

The role of some important metal oxide nanoparticles for wastewater and antibacterial applications: A review



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ABSTRACT

Nanotechnology has great potential to improve water purification and decontamination efficiency. Nanomaterials are efficient at removing organic and inorganic pollutants and heavy metals from wastewater and killing microorganisms. The distinctive characteristics of metal oxides make them the most varied class of materials, and they have properties that cover almost all aspects of solid-state physics and material science. Only a few review articles in the literature specifically address metal oxide nanoparticles and their application in the treatment of wastewater. This review article aims to fill this gap and present a thorough review of the various types of metal oxide nanoparticles in contaminating water impurities. Metal oxide nanoparticles have great potential for treating contaminated water due to their unique properties, such as their large surface areas and low concentration. This paper discussed in detail five metal oxide nanoparticles and their applications for antibacterial and wastewater applications.

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1. Introduction

Clean water is a vital element for all living organisms. The contamination of present water resources, however, has increased globally due to rapid industrialization and the massive population explosion [1]. In agriculture, the demand for and consumption of clean water has increased on a large scale. The consumption of fresh and clean water with a high range of pollutants in industry, household sectors, and other forms of consumption is about 70%, 22%, and 8%, respectively [2].

The main classes of pollutants are heavy metal ions and dyes. Water containing such pollutants should not be used for drinking purposes without purification. Once these heavy metal ions enter water, it is extremely difficult to completely treat it [3]. These aquatic pollutants are hazardous for all living organisms and strongly affect ecosystems. Therefore, these pollutants need to be eliminated from contaminated water to prevent their harmful effects on humans and the environment. Currently, water supply entrances face many diverse challenges. All over the world, about 780 million people do not have access to clean drinking water [4].

In the affected areas, which are mostly developing countries where wastewater management is usually non-existent, urgent action is required. However, existing wastewater management and technologies are improving their ability to provide satisfactory clean water to meet human needs and other environmental needs. Current improvements and advances in nanoscience and nanotechnology suggest opportunities for the development of improved water resources and arrangements [5,6]. The extremely effective, integrated, and multifunctional progress facilitated by nanoscience and nanotechnology are predicted to offer high rates of performance and reasonable and affordable wastewater treatment solutions compared to large infrastructure [7].

The removal of these pollutants through environmentally friendly and efficient methods is crucial [8,9]. In the literature, numerous strategies have been used for wastewater treatment, such as solvent extraction, ultrafiltration, evaporation, and reverse osmosis. These techniques, however, remove impurities from water without making them harmless end products [10]. Full decomposition, either chemically or photochemically, can easily be achieved by oxidation [11]. The purpose of each oxidative process is to generate and use a hydroxyl free radical as a powerful oxidant to reduce the effects of pollutants. After activation, hydrogen peroxide may be used as an oxidant, such as UV radiation [12], as a metal ion, or as a Fenton reagent.

Nanomaterials are mainly used to overcome major water and wastewater problems. The term “nanomaterial” refers to a nanometer that is a trillionth of a meter in size [13]. Nanomaterials are widely used in the fields of environmental detection, biomedicine and pharmaceuticals, electronics and optoelectronics, the clothing industry, and cosmetics. These tiny nanomaterials lead to several changes in its physical properties such as the enhancement of the volume to surface-area ratio and the effect of quantum properties on the particle size. In contrast with conventional materials, the properties of nanoparticles, such as their magnetic, visual, and electrical properties, are significantly different compared to conventional materials. Characteristics such as high adsorption, catalytical activity, and reactivity are associated with nanomaterials [14]. Over the past few decades, nanoparticles have attracted widespread attention and have been applied effectively in different fields, including biology, sensing, medicine, catalytic chemistry, and active research and development [15,16]. Nanoparticles are commonly used in the treatment of wastewater [17]–[19]. Since nanoparticles having a large area and small sizes, they possess a strong adsorption reactivity and capacity [20]. Several pollution sources have been reported worldwide to have disintegrated into various kinds of nanomaterials, including bacteria, emerging pollutants, organic pollutants, and inorganic anions [21]–[22]. Nanoparticles are promising tools for application in different wastewater ecosystems, including carbon nanotubes, zerovalent nanoparticles, metal oxide nanoparticles, and nanocomposites [23]–[24].

Nanomaterials provide new strategies that expand on existing water supply and unconventional water sources. Over the past few years, several techniques for treating wastewater have been developed [9,18,19,25].

Some of the most important methods are reverse osmosis, solvent extraction, sedimentation, gravity separation, microfiltration, ultrafiltration, precipitation, coagulation, distillation, oxidation, adsorption, electrodialysis, electrolysis, flotation, and ion exchange [26].

Review work on metal oxide nanoparticles for wastewater applications is limited in the literature. Vikram et al. [19] discussed the current use of metal oxide nanoparticles and their impact on biological wastewater treatment processes in their review. They also summarised the different methods used to measure the inhibition of nitrification by metal oxide nanoparticles and highlight the corresponding results obtained using these methods. Yang et al. [27] in their review discussed the fate and potential effects of four types of nanoparticles: nano Zinc oxide (ZnO), silver nanoparticles (AgNPs), nano zero valent iron, and nano TiO₂ on wastewater treatment and anaerobic digestion. They discussed the impact of metal and non-metallic oxide nanoparticle on both wastewater and anaerobic sludge digestion. Sing et al. [28] focused on the application of nanoparticles in wastewater treatment. In this review, the authors discussed in detail three metal oxide nanoparticles: TiO₂, ZnO, and iron oxide. Junbai et al. [29] focused on several types of metal oxide nanoparticles including MgO, TiO₂, MnO₂, Fe₃O₄/Fe₂O₃, MnO₂, Al₂O₃, and CeO₂ and their applications in water treatment. In addition, in some reviews (on nanoparticles) some metal oxides have been outlined, but they lack a detailed review on metal oxides for wastewater treatment [30,31]. Abdelbasir et al. discussed in detail nanomaterials for industrial wastewater treatment [32]. In their work, the industrial applications of three metal oxides (TiO₂, Fe₃O₄/Fe₂O₃, and ZnO) are presented. To fill this gap and give a detailed review, this paper focuses on some recent advances and applications in wastewater treatment for metal oxide nanoparticles and highlights the potential uses of such techniques to tackle various challenges confronting existing wastewater treatment technologies. Several types of metal oxide nanoparticles i.e. iron oxide nanoparticles, ZnO nanoparticles, copper oxide nanoparticles, silver oxide nanoparticles, and titanium oxide nanoparticles are discussed in detail. The reason why these nanoparticles are chosen for wastewater treatment is because of their capability to be oxidized or dissolved in water and release metal ions, leading to metal toxicity. These metal oxide nanoparticles are chemically stable (have no adverse effects) and are used in a variety of different applications such as adsorption, photocatalytic activities, antibacterial and antifungal activities.

2. Wastewater

Wastewater is a municipal waste liquid product that contains contaminants such as organic materials, microorganisms, inorganic soluble compounds, and the use of toxic heavy metals. The chemical, biological and physical characteristics of clean water are changed by these contaminants [33]. Wastewater may be divided into municipal and industrial wastewater from waste sources, frequently contains feces and urine, industrial and agricultural wastewater sources as well as domestic compositions and organic and inorganic chemicals [34]. Fig. 1 shows different sources of wastewater. Different sources of wastewater are shown in Fig. 2.)

Wastewater contains large microorganisms, including viruses, bacteria, and protozoa and toxic substances, such as heavy metals, radionuclides, and trace elements. Wastewater is also one of the leading causes of water-borne diseases, including deadly conditions such as typhoid and cholera. Polluted water caused the death of more than 1.5 million children under the age of five in 2004 [35,36]. Fig. 1 shows the different pollutants found in water.

Wastewater treatment has become essential nowadays because of the toxic effects of pathogens and the hazards of wastewater pollution on humans, agriculture, and animals. Wastewater treatment at the personal and government level must be taken into account to protect the environment from pollution. The treatment of wastewater can involve physical, chemical, and biological procedures for water purification from various contaminants [37,38].

There are many physical characteristics of wastewater, including total solids, dye, and others (fixed, volatile, dissolved, and suspended) [39].

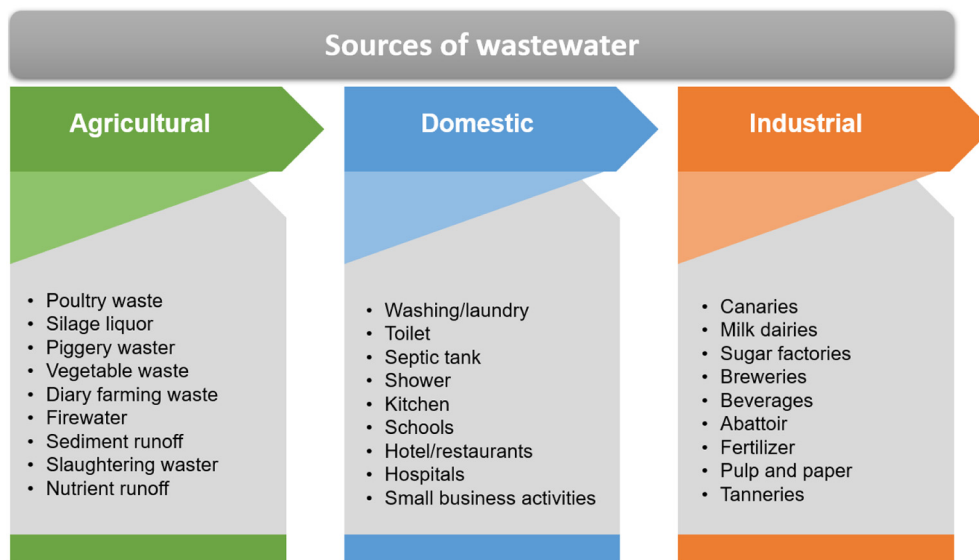


Fig. 1. Different sources of wastewater.

Total dissolved solids are dissolved matters in wastewater and can potentially include inorganic salts and metals such as bicarbonates, chlorides, calcium, magnesium, potassium, and sodium as well as small amounts of organic materials. These particle sizes range from 0.01 to 1.00 μm for dissolved solids [40,41].

The chemical pollutants in wastewater can be classified into organic, inorganic, and gaseous chemicals. Organic impurities in wastewater are generally fats and oils, carbohydrates, and proteins at about 50%, 40%, and 10%, respectively [42]. Primary impurities, surfactants, and impurities are the organic pollutants present in wastewater [37]. Chemical oxygen demand and biological oxygen demand are the most practical indicators of the quality of organic pollutants in water. Wastewater contains many inorganic pollutants such as nitrogen compounds, heavy metals, phosphorus trace elements, and other toxic inorganic constituents.

In addition to the chemical and physical characteristics, there are biological characteristics present in wastewater. The biological pollutants are living pathogenic microorganisms that exist in wastewater. The main

wastewater microorganisms are bacteria, viruses, and protozoa, which can cause acute and chronic health effects. The different kinds of bacteria in wastewater can cause different waterborne diseases such as cholera, typhoid, and shigella. However, many types of bacteria can exist in wastewater that have less serious impacts such as *Escherichia coli*, *Enterobacter*, *Klebsiella pneumonia*, *Streptococcus faecalis* and others.

Nanotechnology offers innovative water treatment solutions for catalysis, adsorption, electrostatic, reactivity and adjustable pore volume, sensors and optical electronics with high aspect ratio, hydrophilic and hydrophobic interactions [43]. Nanotechnology-based processes are effective, modular and versatile, offering high-performance, low-cost water, and wastewater solutions. In addition, nanotechnology can be expanded economically to clean up and restore unusual water sources. A summary of the technologies used for removing contaminants from wastewater is presented in Table 1. The major limitations of conventional water purification methods are presented in Table 2.

3. Nanoparticles

The main parameters of nanoparticles are their shape and size and the morphologic substructure of the substance (including the aspect ratio where appropriate). Surface and interfacial properties may be modified in

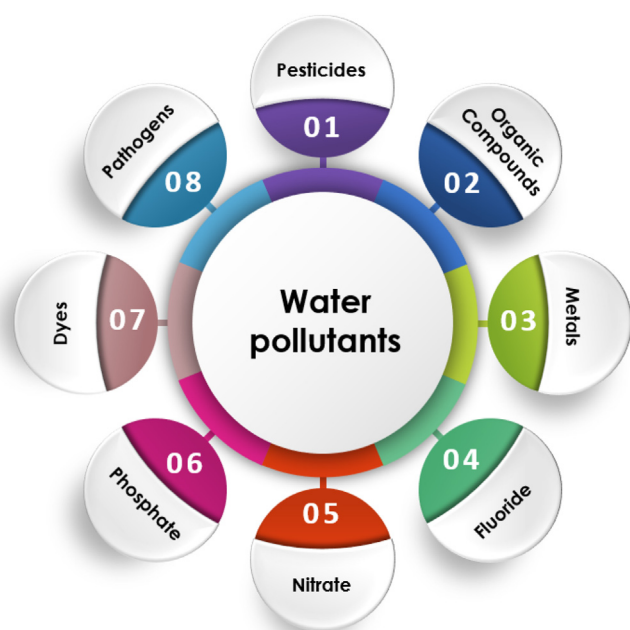


Fig. 2. Different pollutants found in water.

Table 1

Conventional techniques used for the wastewater treatment.

Method	Main characteristic(s)
Chemical precipitation	Separation of the products formed/uptake of the pollutants
Flocculation/ Coagulation	Separation of the products formed/uptake of the pollutants
Flotation	Separation process
Chemical oxidation	Use of an oxidant (e.g., Cl_2 , O_3 , KMnO_4 , ClO_2 , H_2O_2)
Biological treatment	Use of biological (mixed or pure) cultures
Filtration (Nano, Ultra, Micro and Carbon filter)	Use of a solid material and nondestructive process
Ion exchange	Nondestructive process
Incineration / Thermal oxidation	Destruction by combustion
Electrochemistry	Electrolysis (E)
Membrane filtration	Nondestructive separation
Evaporation	Separation process, Thermal process and Concentration technique,
Liquid-liquid (solvent) extraction	Separation technology
Advanced oxidation processes (AOP) Photolysis	Emerging processes, Destructive techniques

Table 2
Limitations of conventional methods of water purification.

Method	Limitations
Flotation	High initial capital cost, pH dependent selectivity and high maintenance and operational cost.
Chemical precipitation	An excess of the reagent is necessary. Its product can be of low-quality mixture that can limit its usage. It's not a very selective method.
Flocculation and coagulation	This method, which needs alkaline additives to achieve an optimum pH, is complex and inefficient.
Biological treatment	Micro-organisms are environmentally sensitive. Intermediates can destroy microbial cells. This method is expensive and time-consuming.
Ion exchange	High initial capital cost and high maintenance and operational cost.
Incineration / Thermal oxidation	High initial capital cost, high running cost.
Electrochemistry	High initial cost of the equipment, high maintenance cost.
Membrane filtration	High investment costs for small and medium-sized industries. High energy needs. The design of membrane filtration systems can vary considerably.
Evaporation	Expensive for high volumes of wastewater. High investment costs for small and medium-sized industries.
Liquid–liquid (solvent) extraction	High investment (equipment)
Advanced oxidation processes (AOP)	Economically non-viable for medium and small-sized industries, technical constraints and low throughput.
Photolysis	
Nanofiltration	This method requires pre-treatment and high-water cleaning energy. Salt and univalent ions had limited retention.
Ultrafiltration	This method requires high energy and fails to remove dissolved inorganics.
Microfiltration	This method fails to remove metals, fluoride, sodium, nitrates, volatile organics, colors and so on. This method require regular cleaning and membrane fouling can also occur.
Carbon filter	This method fails to remove fluoride, nitrates, sodium, metals and so on. This is susceptible to mold and clogging occurs with undissolved solids.

the presence of chemical agents (surfactants). These agents can indirectly stabilize against aggregation by preserving the particle charge and changing the particle's outermost layer. A very complex structure should be predicted depending on the route of growth and lifetime of a nanoparticle. For example, many different agents are condensed on the particle during cooling in the typical manufacturing process of nanoparticles, which is the subject of different ambient atmospheres. Consequently, complex surface chemical processes are required, and only a limited number of particulate model systems have been reported. Polyelectrolytes have been employed at the nanoparticles-liquid interface to alter the surface properties and interactions between particles and their surroundings. They are used in different ways in industries, including for lubrication, stabilization, adhesion, and the controlled flocculation of colloidal dispersions [44].

Depending on their size, shape and chemical and physical properties, nanoparticles can be categorized into different groups. Some of them are considered semiconductor nanoparticles, ceramic nanoparticles, polymeric nanoparticles, carbon-based nanoparticles, lipid-based nanoparticles, and metal nanoparticles. Fig. 3 shows some commonly used nanoparticles. Nanoparticles have complex chemical and physical properties based on their atomic and molecular origins. The electronic and optical characteristics and reactivity of small clusters differ entirely from the more familiar properties of each component on a large or extended surface. The weak forces of Waals Van, stronger polar and electrostatic interactions, and covalent interactions can influence the interactions between particles at the nanometer scale. The interaction between nanoparticles can determine particle aggregation in accordance with the viscosity and polarisation of the fluid. The propensity of a coagulating colloid can be improved or diminished by surface modification.

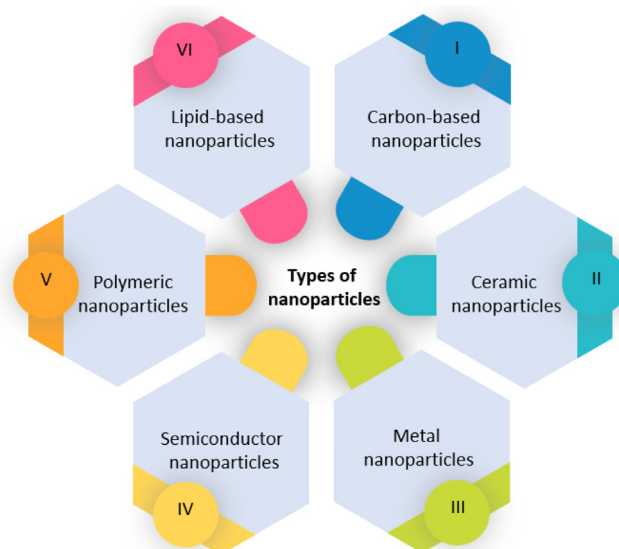


Fig. 3. Commonly used types of nanoparticles.

Details on the interactions between nanoparticles-nanoparticles and interactions between nanoparticle-fluid are very important for understanding chemical and physical processes. The small number of molecules in the active surface layer makes it difficult to characterize their properties. The relevant parameters to be considered are surface energy, charge, and solvation. Attractive or repulsive forces of interaction are necessary for regulating individual and collective nanoparticles. This relationship between nanoparticles leads to aggregates and/or agglomerates, which can affect their behaviors. The nanoparticles are designed for water attraction and are highly porous and absorb water, like a sponge, repelling dissolved salts and other impurities. Hydrophilic nanoparticles embedded in the membrane repel organic compounds and bacteria that are more likely to obstruct conventional membranes over time [45]. Different types of nanoparticles are used in wastewater treatment. Fig. 4 shows some commonly used ways to treat wastewater.

3.1. Metal oxide nanoparticles

Metal oxide nanoparticles (MONPs) are made of purely metal precursors. These nanoparticles play a significant role in many areas of physics, chemistry, and material sciences. Thermal elements are capable of forming a wide range of oxide compounds. These can adopt a vast number of structural geometries with an electronic structure that can exhibit insulator, semiconductor, or metallic characteristics. These nanoparticles have unique opto-electrical features because of their well-known localized surface plasmon resonance characteristics. Alkali nanoparticles and noble metals such as Ag, Au, and Cu have a wide absorption band in the visible electromagnetic spectrum zone. In today's state-of-the-art materials, the facet, size, and shape of the synthesis of metal nanoparticles have significant importance [46].

Owing to their advanced optical properties, metal nanoparticles have applications in many fields of research. Gold nanoparticle coating is commonly used to analyze SEM to increase the electronic stream that allows high-quality SEM images to be obtained. Nanosized metal oxides have many outstanding properties, including a high removal capacity and heavy metals selectivity. As promising adsorbents to heavy metals, they have great potential. Metal oxide-based nanomaterials include manganese oxides, nanosized iron oxides, titanium oxides, cerium oxides, ZnOs, magnesium oxides, aluminum oxides, and zirconium oxides. Fig. 5 shows some commonly used types and applications of metal oxide nanoparticles. This paper only discusses five types of metal oxide nanoparticles and their applications in wastewater treatment, as shown in Fig. 6.

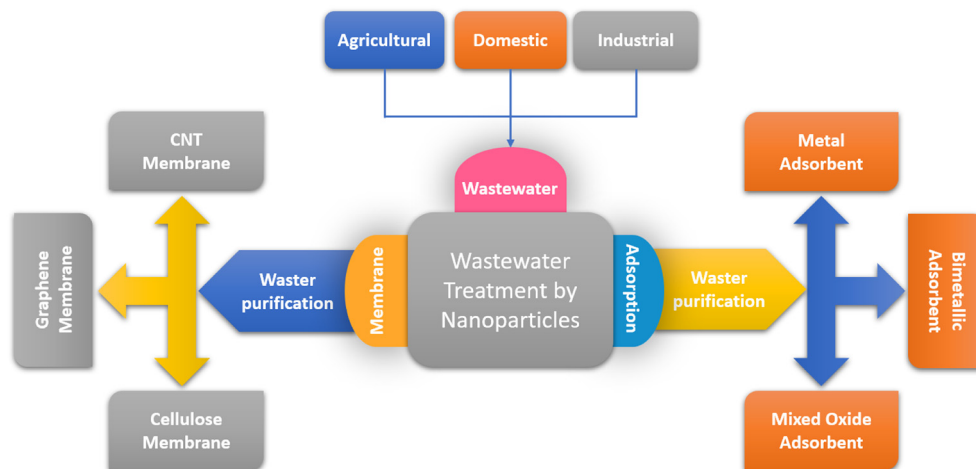


Fig. 4. Wastewater treatment by nanoparticles.

For both antibacterial activities and the removal of dye from wastewater, MONP depends on a variety of factors such as morphology, size, and aggregation. This is why synthesis techniques mainly focus on size, morphological configuration, stability, and distribution.

In this review article, we mainly focused on antimicrobial activities (mostly antibacterial), adsorption, and photocatalytic degradation. In antibacterial activities, cell wall and membrane are the significant defensive obstacles to bacterial resistance in the outside environment. In particular, the bacterial cell wall plays an important role in the preservation of the bacterial natural form. Cell-membrane-elements produce distinct gram-positive and gram-negative bacteria adsorption mechanisms. Lipopolysaccharides (LPS) is a distinctive component of a bacterial gram-negative cell wall which provides a highly loaded region that attracts nanoparticles. Teichoic acid, however only reflects gram-positive bacteria in the cell wall, so that nanoparticle is spread along the molecular chain of phosphate, stopping their aggregation. Several studies have shown that nanoparticles are more active against gram-positive bacteria than gram-negative bacteria as lipopolysaccharides, lipoproteins and phospho-lipids are part of the gram-negative cell wall, which create a binding obstacle that allows only macromolecules to enter. In comparison, the cell wall of the gram-positive bacteria contains a slim, peptidoglycan, teichoic acid and abundant pores, which allow foreign molecules to enter, which result in damage to the cell membrane and death of cells. Indeed, unlike bacteria which are gram-negative, gram-positive fungi have increased adverse stress on the cell wall surface, which can hold the nanoparticles. The process of causing bacterial mortality for nanoparticles depends on the bacterial cell's parts and components.

Adsorption kinetics is essential in describing the solvent absorption rate and the time needed for adsorption. Kinetic studies are usually conducted at

different intervals for adsorption using linear and non-linear pseudo-first- and second-order kinetics. In adsorption, the main focus is on the effect of pH, removal efficiency, and initial contact time. Isotherms of adsorption are also essential to describe how pollutants interact with adsorbent surfaces, as well as how adsorbents are used to optimize the use of pollutant removal from aqueous solutions.

Photocatalysis is the activity that happens when a light source interacts with the surface of the photocatalyst, the semiconductor materials. At least two simultaneous reactions, oxidation from photogenerated holes and a reduction of photogenerated electrons, must occur during this process.

4. Zinc oxide nanoparticles as a disinfectant

ZnO is regarded as a good photocatalyst due to its high chemical stability and excellent photocatalytic activity when removing water pollutants. ZnO possesses a wide bandgap (3.37 eV), and also the exciton binding energy (60 meV) is large at room temperature. Different ZnO nanostructures, including nanosheets, nanowires, nanobelts, nanorods, and complex hybrid structures, can be developed. Hollow spheres are of particular concern among these nanostructures due to their light-harvesting efficiencies and highly enhanced photocatalytic activity as well as their high surface area, low density, and good surface permeability.

ZnO is a promising material for conducting photocatalytic activity because of its physical and chemical characteristics such as its high electrochemical stability, super oxidative capability, and low toxicity. ZnO is therefore the first and most commonly used material for heterogeneous photocatalysis among other metal oxides. Although the photocatalytic method provides numerous benefits, a rapid recombinant photo-excited carrier in ZnO hinders the photocatalytic efficiency and generation of photocurrent. In-depth studies are now being conducted to modify ZnO with metal and non-metal additions to increase its electrical and optical properties to enhance the photocatalytic performance of ZnO.

Due to its specific properties, ZnO is considered one of the most promising catalysts for contaminated water treatment. These properties are:

1. Low cost
2. Abundance
3. Nontoxicity

5. High surface activity

In electro-mechanical nanoscale production, ZnO has great potential. The hexagonal structure (wurtzite) helps to suit and control growth. Positive Zn surfaces and negative O surfaces create electronic dipoles that allow the polarisation of voltage and temperature in certain directions and planes. At present, nanoparticles are not considered contaminants, but their water stability is crucial to assessing potential risks, since in the

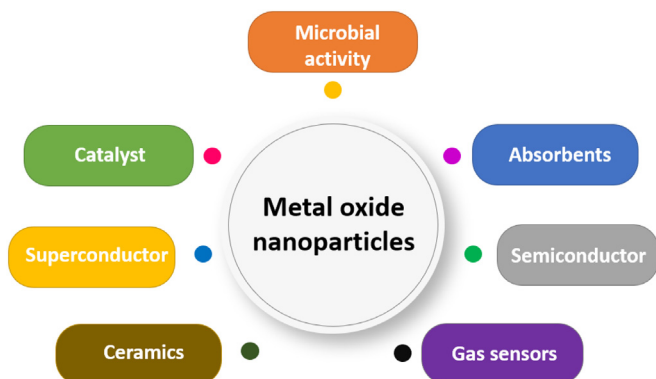


Fig. 5. Common applications of metal oxide nanoparticles.

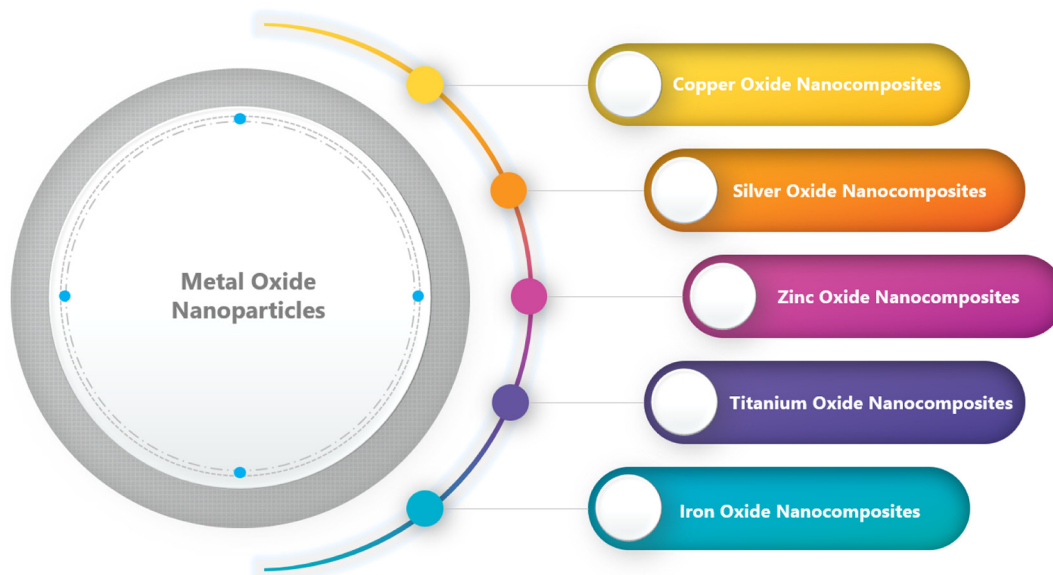


Fig. 6. Five types of metal oxide nanoparticles discussed in this paper.

future, water plants may have difficulty removing nanoparticles. The composition of nanoparticles may change their properties. Therefore, their reactivity, translocation, and penetration within the plant can lead plants to have different responses to the same nanoparticle [47]. Barrios et al. [48] showed that capping nanoparticles has an impact on plant reactions compared to exposure to bare nanoparticles. Plants interact continuously with soil, air, and water, all of which may contain engineered nanoparticles. Since plants are also consumed by animals, nanoparticles may be transferred to them. There is also a risk that nanoparticles could invade the food chain and become hazardous to humans. We refer the interested reader to article [50] for further reading on the impacts of metal oxide nanoparticles on plants.

For ZnO synthesis, a variety of techniques are used. In general, these techniques can be divided into three types: physical, biological, and chemical techniques, as shown in Fig. 7. Chemical synthesis can also be divided into the synthesis of a gas phase and liquid phase. Physical synthesis can also be divided into laser ablation, high-energy ball milling, and chemical, physical and solid vapor deposition.

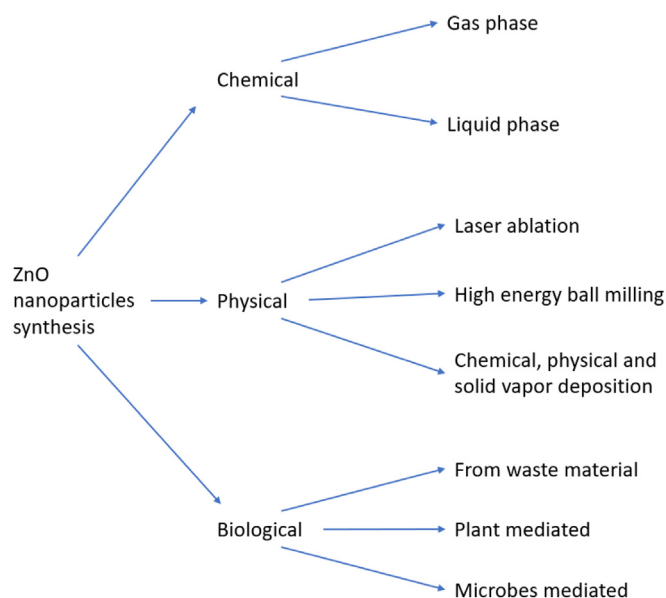


Fig. 7. Major synthetic techniques used for ZnO nanoparticles.

physical, and solid vapor deposition. Physical synthesis can also be divided into plant mediated, microbe mediated, and from waste materials.

Baruah et al. extensively examined the prospects of photocatalysis as a successful disinfectant method [49]. A metal oxide for oxidizing pollutants in carbon dioxide and water is included as a photocatalyst. If light energy, such as UV and visible light, is irradiated by a photocatalyst that has higher gap energy than the material, electrons from the valence strip are excited to the conduction strip, leaving positive lengths on the valence strip. The electrons are generated with the valence belt hole and conduction band. In the degradation of microbial contaminants, photogenerated electrons and holes were found.

The photocatalytic activity of various ZnO concentrations in infected water has been studied by [50]. ZnO has a particle size of 20–40 nm and has been synthesized from $\text{Zn}(\text{NO}_3)_2$ and $(\text{NH}_4)_2\text{CO}_3$. Increased photocatalytic efficiency leading to an increase in the constant decay of bacteria has been reported as a result of the reduced particle size and the effect of quantum containment activating ZnO to produce reactive oxygen species (ROS).

In an experiment in [37], ZnO concentrations were lower than in other studies with antibacterial activity. This was supposedly due to the smaller particle sizes or the relatively low salt/protein growth medium (minimum Davis medium) that minimized the coagulation of nanoparticles. The study also showed that gram-positive *Bacillus Subtilis* was deeper than gram-negative *Escherichia coli* by adding nanoparticles.

Ray et al. [51] studied the possibility that a small amount of UV or fluorescent light known to emit 4% of UV light can activate ZnO in the laboratory. Results show that environmental laboratory conditions for ZnO nanoparticulate are sufficient for biocidal activity and that they probably depend upon the size of the nanoparticles. The results demonstrate that ZnO can be used in visible light as an antibacterial.

Removal methods of commercial nanoparticles for water treatment have not been thoroughly investigated. Zhang et al. [52] tested and removed commercial metal oxide nanoparticles Fe_2O_3 , ZnO, NiO, and SiO_2 for their properties, discursiveness, and water stability. Nanoparticles are quickly combined in tap water as well as silica and are not destroyed by ultrasound or chemical dispersants. The quantity of alcoholic material removed is less than 80% of the nanoparticles' total mass. The behavior of metal oxide nanoparticles in water has been reported to depend on their physical properties and interaction with other water components [53].

Tso et al. [54] in their study showed that nanoparticles quickly aggregate and precipitate into pure water on the stability of three metal oxide nanoparticles: SiO_2 , ZnO, and TiO_2 , in an aqueous solution. The most efficient method for partitioning nanoparticles in water is using ultrasound

effects. The results show that nanoparticles changed their stability under various water conditions. The presence of organic colloids (surfactants and humic compounds) means that samples of water and wastewater aggregate faster than pure water. This is in line with Li et al.'s [55], finding that the toxicity of ZnO nanoparticles in water is water chemistry.

Esmailzadeh et al. [56] tested nanocomposites by mixing low-density polyethylene and ZnO with *Bacillus subtilis*, a common food spoiler bacterium, and *Enterobacter aerogenes* (food and water) produced on pathogens. For gram-positive bacteria, antibacterial effects appear more pronounced. The study also showed a direct relationship between the antimicrobial effect and the ZnO nanoparticle concentration in the composition.

ZnO polyurethane nanocomposite (ZPN) gram-negative (*Escherichia coli*) and gram-positive (*Bacillus subtilis*) coatings with a different concentration of ZnO nanoparticles of 0.1% to 2.0% ZPN coating antibacterial activities of bacteria. In El Saeed et al. [57] gram-positive was observed to be more sensitive to gram-negative bacteria under 2% ZnO nanoparticles.

Motshelga et al. [58] developed a nanocomposite in which bentonite-supported silver and ZnO nanoparticles are synthesized and distributed to chitosan. To test antibacterial activity, *Enterococcus faecalis* bacteria were used. Bentonite-chitosan nanocomposites containing both silver and ZnO showed good antibacterial activity. ZnO, however, showed the best antibacterial activity with a removal efficiency of at least 78%. It has also been suggested that nanoparticles' antibacterial activity is also affected by the bacterial concentration.

Adams et al. reported that *Bacillus subtilis* and *Escherichia coli* are antibacterial to ZnO nanoparticles [37]. The studies reported that antibacterial activity is not affected by different sizes of particles, and their activity is similar in light and dark activity on *Bacillus subtilis*, but there is more light activity on *Escherichia coli* [37]. For *Bacillus subtilis*, 90% growth reduction was observed at 10 ppm. For *Escherichia coli*, only 48% growth reduction was observed at 1000 ppm. Premanathan et al. reported that bacterial ZnO nanoparticles activity on gram-positive bacteria such as *Staphylococcus aureus* is better than on gram-negative bacteria such as *Escherichia coli* [59]. It was observed that the effect on the Gram-positive bacterium *Staphylococcus aureus* were more than Gram-negative bacteria *Escherichia coli* and *Pseudomonas aeruginosa*. Table 3 presents existing methods of ZnO nanoparticles and their application for wastewater treatment. ZnO nanoparticle synthesis methods with the size range are presented in Table 4.)

6. Copper oxide nanoparticles

The exact period of the discovery of copper is not known, but it is estimated at about 9000 BCE in the Middle East [62]. Copper is the oldest metal used by humans, as it was first used by Egyptians in about 2000 BCE for the sterilization of wounds and water. Copper has many positive features such as good corrosion resistance, low cost, and antimicrobial activity [72].

Copper (II) oxide is a monoclinically structured semiconductor. It has many useful chemical and physical characteristics, such as superconductivity at high temperatures, solar energy efficiency, relative stability, low cost, and antibacterial activity [73]. Due to their high electrochemical capabilities, CuO nanoparticles have different technical applications such as catalysis and can be used in batteries. Nanoparticles of diverse sizes and shapes can be synthesized using different methods such as sonochemical techniques, electrochemical techniques, high-temperature combustion, and new quick precipitation techniques.

The use of CuO nanoparticles with a narrow size distribution for these applications would further promote the nanoparticles' chemical reactivity because the size of the particle decreases the surface-to-volume ratio and thus the number of reactive sites increases [74]. CuO's electronic and optical properties are therefore improved to the bulk of their equivalent [77]. As a result, many methods for the synthesis of CuO nanoparticles of different dimensions have been developed as shown in Table 2.

Zhu et al. [89] prepared highly dispersed CuO nanoparticles using precursor and sodium hydroxide as a reduction agent. The synthesized CuO nanoparticle had an average size of 6 nm. The solution's plasma method

can be used to synthesize CuO nanoparticle with a good performance [42]. The advantages of this method are that the use of complicated devices is not necessary and the size and shape of the CuO nanoparticle can be easily controlled. CuO nanoparticle is produced as a copper cable cathode, and the electrolyte is a citrate buffer K_2CO_3 (pH: 4.8), with a voltage of 105–130 V. CuO nanoparticle flower is below 100 nm. As the concentration of K_2CO_3 electrolyte decreases, the size of CuO nanoparticle decreases, and spherical and porous spherical CuO nanoparticle is obtained when voltages of 105 and 130 V are applied, respectively.

A copper hydroxide/oxide layer on Cu nanocube surfaces has been found to play a major part in enhancing the hydrocatalyst's electrocatalytic activity and stability. Other researchers successfully applied Cu_2O nanotubes wrapped in nanoscales of graphene (Cu_2O / GNs), which represent promising, highly sensitive, enzyme-free glucose, and hydrogen peroxide sensors [92].

Due to its inherent compatibility, low-cost manufacturing, and excellent electrochemical properties, copper oxide nanomaterial research has grown significantly over the past year. Copper oxide shows two types of copper oxide (Cu_2O) and copper oxide (CuO) polymorphs. The two main stoichiometric compounds in the CuO system are these oxides. Pure copper oxide is a 6.4 g / cm^3 black solid. It also has a melting point of 1330°C and is water-insoluble. CuO is a relatively small bandgap (1.2–1.85 eV) intrinsic p-type semiconductor and has many attractive features that allow it to be used in various applications.

Bacterial disinfectants are one of the major applications of copper and copper compounds, which are critical to low levels of human and biological activity due to their versatility and low cost [75]. V.B.P. Sudha et al. tested the effect of a copper device (15.2 cm^2 / L area of a the copper coil on the volume of water) on *Escherichia coli*, *Vibrio cholerae*, and *Salmonella typhi* in water, and the result showed that when the device was on a glass bottle, there was no growth of *Vibrio cholerae*, *Salmonella typhi* and *Escherichia coli* after overnight incubation, with 935, 688 and 502 CFU, respectively, before incubation [90]. Gustavo Faúndez et al. [91] studied the activity of copper against suspension *Campylobacter jejuni* and *Salmonella enterica* at different temperatures: 10°C and 25°C . The result showed that the copper surface has good antibacterial activity at these temperatures, but higher efficacy at 25°C [75].

Fathima et al. used sodium dodecyl sulphate (SDS), TritonX-100, dodecyl trimethyl ammonium bromide, and other fast deposition methods as anions, cations, and neutral surfactants, respectively. CuO nanorods have been extensively studied. Depending on the negative charge of the surfactant, the ion SDS has the strongest interaction with cationic CuO nanoparticles. Other results show, however, that surfactants play an important role in nanomaterial form and application [92].

Few studies have been conducted on the antibacterial activity of CuO nanoparticles. Augustin et al. showed that synthesized CuO nanoparticles have good antibacterial activity against methicillin-resistant *Staphylococcus aureus* and *Escherichia coli*, with minimum bactericidal concentrations [76]. Although they have an antibacterial effect on CuO, NiO, ZnO, and Sb_2O_3 nanoparticles, CuO nanoparticles have been shown to be the most toxic on gram-negative bacteria like *Bacillus subtilis* among these metal oxide nanoparticles. In turn, CuO nanoparticles are more active than gram-positive bacteria against *Escherichia coli* [39]. The effects on *Vibrio fischeri*, *Daphnia magna*, and *Thamnocephalus platyurus* were investigated by bulk and nano-CuO. The results show that CuO nanoparticles are more anti-bacterial than CuO bulk [70].

The exact mechanism of bacterial disinfection by CuO is not clear. However, limited suggestion mechanisms are reported, with one of these mechanisms being that the released Cu ions from the nanoparticles come into contact with the bacterial cell membrane, which damages bacterial cell membranes. Ruparelia et al. suggested that the released Cu ions can lead to disorder in the DNA helical structure through the interaction of the ions with DNA molecules [93]. The latest proposed mechanism is oxidative stress. Ivask et al. [71] reported that ROS can be induced by CuO nanoparticles, depending on the CuO nanoparticle decomposition rate, where ROS can damage the bacterial cell structure. However, the mechanism is only

Table 3
Existing methods of ZnO nanoparticles and their application for wastewater treatment.

S. No.	Year	Application	Characteristics	Reference
1	2012	Antimicrobial Activity: <i>Staphylococcus aureus</i> and <i>Escherichia coli</i>	Surface area: Sample1 = 34.27 cm ² , Sample2 = 47.54 cm ² and Sample3 = 39.12 cm ² . Particle type: nanorods. Average particle size 5–7 nm. Under sunlight, 99% <i>Staphylococcus aureus</i> and <i>Escherichia coli</i> could be immobilized. 80% of <i>Escherichia coli</i> and 59% of <i>Staphylococcus aureus</i> cells could be inactivated under room lighting conditions.	[52]
2	2010	Antimicrobial Activity: <i>Escherichia coli</i> Photocatalytic degradation: methylene blue	Particle sizes 80 and 260 nm. Particle type: nanorods. For <i>Escherichia coli</i> , maximum inhibition zone was 4.4 cm ² . Under white-light irradiation at 963 Wm ⁻² , Methylene blue photodegradation was 93%, methyl orange photodegradation was 35%.	[60]
3	2010	Photocatalytic degradation: methylene blue	Average particle size 5–7 nm. Particle type: nanorods. Surface area: Sample1 = 34.27 cm ² , Sample2 = 47.54 cm ² and Sample3 = 39.12 cm ² . The photocatalytic activity of methylene blue was observed to improve by ≈ 8%.	[61]
4	2011	Antimicrobial Activity: <i>Escherichia coli</i>	Average particle size 20–40 nm. A quite high (0.24 min ⁻¹) bacterial decay constant was observed for antibacterial activity.	[50]
5	2006	Antimicrobial Activity: <i>Escherichia coli</i> and <i>Bacillus subtilis</i>	Particle sizes 67 and 820 nm. 90% growth reduction of <i>Bacillus subtilis</i> was observed at 10 ppm. For <i>Escherichia coli</i> , only 48% growth reduction was observed at 1000 ppm.	[34]
6	2008	Antimicrobial Activity: <i>Staphylococcus aureus</i>	Average particle size 84 nm. Nanoparticles with smaller particle sizes had 95% growth inhibition at 1 mM concentration (0.008%), with relatively larger particle sizes, with 5 mM of ZnO showed only 40–50% growth inhibition.	[51]
7	2011	Antimicrobial Activity: <i>Escherichia coli</i>	Average particle size 19 ± 7 nm. The toxicity to <i>Escherichia coli</i> of nano-ZnO in the 5 media deceased as follows: ultrapure water > NaCl > minimal Davis > Luria-Bertani > phosphate-buffered saline.	[55]
8	2015	Antimicrobial Activity: <i>Enterobacter aerogenes</i> and <i>Bacillus subtilis</i>	Length and diameter of particle 400 and 50 nm. Nanocomposites containing 2 and 4 wt% ZnO can reduce the growth of both <i>Bacillus subtilis</i> and <i>Enterobacter aerogenes</i> . Stronger inhibitory effect was found for 4 wt% ZnO containing nanocomposite.	[56]
9	2015	Antimicrobial Activity: <i>Escherichia coli</i> and <i>Bacillus subtilis</i>	Average particle size 20 nm. at 2.0 wt% loading level, the ZnO nanoparticles showed the obvious inhibitory effect on the growth of both <i>Bacillus subtilis</i> and <i>Escherichia coli</i>	[57]
10	2015	Antimicrobial Activity: <i>Escherichia coli</i> and <i>Enterococcus faecalis</i>	Average particle size 86 nm. Maximum removal efficiency was found to be 78%.	[58]
11	2013	Antimicrobial Activity: <i>Escherichia coli</i>	The pH ranging from 5.7 to 8.7 showed no viable effect. Sharp decrease of bacterial mortality was observed from 80 to 90% at pH 8.3 to about 10–20% at pH 8.7.	[62]
12	2011	Antimicrobial Activity: <i>Pseudomonas aeruginosa</i> , <i>Staphylococcus aureus</i> and <i>Escherichia coli</i>	Average particle size 25 nm. The effect on the Gram-positive bacterium <i>Staphylococcus aureus</i> were more than Gram-negative bacteria <i>Pseudomonas aeruginosa</i> and <i>Escherichia coli</i> .	[59]
13	2018	Antimicrobial Activity: <i>Escherichia coli</i> and <i>Salmonella typhimurium</i> Photocatalytic degradation: methylene blue	Particle size between 2 and 50 nm. MB equilibrium data was best fitted to the Dubinin-Radushkevich model. The adsorption energy (E) was between 1.76 and 2.00 kJ/mol.	[63]
14	2019	Adsorption: Cr(VI)	Average particle size 31 nm. The maximum adsorption capacity for Cr(VI) was at pH of 2. Increasing the pH value results in decreasing adsorption capacity. The maximum monolayer adsorption capacity was 139.47 mg/g at 50°.	[64]
15	2018	Adsorption: Reactive Blue 19 (RB19) and Acid Black 210 (AB210) dyes	Average particle size 12 nm. Fast Adsorption rate with an equilibrium adsorption established after 15 min of shaking times. RB19 removal efficiency was highest at pH ~3. Both dyes followed the pseudo-second-order model. The isothermal model Langmuir was best suited to the experimental data. The maximum of adsorption capacities for RB19 and AB210 were 38.02 and 34.13 mg/g.	[65]
16	2017	Adsorption: Pb(II)	Particle sizes 10.01 ± 2.6 nm The pseudo-second-order model was followed by the adsorption process. The maximum removal was observed to be 93% at pH 5. The adsorption process was endothermic and spontaneous.	[66]
17	2018	Adsorption: As(III)	The pseudo-second-order model was followed by the adsorption process. The maximum removal was observed to be 52.63 mg/g at pH 7. The adsorption process was endothermic and spontaneous. The isothermal model Langmuir was best suited to the experimental data.	[67]
18	2019	Adsorption: Azo dyes	Particle sizes 75–150 nm. ZnO-NPs of amount 0.3 g showed maximum removal efficiency of each dye (40 ppm) at pH 6. The isothermal model Langmuir was best suited to the experimental data. The adsorption process followed the pseudo-second-order model.	[68]
19	2019	Adsorption: Arsenic (As(V))	Average width of particle around 7 nm and average length about 80 nm. The maximum capacity of 4421 mg/g at neutral pH (7). The isothermal model Langmuir was best suited to the experimental data.	[69]
20	2008	Antimicrobial Activity: <i>Crustaceans Daphnia magna</i> , <i>Vibrio fischeri</i> , and <i>Thamnocephalus platyurus</i>	Particle sizes 25–70 nm. All Zn formulations were very toxic: L(E)C ₅₀ (mg l ⁻¹) for nano-ZnO, bulk ZnO and ZnSO ₄ 7H ₂ O: 8.8, 3.2, 6.1 (<i>Daphnia magna</i>); 1.8, 1.9, 1.1 (<i>Vibrio fischeri</i>) and 0.24, 0.18, 0.98 (<i>Thamnocephalus platyurus</i>), respectively.	[70]
21	2010	Antimicrobial Activity: <i>Escherichia coli</i>	Average particle size 30 nm. Surface area (m ² g ⁻¹) 12.9 Toxicity (30-min and 2-h EC ₅₀ , mg compound l ⁻¹) of bZnO for <i>E. coli</i> AB1157 = 849 ± 180, <i>E. coli</i> J1130 = 932 ± 143, <i>E. coli</i> J1131 = 624 ± 81, <i>E. coli</i> AS393 = 612 ± 107, <i>E. coli</i> J1132 = 43 ± 6.9, <i>E. coli</i> AS391 = 16 ± 6.1.	[71]

Table 4

ZnO nanoparticle synthesis methods with the size range [74].

Preparation method	Size (nm)	Reference
Sol-gel	1–10	[75]
Microwave irradiation	3–5	[76]
Colloid-thermal synthesis	3 and 5	[77]
Alcoholthermal decomposition of copper acetate	3–9	[78]
Electromechanical synthesis	4	[79]
Precipitation synthesis	4	[80]
Microemulsion system	5–35	[81]
Alcoholthermal decomposition of copper acetate	6	[82]
Sol-gel techniques	7–9	[83]
Sonochemical synthesis	10 to several microns	[84]
Precipitation pyrolysis	11–35	[85]
Solid state reaction	15–20	[86]
Thermal decomposition	15–30	[87]
Spinning disk reactor	20–30	[88]

used on *Escherichia coli* bacteria. Nwanya et al. [94] obtained Cu₂O nanoparticles in red-colored cubic by a simple green-synthesis route that is eco-friendly. At 600 °C, the Cu₂O nanoparticles oxidize thermally to pure monoclinic CuO nanoparticles. Copper oxide nanoparticles confirm their efficacy in the photodegradation of wastewater and anti-microbial activity. 91% and 90% degradation of wastewater was observed for BM and TE, respectively. Although CuO is effective in stopping the growth of *Escherichia coli* and *Staphylococcus aureus*, it is better suited for *Pseudomonas aeruginosa* and *Bacillus licheniformis*.

Mousa et al. [95] prepared CuO nanoparticles by the rapid precipitation method in the absence and presence of tetraocylammonium bromide (TOAB), which is used as a stabilizer for controlling the size of the nanoparticle. The stabilized nanoparticles of CuO-TOAB displayed greater antibacterial activity than without using TOAB. Remarkably high nanoparticles surfactant activity occurred when samples of wastewater were tested at 25 °C and 35 °C. CuO nanoparticles with and without a surfactant TOAB improved antibacterial activity marginally by reducing the pH value for wastewater. In contrast to anti-bacterial activity with shaking, the antibacterial activity of CuO nanoparticles without shaking demonstrated between 70 and 90% lower activity for CuO nanoparticles without and with the TOAB surfactive. Table 5 presents existing methods for working with CuO nanoparticles and their application for wastewater treatment.

7. Silver oxide nanoparticles

Silver oxide (Ag₂O) nanoparticles are spherical or faceted high surface area oxide magnetic nanostructured particles. The particle size of silver oxide nanoparticles is usually between 20 and 80 nm with a surface area between 10 and 50 m²/g. Silver oxide nanoparticles are also available in coated and dispersed, high purity, ultra-high purity, and transparent forms. They can also be dispersed in the production group of AE nanofluid. Nanofluids are generally defined using surfactants or surface charge technology in the solution as suspended nanoparticles. Other nanostructures include nanohorns, nanocomposites, nanorods, nanopyramids, and nanowhiskers. Silver oxide nanoparticles, which have excellent antibacterial activity, are being studied and are currently being used for many commercial products and are among many of the nanomaterials reported as antibacterial agents [107].

Ag₂O nanoparticles have been synthesized using different methods that, as shown in Fig. 8, can be classified as gas, solid, and liquid-phase routes. The methods of chemical and physical synthesis are well known for Ag₂O nanoparticles [108].

Jiang et al. [109] synthesized a silver oxide nanoparticle aggregation. The synthesis showed a superb photocatalytic performance under artificial light sources and sunlight. Results indicate that methyl orange was decomposed completely in 40 min under near-infrared light and in 120 s under sunlight, artificial ultraviolet light, and the irradiation of artificial visible light. Li et al. [110] proposed a green combustion synthesis of Ag₂O nanoparticles using *Lippia citriodora* plant powder. The Ag₂O

nanoparticles have excellent antifungal activity against *A. aureus* and are antibacterial against *S. aureus*.

Shah et al. [111] used *Paeonia emodi* fresh leaves extract as a reducing agent to synthesize Ag₂O nanoparticles. The synthesized nanoparticles reduced 97.78% methylene blue in 180 min. The antibacterial activity of the Ag₂O nanoparticle was tested against two gram-positive and two gram-negative bacteria. The synthesized Ag₂O nanoparticles have strong growth inhibitors of gram-negative bacteria.

Vithiya et al. [112] used *Bacillus thuringiensis* SSV1 to synthesize Ag₂O nanoparticles. The synthesized nanoparticles were spherical and monodispersed. The silver oxide nanoparticles showed inhibitory effects with *Staphylococcus aureus* (Gram-positive) and *Escherichia coli*, *Pseudomonas aeruginosa*, *Proteus mirabilis*, *Enterococcus faecalis* (Gram-negative). Li et al. [113] presented a green synthesis of Ag₂O nanoparticles using various plant extracts. The synthesized nanoparticles showed strong growth-inhibiting activity against *Staphylococcus aureus*.

Manikandan et al. [113] synthesized Ag₂O nanoparticles using *Ficus benghalensis* prop root extract. The synthesized nanoparticles showed excellent antibacterial activities against *Streptococcus mutans* and *Lactobacilli* sp. Abdulrazzak et al. [114] synthesized Ag nanoparticles as a mixture of AgO, Ag₂O, and Ag from silver nitrate and hydrogen peroxide. The synthesized nanoparticles showed an inhibitory effect on *Acinetobacter baumannii* bacteria. Table 6 presents existing methods of Ag₂O nanoparticles and their application for wastewater treatment.

8. Titanium oxide nanoparticles

In recent decades, the most comprehensively studied metal oxides have been titanium oxide (TiO₂) nanoparticles. Owing to its photostability, reasonable price, high photocatalytic activity, and biological and chemical stability [117], TiO₂ is to date the most exceptional photocatalyst. Usually, charge separation within particles is induced in TiO₂ because of the large bandgap energy (3.2 eV) and ultraviolet (UV) excitation. TiO₂ nanoparticles possess little selectivity, which makes these nanoparticles suitable for the degradation of all kinds of contaminants, such as polycyclic aromatic hydrocarbons [118], chlorinated organic compounds [119], dyes [120], pesticides [122], phenols [121], cyanide [122], arsenic [123] and heavy metals [124]. The photocatalytic properties of TiO₂ nanoparticles can kill a wide array of microorganisms, such as gram-positive and gram-negative bacteria as well as viruses, algae, fungi, and protozoa [125].

Titanium oxide is also used in a variety of applications such as disinfection agents and white pigment, food color flavor enhancer additives, and in the decomposition of organic compounds [126]–[128]. The three different crystalline forms of TiO₂ are anatase, rutile, and brookite. The pure form of rutile and anatase can be synthesized at low temperatures and are preferred for the photocatalytic process [129]. Ti₂O nanoparticles have been synthesized using different methods, as shown in Fig. 9.

TiO₂ nanometal is relatively less expensive than any other nanomaterial and exhibits good thermal and chemical stability and low human toxicity [129]. In addition to their photocatalytic properties, they are widely used in wastewater purification and anti-biofouling. The main advantage of TiO₂ nanoparticles is that they have an endless lifetime and remain unchanged during the degradation process of microorganisms and organic compounds [130].

Some of TiO₂'s other benefits in terms of photo-induced hydrophilicity and high oxidant power make this membrane process attractive. The creation of self-cleaning membranes can reduce fouling and preserve the permeation of membrane water. In the literature on preparing TiO, several methods have been reported. The nanoparticles are filled-membrane systems; for example, the membrane surface is immobilized or TiO is added to the casting solutions. Indeed, blending, dip or spin coating, physical or chemical cross-linking, and hot pressing are some of the commonly used methods for incorporating nanoparticles on membrane surfaces [131,132].

Madaeni et al. [133] reported two membrane preparation procedures using TiO. In the first method, green chemistry is used, where a

Table 5

Existing methods for working with CuO nanoparticles and their application for wastewater treatment.

S. No.	Year	Application	Characteristics	Reference
1	2009	Antimicrobial Activity: <i>Escherichia coli</i> and <i>Staphylococcus aureus</i>	Particle sizes in the range 20–95 nm. Mean surface area 15.69 m ² /g. In the presence of 1000 g/ml of CuO, populations of Gram-negative (× 3 strains) and Gram-positive (× 4 strains) organisms tested were reduced by 65% and 68%, respectively,	[73]
2	2008	Antimicrobial Activity: <i>Bacillus subtilis</i> , <i>Staphylococcus aureus</i> , and <i>Escherichia coli</i>	Average particle size 9 nm. Disk diffusion studies with <i>Staphylococcus aureus</i> , and <i>Escherichia coli</i> revealed effectiveness with CuO. The highest sensitivity to CuO nanoparticle was depicted by <i>Bacillus subtilis</i> .	[93]
3	2010	Antimicrobial Activity: <i>Escherichia coli</i>	Average particle size 30 nm. Surface area (m ² g ⁻¹) 12.9 Toxicity (30-min and 2-h EC ₅₀ , mg compound l ⁻¹) of cCuO for <i>E. coli</i> AB1157 = 50.5 ± 15, <i>E. coli</i> JI130 = 39.7 ± 16, <i>E. coli</i> JI131 33.0 ± 1.9, <i>E. coli</i> AS393 = 47.6 ± 5.5, <i>E. coli</i> JI132 = 14.8 ± 0.1, <i>E. coli</i> AS391 = 11.4 ± 5.4.	[74]
4	2019	Photocatalytic degradation: methylene blue (MB) and textile effluent (TE) Antimicrobial Activity: <i>Staphylococcus aureus</i> , <i>Escherichia coli</i> , <i>Bacillus licheniformis</i> and <i>Pseudomonas aeruginosa</i>	The nanoparticle sizes range from 10 to 26 nm, 36–73 nm and 30–90 nm for the unannealed Cu ₂ O, 300 °C and 600 °C annealed CuO respectively. The best degradation ability was shown by 600 °C annealed CuO, methylene blue (MB) = 91%, textile effluent (TE) = 90%. 300 °C annealed CuO showed best antimicrobial activities on <i>Staphylococcus aureus</i> , <i>Escherichia coli</i> , <i>Bacillus licheniformis</i> and <i>Pseudomonas aeruginosa</i> .	[94]
5	2015	Antimicrobial Activity: <i>Enterococcus faecalis</i> , <i>Fecal coliform</i> and <i>Total coliform</i>	Particle sizes in the range 7–12 nm. The best activity on <i>Enterococcus faecalis</i> = 92%, <i>Fecal coliform</i> = 89% and <i>Total coliform</i> = 88% was at pH = 6. The bacteraial inhibition growth rate was decreased when pH is increased after 6.	[95]
6	2018	Antimicrobial Activity: <i>Vibrio anguillarum</i> , <i>Proteus mirabilis</i> , <i>Bacillus cereus</i> , <i>Edwardsiella tarda</i> , <i>Staphylococcus aureus</i> , <i>Aeromonas hydrophila</i> , and <i>Aeromonas caviae</i>	Average particle size 61.7 nm. <i>Bacillus cereus</i> was more susceptible to biosynthesized CuO NPs than all other pathogens tested. Best inhibition zone was found to be 25.3 ± 1.80 for <i>Bacillus cereus</i> using 100 (µg/ml) of CuO nanoparticle.	[96]
7	2019	Antimicrobial Activity: <i>Staphylococcus aureus</i> and <i>Escherichia coli</i>	Particle sizes in the range 7–14 nm. The CuO nanoparticle's minimum inhibitory concentration (MIC) against <i>Escherichia coli</i> and <i>Staphylococcus aureus</i> were 3.75 and 2.50 mg/ml, respectively.	[97]
8	2014	Antimicrobial Activity: <i>Salmonella typhimurium</i> , <i>Klebsiella pneumoniae</i> and <i>Enterobacter aerogenes</i>	Particle sizes in the range 5–8 nm. The CuO nanoparticle's minimum inhibitory concentration (MIC) against <i>Salmonella typhimurium</i> , <i>Klebsiella pneumoniae</i> and <i>Enterobacter aerogenes</i> were 0.15, 0.55, and 0.30 µmg/ml, respectively.	[98]
9	2014	Antimicrobial Activity: <i>Pseudomonas aeruginosa</i> , <i>Escherichia coli</i> , <i>Enterococcus faecalis</i> , <i>Klebsiella pneumoniae</i> , <i>Proteus vulgaris</i> , <i>Shigella flexneri</i> , <i>Salmonella typhimurium</i> , and <i>Staphylococcus aureus</i>	Average particle size 23 nm. <i>Escherichia coli</i> , and <i>Enterococcus faecalis</i> exhibited the highest sensitivity to CuO nanoparticles <i>Klebsiella pneumoniae</i> was the least sensitive.	[99]
10	2019	Antimicrobial Activity: <i>Escherichia coli</i> and <i>Salmonella typhimurium</i>	Particle size 190.93 ± 2.84 nm. CuO nanoparticle showed effective antibacterial nanomaterial, significantly inhibiting the growth of both <i>Escherichia coli</i> and <i>Salmonella typhimurium</i> bacteria.	[100]
11	2014	Antimicrobial Activity: <i>Bacillus anthracis</i>	Average particle size 60 nm. Best efficacy was 99.92% of 7 × 10 ⁵ CFU/ml <i>Bacillus anthracis</i> cells within 30 min for CuO of 0.5 mg/ml.	[101]
12	2016	Adsorption: basic red 14 (BR 14) and basic violet 16 (BV 16)	Average particle size 9 nm. The optimum conditions were selected as adsorbent dosage of 0.5 g/l, pH 7 and contact time of 30 min. The isothermal model Langmuir was best suited to the experimental data. Maximum adsorption capacity for BR 14 = 27.4(mg/g) and BV 16 = 16.8 (mg/g).	[102]
13	2017	Adsorption: malachite green oxalate (MGO) and methyl orange (MO)	The pseudo-second-order model was followed by the adsorption process. The pseudo-second-order model was followed by the adsorption process. The adsorption process was endothermic and spontaneous. The isothermal model Freundlich was best suited to the experimental data. For CuO nanoparticle, the maximum dye removal was observed at pH 8 for MGO (83.4%) and at pH 2 for MO (93.2%).	[103]
14	2012	Adsorption: Arsenic (As(V))	Average particle size 40 nm. The isothermal model Langmuir was best suited to the experimental data. The adsorption process was endothermic and spontaneous. 100% arsenic was removed from water at pH more than 8.	[104]
15	2014	Adsorption: Pb (II)	Average width of particle around 5 nm and average length about 50 nm. The adsorption process was endothermic and spontaneous. 90% removal efficiency was found at basic pH (9.0).	[105]
16	2019	Adsorption: Lead (II)	Average particle size 20 nm. 95% removal efficiency was found at basic pH (6).	[106]

grafting polymerization reaction in the aqueous phase is used to prepare PAA-PVDF membranes. Through dipping membranes of 0.05% WT per- cent TiO₂ colloidal suspension, TiO nanoparticles (20 nm) were self-

assembled to the surface of prepared PAA-PVDF. Finally, UV radiation of the membranes (160 W) was applied on the surface of the hydropho- bic PVDF membrane to bind TiO₂ nanoparticles. In the second method, a

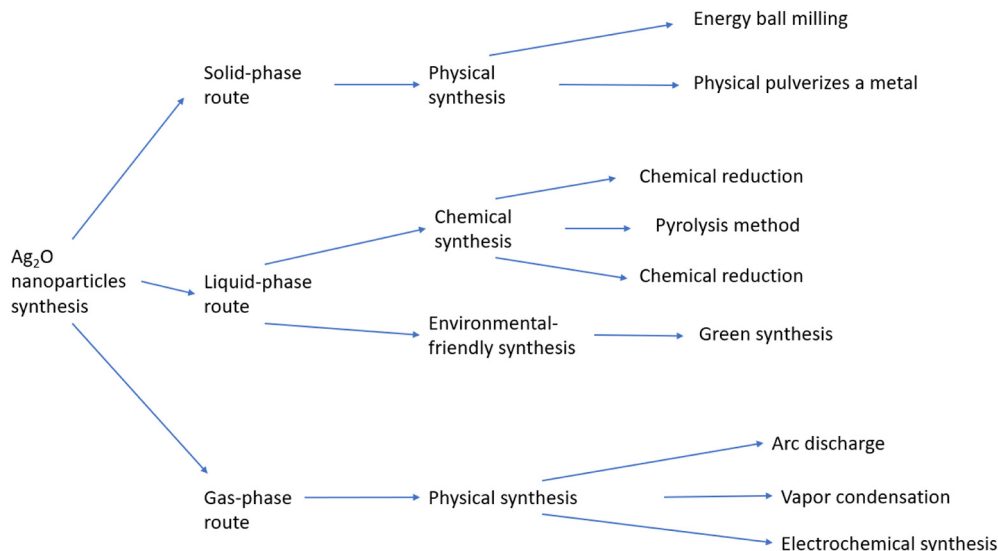
Fig. 8. Major synthetic techniques used with Ag₂O nanoparticles.

Table 6

Existing methods of Ag₂O nanoparticles and their application for wastewater treatment.

S. No.	Year	Application	Reference
1	2014	Antimicrobial Activity: <i>Bacillus subtilis</i> , <i>Staphylococcus aureus</i> , <i>Pseudomonas aeruginosa</i> , <i>Escherichia coli</i> , <i>Candida albicans</i> and <i>Aspergillus niger</i> .	Average particle size 772 nm. The highest inhibition zone values were observed for 300 (µg/ml) of Ag nanoparticles. [112]
2	2015	Photocatalytic Activity: methyl orange (MO)	Average particle size 8.33 nm. Surface area of 0.4726 m ² /g. Methyl orange was completely degraded by Ag ₂ O under UV or visible light irradiation in 120 s. [109]
3	2019	Photocatalytic Activity: Methylene Blue Antimicrobial Activity: <i>Pseudomonas aeruginosa</i> , <i>Staphylococcus aureus</i> , <i>Bacillus subtilis</i> , <i>Escherichia coli</i>	Average particle size 38.29 nm. 97.78% Methyl blue was degraded by Ag ₂ O under UV or visible light irradiation in 180 min. Ag ₂ O nanoparticles have shown strong growth inhibiting activity against Gram-negative bacteria than Gram-positive bacteria. [111]
4	2017	Antibacterial activity: <i>Streptococcus mutans</i> and <i>Lactobacilli sp.</i>	Average particle size 42.7 nm. The maximum inhibition zones on <i>Streptococcus mutans</i> and <i>Lactobacilli sp</i> was found at higher concentrations of 250 µg. [113]
5	2011	Antibacterial activity: <i>Staphylococcus aureus</i> and <i>Escherichia coli</i>	Particle sizes in the range 10–20 nm. The synthesized Ag ₂ O nanoparticles remarkable antibacterial activity against <i>Staphylococcus aureus</i> and <i>Escherichia coli</i> at PH 5 and 7. [115]
6	2019	Antibacterial activity: <i>Staphylococcus aureus</i> Photocatalytic activity: AO8 dye	Average particle size 20 nm. Nearly 95% AO8 dye was degraded by Ag ₂ O in 180 min. The Ag ₂ O nanoparticle showed excellent antibacterial against <i>Staphylococcus aureus</i> . [110]
7	2016	Adsorption of malachite green (MG)	Average particle size 55 nm. Maximum adsorption capacity 90.909 mg/g. The Langmuir isotherm model was best fitted on the experimental data. The pseudo-second-order model was followed by the adsorption process. [116]

TiO of 0.05 wt. percent was added to the monomer of acrylic acid, followed by adding an initiator and a cross-linking agent. This reactive solution was dipped with PVDF membranes, and the same methodology was used for the first method [127]. Also, in different oxidation reactions, nanoparticles play an excellent role as a catalyst. They demonstrate high catalytic sensitivity towards pollutant molecules and transform them into environmentally friendly products. [134]. Some unique properties are present in these nanomaterials such as greater surface area, high reactivity, and nano size. In particular, TiO₂ photocatalysis plays a key role in the removal of various impurities from surface water. Many researchers have photodegraded various types of pollutants such as pharmaceutical products, organic dyes, and organic pesticides under various conditions such as UV or visible-light, doped or undoped nanoparticles, and metal / non-metal doping.

Engates et al. [135] synthesized TiO₂ nanoparticles for use as a contaminant removal substrate. Nanoparticles were able to remove multiple metals (Ni, Cu, Zn, Pb, and Cd) from both San Antonio tap water and a pH 8 solution simultaneously. Youssef et al. [136] synthesized TiO₂ nanowire using hydrothermal methods. The synthesized nanoparticle was used for the removal of heavy metal (Pb²⁺, Cu²⁺, Fe³⁺, Cd²⁺, and Zn²⁺) residues from contaminated water.

Yu et al. synthesized anatase mesoporous TiO₂ (MTiO₂) and TiO₂ nanometer thin films on soda-lime glass [137]. Higher photocatalytic activity was shown by MTiO₂ thin films than by TiO₂ thin films. Shieh et al. developed a photocatalyst thin film. This photocatalyst showed a strong antibacterial action in visible light [138]. Sunada et al. illustrated a mechanism for the antibacterial activity of *Escherichia coli* cells on TiO₂ thin films. The survival of intact cells and spheroplasts was investigated as a function of the

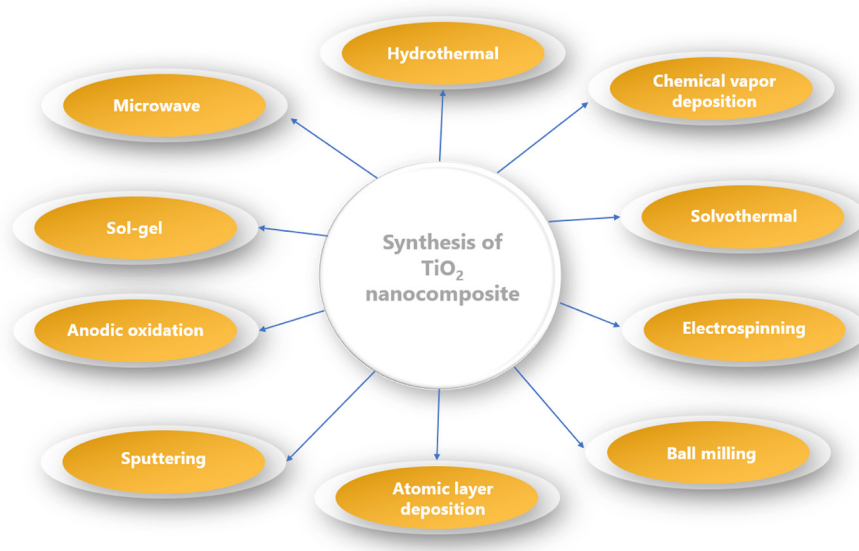


Fig. 9. Major synthetic techniques used for Ti_2O nanoparticles.

photo-illumination time [139]. Kiwi et al. used attenuated total reflectance (ATR) - FTIR spectroscopy on TiO_2 to study the photocatalytic peroxidation of *Escherichia coli* cells, peptidoglycan, lipopolysaccharide, and phosphatidylethanolamine of the *Escherichia coli* membrane wall [140]. Table 7 presents existing methods of TiO_2 nanoparticles and their application for wastewater treatment.

9. Iron Oxides Nanoparticles

In recent years, iron oxide nanoparticles, because of their simplicity and availability, have been widely used for removing heavy metals. Iron oxide-based nanomaterials possess favorable properties such as improved membrane properties, high surface area, high tensile strength, and small particle

Table 7

Existing methods of TiO_2 nanoparticles and their application for wastewater treatment.

S. No.	Year	Application	Characteristics	Reference
1	2012	Antibacterial activity: <i>Escherichia coli</i> and human pathogens	The synthesized TiO_2 nanoparticles showed promising disinfection of <i>Escherichia coli</i> and human pathogens.	[130]
2	2011	Adsorption: Cu, Zn, Pb, Cd, and Ni	Average particle size of nano TiO_2 8.3 nm and bulk TiO_2 329.8. Surface area of nano TiO_2 9.5 m^2/g and bulk TiO_2 185.5 m^2/g . At pH 6, both the nanoparticles particles were exhausted at but at pH 8, exhaustion did not occur.	[135]
3	2008	Antimicrobial Activity: <i>Thamnocephalus platyurus</i> , crustaceans <i>Daphnia magna</i> , and <i>Vibrio fischeri</i>	Particle sizes 25–70 nm. For synthesized TiO_2 nanoparticles <i>Thamnocephalus platyurus</i> was more sensitive than <i>Daphnia magna</i> .	[73]
4	2010	Antimicrobial Activity: <i>Escherichia coli</i>	Average particle size 30 nm. Surface area (m^2g^{-1}) 12.9. Toxicity (30-min and 2-h EC_{50} , $\text{mg compound l}^{-1}$) of b TiO_2 for E. coli AB1157 $\geq 20,000$, E. coli JI130 $\geq 20,000$, E. coli JI131 $\geq 20,000$, E. coli AS393 $\geq 20,000$, E. coli JI132 = 94 ± 12 , E. coli AS391 = 118 ± 43 .	[74]
5	2013	Photocatalytic activity: Methylene Blue (MB) and Rhodamine B (RhB)	Average particle size 10 nm. Surface area (m^2g^{-1}) 132. At theoretical pH 5.69 the degree of ionization was 96.9 for 1 mol l^{-1} RhB. Both MB and RhB followed pseudo-first order kinetics.	[8]
6	2006	Antibacterial activity: <i>Bacillus subtilis</i> and <i>Escherichia coli</i>	Average particle size 330 nm. The synthesized TiO_2 exhibited higher toxicity on <i>Bacillus subtilis</i> than <i>Escherichia coli</i> .	[37]
7	2018	Removal of Zn (II) and Sr (II) ions	Average particle size 25 nm. Surface area (m^2g^{-1}) >14. Highest sorption efficiency was achieved at pH 8.	[141]
8	2014	Removal of heavy metal (Pb^{2+} , Cu^{2+} , Fe^{3+} , Cd^{2+}) and Zn^{2+} .	Average particle size 30 nm. The absorption efficiency of TiO_2 was: Pb^{2+} = 97.6, Cu^{2+} = 75.24, Fe^{3+} = 79.77, Cd^{2+} = 64.89 and Zn^{2+} = 35.18%	[136]
9	2006	Antibacterial activity: <i>Escherichia coli</i>	120 nm film thickness. 99.99% removal of <i>Escherichia coli</i> was achieved in visible light.	[138]
10	2005	Antibacterial activity: <i>Escherichia coli</i>	Surface area 50 m^2/g . Effective photocatalytic peroxidation of <i>Escherichia coli</i> cell.	[140]
11	2016	Photocatalytic activity: Methylene Orange (MO)	Average particle size 7.30 ± 1.70 nm. After UV irradiation for 60 min, 98% of the 10 ppm MO was degraded.	[117]
12	2016	Photodegradation of 4-chlorophenol (4-CP), phenol and o-cresol	Average particle size 50 nm. Surface area (m^2g^{-1}) 53.3. A maximum degradation of 99% for 0.5 mM 4-CP (2.5 h), 94% for 0.5 mM o-cresol (3 h) and 97% for 0.5 mM phenol (3 h) was achieved.	[121]
13	2016	Removal of removal of Cr(VI) and Cr(III)	Particle sizes 6–14 nm. The maximum Cr(total) removal efficiency of the material is 99.02% in 60 min under sunlight irradiation.	[124]

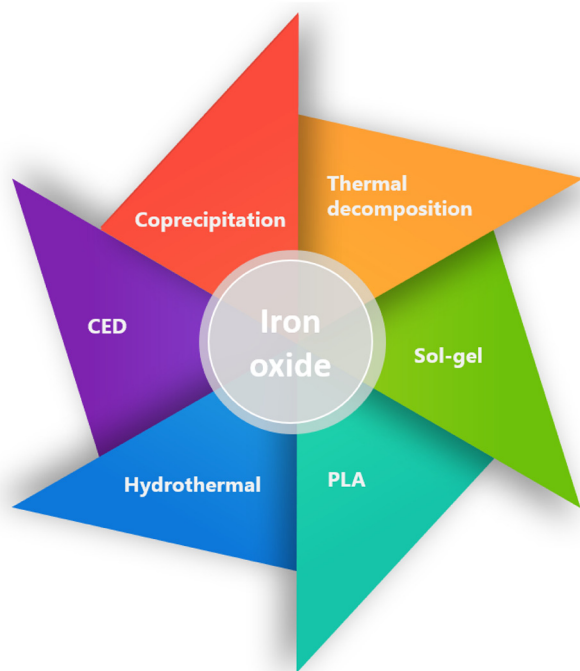


Fig. 10. Major synthetic techniques used for iron oxide nanoparticles.

size [142]. Magnetic magnetite (Fe_3O_4), nonmagnetic hematite ($\alpha\text{-Fe}_2\text{O}_3$) and magnetic maghemite ($\gamma\text{-Fe}_2\text{O}_4$) are often used as nano adsorbents. The separation and recovery from contaminated water are important challenges for water treatment due to the small size of nano sorbent materials. On the other hand, Fe_3O_4 and $\gamma\text{-Fe}_2\text{O}_4$, are easy to separate and recover from the system. Both have successfully been used to remove different heavy metals from wastewater as sorbent material [143]–[144]. Iron

oxide nanoparticles have been synthesized using different methods, as shown in Fig. 10.

Nanoparticles with iron oxides have been designed to adapt their adsorption properties by using different ligands (e.g., L-glutathione, ethylenediaminetetraacetic acid, α -thio- ω -(propionic acid) hepta(ethylene glycol) (PEG-SH), mercaptobutyric acid, and *meso*-2,3-dimercaptosuccinic acid) [145] or polymers (e.g., copolymers of crotonic acid and acrylic acid) [146]. There have been reports of a flexible ligand shell that allows the incorporation of a broad range of functional groups into the shell and ensures that Fe_3O_4 nanoparticles have their properties intact. [147]. Furthermore, a polymer shell has been found that can prevent particles from aggregating and improve nanostructural dispersion stability [150]. Polymer molecules could serve as binders for metal ions, thereby becoming a “carrier” of metal ions from treated water [151].

Iron-based nanomaterials have recently shown a remarkable sorption potential due to their high porosity, specific surface area, and strong magnetic response, which results in an extraordinary sorption capacity [148,149]. In recent years, iron-based nanomaterials have gained widespread attention because of their high BET surface area, super magnetic properties, and high pore volume [40]. Different carbon forms such as (multiwall nanotubes expanded carbon, graphene) and paramagnetic particles (Fe_2O_3) are found to be effective for the removal of toxic heavy metals such as Cd(II), Cu(II), and Pb(II) [150].

Iron oxide nanomaterials have recently been used to effectively remove wastewater from dyes. The super magnetic Fe_3O_4 nanoparticles were synthesized by Singh et al. [151] and coated with green tea polyphenols for the removal of dye from an aqueous solution. Es'haghzade et al. [152] synthesized nanoparticles of magnetic iron oxide and used them to absorb azo dye. The synthesized Fe_3O_4 nanoparticles have been shown to have a good dye removal performance in a large range of pH levels. Angamuthu et al. [153] prepared nanomaterial from an Fe_3O_4 mesoporous carbon shell and used it to degrade methylene blue dye. There was an excellent catalytic activity towards the degradation of methylene blue dye with this synthesized nanomaterial. According to Ebrahiminezhad et al. [154], processed nanoparticles of iron showed a high potential for dye removal. At six hours,

Table 8

Existing methods of working with iron oxide nanoparticles and their application for wastewater treatment.

S. No.	Year	Application	Characteristics	Reference
1	2012	Removal of Eu^{3+} , La^{3+} , Co^{2+} and Ni^{2+} ions	Average particle sizes 4–15 nm. Surface area (m^2g^{-1}) 170. 83% of cations were adsorbed after 1 min.	[144]
2	2010	Heavy metal in river water	Average particle size 8 nm. Surface area (m^2g^{-1}) >100. The adsorbant capabilities of nanoparticles were found to be superior to commercially available sorbent materials.	[145]
3	2012	Heavy metal ions removal: Cu^{2+} , Zn^{2+} , Pb^{2+} , Cd^{2+}	Particle sizes 15–20 nm. The maximum adsorption capacity was at pH 5.5.	[146]
4	2019	Removal of heavy metal	Particle sizes 10–20 nm. Surface area (m^2g^{-1}) 177. Removal efficiency 99%.	[142]
5	2009	Adsorption of Cu (II) and Cr (VI) ions	Mean diameter of 11.2 ± 2.8 nm. The maximum adsorption capacities of Cu(II) ions was 12.43 mg/g and Cu(II) ions was 11.24 mg/g.	[148]
6	2011	Adsorption of Cr (VI) ions	Average particle size 40 nm. Optimal adsorption was achieved at pH of 2–3.	[149]
7	2017	Removal of methylene blue (MB) dye	Average particle size 10 ± 3 nm. Highest adsorption capacity of 7.25 mg/g was achieved. The isothermal model Langmuir was best suited to the experimental data. The pseudo-second-order model was followed by the adsorption process.	[151]
8	2017	Removal of azo dyes	Average diameter 40–45 nm. Best removal efficiency was achieved at pH 3 and pH 11.	[152]
9	2017	Photocatalytic degradation: methylene blue (MB)	Average diameter 17 nm. Best removal efficiency was achieved at pH 3.	[153]
10	2018	Photocatalytic degradation: methylene blue (MB)	Average diameter 19 nm.	[154]
11	2017	Adsorption of malachite green and brilliant green	The maximum removal efficiency of 95% was achieved in 6 h. Average particle size 20 nm. The maximum removal efficiencies were obtained to be 99.50% and 99.00% malachite green and brilliant green.	[155]

the efficiency of decolorization in methyl orange removal was 95%. Asfaram et al. [155] investigated ultrasound-assisted brilliant green and malachite green onto Mn-doped Fe₃O₄ carbon nanoparticles. Table 8 presents the existing methods of working with iron oxide nanoparticles and their application for wastewater treatment.

10. Future Research

In the 21st century, it appears that metal-oxide nanoparticles will have a crucial role in nanomedicine and other biological applications. These types of nanoparticles can be fabricated using several synthetic routes and used effectively in various nano-medical and biological applications. However, there is still a need to prepare these nanoparticles on a commercial scale to reduce costs. Natural resources for the preparation of these nanoparticles should be sustainable, low-cost, environmentally friendly and free from toxic chemicals. For future research, it is important to produce monodispersed nanoparticles. However, the mechanism for these nanoparticles' synthesis is not currently clear. Future research should therefore focus on the mechanism by which nanoparticles can be controlled in their size and shape. The need to expand the use of nanoparticles in therapeutic applications and reduce toxicity is also an important challenge. New strategies are being developed to overcome such challenges through the use of noble metal nanoparticles through developments in nanoscience; however, their impacts on anthropological health factors must be taken into account before their extensive use. Given that most nano-materials so far have been inexpensive compared to traditional materials such as activated carbon, future applications will focus on efficient processes, where only small quantities of nanomaterials of metal oxide will be needed. In addition, more work is necessary to develop cost-effective methods of synthesis, and large-scale testing is required for the successful field application of metal oxide nanomaterials.

11. Conclusions

To date, many studies have been carried out to improve the quality of drinking water, which is directly linked to the health and safety of human beings and the environment. Nanomaterials with specific physical and chemical properties have the ability to effectively kill pollutants. The concept of nanomaterial production has been raised to give priority to opportunities for their implementation.

Metal oxide nanomaterials are preferred for the absorption of heavy metals and organic pollutants, as they have shown promising results when they are used in various applications. These types of nanoparticles are called immobilization carriers and can also be used as support carriers for biosensors and bio sorbents, though they are rarely discussed. Their success has been attributed to their physical and chemical properties, but their application to wastewater treatment is still limited.

In this review, we presented a detailed overview of five metal oxide nanoparticles: copper oxide, silver oxide, zinc oxide, iron oxide, and titanium oxide. Among these five metal oxide nanoparticles, ZnO is mostly used for different types of wastewater treatment techniques. After ZnO, CuO and TiO₂ are the most widely used metal oxides for wastewater treatment. In the literature, ZnO, CuO, and TiO₂ are used in a variety of different applications such as adsorption, photocatalytic activities, antibacterial and antifungal activities. Silver oxide is mostly used in antimicrobial and photocatalytic activities. Iron oxide is mostly used in adsorption.

Although metal oxide nanoparticles, like other nanoparticles, are useful for many applications, there are still some health hazard concerns due to their uncontrollable usage and release to the natural environment. These concerns should be addressed to make the use of nanoparticles more effective and environmentally friendly.

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