for the removal of heavy metals and dyes from wastewater based on their special features. For the removal of both organic and inorganic contaminants, adsorption techniques utilising nanoparticles are extremely efficient, practicable, and employed. These adsorbents' potential for widespread commercial use in wastewater treatment in the near future seems extremely likely.

High stability, well-dispersion in aqueous medium, high adsorption capacity, easily separation from the liquid medium, reuse in multiple cycles are important properties for the nanoparticle absorbents.

To cut prices, these nanoparticles must still be manufactured on a large scale commercially. Natural resources should be abundant, affordable, environmentally friendly, and devoid of hazardous chemicals when used for manufacturing these nanoparticles. Monodispersed nanoparticle production requires further researches.

Despite their widespread use, nanoparticles could be dangerous for humans due to their nanotoxicity. Therefore, it is essential to analyse the toxicity of nanoparticles in order to determine the relative health hazards connected to consumer exposure. Metal oxide nanoparticles, like other nanoparticles, are valuable for many applications, but due to their unrestrained use and release into the environment, there are still some health hazard concerns. To make the usage of nanoparticles more efficient and environmentally beneficial, several issues should be solved.

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