

# Clay Minerals and Metal Oxides Based Green Nanocomposites for Sustainable Water Remediation: A Review

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Received: October 20, 2023

Accepted: December 22, 2023

Published: December 28, 2023

**Citation:** Pareek P, Ledwani L. 2023. Clay Minerals and Metal Oxides Based Green Nanocomposites for Sustainable Water Remediation: A Review. *NanoWorld J* 9(S5): S438-S450.

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## Abstract

The textile, paper, and plastics industries are facing the challenges of water scarcity for dyeing, processing, and washing. A large volume of colored and heavy metal-based wastewater results from its operations. Water recycling and reuse are practical options for improving water availability while reducing environmental impacts from discharging untreated colored wastewater. Several methods are used to recycle chemical-rich wastewater: biological, chemical, and physical. Physical treatment of wastewater is the most favorable method over chemical and biological treatment because of its low cost, simple, and effective operating system, which offers different methods such as adsorption, filtration, sedimentation, distillation, etc. This review article is based on the physical method and uses various natural and low-cost adsorbents and green nanocomposites to remove dyes and heavy metals. However, several key concepts discussed here reveal the better method, including the green method, nanocomposites, water quality enhancement, low environmental impacts, and technology integration. Including this, environmentally friendly, inexpensive, natural, and achievable water conservation and treatment strategies are also discussed, highlighting the current limitations and opportunities for treating wastewater with sustainable and cost-effective methods.

## Keywords

Sustainable wastewater treatment, Adsorption, Nanocomposites, Natural adsorbent

## Introduction

The destruction of the environment is one of the most controversial problems in the world and has led to the rapid development of modern technologies. Protecting the environment and natural water sources is one of the primary responsibilities. Natural sources of water are used in various industrial processes [1]. The provision of drinking water is a fundamental necessity for human society. In addition, dangerous components are used to purify water sources. These toxic substances have caused a significant increase in unwanted compounds in soft water [2]. Rapid industrialization has led to several environmental problems, namely the accumulation of harmful substances. Our environment is believed to be contaminated with more than 700 harmful inorganic and organic microbes, which can lead to cancer and pose serious health risks to humans and animals [3]. The primary source of these irritating substances is industrial activity. Immediate manipulation is essential to tackle this problem and protect our environment. In addition to inorganic elements, even organic deterrents can be toxic and harmful [4].

Certain chemicals, such as dyes, pigments, and others, are poorly soluble in biological systems. These organic irritants can live in water and land, infect

humans, and cause health problems. Inorganic pollutants such as Cr(VI), Pb(II), Hg, As(V), and Cd(II) are considered toxic [5]. They can be consumed from numerous sources, including water, soil, and air. Contact with these elements can lead to innumerable health complications, such as cancer, damage to the nervous system, and developmental problems. Contact with organic irritants can cause skin issues, asthma, and other health hitches [6]. The influencing factors of diseases are the primary sources of diseases. These microbes have been linked to several illnesses, including cholera, typhus, diarrhea, and hepatitis A [7]. To resolve this complicated problem, simple and efficient solutions are required. Protecting the community and accessing clean, healthy air and water has become a priority. A practical and effective water treatment method is necessary to achieve this goal. However, it is also essential to consider eco-friendly treatment and pollution-free materials [8]. While conventional methods have been influential in the past, we must take action to protect the environment and our health. However, this technique has specific weaknesses. One disadvantage is the high operation and maintenance [9].

Another problem is residual toxicity, which may persist after treatment. In addition, these techniques often do not allow the recovery of valuable substances that may have been trapped in the adsorbent [10]. Because of these losses, interest in developing more sustainable and economical methods for treating water and sewage is increasing. This new method aims to reduce or eliminate the adverse effects of conventional techniques and, at the same time, improve overall water quality [11].

Nanotechnology has become the preferred solution for treating used water and water purification [12]. Nanotechnology has demonstrated enormous potential to solve the problem of water pollution among the various technologies used for water purification. Among the applied methods of water purification based on nanotechnologies, the use of nanomaterials prevails to eliminate water pollution at the molecular level. Nanocomposites based on clay can be extremely valuable in water and sewage purification because of their unique characteristics [13]. These nanomaterials have great potential for secondary processing, which makes them more sought after than traditional processing methods. Clay-based nanocomposites can effectively remove impurities from water and industrial water [14]. Nanotechnology is an area concerned with developing materials on a nanometric scale. The application of nanotechnologies has paved the way for the development of reflective materials and approaches to solving these thorny problems. The elimination of substances that pollute natural resources such as air and water is a severe problem that modern society must deal with. A promising approach to this problem is using natural materials containing nanomaterials and forming nanocomposites [15]. Nanocomposites are presented as functional solutions for the purification and remediation of wastewater. The nanoparticles have exclusive material properties, such as a large surface area, and are photo-catalytically active, effectively removing inorganic, organic, and microbial contaminants from the water. For this purpose, numerous nanomaterials have been identified that offer higher efficiency in this process. Carbon nanomaterials, polymers, zeolites, and metals containing

nanoparticles are among the nanomaterials used for water purification [16-18].

Nanocomposites can efficiently remove contaminants from the wastewater. Nanotechnology is an area concerned with developing materials on a nanometric scale. The application of nanotechnologies has covered the way for developing reflective materials and approaches to solving these thorny problems [19]. Removing elements contaminating natural resources such as air and water is a severe problem that modern society must face. A debatable approach to solving these problems is using natural materials, nanomaterials, and nanocomposites. The nanometric particles observed have been presented as valuable solutions for the purification and use of water. The nanoparticles possess unique material properties, such as a large surface, which effectively removes the water's inorganic, organic, and microbiological contaminants [20]. To overcome this issue, various nanomaterials have been determined to offer greater efficacy in the process. Carbon nanomaterials, polymers, zeolites, and metals that comprise nanoparticles form part of the nanomaterials used for water decontamination.

Researchers have used various specialized materials to remove hazardous contaminants from contaminant water. These materials include electro dialysis, adsorption, reverse osmosis, electrolysis, membrane, chemical oxidation, conventional and chemical precipitation, and coagulation [10, 21, 22]. Each of these techniques has its advantages and disadvantages. Access to affordable options is essential to give everyone a fair chance to succeed. The many multidisciplinary approaches can be costly and inaccessible to low-income earners. Chemical precipitation and coagulation techniques are commonly used in wastewater treatment processes to remove suspended solids and other contaminants from wastewater. However, these procedures can have unintended consequences, require treatments, and increase overall treatment costs. Adsorption is an environmentally friendly process to remove pollutants from wastewater. This inexpensive and convenient method allows for the treatment of large amounts of contaminants [23]. The best drying material is essential to getting the desired results. The chosen absorbent must meet specific criteria to be effective, including availability, cost, and selection. Adsorbents based on natural materials are a reliable and eco-friendly method for the efficient removal of pollutants from water [24].

Polymerous materials, industrial by-products, agricultural wastes, natural clay minerals, activated carbon, zeolite, and biomass are the most common adsorption materials [25]. The unique properties of clay minerals and kaolinite make them more efficient in water treatment [26]. They are still easy to use and amazing. However, it is useful due to certain factors. One of these factors is the low specific surface area of the ceramic material, which limits the adsorption amount. The ineffectiveness of the ceramic material as an adsorbent medium is also due to the low electronegativity of the adsorbent medium surface and the attraction of the molecules. To consume common consumables, researchers rely on materials based on nanocomposites. These products offer the unique advantages of naturally occurring minerals and other composite materials. The new poly-adsorbent effectively removes environmental

pollutants and has various advantages [27]. It is a promising material due to its high adsorption efficiency, specific surface area, thermal stability, and regenerative efficiency. It is a highly recommended ingredient for a variety of environmental cleaners. This comprehensive review describes the latest experimental parameters to be used for the removal of water pollutants [28]. The new poly-sorbent materials are removed environmentally and efficiently and have various advantages. It is a highly recommended ingredient for a variety of environmental cleaners. This comprehensive review describes the latest research protocols applied to nano-clay composites for water decontamination [28].

### Clay minerals

These sedimentary rocks are a known source of hydrated layered silicates and other minerals. This mineral consists of a  $\text{SiO}_4$  tetrahedron consisting of three oxygen molecules separated by tetrahedrons in succession [29]. A unique feature of clay minerals is that they absorb and adsorb cations equally well as heavy metals and cationic trace elements. They are highly surfaced due to their sheet-like structure which creates a negative charge on their structure. Such minerals, these stones, which are kaolin, montmorillonite, along with bentonite, illite, kaolinite, and mica are, can adsorb and capture cations from metallic and cationic trace elements effectively [30]. These are considered low-cost, nano-ceramic materials. Furthermore, these nanocrystalline minerals have properties such as porosity, rough surface, and retention capacity of adsorbed impurities [31].

### Utilization of nanomaterial in water remediation

The field of nanotechnology has seen growth, leading to the development of tools at the nanoscale level. We must take action to protect the environment. Nanotechnology offers promising solutions for improving water treatment processes and safeguarding our resources for generations [32]. By harnessing the benefits of nanotechnology, we can significantly enhance water treatment methods. Ensure environmental protection for future generations. This advanced approach is more efficient and economically sustainable in removing pollutants from water sources. Magnetic units and nanoparticles play a role in adsorbing pollutants from wastewater. Moreover, nanoparticles employ photochemistry to eliminate contaminants [33]. Due to their properties, such as increased porosity and smaller size, nanoparticles hold tremendous potential in managing water resources effectively.

## Types of Nanocomposites

The creation of materials includes types of nanomaterials. These materials stand apart from composites due to their grain size and larger surface area [34]. Each segment of a nanocomposite component has unique dimensional properties and can exist in different phases. Advanced properties of this material include electrochemical, electrically conductive, and thermal properties. Natural clay nanocomposites are divided into three main types: metallic based, carbonized, and polymeric nanocomposites, which differ based on matrix and structural organization [35].

### Metallic nano-clay composites

Metallic clay composites have unique properties such as superparamagnetic properties, high surface area, and high mechanical strength. They are excellent at absorbing contaminants in water and wastewater, making them ideal for environmental applications. Popular composites include nanomaterials such as zinc oxide/argillite, zinc oxide/argillite, nickel oxide/argillite, and titanium oxide/argillite.

### Magnetic iron oxide/clay nanocomposite

Scientists are currently focusing their efforts on the nanometric level and believe that nanomaterials have unlimited capacity to reduce the semi-surface area of water. Carbon dioxide nanoparticles clean the water very efficiently. These materials have unique properties, such as greater mobility, reactivity, adsorption capacity, catalytic properties, pre-cum, and magnetic, electrical, and optical properties [36]. Magnetite ( $\text{Fe}_3\text{O}_4$ ) and Maghemite ( $-\text{Fe}_2\text{O}_3$ ) are surprising materials given recent attention. Large surface area, exceptional thermal stability, strong mechanical properties, chemical stability, and superparamagnetic properties are common properties that do not manifest on the nanometric scale. Scientists from all over the world study these magnetic nanomaterials to reveal their potential and find new applications. Due to extensive research on nanoparticles of ferromagnetic oxide, it has been discovered that they possess or have an exceptional saturation orientation, which makes this face very valuable for various applications [37]. Clay minerals nanocomposites are an economical option for adsorption and photocatalysis since they can be quickly recovered and re-utilized. Modifying their surfaces and associated with material supports can increase their adsorption capacity and selectivity against a specific contaminant.

Recent studies have shown that various carrier top materials, in which nanoparticles are magnetically effective, participate in developing Nano-clay parallel to the first development plan. This is due to the structural arrangement of the minerals and the negative net charge, which leads to a greater specific surface area and improved adsorption capacity. The mineral has exceptional properties and is perfect because it contains buoyant pollutants. These natural minerals have a large surface area, which makes them very porous and ideal for adsorption. The configuration of the gelling layer, the distribution of the porosity of the particles in the nanocomposite, and the ability to exchange cations are vital factors in the removal of specific molecules when purifying water [38].

### Preparation of magnetic iron oxide/clay nanocomposites

Magnetic iron oxide/clay nanocomposite is typically prepared through co-precipitation and the intercalation of magnetic nanoparticles (MNPs) into a clay structure [39]. Co-precipitation is preferred because it promotes chemical reactions and interactions between clay particles in the aqueous dispersion used to create MNPs.

Figure 1 depicts the real-time production and crystallization of nanoparticles during the solvent diffusion of clay crystals. MNPs must be mixed with clay minerals in

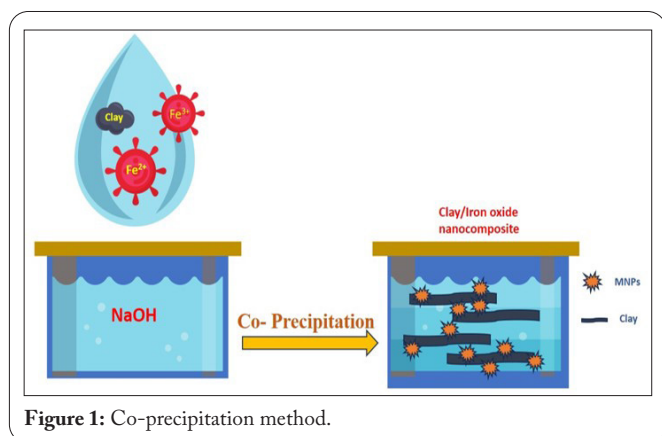


Figure 1: Co-precipitation method.

an alkaline environment to create magnetic iron oxide/clay nanocomposites [40]. A sol-gel pattern produced by  $\text{NH}_4\text{OH}$  or  $\text{NaOH}$  is crucial for successful Synthesis. It is essential to use refluxing and microwave irradiation procedures to successfully create composites of clay minerals and magnetic nanoparticles. Critical procedures include drying the sample after the reaction and calcining it. Slovakian bentonite and the  $\gamma\text{-Fe}_2\text{O}_3$  phase may produce a 35 - 42 emu/g magnetization range at 85 °C. A homogenous magnetic nanoparticle phase may be formed by merging oxidation and co-precipitation. Natural kaolinite is required to get maximum outcomes in wastewater treatment utilizing clay minerals [41]. The co-precipitated reaction approach with a one-to-one structural arrangement is advised to produce the best results while working with clay and MNPs. The adsorption capacity of nanoparticles is minor, and they may spread onto the outer and interlayer surfaces during precipitation.

#### Adsorption phenomenon of magnetic iron/clay nanocomposite

Adsorption, photo-oxidation, oxidation, and chemical techniques have all been utilized to remove pollutants from water and wastewater using magnetic iron clay nanocomposite. On the other hand, chemical pollutants, such as heavy metals, pigments, dyes, and other organic contaminants, are incredibly damaging and must be removed from water and wastewater [42]. Surprisingly, magnetic iron/clay nanocomposites are more successful at adsorption than advanced photocatalysis and oxidation approaches. This technology combines the adsorption behavior of natural nano clays with the magnetic features of MNPs from unique magnetic iron/clay nanocomposite adsorbents, resulting in a valuable instrument for eliminating pollutants from water. The data shown below contain a thorough list of magnetic iron/clay nanocomposites that may be used to replace pollutants and the processes that can be used to avoid them.

Surface functionalization has improved the adsorption capacities of these nanocomposites. Surfactants, polymers, and other chemical compounds with functional groups have been added to attract the adsorbate. Adsorption performance has been investigated using isotherm models such as the Langmuir, Freundlich, and Dubinin-Radushkevich [43]. According to the Langmuir isotherm model, monolayer adsorption occurs on a homogenous exterior, where the adsorbent pollutants do not interact with the adsorbent surface. The magnetic iron/

clay nanocomposite adsorption technique for pollutants is shown in figure 2.

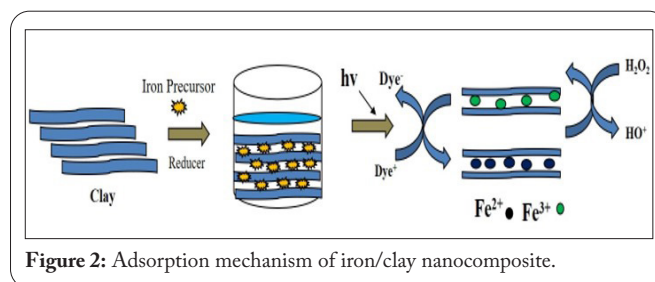


Figure 2: Adsorption mechanism of iron/clay nanocomposite.

#### Physio-chemical description of magnetic iron/clay nanocomposite

It is essential to consider the pore size distribution and specific surface area when using adsorbent and photocatalyst applications in water and wastewater treatment. More adsorption sites are accessible at more significant surface areas, strengthening the connection with the targeted pollutants. Experts often use the Brunauer-Emmett-Teller technique, which uses  $\text{N}_2$  adsorption/desorption isotherm data to calculate a given surface area precisely. The interaction between water-loving and water-hating adsorbent surfaces, the porosity of particle distribution, and the specific surface area are additional parameters that should be considered [44]. The quantity of adsorbent or catalyst, contact time duration, pH level, and size of the adsorbent particle are all parameters that impact the process of concentrated adsorption in molecules. Among these variables, the pH value significantly impacts the adsorption process during surface infarction. The zeta potential, which governs the force of attraction or repulsion between particles in a magnetic field, is another crucial variable [45]. Higher adsorption capacity and pollutant elimination are reported when a greater magnetic field is applied. Furthermore, a transmission electron microscope investigation demonstrated that the size of nanoparticles substantially influences the strength of the magnetic field.

#### Kaolin/zinc oxide nanocomposite

Activated carbon, graphene oxide, agricultural wastes, metals/metal oxide nanoparticles, zeolites, industrial byproducts, and many other adsorbents have all been effectively used to remove harmful waste from wastewater [46]. A zinc oxide nanoparticle is one such adsorbent; it is a semiconductor metal oxide with outstanding adsorption and photocatalytic activity, whether light is present or not (Table 1).

Zinc oxide nanoparticles are non-toxic, highly catalytic, chemically stable, and corrosion resistant. Zinc nitrate, zinc sulfate, zinc chloride, hexahydrate, zinc acetylacetonate, and zinc acetate dihydrate are all precursors that may be created chemically or physically. It is critical to recognize that zinc oxide nanoparticles have unique properties that make recovery from the aqueous phase difficult [58]. These nanoparticles are difficult to recover because of their low specific surface area, tiny particle size dispersion, and rapid aggregation. Furthermore, releasing these nanoparticles into the atmosphere might endanger human health and the ecosystem.

**Table 1:** Magnetic iron oxide/clay nanocomposite utilized for pollutant adsorption.

S. No.	Magnetic iron oxide/clay nanocomposite	Adsorbate	pH	Removal capacity (mg/g)	Isotherm model	Ref.
1.	Bentonite/iron oxide magnetic	Sr(II) and Ba(II)	9	124.8 and 120.0	Langmuir	[47]
2.	MnO <sub>2</sub> /Fe <sub>3</sub> O <sub>4</sub> NPs/diatomite	Methylene blue	4 - 8	Nearly 100%	-	[48]
3.	Fe <sub>3</sub> O <sub>4</sub> /kaolinite	Anionic Direct red 23	7	22.88	Langmuir	[49]
4.	γ-Fe <sub>2</sub> O <sub>3</sub> /halloysite	Aniline, 2,6-dichloroaniline, and 2-chloroaniline	-	-	Langmuir	[50]
5.	Fe <sub>3</sub> O <sub>4</sub> /bentonite	Cu and Pb	-	82.3 and 97.8%	-	[51]
6.	Fe <sub>3</sub> O <sub>4</sub> /halloysite	As(III) and As(V)	6	408.71 and 427.72	Langmuir	[52]
7.	Fe <sub>3</sub> O <sub>4</sub> /Al-pillared bentonite	Orange II	3	-	-	[53]
8.	Fe <sub>3</sub> O <sub>4</sub> -sepiolite	Methylene blue	10	98.5%	Freundlich	[54]
9.	Fe <sub>3</sub> O <sub>4</sub> /montmorillonite	Methylene blue	9	99.47%	Langmuir	[55]
10.	Montmorillonite clay/Fe <sub>3</sub> O <sub>4</sub> /sepiolite	Diuron	3	-	-	[56]
11.	Graphene/magnetite/montmorillonite	Methylene blue	11	225.0	Langmuir	[57]

Thus, it is essential to avoid these problems by using a suitable medium for the nanoparticles to cling to. Clay minerals include reinforced nanoparticles with exceptional mechanical properties, a high adsorption capacity, and thermal stability [59]. Clay minerals are the best option since they are readily available and affordable. Understanding a substance's molecular structure is essential for identifying its unique chemical and physical features. These include their ability to store water, participate in chemical reactions, and exchange ions. Ion exchange and adsorption processes can remove contaminants from wastewater, but it is crucial to consider the load. It is essential to study how effectively the adsorption technique works under different experimental conditions to ensure its efficiency.

Therefore, the ability of kaolin and hybrid kaolin clay needs to be studied carefully to remove contaminants from wastewater [60]. Natural clay minerals were modified to dope zinc oxide nanoparticles over the surface. The entire surface of the nanocomposite is to be studied for its ability to act as an adsorbent for extracting pollutants from wastewater. The aim was to remove heavy metal ions from the wastewater. The resulting zinc oxide nanoparticles had a 19.65 mg/g capacity and a remarkable removal efficiency of 93%. The study also identified the best fecal settings, which included a pH of 5.0, an initial ion concentration of 10 mg/L, a contact time of 1 h, an adsorbent dose of 0.1 g, and a temperature of 70 °C. Kaolin and hybrid clay's efficiency in removing contaminants from water and wastewater is illustrated through these findings [61]. Findings demonstrate that the carbon nanopores coated with zinc oxide possess maximum sorption ability. This threshold was 2.5 mmol/g, enabling efficient elimination of 90% of Pb(II). Unlike zinc oxide, the sorption capacity was 2.38 mmol/g, and the removal efficiency reached 78%. By investigating how zinc oxide/graphene oxide composites purge hazardous toxins from water, scientists aimed to identify new techniques for water treatment. These contaminants consisted of copper. The enhanced adsorptive ability of zinc

oxide nanoparticles was determined to be attributable to the graphene oxide and polyaniline polymer-support materials [62]. The investigation showed that a zinc oxide and bentonite mixture could successfully remove safranin pigments from aquatic environments. Nanotechnology is employed to address the problem of metal pollutants in wastewater such as, the surface area of zinc oxide and montmorillonite composite was optimized and applied as a more effective absorption of copper and lead molecules [63].

#### Synthesis of kaolin-based zinc oxide nanoparticles

It was taught how to make clay and zinc oxide nanocomposites utilizing sol-gel and wet impregnation. The presence of zinc oxide nanoparticles was determined to be responsible for the zinc oxide/clay nanocomposite's excellent adsorption capabilities. A NaOH solution was added dropwise to correct the pH after dissolving 5.0 g of Zn(C<sub>2</sub>H<sub>3</sub>O<sub>2</sub>)<sub>2</sub> in 50.0 cm<sup>3</sup> of distilled water. After 30 min of churning, a white colloidal slurry was generated, which included approximately 0.5 g of the kaolin clay particles and was churned for up to 1 h at 150 rpm. Through rigorous testing and persistent labor, a homogenous gel was made utilizing clay and zinc oxide. It was cleansed with distilled water and then gently dried in the oven at a specific temperature after being given time to grow in the environment. After a vigorous heating procedure, the resultant structure may be examined using X-ray diffraction (XRD) pattern analysis. The findings demonstrate that the hexagonal zincite structure was visible by diffraction and that the zinc oxide nanoparticles were uniformly distributed throughout the kaolin matrix. The low-intensity values further supported these results, corresponding to the conventional data for crystal structures at 2. The XRD pattern findings point to a zinc oxide and kaolin nanocomposite combination. The nanocomposites were very crystalline based on the technology used to get these findings. It was found that some of the nanocomposites included kaolinite and zinc oxide in them, whereas others were primarily composed of zinc oxide [64]. Because of the tiny

and intense diffraction patterns at the 12.0° and 26.9° peaks, kaolin clay was present in the nanocomposite samples. When the zinc oxide/kaolin nanocomposites were made, the pH level was 8.0. The Fourier transform infrared spectra of the kaolin/zinc oxide nanocomposite made at varied pH values and calcinated at pH 8.0 clearly showed the distinctive reflection structure of zinc oxide, suited for high-resolution transmission electron microscope analysis.

### Titanium dioxide/kaolin clay nanocomposite

#### Titanium dioxide/kaolin nanocomposite preparation

A titanium dioxide/kaolin nanocomposite was made using kaolin clay with particles smaller than 5 micrometers. It was accomplished by introducing HCl to the powder and stirring it for three hours at room temperature, resulting in a yellow-red solution that was separated via decantation. The kaolin clay units were mechanically agitated with NaOH for approximately 30 min, followed by decantation, filtration, and washing with distilled water to create the required white powder [65]. Finally, the sample was dried in an oven set to 80 °C for four hours. The authors discovered that the fundamental compositions of the synthesized titanium dioxide/kaolinite nanocomposite and the raw kaolin material differed after completing chemical analyses using XRD, scanning electron microscope, and X-ray fluorescence spectroscopy.

The nanocomposite had K<sub>2</sub>O, MgO, CaO, SiO<sub>2</sub>, Na<sub>2</sub>O, and a little degree of loss of ignition, whereas the raw kaolin clay had a SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio of 2.69. On the other hand, the titanium dioxide/kaolinite nanocomposite exhibited a SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio of 3.87. The raw kaolin clay was discovered to predominantly consist of kaolinite and metal oxides during X-ray fluorescence spectroscopy and XRD examinations. HCl and NaOH solutions were used to remove any impurities or oxides and purify the kaolin minerals. This resulted in the formation of a nanocomposite mainly composed of titanium dioxide and aluminum silicates, with titanium dioxide dominating.

The metal components were extracted from raw kaolin clay using hydrochloric acid and sodium hydroxide, which changed its chemical composition and enhanced its surface area and adsorption capacity. Experiments using titanium dioxide and kaolinite were carried out to generate a variety of nanocomposites. The author studied the crystal structure of the titanium dioxide nanoparticles and minerals by integrating the kaolin clay matrix, which resulted in outstanding catalytic characteristics. In addition, acid-activated clay was created by chemically activating coal-bearing kaolin. Adding titanium dioxide nanoparticles improved the clay's capacity to absorb and remove colors, resulting in an efficient solution. A comprehensive examination of the preparation of titanium dioxide/kaolinite nanocomposites revealed the significance of the shape of the catalysts in producing this composite material [66]. The study aimed to evaluate the photocatalytic effects on substances using clay, and the results were favorable. The secrets of this one-of-a-kind procedure were revealed by combining titanium oxide sulfate with clay and heating it in a furnace to 600 °C. Combining titanium dioxide

and kaolin clay may significantly improve their distinctive properties and adsorption capacities, paving the way for a bright future in organic pollutant degradation. Using kaolin clay as a supporting material in producing titanium dioxide nanoparticles is beneficial since this results in finer dispersion and smaller particle size than utilizing individual titanium dioxide samples. It is intriguing to think about how kaolin clays might organize the size of titanium dioxide particles, provide more active sites for catalyzing processes, and eventually improve the photocatalytic properties of these materials. The field of pollutant degradation may undergo a revolution due to this discovery.

### Clay/polymers nanocomposite

Minerals such as kaolinite and clay have enormous potential to improve polymeric goods' functioning and mechanical qualities. These clay-polymer nanocomposites are changing the way we tackle pollution in the environment. Three kinds of phase-separated micro composites exist intercalated nanocomposites, exfoliated nanocomposites, and phase-separated micro composites [67]. Exfoliated nanocomposites stand out because they equally distribute clay particles throughout the inside of the polymer matrix while having a thickness of just around 1 nm.

A clay/polymer nanocomposite is a material formed by combining silicate crystals with a polymer chain. This method produces a micro-composite composed of a polymer surrounded by silicate layers. Biodegradability, deformability, chemical resistance, thermal stability, and mechanical strength are all remarkable properties of nanocomposite polymers. Polypropylene polyester, polyvinyl chloride, chitosan, epoxy, polyurethane, polystyrene, and other polymers can form this nanocomposite [68]. Polymer-clay nanocomposites are synthesized using three distinct techniques: solution-blending, melt-bending, and *in-situ* polymerization [69].

#### Solution-blending technique

The solution-blending technique is a simple and less time-consuming process. Firstly, the polymer is kept in a solvent to harden and make the material more substantial, while the clay material is dispersed in the same solvent. Once the clay is evenly distributed, it is then added to the polymer solution. The mixture is then allowed to homogenize for a while before being cast onto a level support, and the solvent is evaporated. After solvent evaporation, the final product can be used as a clay-polymer nanocomposite after appropriate characterization. The solution blending approach is shown in figure 3.

#### Melt-bending method

The second process of composite preparation is a melt-blending method; it improves the mixing of the clay and polymer components and employs a melt-integration technique to strengthen the clay particles towards the polymer matrix. This enables the resultant nanocomposite to strengthen operations that generally demand higher temperatures than the polymer melting point. Figure 4 depicts the processes involved in the melt-blending operation.

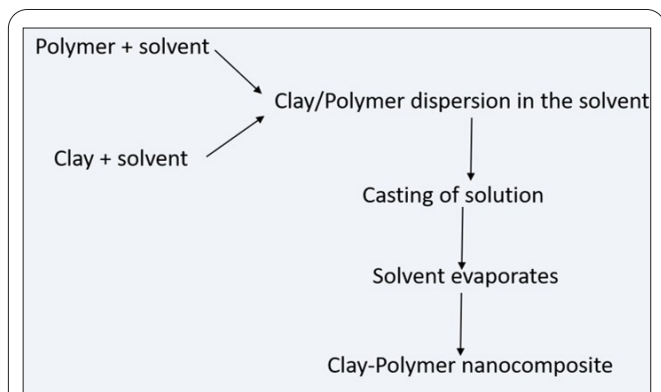


Figure 3: Synthesis of clay-polymer nanocomposite adsorbent by solution method.

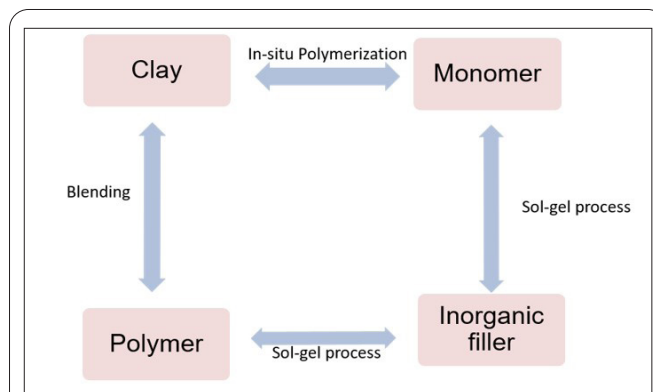


Figure 4: Different ways of synthesis of clay/polymer nanocomposite.

### In-situ polymerization technique

In-situ polymerization is a crucial method to investigate polymer preparation. It may significantly enhance the compatibility of clay and polymer components. This technique uses a monomer solution that goes through polymerization to disperse clay molecules evenly inside the polymer matrix. The result is a thin polymer layer containing clay atoms that offer the polymer matrix extraordinary support. Due to its adaptability, the material is ideal for various processing methods, including extrusion and compression casting. Figure 4 explains the in-situ polymerization procedures.

Metal ions, bacteria, phenols, pesticides, pigments, and dyes may be removed using clay/polymer nanocomposites, which can filter water and wastewater [70]. It is critical to guarantee our drinking water's safety and avoid water-borne diseases caused by hazardous germs, such as typhoid, diarrhea, and dysentery. Although chlorination was initially the primary approach for pathogen reduction, it is now less often utilized owing to the development of additional hazardous pollutants from soluble organic contaminants such as halo acetic acids, trihalomethanes, and chlorophenols. On the other hand, nanocomposite technology is a promising and inspirational alternative that does not produce nitrosamines.

Sand has traditionally been used for slow filtering. However, this approach has proven less successful since it is slow and prone to clogging. A novel method has been developed that uses a clay/polymer combination. A significant promise for removing germs and other contaminants has been shown for this combination. Copper-doped montmorillonite polyethylene composites have been tested for their capacity to destroy *Escherichia coli* germs. It contains a montmorillonite-Cu<sup>+2</sup> complex with antibacterial solid effects that can eliminate up to 99% of *Escherichia coli*. Numerous microorganisms are inhibited by other clay/polymer composites, including montmorillonite/poly dimethyl oxane/chlorhexidine acetate and clay/poly dimethyl oxane/chitosan/silver [71]. *E. coli* can be removed using starch graft-based surface-modified bentonite clay. Cationic molecules on the clay's surface are effective for microbial clearance. Monomers on the clay surface improve the nanocomposite with positive charges that electrostatically attract the microbial cell negative phospholipids, producing cell interference. In addition,

chitosan/montmorillonite nanocomposites have antibacterial action and can eliminate *E. coli* and *Staphylococcus aureus*. Several clay/polymer nanocomposites were synthesized as tablets or powders utilizing thin-coated layer techniques. This approach increases the reusability of the adsorbent after the adsorption process. Various organic and inorganic pollutants may be removed from wastewater using clay/polymer, as shown in table 2. Directly discharging organic, microbiological, and inorganic contaminants from water sources has a significant environmental effect. Polyphenolics, tannic acid, dyes, pesticides, chromium, and colors have threatening effects on marine life. Among those pollutants, tannic acid compounds are often found in water and on the soil surface, which is harmful to aquatic as well as terrestrial life. Chitosan-polymer can remove tannic acid from wastewater aided the sorption. Interestingly, the synthesized nanocomposite absorbed more tannic acid molecules due to H-bonding, electrostatic interactions, and van der Waals forces on the protonated clay surface in the solid medium with chitosan resin polymer dispersion.

Similarly, polymer reinforcement with a clay surface reformed with a hexadecyl trimethyl ammonium surfactant was used to remove organic phenolic chemicals such as phenols and 4-chlorophenol. This adsorbent's elimination capacity for phenols and 4-chlorophenol was 0.335 mg/g and 0.119 mg/g, respectively. Similarly, trinitrophenol and trichlorophenol were removed using montmorillonite, synthesized from poly-4-vinylpyridine-co-styrene, with 99.6% and 60% removal efficacy, respectively [72]. According to this article, the adsorption process generates a weak van der Waals force. Similarly, the elimination of phenolic chemicals was explored by altering montmorillonite clay with a nanocomposite of cetyltrimethylammonium poly (diallyl dimethylammonium). The ability to absorb phenols was improved. Many studies have been done to help remove colors from water and wastewater using a clay/polymer nanocomposite.

In this study, many researchers proposed using chitosan polymer as a promising adsorbent. Findings suggested that pH is the most critical factor determining adsorption. A low pH provided a positive charge and allowed electrostatic contact between the nanocomposite surface and the dye molecules. At maximum pH, the charge becomes negative and decreases adsorption due to repulsive interactions between the dye

**Table 2:** Clay-polymer nanocomposite adsorbent for the removal of different contaminants from wastewater.

S. No	Adsorbent type	Adsorbate	pH	Temp. (K)	Adsorption capacity (mg/g)	Isotherm model	Kinetic model	Ref.
1	Chitosan/bentonite	Cr(VI)	3	298	133.85	Langmuir	Pseudo-second order	[74]
2	Chitosan-coated attapulgite	Cr(VI)	2	298	294.12	Langmuir	Pseudo-second order	[75]
3	Chitosan-g-poly (acrylic acid)/ montmorillonite	Methylene blue	6.5	303	1859	Langmuir	Pseudo-second order	[76]
4	Palygorskite/chitosan resin microspheres	Tannic acid	8	303	455	Langmuir	Pseudo-second order	[77]
5	Amino-modified polyacrylamide-bentonite	Humic acid	4-7	303	103.91	Langmuir	Pseudo-second order	[78]
6	Chitosan/montmorillonite	Basic blue 9, Basic blue 66, and Basic yellow 1	-	293	46 - 49	-	-	[79]
7	Humic acid-modified bentonite	o NH <sup>4+</sup> -N and NO <sup>3-</sup> -N	7	-	96.4%	Langmuir	Pseudo-second order	[80]
8	Polyaniline/montmorillonite clay nanocomposites	Acid green 25	6	293	100%	Langmuir	Pseudo-second order	[81]
9	Kappacarrageenan-poly(acrylamide)/sepiolite	Cationic crystal violet	10	-	-	Langmuir	Pseudo-second order	[82]
10	Polyaniline/montmorillonite clay nanocomposites	Methylene blue	-	-	0.062	-	-	[83]
11	Montmorillonite/Poly(acrylic acid-co-2-acrylamido-2-methyl-1-propane sulfonic acid) Superabsorbent Composite	Ni <sup>2+</sup> , Cu <sup>2+</sup> , and Pb <sup>2+</sup>	-	-	211.0, 159.6, and 1646.0	Langmuir and Freundlich	-	[84]
12	Poly(AA-co AMPS)/montmorillonite nanocomposite hydrogel	Methylene blue	10	328	215	Redlich-Peterson	Pseudo-second order	[27]
13	Na-montmorillonite//uracil-poly(oxyethylene) nano-composite	Methylene blue	-	-	39.6	Langmuir	Pseudo-first order	[85]
14	Polyaniline modified bentonite	U(VI)	< 6.5	333	17.5	Langmuir	Pseudo-second order	[81]
15	Glutaraldehyde cross-linked chitosan-coated bentonite clay capsules	Cr(VI)	2	298	106.444	Scatchard adsorption isotherm	-	[86]
16	Alginate-montmorillonite/polyaniline nanocomposite	Cr(VI)	< 4	328	29.89	Freundlich	Pseudo-second order	[87]
17	Chitosan/attapulgite/poly(acrylic acid) hydrogel	Pb <sup>2+</sup>	4.8	-	531.9	Langmuir	Pseudo-second order	[88]
18	Bentonite/ thiourea formaldehyde	Pb(II), Mn(VII), and Cr(VI)	4, 4, and 2	-	13.38, 14.81, and 4.20	Langmuir	Pseudo-second order	[89]
19	Chitosan-g-poly(acrylic acid)-bentonite composite	Cu, Zn, Cd, and Ni	6, 7, 6, and 8	-	88.5, 72.9, 51.5, and 48.5	Langmuir	-	[90]
20	Chitosan/clinoptilolite (with penta ethylene hexamine)	Nitrate	4	-	277.77	Freundlich	Pseudo-second order	[91]
21	Attapulgite/poly(acrylic acid-co hybrid hydrogels	Pb <sup>2+</sup> and Cu <sup>2+</sup>	5	-	34.98 and 32.60	Freundlich	Pseudo-second order	[92]

molecules and the nanocomposite surfaces. The cross-linked chitosan in the composite is composed of epichlorohydrin, making it water-insoluble and increasing its adsorption ability. Researchers studied the microbiological activity of chitosan montmorillonite and chitosan/clay composites with silver nanoparticle surface modification. Multiple treatments, including HCl, steam, and sodium hypochlorite, can prevent microbial growth and reuse clay/polymer composites. Clay/polymer composites increase antibacterial activity and

efficiently purify polluted water [73].

Furthermore, the researchers developed clay/chitosan nanocomposites with a 446.4 mg/g sorption capacity for removing the rhodamin-6 G dye [93]. The chemical adsorption kinetics of dye molecules were rationalized using a pseudo-second-order kinetic model. In addition, researchers investigated methylene blue removal using a clay/chitosan nanocomposite. The most effective removal efficiency was 99.99%, while the most effective adsorption capacity was 152.3

mg/g [94]. The Langmuir isotherm followed the adsorption equilibrium experimental data, and pH was used to estimate the adsorption performance. Researchers also developed chitosan-modified nano-clays with adsorption capabilities of 477 mg/g and 440 mg/g, respectively, to remove reactive blue-21 and red-141 dyes [95]. The experimental adsorption results for the equilibrium of the two dyes fit the pseudo-second-order kinetic model [96].

Overall, the chitosan-modified clay nanocomposite's reactive yellow-3 RS dye adsorption capacity was 71.39 mg/g, much greater than the 6.4 mg/g of the unmodified clay materials [97]. Due to repulsion forces between similar charges, the adsorption process slowed down at the highest pH level. However, the chitosan surface's positive charge at the lowest pH level enhanced the adsorption process through electrostatic attraction. To remove methylene blue from an aqueous solution, sodium bentonite and poly-uracil (ethylene oxide) were recently used to generate a clay-polymer composite. It is incredible how well methylene blue is separated throughout adsorption. Microbial metalloids and toxic metals, including Cr, As, Cd, Ni, Sb, Cu, U, Se, and Pb, are the leading causes of heavy metal pollution in water. Heavy metals and metalloids pose a severe risk to humans and wildlife, as they can accumulate in various species and lead to harmful consequences. As a result, eliminating them from water sources and preserving a healthy ecology is critical. Clay minerals and chitosan have recently been used to remove polluted heavy metals from water supplies. It is encouraging to see that we are taking steps to solve this problem and protect our planet. In a recent study, researchers prepared a clay-chitosan/silver nanocomposite that was highly effective at eliminating Cu(II) ions. The best results were obtained at pH 7.0 with a remarkable adsorption capacity of 181.5 mg/g [98].

Interestingly, the main reason for the substantial adsorption at pH 7.0 was the interaction of Cu(II) ions with chitosan particles. A poly/methacrylic acid/chitosan/bentonite nanocomposite was developed to remove various heavy metal ions such as Hg(II), Pb(II), and Cd(II). This combination had excellent removal efficiencies of 94%, 89%, and 78% for Hg(II), Pb(II), and Cd(II), respectively, at high pH. Fe(III) and Cr(II) ions could be successfully removed from an aqueous solution using a chitosan/attapulgitite composite [99]. These results provide insights into innovative materials for effective heavy metal ions removal. According to the study results, Fe(III) and Cr(II) had optimal adsorption capacities of 10.41 mg/g and 11.65 mg/g, respectively, at adsorbent doses of 0.2 g/L [100]. This is due to the covalent interaction between the adsorption surfaces and the metals. Researchers also investigated using a chitosan/Al-pillar montmorillonite composite with a 15.7 mg/g adsorption capacity for Cr(VI) removal [101]. In addition, the composite synthesized from clay/chitosan at a ratio of 1:0.45 successfully removed Cu(II) and Pb(II) ions at pH 6.5 using chemisorption techniques with removal efficiencies of 96% and 99.5%, respectively [102]. These findings show that these composites have the potential to be employed as excellent adsorbents for heavy metal removal from polluted water sources. Experiments were conducted to investigate the efficacy of altering the

mass clay-to-chitosan ratios in eliminating lead compounds from aqueous solutions. They investigated the 1:1, 1:2, and 2:1 ratio and discovered that the most efficient ratio was 1:1, eliminating 201.5 mg/g of lead ions [103]. This achievement is likely due to the polymer and clay components' pores and vast surface area promoting Pb ion removal. Researchers observed in a similar investigation that increasing the weight of clay in a polystyrene polymer composite by 5% increased Pb(II) adsorption from aqueous solutions. A chitosan-grafted polyacrylic acid bentonite nanocomposite was also shown to be efficient in eliminating metals such as Cd(II), Zn(II), Ni(II), and Cu(II) [104]. It promises to eliminate heavy metals from water sources, reducing their detrimental effects on human health and the environment. This knowledge might open the path for developing low-cost technologies to enhance water quality in communities throughout the globe. The best pH values for eliminating Cd(II), Ni(II), Cu(II), and Zn(II) were discovered to be 6, 8, 6, and 7, respectively [105]. A negative ion with a positive heavy metal charge on the adsorbent surface allows maximal adsorption at a higher pH value.

Furthermore, the researchers evaluated the removal processes of Pb(II), Ni(II), and Cu(II) using chitosan immobilized on bentonite clay using ethylene glycol diglycidyl ether. The maximum adsorption capacity for Ni(II), Cu(II), and Pb(II), ions was found to be 15.82 mg/g, 21.55 mg/g, and 26.38 mg/g, respectively [106]. The study also revealed that an exothermic adsorption process occurred for both metals, and the entropy behavior decreased.

## Nano-clay Adsorption Mechanism in Water Treatment

According to the current study, nano-clay adsorbents are a better solution for absorbing contaminants from water and wastewater. This is because the surface adsorption process is aided by using nano clay. Previous research has demonstrated that these low-cost materials are non-toxic and have a higher ion exchangeability potential [107]. Furthermore, kaolinite, bentonite, illite, pyrophyllite, sepiolite, and montmorillonite have been recognized as helpful for eliminating contaminants from wastewater. Nano-clay minerals are beneficial because they have a larger specific particle surface area and can remove more pollutants.

Furthermore, they are suitable for different pollutants' adsorption because of their thermal stability, chemical resistance, and structural, and mechanical properties. The presence of charged molecules on the clay surface has a complex relationship with the adsorption process. Several techniques have been proposed for the adsorption of metal ions utilizing nano-clay materials, including chemisorption, physical adsorption, and micro-precipitation. Clay made of organic molecules may be created via the interchange of organic molecules with cations.

## Conclusion and Future Prospective

Using nano-clay materials to produce novel chemicals is a cost-effective, long-term health and environmental

protection solution. Low-cost materials can replace standard decontamination processes to treat water and wastewater efficiently and environmentally. Clay composites doped with different compounds are more likely to remove impurities. According to the literature, integrating new materials at the nanoscale level may increase clay mineral proficiency and allow for the creation of stable clay nanocomposites with improved properties. Stable adsorbents, photocatalysts, and catalytic nanomaterials may be created by incorporating functional and active components onto nano-clay surfaces.

These nanomaterials are well-suited for surface modification, particle geometry modification, and enhanced surface area, making them ideal for adsorption. Clay-based nanocomposites have received much interest in wastewater treatment and have been used in several ways to treat contaminated water. This approach has enhanced adsorbent recovery and process sustainability in wastewater treatment. The existing status of advancement in mass manufacturing and sophisticated technology applications must be improved. Furthermore, scientists frequently tested the nanocomposites with an artificial wastewater solution. It is critical to thoroughly evaluate the application of clay-based nanocomposites in treating industrial waste products. Industry specialists must extensively study the possibility of large-scale production in this area.

## Acknowledgements

The authors want to acknowledge the help, resources, and facilities provided by the Department of Chemistry, School of Basic Sciences, Manipal University Jaipur.

## Conflict of Interest

The authors state that they have no known conflicts of interest or personal ties that may have influenced the work disclosed in this study.

## Funding

The authors thank the Department of Science and Technology, Rajasthan (ref. no. P.7(3)V.Pro./R&D/2016/4895 dated 07.07.2017) for their financial assistance..

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